HYDROGEOCHEMICAL EXPLORATION AT TSUMEB

J.W. MARCHANT

Thesis submitted in fulfilment of the requirements for the
degree of Ph.D at the University of Cape Town

VOLUME Ic

containing a supplement entitled

"A REVIEW OF THE HISTORY AND LITERATURE OF
GROUNDWATER HYDROGEOCHEMICAL EXPLORATION FOR ORES"

and

NUMERICAL AND ALPHABETICAL LISTINGS OF REFERENCES

cited in either the main body of or appendixes to the thesis,
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PART 1

A REVIEW OF THE HISTORY AND LITERATURE OF GROUNDWATER HYDROGEOCHEMICAL EXPLORATION FOR ORES

SUMMARY

MARCHANT, J.W. A review of the history and literature of groundwater hydrogeochemical exploration for ores. Supplement to Ph.D. thesis, University of Cape Town, 1980.

A review is given of the historical development and current status of groundwater hydrogeochemical exploration. Over one thousand references are cited. The review concentrates particularly on the search for uranium and base metals and to a lesser degree on the location of other types of mineralization. The questions of hydrogeochemical prospecting for petroleum and the significance to exploration of the isotopic composition of the constituents of natural waters are not examined in detail.

It is intended that this review will serve as an expanded index to the available literature. Thus it does not take the form of a conventional review or textbook, in that no attempt is made to draw overall conclusions about particular concepts, techniques or principles. On the contrary, the objective of the reviewer has been to summarize the individual works and views of numerous authors in various historical periods, and the reader is left to draw his own conclusions and to undertake additional reading of pertinent original reference material.

The above notwithstanding, it is suggested that the fundamental nature of the science of hydrogeochemical exploration can be adequately described in terms of a few essential characteristics:

(A) It is very much an applied science and is most frequently employed in direct and aggressive searches for mineral wealth, rather than in an ivory tower environment. Most of the data remain unpublished.

(B) It is highly multi-disciplinary science, which has continuously absorbed and benefitted from advances in a host of related fields such as

SUMMARY (Cont.)

chemistry, geology, physics, hydrology and applied mathematics, and from new technological developments of all kinds.

(C) It is a strongly empirical science. Although the principles are everywhere the same, the optimum methods of applying them often vary strongly from one area to another, and great attention is always paid to local features of geology, physical hydrology, quality of water, topography, climate, types of ores, problems of sampling and contamination, etc. In almost every instance the nature, magnitude and effect of these variations must be determined by empirical orientation studies.

These three characteristics are, in general, common to all branches of geochemical exploration, but the hydrogeochemical method has two other features that set it apart as the *enfant terrible* of the group:

(D) Extremely minute amounts of elements are often sought in water samples. This involves particularly sophisticated and potentially troublesome procedures for sample collection and analysis.

(E) The method yields results that tend to be particularly difficult to interpret - this owing to the high mobility and reactivity of water.

The first (introductory) part of the text is very general and examines the emergence of groundwater prospecting as a branch of geochemical exploration. Hydrogeochemistry is one of the oldest branches of mineral exploration. The principles involved were clearly understood by sixteenth century writers and a sprinkling of references to water sampling in prospecting is found throughout what might be termed the "Dark Ages" of geochemical exploration (c. 1600-1932). However, hydrogeochemistry was a slow starter in the "Renaissance" of the nineteen-thirties, and it was only after 1939 that real interest in the method reawakened. Groundwater hydrogeochemistry became a recognizably separate branch of the art only after 1945.

SUMMARY (Cont.)

The second (main) part of the test is concerned specifically with the literature of groundwater hydrogeochemical exploration and is subdivided into several sections, covering various historical periods. There is apparently very little of significance in the literature between the amazing writings of Agricola (1546) and those of the modern pioneer Henniger (1925), who attempted to use the sulphur content of brines as an exploration guide to petroleum. Nevertheless numerous indirect advances, which would later serve as a foundation, were made in many related disciplines, viz: analytical techniques, studies of the compositions of natural waters, principles of water/rock interaction and the development of the main concepts of the science of geochemistry. Of notable importance here are the early investigations of the radioactivity of spring waters (c. 1905-1940), and the pioneering works of Vogt (1939), Feigelson (1940) and Miholic (1940). Real expansion of the literature of groundwater hydrogeochemical exploration began after 1948. In the period 1949-1952 Japanese pioneers were very active and in the West classic papers were published by authors such as Hawkes, Webb, Huff and Lovering. Most of this activity was directed towards finding base metals. Steady growth continued during 1953-4 and many Western workers began to take an interest in the hydrogeochemistry of uranium. The Japanese studies proceeded but very few papers from the Soviet Union reached the West. Nevertheless it is obvious that Russian scientists were engaged in a good deal of fundamental research. By 1955-7 the flow of translated Russian literature had increased greatly and it is clear that the Soviets were world leaders in the field by that time, especially in the search for base metals. In the West during this period there was a great increase in the application of the hydrogeochemical method in prospecting for uranium. Thus, by 1957, groundwater hydrogeochemical exploration was firmly

SUMMARY (Cont.)

established as a modern science. Between 1958 and 1962 hundreds of papers on this subject appeared, both in the East and the West. Again, the search for uranium and base metals predominated, but important advances were made in using groundwater sampling to locate a host of other mineral substances. Notable attempts were made to make the science more rigorous through studies of Eh-pH, solution equilibria and reaction kinetics.

The "current modern period" of the science is considered to be from 1963 to 1978. Many hundreds of papers and reports appeared during these years and those that are reviewed in this work are grouped into eight categories:

(1) Reviews and overviews (2) Methodology, sampling, contamination problems, analysis, data handling, interpretation (3) Hydrogeology, water-rock interaction, pH/Eh, complex ion formation (4) Organic matter, gases, particulate matter, thermal/mineral waters (5) Anions, halogens, sulphate, boron, alkali metals, alkaline earths. (6) Some less commonly sought elements: As, V, Cd, Se, Te, Sb, Sn, Ag, Au, Hg (7) The common base metals, sulphide bodies, polymetallic ores, copper/molybdenum porphyry bodies (8) Uranium and associated elements.

The most important development of this modern period has been the advent of computer-generated, mathematical/physico-chemical "modelling" of groundwater systems and the attempted application of these models in mineral exploration. It is predicted that this trend towards quantification and mass data handling by computers will dominate the next decade of research in groundwater hydrogeochemical exploration.

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The cartoon is by Brant Parker and Johnny Hart /157/.


THEN ALL WE DO IS SURROUND THEM!
I SAY ...WHAT ARE WE FIGHTING THIS WAR FOR ANYWAY?
GOLD, DUKE, GOLD!
FOR A SILLY PIECE OF YELLOW METAL, WE'RE FIGHTING A WAR?
YOU WANT WE SHOULD FIGHT OVER ZINC?

"Now I will discuss that kind of minerals for which it is not necessary to dig, because the force of water carries them out of the veins. Of these there are two kind, minerals - and their fragments - and juices. When there are springs at the outcrop of the veins from which, as I have already said, the abovementioned products are emitted, the miner should consider these first, to see whether there are metals or gems mixed with the sand, or whether the waters discharged are filled with juices. In case metals or gems have settled in the pool of the spring, not only should the sand from it be washed, but also that from the streams which flow from these springs, and even from the river itself into which they again discharge. If the springs discharge water containing some juice, this also should be collected; the further such a stream has flowed from the source, the more it receives plain water and the more diluted does it become, and so much the more deficient in strength. If the stream receives no water of another kind, or scarcely any, not only the rivers, but likewise the lakes which receive these waters, are of the same nature as the springs, and serve the same uses." And further, "The waters of springs taste according to the juice they contain, and they differ greatly in this respect. There are six kinds of these tastes which the worker especially observes and examines; there is the salty kind, which shows that salt may be obtained by evaporation; the nitrous, which indicates soda; the aluminous kind, which indicates alum; the vitrioline, which indicates vitriol; the sulphurous kind, which indicates sulphur; and as for the bituminous juice, out of which bitumen is melted down, the colour itself proclaims it to the worker who is evaporating it."

AGRICOLA, (1546/689/)

"De Re Metallica", BASEL.

1. FOREWORD

Groundwater hydrogeochemical prospecting cannot be viewed as an isolated subject. On the contrary, it can be properly understood only in the context of the development of exploration geochemistry as a whole, and especially of the art of hydrogeochemical prospecting in general. This review will therefore have an historical emphasis and will proceed more or less chronologically from the earliest records to the present literature, beginning with the development of basic exploration principles, proceeding to the emergence of hydrogeochemistry as a branch of the art, and finally narrowing down to a consideration of very specialised modern publications dealing with the detection and location of orebodies through chemical analysis of subterranean waters.

In this review a reference to any publication is enclosed in slashes; thus /49/ means "refer to item number 49 in the reference list". Where it is considered appropriate, the author may be cited, e.g. Thompson /422/, or both the author and the year of publication may be given, e.g. Levinson (1974/51/).

Many figures and tables have been reproduced from the cited references. In most cases the original numbering of these figures and tables has been retained. Any such figures or tables will be found on the page or pages immediately following the first reference made to them in the text of this review.

2. INTRODUCTION

The most important observation emerging from the literature on groundwater hydrogeochemical prospecting is the emphasis on an empirical approach to the subject. There is no one element or group of elements or any set procedure, no *idee fixe*, that has universal applicability. Each area will have its own characteristic geology, physical hydrology, water quality, topography, climate, ore types, problems of sampling and contamination, etc. that will dictate the special formula for a successful approach to a groundwater hydrogeochemical prospecting programme in that area. This formula will almost never be transferable to a different type-area without some (usually substantial) modification. In his recent assessment of the state of the art, Levinson (1974/51 p. 155/ writes :

> "Clearly, the mobility of elements in groundwater cannot always be predicted or explained solely on the basis of pH and Eh, as other natural phenomena are important, particularly on a local level".

And again /51 p. 160/

> "Clearly, the recognition of anomalous metal values in groundwater is not a simple process, but rather one which requires careful attention to all the local and geochemical parameters and, ideally, access to a considerable amount of reliable data".

It is difficult to isolate the origin of the concepts of groundwater hydrogeochemical prospecting because they are so intricately rooted in the fundamentals of geology, soil science, theoretical geochemistry, theoretical and applied exploration geochemistry, hydrology, theoretical chemistry and theoretical and applied analytical chemistry. Thus it is not easy to find basic tenets applying to or procedures used in groundwater prospecting that were developed *in principio* for application to such methods of geochemical exploration. Naturally, it is beyond the scope of this volume to review the history of all the separate fields of knowledge that are mentioned above.* In the following assessment of the development of groundwater prospecting techniques, references to essential parallel advances in these associated disciplines will be made only where they are pertinent.

In the next section we will look briefly at the overall historical advance of groundwater prospecting as a sub-discipline of geochemical exploration in general. In Section 4, which follows on, we will examine some of the key publications on groundwater hydrogeochemical prospecting and pertinent related subjects.

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* Of the abovementioned topics, hydrology is most often *terra incognita* to the geochemist. An excellent introductory account (with bibliography) of many hydrological topics (and indeed of many subjects relevant to groundwater prospecting) is given in The Encyclopedia of Geochemistry and Environmental Science /61/. Three additional useful texts are those by Davis and De Wiest /62/, De Wiest /63/ and Heath and Trainer /64/.

3. A BRIEF HISTORICAL SKETCH OF THE EMERGENCE OF GROUNDWATER PROSPECTING AS A BRANCH OF GEOCHEMICAL EXPLORATION

If one adopts the common practice of including geobotany as a branch of geochemical exploration, then the earliest exponents of this art were apparently the Chinese, who by the 8th or 9th century A.D. had identified and were utilising certain indicator plants associated with deposits of silver, gold, copper and tin /59, 51/.

Although not so ancient, the idea of using underground water as a guide to hidden ore is certainly hundreds of years old. One of the earliest written references to geochemical prospecting, by Agricola (1546*/689/, translated by Hoover and Hoover, 1912/690/, quoted by Boyle /59/ and Levinson /51/), deals with the chemical analysis of spring water. It is clear from this quotation (see page 1 of this review) that Agricola understood the principles of exploration hydrogeochemistry. His approach was not fundamentally different from ours - only the technology has changed.

An even earlier work by Biringuccio (1540**/691/, quoted by Hawkes /53/) deals with the analysis of the dissolved and/or precipitated/suspended fractions of natural fresh waters, but the quotation is not

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* Levinson refers to "Agricola's "De Re Metallica" ..... first published in 1546 in Basel ..... ". Kerr, writing under the title "United Nations" /655, p. 58/, refers to "Georgius Agricola de re metallica: trans. from the first Latin edition of 1556, London ..... ".

** Hawkes describes Biringuccio's text as "probably the first written record of the use of chemical analyses in prospecting ..... ". It is not clear whether the earliest Chinese records are extant.

very specific* about the source of the water.

After this remarkably early start, nothing of note happened in the field of geochemical prospecting generally for more than three hundred years, although there are a few scattered written references to chemical or biogeochemical methods of exploration from this period, especially after 1600 /51/. From the 1890's to 1932 a handful of "modern" papers (for example /53, 79, 154, 358-62, 390, 414/; see also Hawkes /118/) appeared which we can regard as being about, or directly related to, geochemical prospecting /57/. Many of them noted relationships between geology and vegetation and Vysotzkii /362/ used the term "geobotanical methods" as early as 1904.

The impact of these developments was apparently not very great. For example, Fersman /54/, in his classic book reviewing the advance of Soviet geochemistry up to 1934, makes only one passing reference to prospecting, which he terms "applied geochemistry". Moreover, amongst these pioneering works reference to groundwater is conspicuous by its absence. This is not to say that there was no interest in the chemistry of groundwater for other purposes, principally those of agricultural and domestic water supply, and many analyses of groundwaters and other natural waters can be found in the literature prior to 1940 (for example /142-5/). Although very few of these can be considered to be in any

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* The reference is to "certain residues that waters make where they are still, and after having stood for several days, frequently warmed by the rays of the sun ..... ". The remainder of the quotation makes it difficult to decide whether Biringuccio is talking about *in situ* standing surface waters (e.g. lakes) or about the residues left by any natural water that is allowed to stand quietly in the sun after collection.

way related* to prospecting, in some instances, where analytical methods were developed and the geochemistry of mine waters investigated (e.g. /177-80/), an important foundation was thus laid.

Between 1932 and 1939 geochemical exploration emerged as a "recognized" science, first in the U.S.S.R. and shortly thereafter in Scandanavia, Canada and the U.S.A. /49, 51, 53/, so that only five or six years after his perfunctory comment about applied geochemistry Fersman had produced (in Russian) works entitled "Geochemical and Mineralogical Methods of Prospecting for Mineral Deposits" /366/ and "Geochemical and Mineralogical Methods of Prospecting for Useful Minerals" /81/. These were complimented by the first of what might be loosely termed "summaries" or "reviews" of geochemical exploration (e.g. /85, 365, 388-9/). As had been the case with the publications of the first third of the century, much of the work of this period was biogeochemical or geobotanical, with some lithogeochemical and pedogeochemical reports. Once again papers related to hydrogeochemical prospecting generally, and groundwater prospecting specifically, are all but absent.

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* This trend continues today. Although it is difficult to extract exact figures, it is almost certain that less than 1% of modern analyses of groundwater are for purposes of mineral exploration. Today interest in groundwater composition (including fresh interstitial waters) extends to additional fields of study such as pollution /146, 310/, military applications /147/ and theoretical geochemistry and geology /50, 149, 228, 661/. Hydrologists and water supply engineers in particular are frequently concerned with analyses of groundwater to establish potability. Ferris and Sayre (1955/627/) have presented a comprehensive review of the early literature on quantitative analysis of groundwater from the point of view of the water engineer. They give 221 references dating from 1797. The bibliography of Klaer *et al.* (1948/628/) will also prove of interest to the student of the earlier history of this and related subjects.

It is interesting to note /53/ that even at this early stage only a small proportion of the total amount of geochemical prospecting work was ever published.* In addition, very little of the Russian literature was ever translated into English.

What we might call the "Modern Era" of geochemical prospecting commenced about 1939 and rapid expansion occurred after the end of the Second World War (1945). It was during this modern period that hydrogeochemical prospecting first emerged as a distinct sub-discipline, within which groundwater sampling has, to some degree, become a separate branch of the art /51/.

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* This trend, too, continues today. For example, in southern Africa alone there have been hundreds of exploration surveys (mainly pedogeochemical) in the last ten years, involving several million assays, the results of which have never been formally published and very probably never will be.

4. AN EXAMINATION OF SOME IMPORTANT PUBLICATIONS IN THE HISTORICAL AND RECENT DEVELOPMENT OF HYDROGEOCHEMICAL EXPLORATION

In this section the literature of the art of groundwater hydrogeochemical prospecting will be examined chronologically. It is convenient to divide this progress into a few consecutive sections corresponding, more or less, to a small number of benchmark textbooks, reviews or bibliographies of geochemical prospecting literature.

4.1	Antiquity to 1952*			Harbaugh /57/
4.2	1953	-	1954	Erikson /58/, Lang /1118/
4.3	1955	-	1957	Hawkes /53/, Ginzberg /55/
4.4	1958	-	1962	Hawkes and Webb /49/
4.5	1963	-	1978	Hawkes /60/, Levinson /51/

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* A very early prospecting bibliography was compiled by Hawkes in 1948 /175/. This is superseded by Harbaugh's volume /57/.

4.1 PERIOD ANTIQUITY TO 1952

4.11 FOUNDATIONAL WORK UP TO 1940

The mediaeval work of Biringuccio and Agricola has been mentioned. On the subject of hydrogeochemical prospecting, nothing further of note occurred until the second half of the nineteenth century*, when early workers (c. 1880 to 1940) began recording observations, concepts and technical advances that can be seen as the foundation for present day methods of prospecting that make use of aqueous samples. Although it is generally agreed that exploration geochemistry is a "recognized" science dates from between 1932 and 1938 /49, 51, 53/, some of the principle concepts involved were enunciated surprisingly early in the century. Some examples follow of significant publications from this foundational era; these can be conveniently treated under various subheadings, which must not, however, be regarded as mutually exclusive, nor in any important sequence.

(A) The development of analytical methods and the systematic investigation of the composition of natural fresh waters

A *sine qua non* of the progress of interest in water-rock interaction (certain pertinent aspects of which will be described hereafter) was the prior development of suitable analytical technology and an accumulation of a body of empirical knowledge of the composition of natural fresh waters in general. It is apparent that quite sophisticated water analysis was possible well before 1900 /444-446/, but for the most part these pioneering analyses had to be performed by (in many instances very

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* Clearly, these modern developments in exploration techniques and ideas did not appear out of thin air, but were based on multiple general advances in all related sciences. For example, the twentieth century discovery of the effect of cation exchange on geochemical mobility presupposes an understanding of the concepts and mechanisms of ion-exchange, which was first described in 1850 (Thompson /422/, Way /423/).

tedious) wet chemical techniques /118/. These wet chemical methods were constantly updated (e.g. /178/) and remained very important up to and through the nineteen-forties*.

The advent of spectrographic techniques was an important advance. For example, Gerlach and Riedl (1934/664/) published a simple spectrographic method for the determination of uranium in natural waters. (Although the method was reproducible, speedy and economical, it never became popular for the direct determination of U, probably because the detection limit was not low enough for many applications). Spectrographic analysis of water was in routine use by 1935 and proved to be a great boon to the determination of many of the less commonly assayed elements /143/. By 1939 the Soviets had perfected portable spectrographic equipment for field use /118, 413/, but there is no evidence that it was used extensively for water analyses.

Other potentially useful analytical techniques were developed and used during these foundational decades, for example electrometric titration (/449/1927) and conductivity measurements (/450/1925, /451/1932).

Despite the lack of rapid, sophisticated analytical methods, between 1880 and 1940 water chemists had accumulated a sound body of data describing the composition of, *inter alia*, spring water (e.g. /443-4, 446, 448, 452, 518-9/), river and lake waters /143-5, 404, 454-7, 747/, swamp waters /460/, rainwater /458-9, 488/, mine waters /443, 447, 524/, groundwaters /442, 462-70, 643-4/, including oilfield waters (Engler and Hofer 1909/461/, Renick 1924/684/, Reistle 1927/290/ and others /478-9/) and various unusual waters such as spring waters and groundwaters rich in dissolved organic matter (Pischel 1912/660/).

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* and are of course still used today for a great many water analyses all over the world.

Many of the principles of geohydrology evolved as these chemical studies of groundwaters were complimented by early hydrological investigations of various kinds (e.g. Slichter 1899/309/ and others /307-8, 310, 471-3/).

In response to the inevitable needs generated by this growing bulk of hydrogeochemical data, publications began to appear on the practical aspects of the examination of natural waters (Leighton 1905/474/ and others /475-6/), including problems arising from contamination of samples (Collins and Riffenburg 1923/477/), and on the practical interpretation and graphical presentation of water assays (Tickell 1921/289/, Reistle 1927/290/ and others /478-9/).

Amongst these earliest publications we can identify several that can reasonably be classified as "applied geochemistry" in that the analysis of waters was directed towards the attainment of some immediate practical goal, usually agricultural (e.g. Kelley and Brown 1928/481/ and others /480, 483/) or related to industrial applications or engineering problems such as fouling of boilers /457, 482/, but very few can be viewed as direct contributions to "mineral exploration" *per se*.

(B) General principles of water-mineral/rock reactions and the initial alteration of minerals and ores in the secondary environment

That groundwaters react chemically with rocks has been clearly understood since at least the time of Agricola. Around the turn of the century geochemists turned their serious attention to a closer investigation of the nature of this interaction (e.g. /424-441, 521, 526/, 1902 to 1940). Their researches consisted, on the one hand, of quite specific studies of the reaction between water as a chemical reagent and given minerals, compounds and elements, and on the other, of more general observations of the interaction between rocks or groups of rocks and the natural waters passing through or over them. With regard to the firstmentioned researches, early examples of studies of the reaction between mineral substances and water are the works of (i) Weigel (1906-7/439-40/), who discussed the behaviour of sulphides of heavy metals in aqueous solution, (ii) Kohlrausch (1908/432/), on the properties of saturated aqueous solutions of slightly soluble salts and (iii) Lenher and Merril (1917/434/), on the solubility of silica in water.

Of parallel importance to the development of basic hydrogeochemical concepts, and also dating back to the first years of the century, are pioneering studies of the processes controlling the oxidation of sulphide minerals exposed to the influences of water and the atmosphere (e.g. Gottschalk and Buehler 1912/152/ and Emmons 1913/447/).

Concerning the second category - the study, in more general terms, of the interactions of rocks and waters as co-existing natural materials within the crust of the earth - it is worth noting that analyses of mine waters were first reported at least a hundred years ago. Pattinson (1879, cited by Watson 1918/1051/) analysed spring water emerging from an old drift that had been made in searching for a lead ore near Newcastle-on-Tyne and noted that this water precipitated "black muds" containing nearly 70% Mn oxides.

Darton /442/ wrote in 1906 that "all waters which flow over or through rocks and soil dissolve various chemical compounds, the nature and amount of which have great local variation. Water passing through pure sand or sandstone can be contaminated only to a very slight degree, but nearly all rocks contain soluble minerals, especially shales, limestones, and red beds, which often yield a large amount of salts to waters which come in contact with them".

Sullivan (1907/151/) discussed how the reaction between minerals and natural waters affects geological phenomena, while Palmer (1911/441/) wrote extensively on the geological interpretation of water composition. In his description of the artesian waters of Australia, Pittman (1908-1915/1008-1010/) showed that he had clearly grasped the basic principles of water-rock reaction and the factors that control the chemical composition of shallow groundwaters. (It is equally clear that some of his opponents, e.g. Cregory (1906/1011/) still held some very weird views on this subject - for instance, that the artesian waters of Australia, because of their chemical composition, had to be of "plutonic" origin). Pittman understood the control that lithology exercises over water chemistry and was aware of the phenomenon of the "metamorphism" of water as it migrates laterally from the point of recharge and reacts continuously with the country rocks. He also understood the problems of sample contamination and records an instance where well waters were contaminated by zinc from a galvanized casing pipe (/1009/1914).

In 1913 Emmons and Harrington /443/ produced a remarkable comparison of analyses of waters of mines and of hot* springs "with a view of ascertaining the composition of waters that have been assumed to be the agents of deposition of sulphide lode ores". The significant corrolary that is now, with hindsight, so obvious to us, appears to have escaped the authors.

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* The origin, chemistry and significance in prospecting of hot (thermal), as opposed to cold, springs is generally very different /49, 286, 525/. The geochemistry of thermal waters will not be considered in detail in this review. Readers interested in the earlier literature of the chemistry of thermal waters and associated deposits might consult White (1955/626/) who has reviewed this subject at length and has compiled a list of 149 references dating from 1847.

Nevertheless, much of the content of this paper has a very familiar ring to the modern exploration geochemist. The authors present a review of the relatively few analyses of mine waters and spring waters available at that time (e.g. /444-6/, 1884-1888 and /447-8/ 1912-1913). There are analyses of some thirty-odd aqueous species including As, Sb, Mn, Co, Cd, Ni, Pb, Cu and Zn, many at sub-ppm levels of concentration. Mine waters and various spring waters are shown to contain similar elements but in very different amounts. Chloride, sulphate and carbonate predominated in mine waters and the heavy metal content was markedly greater than for waters of hot springs. The authors assert clearly that sulphides are the obvious source of the sulphate ion, but the implication is not pursued to its logical conclusion. The paper includes a long discussion of the ability of various "subterranean water types" (for example alkali-chloride waters, alkali-sulphate waters) to dissolve, transport and deposit various metals. The probable importance of mobile complexes such as $KClAuCl_3$ is emphasized, as are other natural interactions affecting mobility, for example, the influence of pCO_2 on the solubility of Pb and the effect of [As] and [Sb] on the solubility of copper. The causes of precipitation of metals from groundwaters are said to include "reactions with wall rock, decrease of temperature, decrease of pressure, dilution and oxidation".

Another significant early account of this type is that of Watson (1918/1051/). He analysed water that was emerging from a fissure in the City and Suburban gold mine (Johannesburg) to see if the water was potable. The water contained appreciable amounts of Mn (40 mg/l) and Li (2 mg/l). The fissure was resampled seven weeks later and little change in composition was evident. There is no indication that the author sought to use the data for prospecting purposes.

Somewhat later than in the West, Russian speaking authors began to show an increasing interest in the chemistry of water-rock reaction, on both the global (i.e. historical) as well as the local scale. Vernadski, for instance, wrote about the essential characteristics of the reactions that occur between solid substances (rock) and (natural) aqueous solutions and emphasized the severe time-dependancy of many of the slower processes (1927/646/). Later he wrote extensively on the history of natural waters as components of the lithosphere (1933-34/180/), while other Soviet writers described the factors that characterize the chemistry of groundwaters associated with specific lithological formations (Smurov 1938/179/) and the composition and geochemistry of mine waters in sulphidic ores (Khitarov and Mulikovskaia 1935/177/).

(C) The development of key geochemical concepts of importance to exploration

Even before the Great War studies had been undertaken of the processes of aqueous dispersion and the factors influencing the migration, mobilization and immobilization of various elements in the secondary environment /83, 85, 142, 391, 394-5/. The geochemical role of colloidal substances in natural solutions was amongst the first fields of interest, probably because of the link with agricultural science (e.g. Gedroiz 1912/484/ and others /485, 510/). Smyth (1913/391) conducted early investigations of the mobility of the chemical constituents of rocks.

It was noted that the concentration and mobility of aqueous metals could be profoundly affected by the presence of living organisms or their remains (Lovering 1927/396/ and others /402-4/. Thus Freise (1930-1/778-9/) and Fetzer (1934/777/) showed that gold could be dissolved and transported by humic complexes ("soil acids") and redeposited in significant concentrations in certain favourable locations such as placers.

It was understood that mobility is also influenced by ion exchange reactions with natural materials (e.g. Renick 1925/467/, Noll 1931/397/, Vanselow 1932/485/ and Schoeller 1935/472/), or by general, non-differentiated sorption reactions with sedimentary rocks (Arstinoff 1914/743/, Vernadsky 1935/742/ and others /744-5/).

In 1925 Doerner and Hoskins /600/ demonstrated that radium may be co-precipitated with barium sulphate during the reaction

$$Ba^{++} + SO_4^{=} \rightarrow BaSO_4$$

even when the solubility product of no radium compound is exceeded. This scavenging (sorption-coprecipitation) of many species of metallic ions from solution by the precipitation of certain other compounds (notably hydroxides of Fe and Mn) was reported in detail by Goldschmidt and co-workers in 1934 /394-5/.

Murata (1939/149/) was one of the earlier writers who emphasized the important relationship between environmental oxidation potential and the mobility of polyvalent elements.

(D) Early relevant prospecting experiments and applications

A variety of early geochemical experiments can be considered to be the recent ancestors of modern groundwater hydrogeochemical prospecting methods. Some of these experiments, although highly significant, did not directly involve any water sampling, and we will briefly examine a few of the more important "indirect" contributions before proceeding to a consideration of the first true hydrogeochemical studies.

Raeburn and Milner (1927/80/), in their work on the exploration of placer deposits, implicitly anticipated many of the principles of stream hydrogeochemical prospecting, such as dilution of anomalies with barren material and the variable mobility of different elements in river channels.

Flerov, during his work on the search for tin in the U.S.S.R. (1935/82/), developed, and stressed the need for, rational and systematic sampling patterns and techniques.

A noteworthy concept developed by the Soviets in the mid-thirties was the possibility of the practical exploitation of the relationship between economic mineral deposits, especially oxidizing sulphide bodies, and dispersed anions. However, the anions were sought in the soil and usually by now-obsolete electrometric methods using special electrodes inserted into the ground - essentially a geophysical method, which had its heyday between 1936 and 1941 /363-5/. For example, in 1936 Safronov and Sergeev /84/ used this kind of approach to relate soil chloride and sulphate to hidden sulphide bodies. In the same year Sofronov /85/ reported the use of a similar "physico-chemical method of prospecting" to detect "salt aureoles of dissemination". In 1937 Sergeyev* and Solovov /87/ described a similar soil-electrode potentiometric method for investigating the "sulphate and sulphuric acid concentrations in soil moisture as a means of prospecting for oxidizing sulphide deposits".

Interest was not restricted to sulphate and chloride only; for example in 1936 Sergeyev /88/ described colorimetric tests for borate ions, with a view to locating borate deposits in the U.S.S.R.

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* Translated variously as Sergeyev, Ye.A. or Sergeev, E.A. Some Russian names can be found in more than one English form because of the incomplete correspondence between the letters of the Cyrillic and Latin alphabets.

It is probable that we have not yet heard the last word about the identity of the "first true twentieth century hydrogeochemical prospector" /1122/. There can be little doubt that pioneering investigations of the radioactivity of spring waters and associated precipitates and deposits in the U.S.A., Sweden, Japan (Boltwood 1905/619/, Sjögren and Sahlbom 1907/507/, Sahlbom 1915/508/, Ohashi 1920/614/, Nakai 1940/509/) and elsewhere /601-2, 615, 630/ portended later methods of uranium prospecting. Were these papers to appear today, we should probably not hesitate to describe some of them as "orientation studies". But they were written before this phrase was coined, and one could legitimately argue that the authors of these works do not, in so many words, suggest that their methods be employed to locate exploitable concentrations of uranium ore. This possibility is nevertheless clearly implied in at least some of these earliest works, as the following quotations will show: "It can be anticipated that waters which rise through strata containing appreciable quantities of uranium minerals will be found more highly radio-active than any which have thus far been described. The results obtained from the examination of waters from springs in well-known uranium localities can be looked forward to with interest" (Boltwood 1904/1123/).

"The foregoing argument of course does not exclude the possibility of the underground flow touching very active substances but the presence of such material can not be inferred from the existence of a highly radioactive water or gas without other evidence. If a spring happened to be so situated that its water came in contact with a material which could be classified as even a low grade radioactive ore, and further if it had the large and rapid flow characteristic of most of the springs examined in this work, it seems fairly certain that it would show an activity of a different order of magnitude from those recorded in the tables above" (Lester 1915/1124/).

To the writer's knowledge, the oldest "modern" publication that is indisputably concerned with hydrogeochemical exploration was written by Henniger in 1925 /1125/. This neglected paper is highly significant because, as the author has pointed out elsewhere /1122/, it makes "modern" geochemical prospecting ten years older than any historians of the subject had hitherto suspected. It is worth quoting at length from Henniger: "For several years the Gulf Production Company has been collecting and analyzing samples of waters from the known salt domes throughout the Coastal Belt of Texas and Louisiana. The object of the work was to determine whether or not waters with high mineral content were of local occurrence over the salt cores of domes, and to compare the results obtained with shallow waters from other localities which appear to be highly mineralized. Approximately one thousand waters have been analyzed in this work ..... The analyses show that the waters of the known domes vary widely in their sulphur content ..... In the case of Barber's Hill ..... an average of all the analyses gives a higher concentration over the salt core than the analyses of the waters taken off the salt ..... By a process of averaging, a small area of high concentration can be detected over the salt both at Hockley and Damon Mound ..... In the investigation of waters from localities other than salt domes, several small areas were found which are characterized by an abnormally high percentage of both sulphate and sulphuretted hydrogen. In some instances, shallow waters are as highly impregnated with these constituents as any dome waters that have come to our attention ..... Actual drilling of these prospects has given no evidence that the sulphur content of these waters is due to a buried dome ..... it is quite probable that the sulphur water in a number of localities more or less remote from known salt domes has the same source as the deep waters directly associated with a buried salt core ..... Actual drilling has practically condemned several of the most promising areas, emphasizing the fact that new salt domes are not readily located on the evidence of sulphur water alone .....".

Only a handful of additional studies of the use of water sampling in geochemical prospecting took place before 1941. These include the earliest "modern" record of a true surface hydrogeochemical survey, by Vogt, in 1939 /86/. He showed that stream waters draining certain Norwegian sulphide deposits contained easily detectable amounts of sulphate ion and suggested that this fact be utilized for exploration purposes. A paper by Feigelson in 1940 /89/ deals with the analysis of surface and underground brines in the search for borate deposits. He also makes the important observation that the ratio of components of these waters (in the case H_3BO_3/Br) may be a more useful indicator than absolute concentrations.

The year 1940 saw also a remarkably advanced groundwater study by Miholić in northwestern Croatia (Yugoslavia), but the results were not published until 1950 /108/. He used heavy metal analysis of mineral and thermal waters as a regional reconnaissance prospecting guide, to single out regions favourable for further study. Apparently this method enjoyed some success in the search for copper deposits.

4.12 THE INFANT SCIENCE : 1941 TO 1948

It can scarcely be said that the work of Henniger, Micholic, etc. took the world of mineral exploration by storm and, to be seen in perspective, these earliest descriptions of applied studies in groundwater hydrogeochemical exploration must be viewed as isolated examples. The same appears to be true for the interval 1941-1948. Reports concerned with groundwater hydrogeochemical exploration were still a rarity during this period and few of them can be considered to have constituted an important advance in the science. A possible exception is the work of Tageeva (1942/181/), who recorded the results of his enquiry into the connection between petroleum* deposits and the fluorine and boron contents of natural waters. As had been the case in the thirties, most of the historically important developments were made in various peripheral areas of study rather than as a result of case histories involving the sampling of groundwaters. For purposes of review this progress can conveniently be considered within a few loose groups of topics:

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* A substantial literature has evolved around this subject /51, 78, 118, 155, 386-7/, which includes (usually deep-well) groundwater sampling as an aid to the location of deposits of petroleum or gas. These techniques have little bearing on the search for relatively shallowly buried mineral ores (cf. the work of Ignatovich /645/, subsection (A) following), and the historical growth of the literature of petroleum exploration geochemistry will not be followed in great detail here.

(A) Supplementary studies of subterranean waters, pertinent to the understanding of the relationship between the chemistry of such waters and rocks and ores

These studies (e.g. /50, 182, 368-9, 653/), which can be compared to those of earlier workers such as Khitarov and Mulikovskaia /177/ and Emmons and Harrington /443/, might, loosely speaking, be termed "non--case histories", and were mainly concerned with geohydrology, water/rock reactions or the geochemistry of mine waters.

Jarrell /50/ published a remarkable paper recording a study of oxidation processes in an oxide/sulphide copper mine at Chuquicamata in Chile. He attempts to relate these processes to the properties of the ore minerals and to hydrological factors. He lists several groundwater analyses for sulphate, chloride, nitrate, silicate, Na, K, Mg, Ca, Fe, Cu, Mn, Mo and free acid. He discusses these results at length and clearly understood that the chemical composition of the groundwater was a reflection of oxidization processes in the adjacent ore. What is remarkable is that he gives no indication that he was aware of the potential application of his findings.

Mulikovskaia (1947/182/) conducted a similar study of the geochemical characteristics of mine waters in sulphide ore bodies but his conclusions are no more penetrating than those of Jarrell.

In an important paper in 1944, Ignatovich /645/ recognized that the distribution of dissolved components (including salts and gases), organic matter and micro-organisms in groundwaters are distinctively and regularly zoned within the vertical cross-section through the first five kilometers of the lithosphere. The characteristics of the shallower groundwaters with which the prospector will normally be dealing are very different from waters occurring or originating in the deeper crust.

(B) Investigations of natural phenomena which affect the concentration and movement of the chemical elements in water

During this period the theoretical and empirical understanding of the controls of secondary dispersion and mobility became quite sophisticated (e.g. /371-3, 392-3/).

One crucial development of the early forties (which followed logically from pioneering chemical sedimentological work /149/ and classic studies by geochemists such as Goldschmidt and others /394-5/ in the thirties) was the full recognition of the importance, to hydrogeochemical prospecting, of co-precipitation and sorption effects in natural aquatic systems /104-5, 148, 370, 399, 400/. Thus Vogt 1942/104/ showed that Zn, Cu, Ni, Co and Ca are co-precipitated with manganese hydroxides in bogs and lakes. He suggested that these precipitates might be used as a prospecting medium.

It was also clearly understood that the formation of the initial precipitates such as manganese hydroxide was in turn very dependent on ambient pH. In 1942 Britton (Table 1) published data for the pH of hydrolysis of many metals:

TABLE 1.

pH OF PRECIPITATION OF HYDROXIDE OR BASIC SALT FROM DILUTE SOLUTIONS

(BRITTON 1942/415/)

Silver	7.5 - 8.0	Ferrous	5.5
Zinc	7.0	Cupric	5.3
Cobalt	6.8	Aluminium	4.1
Nickel	6.7	Stannous	2
Lead	6.0	Ferric	2

In addition to the complex phenomena of precipitation/co-precipitation, principle areas of relevant research were the role of redox potential in the accumulation and dispersion of the elements (e.g. Kryukov and Levchenko 1947-8/650-2/ and others /367, 411/), the effect of ion exchange between dissolved elements and organic and colloidal substances /371, 401, 523/ and the general, complex interactions that may occur between organisms or organic matter and trace elements /405-10, 530/. Volkova (1946/649/), for instance, showed that the microflora of (and other biological processes in) certain mineral springs had an important role in the conditioning of these waters.

Some notable empirical studies of mobility in surface waters were reported. Thus Sergeev (1946/347/) discussed the role of pH in the control of metal concentrations in stream waters and showed that the mobility of Cu in some mineralized areas was greatly enhanced by a relatively small decrease in pH.

In 1947 Leach /93/ reported on the relative persistence of Cu, Zn and Fe at sub-ppm levels up to 32 miles downstream from the source of the metals. He confirmed that Cu is very immobile at pH values above 5,5.

In an interesting study in 1948, Bryant and Slater /522/ noted that the soluble salt content of runoff waters was highest just after the first rains of the season, as accumulated salts were swept away, and that the salt concentration decreased again with subsequent additional runoff.

(C) Analytical techniques

Important advances were achieved between 1941 and 1948. Spectrographic techniques were the order of the day, at least in the early forties

/118/, because they offered the advantages of speed and simultaneous multi-element capability. Of the additional methods of analysis that appeared before 1948* (e.g. /118, 123, 416/) the one that offered to be of most use in hydrogeochemical prospecting was polarography /364, 417/, but in fact it never developed into an important tool for this purpose /51/.

Despite these advances, interest in hydrogeochemical methods was still largely inhibited by the lack of simple, sensitive analytical procedures /49, 53, 55, 91/. Therefore by far the most important breakthrough of the forties was the application of colorimetric analysis** to prospecting and most especially to the examination of waters. This innovation was heralded by Huff's paper (1948/91/) describing the use of

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* XRF analysis for prospecting purposes also began in the 1940's (e.g. Vogt and Bugge 1943/418/). Although this technique has become of immense importance in geochemical analysis, it has never found large scale application in the examination of natural waters.

** The use of colorimetric methods was a logical sequel to the spot tests /364, 419/ that had been used by Soviet exploration geochemists since 1941, but it was in the U.S.A. that colorimetric analysis of exploration samples was first perfected and applied on a large scale in 1948-1949, generally for the examination of solid material /412, 420-1/.

dithizone* for the colorimetric detection of many heavy metals in water. Almost immediately modifications of or improvements to Huff's dithizone tests, and new colorimetric methods, began to appear /90, 109-11, 120, 150/.

(D) Surface hydrogeochemical prospecting

It is certainly true that for most of the forties general advances in applied hydrogeochemical prospecting resulted principally from studies of surface waters (e.g. /90, 96-7, 183, 347/). During the period 1941-1948 the number of publications on or with strong affinities to surface hydrogeochemical prospecting averaged about two per annum.

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* The dithizone (diphenylthiocarbazone) analytical method was first popularized by Sandell /123/ in 1944. Huff's paper is generally recognized as the first English language paper to record the use of dithizone for hydrogeochemical prospecting purposes. It is possible that Sergeev used colorimetry as early as 1946 /53, 96/ but his analytical method is not stated and his paper was, in any case, not available in English until 1950. Few people are aware that Sugawara *et al.* /124/ also published a paper in 1948 recording the use of a modified version of Sandell's method for studying the "natural distribution of Cu and Zn". This paper is in Japanese and it is not clear whether water was analyzed or if the Japanese workers and Huff were aware of each other's work. In this regard it is interesting to note the comment made by Murakami *et al.* /640 p. 131/ in their synopsis of the origins (1943 onward) of Japanese 'chemical prospecting':"It is to be noticed that chemical prospecting was developed in Japan independently of the activities of other countries and the use of diphenylthiocarbazone (dithizone) as the most useful reagent was a strange and unexpected coincidence".

For instance, in 1941, Konovalov /92/ determined Au in stream waters and demonstrated that the concentration varied with time at a given locality. Also in 1941, Rosenqvist and Vogt /95/ applied newly-developed trace-analytical methods for copper to stream waters at Roros in Norway. They studied the concentration of Cu in sixty-nine streams and selected six with copper contents from 0,05 to 0,185 mg/l as anomalous. One new orebody was discovered in the follow-up work.

Lundberg (1941/94/) recorded the first laboratory experiments to attempt to relate the solubility of natural economically important minerals (placer gold and cassiterite) to the concentration of elements (Au and Sn) in stream waters.

In a now-famous study in 1946, Sergeyev /347/ demonstrated that the decay of heavy metal anomalies in streams is a function (*inter alia*) of dilution from tributaries, and that sudden, massive influxes of water from large, confluent rivers can rapidly dilute the anomaly to background levels.

A study by Huff (reported in /53 . p. 310/) in 1947 at Butte, Montana, showed that the Cu/Zn ratio in stream waters could be used to distinguish between decreases in [Cu] due to precipitation as opposed to dilution. Severe loss of copper occurred when the pH of acid mine waters, draining away via nearby streams, rose above 5,0. In 1948 Huff /91/ presented a paper (referred to briefly in the preceding subsection on Analytical methods) that has become one of the classics of exploration geochemistry. He records dithizone analyses of spring, mine and groundwaters as well as stream waters in the U.S.A. during 1947, but provides no detailed description of the use, for prospecting purposes, of waters other than stream waters. He also makes a clear statement of the concept of the relative secondary mobility of various heavy metals in natural water.

It is difficult for the English speaking reader to establish exactly what the Japanese were doing in this field during this period, but it is clear that they were very active. The analysis of stream waters by means of carbon tetrachloride solutions of dithizone became a very popular method of exploration /640/. In typical cases most of the metal present was zinc, at background values of 10^{-9} or 10^{-10} g/l. Exposed or blind orebodies would, under favourable circumstances, cause the metal value to rise to 10^{-8} g/l, but this anomaly would be diluted to background values once again within 30 to 40 m downstream. "Chemical prospecting by this method was carried out for more than one hundred sulfide ore mines from 1946 to 1957" /640 p. 132, 710/.

Huff's paper and the work of the Japanese show that by 1948 stream water prospecting was a mature art and that most of the principles, if not all the details, of hydrogeochemical prospecting in general were well known. This increasing sophistication was reflected also in newer procedures for data handling, etc. For example Sergeev (1946/347/) described detailed sampling patterns for stream waters. A number of writers /91, 102, 486-7, 491-3/ from Langelier and Ludwig (1942/489/) to Larson (1949/490/) contributed to the development of graphical and cartographical methods for the presentation and interpretation of hydrogeochemical data.

(E) Reviews

An indication of the coming of age of applied hydrogeochemistry was the appearance in the late forties of several review papers that comment on the advance of exploration methods using natural waters /98-103/. They deal principally with surface waters; groundwater prospecting, as we have seen, had still not yet made much progress. The most important of these reviews is the one by Hawkes /99/ in 1949. He makes a clear distinction between groundwater and surface water prospecting, but can list only a few examples of previous groundwater prospecting investigations - viz. Huff's work /91/ and the following unpublished surveys: one by the U.S.G.S. of Zn in springs in south-western Wisconsin and well water in New York State, and the other of Cu in drill-hole water near a copper deposit in Arizona (this last survey later published by Lovering *et al.* /106/). Hawkes concludes, in summary, that Zn has good potential for application in groundwater prospecting, Cu is promising, Pb gives negative response and no information* is available for Sn, W, Mo, Ni and Cr.

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* The earlier Soviet work described in the foregoing was presumably not available to Hawkes at that time.

4.13 EXPANSION : 1949 TO 1952

> "Geochemical methods of prospecting are based on the premise that a chemical pattern exists in relatively accessible natural material that can be used as a guide in locating relatively inaccessible deposits of valuable minerals. Such chemical patterns are most commonly the result of the dispersion of elements and compounds from the site of primary deposition of the ore, and they assume shapes controlled by the characteristics of the agents of dispersion and the structure and composition of the material through which the dispersion takes place.
>
> .... In spite of the apparent complexity of the problem, considerable progress has been made in the past 15 years both in understanding the basic principles and in applying chemical techniques to practical exploration problems".

Hawkes, 1950/118 pp.537-8/.

After 1948 groundwater geochemical exploration, although very young, could reasonably if arbitrarily be regarded as a distinct sub-discipline of hydrogeochemical prospecting, with an embryonic literature separate from that of surface water sampling methods. The rate of publication of papers describing groundwater sampling projects increased sharply.

4.131 THE EAST

Thus the year 1949 marks the beginning of a series of papers on groundwater prospecting studies by workers in Japan /107, 112-116, 122/. Most of this literature is in Japanese and it is difficult to say how much cross-communication there was between East and West on this topic at that time. Yokoyama (1949/107/) found a relationship between Mn deposits at Awano and the Mn content of local underground and surface waters and suggested that this fact be employed in prospecting. Hayashi *et al.* (1951/114) determined pH, Zn, Cu and sulphate in ground- and surface waters from various areas; on the basis of these measurements they regarded some regions as having more promise than others for further exploration. Kimura *et al.* (1951/115/) determined Cu and Zn in ground- and surface waters near the Takara mine. They concluded that Zn is more mobile than Cu, but that copper was a better indicator of nearly mineralisation.

Ohashi (1951/116/) showed that the composition of waters of the Ogoya mine were highly variable (sulphate from 56 to 2280 mg/l, Cu 0,2-123 mg/l, Zn 1,1-339 mg/l). He also showed that the release of these substances was greater near the oxidized upper levels than lower down the ore vein.

Kimura *et al.* (1951/113/), in what was probably the first study of its type, demonstrated a correlation between the iron, sulphate and Zn contents of waters from subsurface fractures and the occurrence of ore above underground workings in the Hosokra Mine district of Japan. Apparently a new orebody was discovered by this approach /53/.

An interesting investigation of the lithium and caesium contents of natural waters and hot springs was conducted by Yamagata (1951/639/), "for the purpose of finding new sources of these elements" /640 p. 131/.

Soviet contributions* of importance during the early fifties are a detailed discussion by Valyashko and Korenenko (1950/117/) of the significance, in exploration, of concentration ratios in natural waters, and an attempt by Makarenko (1950/184/) to correlate the compositions of regional groundwaters with those of local stream waters.

4.132 THE WEST

In the West, as in the U.S.S.R., a small number of important papers appeared during this period. In 1950 Hawkes /118/, in an expansion of his 1949 summary of geochemical exploration /99/, discussed many modern theoretical concepts related to dispersion and mobility in groundwater, e.g. the effects of hydrological factors, pH, Eh, hydrolysis, complex formation and ion exchange in water/trace-metal systems, and trace element scavenging by hydroxides of iron and manganese. He discusses special problems of groundwater sampling, such as dearth of sampling points, restricted release of metals from orebodies below the water table and problems of contamination of wells by metal fittings. (He notes that springs are generally free of contamination). Hawkes also gives a summary of the analytical methods available at that time (colorimetric methods still being the most important) and summarizes the potential application, in hydrogeochemical exploration, of various chemical species as follows: Zn - most promising, sulphate - promising, Cu - generally not useful but may be in special cases.

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* Akhmedsafin (1951/378/) wrote about the "prospecting signs of underground water in sandy deserts". The paper is unfortunately unobtainable and its significance remains unknown. He may well have meant prospecting for water rather than using water for prospecting.

Also in 1950, Webb and Millman /119, 137/ used dithizone to detect anomalous concentrations of heavy metals in springs issuing from ore lodes in the lead-zinc belt of Nigeria. These waters contained up to 0,5 mg/l heavy metals and the authors clearly recognized the distinction between the dissolved and suspended metal fractions of the samples. The results were viewed by the writers as promising.

In 1951 Huff studied the Pb content of acid springs in the Judith mountain area of Montana, but apparently did not publish his results /53/. Considerable concentrations (up to 1%) of lead were precipitated near these springs. In the same year, Warren *et al.* /150/ reported the analysis of spring water in the Pacific northwest of the U.S.A. They regarded 5 μg/l heavy metals as promising.

During a project similar to that pioneered by Kimura *et al.* /113/, Lovering (1952/511/) demonstrated a definite relationship between the metal content of descending mine waters and both the degree of mineralization (ore grade) of the channels of migration and the distance thus travelled.

In contrast to all this optimism, Lovering *et al.* (1950/106/) gave an example of a case history (Pinal County, Arizona) where groundwater prospecting did not work.* It was found that the waters from boreholes in copper deposits there had a pH of about 9 and that hydrolysis had lowered the copper concentration to undetectable values.

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* This negative result is significant in view of the fairly widespread scepticism about wellwater hydrogeochemical prospecting that was to develop in the following few years. Thus Hawkes /53/ stated in 1957 that "results of experimental wellwater sampling have to date been uniformly discouraging .....".

In an important paper in 1952, Morris and Lovering /121/ studied the mobility of metals in groundwater as part of their investigation of dispersion of heavy metals in wall rocks near ore bodies in the Tintic District of Utah. They applied data and concepts from theoretical chemistry to predict the mobility of several metals in groundwater in contact with carbonate and other country rocks. In carbonate rocks predicted mobilities are as follows:

Zn - fairly immobile
Ag, Cu - very immobile
Au, Pb - exceedingly immobile.

This is in excellent agreement with their field observations and those of previous workers. Morris and Lovering also recorded theoretical aqueous pH values at which various metals normally hydrolyse and precipitate. These pH values may be modified by precipitation of insoluble species (e.g. Ag^+ in presence of Cl^-) or by complex formation. In natural fresh waters of pH >6,5 the following elements will not normally be stable in solution: Fe(III,II), Cu(II,I), Cr, U(IV), Al, Hg(I), Th, Sn(II), Zn, Ag and Mn(II). At pH >6,8 Pb, Cd, Ni and Co are likewise unstable.

It is noteworthy that these observations on pH of hydrolysis partly contradict the earlier data of Britton (1942/415/). This is an early example of the difficulties inherent in attempts to quantify natural hydrogeochemical phenomena by means of laboratory experiments, the results of which are generally only a function of how the experimenter defines and attempts to duplicate a "natural fresh water system".

The rest of the world, at this time, still appeared to be taking very little interest in topics related to groundwater prospecting. In an isolated contribution in 1952, Wilska /153/ reported in detail on the composition of Finnish ground and mine waters. A few publications from various countries dealt with phenomena related to radio-elements in groundwater /603-4, 607, 620-2/. The radioactivity of spring waters, for example, was linked to water temperature (Rosenblatt and Lindeman, Israel, 1952-603/) or to the influence of the lithology and geological age of the source rocks (Miholic, Yugoslavia, 1952/604/). Hurley (1950/617/) demonstrated that much (90%) of the uranium and thorium in samples of various igneous rocks was readily elutriated by dilute acids and that Ca, Fe, Al, Si, P, K, and some Na, Mg and Ti were initially mobilized with the uranium. Gruner (1952/736/) conducted early experiments on the synthesis of secondary uranium minerals and showed that uranyl ions in natural solution are readily reduced by Fe^{++} to yield Fe_2O_3 and insoluble U(IV) compounds.

Noteworthy publications of the early fifties on peripheral issues covered subjects such as sampling statistics /374/, advances in analytical techniques for natural waters /375, 711/, the physical chemistry of precipitation/sorption phenomena and the elucidation of the molecular and ionic forms in which elutriated metal atoms migrate away from the oxidized zone of ore deposits /376, 379-81/.

In summary, one might say that by 1952 groundwater exploration was firmly established, but that it had as yet no well developed tradition. This is evident from the fact that although some contemporary reviews (e.g. /118/ of geochemical exploration are quite informative about groundwater prospecting techniques, there were still published at that time standard texts and field manuals that had almost nothing to say on the subject (e.g. 120, 377, 382/).

4.2 PERIOD 1953 - 1954

The choice of the year 1953 as an historical division is to a degree arbitrary, but it does more or less close the period during which the modern foundations of groundwater hydrogeochemical prospecting had been laid. Subsequently, papers making some fundamental new theoretical contribution to the subject became less numerous than those describing practical studies and case histories.

Webb's review of American geochemical exploration (1953/382/) creates a succinct picture of the state of hydrogeochemistry in general at that time: Stream water studies predominated; problems included the vagaries of metal concentrations at the hands of influences such as pH, colloids, clays, organic matter and bacteria; aqueous metal concentrations were often found to be very low, so that analytical methods were frequently tedious and not always sufficiently reliable (for example in dithizone tests results of ± 100% precision were often regarded as adequate); wet chemical, colorimetric, distillation extraction, fluorimetric, turbidometric, optical spectrographic and spot test methods held sway (Atomic Absorption was unheard of); the U.S.G.S. was constructing a mobile spectrographic laboratory for field use and newer analytical techniques were being rapidly developed; the total volume of exploration work being performed was difficult to assess, because of non-publication of privately-held information.

4.21 URANIUM

Against this background, a moderate expansion continued, especially in methods of hydrogeochemical prospecting for radioactive elements (e.g. /515/). Arndt and Kuroda (1953/138/) reported the first (indisputable) radiochemical prospecting survey of surface and groundwaters (in Arkansas).

They concluded that the detection of radon in spring water and groundwater is potentially applicable to the location of low-grade uranium deposits. Several other workers showed interest in radio-elements in groundwaters, spring waters and spring deposits /605-6, 616, 623-5/. For instance, Staatz and Bauer (1953/605/) showed that some spring waters (in Nevada) that are relatively highly radioactive contain but little uranium and that the radio-source, presumably radon, is rapidly lost from standing samples. Similarly, and in the same year, Gott and Hill /670/ noted that not all radioactive waters necessarily indicate the presence of local uranium occurrences. Small amounts of highly radioactive radium may be present even when uranium is all but absent. Uranium may never have been present in the water, or it may have been lost. The authors give an example (oilfields of southwestern Kansas) of uranium being precipitated from solution as a result of the mixing of groundwaters of different compositions.

An unusual relationship between radioactivity and the Mn content of hot springs in the San Juan Mountains of Colorado was demonstrated by Burbank and Pierson /606/ in 1953. On the experimental side, Larsen and Phair /631/ reported that "commonly close to 40 per cent of the uranium in most fresh-appearing igneous rocks is readily leachable". (Hurley's earlier estimates /617/ had been as high as 90%).

Isotopic* studies /577, 665/ of uranium minerals undergoing acidic oxidizing attack suggested that uranium will be preferentially leached

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* Isotope geochemistry is a specialized field and "groundwater isotopic hydrogeochemical prospecting" is so esoteric that it hardly exists. No studies of isotopes are detailed in this review and reference to isotopic work is made only in special cases. For an introduction to the subject of exploration isotope geochemistry see Adler (1958/667/) and Rosholt (1958/669/).

with respect to Ra and Pb. During 1953-4 a few Russian language papers appeared that dealt with the isotope- and radio-chemistry of groundwaters /186-8/. These investigations constituted an attempt to establish the origin of groundwaters and to recognize waters of differing sources, especially those rising from depth along faults. The significance of such studies for deep prospecting was emphasized later by Ginzberg /55/.

On the analytical side, Yoe *et al.* (1953/641/) described the use of dibenzoylmethane, which was to become a fairly important reagent for the colorimetric determination of uranium in natural waters. The fluorimetric method (Grimaldi *et al.* 1952/513/) was, however, destined to become the most widely used technique in the nineteen-fifties.

4.22 THE WEST : NON-RADIOACTIVE MINERALS

In the West, the study of the hydrogeochemistry of non-radioactive species produced several interesting papers.* Fricke (1953/139/) reported the concentration of Cl, CO_2, Li, Cs, Ru, Ni and Co in mineral springs and speculated about the extent to which these substances were contributed by juvenile sources as opposed to existing ore lodes. Marmo published two papers on work in 1953 in Finland /140-1/. He reported that in the

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* It is worth noting once again that not all of the results of such studies were published. Brotzen /704/ reported that tests for heavy metals in waters (following Huff /91/) began in northern Sweden in 1953. Although positive results were obtained from some stream, bog and spring waters associated with unmined lead deposits, the results obtained in other areas were totally negative and water testing was therefore discontinued. Apparently no detailed account of this project is available.

Nokia area copper and zinc bearing phyllite was followed by analysing groundwater from 300 wells. Copper was determined by the method of rate of catalysis of Fe III to Fe II and [Zn] was measured polarographically. He could correlate the pH of these waters with the sulphide content of the adjacent rocks. The Cu concentration of the waters ranged from 0,5 to 5 mg/1 (sic) and seemed too sensitive to insignificant variations in the copper content of the country rocks to be of use. The concentration of Zn in the water varied from less than 5 to 20 mg/1. In the Toivakka area he found that very small fluctuations in the Cu and Zn content of the bedrock were reflected in the corresponding groundwater. The author reported the occurrence of some remarkable false anomalies, where the weathering of pyritiferous but otherwise barren granites produced acidic groundwaters (wellwaters) with up to ,5 mg/1 Cu and 5 mg/1 Zn. Marmo's chief interest, in this work, was to demonstrate the concatenation (concentration of plant metal) = f (groundwater metal) = f (bedrock metal), rather than to utilize the groundwater as a prospecting tool.

Huff (/53/p.309) noted that the zinc content of waters from mine workings at Jerome, Arizona, decreased to one twenty-fifth of the original value after travelling only three miles along an irrigation ditch, although no pH change or dilution occurred. The loss was attributed to ion-exchange.

Two important formal statements of concepts (not necessarily developed in connection with hydrogeochemistry) made during 1953 were those of the orientation survey (Hawkes /125/) and pathfinders (Warren and Delavault /126/), although both ideas are implicit in earlier works by many authors.

Additional and improved methods of water testing, generally colorimetric, continued to appear regularly /127-32/ and by 1954 a water testing colorimetric kit became available commercially /132/.

4.23 THE EAST : NON-RADIOACTIVE MINERALS

Japanese interest in groundwater prospecting continued /127, 133-6, 185/. Shima (1953/127/) used the concentration of Mn in pit waters as a rapid gauge of the quality of the adjacent ore. Umemoto (1953/136/) found that sulphate, chloride and temperature identified springs that were related to mineralization. Yokosuka *et al.* (1953/185) determined criteria for distinguishing, in swamp areas fed by springs, anomalies due to sulphide as opposed to gypsum mineralization. The two types could be differentiated by the Zn, sulphate, Ca and Cu contents.

The output of Soviet publications during this period was relatively small*, although it is clear from Ginzberg's monograph /55/ that they were fairly active in groundwater prospecting at the time. Dolukhanova (alias Kolukhanova, 1956/496/), for example, was responsible for what was probably the earliest (1953) major investigation of Mo in groundwaters (in Armenia). Molybdenum tended to remain soluble in the pH range 2 to 8 and concentrations of Mo from 0,001 to 1,6 mg/l were observed in the groundwaters of the mineralized zones. This work is also an early example of observations of inter-component reactions as a factor influencing mobility - Dolukhanova found that the concentration of Mo was strongly dependent on the sulphide (sic) content of the water but inversely proportional to [Ca]**.

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* Alternatively, notice of such publications did not circulate widely in the English speaking countries.

** Ginzberg /55/ pointed out that these results contradict the then widely held belief that $MoO_4^{=}$ is mobile in "alkaline and limey waters". It will be seen, in subsequent sections of this review, that neither of these authors had the last word in this controversy.

Other Soviet workers continued to expand their investigations to cover lesser known groundwater components, for example fluorine /352/.

Kamenskii *et al.* (1953/176/) discussed the hydrology of mineral deposits and the implications for exploration.

The Soviets were also actively investigating aspects of molecular level phenomena and the physical chemistry of processes of importance to hydrogeochemistry /383-5/, such as the forms taken by various metals in natural waters and the mechanisms of fixation of metals by substances such as colloids, clays etc. Yatsimirskii (1953/776/), for example, showed that complex formation greatly increased the mobility of some elements, and that the solubility products and redox potentials of ions could be sharply changed by complexing.

4.3 THE PERIOD 1955 - 1957

After 1954 Soviet interest in the use of groundwater as a sampling medium mushroomed and, with the exception of the study of uranium in subsurface waters, the period 1955-7 is dominated by Russian language publications on this topic (for example /55, 344-5, 349-57, 495/). A fair amount of this work is now available in English translation.

4.31 SOVIET UNION : Zn, Cu and Pb in Groundwaters

The Soviets were at this time particularly interested in hydrogeochemical prospecting for base metals and their methods had become quite sophisticated, as the following examples will show.

In 1957 Bughelskii /782/ described the groundwater hydrogeochemistry of the complex (polymetallic) ores of arid Central Kazakhstan, where a major study had been undertaken in the early fifties. He stated that "hydrochemical methods of exploration of mineral deposits are acquiring ever greater significance at the present time. The possibility of their use over concealed bedrock ..... and their simplicity cannot be surpassed by other currently used methods". He notes, however, that "one of the main problems faced in the development of geochemical criteria (for polymetallic ore deposits) is a study of supergene migration of zinc, lead and copper".

The rainfall in this study area is only 200-300 mm/annum and Total Dissolved Solids (TDS) in the groundwaters varies from 200-1300 mg/l. Interestingly, TDS appears to be relatively higher in anomalous groundwaters, which also show characteristic lowering of pH and elevation of [sulphate] . Zinc, Cu and Pb form "an aqueous aureole with abnormal metallic contents". Following the standard pattern, lead is least mobile and zinc most mobile, being detectable in anomalous concentrations at distances of up to 900 m from the orebody. Background values differ in various regions (Zn 40-100 μg/l, Cu trace -60 μg/l, Pb traces only). "Aureoles" were found to contain up to 20 μg Pb, 200 μg Zn and 100 μg Cu per litre. Waters within orebodies could yield considerably higher values.

Bughelskii attempted to identify some of the factors controlling the intensity of the aureoles by comparing many data from several orebodies in different geological settings. The most critical factor was the "intensity of the development of the oxidized zone" of the orebody, since this process controlled the release of ions. Also important were any factors such as topographic slope or "tectonic breaks", which favour accelerated percolation. Once free of the orebody, the anomalous waters immediately become subject to reactions with the barren country rocks, which will tend to rob the waters of some of their excess metal. The rate of attenuation of the anomaly depended on the lithology of the country rocks and increased in the following sequence: Hornfelses, skarns/ sandstones, carbonates/carbonated sandy shales. In hornfelses, for example, lead anomalies up to 400 m wide were detected, but in carbonate rocks this distance was typically 30-60 m.

Like many previous authors, Bughelskii notes that a lower pH promotes mobility, but he has some radical views on the subject. He shows that a plot of pH versus the maximum metal concentration observed at that pH shows a regular trend with the lines for Cu and Pb, on the one hand, and Zn, on the other, separating sharply at concentrations above 150 µg/1. "Metal contents in ground waters generally do not exceed such figures and, therefore, there is obviously no relationship between the pH and very small metal contents ..... The effect of pH becomes notable only for higher metal contents. Thus *upper limits* of metal contents tend to decrease as pH increases" (reviewer's emphasis). This effect is more strongly felt by copper and lead, which are thus seldom encountered in abnormally high concentrations in neutral waters.

Sveshnikov (1955-7/356, 537/) described the chemical composition of very dilute (low TDS) bicarbonate-chloride groundwaters of pH 7,1 to 8,1 that are typically associated with the polymetallic deposits of Rudnyi Altai. Copper, Pb and Zn were again ubiquitous at concentrations below one milligram per litre. They frequently showed peak values of the order of at least ten times background, which was usually around $n \times 10^{-3}$ to $n \times 10^{-2}$ mg/l. Zinc was found to be a good pathfinder. Eh values (standard hydrogen electrode) varied between +250 and +420 millivolts.

An interesting development in the Soviet work of this period was the beginning of a concerted effort to consolidate available data on the chemistry of groundwater so that generalized conclusions could be drawn and guidelines laid down for further application studies. Tables 1 and 2 (/344/ and /345/ respectively, both 1956) are typical examples of consolidated data from Russian language papers of this ilk, some of which are reviewed briefly below:

Brodskii's work (1956/345/) on the waters of copper deposits is a good illustration of how much progress the Soviets had made in organising their hydrogeochemical data into general statements of empirical principles* concerning background and threshold values and the classification of various elements according to their mobility and behaviour in various natural waters. In terms of prospecting for copper, he divides water into three types: (a) surface waters associated with oxidizing ore, (b) surface waters associated with the oxidization of dispersed sulphides in "enriched mineralization fields" (i.e. metallogenic provinces) and (c) groundwaters infiltrated by either (a) or (b). The pH of these groundwaters is generally nearly neutral.

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* This approach was, however, not universally acceptable. There is for instance the important and forceful assertion by Germanov and Saukov (1955/349/) that there is no universal background value for hydrogeochemical surveys and that this value will have to be re-established for each new combination of circumstances. These authors regard the following to be the chief factors controlling background and anomalous values:
(a) Geological structure (in a very general sense, including lithology). (b) Geomorphology. (c) Properties of the ore sought. (d) The overall character of the local waters. (e) The hydrological relationships between the groundwater and the ore and (f) The climate (including the season) and other environmental factors peculiar to the given district.

TABLE 1/344/

Comparison of mineralized and non-mineralized waters

Metal	Concentration (g/1.) in surface and underground drainage	
	Non-mineralized areas	Mineralized areas
Nickel	$1 \times 10^{-6} - 1 \times 10^{-5}$	$1 \times 10^{-5} - 1 \times 10^{-3}$
Cobalt	$1 \times 10^{-7} - 1 \times 10^{-5}$	$1 \times 10^{-5} - 1 \times 10^{-3}$
Zinc	$1 \times 10^{-7} - 1 \times 10^{-5}$	$1 \times 10^{-5} - 1 \times 10^{-3}$
Copper	$1 \times 10^{-6} - 1 \times 10^{-5}$	$1 \times 10^{-5} - 1 \times 10^{-3}$
Uranium	$1 \times 10^{-8} - 1 \times 10^{-5}$	$1 \times 10^{-5} - 1 \times 10^{-3}$
Molybdenum	$1 \times 10^{-7} - 1 \times 10^{-6}$	1×10^{-4}
Lead	$1 \times 10^{-7} - 1 \times 10^{-6}$	$1 \times 10^{-5} - 1 \times 10^{-3}$

TABLE 2/345/

Comparison of mineralized and non-mineralized waters

Element	Weight clarke** in the lithosphere (%)	Sensitivity of detection (%)	"Abundance percentage"* In lead-zinc districts	"Abundance percentage"* Outside of lead-zinc districts
Pb	1.6×10^{-3}	1×10^{-3}	80	29
Mo	3.0×10^{-4}	3×10^{-4}	71	25
Mn	9×10^{-2}	1×10^{-1}	28	19
Cu	1×10^{-2}	1×10^{-2}	12	0
Cr	2×10^{-2}	1×10^{-2}	12	13
Be	6×10^{-4}	3×10^{-4}	9	4
Ga	1.5×10^{-3}	1×10^{-3}	8	5
Ti	6×10^{-1}	1×10^{-1}	8	3
Ni	8×10^{-3}	1×10^{-2}	4	1
Sn	4×10^{-3}	1×10^{-3}	4	1
V	1.5×10^{-2}	1×10^{-2}	0.6	0
Zr	2×10^{-2}	1×10^{-2}	0.3	0
Co	3×10^{-3}	3×10^{-3}	Trace	0
Nb	1×10^{-3}	1×10^{-3}	0	0
Ag	1×10^{-5}	3×10^{-4}	76	64
W	1×10^{-4}	3×10^{-3}	Trace	0
Zn	5×10^{-3}	1×10^{-1}	11	8
Bi	2×10^{-5}	1×10^{-3}	0.1	0
Sb	4×10^{-5}	3×10^{-3}	0.3	0
In	1×10^{-5}	1×10^{-3}	Trace	0
Ge	7×10^{-4}	1×10^{-2}	Trace	0
Ta	2×10^{-4}	1×10^{-2}	0	0
Cd	2×10^{-5}	1×10^{-2}	1.8	0
As	5×10^{-4}	1×10^{-1}	0	0

* number of cases per 100 in which the given element was detected.

** Clarke = average percentage of the element in the crust of the earth /346/.

He records values for the mean copper contents of various waters such as stream waters, marsh waters and groundwaters at various distances from Cu deposits (generally around 30-40 μg/l in all instances). On the other hand, the Cu content of mine waters can be very varied, from as low as 0,003 to about 7 mg/l in some (neutral?) mine waters, but up to thousands of mg/l in some others more acid than pH 5,3. However, the great majority of such determinations fall below 100 mg/l. At pH > 6,0 [Cu] generally falls sharply to sub-mg/l values. Although useful generalizations of this type are drawn, Brodskii does admit to the fact that real variations will be found between copper provinces and even within them. Thus peak and background values and their ratios change somewhat from one deposit to another and Cu anomalies around given orebodies can be discontinuous, erratic, or attenuated by high relief and abundant precipitation. He states that the generalized ideal behaviour of Cu in groundwater can be radically altered by factors such as the presence of organic matter (increased mobility of Cu) or high Total Dissolved Solids (decreased mobility). Depth of burial of the ore is also critical, more copper being released to the groundwater by the shallower bodies. In his experience, one may expect to detect the groundwater dispersion haloes of copper deposits at a maximum distance of between 50 m and 5 km from the source, depending on the conditions. In most cases the mean decrease in [Cu] as one moves outward from the source is quite regular and hyperbolic, but the rate of "decay" varies widely for different orebodies. Amongst the elements that Brodskii found to be commonly associated with Cu anomalies in groundwater are Zn, Ag and sometimes Mo. Zinc in particular often forms a halo broader than that of copper, and [Zn] usually obeys a similar hyperbolic decay law.*

4.32 SOVIET UNION : Anionic Pathfinders

It is implied in some of the foregoing examples that the Soviet writers were at this time also very interested in the use of anionic pathfinders for base metal sulphides (e.g. Krasnikov, 1955/495/). Brodskii /345/, although primarily interested in metal anomalies, stressed that low pH and high [$SO_4^=$] are both very important criteria for the recognition of anomalous groundwaters associated with copper mineralization. In his subsequent summaries of the general principles of hydrogeochemical prospecting (e.g. /784/), he repeatedly underscored the importance of the sulphate/chloride ratio as a generally useful tool. Indeed, the use of anionic pathfinders may be mandatory in some cases. Krainov (alias Kraynov) (1956/351/, 1957/536/) records that some types of sulphidic deposits could not always be located by searching for the traditional combination of low pH and high aqueous concentrations of metal (whether Cu, Pb or Zn), especially in regions where the natural background metal values in groundwater are high. He found that the mean background value for sulphate ion in groundwaters in Armenia was 6-10 mg/l

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* The cynic will be forgiven for making the observation that, by the time he is finished, Brodskii sounds suspiciously like Germanov and Saukov. Brodskii (apparently) partly contradicts himself by erecting a general system for classifying elements and ions according to mobility, in which he describes Zn and $SO_4^=$ as "accumulating" (i.e. rapidly collected by groundwater and forming wide but possibly erratic haloes), Mo as "non-dispersible" (i.e. forming clearly defined haloes) and most of the rest of the ore-forming elements as "dispersible" (i.e. dissipated from groundwater by various processes (including, presumably, precipitation), and therefore not given to forming clear haloes).

and that anomalous peaks of 15 to 200 mg/l could be expected, depending on the type of mineralization, with chalcopyrite deposits giving the highest values. He noted, however, that the sulphate/chloride ratio was a more reliable pathfinder, especially for polymetallic deposits as opposed to chalcopyrite or sulphur-pyrite bodies. High values for the ratio are a favourable sign, but care must be taken to correct for seasonal factors, since cyclic changes in the ratio value bespeak an unequal climatic influence on the respective absolute concentrations of the chloride and sulphate ions. Krainov found that the ratio in background areas varied from 1,5 in early summer to 2,5 in autumn. There are obviously other limitations, too, since the appearance of sulphatic waters can be the result of many different agencies, not all of which are connected with economically important concentrations of minerals. Gypsum is one obvious source of the ion and Brodskii /345/ pointed out that pyrite bodies can generate sulphate anomalies that are identical to those associated with sulphides of the rarer metals.

4.33 SOVIET UNION : Less-common Pathfinders and Minerals

One can gauge from the foregoing that by 1957 the traditional combination of Cu-Pb-Zn and sulphate had almost become the "touchstone" of groundwater hydrogeochemical exploration in the U.S.S.R. /55 p. 227/. However the Soviet researchers had also accumulated a substantial body of data on the use of additional pathfinders and on prospecting for a wide range of mineral deposits. Note, for example, that Tables 1 and 2 (pp. 46 - 47) list data for twenty-five elements. In addition, the Soviets were experimenting and prospecting with parameters and species such as phosphate /350/, fluorine /352/, pO_2 /353/, Eh /356 and 55 p. 217/

and boron /354 and 55 p. 227/. Vinogradov (1957/528/), for example, found as much as 10 μg Mo/l in springs issuing from well-fractured biotite granite on steep slopes below contact molybdenite bodies at the Lyangar deposits of the Caucasus. Classic fan-shaped anomalies, such as those normally associated only with pedogeochemical dispersion, are well developed in the groundwaters below the ore lodes. The favourable conditions that permitted this phenomenon were the even slope of the water table and the high and uniform permiability of the ground.

It was reported by Ginzburg /55/ that the spectra of residues of waters of quartz-tungsten bodies included weak lines of Be, Pb, Sn, Ag and W and stronger lines of Mo and especially copper. Similar treatment of waters from Mo deposits yielded weak Sn, Cu, Mo, Co, W and Ti spectra, but strong Sr lines. Although [Mo] was low, it nevertheless served well as an indicator of molybdenum deposits.

Ginzburg added his own general conclusions about the association of certain elements in groundwaters with a broad category of orebodies (the pyritic deposits of the southern Urals). Silver (<1 to 260 mg/l), Cu (<1 to 21,5 mg/l), Pb (<1 mg/l), Zn (<1 to 35,6 mg/l), As (<1 to 22 mg/l), Hg (<1 to 1,6 mg/l), Mn (<1 to 7,4 mg/l), Ag (about 40 μg/l) and Au (about 3 μg/l) were noted, but antimony could not be detected in any sample by wet chemical tests. By arcing dry residues of these waters the elements Co, Mo, Ba and Sr were commonly detected and occasionally Cr, V, Cd and Ga. Tin was always undetectable. Ginzburg also records wet chemical as well as optical spectrographic (dry residue) analyses of waters (apparently both surface and groundwaters) associated with the polymetallic deposits of Central Kazakhstan. Copper, Pb and zinc were generally found at concentrations below 1 mg/l and Mo and Ag from

$n \times 10^{-4}$ to $n \times 10^{-3}$ % of the dry residues. Molybdenum appeared to be very mobile. Beryllium, P, Sb, W, Ge and In could not be detected and Sn and As were very rare.

In addition, Ginzburg claimed that boron is a useful pathfinder for Cu, Fe and Pb-Zn ores in addition to borax deposits. Albov (1957/781/), on the other hand, summarized the information available about boron in the groundwaters of Crimea, and came to the opposite conclusion. He showed that detectable concentrations of boron ranged from about $n \times 10^{-2}$ to $n \times 10^{3}$ mg/l, that the principle sources of boron were volcanic activity and marine sedimentary rocks, and that there was nothing to suggest that boron was a useful hydrogeochemical pathfinder in this region.

Beus (1956/780/) discovered that mine water at pegmatite deposits in the U.S.S.R. initially contained as much as 10 μgBe/l, but that precipitation, or co-precipitation with oxides of Fe, Mn and Al, soon lowered the concentration of beryllium to below the detection limit.

4.34 SOVIET UNION : Miscellaneous Studies

In addition to emphasizing the typical relationships which might be expected to exist between orebodies and trace elements dissolved in associated natural waters, the Soviet prospectors were very interested in the overall characterization and classification of natural waters as a basic exploration tool. Whereas Brodskii /345/ had divided certain anomalous waters into three types, Udodov and Onufrienko /55 p. 127 *et seq.*/ classified the natural waters associated with or adjacent to mineralized areas in the mountains of western Siberia into fifteen kinds, on the basis of (a) "Type of natural water", for example, surface- or groundwater, (b) " ..... Environment with respect to sources of mineralization", e.g. waters of mineralized zones or waters of pedologically dispersed regolith and (c) "Types of rock, in reference to their effect on pH of natural water", i.e. either "active or inactive". For each kind of natural water they list the typical pH range found, [$SO_4^{=}$], [heavy metals] and additional information such as the presence of free oxygen and similarities and differences between the heavy metal associations in the water, on the one hand, and in the parent rock on the other.

It is appropriate at this point to mention the work of Chebotarev (1955/1048-1050/). Chebotarev was not a prospector, but he was one of the "fathers" of a hydrochemical concept that would later prove to be of great importance in groundwater hydrogeochemical exploration, especially for uranium. His work is another reminder that this science has always borrowed from, and been enriched by, many related disciplines. Chebotarev was concerned with the fundamental classification of groundwater types and with the essential chemical characteristics of the subterranean hydrological cycle. His conclusions were based on analyses and hydrological data for 10 000 groundwater samples from many parts of the Soviet Union. The main point he made is that, all else being equal, the chemistry of groundwater is a function of its age, and that waters "metamorphoze" as they pass through the hydrological cycle of recharge, transportation and discharge. Thus young, fresh waters in upland recharge areas tend to have common chemical characteristics and old or well-travelled waters all tend to be chloride-rich and brackish. The normal course of this metamorphic sequence is bicarbonate waters → HCO_3-Cl waters → Cl-HCO_3 waters → Cl-SO_4 waters → SO_4-Cl waters → sulphate waters

→ chloride waters. Variations occur within the general scheme, but its overall direction is not reversible unless significant dilution takes place. We will see, in subsequent sections of this review, that many workers have made very good use of Chebotarev's principles in groundwater hydrogeochemical exploration.

There was a renewed Soviet interest at this time in the possibility of substituting rapid geophysical (conductivity, resistivity) measurements for certain chemical tests, for example in areas where H_2SO_4 from pyritic mineralization increases the specific conductivity of groundwaters (/494/1955, cited reference published 1958).

Undiminished attention was paid to the physico-chemical controls of solubility and mobility and on the inter-relationships between various species present in different water types (e.g. /504-5/). Beliakova (alias Belyakova, 1956/348, 785/, 1957/533/), for example, found that in groundwaters associated with molybdenum mineralization in central Kazakhstan [Mo] was independant of $[SO_4^=]$ and pH and that high levels of calcium bicarbonate resulted in the precipitation of calcium molybdate - the latter observation confirming the earlier findings of Dolukhanova (alias Kolukhanova, /496/). Plotnikov and Syrovatko (1957/794/) studied groundwaters in various orebodies and emphasised the very powerful role that co-precipitation plays in controlling the abundances of metals in these waters. Various workers (e.g. /356-7, 506/) discussed the importance of Eh with reference to the solubility of sulphide minerals and the dispersion of liberated heavy metals. Shcherbakov (1956/654/) presented an extensive summary of the geochemical significance of Eh conditions and redox reactions in the "underground hydrosphere".

One of the most noteworthy examples in this category is the treatise by Shcherbina (1956/775/) on the role played by complex ions in the transfer of elements in the supergene zone. This subject had been relatively little studied before 1956, but it was clear that "many elements such as ferric ion, titanium, zirconium, uranium, tin, scandium, silver, gold and others are immobile in the form of simple cations in the neutral aqueous solutions of the supergene zone, and if it were not for the existence of other modes of transfer, they could not participate in geochemical migration. When these cations form complex ions, usually changing to anions in the process, their chemical properties change greatly. In most cases their solubility increases, the oxidation--reduction potential changes sharply and the elements become very mobile ..." (p. 486). The example that Shcherbina uses to illustrate this concept is the normally-insoluble compound AgCl, which nevertheless dissolves readily in brine:

$$AgCl + NaCl \rightleftarrows Na(AgCl_2)$$

The "instability constant" (K) for this complex is defined for

$$(AgCl_2)^- \rightleftarrows Ag^+ + 2Cl^-$$

and

$$K = \frac{[Ag^+][Cl^-]}{[(AgCl)_2^-]} = 2{,}3 \times 10^{-6}$$

The smaller the value of K, the more stable the complex. Thus the above chloro-complex is not dissociated by either phosphate or carbonate ions, but will be destroyed by sulphide ion, because the value for the solubility product of Ag_2S is extremely small (10^{-51}). Other complexing agents and other metals will, of course, behave differently. Thus "the sulphide ion does not precipitate iron from its stable complexes with soil acids, because the concentration of iron ions resulting from the dissociation of these complexes is lower than the concentration of iron ions corresponding to the solubility product constant of FeS ($3,7 \times 10^{-19}$) ..... Complex ions play the role of solvents, if the new complex ion resulting from solution is less dissociated than the first one ..... The reactions occurring according to this principle may be much more common than is known at present" (p. 487).

The tendency for elements to participate in complex formation varies rather regularly with periodic properties, increasing with both increasing valence and decreasing ionic radius. The characteristic complex-forming elements are Be, Al, Fe^{+++}, Ti, Zr, Sn^{4+}, V^{4+}, U^{6+} and to a lesser degree Cr^{3+}, V^{3+}, Sc^{3+}, the rare earths, U^{4+}, W^{6+}, Mo^{6+} and possibly Nb and Ta, all of which have ionic potentials (IP) of intermediate magnitudes. The ions with high IP (S, P, Si, N) tend to form simple anions similar to PO_4^{3-} and $SO_4^{=}$, while the metals with low IP's, such as Rb, Ba, Na, Li etc. do not readily form complexes.

The more stable complex ions will tend to migrate the furthest, since they will be less susceptible to destruction by newly-encountered chemical agents or changes in physico-chemical parameters. For example, the redox potential for the reaction $Au \rightarrow Au^{3+} + 3e^-$ is 1,50v. but for the reaction $Au \rightarrow AuCl_4^-$ it is only 1,0v. Moreover, "it is possible that gold may form complex ions with still smaller instability constants with one of the numerous soil acid" (cf. Freise, 1930-1934/778-9/).

Nevertheless, no complex can exist indefinitely under thermodynamic conditions that are very unfavourable to it. Thus free atmospheric oxygen, for instance, is eventually fatal to humic or other organic complexes of many metals, notably iron.

Other notable Russian publications from this period include general articles or reviews that summarize aspects of hydrogeochemical prospecting

/497-8, 710/ and discussions of procedures for sample collection, analysis and interpretation, including the construction of hydrogeochemical maps /499-503, 534/.

Ginzburg's text (1957/55/, English translation 1960) is of course the most important publication of this period and remains a standard text for the hydrogeochemical prospector. His discussions range over a very broad array of topics of interest and include two chapters devoted entirely to the use of natural waters in geochemical exploration. Krasnikov (1957/535/) edited a volume on geochemical prospecting that included observations about groundwater hydrogeochemistry but the book was not translated into English.

4.35 EASTERN COUNTRIES OTHER THAN THE SOVIET UNION

Very little of the literature of groundwater hydrogeochemical exploration that was published between 1955 and 1957 in Eastern countries other than the Soviet Union is available in English. Fukuo and Ohtsuka (1955/1120/) measured the sulphate contents of river, spring and well waters in the Noto Peninsula of Japan. They found that the distribution and abundance of this ion was closely correlated with the occurrence of gypsum beds and suggested that these facts be used for "geochemical prospecting of gypsum".

4.36 THE WEST : NON-RADIOACTIVE ORES

During 1955-7 Western publications on groundwater prospecting for elements other than uranium were predominantly from North America.

Boyle and co-workers (1955-6/527, 819/) studied heavy metal anomalies in the waters of springs and streams in Yukon Territory, Canada. They demonstrated that the liberation of dissolved metals into groundwater was favoured by fracturing or shearing of the ore and host rocks and that disrupted, disseminated or permeable ores tend to show stronger hydrogeochemical anomalies than those associated with compact, massive ores of equivalent mass and grade.

Zinc is the principal heavy metal in the spring waters of the lead-zinc-silver province of Galena Hill-Mount Haldane. Some Pb and Cu are encountered in waters issuing from rocks in the vicinity of ore shoots containing galena, sphalerite and freibergite. The pH of the waters in this region was nearly 7 and the temperatures were 2-4^{o}C. Zinc concentrations of as much as 80 mg/l were found, but [Cu] and [Pb] did not exceed 1 mg/l. The anomalous spring waters were related to oxidized ore bodies within Galena Hill and were thought to have drained towards the eyes of the springs via mine workings, fractures and faults. If the moving water drains through an appreciable thickness of surface muskeg before emerging, then the zinc content is radically diminished. As usual, zinc was more mobile and a better pathfinder than either copper or lead. The authors emphasize the importance of locating "iron springs" - i.e. those precipitating Fe/Mn hydroxides - since these are often very important indications of mineralization in the vicinity. It must be remembered, however, that displaced anomalies are a common occurrence in faulted rocks. Spring waters may reach the surface via devious paths, for example along fractures that are not directly related to the ore lode. A useful general observation is that, unless the circumstances are very peculiar, the springs always issue below the topographic level of the orebody. False anomalies may be caused by disseminated mineralization and there appears to be no hydrogeochemical method of telling when this is the case. Despite the partial permafrost conditions that prevail in this region, the hydrogeochemical method works very well and few of the lodes are premanently "frozen in". A more serious limitation is perhaps contamination, for in some areas the metal content of spring waters has been abnormally enhanced by mining. Several new anomalies were noted in this area, and these were regarded as being well worthy of attention.

Results similar to those described above were obtained in the adjacent Keno Hill-Galena Hill area. Zinc and sulphate are the main pathfinders. Deeply drawing springs generally give better heavy metal anomalies than those of shallower origin and usually have higher TDS. Certain waters with high TDS, [Mn] and [Fe] are related to mine workings and deep ore lodes. The relatively dilute waters of shallower springs may have high zinc contents if they come into contact with the upper portions of ore veins. Several anomalies were noted in the area, some related to known occurrences of sulphides and others attributable to unknown causes. Not all of the known orebodies are associated with anomalous springs. In at least one case spring waters gave anomalies while the associated stream waters did not.

Sampling of spring waters and groundwater seepage played a minor role in a very successful prospecting campaign in New Brunswick between 1952 and 1954 (Hawkes *et al.* 1956/531/).

Conversely, a study by Kennedy (1956/520/) served to emphasize the unfortunate but ineluctable fact that groundwater prospecting may be quite useless in many cases where sulphide orebodies are not exposed to oxidation, for instance in deep lodes below the zone of weathering or perhaps shut off by a high water table. He found that the waters from drill holes in a deep, undeveloped zinc sulphide body in south-western Wisconsin were not anomalous in zinc, even though spring waters issuing from these deposits contained as much as 50 µg/l heavy metals. He demonstrated that the stability of heavy metals in spring waters of the Potosi area (Wisconsin) was pH sensitive and that the metals were rapidly precipitated as the pH rose in the streams fed by the spring waters. Kennedy's studies in Wisconsin confirmed the importance of zinc in groundwater hydrogeochemical prospecting for sulphidic ores.

Cannon (1955/398/) showed that disseminated zinc (sphalerite) in the Lockport dolomite was transferred via the medium of groundwaters and spring waters to the nearby Manning bog (New York State), where the metal was immediately immobilised by peat and organic muck*, which contained more than 10% Zn (dry weight) in some places. Ermengen (1957/529/) reported similar findings.

During the period reviewed here there are but few American parallels to the Soviet attempts to lump together data and draw statistical conclusions about the nature and composition of groundwaters sampled in mineral exploration. One example is the work of Gill and Denson (1957/532/) who presented a useful compendium of water analyses from Montana and the Dakota states.

Hawkes' "Principles of Geochemical Prospecting" /53/, which can be regarded as an American parallel to Ginzburg's book /55/, is, in terms of hydrogeochemical prospecting at least, far behind the Soviet work. Hawkes fails to back up many of his conclusions with data or literature references, but he does draw some useful general conclusions. Unlike the Soviets, he

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* This was not a new discovery. Staker and Cummings (1941/530/) and Forrester (1942/399/) had reported essentially similar results.

is not too enthusiastic about the possibilities of groundwater hydrogeochemical prospecting but concedes that considerable masses of certain elements such as Zn and Co do move in groundwaters either in solution or colloidal suspension. He blames contamination for the poor success rate enjoyed by well and borehole sampling projects and suggests that spring water sampling may prove to be more successful, because contamination problems are greatly reduced and because spring water is generally drawn from near the water table, where oxidation is at a maximum. Hawkes is more specific about practical procedures in groundwater sampling than Ginzburg is. He notes the ease with which the trace element concentrations of water samples can be altered by contamination or removal of ions by precipitation, adsorption and other phenomena, and recommends that samples be acidified immediately after collection to minimize this last problem. Borosilicate glass or plastic containers are recommended, but he makes the regrettable claim that "Polyethylene bottles ..... are ..... free of contamination".

Few important publications appeared at this time in Western countries outside North America. A notable exception is Schoeller's work on the geochemistry of subterranean waters (1955/663/). Although he was concerned principally with oilfield waters, his graphical methods and hydrochemical observations have been widely adopted in hydrogeochemical exploration.

Kauraune's account (1955/1119/) of hydrogeochemical prospecting for Cu and Zn in northern Norway is notable for its assessment of errors caused by the ion-exchange capacity of the plastic sampling bottles used.

Leutwein (1957/1019/) showed that the analysis of groundwaters could be used for finding bismuth ores.

Finally, brief mention must be made of a publication by Ferris and Sayre (1955/627/), which contains many references to early publications that are of importance in the development of the scientitic principles of hydrogeochemistry. These authors also predicted correctly that the nineteen-sixties would witness an unprecedented growth of quantitative investigations in all branches of hydrogeochemistry.

4.37 URANIUM : MIGRATION AND ACCRETION

After 1955 there occurred a worldwide surge in the number of publications on hydrogeochemical prospecting for uranium*, mainly in surface waters (e.g. /702/), but also in subterranean waters. The enthusiasm with which this technique was adopted in both the East and the West is illustrated by the following quotations:

"Water sampling of both ground water and surface streams shows that favourable uranium formations and districts can be outlined by this method" (United States Atomic Energy Commission 1956/636 p. 753/).

" ..... hydrogeochemical methods (of prospecting for uranium), because of the deep penetration by water underground far below the instrumental zone, also can be used in some places where other methods fail" (Fix 1956/541 p. 788/).

"The hydrogeochemical method is used as the primary method of exploring unfamiliar territories" (in the U.S.S.R., for uranium) (Vinogradov 1956/637 p. 824/).

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* Readers interested in the early (pre-1955) development of this subject in the West can consult the following pioneering references: /507-9, 513-17/. Very little of the earliest Soviet work has been translated, but an important review in English of the origins of uranium hydrogeochemistry in the U.S.S.R. has been given by Starik *et al.* /1121/. Preliminary work on uranium hydrogeochemistry began in 1941, and large scale regional surveys were made between 1948 and 1958.

The efficacy of the hydrogeochemical method for locating uranium deposits is due largely to the unique secondary mobility of this element. Between 1955 and 1957 many authors /528-46, 637, 1095/ showed that uranium could become mobile under alkaline oxidizing conditions in the form of species such as UO_2^{++}, $UO_2(CO_3)_3^{4-}$ or $UO_2(CO_3)_2^{2-}$, and that consequently the analysis of U in fresh waters is a powerful reconnaissance method. Blake (1955/598/) investigated the latter two complex ions and found that they were stable above pH 7,0 as alkali or alkaline earth uranyl carbonates*. Shcherbina /775/ stated that "the presence in nature of the complex carbonates of hexavalent uranium ..... is convincing proof of supergene migration of uranium in the form of $Na_4(UO_2(CO_3)_3)$"(p.491).

Starik and Kaliadin (1957/685/) showed experimentally that the rate of loss of uranium from natural solutions was reduced by high values of pH and $[HCO_3^-]$. Additional confirmation of the importance of the carbonate and bicarbonate ions in the mobilization of uranium in natural waters is well documented /e.g. 686-8/. Thatcher and Barker /638/ demonstrated that, in addition to bicarbonate ion, sulphate and chloride ions also tend to stabilize dissolved uranium in the form of complexes. Experiment has shown that uranium could also exist as a soluble organic complex below pH 2,2 to 2,18 (Breger and co-workers, 1955/579, 588/). But whatever form it might take, "the mobility of the uranyl ion is very high and hence its direct detection holds promise for locating ore deposits" (United States Atomic Energy Commission /636 p. 753/).

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* The U(IV) ion is not known to form carbonate complexes, and this may explain why the uranous ion (and also Ra and Pb) are more slowly elutriated by acidic solutions than is U(VI) /577/. The following references deal exhaustively with the solution and carbonate chemistry or uranium: /687-8, 750-4, 756-63/.

The second factor that favours the application of hydrogeochemical methods in the search for uranium is the ready solubility of many common uranium-bearing minerals. Brown and Silver* (1955-56/618, 697/), for example demonstrated that up to 40% of the U (and 42% of the Th) in some igneous rocks were readily solubilized by weak acids.

The third important phenomenon that characterizes the unique behaviour of aqueous uranium is the fact that, under the appropriate respective conditions, the element is immobilised with the same alacrity with which it is solubilized. Unlike most elements that are commonly associated with sulphidic ores and that are generally irreversibly dispersed when such ores are weathered, uranium has the ability to dissolve into groundwater, migrate and, under suitable conditions, reprecipitate in economically attractive concentrations. This process of "multiple migration-accretion" (Gruner /568/)** was intensively documented between 1955 and 1957/564-73

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* cf. the earlier work of Larsen and Phair (1954/631/) and Hurley (1950/617/)

** HISTORICAL NOTE: This theory of migration in groundwater and relocation or uranium has venerable roots, viz: (i) Butler *et al.* 1920/551 p. 158/): "The deposits are regarded as having been formed by circulating waters that collected the metals disseminated through the sedimentary rocks and deposited them on contact with carbonaceous material. The circulation in some places is believed to have been of artesian character and to have been controlled to a large extent by structural features". (ii) Lindgren (1933/552 p. 414/): "it (uranium) has been concentrated with vanadium, by meteoric waters, which derived the metals from terrigenous sediments resulting from the disintegration of pre-Cambrian igneous rocks and pegmatites".
See also Koeberlin (1938/553/), Fischer (1942/554/) and more recently (1950 to 1954) the following references describing both field studies /550, 555-63, 582-4/ and laboratory experiments /577, 582-3, 593, 599/. The migration-accretion theory may, of course, not be correct in some cases, or it may be wholly wrong. Hess /671/ proposed in 1922 that the Colorado Plateau ores are hydrothermal and this theory received new support in 1958 from Kerr /672/ and Gabelman and Boyer /674/. Other mechanisms have also been proposed /672/, for example a syngenetic origin /673/, but this seems improbable in view of isotopic ages obtained for many of these Colorado Plateau-type ores /733-4/.

578-81, 585-92, 595-7, 629, 692-3, 695-6/. Although it can be argued that the *creation* of orebodies is hardly the direct concern of the exploration geochemist, some discussion of the formation of secondary uranium ores is essential to a proper understanding of the hydrogeochemistry of this element.

Gruner (/568 pp. 496 and 513/) has elaborated as follows: "Concentration of large deposits could proceed by several stages of oxidation-solution-migration-accretion, a kind of 'recycling' action ....." and that, given enough time, this process will "produce large individual ore concentrations by ordinary groundwater leaching".

Klepper and Wyant /632 p. 218, 633/, while describing acid-soluble minerals dispersed in certain igneous rocks, observed that the presence of "this acid-soluble material ..... has several important implications. First, it suggest that significant quantities of uranium can be leached from igneous rocks by ground water and perhaps later be concentrated as secondary deposits in coal or other sedimentary rocks".

Gill and Moore (1955/596/) suggested that percolating meteoritic waters dissolved both uranium and vanadium from the tuffaceous Arikaree formation (Miocene, South Dakota) and redeposited these elements in an underlying sandstone, along the base of a water table perched on bentonite. Spring waters from this perched water table yielded from 42 to 172 μgU and 290 μgV per litre. The waters from formations below the bentonite are poor in uranium.

There has been much theorizing and speculation and some experimental work on the causes of this secondary deposition. Several mechanisms may be responsible, for example reaction with organic matter /632-3, 731, 741/, reaction with inorganic dissolved substances /683/, or changes in pH /579/.

Thompson and Krauskopf /566/ expressed the belief that such reprecipitation of uranium can only occur from solutions stronger than 10 µg/l and studies of sulphur isotopes associated with some secondary deposits suggest that in many cases the agent responsible for the reprecipitation was probably H_2S produced by anaerobic bacteria (Jensen 1957/567/). In 1956 Sims /683/ had shown that even traces of H_2S (in natural gases) caused significant precipitation of uranium from aqueous solutions, especially in the presence of Fe^{++} (see also /568/). Miller's experiments /724/ showed that organically derived H_2S reduces U^{6+} to U^{4+}, which is then precipitated and ages in the following sequence:

$$UO^{++} \rightarrow UO(OH)_2 \rightarrow UO_2$$

Lovering (1955/575/) proposed an alternative mechanism that may operate in some instances of ore formation by immobilization of uranium migrating in groundwaters. He notes that uranium associated with oxidizing sulphides* will enter the groundwater as uranyl sulphate and will migrate together with ferric sulphate derived from the weathering ore.

$$UO_3 + H_2SO_4 \xrightarrow{pH\ 2,5} UO_2^{++} + SO_4^{=} + H_2O$$

Upon neutralisation of these acid waters, the ferric sulphate is hydrolyzed to colloidal hydrated ferric oxide, which co-precipitates the uranium. This postulated mechanism was successfully duplicated in laboratory experiments /568, 576/.

On the other hand, Breger *et al.* /579/ have pointed out that alkaline uranyl carbonates in solution are sensitive to (any acids). For example,

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* Grutt (1957/680/) showed that the solubility of U in shallow sandstones is increased by the presence of dispersed pyrite, which is oxidized and thereby increases groundwater acidity.

such uranium compounds, upon encountering acidic lignite beds, will decompose with the formation of insoluble sodium and uranyl humates.

$$Na_4(UO_2)(CO_3)_3 + 6HA \rightarrow 4NaA + UO_2A_2 + 3CO_2 + 3H_2O$$

Shcherbina and Ignatova (1956/648/) felt that the precipitation of secondary uranium minerals was most probable when the concentration of uranium in groundwaters exceeded one milligram per litre (a value 100 times greater than that proposed by Thompson and Krauskopf /566/) and that the observed precipitation of secondary compounds such as phosphates was "in agreement" with measured solubility products. Shcherbina (1956/775/) noted that Cu^{2+} (and by implication possibly other ions) is a powerful agent for the precipitation of uranium from its complexes.

4.38 URANIUM : CASE-HISTORIES AND RELATED STUDIES

The very high mobility of uranium under certain oxidizing conditions /53/ is confirmed by empirical observations of the concentration of uranium in various natural waters, and the ubiquitous concurrence of uraniferous rocks and uranium-rich formational waters is certainly very well established through case histories and other practical hydrogeochemical studies. Many of the authors mentioned in the foregoing (notably Murphy /548/, Walton /549/ and Fix /512, 541/) have observed that, as a rule, relatively high concentrations of uranium occur in spring waters in regions in which uranium deposits are located. Denson and his co-workers /540, 547/ showed that spring waters from abnormally radioactive volcanic rocks (especially acid pyroclastics) contain from 10 to 100 times more uranium than most "ordinary groundwaters". Springs issuing from granitic rocks at high altitudes (1500 to 2400 m) in the Sierra Nevada of California carry significant amounts of uranium, which is dissolved out of these rocks and precipitated in nearby bogs, where concentrations of up to 0,7% U_3O_8 occur /581/. The general principle is recognized in numerous additional publications dealing with the radioactivity or radioelemental composition of spring waters or the precipitates therefrom /608-10, 613/.

Denson and co-workers have recorded some interesting observations of the distribution of uranium in subterranean waters /539-40/. In the Colorado Front Range they conducted a reconnaissance survey by measuring U in streams, springs and wells and found from 0,5 to 120 μg/l. They stated that "uranium contents of 120 to 50 parts per billion in formation waters are believed to indicate destruction of a uranium deposit".

In Nebraska and neighbouring States subterranean waters from Tertiary strata, including radioactive tuffs, contain from 4 to 110 micrograms of U per litre (Tourtelot 1955/612/).

Some interesting practical and theoretical work on U in groundwaters appeared in the French language /662-3, 713/. Coulomb and Goldstein /662/, for example, examined the uranium content of waters associated with known deposits of granitic massifs in France, with the aim of developing "practical methods of geochemical prospecting" for the metal.

Of those papers that may be termed overviews of uranium in groundwater, the most important are those by Fix /512, 541/ and Saukov /544/. Both authors attempted to draw statistical conclusions about the abundance and distribution of uranium in various natural waters. Fix reported that the uranium content of natural waters was a function of pH and overall chemical composition of the water, and also of lithological criteria, and that [U] in water served as a rough measure of the uranium content of contiguous geological materials. Fix states that background values for U in subterranean waters in the U.S.A. were generally of the order of 10 μg/l and that analyses of groundwater from uraniferous areas ordinarily yielded values of one to 120 µg/l. However, acidic mine waters can carry several milligrams of uranium per litre. Threshold values for [U] are usually set at from three to ten times background and anomalies ranging from as little as 5 to several hundred µg/l have been reported. Fix also makes an important observation concerning the effects of man's activities on groundwaters: "Where ground has been disturbed by mining or water has been partly evaporated, abnormally high uranium concentrations may result, and interpretation must always be made with such factors in mind".

Saukov /544/ presented an excellent short summary of the principles of uranium hydrogeochemistry, or, as he also calls it, radiohydrogeology. He notes that: (1) Reducing waters will not promote the migration of uranium. (2) "The natural hydrogeochemical background is not established as a set of universally applicable values of the radioelement content of underground waters, but is determined separately for each definite type of rock, and for each district".(3) The interpretation of the collected data remains the most difficult part of any prospecting survey of U in water, and (4) the Soviets think highly enough of groundwater exploration for uranium to go to the trouble of sinking pits to get groundwater samples in certain areas where subterranean waters are not otherwise procurable. Saukov states that [U] in groundwaters (presumably mainly from data gathered in the U.S.S.R.)shows a wide range, from n x 10^{-2} µg/l to n x 10^{-2} g/l, reaching a maximum in either the waters of uranium mines or in regions containing numerous uranium deposits.

Several students of hydrogeochemistry stressed the role of hydrological factors such as the position of the water table (or water-oil interfaces /731/) and hydraulic gradient in the determination of the behaviour of U in underground waters. The United States Atomic Energy Commission stated that "The sampling of groundwater and measurement of its uranium content has (sic) resulted in the indication of areas favourable for uranium that have subsequently been found to contain commercial deposits. Most ground water contains less than 2 parts per 10^9 (ppb) uranium and water from volcanic and tuffaceous sediments (sic) considered favourable for uranium deposits may contain 10 to 250 ppb. In many areas sampling indicates that leaching of uranium is taking place at the present time. Variations in the uranium content of ground waters vary with the transmissivity of the host rock which, in turn, effects (sic) the availability of uranium" /636 p. 753/. Germanov (1956/647/) studied the hydrogeochemistry of shallow, oxidizing orebodies (uranium and other metals) and found that uranium tends to be most strongly leached from that part of the rock/soil profile within which seasonal fluctuation of the water table is strongest. Leaching is relatively restricted both below and above this zone. Garrels and Christ /681-2/ presented evidence to show that the degree of oxidation (and hence mobilization) of uranium ores was in some instances dependant on the relationship between the deposit and the modern water table, oxidation being promoted by a subsiding groundwater table.

Because of the widely-accepted view that the Colorado uranium deposits are related to groundwaters, several extensive studies with a hydrological or hydrological/chemical emphasis were undertaken in this area in the search for uranium /675-7/. Kelley /679/, for example, felt that persistent favourable hydraulic gradients are necessary for large-scale and/or long distance lateral migration of uraniferous solutions.

Miholic (1956/611/) stated that [U], like the concentration of many other elements in groundwaters, is dependant on climate and season and that provided all other factors are equal, the radioactivity of springs tends to be higher in wetter years. Both U and Ra tend to be unevenly distributed within the different springs of any district. Klepper and Wyant /632 p. 219/ have speculated that the process of "migration-accretion" is probably largely controlled by climate and that the redeposition of uranium is favoured by arid conditions with minimal rainfall runoff (surface or subsurface) and export to the sea.

There was relatively little comment in the literature at this time concerning the relationship between uranium and other trace elements in groundwater. Denson (1955/547/) investigated waters from uraniferous formations (Oligocene and Miocene tuffs) in Montana and the Dakota States and found that the elements As, Cu, P, V and Mo were associated in significant amounts with uranium in solution. High [Zn] may also be present (Denson *et al.* 1956/540/).

McKelvey and Carswell (1956/574, 694/) noted a relationship between phosphorus and uranium in groundwaters associated with some uranium-enriched rocks. They stated that [U] in the waters of phosphate mineswas as high as 22 μg/l, whereas most groundwaters contain less than one microgram of uranium per litre.

Austin (1957/594, 728/) observed that acid-sulphate solutions could mobilise U from primary deposits of urananite plus sulphides (of Fe, Cd, Pb and Cu) and that Mn, Co, Mo and Cu (but not Pb) are redistributed by the same process (but not necessarily to the same places). Isotopic studies /666 p. 225/ of secondary uranium minerals, which contain a small amount of lead as a lattice impurity, indicated that only "a small fraction of the amount (of lead) originally associated with uranium in the source rock" is included in the new minerals. This was in agreement with earlier isotopic studies /577, 665/ that suggested the relative immobility of Pb (and Ra) with respect to uranium. Rosholt (1957/727/) showed that certain long-lived daughter products such as Th^{230} and Pa^{231} are leached from sulphide ores along with uranium, but their concentrations are so low that they cannot seriously be considered as additional pathfinders for uranium.

Denson /539/ determined Ca, Mg, Na, K, bicarbonate, carbonate, chloride, pH, Total Dissolved Solids and conductivity in waters that had leached salts from soils around uranium deposits in clays, mudstones, silts and sands in the Black Hills of Dakota. He concluded that only the Ca/Mg ratio had any significant relationship to the concentration of U, but that this was probably a purely local phenomenon.

Amiel and Winsberg /538/ made an interesting attempt to predict quantitatively the size of uranium orebodies from the helium and krypton content of spring waters supposedly connected with such deposits.

Radium and radon frequently accompany* uranium in groundwaters /544/, and the radon especially accounts for the radioactivity of most "radioactive" thermal waters. Stead (1956/635 p. 717/) noted that "In gamma-ray logging of exploratory drill holes on the Colorado Plateau, small sharp anomalies detected several hundred feet down dip from carnotite ores are presumably caused by radon migrating in ground waters along permeable channels". Lovering /613/ pointed out that the radioactivity of spring waters was more often a function of [Rn] rather than of [U]. Radon has a short half-life and therefore the radioactivity of spring waters is dependent upon both the degree of radioactivity of the source rocks and the speed of circulation of the groundwater.

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* confirms earlier observations /605, 670/.

4.39 URANIUM : ANALYSIS AND REVIEW ARTICLES

The analysis of uranium was apparently a very tedious business in the mid-fifties, if one judges the method used by Denson /540/ as an example: "Pint samples of ground water were collected ..... The uranium content was determined by ethyl acetate extraction method (sic) as follows: an acidified aliquot of the sample, usually 100 to 500 ml, was evaporated nearly to dryness and then diluted with distilled water acidified with nitric acid. An aliquot of this solution was salted with recrystallized aluminum (sic) nitrate, and the uranium was extracted with ethyl acetate. The extract was then burned off in a platinum dish, and the residue was fused with a sodium fluoride carbonate flux. A sensitive fluorimeter was used to compare the fluorescence of the unknown with standards treated in a similar manner, and the uranium content was calculated ....." Hydrogeochemists were aided by the introduction around 1956-7 of simpler and less tedious analytical procedures for uranium /668/. Some important papers on the determination of U and other radioelements were presented by Ward and Marranzino /546/, Thatcher and Barker /638/ and Barker and Thatcher /642/ (all 1957).

Predictably enough, many important review papers or textbooks /655-9, 678, 698/ on the subject of "radiohydrogeochemistry" appeared during this period. The most important of these, for the English speaking reader, is Volume VI of the (First) United Nations International Conference on the Peaceful Uses of Atomic Energy /636/. Others, although potentially of great interest and value, have apparently not been translated from Russian.

It should be noted that, despite the great number of investigations of the uranium content of natural waters in many parts of the world during this period, there were at this time many who regarded the hydrogeochemical method as being of minor interest only when compared to radiometric methods of prospecting. Lang (1956/634 p. 667/), for example, in his summary of uranium prospecting in Canada, merely suggests that "geochemical methods of prospecting for uranium hold possibilities ....." and he dismisses the entire subject, including the applied geochemistry of natural waters, in less than twenty lines.

4.4 THE PERIOD 1958-1962

4.41 URANIUM : INTRODUCTION

Around 1960 the science of groundwater hydrogeochemical prospecting was still dominated by the search for uranium, but, as in earlier years, these methods were not accepted everywhere with equal enthusiasm. Therefore, in order that the advances of this period might be seen in the correct perspective, it may be as well to begin with a short summary of the reservations expressed or problems defined by contemporary authors. Hawkes (1959/705/), for example, remained cautious in his attitude to groundwater prospecting - mainly because of its relatively undistinguished track record - but conceded that "the analysis of ground water for the soluble weathering products of ores probably shows as much promise as any other geochemical method". Hawkes nevertheless devotes no more than eleven lines to the subject. In similar vein, Page (1958/707/) stated that "no significant discoveries of uranium deposits during the past three years can be attributed to hydrogeochemistry ..... However, this continues to be of interest as a potential method of finding new and major deposits .... but more precise delimitation of tectonic provinces and more sample data will be needed before ground-water sampling can be considered an important or economic reconnaissance tool". Everhart /729/, in his summary in 1958 of "Unsolved Problems in Uranium Geology" lists, *inter alia*, the lack of adequate data for uranium in some geographical or climatic situations, for instance in natural fresh tropical waters. It was his opinion that the hydrogeochemistry of uranium transportation was one of the more important subjects requiring concentrated additional research.

Within the next few years a great deal of additional work did go into solving some of these problems and an examination of the Literature reveals substantial theoretical and practical progress in the form of several important papers from this period. These will be examined in some detail in subsections 4.42 *et seq.*, which follow. A number of shorter or less significant items from this period, and some collected works, are summarized below:

(i) Abundances and distribution of uranium and other radioelements in groundwater and the application of these data to geochemical prospecting /738, 746, 764, 938-9, 1063/.

(ii) Statistical methods for the interpretation of uranium hydrogeochemical data /739/.

(iii) Summary of the abundances and distribution of U, Ra and other radioelements in oilfield waters of the U.S.S.R. /790/.

(iv) Important collected volumes: (First) U.N. Conference on the Peaceful Uses of Atomic Energy (1958/706/) and Heinrich (1958/699/).

4.42 URANIUM : THE WEST

Illsley *et al.* (1958/708/) were responsible for a short, powerful presentation of concepts of importance to the hydrogeochemical prospector. These authors worked mainly with samples of surface waters, but their observations are relevant to hydrogeochemistry in general. They comment as follows on the role of pH in the mobilization of chemical elements:

"Most chemical reactions in the zone of weathering occur in nearly neutral waters. The pH of surface films, however, is very different from the gross pH as measured in natural waters. The chemical reaction between water and granitic rocks may take place because the pH at the interface is relatively high (8 or 9); yet, upon dilution the gross pH is nearly neutral. If this film could be isolated in an undiluted state and the pH lowered, the dissolved solids would probably precipitate. However, in nature, this material remains in solution as a result of dilution and not because the measured pH indicates an effective solvent. Thus, conclusions drawn from natural--water pH values may be misleading without proper evaluation.

Pure water in contact with granite dissolves as much as 30 x 10 $^{-6}$ grams of U_3O_8 per liter. These solutions are stable although the pH is only 6.5. A possible explanation for the stability of such solutions is the presence of colloidal silica and ferric hydroxide. Colloidal materials that absorb uranium contribute to its presence in water as long as the colloids remain dispersed. Waters from semiarid regions commonly contain enough bicarbonate ion to stabilize uranium as a carbonate complex. Uranium present in waters containing abundant organic matter is kept in solution by the fulvic-acid fraction of the organic material.

Various processes combine to remove considerable amounts of uranium from solution. The soluble uranium complexes are readily absorbed on the hydroxide gels of iron, aluminum and manganese, and possibly on silica gel, since size and charge limitations are fulfilled by hexavalent uranium. This absorbed uranium is retained by these hydroxides during coagulation and precipitation and thus becomes enriched in sediments" (p. 126).

Amongst the practical aspects that were stressed were the use of polyethylene containers for contamination-free sampling, the importance of orientation before reconnaissance work and the need for better analytical sensitivity in reconnaissance work than in subsequent detailed work. The authors emphasized the need to characterize the bulk chemistry of the "specific type of waters in the region as a whole" and noted that, with respect to the major components, "greater value was placed on obtaining chemical correlation with uranium content than in obtaining empirical relationships with other trace elements" (p. 126). For purposes of interpreting hydrogeochemical uranium analyses, Illsley *et al.* used graphical profiling, isograd maps, frequency-distribution diagrams and methods of statistical evaluation that were based on the proposal by Ahrens (1954/709/) that elements in barren material follow a lognormal concentration distribution. Thus significant analytical values were identified by plotting, separately, the frequency distribution of the logarithms of the uranium/bicarbonate ratios and the logarithms of the uranium/specific conductance ratios (the specific conductance, K, being a good estimate of Total Dissolved Solids). These plots were in good agreement with the theoretical normal curve. For both plots the threshold was arbitrarily set at a point equal to the mean plus one and one half standard deviations, which eliminated thirteen out of every fourteen samples. The authors describe a typical example of a detailed investigation of one of the most anomalous values isolated by the abovementioned procedures. Water from the Schweitzer Creek contained 0,6 µg U/1 - twice background. The anomaly was traced back to a bog. An investigation of well waters in the area showed that groundwaters near the bog were anomalous, and there was a positive statistical correlation "between uranium concentration and organic material from uraniferous groundwater". The source of the uranium was thought to be in the local bedrock.

Gangloff *et al.* (1958/712/) stated that "until now it has always appeared that the direct search for uranium is preferable to the search for the other elements which go with it; in some cases, the determination of lead or of vanadium, for instance, brought no new information" (p. 143). Further: " ..... hydrogeochemistry, though empirical, brings a very substantial addition to conventional methods" (p. 143). These authors were interested mainly in surface waters (like so many other uranium hydrogeochemists at that time) but they added that springs "may provide interesting guides" (p. 147). The following additional points raised by Gangloff *et al.* are worthy of note: (i) Hydrogeochemistry is a powerful means of *preliminary* survey. (ii) Uranium hydrogeochemistry is strongly influenced by climate. (iii) Hydrogeochemical data can rarely be adequately contoured and some reference to local background becomes essential when different geological formations are encountered. Additionally, some kind of "correction coefficient" based on overall chemical composition of the waters must be applied to the raw uranium data.

Smith and Chandler (1958/714/) described in detail a dibenzoylmethane colorimetric method for the determination of uranium in natural waters in the field. They were concerned about the consequences of loss of uranium from solution as a result of natural phenomena or sampling procedures. It was their view that the most serious natural loss of uranium from most solutions was due to co-precipitation with ferric hydroxide or kaolin. Loss of dissolved uranium from samples stored in either glass or polyethylene bottles was negligible if the solution was acidified or if complex-forming anions were present.

The reader will be aware by now that there is no dearth of authors who have stated that a thorough knowledge of the regional distribution of hydrogeochemical background values for uranium is essential to the most effective utilization of the data of water testing surveys (e.g. /640, 707-8, 712, 716-7/). Scott and Barker (1958/715/) were the first to tackle this problem on a large scale when they presented a synthesis of uranium analyses of groundwaters from "most major and some minor aquifers throughout the United States". To facilitate the interpretation of the data, they divided the conterminous U.S.A. into ten "geotectonic regions" on the basis of tectonics, geology, physiography and groundwater provinces /643-4/. Contrary to the results of some more local studies (e.g. /712/), they found, on the broader scale, that there was no good correlation between either [U] or [Ra] and the concentration of any major components, but that the concentration of uranium was "somewhat higher" in bicarbonate waters. The concentrations of both U and Ra were approximately log-normally distributed in those regions for which sufficient data were available. There were considerable variations in abundances between regions, and the threshold of anomalous concentrations - set by them at the mean plus two standard deviations - varied from 2,8 to 45 μg/l.* Samples with concentrations above the threshold values "suggest local areas where uranium prospecting might be profitable". The authors also recommended consideration of the U/TDS ratio as a useful tool in those cases where interpretation is complicated by the influence of variable dilution.

Landis (1960/801/) undertook a comprehensive study of the uranium content of the groundwaters of the central U.S.A., with the object of delineating potentially uraniferous areas. His main conclusions are as follows: (i) The mobility of uranium is favoured by waters with a markedly

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* In their Table, p. 155, this is misprinted μμg/l, or picograms per litre, which would be equivalent to one part in 10^{15}.

non-neutral pH. (ii) The background value for the concentration of uranium is very clearly influenced by lithology. Mean values for the waters of various rock units range from less than 1,0 to 38 μg/1. (iii) Anomalies could be recognized by comparing the uranium content of an individual water to the range of values found in the waters of a given local area or lithological unit.

Landis did not attempt to analyse his data in terms of variables other than lithology, and none of the anomalies that were noted were followed up.

Phoenix (1960/898/) gave a broad general account of the chemistry of certain groundwaters in the Colorado Plateau region, which is one of the principle uranium provinces of the world.

Walton (1960/1091/) discussed geochemical exploration for uranium in the Rocky Mountains. Water sampling (springs etc.) was usually a better guide than the measurement of radioactivity because severe radioactive disequilibrium occurs in this area. However, the hydrogeochemical method was not one hundred percent effective either.

Hecht *et al.* (1958/716/) adopted an interesting approach to the interpretation of the uranium content of the many Austrian spring and other subterranean waters that they analysed. Not only did they contrast the spring waters of tectonically different areas, but they were able to declare some areas to be anomalous by comparison of the groundwater data with the uranium content of the Danube. Many uraniferous springs were associated with major Alpine fault lines and the Alpine sedimentary formations generally yielded higher values than the associated crystalline rocks.

Chervet and Coulomb (1958/723/) were responsible for important experimental and field work on the mobilization, migration and fixation of uranium. They found that under suitable conditions - for instance where pyrite and free oxygen are abundant - the destruction of uranium minerals can be very rapid. A test sample of 10 g of weathering pitchblende yielded 142 mg of mobilized uranium in 20 days. Such conditions prevail in many (French?) uranium mines, where the authors detected between 0,5 and 90 mg U per litre in mine waters. Their experiments with ion exchangers showed that at pH 1,5 to 4 the uranium from ores dissolved into water under laboratory conditions exists as $UO_2(SO_4)_2^=$, $UO_2(SO_4)_3^{4-}$ and UO_2^{++} in the proportion 2:2:1. At pH values somewhat above 4 UO_2^{++} and $UO_2(OH)^+$ predominate. If enough carbonate ion is present, then $UO_2(CO_3)_2^=$ will exist from pH 4,5 to 6,5 and $UO_2(CO_3)_3^{4-}$ up to pH 11. Although uranium phosphate complexes may also be important in some instances, the authors conclude that any significant migration of uranium in near-neutral natural waters must be in the form of carbonate complexes and that therefore "the long range circulation of uranium is controlled by the carbonate concentration of the waters ....." In their supplementary field work the authors were not particularly concerned with the distinction between surface and sub-surface waters. They found the "normal" range of [U] in uraniferous provinces to be from 0,1 to 1000 µg/l, with hidden mineralization indicated by values of 1 - 20 mg/l spread over several kilometres (a high concentration by worldwide standards). Total analyses of uranium-rich well-water from Verneix revealed that the abundance of uranium (2 to 1242 µg/l) was "related to the amount of bicarbonate" in solution and to TDS, but not to any other individual major components. According to the authors, the important processes that immobilize uranium are as follows: (i) evaporation. (ii) precipitation as insoluble phosphate (autunite), commonly as a result of indirect reaction with apatite:

$2CaHPO_4 + 3CO_2 + 3H_2O + 2Na_2(UO_2)(CO_3)_3 \rightarrow Ca(UO_2)_2(PO_4)_2 + 8NaHCO_3 + CaCO_3$ (iii) formation of uranium silicates and oxides, especially when $[HCO_3^-]$ is low. (iv) encounter with reducing substances such as organic matter or H_2S (cf. Miller /724/). (v) fixation on natural anionic or cationic exchangers such as clays and humus. These reactions are generally reversible, at least to a degree. (vi) accumulation by plants.

Mingarro and Catalina (1958/725) used optical spectrographic techniques to determine Cu, V, Fe, Ag, Ca, Na, Mg, K, Si, Al and U (the latter by the method of Gerlach and Riedl /664/) in waters from Spanish wells, springs, mines and other sources. They found that the type of ore lode responsible for a hydrogeochemical uranium anomaly could be identified by studying the association of V, Fe, Cu, Ag and U. Threshold values (three times background) were established at 1 mg/l for U and 0,01 mg/l for silver, and samples anomalous in either of these elements were plotted on a ternary Fe - Cu(x10) - V(x100) diagram. Certain regularities were noted; thus samples anomalous in both U and Ag always contained significant amounts of each of V, Fe and Cu. Changes in the lithology of the formations yielding the water samples were clearly reflected in the diagram. The authors were careful to stress that these phenomena may be no more than a local "geochemical characteristic of the region" studied.

Dall'Aglio and Tonani (Italy; 1962/891/) demonstrated that the concentration of uranium in natural waters generally increases linearly with TDS. This suggests that under most conditions the rate of dissolution of uranium proceeds at about the same rate as the dissolution of the other major mineral components of rocks.

A summary of the available information on the leaching and migration of radioelements normally associated with uranium was given by Adler

(1958/667/). He noted that the dearth of Pb and Ra in secondary uranium deposits corresponded well with the tendency of these two elements to stay behind and concentrate in oxidizing primary deposits. Lead is very insoluble in carbonate/sulphate solutions, which are possibly the most frequent transporters* of uranium, and Pb is also readily absorbed from groundwaters by iron and manganese oxides in fine grained sedimentary materials. However, small amounts of barite containing Ra may be deposited from groundwaters along with secondary uranium minerals. Although the small amounts of Pb and Ra transported with U are generally sufficient for very careful isotopic studies that may have both theoretical and economic significance, they are nevertheless inadequate as hydrogeochemical pathfinders.

Isotopic studies by Rosholt /669/ tended to confirm that the use of elements such as radium fell more naturally into the realm of the geophysicist, and that physical (radiometric) rather than chemical measurements of such radionuclides in spring and groundwaters are of greater interest to the prospector.

Woodmansee (1958/730/), following earlier workers /636, 647, 681-2, 675-7, 679/, presented the results of a detailed investigation of the relationship between the groundwater regime and some uranium deposits of the U.S.A. He states that "Many uranium deposits in clastic sedimentary host rocks are associated with present-day water tables or are considered to have been related to past groundwater conditions". If one accepts that these "deposits were formed and are forming epigenetically by the precipitation and concentration of uranium from groundwater solutions", then

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* Miller (1958/944/) showed that under certain circumstances U is transported as a sulphato- complex.

"directions of migration and positions of water tables, both past and present, are important in the search for ore deposits in uranium-mining districts". However, "such ..... investigations are difficult or impracticable in many districts ....." and by 1958 little attention had been given to the problems involved. Woodmansee summarizes the available information in respect of orebodies related to present day water tables (from near surface to very deep) in several locations in the U.S.A. He notes that such studies can have an immediate and considerable practical importance, since "drilling to zones where favourable formations are intersected by water tables at depths of 200 to 800 feet has produced highly favourable results in exploration for uranium" in certain strata. This association of uranium ore with the water table may be the result of supergene/lateral enrichment processes following the relatively rapid destruction of uranium minerals in the presence of free oxygen and vadose water, the uranium being redeposited in the relatively reducing environment below the water table. (The redox potential for $U^{4+} \rightarrow U^{6+}$ is 0,41v /737/). This relationship is well illustrated, for example, in the Maybell district of Colorado, where the location and degree of oxidation of the uranium is clearly related to the present water table. In addition, "pre-existing water tables are indicated by the presence of uranium, carbonate and iron-oxide mineral concentrations in horizontal layers, transverse to dip and crossbedding in the host sandstone". Similar "magic contours" defining the location of uranium ore deposits have been noted in several uranium-petroleum districts of the U.S.A. (for example San Rafael Swell, Utah) and are ascribed to paleo-oil/water or gas/water interfaces /731-2/. However, not all uranium deposits in petroliferous areas are directly related to the petroleum deposits, since reducing oilfield waters often migrate for some distance before encountering and mingling with oxidizing, uranium-rich solutions.

Woodmansee also points out that it may be profitable to pay attention to the details of past or present lateral movement of groundwater via paleochannels, etc., since these may have some control over the eventual location of secondary accumulation of uranium minerals.

The first major hydrogeochemical study of uranium in Sweden was carried out by Armands and Landergren in 1958-1960 and was reported by them in 1960 and 1967 (/700-1/). These authors still regarded this work as something of a novelty: "In addition to the more coventional methods used in prospecting for uranium, geochemical investigations were also carried out in the region ....." They analysed soil, vegetation and water from bog deposits north of the Arctic Circle. The local geology is complex and includes granites, mafics, sedimentary rocks, ironstones and quaternary moraine deposits. Uranium was determined in the water by fluorimetry, in conjunction, at times, with ion-exchange concentration. Radon, pH and Eh were also measured, together with total hardness, specific conductance and, in some samples, total major ion abundances. TDS was typically around 150 mg/l, Ca^{++} up to 16 and bicarbonate ion up to 94 mg/l, with pH values of around 7,7. In this wet climate the source of the water sample had a very important bearing on uranium prospecting. Samples could be obtained from seepages, drill holes, reservoirs, streams, rivers, wells and springs, but the last two of these sources were easily the most important for prospecting purposes. The most highly radioactive waters that they encountered were emerging from the bedrock via springs or drill holes and contained up to 1,8 mg U per litre. However, many samples of water extracted from soils upslope of the bog contained only 2-3 µg/l, and the authors observed that "..... the mineralogical composition of the surface soils has a decisive (negative) effect on the content of metal cations in the groundwater ....". Water from the bog itself contained considerably more uranium, often up to 100 μg/l and sometimes more.

Some 250 springs occur in the area and the water issuing from them "partially consists of intra-bedrock water". Apparently most of these springs were sampled and analysed for U and Rn. There was a marked general elevation of [U] and/or [Rn] in "springs .... located in topographically marked depressions which are to some extent due to contact zones between rocks, or to fracture and fissure zones in the bedrock ...." However, there was no general relationship between the concentrations of radon and uranium and although both elements proved to be good pathfinders, the latter was apparently superior in this respect. Like several other investigators, Armands and Landergren found that the presence of bicarbonate was essential for the mobilization of uranium, and they considered the mobile species to be uranyl bicarbonate. Table 1 is a summary of the uranium data for the spring waters. These contrast sharply with analyses of waters from a background area (Table 2).

Several other elements were sought in these spring waters by spectrographic methods, with detection limits of 0,05 to 0,3 mg/l. The important ones were Sr, Ba, Fe, Ni, Cu, Mn, Pb, and P but none of these proved to be a useful auxiliary pathfinder.

The authors were also interested in what occurred when the spring ("intrabedrock") waters mixed with relatively acidic manganiferous peat waters of the bogs. It was found that several radioelements were commonly co-precipitated along with hydroxides and oxides of Mn, but not the uranium, which survived for a little longer and then became fixed by the peat itself. This was wholly in agreement with the earlier findings of Szalay (1951-1958 /582, 703, 718-721/) and others /722, 543, 593/ who had shown clearly that

TABLE 1/700/

Distribution of uranium in spring water from the Masugnsbyn area

Concentration range (μg U/l)	Average content of uranium (μg/l)	Number of springs	Percentage distribution of springs
>100	730	4	1,8
100-50	74	3	1,3
50-25	39	15	6,7
25-15	17	14	6,2
<15	6[a]	190	84,0

[a] Background value

TABLE 2/700/

Uranium content, pH, and specific conductivity of water from central Sweden

Kind of water	Average content of uranium (µg/1)	pH range	Specific conductivity (10^{-6} mho/cm)
Surface water from non-radioactive regions	<1,0	7,3-4,5	31,7
Surface water from radioactive regions	3,0	7,0-6,7	31,7
Water from iron ore mines[a] (non-radioactive)	0,5-2,0	7,7-6,4	about 100
Spring water	3,5	7,6-6,2	41

[a] Water from mines containing uranium minerals may have contents >2000 µg U/1, e.g. the Stripa Mine in central Sweden.

peat and certain other biological materials were capable, through reversible ion-exchange processes, of geochemical enrichments by factors of 10^4 with respect to dilute solutions of uranyl ions. In this way the Swedish peat bog had served as a collector for uranium and the ore in the bog was formed by a continuous, slow removal of this element from the intrabedrock water.

The authors showed in leaching experiments using local waters and crushed local rocks that leaching by groundwater could account for all of the uranium present in the bog ores. However, they recognized the possibility that some of the uranium may have come from an unknown local uranium orebody, probably pegmatitic. They did not proceed to a search for this potential lode, but concluded that "a thorough investigation of the water systems, including their chemistry, origin, flow and drainage patterns, are most important in the search for the uranium source" (p. 153).

Keys and Dodd (1958/735/) summarized the pertinent information available at that time on the hydrochemical movement of uranium through sedimentary rocks. They noted that for any long-range transportation of uranium to occur the transporting solutions would have to be in fairly good chemical equilibrium with the country rocks. Citing earlier works /568, 575, 683, 736/, they stressed the role of Fe^{++} and clays in the diminution of [U] in natural waters. Clays may immobilize U^{6+} through base exchange, absorption, or through the "reductive capacity of trapped ions in the clay" (p. 377).

Peacock (1961/834/) summarized hydrogeochemical data (surface and underground water) accumulated during the assessment of uranium mineral resources in Great Britain (1956-1958), and gives references to several papers dealing with colorimetric and fluorimetric analytical methods. The average background value in waters from various regions was 1 μg/l or less.

Higher background values were noted in dolomitic areas and this was attributed to a combination of high [HCO_3^-] and elevated concentrations of uraniferous phosphates in the limestone. It is interesting to note that in at least one anomalous water (50 µg/l) the greater part of the mass of uranium was in suspension, and only about 1 µg/l was in true solution. Waters from a disused uranium mine contained 30-50 µg/ U/l, but the concentration of U in thermal waters was not in any way peculiar. In some areas [U] was strongly dependant upon the "weather conditions immediately preceding the collection of samples". The concentration of uranium in natural waters shows a sharp positive pulse after rain that follows a dry spell. This phenomenon was attributed to the leaching of accumulated uranium salts that had been released during the dry season by the slow oxidation of uranium minerals. At Saint Annes Well the concentration of uranium in groundwaters varied from 2,5 to 6,0 µg/l during a period of several weeks, and this was attributed to climatic factors that resulted in "varying ground-water circulation".

4.43 URANIUM : THE EAST

A number of accounts of hydrogeochemical exploration for uranium in the U.S.S.R. between 1958 and 1962 are available in English.

Serebrennikov (1959/1092/) claimed that several uranium deposits have been discovered by the hydrogeochemical method in the Soviet Union. He reported that [U] in groundwater was significantly affected by seasonal changes in areas of unreliable rainfall. The observed background values varied by a factor of 2-3, but in anomalous waters this seasonal variation was as high as tenfold. Under arid conditions the "zone of infiltration" contains only capillary water for much of the year, and this water tends to become relatively salty as a result of slow chemical reaction with contiguous minerals. If uranium ore is present, this metal will be strongly concentrated in the capillary water, which will thus produce a local "pulse" in [U] when it is periodically washed down and diluted. If the uranium ore is associated with sulphides, then the concentration of sulphate ion will often show the same exaggerated seasonal variation.

Germanov *et al.* (1958/717/) communicated a powerful summary of the state of the art in the U.S.S.R. at that time. Uranium, Ra, Rn, major components and "gas content" were routinely determined in water samples, and particular attention was paid to pO_2 and Eh. Special equipment was needed to get good results for the last two of these measurements, and they were made *in situ* wherever possible. The authors believed that redox conditions "are one of the main agents determining behaviour of uranium in underground solutions" and noted the strong influence of dissolved gases and micro-organisms on Eh. Like their Western colleagues, they were convinced that uranium could not be examined as "an isolated compoment" and they emphasized the need for "close co-ordination" of hydrogeochemical and hydrological data with those of ancillary disciplines such as mineralogy, geomorphology, tectonics and climatology.

Following Ignatovich /645/, Germanov *et al.* conclude that the distribution of dissolved uranium is not uniform through the *depth* of the lithosphere because of the influence of oxygen in the atmosphere. In prospecting, therefore, it is essential to distinguish between shallower, oxygenated waters and reducing waters ascending from deeper levels, as the respective uranium contents of these waters will not be immediately comparable. It is stressed that "shallow" and "deep" waters are relative concepts, the definition of which will vary with the geological history and geotectonics of each region. For example, "water exchange" is more intense in mountainous areas than in plains regions and the depth of penetration of free oxygen is as a consequence higher in the former case (up to 3 km deep). In regions that are relatively rich in organic matter - such as sedimentary basins, oilfields and coal-bearing provinces - there will be a sharp borderline between the two types of waters at moderate depths. In igneous rocks and other formations low in organic content the groundwater may be quite well oxygenated far below the water table.

All else being equal, the shallower waters, especially those poor in organic matter, will normally carry more uranium. If oxygen is freely available, Eh may be around +300 mV and intense mobilization of U can take place as U^{4+} is oxidized to U^{6+}. Some of this uranium is soon reprecipitated as phosphates, silicates, etc., but part of it survives in solution and is carried away from the orebody. As a result, [U] in shallower waters is (*inter alia*) a function of [U] in the local rocks. The authors show that plots of [U] in rocks vs. [U] in formational waters, for various rock types, yield straight line plots with slopes of around 45°. Naturally, background and threshold values will vary with the "peculiarities of each region".

In deeper waters, where the equivalent of tens or even hundreds of mg/l of organic matter may be present, Eh may vary from -100 to -500 mv and the concentration of uranium is typically lowered to $n \times 10^{-4}$ mg/l or less, irrespective of whether the formation that is host to these waters is uraniferous or not. Here the authors note an exception in the case of waters enriched in H_2S, which may have relatively high [U] irrespective of Eh. Interestingly, the reverse situation holds for radium, which is strongly enriched in waters of the "deeper" type. This is partly due to a suppression at these depths of the adsorption/precipitation of radium on hydroxide, a mechanism known to strip the element from shallower waters. It follows that radium comes into its own as an important pathfinder for fissile materials when reducing or organic-rich waters are encountered.

Germanov *et al.* state that experience throughout the vast territory of the U.S.S.R. has shown that the uranium background level of subterranean waters is strongly dependant on climate. As a first approximation, [U] is directly proportional to the degree of "mineralization" of the water (i.e. TDS), which is in turn inversely proportional to the mean annual rainfall. Thus arid areas are likely to have higher threshold values for uranium than humid regions. Variations in local climate may be effected by changes in latitude, longitude or altitude. In the Tien Shan Ranges, for example, the authors found that the uranium content of groundwaters decreased with altitude, but that for any given zone of comparable rock type the "modulus of underground runoff" (litre/sec/km^2) x aqueous [U] is approximately constant, and represents the "modulus of chemical runoff" (g/year/km^2). This latter modulus is very useful for the identification of uraniferous formations or areas.

Using data from the northern Tien Shan, they show that various climatic/altitudinal zones are also readily distinguishable on plots of [U] vs. TDS, in which the data from different zones separate out simply into discrete groups. The "amount" (ratio) of uranium in the dried residues of these various water types varied from about 1,5 x 10^{-4} to 4 x 10^{-3}%, irrespective of the absolute values of the numerator ([U]) or the denominator ([TDS]), even when the latter reached 10 000 mg/l. Above this value, in the special case of extremely saline waters formed by intensive evaporation, the proportionate concentration of uranium tends to "lag behind the growth of dry residues", even though absolute uranium values may be high. On the other hand, saline waters formed by the leaching of salt bearing rocks (evaporites) do not usually contain appreciable amounts of uranium (n x 10^{-6} g/l).

An interesting and important observation reported by Germanov and his colleagues is the fact that climatic factors seem to have a stronger influence on some anomalous groundwaters than on waters with no more than background concentrations of uranium. In barren springs that they studied [U] varied by a factor of two over a period of twelve months, whereas in anomalous spring waters the difference was often tenfold. This was probably a local phenomenon related to periodic "flushing" of uranium dissolved out of weathering uranium minerals situated above the water table. This hypothesis also accounts for the sympathetic rise of [$SO_4^=$], [U] and the yield (l/sec) of the spring. The authors point out that the solubility of atmospheric oxygen in water is only about 12 ppm and cannot produce, by oxidation, more than 15 mg/l of sulphate in phreatic waters. Therefore any sharp upward pulse in [$SO_4^=$] indicates the influx of vadose water to the spring.

Other factors that jointly or severally influence the rate of discharge of uranium and commonly associated elements (Pb, P, Si and As) from the orebody, and hence influence the intensity of the aqueous halo, are the mineralogy and permeability of the orebody and the host rocks, and the position of the lode relative to the water table. Thus fractured ores dissolve more readily than massive ores, secondary uranium minerals resist dissolution better than primary ones, and the most favourable situation for massive mobilization of uranium from an orebody is just above the saturated zone of the water table.

Hydrogen ion concentration is important too, but the effect of pH is in turn dependant on other factors. For example, fluctuations in pH have a much stronger effect on [U] in waters of the "leakage zone" (presumably shallow, vadose water) than on phreatic waters. Furthermore, pH influences the rate of dissolution of some types of ore more than others, although not always in a rational manner. Uranium is soluble to some extent throughout the pH range below 8, but [U] will rise very rapidly if the pH of the water of the orebody falls below 3. These extreme conditions may be approached during the oxidation of uranium ore rich in pyrite, under which circumstances the pH of adjacent waters will often drop to less than 4. Uranium then becomes extremely mobile and will usually be dissipated from the vicinity of the orebody by groundwaters without the formation of secondary minerals. If the leaching has been particularly intense and complete, then these orebodies may be quite difficult to locate. The authors have noted cases where continuous movement of groundwater through a weathering body of rock has leached out practically all the uranium, and as a result the formation waters now contain very little of the metal.

If the ore is somewhat less pyritic, or if more carbonates are present to allow some neutralization of the acidic waters of oxidation, then secondary phosphates, arsenates, vandates etc. of uranium may form nearby. These secondary minerals, if traced by hydrogeochemical methods, may point to the presence of a mother lode. Germanov and his co-workers have found that the amount and nature of this secondary mineralization and/or uranium dispersion is highly dependant on the interaction of a large number of local chemical and physical factors, for example pO_2, the presence or absence of H_2S, hydrocarbons, oxides and hydroxides of iron and manganese and the influence of "pumping"* of the water table.

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* The word "pumping" is apparently intended to mean natural cyclic changes in the level of the water table, and not artificial changes due to the activities of man.

With respect to the mobility of uranium beyond the immediate vicinity of the orebody, Germanov *et al.* claim that "the chemical composition of underground waters .... as regards the usually determined six components - Na, Ca, Mg, Cl^-, HCO_3^-, $SO_4^=$-does not exert any essential influence on uranium migration". The reader will recall from the foregoing that Western hydrogeochemists, barring a few dissenters such as Scott and Barker /715/, would take exception to this claim.

Germanov *et al.* provide a unique example demonstrating that dilution/adsorption effects may operate in underground waters in a manner similar to that in stream waters. They describe a sloping aquifer of alluvial sediments on the floor of a dry valley, with springs at the head of the valley that carry 1 µg U/l and feed the aquifer. Water from this aquifer becomes systematically poorer in uranium further down the valley. In another example, they describe the sampling of seepage mine water from a drive that was being developed towards a known uranium lode. As the drive advanced the uranium content of the waters increased because the nett effect of anomalous waters derived from the orebody began to outweigh the effect of barren waters entering the drive from the country rock on either side.

Grammakov *et al.* (1958/764/), expanding on some aspects of the earlier work of Germanov (1956/647/), found definite general relationships between, on the one hand, ions released into the groundwater system and, on the other, the mineralogy and depth of uranium ore lodes and the position of these relative to the water table. Vadose water associated with Soviet uranium deposits always contains important amounts of UO_2^{++} (sic)* and, depending on the ore mineralogy, may also contain considerable

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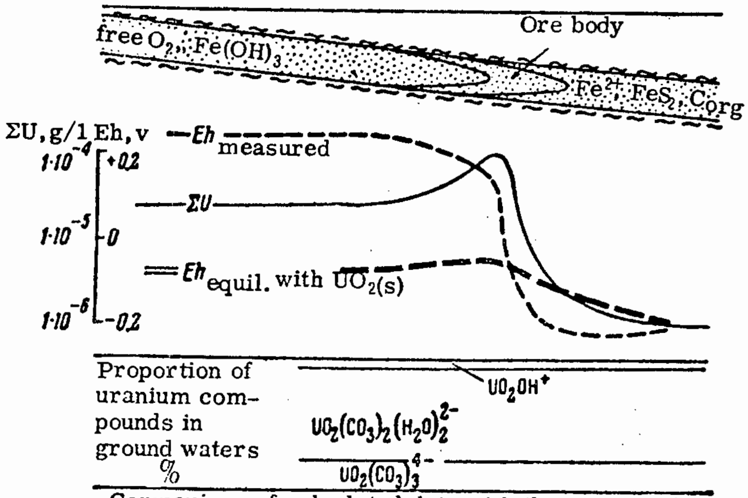
* It is doubtful that the authors literally mean the species UO_2^{++} *per se*. i.e., read as "uranium".

concentrations of OH^-, HCO_3^-, $CO_3^=$, $SiO_3^=$ and $SO_4^=$. Shallow phreatic waters always contain UO_2^{++}, bicarbonate and carbonate and sometimes sulphate. Deeper phreatic waters always contain UO_2^{++}, bicarbonate and carbonate, and may in addition contain significant amounts of OH^- and sulphate ion.

Lisitsin (1962/799/) pointed out that the determination of uranium speciation in groundwaters was a difficult problem but that it was possible to calculate the theoretical proportions in which uranium compounds may be present in water in contact with UO_2 at different values of Eh and pH. The calculations suggest that U^{6+} carbonate species are favoured above sulphate or chloride complexes even in Cl^- and $SO_4^=$ rich waters. For example, at pH 6,6, Eh -0,125v, 20°C and TDS not exceeding 6 g/l, uranium carbonate species constitute practically 100% of the total mass of dissolved uranium, and all other species together account for only about 10^{-10}% of the total. There are extremely few natural waters in which more than one of every 10 000 ions of dissolved uranium is U^{4+}. For the reaction

$$UO_2 + 2CO_3^{2-} + 2H_2O \leftrightarrow UO_2(CO_3)_2(H_2O)_2^{2-} + 2e^- \quad (25^\circ C,\ 1\ \text{atmosphere})$$

the standard potential ought to be -0,13 mv. In actual geological situations it was determined as -0,125v. Theory suggests that the Eh of precipitation of UO_2 is a function of the major component composition of water and of pH and [U]. This potential typically ranges from 0 to -,2v. Lisitsin makes the interesting observation that uranium orebodies, if they are still actively growing, may deplete rather than enhance the uranium content of the groundwaters passing through them. This is most likely to occur when the uranium lode constitutes a redox "front" at which a marked drop in Eh takes place. This concept is illustrated in the following diagram:



Comparison of calculated data with the results of hydrogeochemical investigations (generalized outline).

Rozhkova *et al.* /746/ observed that phosphates, especially calcium phosphates, might be an important immobilizer of aqueous uranium. Their data are as follows:

calcium phosphate	absorbs	0,244 meq UO_2^{++} /g	at a pH of about	4.
apatite	"	0,029	"	4.
ferric hydroxide	"	0,286	"	2 to 3.

These values were scarcely affected by changes in the concentrations of K, Na, Ca and Mg. Tests showed that the absorption of dissolved uranium was a function of the specific surface area of the materials in contact with the solution, as well as a direct result of the chemical affinity.

Shcherbina /726/ carefully examined the complex geochemical processes which occur in oxidizing uranium ore bodies. His results confirmed that although the reactions that take place, and the intermediate minerals that may form, are highly dependant on local factors such as pH, lithology and climate, any meaningful migration of uranium will be in the form of carbonate complexes. Shcherbina also observed that certain ions such as Cu^{++} can limit the mobility of uranium by competing for the CO_2 in the uranocarbonate complex.

A handful of less important Russian publications dealt with experimental studies of uranium dissolution and transportation /740, 746, 749/.

Barabas and Kiss (1958/737/) were the authors of an interesting attempt to reconstruct the conditions under which uranium was precipitated from natural waters to form the ore horizons of the Mecsek Mountains in Hungary. This involved the construction of a model relating chemical compositions, O/Fe ratios and the Eh-pH fields for various minerals present. It is doubtful whether this approach is of any direct use to the prospector, but it does suggest the potential practical application of quantitative models that relate [U] to variations in pH-Eh conditions in natural waters.

Field and laboratory studies in Hungary by Szalay /703/ corroborated previous conclusions /717/ about the sensitivity of uranium to certain organic compounds. Szalay found that background values for [U] in Hungarian natural waters was 1-2 μg/l, with exceptional anomalies of 100 μg/l. In the absence of oxygen and in the pH range 3-6, humic substances will, within seconds, reduce the uranium content of such waters to almost nothing. It follows that the migration of uranium in organic-rich sedimentary materials may be extremely limited.

Murakami *et al.* (1958/640/) reported the results of the first major systematic hydrogeochemical search for uranium in Japan. They analysed many stream waters and some mine waters in order to identify pathfinders for a specific type of uraniferous conglomerate typified by the Ningyo-Toge ore deposit (a phosphatic/sulphidic uranium ore). The detection limits were: U 0,1 µg/l (fluorimetry); PO_4^{3-} 0,01 µg/l (Mo blue) and Zn 0,1 µg/l (dithizone). The "statistical indication" that they used to distinguish between "barren, promising and most promising" streams was:

> (number of samples from that stream with concentrations greater than background)/(total number of samples from that stream).

"The results of direct analysis of the uranium content of rivulet water coincided completely with those of indirect analysis of zinc and phosphate, and, moreover, the statistical interpretation of the concentrations of the three elements in rivulet water was found to be very useful for the determination of the location of ore deposits ....." (p. 132). This paper includes a summary of the uranium contents of various selected natural fresh waters of Japan. Hot springs yielded values of around 0,2 µg U/l and the concentration in most other waters was about 1 µg/l. The authors emphasized that regional differences in background would be of vital concern in reconnaissance surveys, and that peak-to-background ratios were far more important than absolute concentrations. They noted that climate and weather strongly influence the concentration of uranium in stream waters.

4.44 NON-RADIOACTIVE MINERALS : THE WEST

There are few outstanding or original Western publications in this category from the years 1958-1962. The standard reference work for the period is the textbook by Hawkes and Webb (1962/49/).

Boyle /1127-8/ presented a useful summary of all of the earlier investigations (e.g. /527, 819/ of spring water anomalies near lead-zinc deposits in the permafrost regions of Yukon Territory, Canada.

Sato (1960/1061/ and Sato and Mooney (1960/769/) gave excellent accounts of the importance of Eh and pH in groundwater chemistry and the effect that these two parameters have on the mobilization of elements at various depths in sulphidic orebodies. It is rather difficult to get good experimental data for redox potentials of natural waters, because even in deep mines the Eh measurements are often biased by oxygen contamination. Much of the reported experiment data was collected in copper mines in the U.S.A. In zones of primary ore the mine waters have typical pH and Eh ranges of 5 to 9 and +0,1 to +0,3v, but in the oxidized zones the corresponding ranges are 2 to 8,5 and +0,2 to +0,7v. A plot of all observed values on an Eh-pH diagram revealed a strong trend parallel to but below the line of the equation for the reaction

$$2H_2O = O_2 + 4H^+ + 4e^-, \ Eh = 1,219 - 0,051 \ pH,$$

which defines the upper boundary of the stability field for water. In practice this boundary is never approached by natural waters because in the presence of O_2 the Eh is powerfully influenced by the redox couple $(H_2O_2 - O_2)$:

$$H_2O_2 = O_2 + 2H^+ + 2e^-, \ Eh = 0,682 - 0,0591 \ pH.$$

Only a small amount of free oxygen is necessary to maintain oxidizing conditions in subterranean waters. The authors also discussed some natural pH buffers and note that extremely low pH values in mine waters are prevented by the reaction

$$Fe(OH)_3 + 3H^+ \leftrightarrow Fe^{3+} + 3H_2O.$$

Like the Soviet prospectors, the Americans began to conceive of the groundwater anomaly as simply a special conditon within the slowly evolving subterranean hydrogeochemical cycle. Seaber (1960/862/) used the term "hydrochemical facies" to describe "regional relations between chemical content of ground water, lithology, topography, and flow patterns .....". Systematic changes occur as the water migrates and ages (cf. Chebotarev 1955/1048-50/) and TDS increases as the water moves downwards and laterally from the point of recharge. One of the commonest features of the metamorphism of groundwater is the accumulation of sodium ions and this appears to be due largely to the ion-exchange capacity of clay minerals, expecially montmorillonite. Wyrick (1960/863/) stated that, in addition to commonly used hydrological techniques such as water level contour maps, "chemical data from readily available water samples may be used ..... as a means of defining ground-water flow patterns". In Martin County, North Carolina, he used pH and Eh directly to "separate areas of ground-water recharge". In Volusia County, Florida, he found that the chloride content of water samples was an excellent index of flow patterns in groundwater. The potential importance of these concepts to hydrogeochemical exploration is self-evident, but considerable practical difficulties may be involved in applying them and there have been very few attempts to pursue the idea (See Section 4.5).

Brown and Floyd (1960/861/) found that the chemical composition of groundwaters in the Coastal Plain of North Carolina reflected the chemical nature of the host rocks but that this relationship was frequently complicated by processes of rock decomposition and metamorphic changes that occurred in the groundwater as it aged. "The minor elements in natural ground waters can be used to trace ground-water flow, to define geohydrologic environments, and to indicate the presence of mineral deposits of potential economic value". The elements Li, B, Br, F and I were said to be particularly useful in this respect.

Erikson and Marranzino (1961/1097/) described spring water anomalies from Eureka County in Nevada. Near the head of the Eureka Canyon the waters are acidic/sulphatic, with up to 300 μg/1 of (Zn + Cu + Pb). But at the mouth of the Canyon the waters are of the alkaline/bicarbonate type and here [Mo] had risen very significantly, to as much as 60 μg/1.

Ward *et al.* (1960/1082/) demonstrated that molybdenum anomalies in groundwater could be severely displaced from the source of the metal, sometimes by several miles.

Brown (1958/1098/) showed that it is possible to use groundwater hydrogeochemistry to prospect for deposits of ore-grade phosphatic sands intercalated with limestone bands. Bromine and iodine were suitable pathfinders.

Tooms and Webb (1961/772/) sampled groundwaters in the Zambian copper belt, where primary minerals have been destroyed to a great depth by intense tropical weathering. They concluded that the anomalies that they found must have been due to the remobilization of secondary minerals.

Additional publications of some interest dealt with (i) The relationship between ore deposits and either manganese /1058/ or strontium /1059/ in spring waters, and (ii) The occurrence of Ba, B, Br and F in groundwaters as a guide to deposits of the minerals of these elements /1060/.

The contamination of individual water samples has always been a problem in groundwater hydrogeochemistry, but prior to about 1960 there are few accounts of problems resulting from the wholesale pollution of regional groundwaters. This happy state of affairs has ended and the prospector can no longer always assume that widely distributed chemical species in groundwater have a natural origin. Anthropogenic components are becoming increasingly common in many agricultural and industrial areas. Krieger and Hendrickson (1960/864/), for instance, showed that chloride-rich waste water, which was discarded on the surface near oilfields in Kentucky, penetrated back into the ground and contaminated groundwaters as much as 100 miles away.

Workers such as Garrells (1960/770/), Silman (1958/771/) and others /773/ continued with the complex task of constructing quantitative models that could describe and predict the mode of occurrence and mechanisms of transportation of metals in groundwaters. A number of papers dealt with the transportation of copper and cobalt in subterranean waters and the circumstances under which they are immobilized by organic matter or clay minerals /765-7/. Hawkes and Webb /49/ noted that "precipitation of insoluble compounds in areas where natural waters pass from one chemical environment into another tends to remove metal from solution, and thus cause an accelerated decay of hydrochemical anomalies. Updrainage from a precipitation barrier of this kind, hydromorphic anomalies show up more clearly in the water. Below the barrier, they are stronger in the clastic material with which the water is in contact. Within the area where precipitation is actively taking place, anomalous values arising from material precipitated in the clastic matrix will be at a maximum".

An important handbook of sampling and analytical techniques in hydrogeochemistry was prepared by Rainwater and Thatcher (1960/210/). Another notable contribution was the use of ion-exchange resins for preconcentrating hydrogeochemical samples in the field (Canney and Hawkins (1960/1066/).

4.45 NON-RADIOACTIVE MINERALS : THE EAST

Belyakova (1958/783/) investigated the migration of elements from the Cu-Pb-Ba-Mn-W deposits of Upper Kairakty, in central Kazakhstan.

Here the strong relief and low permeability of the schistose country rocks favour an "active water exchange", so that TDS in the groundwaters is typically less than 500 mg/l. Most of the groundwaters occurring below the regolithic layer are of either the calcium bicarbonate or sodium bicarbonate types. Around the orebodies the relationships between the major ions changes in a complex manner, with magnesium, sulphate and chloride becoming important constituents in the waters adjacent to the ore. These changes can generally be related to changes in bedrock geology but the increased abundance of sulphate ion is apparently due to the oxidation of the sulphidic ore. The range of pH values in these waters is typically 5,8 to 7,8. Interestingly, the waters of alluvial sediments in the same area are "highly mineralized saline" (TDS from 1 000 to 3 000 mg/l), with sodium, sulphate and chloride ions predominating. Belyakova ascribes this marked change in water type to evaporation.

Unlike many other Soviet hydrogeochemists (e.g. /345, 351, 784/), Belyakova was cautious about the use of elevated sulphate/chloride ratios as an indicator of mineralization. He in fact found that the ratio dropped significantly as one approached some orebodies at Upper Kairakty. Many trace elements were determined in these groundwaters, mainly by spectrographic methods. The dried residues of the waters contained detectable amounts of Cu, Pb, V, Ni, Co, Cr, Ti, Mn, Sr, La, Zr, Cd, Bi, Ga, Ag, Sb, Li, Ba, Zn, Be, W and occasionally As, Sn and Mo. "The metals found in waters representing different lithologic-stratigraphic complexes reflect the ores of the aquifer rocks. Qualitatively, the metallogenic fields ..... are clearly outlined in the district by the complex of the metals they contain" (p. 181). The Kairakty lead-copper-barite deposit, for example, is accompanied by a relatively uncomplicated Pb-Cu-Ba groundwater anomaly. This anomaly is rather large in comparison to the size of the deposit and has a width of at least 8 km. Belyakova implies that it is not always an advantage to have a very broad an intense anomaly associated with a small deposit, since this may "mask" similar orebodies in the immediate vicinity.

The waters of the Upper Kairakty tungsten deposit were investigated in detail and compared to the waters that are typical of the region. The associations that were noted are compiled in Table 1, following:

TABLE 1.

Occurrence of trace elements in "Ore waters" and "Regional waters" at the Upper Kairakty tungsten deposit

Element	Ore waters	Regional waters
Cu*, Mn, Ti, Sr	very common	very common
Mo	very common	common
Zr, Zn**, Cu**, Pb	rare	common
Ni, Ag, V, Ba	rare	locally common
W, Y, La	common	rare
Sn, Cr	rare	rare
Ga, As, Zn*	absent	rare

These associations are rather unusual. Note that zinc is here a poor pathfinder whereas W, Y and La, which are practically unheard of in Western hydrogeochemical exploration, are considered to be good pathfinders. The following background values were established:

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* and **. Note that Cu and Zn are repeated. This sort of mistake is quite common in the English translations of Russian papers. The error may or may not be in the original. After studying Belyakova's tabular data the reviewer is of the opinion that the duplicates marked with two asterisks are incorrect.

n x 10^{-3} mg/l or less.	Mo, Cu, Pb, Ni, Ag, V
n x 10^{-2} mg/l or less.	Zr, Mn, Ti, Ba
n x 10^{-1} mg/l or less.	Sr, Zn
<n x 10^{-2} mg/l.	W

The following typical anomalous values were claimed:

n x 10 μg/l	V
n x 100 μg/l	Mo, W
n x 10 mg/l	La

The values for W and La are inordinately high and are probably related to the very unusual mineralogy of the ores at Kairakty.

After giving consideration to the requirements of adequate contrast and ease of analysis, Belyakova selected Mo and W as the potential pathfinders for these particular ores but added that tungsten could not be used in practice because of its restricted mobility. The background range of [Mo] in this area was 5 to 20 μg/l and anomalies in the vicinity of known orebodies ranged from 30 to 80 μg/l. These anomalies were sometimes displaced by lateral flow of groundwaters. Belyakova's study at Kairakty supported the well-established belief /348, 785, 533, 496/ that bicarbonate ions tend to precipitate molybdenum and that the Mo/bicarbonate ratio had to be considered during the interpreation of raw molybdenum data. Spurious molybdenum anomalies could be generated in alluvial sediments by evaporation, but these could be identified by examining the Mo/TDS ratio.

Erskov and Shcheglova (1958/786/) investigated the germanium content of several coal mines in the Kizelov basin of the U.S.S.R. Here sulphuric acid waters from coal seams and sandy host beds mix with neutral waters from superjacent carbonate strata. Values ranged from <1 to 3 μg Ge/litre and were neither seasonally dependant nor clearly related to [H_2SO_4].

Nevertheless the germanium content of the mine waters was anomalous when compared to underground waters of the adjacent regions and the authors were of the opinion* that systematic analysis of waters of coal mines might "result in discoveries of (economic) sources of the metal".

Chernyayev and Chernyayev (1962/800/) studied the process of hydrogeochemical dispersion at a polymetallic (Fe-Cu-Zn) mineral deposit in volcanic and sedimentary rocks at Gaisk in the U.S.S.R. They believed that their findings were quite typical of deposits of this kind. The local waters are of the chloride type but are extensively modified in the vicinity of the orebody, which is deeply oxidized (to depths of as much as 115 m). Thus the waters directly over the ore are sulphate-rich and have an elevated [TDS]. They are enriched in Fe, Cu and Zn (respective maximum concentrations being 3 800, 674 and 500 mg/l) and also carry abnormal amounts of Al, Mn, Pb, Mo, As, Ag, and Hg. This body of anomalous sulphatic water is elongated in the direction of groundwater flow and mingles with the regional chloride-rich waters to form a mixed zone, which may be situated as much as 3 km from the orebody. At the inner edges of this mixed zone the concentrations of Cu, Fe and Zn still exceed 1 mg/l.

The most interesting aspect of this study is that the authors have examined the abovementioned anomaly in three dimensions rather than simply as an areal feature. They established from samples collected at depths between 0 and 800 m that the anomaly decays vertically as well as laterally. The most serious dilution occurs within clearly defined zones at depths

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* This is probably only true for meteoritic waters. One ought to exercise care in using Ge as a pathfinder in hydrogeochemical exploration of geothermal areas. Studies of germanium in spring waters suggested that the element is generally correlated with B and As and that [Ge] is principally a function of temperature /787/.

of 250 and 550 m, where "water exchange" is intense. However, even at a depth of 700 m the trace element content of the sulphatic waters is still anomalous. The authors do not believe that this metal could have migrated all the way from the oxidized surface zone, and they ascribe the phenomenon to oxidation of deeply buried, disseminated sulphides by slightly oxygenated waters, which circulate* "to great depth ..... in fractures".

Kulikova (1962/795/) took a somewhat novel look at the question of secondary dispersion from orebodies and raised an interesting point. He attempted to assess the mobility of the components of ore by examining what was left behind once the processes of oxidation and leaching were essentially completed. He studied many polymetallic deposits in the U.S.S.R. and concluded that several elements showed a general tendency to disappear from these primary ores during oxidation. Amongst these are some elements, particularly Tl, In, Cd and Ga, that are not considered to be either very mobile or good pathfinders. The degree to which these elements are leached is dependant upon the composition of the groundwater, the level of acidity during oxidation and geological factors - for example, dispersion is reduced if the country rocks are carbonates. Kulikova does not speculate about what becomes of these elements or why they do not

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* Reviewers Note: They fail to explain why oxygen can migrate to great depth in fractures while trace elements cannot. This appears to be an illogical deduction. The reader should also note another important observation made in passing by these authors: "...... the natural hydrogeological conditions in the ..... area have been considerably modified by mining". They do not elaborate and, especially, they do not comment on the possible effects that mining and dewatering may have had on the deep circulation of metals and the three-dimensional shape of the anomaly.

form useful haloes* as they migrate away from the orebody.

Kovalev *et al.* /797/ studied the hydrogeochemistry of the Sibai' copper sulphide (chalcopyrite) deposit of the southern Urals, which is interesting because of the high carbonate content of the ore and wall rocks (altered volcanics). The deposit lies on a plain to the east of and some 300-400 m below the Karmalytash massif. The massif range is dissected and fissured, and water exchange is active although rainfall is low and seasonal. On the other hand the plains have little water and it circulates slowly except where Carboniferous limestones are encountered. There is a definite relationship between the hydrogeochemistry and the hydrogeology of the whole region and this is demonstrated by the contrasting types of water that occur in different topographic zones. The massif yields dilute carbonate groundwaters in which TDS ranges up to 3 000 mg/l. The principle ions are bicarbonate, and sulphate derived from disseminated sulphides but the concentration of chloride is only about 9 mg/l. In the acidic or siliceous rocks Na and Ca are the main cations but in basic rocks Ca and Mg predominate.

The more sluggish waters of the plain are also of the carbonate type but are more varied in composition, more saline and richer in chloride. High concentrations of sulphate occur only near sulphide deposits. Alkalis tend to accumulate as a result of intense cation exchange and this process is inclined to obscure the relationship between lithology and the cationic content of the waters. The typical background metal concentrations in these groundwaters are rather high and the maximum values for Fe, Cu and Zn

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* It is probable that dilution rapidly reduces the concentrations of these elements to values that are either indistinguishable from the background levels or below the detection limits.

are 60 mg/l, 600 μg/l and 30 mg/l respectively. Mine waters around the Sibai' deposit contain 9 000 mg/$SO_4^=$, 1 600 mg Fe, 160 mg Si, 1 750 μg Cu, 1 350 μg Zn and 5 μg Mo per litre. Other indicators, determined spectrographically, were Pb, Co, Ni, Ga, In and bismuth. A notable feature of these anomalous groundwaters is the absence of free H_2SO_4. Even water drawn directly from oxidizing ore is neutral (pH 6,8 to 7,7).

The following parameters (threshold values in parentheses) were considered to be important hydrogeochemical indicators of mineralization: Cu (10 μg/l), Zn (100 μg/l), sulphate/chloride (2) and sulphate/bicarbonate (0,1). High [Zn] or [Cu] coupled with high [$SO_4^=$] was regarded as a favourable index and some significance was also attached to high Zn/Cu ratios. The authors noted that the values of several of these indicators are linearly related and that some of them, like the major components of the anomalous waters, are direct functions of TDS.

Udodov and Parilov /792/ studied the metal contents of 4 500 samples of shallow groundwaters and stream waters collected in the vicinity of several Siberian orebodies. The metals in these waters were concentrated by co-precipitation with sodium sulphide/aluminium hydroxide and the metalliferous residues were then analysed by optical spectrography. Most values were in the μg or ng range. The authors concluded that As, Ag, Mo, Cd, Zn, Cu, P, Pb and Sb were relatively mobile and that the concentrations of these elements were usually independent of the pH. Bismuth, Cr, Be, Ni, Co, V, W and Mn were considered to be immobile and Ba, Ti, Sn, Sr and Zr were very immobile.

The more mobile elements formed groundwater anomalies that stretched as far as 500 - 1200 m from the source but the haloes of the immobile elements were typically 300 - 500 m wide. Threshold values were set at between 5 and 10 times background. Multielement associations tended to be more common in anomalous waters than elsewhere - i.e. it was possible to identify an anomaly by summing the concentrations of several elements even though the individual [metal] values were not substantially greater than the background levels.

The authors warn that one must exercise caution in assessing hydrogeochemical data from placers because particularly intense mobilization of metals occurs in groundwater flowing through these deposits.

Germanov *et al.* (1959/791/) noted that Eh is a critical factor controlling the mobility of metals but showed that the determination of meaningful experimental Eh values *in situ* was extremely difficult. The authors made an airtight connection between an Eh meter and a free-flowing underground borehole and measured the Eh of the running water for 15 days. The reading dropped systematically from +495 to +260 mv and stabilized at the latter value only after eight days. The authors offered no explicit explanation of this phenomenon but the implication is self-evident.

They carefully measured the Eh of more than 300 flowing waters in Soviet mines and elsewhere. The range of values (all relative to the standard hydrogen electrode) was +550 to -480 mv. The Eh value was a function of the concentration of dissolved gases and if free oxygen was present the value was generally greater than 300 mv. The highest Eh value (+550 mv) occurred in extremely acid water (pH = 2). Waters containing dissolved H_2S, CH_4 or other reducing compounds had negative Eh values (-30 to -480 mv). Thus it appears that the two factors that decisively influence the redox potential of underground waters are the concentrations of free oxygen and organic matter.

Rasskazov (1962/1093/) undertook a comprehensive study of the seasonal changes in the concentrations of dissolved species in underground waters. Shallow waters were more affected than deeper waters. In his study areas the changes were so large that special seasonal corrections were necessary during the plotting of data from exploration surveys. But provided these changes were made the data obtained in different seasons could be meaningfully compared.

Polikarpochkin *et al.* (1958/774/) investigated the total heavy metal content of groundwaters in the semi-arid Transbaykal region of the U.S.S.R., where several small Pb-Zn deposits are scattered over an area of some 80 km^2. These deposits do not have readily-identifiable individual hydromorphic anomalies but the outline of the area of anomalous waters is virtually coextensive with the boundary of the larger mineralized district.

Krainov and Korol'kova (1962/1069/) summarized the basic principles of hydrogeochemical prospecting for base metals (Zn, Cu, Pb, As) in areas of carbonate waters, using examples from the Lower Caucasus of the U.S.S.R. The compositions and total dissolved solids contents of these waters are quite variable, and they show all of the typical aqueous metamorphic transitions (i.e. HCO_3 - Ca waters → HCO_3-Na → HCO_3-Cl-Na → Cl-HCO_3-Na → NaCl brines). In these groundwaters Zn, Cu and Pb are present as cations but arsenic migrates as an anion or a molecular compound and is sensitive to valence changes. High [Zn], [Cu] and [Pb] were associated with sulphide mineralization but also with certain rock types and/or water types in areas not known to be mineralized, i.e. there are certain waters that naturally have abnormally high metal contents (background values), which appear as false anomalies. These factors must be clearly understood in order to select real hydrogeochemical anomalies. All else being equal, in the Lesser Caucasus carbonate springs with high contents of heavy metals are localized along ore-bearing faults. Carbonate waters issuing from barren faults are generally very poor in these metals. Useful spring water anomalies can be up to several square kilometres in area and can sometimes reflect the presence of completely blind ore deposits.

Arsenic concentrations of up to 20 mg/l were noted but the "normal" range did not exceed 100 μg/l. Arsenic was sometimes a hydrogeochemical indicator of sulphide mineralization but the element also occurred in high concentrations in some other waters*, e.g. in artesian waters "affected by" vulcanism. Here [As] was clearly related to both TDS and [Cl], and increased systematically "with distance from the catchment area". Where As in groundwater was associated with sulphides Zn, Cu and Pb were also frequently anomalously concentrated. The authors note that similar results have been obtained in many areas of carbonate groundwaters both within the U.S.S.R. and elsewhere. They found that arsenic was very mobile in carbonate waters.

This paper by Krainov and Korol'kova is also important for its statement of the concepts of "paleohydrogeochemistry". It is the belief of some Russian authors that the existence of certain regional water types is a

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* The authors note that occurrences of high background values in groundwaters are of two types:
(1) Accumulation of metals with increasing TDS. Here [Cl^-] is very important as a complexing ion. Arsenic is especially affected in this manner.
(2) Stripping of metals by dilute waters (a) in rocks with abnormally high concentrations of dispersed metals, e.g. in tuffs, or (b) under unusual physical conditions. For example, high [As] in groundwaters is typical of regions of Alpine folding and metamorphism and this is probably due to the ready release of arsenic from sedimentary materials to waters under conditions of deep regional heating. "It is evident, then, that in hydrogeochemical prospecting carbonate waters must be used differently in gologic (sic-geologic) structures passing through different stages of development".

reflection of past (or indeed modern-day) ore forming processes in those regions. The identification of these special waters - for instance warm, carbonate-rich, metalliferous brines - may delineate a metallogenic region in which ore bodies may recently have formed. This concept is distinct from that of following a secondary hydrogeochemical anomaly to its oxidized source and can be loosely viewed as a form of "primary" hydrogeochemical prospecting. "The principles of utilization of the ore-forming activity of carbonate waters in hydrogeochemical prospecting were formulated by Ovchinnikov ..... In this case, a search is made for areas of emergence of carbonate waters favorable for accumulation of ore minerals, i.e. for areas possessing certain "geochemical barriers" (Perel'man's terminology). In this method hydrogeochemical (paleohydrogeochemical) and metallogenic factors must be taken into account as well as the migration characteristics of each metal under the concrete conditions existing in the artesian basins.

As shown by the data for the Lesser Caucasus, the hydrogeochemical method based on the ore-forming activity of carbonate waters may be used successfully in the search for deposits of arsenic, borates and, to some extent, of borosilicates. The most favorable localities in this respect are the artesian basins of the Armenian folded belt, whose carbonate waters reflect the effects of late Tertiary intrusive activity and Quaternary volcanism." (p. 535).

Brief mention must be made of the work of Krainov and Petrova /796/, who described the problems of hydrogeochemical prospecting in areas of extremely mineralized subterranean waters, and the publication by Safronov *et al.* (1958/768/) about prospecting for gold. The latter workers showed that gold deposits could be identified by spectrochemical analysis of Au in local groundwaters.

A few notable Russian language papers from the period 1958-1962 deal with ancillary hydrogeochemical topics such as the stabilization of metals (Mo, V) in natural solutions. Popova (1961/793/) noted that the effects of co-precipitation on the concentrations of trace metals in water are well known but that little attention had been given to calcium carbonate as a potential precipitant. In his experiments he added solutions of Na_2CO_3 and $CaCl_2$ to waters containing dissolved trace elements. This resulted in the precipitation of 50-75 mg of $CaCO_3$, which was analyzed to find out which of the trace elements had been stripped from solution. The experiments were conducted at temperatures from 20^o upwards. The intensity of stripping was increased at higher temperatures and the elements that form difficultly soluble carbonates showed the greatest tendency to co-precipitate. Cadmium, Pb and Co, at concentrations up to 2000 μg/l, were completely stripped from the water. Zinc and Cu were almost 100% removed from solutions of 25 μg/l and the corresponding value at concentrations of 1-2 mg/l was 75%. Nickel was least but nevertheless substantially affected (66% at 25 μg/l).

Mekhtiyeva (1962/798/) showed that even deeply seated groundwaters commonly have a specialized microflora, of several physiological groups, which actively influences the chemical composition of these waters. If all else be equal, then the concentration of bacteria diminishes with depth. If the composition of subterranean water changes upon encounter with rocks of a different lithology then the characteristics of the bacterial population are also likely to alter. Thus sulphate reducers will normally flourish in gypsum rich strata near petroleum occurrences but an influx of calcium rich water will suppress their numbers.

The Hungarians were quite active in the field of groundwater hydrogeochemical exploration during 1958-1962/922-4/ but little of their published

4.5 THE CURRENT LITERATURE: 1963-1978

4.51 INTRODUCTION

There is obviously no clear division between historical and current publications but one can reasonably regard the past fifteen years as the "modern" or "present-day" phase of geochemical prospecting. Between 1963 and 1978 a large number of papers were written on this or closely related topics /1122/ and at least 500 of these are of direct or indirect interest to the prospector who samples groundwaters. In Section 4.5 an attempt will be made to review a representative cross-section of the current literature of groundwater hydrogeochemical exploration. It is necessary to bring some order into the considerable volume of material examined in this chapter and the various publications have therefore been grouped into the following categories:

1. Reviews and overviews.
2. Methodology, sampling, contamination problems, analysis, data handling, interpretation.
3. Hydrogeology, water-rock interaction, pH/Eh, complex ion formation.
4. Organic matter, gases, particulate matter, thermal/mineral waters.
5. Anions, halogens, sulphate, boron, alkali metals, alkaline earths.
6. Some less commonly sought elements. As, V, Cd, Se, Te, Sb, Sn, Ag, Au, Hg.
7. The common base metals. Sulphide bodies, polymetallic ores, copper/molybdenum porphyry bodies.
8. Uranium and associated elements.

Some overlap is unavoidable and publications dealing with two or more of these topics have been classified according to the main theme of the work. The reader who requires additional material may find something of interest in Compendium A, which is a listing of other potentially useful work is available in English. Opran /1018/ described a method of hydrogeochemical prospecting for salt deposits in Hungary. The useful "indexes" were K/TDS, $SO_4^{=}/Cl^-$ and Mg^{++}/Cl^-.

COMPENDIUM A

Reviews and textbooks:/56, 845, 903, 946, 961, 974, 975, 1014.

General hydrogeochemical principles: /853, 911, 1138.

General groundwater hydrogeochemical studies, including prospecting applications: /827, 868, 911, 940, 971-2, 1126, 1129-32, 1067-8.

Uranium in groundwaters: /804, 844, 847, 866, 889, 900, 930-1, 933-7, 945, 953, 1025, 1053, 1063, 1083, 1086, 1136, 1140.

Uranium isotopes and daughter elements (Ra, Rn, Th, He etc.) in groundwaters: /755, 805, 807, 810, 812, 838, 858, 879-81, 883-5, 917, 1140-1.

Aqueous chemistry of uranium: /789, 815, 835, 989.

Handling of uranium hydrogeochemical data: / 320, 1054, 1087, 1139.

Groundwater hydrogeochemistry of molybdenum: / 850, 1047.

Groundwater hydrogeochemical exploration using boron: /852.

General hydrogeochemistry of mercury: /853-4, 977, 1044.

Groundwater hydrogeochemistry of mercury: /912, 957-61.

Hydrogeochemistry of gold: /857, 1055.

General hydrogeochemistry or groundwater hydrogeochemistry of copper, lead and zinc: /166, 859, 867, 901, 979-81, 989, 1056-7, 1135, 1140.

Groundwater hydrogeochemistry of sulphate: /987.

Hydrogeochemistry of various rarer elements: /848, 911.

Gases in groundwaters: /899, 917-8, 955.

Groundwater hydrogeochemistry near porphyry deposits: /849.

Groundwater hydrogeochemical exploration in areas of permafrost: /166, 849.

Electrochemical control of aqueous dispersion from orebodies: /892, 962.

Eh and pH: /1137.

Methods of sampling and analysis: /976-8, 980.

Mathematical hydrogeochemical models: /983.

references that will not be examined in detail in this review. A number of other recent publications (1965-1975) that have not been reviewed in this work are listed by Hawkes (1976/1070/).

The hydrogeochemist attempting a survey of the recent literature is confronted by many thousands of potentially useful publications, which are widely scattered in many journals and hundreds of less formal reports and bulletins. Interestingly, there have been extremely few published guides describing how one might set about such a survey. The only one known to the reviewer is that of Delfino (1977/1006/). He states that "the multidisciplinary roots of water chemistry have resulted in a variety of publication outlets ....." and attempts to answer his own question: "Where does one find the published information of interest to, and most often needed by, water chemists?" He does this by providing comprehensive lists of "Bibliographic resources" but points out the unfortunate truth that "it is almost impossible for water chemists to go to a single library ..... and expect to find all of the journals that they might need in a broad-based research program ..... The literature of water chemistry is extensive; it appears in a wide variety of journals; and it is covered in varying detail by bibliographic resources. Scholars desiring to pursue research in the field of water chemistry should be aware of these facts, in order to utilize the literature in an efficient, systematic, and comprehensive manner".

4.52 EXAMINATION OF SELECTED GROUPS OF RECENT PUBLICATIONS RELATED TO GROUNDWATER HYDROGEOCHEMICAL EXPLORATION

4.521 Reviews and overviews

In this section we will very briefly examine a few English language publications that provide a general measure of the "state of the art" of groundwater hydrogeochemical exploration.

According to Erickson (1971/846/) the principle changes in geochemical prospecting in the late 1960's included (i) "A shift in emphasis to a climatic-province or environmental approach to exploration ....." and (ii) an upgrading of analytical methods to meet the demand for good trace element data and an increased use of "computer approaches to mineral exploration problems".

Several noteworthy *review papers* date from this period. Bradshaw *et al.* (1972/1117/), for example, reviewed many aspects of hydrogeochemical prospecting and the relationship of this method to the other branches of geochemical exploration. This is not a high-powered summary directed at the specialist in the field of hydrogeochemistry, but is nevertheless a very fine introduction to and summary of the current state of the art. Perhaps the best of the more serious reviews of the historical and current relationship between groundwater hydrogeochemical exploration and the other geochemical prospecting methods is that of Boyle and Garrett (1970/973/). A very useful thumbnail summary of the principle literature sources is given. The authors point out the tremendous impact that atomic absorption spectroscopy has had on hydrogeochemical exploration and the paper includes an important listing of references dealing with the application of atomic absorption analysis in prospecting. They note that groundwater hydrogeochemical exploration is still a relatively neglected subdiscipline (at least in the West), which lacks a background of basic research. The measurement of Eh and pH will probably become automatic in future hydrogeochemical surveys. The use of anions as pathfinders holds much promise but also presents many problems that need to be solved by fundamental research. This paper includes a notable discussion of the application of fluoride anomalies in organic-rich waters. Under permafrost conditions fluoride haloes are at most 100 to 200 m long and displaced anomalies, which appear as fracture springs, are common. Stream water sampling in the same area produced "no abnormal values" of the F/Ox. ratio and was therefore no substitute for the groundwater technique. The latter method was often superior to either geophysical or geological procedures used for locating phlogopite bodies.

The authors note that uranium deposits of the "roll front" type are also known in the U.S.S.R. These bodies are associated with the movement of oxygenated, uraniferous groundwater into a reducing bedrock environment, as shown in Figure 16.

Predictably, a number of important overviews appeared in English *textbooks* or other large volumes dealing partly or wholly with hydrogeochemical methods of mineral exploration /1071-7/. One interesting example is the translated collection of Russian papers on hydrogeochemistry, edited by Bogomolov and Balashov (1963/1072/). This contains long papers on the statistics of the distribution of elements in waters, on hydrothermal zones in the crust of the earth, on the hydrolysis of heavy metals and on several additional interesting topics.

However, the outstanding work in this category is the textbook by Levinson (1974/51/). It is not possible to give a comprehensive account of Levinson's contribution in the space available here, but some of the most important observations that he has made can be summarized in a very condensed form. Levinson holds the view that hydrogeochemical exploration, despite its obvious progress in the last thirty years, is nevertheless still in its infancy. He notes that the method is used chiefly in the U.S.S.R. and that in the West it is applied principally in the search for uranium.

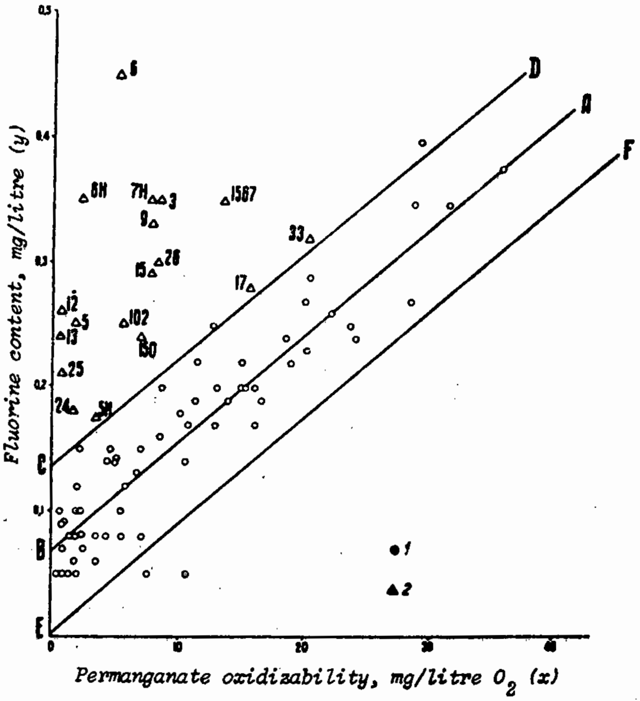

Fig.12. Content of fluorine and organic matter (permanganate oxidizability) in the waters of northern taiga landscapes of the Aldan Upland (81 samples). *1* = water from outside areas of phlogopite deposits; *2* = water from areas of phlogopite deposits.

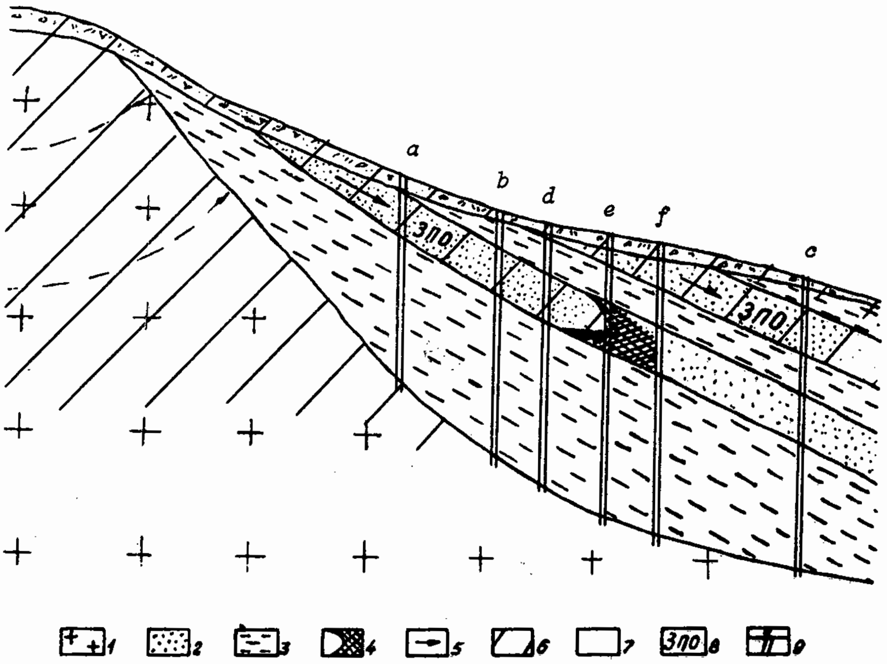

Fig. 16. Diagrammatic depiction of prospecting for undisclosed uranium deposits on the basis of criteria of the theory of epigenetic zonation. *1* = igneous rocks; *2* = aquifers in sedimentary strata – the upraised flanks of an artesian basin (sands, etc.); *3* = horizons in the sedimentary strata impervious to water (clays, etc.); *4* = ore beds at a reduction barrier; *5* = direction of movement of the water; *6* = red, brown, yellow and green rocks with oxidizing conditions (hydroxides of iron, etc.); *7* = grey rocks with reduction conditions (pyrite, organic matter, etc.); *8* = zones of layered oxidation; *9* = test bores on the artesian slope. *a*, *b* = pre-ore bores (in the layered oxidation zone); *c* = post-ore bores (in the zone of reduced rocks); *d*, *e*, *f* = ore bores in the detailed prospecting sector.

The sampling of groundwaters is intrinsically a more powerful method than the sampling of surface waters, since the former waters make more thorough contact with the host rocks and have longer residence times in these formations. Groundwater hydrogeochemistry is therefore recognized as a potentially powerful regional reconnaissance method, especially in those instances where soil sampling is not feasible. Attempts have of course been made to exploit this potential, but in Levinson's opinion the results have generally been rather poor. Even in very recent years the use of the method has been restricted by a number of problems, amongst which are included the following: (i) Seasonal fluctuations in water composition. (ii) Regional variations in water composition. In a sense natural water is not as complex as other earth materials such as soils, because the vast majority of normal dilute waters do not contain more than eight major (>1mg/1) components (Ca, Mg, Na, K, HCO_3, SO_4, Cl, SiO_2). Other elements that are abundant in the crust, such as Al, Fe, Mn and Ti, are not normally concentrated in groundwater because they are hydrolysed or have commonly-occurring insoluble phases. Nevertheless, the composition of groundwaters often varies greatly from one region to another or even from one aquifer to the next, and this can quite severely complicate the interpretation of exploration data. The concept of using the ratios of concentrations of components has lessened this problem considerably, but cannot eliminate it. (iii) With the exception of the major ions, very low concentrations of most components are encountered and almost all of the pathfinders sought in groundwaters are present to the extent of a few parts per billion only. (For a long time this caused severe analytical problems and it is only in the last few years that substantial progress has been made in the field of large-scale, routine, multielement, ultratrace analysis of natural waters).

It is recognized that there are several powerful natural phenomena that suppress the concentrations of many elements in natural solutions. The scavenging effect of certain natural precipitates is now well-known and it has also been recognized that practically all elements, and particularly those given to adopting cationic forms, may be adsorbed or ion-exchanged onto or into various mineral substances such as clays or humic complexes. This leads to problems of interpretation that are not related to analytical difficulties. In those cases where a dispersion halo consists of a relatively minute mass of metal disseminated within a comparatively vast volume of water it is not uncommon for the physical and chemical effects of the country rock, or the aquifer from which the sample is drawn, to override and disguise the influence of any orebody in the vicinity. Nor are we in a position yet to overcome these difficulties by applying equilibrium concepts to calculate the probable results of reactions between groundwaters and host rocks. We lack adequate data for the average abundances of trace elements in groundwaters and our understanding of the theoretical maximum concentrations of these microcomponents in natural solution is very imperfect. The inadequacy of current theory is clearly shown by the fact that observed concentrations very frequently exceed the theoretical maxima. The problem is compounded by the frequent occurrence of disequilibration systems at redox or lithological barriers, or where waters mix. Kinetic and reaction data for these situations are wholly lacking. (iv) High costs of collection and transportation of water samples. (v) Problems of contamination. For obvious reasons water is very easily contaminated both before and after collection. (vi) Sample preservation. It is well-known that several elements or other chemical species are rapidly lost from samples of natural waters and that the stabilization of these samples is often difficult and costly. (vii) In many cases hydrological information is needed in order to interpret the raw chemical data. Methods exist for the measurement of hydraulic gradients and other hydrological parameters (for example, through the use of dyes and radio-isotopic tracers) but it is seldom possible to use them in practical regional exploration projects. Data from a large number of wells are usually required for practical hydrological studies - hydrologists frequently drill many additional holes purely for this purpose - whereas the exploration man will often, for one reason or another, be limited to incomplete information from only a few holes per square kilometer. The problem becomes severe in fractured rocks where the anomalous pattern may be highly complex and not readily contoured or visualized in three dimensions. In extreme cases linear anomalies can result, e.g. as a line of springs along a fault. Similar difficulties may be encountered in highly permiable rocks, in which oxidizing conditions can exist well below the water table. This has the advantage of promoting dispersion, but can also result in displaced or very widely disseminated anomalies. For these and other reasons it has proved to be rather difficult to trace dispersion haloes in groundwater.

Levinson and many other writers have made it quite clear that it is neither possible nor desirable to strictly separate the science of groundwater hydrogeochemical exploration from other chemical studies of subsurface waters. The prospector who samples groundwater stands to benefit greatly if he is aware, in a general sense at least, of the current status of research into groundwater hydrogeochemistry other than that which fall into the narrow field of exploration for mineral deposits. An excellent recent review of the broader topic has been given by Johnson and Meyer (1975/976/). An equally important review of general subsurface hydrogeochemistry in the 1970's is that of Barnes and Hem (1973/1042/). This work is not concerned with prospecting applications but is a very thorough study of all general aspects of the literature dealing with all types of subsurface waters. It is thus a most useful work for the prospector who wishes to view his exploration theories and practices against a broader background of modern hydrogeochemical knowledge. There are 178 references.

4.522 Methodology, sampling, contamination problems, analysis, data handling, interpretation

Several important publications that fall into this class appeared between 1963 and 1978. Some deal with specific aspects of analysis, or computer applications etc. while others have dealt more broadly with general methodology in hydrogeochemical prospecting. Many of these ideas and methods will of course apply equally well to both surface waters and groundwater.

Dall'Aglio (1968/890/) gave a short but useful summary of "the organizational and methodological problems" encountered in hydrogeochemistry. This paper includes aspects of analysis, sample stabilization and computerized data handling practices.

Hem (1970/1028/) produced an updated version of his classic (1959/286/) "Study and interpretation of the chemical characteristics of natural water". This tome (363 pages) remains one of the "holy books" of the water sampler, but unfortunately no attempt can be made here to review a work of this size. Recent useful reviews of aspects of water characterization, sampling procedures, chemical evolution of groundwaters and the stability of samples of groundwaters have been given by Wallish (1977/1112/) and Reed (1977/1113/).

Problems of sample contamination before collection have become more serious in recent years as a result of anthropogenic pollution of groundwaters. Hosking (1971/856/) presented an interesting summary of some of the difficulties associated with the use of hydrogeochemistry (surface and subsurface) in Cornwall, where contamination is almost universal. The problem is generally insoluble in those areas where levels of contamination are very high. Pings (1968/941/) showed that the mine workings of Pennsylvania generate over one million tonnes of "natural" sulphuric acid per year, all of which is dispersed by water. It cannot be imagined that this will be without effect on the local hydrogeochemical regimes. Recent work (1976/943/) in the Transvaal has decisively dispelled any illusion that the relatively under-industrialized nations such as South Africa are free of this problem.

The reviewer, together with Klopper (1976-1978/1001-4/) has presented elsewhere the results of an extensive experimental investigation of the twin problems of sample contamination in storage vessels and sample stabilization. This documentation, which includes a comprehensive literature survey of historical and current publications dealing with the problem, runs to several hundred pages and cannot be adequately repeated here. The principle findings of the experimental studies can be summarized as follows: (i) Hydrogeochemical samples can, in most instances and for most elements, be effectively stabilized by a combination of acidification (pH 1,0) and freezing at -20°C. (ii) Contamination from well-cleaned polyethylene containers, whether they contain acidified or unacidified samples, is negligible even after four years of storage. (iii) Precleaning of polyethylene with 10% nitric acid solution for twenty-four hours is optimal. Stronger acid or longer cleaning is neither necessary nor desirable.
(iv) If the closures of the containers are not made of polyethylene, then care must be taken to ensure that they are not degraded by the acid.
(v) Nitric acid (as opposed to HCl and $HClO_4$) appears to be the best preserving acid for µg/l solutions of Cd, Zn, Tl, Pb, Mn, Ag, Cu, Co, Fe and Cr. Unfrozen samples containing some or all of these elements, if acidified

to pH 1,0 with nitric acid, were completely stable for at least eight weeks.

Other recent studies of the problem of sample stabilization are those of Lo and Wai /1040/, King *et al.* /1041/ and Toribara *et al.* /1043/.

Some mention was made in the previous section of the considerable progress that has been achieved in the field of water analysis in the last fifteen years. Three techniques have come to the fore in the routine analysis of trace or ultratrace amounts of elements in natural water- Atomic absorption spectroscopy, Neutron activation methods, and X-ray fluorescence spectrometry.

Atomic absorption methods have of course produced a complete revolution in this field and since the early 1970's the advent of flameless AAS methods has established this technique very firmly in hydrogeochemical laboratories. One of the earliest comprehensive account of these methods was written by Friedrich *et al.*(1973/871/), but within only a few years the technique was already considered to be well-proven /872 p. 19/.

Despite the well-known disadvantages of neutron activation analysis, the importance of this method in hydrogeochemistry continues to grow rapidly. Provided that the activation facilities are available the use of the method often has advantages that cannot be matched even by AAS methods. The application of neutron activation methods in hydrogeochemistry is well illustrated by the contents of the Preprint Volumes of the Proceedings of the 1976 International Conference on Modern Trends in Activation Analysis /984/. These preprints contain a great deal of information that will interest the hydrogeochemist but no specific articles from these books will be mentioned here, as the foreword of the work specifically forbids this. The items of interest include (i) The use of Indium-EDTA for tracing groundwater movement. (ii) The study of metals in natural ice sheets. This has confirmed that the metals in natural water samples can be totally immobilized for decades by deep freezing. (iii) Neutron activation analysis of water samples, with a section on contamination problems in ultra-trace analysis. It was shown that freezing is indeed the surest means of sample preservation. Samples six months old showed no deterioration whatever. (iv) Several sections of various papers deal with aspects of ultra-clean hydrogeochemical sampling, and sample handling and storage procedures. (v) Neutron activation analysis applied to the study of the mobility of elements in natural water.

Despite the considerable impact that AAS and NAA have had, many hydrogeochemists have remained very interested in the possibility and the obvious advantages of determining microcomponents of water by X-ray fluorescence methods. Green *et al.* (1971/860/), for example, described a combined ion--exchange/XRFS method for the determination of microgram amounts of metals in water samples and other materials. Parslow and Dwairi (1977/1103/) recorded a novel way of preconcentrating uranium from waters in the field by using ion-exchange resins in teabags. This eliminates all of the problems associated with the transportation and storage of unstable water samples.

Several papers described modern computer-oriented procedures for handling large amounts of hydrogeochemical data. Turner (1967/1029/) gave an account of a computer-based data storage system for groundwater hydrogeochemical information. A major publication by Gill and Rosenthal (1975/320/) dealt with Fortran programs for processing hydrogeochemical data. The principles underlying the programmed computations are discussed in detail and there is an important review of previous work on the handling of hydrogeochemical data.

Further information on the subject of computerized data manipulation is given in a publication by Dall'Aglio and Gigli (1972/909/). This paper also records a novel approach to the checking of major component balances in water analyses. Instead of balancing the equivalent masses of cations and anions, these authors calculate the electrical conductivity from the ionic activities of all the ions present and compare this valve to the measured conductivity of the sample.

The ultimate objective of data handling remains interpretation. Hydrogeochemists have adopted many methods of data interpretation and these vary from fairly simple statistical analysis to highly sophisticated mathematical modelling. Smirnov (1963/803/) presented an interesting discussion of the statistical distribution of the concentrations of elements in natural waters. He suggested that the distribution of many species in moderately dilute waters (i.e. typical groundwaters) will be lognormal. Normal distributions are usually restricted to very saline waters such as oilfield waters. It follows that one must be cautious about applying normal statistical tests to groundwater hydrogeochemical data.

A paper by Hitchon *et al.* (1971/1045/) provides a very good example of the advanced mathematical techniques that are being applied to hydrogeochemical data in order to obtain quantitative genetic models of the distribution of various chemical species in typical waters of different geographical regions or lithological zones. Using various statistical procedures, these authors showed that the formation waters of the western Canadian sedimentary basin must originally have consisted of connate seawater, which has been modified to a greater or lesser degree by processes such as dilution by fresh water, membrane filtration, halite dissolution, diffusion of ions to the surface, dolomitization, sulphate reduction, chlorite formation, ion exchange reactions with clay minerals and organic compounds and solution equilibration of salts such as $CaCO_3$ and $SrSO_4$. Advanced modelling of this sort is obviously of enormous potential value to the exploration geochemist.

4.523 Hydrogeology, water-rock interaction, pH/Eh, complex ion formation

Some of the review articles mentioned in Section 4.521 emphasized the need to establish closer links between hydrochemistry, hydrology and hydrogeology in mineral exploration, and in this section the work of some of the writers who have contributed to a closer integration of these disciplines will be examined.

Kovalev *et al.* (1968/1038/) showed that in some groundwater prospecting surveys there is a very real practical need to place the greatest emphasis on the hydrology of the region under investigation. In the Turgai syncline, for example, the waters of crystalline rocks, fracture zones and of the sedimentary mantle occur together but are of very different types. Several subtypes were identified and each had its own characteristic set of threshold values for the various indicator elements (Cu, Zn, Pb, Mo, Cr, Bi, As, Ga, Ag, Te, Sn, Sb), which were determined spectroscopically. A careful analysis of the hydrological relationships between adjacent bodies of waters lead to the location of several anomalies.

Haefeli (1971/1064/) provided a good example of how hydrological parameters can be inferred from hydrogeochemical data if circumstances are favourable. He showed clearly that systematic "metamorphism" of waters occurs during lateral migration through shallow aquifers. If these changes are well understood, then it follows that an outline of the local hydrological cycle can be established by studying the "main hydrogeochemical criteria" of groundwaters. In this manner the "origin, recharge and discharge areas or the flow direction of the groundwater" can be determined. Haefeli notes that $[HCO_3^-]$ is controlled by the carbonic system but that $[Cl^-]$ and $[SO_4^=]$ accumulate as a result of slow dissolution of rock components and seldom reach saturation levels. This is one important reason why the composition of groundwater changes with time. Base exchange is another powerful mechanism that modifies the composition of almost any kind of subterranean water.

Parsons (1967/831/) provides another example of recent progress in the attempt to understand the evolution and movement of groundwaters by establishing the origin of dissolved components. He too found that the patterns of distribution of the major components of groundwaters are clearly related to the lithology of the host rocks, but showed that this was not always revealed by simple ionic ratios. In the Upper Notukeu Creek Basin (Saskatchewan), for example, the Ca/Mg ratio "is erratic ..... and appears to have little or no significance". On the other hand sympathetic changes in the ratios Na/(Ca + Mg) and HCO_3/SO_4 were much more useful.

Hoag and Webber (1975/1089/) described some very interesting experiments in Quebec in which an attempt was made to trace the source of anomalous waters by quantitative measurement of certain hydrogeochemical parameters that are a function of the distance travelled by the groundwater sample. They selected sodium as the indicator and based their calculations on the assumption that this metal was derived from the dissolution of albite. The equation derived was

$$L = (A.\Delta H.M_{Na})^{\frac{1}{2}}$$

where L is the distance travelled by the water, A is an empirical constant determined experimentally, ΔH is the hydraulic head and M_{Na} is the molarity of sodium in the sample. This method was used in conjunction with the distribution of sulphate ion and Si/Na ratios to locate the sources of mineralized waters flowing near volcanogenic mineral deposits.

The abovementioned publications are but a very small indication of the growing importance of understanding the relationship between regional hydrology, hydrogeology and hydrochemistry on the one hand and groundwater hydrogeochemical exploration on the other. It is unfortunately not possible to give many more examples here for want of space but the reader who wishes to obtain extra material can consult the following key references: (1) Toth (1970/1065/), for a description of conceptual models of the groundwater regime and the hydrogeological environment. (2) Burger (1975/1114/) and Back and Zoetl (1975/306/), for a discussion of the principles of water--rock interaction in the carbonic system. (3) The Symposium Volume edited by Cadek and Paces (1976/1115/), for a large number of papers summarizing many aspects of modern thinking on the topic of chemical interaction between rock materials and groundwaters.

The characterization of water-rock interactions in terms of Eh, pH and electrochemistry remains an interesting, fruitful and difficult field of study for the hydrogeochemist. Bolviken *et al.* (1973/869/) described devices for down-hole measurement of pH and Eh. They stressed the importance of these measurements in hydrogeochemical exploration and the difficulties encountered in doing them properly. They found, like previous workers such as Hansuld (1966/870/), that a major problem with Eh measurement in dilute natural waters is the nearly vanishing concentrations of the components of important redox pairs such as Fe^{2+}/Fe^{3+}. Bolviken *et al.* provide a useful list of references to earlier work on Eh in groundwaters.

Sveshnikov (Sveshnikov and Ryss, 1964/811/) followed up his earlier work /357/ on groundwaters of orebodies as components of electrochemical cells. A convincing case is made to support the suggestion that the heavy metal chemistry of mine waters, for example, will be powerfully influenced by electrochemical dissolution and precipitation reactions as well as simple redox reactions. The groundwater in the vicinity of an orebody that is undergoing oxidization acts as a conductor between the cathodic oxidized ore and the anodic primary ore and may have an Eh as high as +0,8v. The mobilities of copper and silver, in particular, are strongly affected by this galvanic reaction, and they are very typical of the zone of secondary enrichment that develops at the base of the cathode. Microgalvanic cells, formed by the local flow of electrons between minerals with different electrode potentials, may cause the dissolution of electronegative compounds such as galena and sphalerite and thus enrich nearby waters with heavy metals. This mechanism is particularly important because it provides for the release of metal ions even in the absence of free oxygen, for example:

$$PbS \rightarrow Pb^{++} + S^{o} + 2e^{-}.$$

This effect is most pronounced in polymineralic-polymetallic deposits. The most important possible result of powerful electrochemical (as opposed to simple oxidative) solution is the formation of a heavy metal groundwater hydrogeochemical anomaly without the presence of high concentrations of sulphate ion, and with the relative concentrations of the dissolved heavy metals very different to their proportions in the ore. This "electrochemical migration" has been very little studied but in at least one case (Rudnyi Altai) it has been responsible for some unusual effects. Here Cu^{++}, Zn^{++}, Fe^{++} and H^{+} reach a maximum concentration in the waters directly over the oxidized zone of the polymetallic ore, while there is a simultaneous and sharp drop in the concentration of dissolved sulphate.

Edmunds (1973/982/) claimed that although there was much mention in the literature about the importance of Eh (and pH) we really know very little about actual Eh conditions and Eh-controlled phenomena in real rock/water systems. The number of studies "in which detailed controlled measurements of redox potentials in ground water have been made" is very small indeed. Most Eh values reported in the literature are probably too high because of oxygen contamination. Edmunds studied the changes in water chemistry that occurred as water moved down dip along a gently sloping limestone horizon in eastern England. The water migrates mainly via fissures and the rate of movement, as deduced from measurements of bomb tritium, is about 2km/year. Samples were readily obtained from artesian boreholes at various points along the dip slope over a distance of sixteen kilometres from the surface outcrop (recharge point) of the aquifer. The procedures for measuring Eh, or for calculating it from other measurements such as Fe^{++} and pH are given. It is important to note that stable Eh readings were obtained only after a minimum period of 35 minutes (max. 105 minutes) as the water flowed continuously through an air-proof measuring system. The reason for this slow stabilization is not discussed.

The results showed that there is a sharp redox barrier in the aquifer some 12 km from the outcrop. The Eh is initially buffered by the presence of free oxygen (+400 mv) but falls sharply as the concentration of free oxygen approaches zero (+150 mv). The water becomes steadily more reducing further down dip as reduced species such as HS^- begin to appear* (+125 mv). At the deepest point for which data are available the Eh approached -100 mv. The changes that occur in the chemistry of the waters along the traverse are summarized in Figures 3, 4 and 5. Unfortunately, it appears that some of the information in these diagrams (e.g. the Ca^{++} graph) is in direct conflict with the ideas expressed in the text and so the data must be assessed with caution.

Note that the "pH barrier" is not coincident with the Eh barrier. All major changes in chemistry occur up to about 4 km beyond the Eh barrier. Sulphate is lost by reduction by organic matter and this results in an

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* Edmunds points out that the human nose is exceedingly sensitive to H_2S and can detect minute quantities equivalent to far less than 1μg/1, so that one must be very careful about judging the redox state of a water from its smell alone.

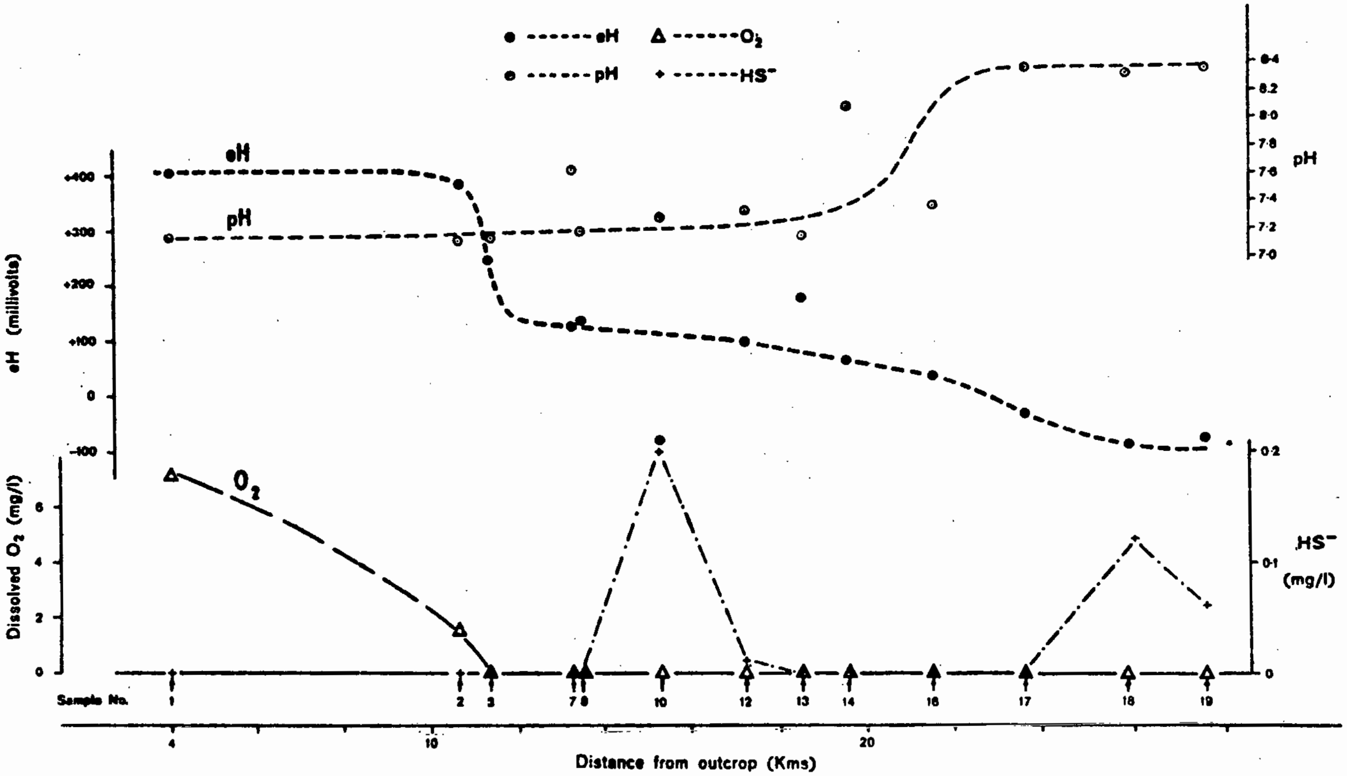

Figure 3. Variation in eH, pH, dissolved oxygen, and sulphide with distance from the outcrop.

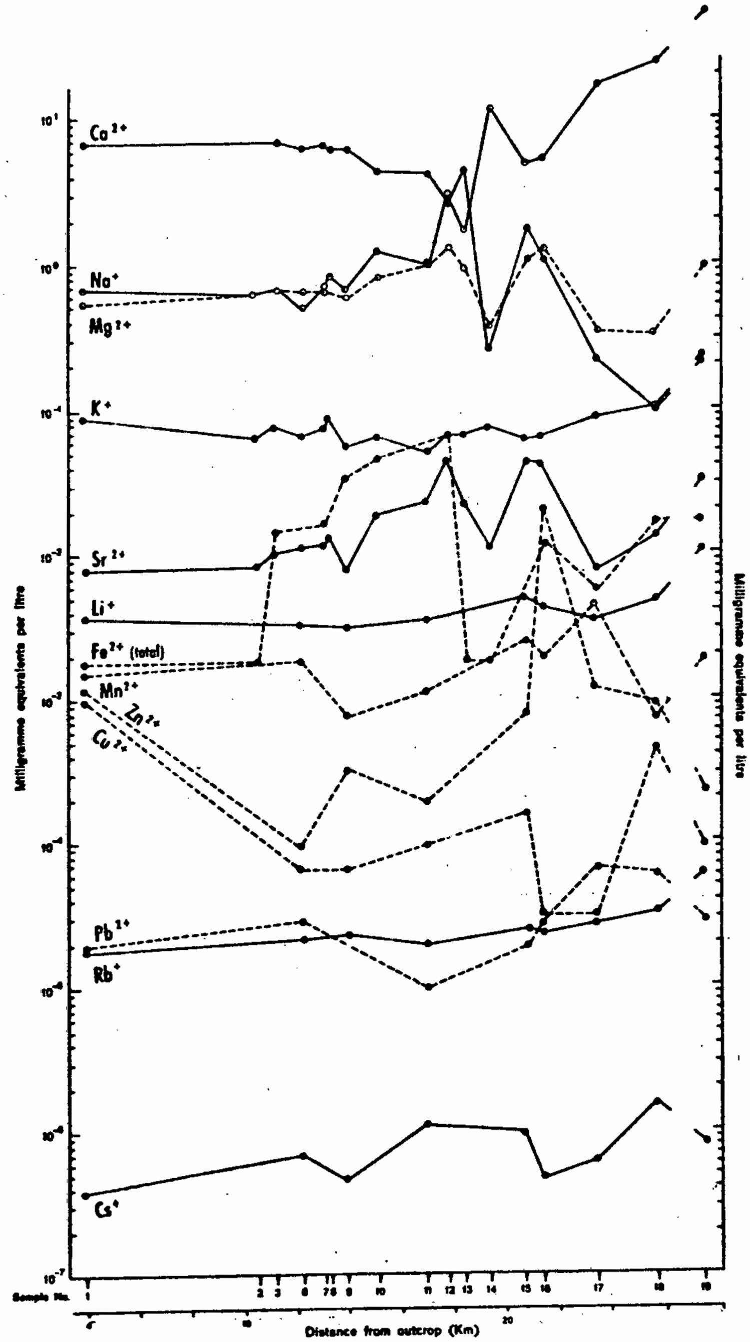

Figure 4. Variation in concentration of major and trace cations with distance from the outcrop.

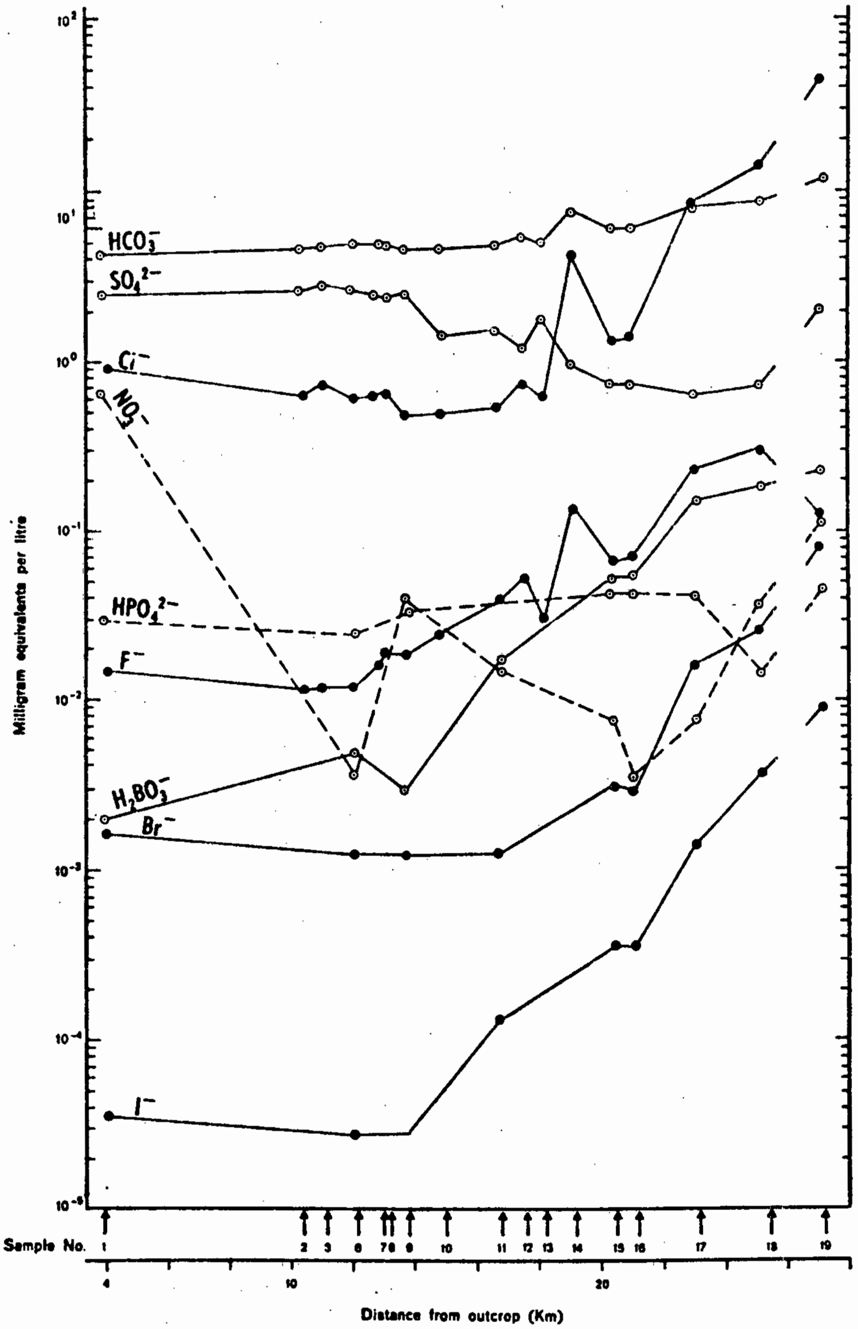

Figure 5. Variation in concentration of major and trace anions with distance from the outcrop.

increase in [HCO_3^-]. Calcium is replaced nearly stoichiometrically according to the reaction $Ca^{++} \leftrightarrow 2Na^+$. The concentration of Fe^{++} rises at first but drops again as the pH rises. The data for the heavy metals (Mn, Zn, Cu, Pb) are the most interesting. "No correlation with variation in total iron or with the processes occurring in the aquifer can be recognized. The level of occurrence of most trace heavy metals will be controlled by the very low solubility of the metal sulphides, as well as by the availability of the elements in the limestone. It is apparent, therefore, that the concentrations of some metals are higher than those expected if equilibrium with sulphides were attained; and this fact can be interpreted as indicating a kinetic delay in reactions at the low concentrations". Although Edmunds does not say so explicitly, the implications for hydrogeochemists who are intent upon applying equilibrium chemical modelling to trace element distributions in natural groundwaters are obviously unfavourable. The actual controls involved are patently very complicated.

Edmunds also records a notable attempt to estimate the degree of metal contamination from a well casing; this is probably one of the few studies of this sort in the literature. A sealed artesian well that had not discharged any water for over a year was opened and allowed to flow freely for twenty-four hours, during which period samples were collected at regular intervals. The concentrations of iron and aluminium decreased with time. The values for F, Cl and Mo remained roughly constant. The amount of Zn and Ni rose, while [Cu], [Mn] and [Pb] fluctuated (Table 3).

Table 3. Variations with time of selected trace metals in ground waters from a borehole at Spalding (No. 18).

Time from start of test (28.5.69)		Start	10 min	40 min	90 min	300 min	480 min	1060 min	1310 min
Cl	mg/l	468	496	510	504	488	512	472	490
F	"	5.6	5.4	5.6	5.6	5.6	5.7	5.7	5.6
Fe	μg/l	700	380	220	140	80	70	60	75
Zn	"	280	260	260	200	200	400	545	320
Cu	"	14	20	16	10	10	20	20	14
Mn	"	2	n.d	n.d	9	n.d	n.d	n.d	2
Al	"	230	n.d	n.d	10	n.d	n.d	n.d	14
Pb	"	3	n.d	n.d	1	n.d	n.d	n.d	7
Ni	"	1	n.d	n.d	1	n.d	n.d	n.d	2
Mo	"	1	n.d	n.d	1	n.d	n.d	n.d	1

n.d = not determined

"It is apparent that there is no large solution of trace metals from the lining tubes when they contain standing water; concentrations higher than those predicted by sulphide solubilities are found, however, equally in static and flowing samples". The reviewer would fain offer the observation that these results indicate once again how frequently hydrogeochemical studies fail to yield rational data that can be used to substantiate reasonably simple models.

The phenomenon of generally decreasing Eh during the cycle of infiltration, migration and discharge of groundwaters was also discussed in somewhat more general terms by Champ *et al.* (1977/986/).

The relationship between complex formation and the hydrogeochemical mobility of the elements has been discussed repeatedly and at length in the earlier sections of this review and for this reason the examination of the subject in this chapter will be restricted to a look at just one outstanding germane publication - that of Shvartsev *et al.* (1975/1102/). These authors recently summarized the vast accumulation of factual data that is now available from the analyses of groundwaters collected in the Soviet Union for purposes of mineral exploration. Table 1 (data columns 1 and 2), which is based on many Soviet studies, is a summary of data for fairly neutral, non-anomalous waters of both oxidizing and reducing (gley) types. These values are quite low and in the opinion of the authors this "reflects the dynamic equilibrium established between the solution and the rocks in the zone of active water replacement as a result of different biogenic process". The waters appear to contain more Zn, B, F and Br and less Ra, Th, Au and Bi than is consistent with this "dynamic equilibrium". The authors point out that many researchers believe that the microcomponents of water come from rocks, but noted that "we do not know actually what part of microcomponents in water are due to soils and what part to enclosing rocks". In some areas at least the contribution of the soil is dominant. The contribution from atmospheric sources is usually insignificant.

This picture changes remarkably in the presence of ore mineralization. Table 1 (data columns 3 and 4) shows the anomalous values and contrast ratios observed in Soviet groundwaters. In almost all cases the contrasts are very high. Thus even in neutral waters, where many microcomponents are subject to hydrolysis, groundwater hydrogeochemical prospecting is clearly a very favourable possibility. It is important to note that equilibrium-solubility calculations very frequently underestimate the amounts of various elements that can exist in solution in neutral waters (cf. Table 1 with Table 2). This is rather sobering, as it underscores the inadequacy of our present models of mineral/solution solubility equilibria in dilute waters. "The mobility of elements is determined by their capacity for complex ion formation" and not by the hydrolysis of the simple aqueous ions. "It is precisely this capacity of elements to form stable complex compounds, which makes it possible to explain the high contents of hydrolysate elements in subsurface waters, and consequently their increased migration ability as well". Thus simple ions of the hydrolysate elements are seldom found, whereas complexes with Cl, F, Br, sulphate and others are very common. For example, strong complexes form between F^- and Be, Nb, Ti and Th in neutral waters. Gold, Hg, Pb and Ag form chloride ligands, hydroxo-complexes solubilize V, Sn, Sb and Zr, and U and Cu enter into combination with the bicarbonate ion. Organic compounds - humic, fulvic, oxalic and lactic acids, and others - also form "easily soluble complex compounds" with many trace metals. Stated in different terms, "the formation of soluble complex compounds raises the pH of precipitation of the hydroxide, and consequently, enhances the migration capacity of the elements .....". The authors note that some elements may migrate in colloidal, i.e. non-dissolved forms.

4.524 Organic matter, gases, particulate matter, thermal/mineral waters

None of these topics is regarded as being part of the mainstream of this review and they will be dealt with only briefly.

According to Bukova and Nikitina (1964/809/) the organic matter of groundwater has been relatively little studied. These workers analysed natural waters from upland areas and found that they contained from 1,2 to 7,2 mg/l* of water soluble organic acids (as CH_3COOH). The concentration

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* Some exploration hydrogeochemists may be slightly amazed to find that organic matter is often, by definition, a major component (>1 mg/l) of groundwater. Organic compounds are usually ignored in exploration surveys and are assumed to be present in negligible amounts. In fact it is not uncommon for the concentration of the soluble organic compounds to exceed those of dozens of metallic microcomponents.

TABLE I

The most frequently encountered and maximum contents (in ppb) of a number of microcomponents in neutral waters of the supergene zone

Element	Most frequently encountered contents		Maximum contents found	Contrast ratio
	Waters of oxidizing setting	Waters of gley setting		
Barium	2.0 — 10	2.0 — 2.5	500	400
Beryllium	0.1 — 0.8	0.05— 0.3	300	3,000
Boron	10 — 50	—	20,000	2,000
Bromine	20 — 200	—	20,000	1,000
Vanadium	0.5 — 2.0	0.5 — 1.5	90	180
Bismuth	<0.1	—	5	50
Tungsten	0.1 — 0.5	—	200	2,000
Germanium	0.3 — 0.8	—	30	100
Gold	0.02— 0.1	—	5	250
Iodine	1 — 10	—	5,000	5,000
Cadmium	0.1 — 0.5	0.05— 0.5	260	5,200
Cobalt	0.5 — 3	0.2 — 10	68	340
Lithium	5 — 10	—	10,000	2,000
Manganese	10 — 50	20 —400	15,000	1,500
Copper	2 — 8	1 — 5	1,000	1,000
Molybdenum	1 — 5	0.2 — 3	8,000	40,000
Arsenic	1 — 5	0.1 — 2.0	100	1,000
Nickel	0.8 — 5	0.5 — 2	200	400
Niobium	0.5 — 2.0	—	—	—
Tin	0.1 — 0.5	0.1 — 0.5	60	600
Radium	$(1–5) \times 10^{-6}$	—	10^{-3}	1,000
Mercury	0.5 — 3.0	—	20	40
Rubidium	0.2 — 5	—	300	1,500
Lead	1 — 8	0.5 — 4	250	500
Selenium	0.5 — 1.0	—	50	100
Silver	0.1 — 0.6	0.05— 0.5	50	1,000
Strontium	5 — 50	3 — 20	1,000	300
Antimony	1 — 10	0.5 — 5.0	1,300	2,600
Titanium	1 — 5	0.5 — 40	3,400	6,800
Thorium	0.02— 0.1	—	—	—
Uranium	0.5 — 5	0.3 — 3.0	500	1,700
Fluorine	50 —1000	—	11,500	230
Chromium	5 — 10	0.1 — 5	600	6,000
Cesium	0.1 — 1	—	50	500
Zinc	5 — 50	2 — 20	2,500	1,250
Zirconium	1 — 10	1 — 5	450	450

TABLE II

Calculation of the maximum possible concentrations of a number of element hydrolysates and elements which form slightly soluble compounds in neutral waters

Element	Form of migration without allowance for complex ion formation	Compound limiting the migration ability	Calculated maximal possible concentration with pH=7.0, ppb
Barium	Ba^{2+}	$BaSO_4$	205*
Beryllium	Be^{2+}	$Be(OH)_2$	1.4×10^{-5}
Vanadium	V^{3+}	$V(OH)_3$	1.0×10^{-4}
Bismuth	BiO^{3+}	Bi_2O_3	2.1
Bismuth	Bi^{3+}	$Bi(OH)_3$	1.2×10^{-4}
Niobium	NbO_2^+	NbO_2OH	0.2
Tin	Sn^{2+}	$Sn(OH)_3$	0.4×10^{-4}
Tin	Sn^{4+}	$Sn(OH)_4$	1.2×10^{-21}
Lead	Pb^{2+}	$PbSO_4$	2700*
Silver	Ag^+	$AgCl$	10**
Scandium	Sc^{3+}	$Sc(OH)_3$	45
Antimony	SbO^+	Sb_2O_3	0.014
Titanium	Ti^{3+}	$Ti(OH)_3$	4.8×10^{-12}
Titanium	TiO_2^+	$TiO(OH)_3$	4.8×10^{-8}
Thorium	Th^{4+}	$Th(OH)_4$	2.3×10^{-3}
Uranium	UO_2^{2+}	$UO_2(OH)_3$	5.0
Chromium	Cr^{3+}	$Cr(OH)_3$	0.035
Zirconium	Zr^{4+}	$Zr(OH)_4$	5.5×10^{-23}
Zirconium	ZrO^{2+}	$ZrO(OH)_3$	2.7×10^{-4}

* At a concentration of SO_4^{2-} = 48 ppm.
** At a concentration of Cl^- = 50 ppm.

of organic acids bore no obvious relationship to the type of water. Other soluble organics included medium molecular mass humic substances and fats. Even very fresh water contains organic acids in quantities in excess of the concentration of many trace metals. It must be assumed that the carboxyl groups of the acids will influence the mobility of many microcomponents by playing "an important role in the formation of chelate compounds". This was corroborated by Varshal and his co-workers (1975/840/), who showed that Ca and many rare earth elements are frequently present in natural waters as organic complexes. The nature of the organic substance present strongly affects the solubility and stability of the metallic complex and hence influences the migration of these elements.

Szalay (1969-1973/919-20/) continued his long series of experimental and observational studies of the effects of organic matter - especially humic compounds - on the mobilities of the elements. Figure 5 is a deceptively simple summary of many years of investigations by this author and his co-workers. Note that Mo and V* are the only metals known to be relatively resistant to sorption by humic molecules.

Dyck and Jonasson (1977/918/) investigated the possibility of using the gaseous components of groundwaters as indicators of mineralization. They studied the distribution of H_2, He, CH_4, O_2, H_2S, CO_2, Rn, N_2, Ne and Ar in various groundwaters around copper and uranium deposits in Quebec. It was possible to explain the distribution of most of these species in terms of geochemical principles but only Rn and He were obviously related to any kind of mineralization (uranium ore).

Kennedy *et al.* (1974/902/) undertook a notable study of the distribution and abundances of metals in particulate matter of varying sizes suspended

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* Dall'Aglio *et al.* (/988/, Section 4.526) disagrees and claims that V is not resistant.

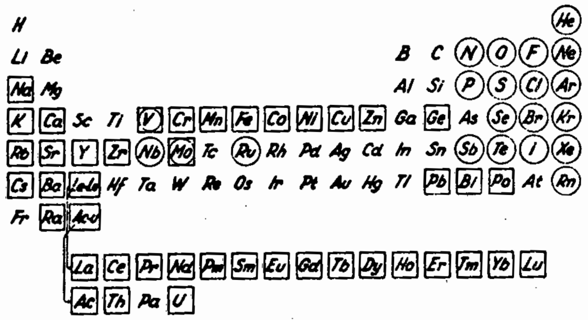

Fig. 5. Interaction of insoluble humic acids with elements of the periodic table. Elements sorbed in squares, those certainly not sorbed in circles. V and Mo are sorbed after reduction of anionic migrating form into cations. Others not investigated yet.

in natural water. Several writers have observed that the microcomponents of natural waters, including subterranean waters, are not always in true solution.

Exploration hydrogeochemists other than oil prospectors have never paid a great deal of attention to thermal or mineral waters and a general feeling has grown that it will not be easy to use these waters as guides to metallic deposits. Dall'Aglio and Tedesco /913-4/ discussed some of the chemical features of the thermal springs of Sicily and, by implication, summarized the reasons why it is difficult to use waters of this type in hydrogeochemical exploration for ores. They studied the composition of Sicilian spring waters between about 1967 and 1969. The distribution of bicarbonate ion was fairly constant, but [NaCl] varied greatly because of the occurrence of buried evaporites in some areas. The concentrations of most of the major components increased with TDS, except for K and Si, which remained remarkably low. Dall'Aglio *et al.* concluded that the composition of the groundwater is extremely dependant on local lithology and that there is an intense exchange between surface waters and groundwaters in some areas. It was noted that earthquakes could cause radical changes in the compositions of spring waters - e.g. marked increases in TDS or in the concentration of a given ion - and that these changes can persist for more than a year after the earthquake episode. Gorbushina *et al.* (1972/925/), who studied the mineral waters of the Tashkent Artesian Basin, confirmed that the accumulation of stress in rocks could cause slow, progressive changes over many years in the composition of groundwaters and that the release of this stress as a shock wave caused radical changes in water chemistry. Uranium and fluorine are amongst the elements particularly affected in this manner.

Despite these and other limitations, further attention might profitably be given to the possible uses of thermal/mineral waters in ore prospecting, especially on the broader regional scale. James (1967/880/) suggested that the metal content of thermal springs might be used to delineate metallogenic provinces.

4.525 Anions, halogens, sulphate, boron, alkali metals, alkaline earths

Over the last fifteen years there has been a fair amount of additional research into the role of anions in groundwater hydrogeochemical exploration. The halogens appear to have received particular attention. The Russians, in particular, seem to have used halogens such as Br and I as pathfinders with some success, but have pointed out that this requires a good understanding of the possible sources and hydrogeochemical behaviour of these elements. Moshkina and Nikol'skaya (1964/814/) studied the behaviour of some halogens and other microcomponents during the metamorphism (natural ageing) of artesian waters of the Irtysh Basin. As the waters change with age and distance of migration Li, I and especially Br accumulate while B and F are removed from solution. The concentration of arsenic remained very low in almost all waters - typically 0,1 to 2 µg/l. Krasintseva (1964/817/) noted that the Cl/Br ratio is lower in the waters of older sedimentary rocks and believed that this reflects an increase in the Cl/Br ratio in the oceans through geological time. It is self-evident that Br background values might have to be adjusted for these effects in certain prospecting surveys.

Of the halogen elements, fluorine has received the most attention in recent years /963-969/. Again, most of the work in this direction has taken place in the U.S.S.R. /1099/. In several related publications (1973-1976 /830, 968, 1099/) Lalonde showed that halogens ought logically to form dispersion haloes around many types of orebodies (note the composition of fluid inclusions in ore minerals) and claimed that fluorine is a particularly promising indicator of such mineralization. He studied the dispersion of this element in the vicinity of fluorite-rich veins in Precambrian carbonate rocks (marbles) near Madoc in Ontario. Two hundred samples were collected from wells and springs and analysed for fluoride by means of an ion-sensitive electrode. A very strong correlation was observed between $[F^-]$ in groundwater and the location of the fluorite bodies. "Fluoride in groundwaters clearly delineates areas of known mineralization as well as other areas of possible interest". Examples of the anomalies observed at the Madoc fluorite veins are shown in Figures 4 and 5. Lalonde warns however that such anomalies "can change drastically within hundreds of feet" and could be missed if sampling points are too widely spaced.

The range of observed fluoride concentrations in the region was from 21 to 1800 μg/l. The distribution of the data was strongly and positively skewed and the author regarded this fact as a sign of mineralization in the area. Background concentrations of fluoride were clearly influenced by lithology and varied from 63 μg/l in volcanics to 115 μg/l in limestones. However, the concentration of fluoride was not influenced by the pH. Replicate collection and analysis of several samples showed that $[F^-]$ varied by less than 10% in a period of six weeks. However, six months later in autumn, after summer rains, both the background and anomalous values had shown a general but not uniform increase. Some values increased tenfold. These changes were attributed to the influence of the rainfall, but the reasons were not clear. The author suggested that the patterns of groundwater flow may have altered.

In the Abitibi area fluoride ion was used as a hydrogeochemical pathfinder for Cu-Zn-Ag sulphides and other deposits possibly buried under a formidable layer of lacustrian clay from 40 to 250 feet thick. Some 2500 orientation and reconnaissance samples were collected over a large area, from wells, drill holes and springs. These samples were analysed for 22 elements, including fluorine. The range of $[F^-]$ was 20 to 1200 μg/l and the threshold was taken to be 200 μg/l. Lalonde regarded the results as extremely favourable. Figures 15 and 16 show the fluoride anomalies associated with two gold deposits. The method has apparently delineated several new and interesting anomalies but the results were regarded as confidential at the time of publication of Lalonde's results. He does however mention that these anomalies, like those at Madoc, are not broad - seldom more than half a mile - and that high sampling densities must therefore be maintained.

Schwartz and Friedrich (1973/1108/) reported the results of hydrogeochemical exploration for fluorite deposits in parts of Spain. Fluoride was determined in spring and other waters. The background concentration was typically below 280 μg/l and values above 1 mg/l were considered to be anomalous. Seasonal variations did not affect $[F^-]$ in springs, but it did in large rivers. Two anomalous springs were found at Puig Rodo, and subsequently a new fluorite occurrence was discovered upslope from the springs.

Jacks *et al.* (1973/963/) investigated the fluoride content of groundwaters near skarn fluorite deposits in Sweden. The authors note that fluorite is quite soluble in water (>1mg/l) and that the concentration of fluoride ion in natural waters is therefore controlled by less soluble compounds such as fluorapatite. Some of these more complex fluorine minerals have solubilities of the order of only 1 μg F^- per litre. Thus the contribution of most minerals to the fluoride content of groundwater is generally

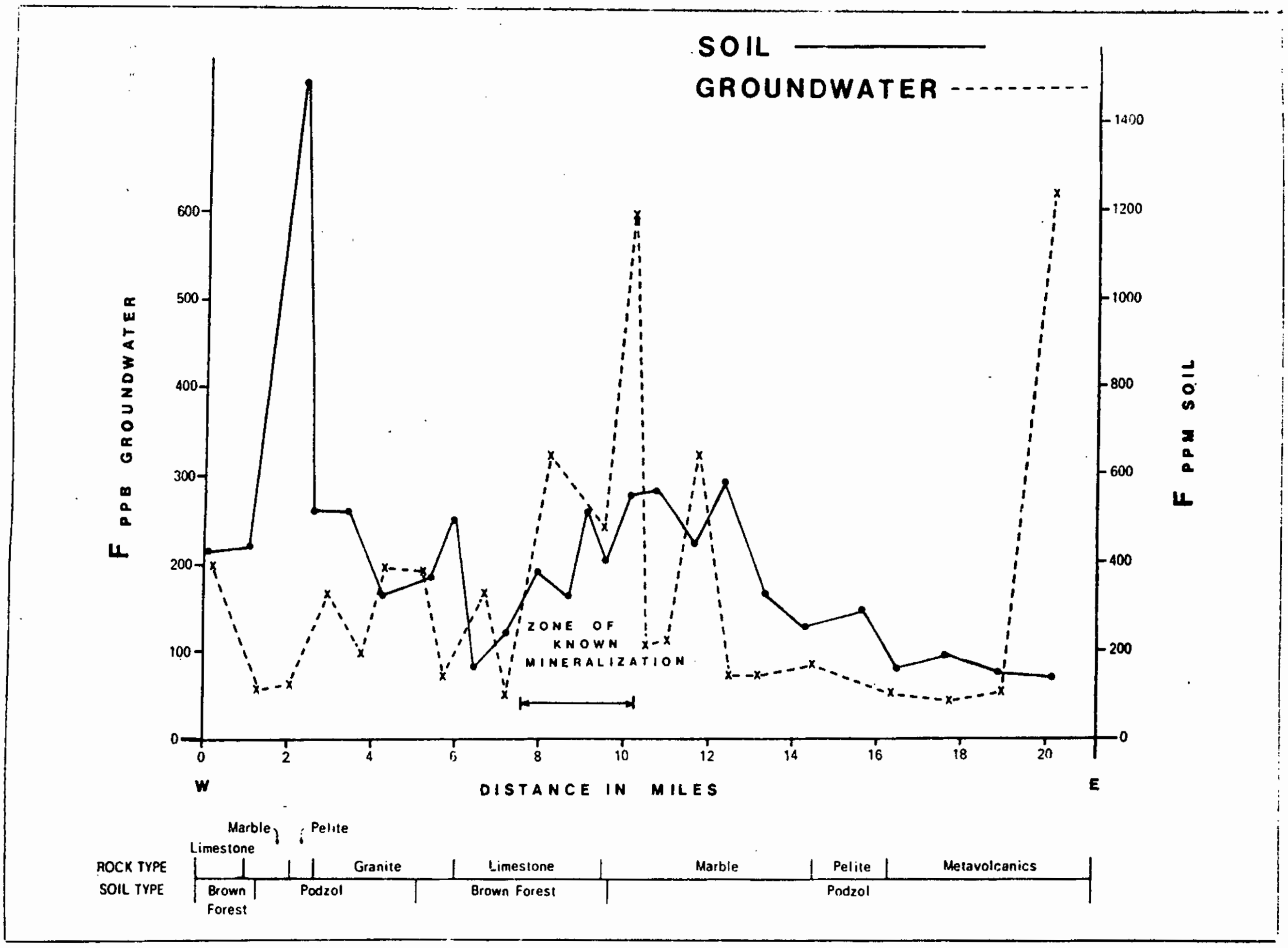

FIGURE 4 — The fluorine content of the A soil horizon and groundwater along the **northern** regional traverse.

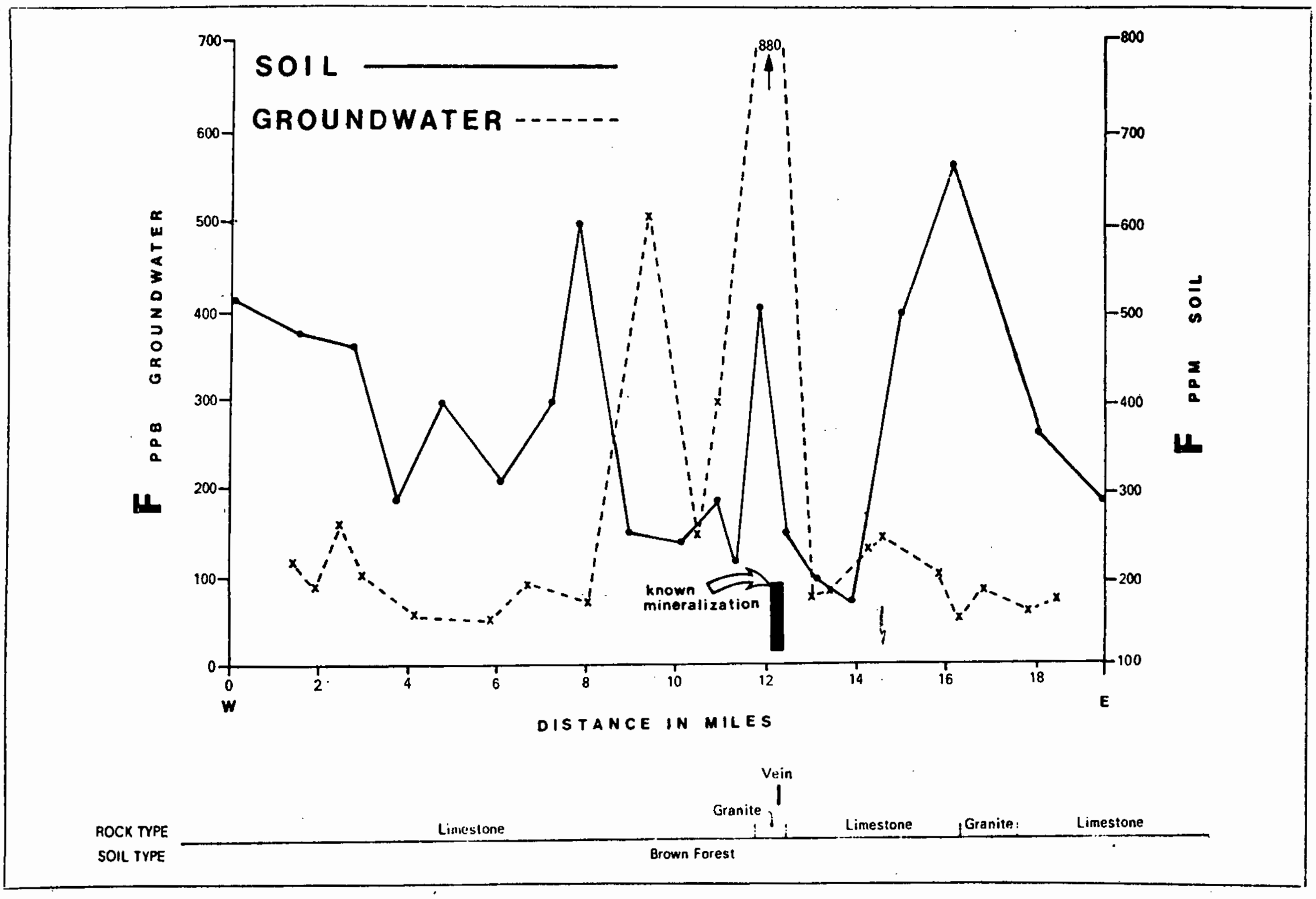

FIGURE 5 — The fluorine content of the A soil horizon and groundwater along the **southern** regional traverse.

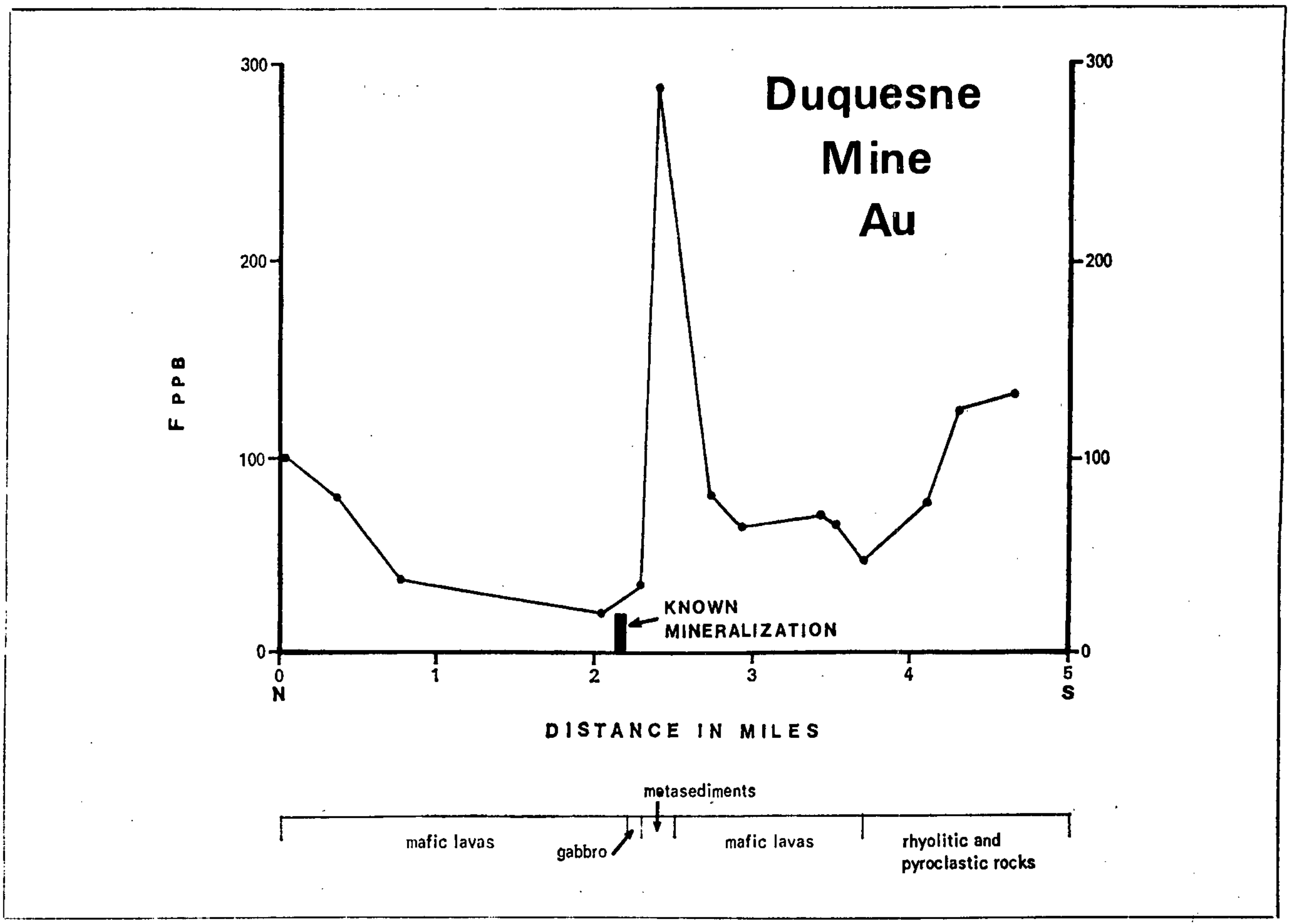

FIGURE 15 — Fluorine in ground waters at the Duquesne mine.

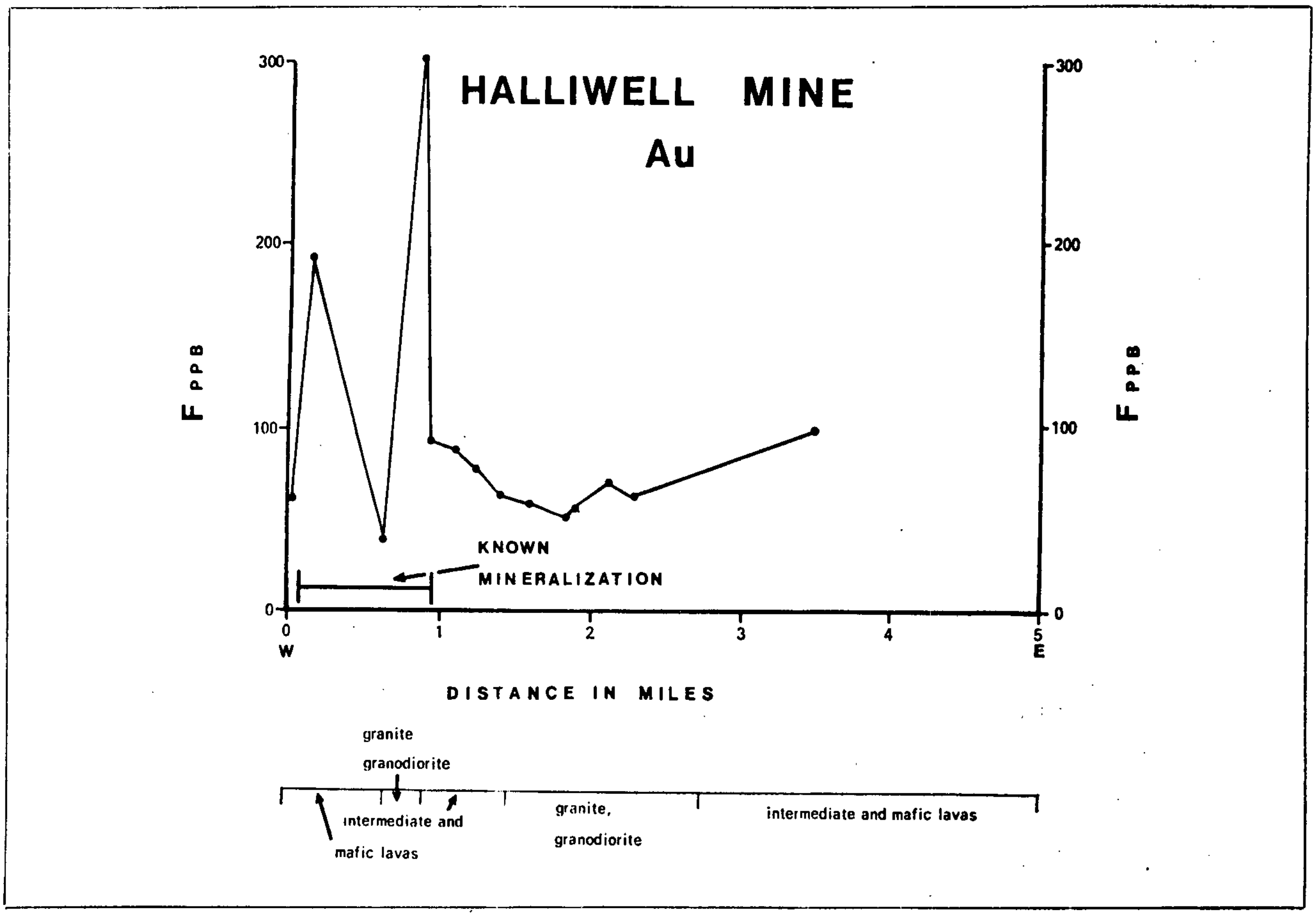

FIGURE 16 — Fluorine in ground waters at the Halliwell mine.

very small and this is why background values for the element are commonly low. Much of the fluorine in circulation in the hydrological cycle is apparently of marine origin and in Swedish well waters the background concentrations of fluoride are highest in marine clays. The fluoride ion is more reactive than the chloride ion and will become attached to many types of solid materials or will form complexes such as MgF^+. Thus the fluoride content of water samples is inherently unstable and analysis should therefore be performed as soon as possible.

In the Swedish study the background concentration of fluoride ion was found to be about 0,2 mg/1. Most samples had less than 0,8 mg/1 but some samples taken near active mines contained as much as 10 mg F^- per litre. Some displacement of the groundwater anomalies was observed. The authors note that fluoride is also associated with some tungsten and molybdenum deposits and that fluorine may therefore serve as a pathfinder for such ores.

Graham *et al.* (1975/964/) observed that fluorine is not a common contaminant of natural waters and that this element therefore enjoys some advantage over zinc in certain situations such as, for example, wellwater surveys. These authors noted greater concentrations of fluorine in groundwaters (up to 15 mg/1 than those observed by Jacks *et al.* (*vide supra*). Their thermodynamic data also differ from those of Jacks *et al.*

It is noted that ions such as Al^{3+}, Fe^{3+} and Si^{4+} have the ability to form soluble complexes with fluorine and thus increase the amount of dissolved fluoride that can exist in equilibrium with fluorite. But since these ions are scarce in natural waters very little F^- is actually present in groundwater in the form of complexes. On the other hand high concentrations of Ca or phosphate ion will definitely depress $[F^-]$. In the Madoc area of

Ontario, where Graham and his colleagues worked, the apparent solubility product of fluorapatite was not exceeded in any but a very few samples. The authors caution that fluorine cannot be used as a pathfinder in those areas where the background concentration of the fluoride ion is severely limited by the insolubility of fluorine minerals. Obviously, if the water is saturated with fluoride even in the absence of fluorine-bearing ores then even a massive excess of fluorine in the orebody cannot produce a hydrogeochemical anomaly.

It is claimed that the "typical range" of $[F^-]$ in groundwaters is 1,5 to 0,05 mg/l. Seasonal variation studies (resampling after six months) at the Madoc deposits were very encouraging. Only thirty percent of the samples showed variations outside of the 95% confidence interval surrounding a perfect correlation between the two sets of data. The "seasonal variation is less than the analytical variation in most cases and ..... long term fluoride surveys are feasible". In the Madoc district the fluoride anomalies are not really displaced but are asymmetric, with a "tail" due to hydrological dispersion in the direction of groundwater movement. The peak-to-background values here were typically between 4 and 10 and the anomalies were a mile wide. Zinc was determined in the same samples but was found to be a poor pathfinder.

Although fluoride has stolen the limelight in recent times, the "grandfather" of the anionic pathfinders, sulphate, was by no means neglected. The most recent comprehensive assessment of the sulphate ion in groundwater hydrogeochemical exploration is that of Hoag and Webber (1976/1100/). They classified the origin of dissolved sulphate (in the absence of species such as $CaSO_4$) into three groups: (i) Atmospheric precipitation. This is equal to 0,04 mmole/l in the study area, which was at the Eustis Mine, Quebec.

(ii) Inorganic subsurface oxidation. This depends upon the solubility of atmospheric O_2 (in the absence of active diffusion or circulation of oxygen) and cannot normally exceed 0,25 mmole/l. (iii) Inorganic/organic surface oxidation. The inorganic reaction rates are significantly moderated by kinetic factors and occur principally at $pH > 4,5$. Below this value the bacterial oxidation processes dominate and the reaction may be very rapid.

It follows that waters with less than 0,29 mmole $SO_4^=/l$ have probably not been in contact with shallow, oxidized sulphides. Intermediate concentrations of sulphate suggest the influence of inorganic surface oxidation. This process will not produce gossans and in such areas supplementary pedogeochemical sampling or trenching might reveal shallow mineralization. High concentrations of sulphate indicate the likelihood of organic oxidation of sulphides and the concomitant development of gossans.

Hoag and Webber also determined zinc in their samples but deemed the results to be dubious because of the effects of contamination. It was suspected that many samples had picked up zinc from borehole hardware or from mine waters.

Gleeson and Boyle (1976/873/) found that sulphate was a useful pathfinder even at very high latitudes. They described a major groundwater hydrogeochemical survey in the Keno Hill area, Yukon, where 172 samples of spring waters and other groundwaters were collected. The authors used moving-average/residual element maps to process the data and found that $[SO_4^=]$ and $[SO_4^=]/[Cl^-]$ outlined the Pb-Zn-Ag mineralization that occurs in the area. The distribution of chloride was governed only by the lithology of the country rocks and was of no significance in prospecting. Spring waters associated with ore were enriched in bicarbonate, sulphate, Si, F and Zn and had high TDS values. Interestingly, they had high pH and were depleted in copper. The authors emphasized that high TDS in wet areas is often an overlooked indication of mineralization. This publication contains a useful list of references to relevant Canadian literature.

Boron is included in this section because of its tendency to form anions. Oborin and Zalkind (1964/816/) published what may still be the definitive article on the supergene geochemistry of boron, with over one hundred (mostly Russian language) references. This is an element little used in hydrogeochemical exploration, although its concentration in, for example, thermal spring waters of oilfield waters may be useful in prospecting for borate deposits. However, boron is also concentrated (3 to 5 times background) in the waters of some metallic orebodies, e.g. those of the western Urals. Values of up to 748 μg/l were observed here, but the authors note that in some humid areas the background values for boron can be as much as 3 mg/l.

Predictably, the elements of Groups IA and IIA received little attention. Kraynov *et al.* (1966/1026/) discussed methods of groundwater hydrogeochemical prospecting for ores of beryllium. Anomalies of up to a few hundred micrograms per litre were observed. The element appears to be mobile in the form of simple and complex cations but its movement is severaly restricted by clays within the zone of weathering. But in deeper waters, especially carbonate waters, beryllium is very much more mobile and can be used to identify, for example, Be bearing pegmatites in felsic rocks.

Skougstad and Horr (1963/954/) gave a comprehensive account of the distribution and geochemical behaviour of strontium in groundwaters. The element could be used to prospect for strontium minerals since it is relatively mobile and easy to detect. There has, however, been very little interest in applying the technique.

Price and Ragland (1970/1116/) conducted a prospecting survey of groundwaters in North Carolina and found that major lithological units were clearly delineated by the chemistry of these waters. The Kings Mountain lithium producing belt was roughly outlined by a belt of lithium-rich groundwater. The [Zn]/[major cations] ratio could be used to distinguish between sulphides and deposits of barite.

4.526 Some less commonly sought elements. As, V, Cd, Se, Te, Sb, Sn, Ag, Au, Hg.

Ratynskii (1975/839/) presented a general but very useful summary of the hydrogeochemical characteristics of several elements that are not commonly used as pathfinders. He claimed that careful work will often show that some of these "obscure" elements are relatively concentrated in anomalous groundwaters near many different types of orebodies, even though their absolute abundances in these waters may be very low. The patterns of concentrations of these obscure elements, like the elements more commonly sought in groundwater, vary greatly from one area to another and the author uses several examples from the U.S.S.R. to demonstrate this fact.

Germanium is more mobile than Si and is often concentrated in thermal waters (often up to 30 μg/1; maximum 200 μg/1). Boron and arsenic also appear to be much more mobile in hotter waters. The concentrations of boron and bromine vary widely in normal groundwaters. Tungsten can form dispersion haloes in alkaline waters near deposits containing the metal (up to 200 μg/1). Beryllium is mobile in carbonate waters (up to 400 μg/1). Niobium may be found in fissure waters in syenites. Its concentration rises as pH, [CO_3] and [Cl] rise and can reach 200 μg/1. The following maximum amounts (μg/1) of rare earth elements have been observed in alkaline waters from the Lovozero Massif: Ce 380, La 260, Yb 3, Y 27. Lithium is most commonly abundant in chloride-rich waters. Little is known about the concentration of Rb in groundwaters. There may be as much as 3 mg Cs/1 in mineral waters, but the metal is very scarce in more dilute subterranean waters. The abundance of strontium in natural waters varies widely. Dilute spring waters contain nanogram amounts of zirconium.

It is concluded that under special circumstances some of these elements may be useful as pathfinders and that geochemists may be underestimating the mobility of many of them. Ratynskii notes that a large number, if not all, of these elements will tend to be stripped from most normal groundwaters by organic matter.

Boyle and Jonasson (1973/1105/) summarized those hydrogeochemical methods that involve the determination of arsenic. Only a few such surveys have been made, but it is clear that the element may be greatly enriched in groundwater near arseniferous ore deposits. Typical data are given in Table XII. The element is amphoteric and stable in either acidic or alkaline water. The speciation and mobility of As are summarized in Figure 3. Some Soviet writers found As to be quite mobile in water and the element was used as a pathfinder for Cu-Pb-Zn deposits. In the Cobalt area of Ontario [As] in both ground- and surface waters was an indicator of Ag-Ni-Co arsenide ores but could not pinpoint the location of the deposits with any accuracy. It was therefore to be regarded as a potentially useful regional reconnaissance tool. In addition, the determination of As in drill hole waters "may be useful in detailed drilling programs to ensure that faults, fractures, and shear zones carrying water and containing orebodies above and below the drill intersections are not missed".

Spring waters and their precipitates are useful guides to ores containing arsenic. However, false anomalies are common in areas of disseminated arseneous pyrite. Arsenic anomalies in thermal waters are frequently false but can sometimes be associated with arsenic ores that are in the process

TABLE XII

Arsenic content of natural waters in parts per billion

Water type	Number of values	Range	$\bar{X}$	S	$\bar{X}g$	Sg ($\log_e$)
Rainwater and snow	48	0.01–13.9	1.44	2.17	0.81	1.11
Normal stream, river, and lake waters	88	0.25–22,400	3.08	5.42	1.75	0.94
Normal ground waters	45	0.01–800	17.9	13.9	8.0	1.80
Ground waters and mine waters near polymetallic sulphide deposits	102	3–400,000	–	–	–	–
Oilfield and other saline waters	11	10–243,000	–	–	–	–
Hot springs	92	0.2–40,000	2,090	2,650	1,500	1.23
Condensed waters from volcanic gases	11	63–812	486	263	395	0.78
Spring waters; mainly cold carbonated waters in volcanic terranes	17	120–37,500	22,200	34,800	5,000	2.29
Spring waters depositing travertine	4	30–500	307	233	197	1.33
Thermal waters associated with epithermal mineral deposits	4	50–200	117	74	99	0.69
Ocean and sea	183	0.0056–11.24	2.57	1.98	1.77	1.11

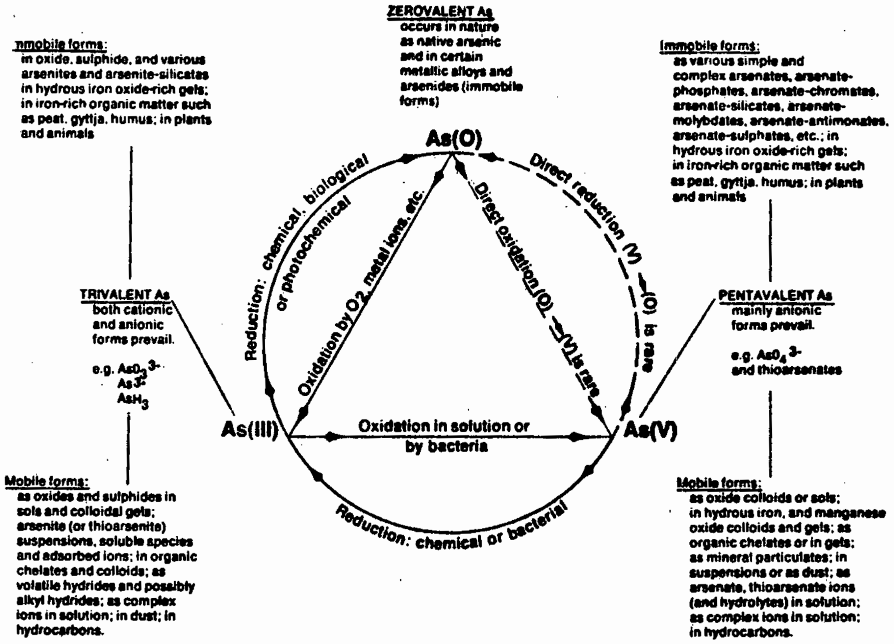

Fig.3. Cycle of arsenic interconversions of nature.

of formation. Arsenic in groundwaters is also a potential regional guide to geothermal fields.

Dall'Aglio *et al.* (1977/988/) described the essential hydrogeochemical properties of vanadium, which is an interesting but little studied element. Under humid tropical conditions vanadium is not as mobile as uranium but under arid conditions the mobilities of the two elements are roughly equal. Dissolved vanadium is particularly sensitive to organic matter.

A publication by Smirnov (1963/802/) was devoted to the behaviour of cadmium in supergene waters. The hydrogeochemistry of this element is poorly known. It is invariably associated with Zn ores and is potentially useful as a pathfinder because its natural background abundance is very low - generally 1-10 μg/1. Elevated concentrations of cadmium are encountered in oxygen-rich waters near several Zn-Pb orebodies in the U.S.S.R. In these waters the metal is more abundant than Ni, Co, Mo, Sn or Be. The destruction of sphalerite leads to the accumulation of large concentrations of cadmium in acidic mine waters (up to 40 mg/1). However, the metal is not quite as mobile as zinc and as a result the Zn/Cd ratio increases in "the supergene solutions as compared with sphalerite". High [Cd] is therefore associated with high [Zn] and low pH, especially below a value of 6,0. This dependence on pH is not related to the hydrolysis of cadmium, which is quite stable at pH 7 or 8, but is due to an indirect control. Smirnov shows that there is an interesting relationship between [Cd] and the sulphate/bicarbonate ratio, which is pH dependent and which the author regards as an "index of metamorphism" of the waters associated with orebodies. This ratio, he claims, is "an exact indicator of the chemical evolution of supergene solutions in a vertical section of the oxidized zones of sulfide deposits", and it drops as the mine waters are diluted or neutralized. It follows that it may be the presence of sulphate and not the pH that is the dominant factor in the migration of Cd (and Zn). Nevertheless, the presence of sulphate is not enough to prevent the precipitation of considerable quantities of zinc and cadmium carbonates between pH 6 and 7. If this precipitation is severe then neither Cd nor Zn will be much mobile than lead. Traces of these metals that remain after precipitation as carbonate or co-precipitation with other carbonates may be removed from solution by adsorption onto wall rock materials. Despite these limiting factors, "cadmium must be regarded as a very important indicator of sulfide mineralization".

Faramazyan and Zar'yan (1964/808/) examined the abundances of S, Te and Se in mine waters in porphyritic granodiorites containing bodies of molybdenite-chalcopyrite ore. The mean concentrations (mg/l) were S 2790, Se 0,64 and Te 0,045 and the relative proportions of the three elements were 60 000:14:1. There has been scant interest in hydrogeochemical prospecting methods involving Te or Se.

Antimony is another element that is hardly ever sought in hydrogeochemical exploration surveys. Boyle (1975/836/) examined the solution chemistry, supergene behaviour and hydrogeochemistry of this element. He determined [Sb] in several hundred water samples from the vicinity of Pb-Zn-Ag ore lodes in the Keno Hill area. These waters have a temperature of two to five degress Celsius and a pH of between 4,4 and 7,8. The Eh is high near the surface but drops rapidly with depth. Minerals containing antimony are readily attacked in the oxidized zone but the element is not dispersed and is largely reprecipitated by a variety of secondary processes, of which hydrolysis seems to be the most important. This accounts for the low absolute and relative abundances of antimony in these waters (Table 9). "The low content of

TABLE 9. Dissolved constituents in underground waters, Keno Hill-Galena Hill area

Constituent	Content (range in ppm)	Constituent	Content (range in ppm)
Calcium (Ca^{2+})......	50 - 200	Cadmium (Cd^{2+})	0,001 - 0,5
Magnesium (Mg^{2+})	10 - 120	Arsenic	0,002
Sodium (Na^{+})	1 - 3	Antimony	<0,001
Potassium (K^{+})	0,4 - 0,6	Silver (Ag^{+})	∿0,001
Iron (Fe) (total) ...	1 - 5	Bicarbonate (HCO_3^-) ...	5 - 100
Aluminium (Al^{3+})	1 - 15	Sulphate (SO_4^{2-})	200 - 1200
Manganese (Mn^{2+})	5 - 25	Chloride (Cl^-)	0,1 - 1,0
Copper (Cu^{2+})	0,001 - 0,65	Fluoride (F^-)	0,3 - 0,5
Zinc (Zn^{2+})	0,001 - 80,0	Nitrate (NO_3^-)	0,1 - 0,6
Lead (Pb^{2+})	0,001 - 1,00	Silica (SiO_2)	5 - 10

antimony in the ground and surface waters makes the element generally unsuitable as an indicator in hydrogeochemical prospecting methods .....".

The reviewer knows of no serious recent proposal to use groundwater hydrogeochemical methods to locate tin deposits. The very little work that has been done on Sn in surface waters would suggest that the prospects for tracing the metal in subterranean solutions are very poor. Heide and Reichardt (1975/837/) summarized the hydrogeochemical properties of tin. The tin content of natural waters is extremely low - generally less than 1 µg/l and it is usual to find that more than half of this is in suspension.

Boyle is one of the few hydrogeochemists who has recently examined the possibility of using the silver content of groundwaters as a pathfinder /818, 826, 1021/. Since the concentration of this metal in natural waters is very low (commonly below 1 µg/l) the chief problem for a long time was simply analytical. For this reason most of the early workers who applied hydrogeochemical methods (springs and streams) of prospecting for silver deposits tended to opt for Zn and Co as practically effective pathfinders. Boyle determined silver and other components in the waters of springs and drill holes near dispersed silver veins in altered sediments in the Cobalt area of Ontario. Both the waters and the precipitates from these springs are enriched in silver and a number of other metals. Some well waters were also anomalous. Cobalt and Ni were particularly abundant, the latter being the less mobile of the two.

In about 1971 a well water survey was undertaken in the vicinity of the Argent silver prospect, near Johannesburg /1143-4/. The samples were preserved with nitric acid (3ml per litre) and stored in polyethylene bottles. No contamination problems were encountered and this was ascribed to the fact that most of the well casings were thickly coated with calcium carbonate deposits. The depth of the boreholes could not as a rule be established and no other hydrological information was available. There was very little apparent biological activity in any of the holes. The water temperatures were almost constant and were roughly the same as the ambient air temperatures. The samples were analysed for chloride, total sulphur, iron, Cu, Zn, Hg, Ag, Co, Sb and In by atomic absorption and neutron activation methods. Some of the results have been released and are compiled in Table A. It was suspected that Sb was being leached from the containers. In general, the samples

TABLE A

Neutron activation analyses (μg/l) of groundwater samples from the Argent prospect

	range	mean
Zn	11 - 159	80
Co	1 - 7	3
Sb	1 - 10	1
Cu	4 - 174	not stated
In	trace	

were found to be rather iron-rich. The concentration of zinc showed marked seasonal variations, especially in those boreholes with high [Zn]. The data were assessed by simple contouring methods and the results of the survey were not encouraging.

Additional studies that were carried out in the Transvaal at the same time, but which were not made public, suggested that Se might serve as a hydrogeochemical pathfinder for platinum deposits such as the Merensky reef in the Bushveld igneous complex.

Borovitskiy *et al.* (1966/1024/) were very encouraged by the results of groundwater hydrogeochemical surveys for gold deposits in the Aldan region the U.S.S.R. They found that the gold haloes were small but well-defined. Silver, Ti, Sn, As, Sb, and Te were supporting pathfinders. The authors believed that the hydrogeochemical technique "has shown its advantage over other known methods of gold determination and indirect indications of gold ore deposits".

These findings are in direct contrast to those of Gosling *et al.* (1971/1027/) who stated that the concentration of dissolved gold had a small range, was not related to the type of water or the lithology of the country rocks, and that direct hydrogeochemical exploration for the metal does not appear to be promising. This apparent discrepancy may be explained in terms of the findings of Ong and Swanson (1969/1046/). Equilibrium calculations show that the concentration of dissolved gold in dilute natural waters ought to be about 2-20 ng/l, and at pH 4 most of this would be $AuCl_2^-$. However, in certain acidic chloride waters [Au] can reach several mg/l. The reduction of this gold compound, by organic acids for instance, will result in the formation of a stable colloidal suspension of metallic gold. The size of the particles in this gold sol will normally be less than 10 nm and each mote will be stabilized by a sheath of organic molecules. It has been shown that this organometallic sol is stable for at least eight months - long enough to make hydrogeochemical prospecting for gold an attractive possibility.

Boyle *et al.* (1975/1133/) also undertook a detailed examination of the solubility of gold. They concluded that gold is indeed transported in the form of humic complexes and that it can also be transported in ferric sulphate solutions derived from the oxidation of sulphides. This paper contains a long list of references to previous studies of the solubility of gold in various natural solutions.

In recent years there has been a revolution in techniques for the determination of minute amounts of mercury /829/ and this has undoubtedly made this element more attractive as a practical indicator in hydrogeochemical exploration. However, the problems involved in using this unique metal as a pathfinder are more than just analytical, as many workers in all branches of exploration geochemistry have found. Zautashvili (1966/1023/) investigated the dispersion of mercury in waters in andesites and shales containing mercury deposits. Although conditions were apparently favourable for dispersion, [Hg] did not exceed 3 μg/l and the "aqueous aureoles" were never wider than fifty metres.

Some of the pitfalls involved have been outlined by Jonasson and Boyle (1972/956/), who presented a fine review of the hydrogeochemistry of mercury and the use of the metal in prospecting. Mercury is associated with many ore deposits and is potentially very mobile. Therefore it can often be used as a powerful pathfinder. However, the liberation of the element and its migration as Hg(I) or Hg(II) is heavily dependent upon the prevailing Eh-pH regime and the anionic species available. Mercury tends to be mobilized by natural oxidants such as Fe(III) and readily forms organometallic compounds. Groundwater typically contains from 0,01 to 0,1 μg Hg/l but for mineral waters the range is wider - from less than 0,01 to 2,5 μg/l. Some mine waters have been found to contain as much as 230 μg Hg/l. The element is stripped from solution by both organic and inorganic absorption reactions and is rapidly immobilized under strongly reducing conditions, especially by H_2S.

Elemental mercury is soluble to the extent of about 60 μg/l in water and is known from mine waters and some springs. Disproportionation occurs readily, according to the reaction

$$2Hg(I) \rightleftarrows Hg(0) + Hg(II)$$

and the speciation of mercury is therefore unusually sensitive to small changes in ambient conditions. For the same reason it is difficult to store aqueous Hg samples.

Mercury usually leaves ore deposits as "carbonate or sulphide-anionic complexes" and perhaps as sulphate complexes. It has been demonstrated that high concentrations of bicarbonate ion promote the dissolution and mobilization of sulphidic Hg. Traces of halides also increase mobility. All of these phenomena are most active at pH values below 4 and tend to be reversed at pH 4 to 7. For this reason one generally finds that groundwater aureoles of mercury "do not persist at great distance from the source" of the metal.

4.527 The common base metals. Sulphide bodies, polymetallic ores, copper-molybdenum porphyry bodies.

In the Soviet Union extensive use is made of groundwater analysis in the search for what are generally termed polymetallic ores. This terms embraces a wide variety of complex mineral assemblages, which usually contain some sulphides and/or base metals.

Alekhin and Zhdanov (1968/1030/) described a spring sampling survey for polymetallic deposits of brucite, smithsonite and barite. The measurement of pH, SO_4 and SO_4/Cl was not found to be helpful, but the direct determination of heavy metals was a good guide. The empirical index (Fe + Pb + Ag + Zn + Mn + Ba)/TDS was used to define anomalies.

Udodov *et al.* (1968/1031/) described hydrogeochemical methods based on the coprecipitation of metals in groundwaters with Na_2S and $Al(OH)_3$. They mention successful uses of groundwater hydrogeochemical methods in the U.S.S.R., leading to the discovery of polymetallic, Cu, U and "other" deposits.

Popova (1965/1094/) recorded the use of groundwater hydrogeochemistry in prospecting for polymetallic ores in carbonate rocks. Carbonates are regarded by prospectors as unfavourable host rocks since they readily neutralize reactive compounds released by the ore. Nevertheless the method worked quite well. The waters studied were of the Ca-Na-bicarbonate type, were well aerated, freely circulating, and had a low TDS content. The pH was uniform and somewhat alkaline and of no significance in exploration. The main pathfinder was copper and the "exploration index" was a 5-10 fold increase in [Cu] in subsurface waters. Sporadic Pb and As anomalies were also noted and an increase in the concentration of sulphate was a favourable sign. The authors felt that Cr, Ni, V, Be and Zr might serve as additional pathfinders.

Skryabin and An (1969/1035/) recorded additional results of groundwater surveys for polymetallic ores in the Lashkerek region. The waters here are of the dilute bicarbonate type. Sulphate ion produced narrow anomalies - seldom more than dozens of metres in width - but sulphate/chloride anomalies were much broader. The ratio sometimes rose to between 4 and 8 in the ore zone but was normally only between 2 and 2,5. Elevated concentrations of Zn, Pb, Cu, As, Ag, Mo, Cd and Ni also occurred here, but these metallic anomalies were apt to decay rapidly. Zinc and molybdenum were more mobile than the other listed elements. Zul'fugarly *et al.* (1970/1036/), on the other hand, had no success at all with metallic pathfinders. They found that there was no regular relationship between the contents of Mn, Fe and Cu and either stratigraphic depth or degree of mineralization in certain Soviet ore zones. Nickel, copper and vanadium could not be detected in these waters.

Kovalev (1964/1096/) showed that sulphide deposits in the Turgai syncline had fairly typical secondary aqueous dispersion haloes. The pH was lowered by the oxidation of sulphides of Fe, Cu and Zn and the waters were thereby enriched in calcium, bicarbonate and sulphate after reacting with the country rock. Beyond the dispersion halo the anomaly is erased by dilution or because the pathfinder elements are removed by additional chemical reactions. The dispersion halo is characterized by particular concentrations of major components as well as by some of the species Fe^{3+}, Fe^{2+}, Al, Ti, Mn, Si, Sr, As, Cu, Zn, Mo, Pb, Sn, Ag, In, Te, Ge, Bi, Ni, Be and others. In general [Cu] and [Zn] and "higher than background amounts of spectrally determinable elements (satellites)" are used as the critical hydrogeochemical prospecting criteria.

Kraynov *et al.* (1966/1022/) sampled springs in the vicinity of sulphide orebodies near the Elbrus volcanic centre, with a view to establishing which elements in these waters were related to the ore and which were derived from the country rocks. It was found that Zn, Cu and Hg were supplied by the leaching of the ore, whereas the concentrations of Li, Rb and Cs were controlled only by "conduit lithology". The abundances of As, Ge and TDS could be related to both the occurrence of ore and "propinquity to the Elbrus volcanic centre". The concentration of zinc was inversely proportional to $[HCO_3^-]$.

Blokhin (1969/1032/) prospected for sulphidic ores by determining sulphate ion and heavy metals in samples from wells and springs in western Baikal.

Sulphate concentrations of up to 140 mg/l were noted in well waters but in spring waters this value never exceeded 70 mg/l. Threshold values for Pb, Sn, Mo and V were of the order of 5 μg/l or more; for Co the value was 0,5 μg/l and for Zn 100 μg/l.

Levashov (1969/1033/) described a very strong anomaly occurring in a highly oxidized sulphide ore zone within which samples had been collected at depths of 15 to 28 m. The following mean values were observed (μg/l): Pb 800, Cu 8, Zn 55, Ba 150. Each of the elements As, Sb, Ag, Ni, Co and Cd were also present in above-average amounts. In some samples spectacular concentrations of Pb, Zn and As were encountered. The author records that high $[SO_4]$, [Cl] and $[HCO_3]$ were also indicative of sulphide mineralization.

Chernyaev *et al.* (1969/1034/) analyzed groundwaters that were collected near sulphide deposits in the Urals and elsewhere. High concentrations of Fe, Cu, Zn, Mo and As were found but Pb, Ag and Hg were absent. The best pathfinders were Zn, Cu and sulphate. Low pH and low $[HCO_3]$ were "secondary indicators".

Groundwater hydrochemistry has been used to locate several types of deposits in the Ukraine (Gavrilenko and Shtogrin, 1968/1039/). These include petroleum, salt deposits, iron ores and base metal bodies. The last of these is generally associated with $Na\text{-}HCO_3\text{-}Cl$ water, within which elevated concentrations of Pb, Zn, Hg or As are regarded as favourable signs.

In the West, during the past fifteen years, hydrogeochemical exploration for base metals appears to have taken second place to the search for uranium. Boyle (1963/822/) stated that the analysis of spring waters from a barite quarry in Nova Scotia revealed Zn-Cu anomalies, and that subsequent drilling intersected a sulphide body beneath the barite. He stressed the necessity of obtaining as such sample coverage as possible in surveys of this type and emphasized the importance of examining the precipitates deposited by springs. Elsewhere /823/ Boyle supplemented his earlier observations about base metal hydrogeochemical anomalies in the Keno Hill-Galena Hill area. Lodes sealed by permafrost do not contribute metals to springwaters, but often these waters will pass through float associated with the deposit and will thus provide a clue to the presence of the orebody. All recent findings had confirmed that zinc was the best indicator element in this area.

Between 1967 and 1969 DeGeoffroy and co-workers produced an important series of publications /45, 48, 970/ documenting a regional reconnaissance springwater survey in Wisconsin. The aim was to locate zinc mineralization in carbonate rocks in the Upper Mississippi Valley zinc-lead district and was an extension of earlier work carried out here between 1945 and the early 1950's /520/. This previous work had been very successful and had led to the discovery of a rich orebody in 1955. However, the method had not been followed up immediately thereafter because of the depression of the price of zinc during the following decade.

DeGeoffroy's survey covered some 400 square miles and involved 3766 springs. In the study area the amount of groundwater recharge amounts to about twenty percent of the annual precipitation. Many shale beds are present within the carbonate and these give rise to a great number of springs because they force the groundwater to move laterally within the carbonate horizons. The primary lithological dispersion haloes of the zinc bodies are very well developed, and groundwater anomalies can be generated either by the orebody *per se* or by the percolation of water through the primary aureole. The metals that are potentially available for dispersion are Cu, Pb, Zn and Ni. Because the groundwaters of the area are rich in bicarbonate ion and are well-buffered, the pH range is generally restricted to between

6,6 and 8,1. The carbonate forms of cations such as Fe, Cu, Zn and Pb are favoured at these pH values. It is to be noted that zinc carbonate is moderately soluble but that lead carbonate is rather insoluble by comparison. Any factor, such as loss of CO_2, that shifts the pH equilibrium towards the alkaline side will cause precipitation of a portion of most of the metals present as the carbonate. It is for this reason that surface water hydrogeochemical prospecting has not been very successful in this area.

Springs were located by applying geological principles, by examination of maps and by following shale bands known for their springs and seeps. Ephemeral springs were avoided. Samples were collected in polyethylene bottles, which were filled to overflowing, and were analysed within a few hours. The pH was measured and [Fe] and [Zn] were determined. The concentration of zinc ranged from 0,05 to 2,5 mg/l and did not vary greatly with the changing seasons. The 75 percentile was 0,27 mg/l and anything above this value was considered to be highly anomalous. The data were interpreted by calculating trend surfaces and residuals. It was found that a value of 0,30 mg/l encompassed all known zinc mining areas and that "ridges" of other high [Zn] values appeared. These seemed to run parallel to the regional fold axes. Contouring of the residuals produced 56 anomalous areas of which 26 were related to known deposits.

By correlating the zinc values with various lithological units in different subareas it could be shown that certain formations and districts had far more potential than others. However, in selecting these favourable zones one had to be very careful of biases caused by the clustering of sample points. Several of the anomalous zones were drilled and ore-grade mineralization was intersected in a few places. It was stressed that very deep, unoxidized lodes were likely to be missed by such a method of exploration. For example, spring sampling reconnaissance would probably not work well in some nearby areas where the potentially mineralized stratigraphic formations are known to be well below the water table. Furthermore, this type of reconnaissance is unsuitable in carbonate terrains within which karstitic conditions are widely developed.

In Section 4.523 mention was made of the work of Hoag and Webber /1101/, who tried to pinpoint the source of anomalous waters by calculating the rate at which sodium was added to these waters during migration. They applied their theories in an attempt to trace the origin of zinc and copper in groundwaters near the Eustis Mine. The background value for each of these metals was about 8 to 10 μg/l. Great care had to be taken in assessing the origin of the sodium present in the water, since the entire process of calculation is critically dependent upon this parameter. It could not be assumed, for example, that Na^+ from albite is balanced by Cl^-, because additional chloride can be derived from plants deficient in sodium. Some additional sodium is contributed by precipitation. The estimated positions of recharge areas, as calculated from data collected at various sampling points, are shown in Figures 1 and 2. In these diagrams the term "Surface waters" refers to seeps, springs and other waters. Anomaly A (zinc) was attributed to contamination from mine shafts in the recharge area. Anomaly B (Zn and Cu) was apparently related to the leaching of contaminated soils. Anomaly C was related to disseminated mineralization.

Some drill hole waters were contaminated with zinc and/or copper but others appeared to indicate additional anomalies. In these cases the sulphate content was also anomalous. The authors estimated that some 50% of the drill hole waters anomalous in copper or zinc were contaminated, 40% were related to known mineralization and 10% were related to unknown causes. The corresponding values for spring waters were 30%, 20%, and 50% respectively.

Mehrtens *et al.* (1973/865/) provided an interesting examination of a displaced groundwater anomaly in Wales, where a low grade copper deposit had been discovered in igneous rocks. The groundwater and bog anomalies associated with this mineralization were situated on both sides of the surface watershed and were strongly influenced by a large, concealed, gravel-filled stream channel that was not suspected before drilling commenced. The situation that was encountered is shown in Figure A (reviewer's diagram).

The authors noted another example of displacement, in British Columbia. Molybdenum dispersed in groundwaters formed anomalies in stream sediments 800 m from the ore, but no anomaly could be detected in the immediate vicinity of the deposit.

Horsnail and Elliot (1971/855/) reviewed many of the aspects of the secondary migration of Mo and Cu in both wet and relatively dry peaty areas in Canada. Both elements form important organometallic compounds under these conditions. The mobility of these and other elements becomes extremely complex when pH, Eh and the concentration and nature of the organic matter varies. All else being equal, Cu tends to be more mobile than Mo at low pH values, while the reverse is true under alkaline conditions.

Coope (1973/1106/) recently summarized the state of the art of hydrogeochemical exploration for porphyry copper deposits. At pH values below 5,5 copper is "very soluble in natural water", but Mo is soluble over a broader range since it forms alkaline inorganic complexes. Molybdenum anomalies have been noted up to eight miles from the source, whereas the dispersion of Cu is generally more limited.* "The distribution of molybdenum

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* There are some exceptions - for example, in the Boise Basin of Idaho (Rostad, 1967/1107/). Here copper "is more extensively dispersed, probably because of complex fixing of molybdenum by iron and ferrimolybdate". In some cases this enhanced release of Cu may be due to bacterial leaching of the porphyry ores. The relative proportions of dissolved Cu and Mo do not seem to be dependent upon Eh.

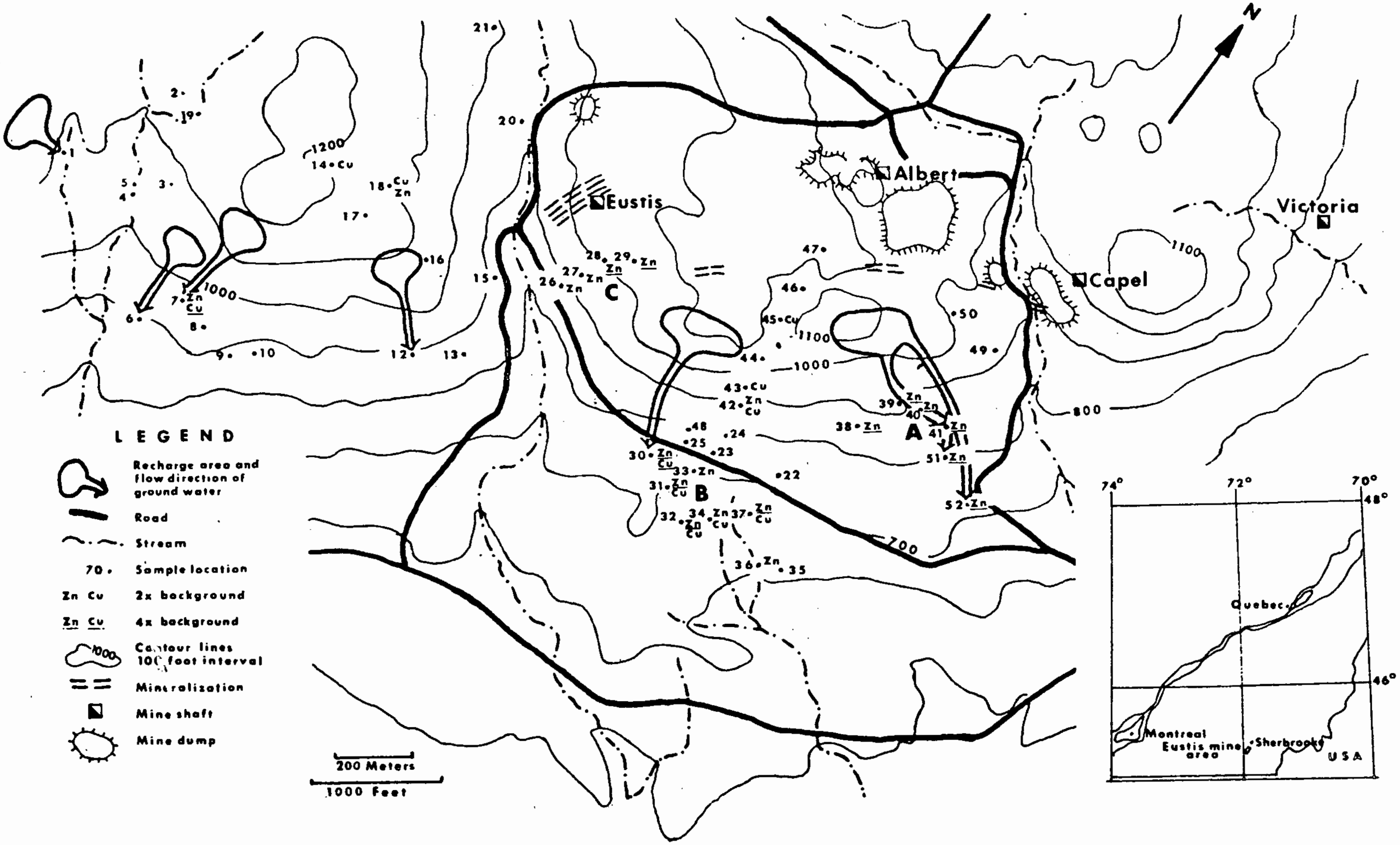


Fig.1. Location of surface waters, recharge areas as determined by the method described in the text, and samples anomalous in Cu and Zn. A, B and C are water anomalies referred to in the text.

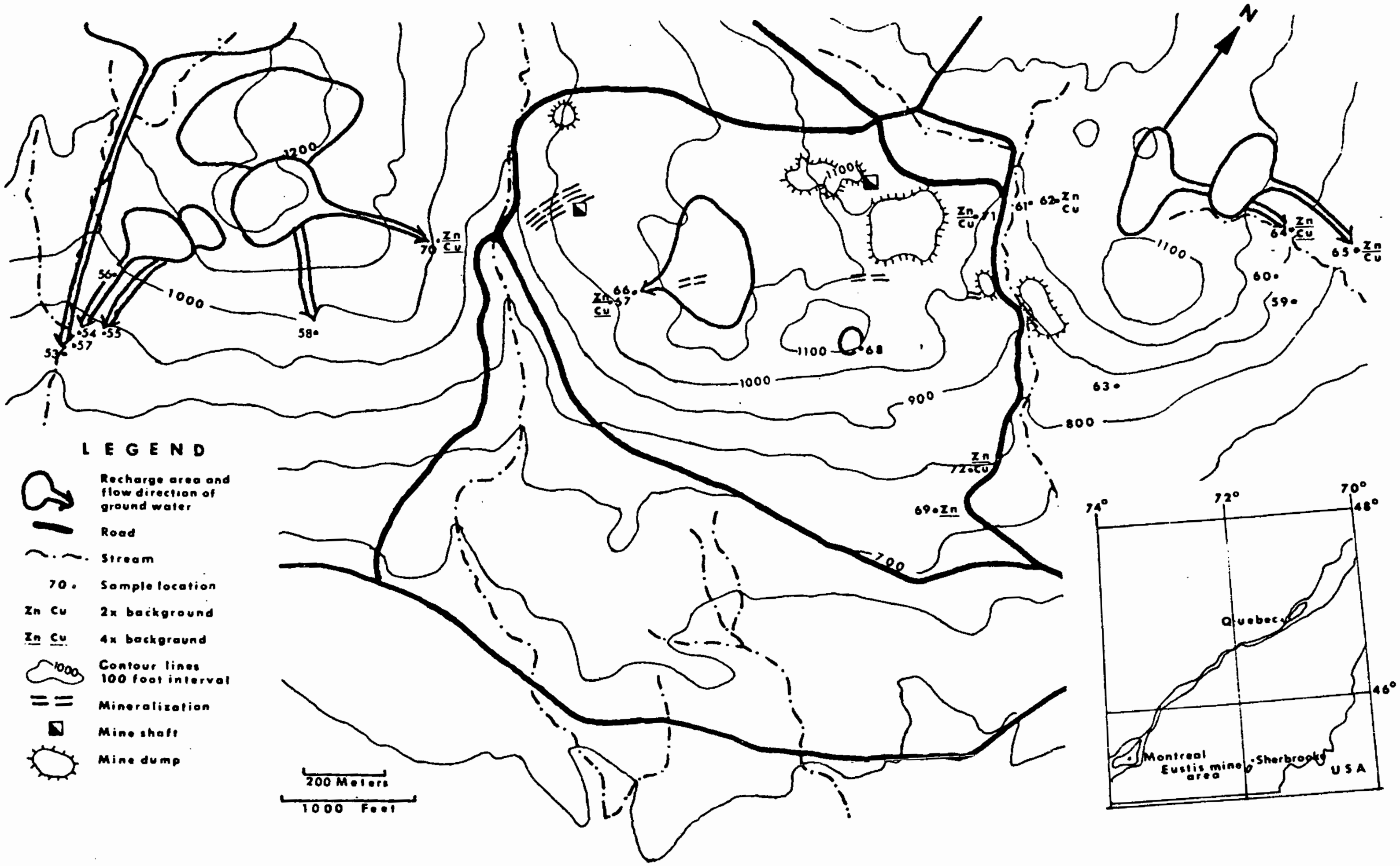
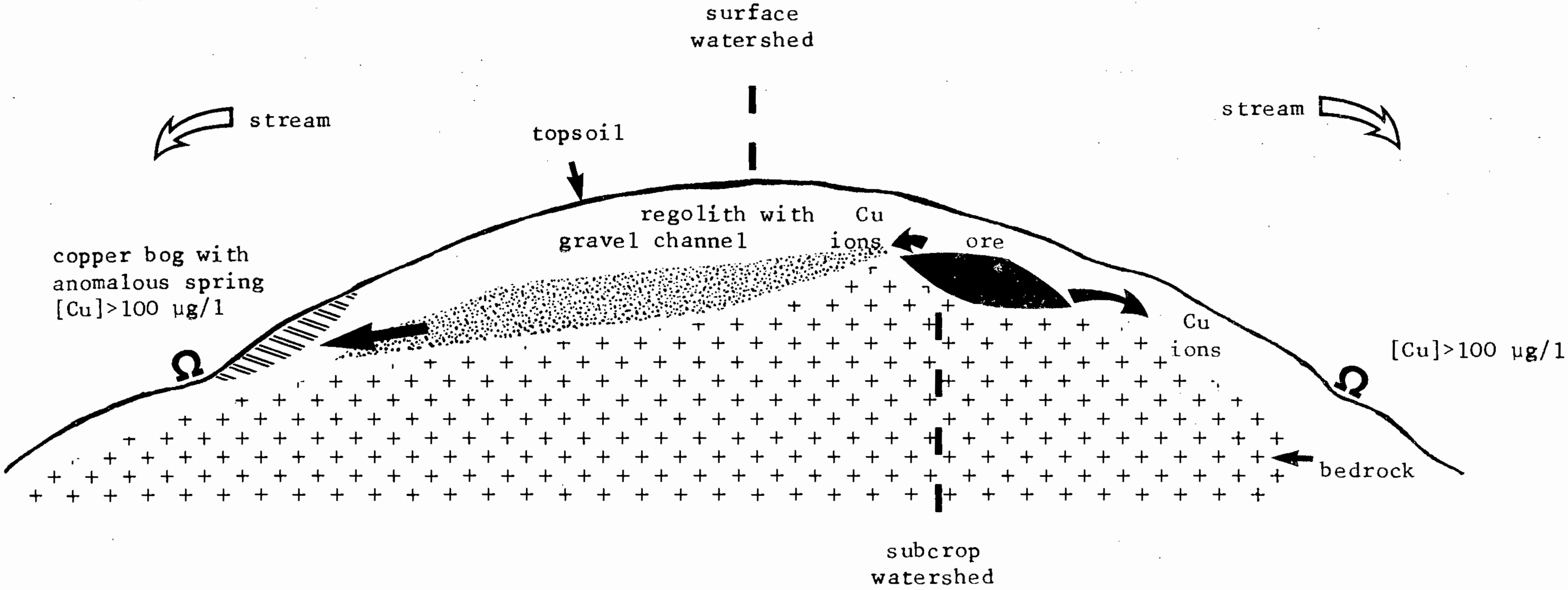


Fig.2. Location of drill-hole waters, recharge areas determined by the method described in the text, and samples anomalous in Cu and Zn.

FIGURE A


surface watershed
stream
stream
topsoil
regolith with gravel channel
Cu ions
ore
copper bog with anomalous spring [Cu]>100 µg/l
Cu ions
[Cu]>100 µg/l
Ω
Ω
bedrock
subcrop watershed

in groundwaters in the Pima District clearly indicates the potential of groundwater sampling as a regional reconnaissance tool for porphyry copper mineralization in areas of transported overburden and poor outcrop. Very little copper is detectable in the groundwaters at San Manuel ..... Measurements elsewhere indicate that in the alkaline environments of the southwestern United States copper occurs in insignificant amounts in alkaline groundwaters in the proximity of mineralization".

Trost and Trautwein (1975/1080/) described the results of groundwater hydrogeochemical exploration for porphyry copper deposits in Arizona. They found that samples could be collected after ten minutes of pumping. Conductivity, Eh, pH, [Mo] and the concentrations of certain other microcomponents were found to be useful parameters. The amount of dissolved molybdenum was not controlled by Eh-pH conditions but was related to TDS. The Eh and pH values did not change near prospecting targets, but [Mo] rose steeply. Thus the Mo/conductivity ratio was a valuable aid to interpretation.

The waters in the study area were neutral to alkaline. As much hydrological data as possible was collected and samples from perched water tables were discarded. Good hydrological control was achieved and groundwater flow rates were found to be from 10 to 40 ft/year. The concentration of molybdenum ranged from less than two to about 50 μg/l.

Huff and Marranzino /1081, 1090/ conducted similar studies around porphyry deposits at Pima Mission and showed that Mo was indeed a much better pathfinder than Cu under most conditions. The anomalies were displaced downslope and covered a broad area.

Nepeina and Sotnikov (1971/1037/) studied the chemistry of groundwaters around six Soviet molybdenum deposits. The waters were characterized by high concentrations of sulphate, calcium and magnesium. The concentration of mercury ranged from 2 to 6 µg/l, but was higher in associated surface waters. The concentrations of Mo and Hg were related. The mercury became immobilized in the presence of H_2S.

4.528 Uranium and associated elements.

Current interest in hydrogeochemical prospecting for uranium remains high. This review will be concerned mainly with developments in the West, but will begin with a brief examination of a few publications by Russian authors.

Lopatkina (1964/806/) summarized most of the basic principles of this technique at the stage of development that had been reached by the early sixties. He noted that it was very difficult to predict the amount of uranium in rocks from the concentration of uranium in associated formational waters because of the critical dependence of aqueous [U] on the mode of occurrence of the metal in minerals, and on climate and regional geochemistry. Lopatkina comments upon certain important changes that occur in the groundwaters of the U.S.S.R. as one traverses north to south from a cold humid to a hot humid terrain. The TDS content rises but remains below 500 mg/l. The concentrations of bicarbonate, chloride, sulphate, calcium and magnesium increase in close proportion to the TDS (Figure 1). Note that bicarbonate remains the principle anion in all waters. This progressive accumulation of salts influences the hydrogeochemistry of uranium. "The content of uranium circulating in igneous rocks in the humid zone is determined, mainly, by two parameters: the concentration of uranium in the rocks and the amount of dissolved rock, which in general is indicated by the total salinity of the waters". Some hydrogeochemists would argue that this view is simplistic but according to Lopatkina's data it is none the less valid. "The effect of each of these factors is clearly observed when one of them remains constant". Figure 2 shows the relationship between $[U]_{rock}$ and $[U]_{water}$ for a group of waters with a TDS content of 50 mg/l. Figures 3 and 4 show the relationships between $[U]_{water}$ and TDS for two groups of rocks with $[U]_{rock}$ equal to 2 μg/g and 6 μg/g respectively. A close correlation is demonstrated in each case. It follows that $[U]_{rock}$ ought to be closely related to $[U]_{water}/\mathrm{TDS}$, i.e. [U] in the "dry residues", and this is in fact found to be the case (Figure 6). Note that the two cruves are not coincident and that the modal value for [U] in the dry residues is about twice that of the concentration of the metal in the associated rocks. This, according to Lopatkina, "is the most probable enrichment factor for uranium in waters circulating through igneous rocks in humid zones". The general equations relating the concentrations of the various abovementioned parameters are

$$[U]_{rock} = 500\ [U]_{water}/[\mathrm{TDS}]$$

$([U]_{rock}$ in μg/g, $[U]_{water}$ in μg/l, [TDS] in mg/l),

or,

$$[U]_{rock} = 357\ [U]_{water}/[\mathrm{HCO}_3^-]$$

if one assumes a constant relationship between [TDS] and the amount of bicarbonate ion.

Several examples are given by the author to show that the wholerock concentration of uranium can be fairly accurately predicted for various bodies of rock* by this method (Figure 7). It is suggested that the abovementioned mathematical relationships could be used to calculate the regional background for a given unit of rock and its associated formational waters if [TDS], $[\mathrm{HCO}_3^-]$ and $[U]_{rock}$ are known. Note that this calculated value

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* These are naturally fairly large bodies. The method will obviously break down if relatively small volumes of rock are considered.

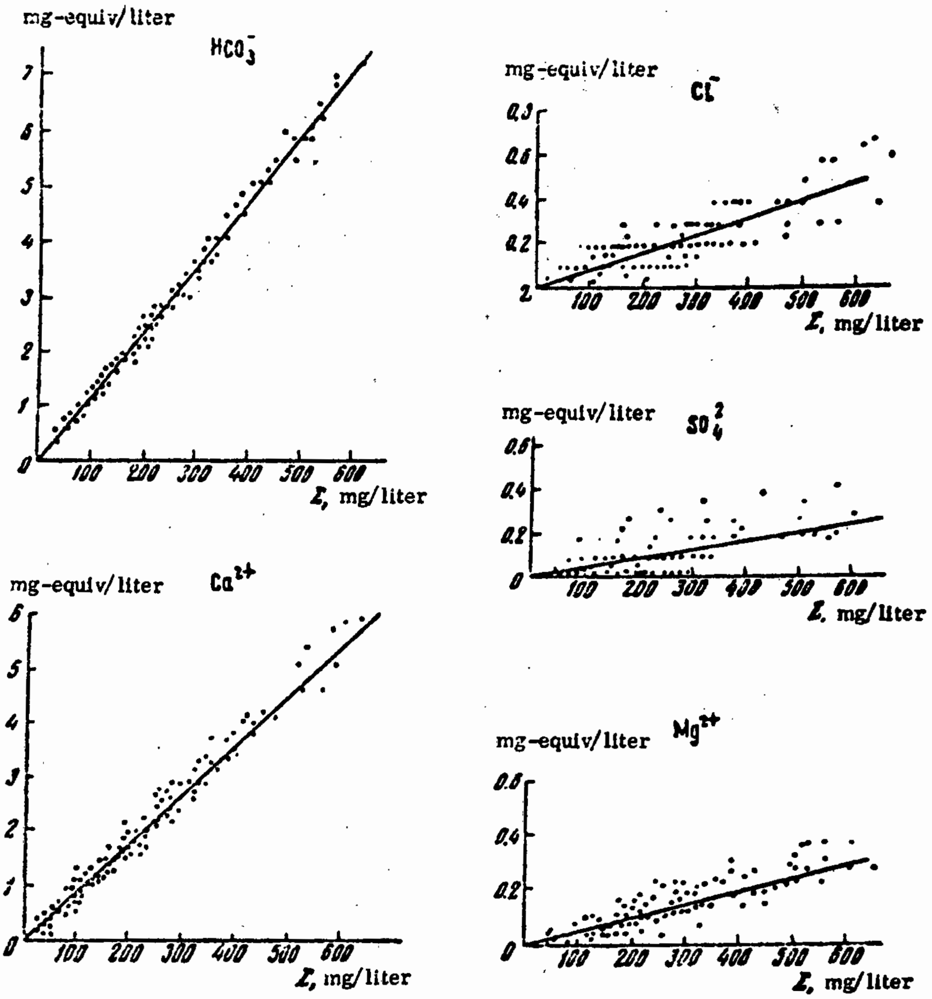

Fig. 1. Relation between the contents of HCO$_3^-$, Ca^{2+}, Cl$^-$, SO$_4^{2-}$ and Mg^{2+} and total salinity.

x—average content of ions for the interval.

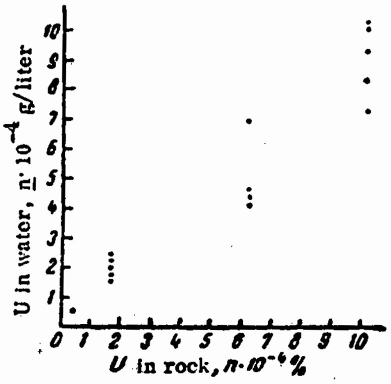

Fig. 2. Relation between uranium content in water with salinity of ~50 mg/liter and its content in rocks.

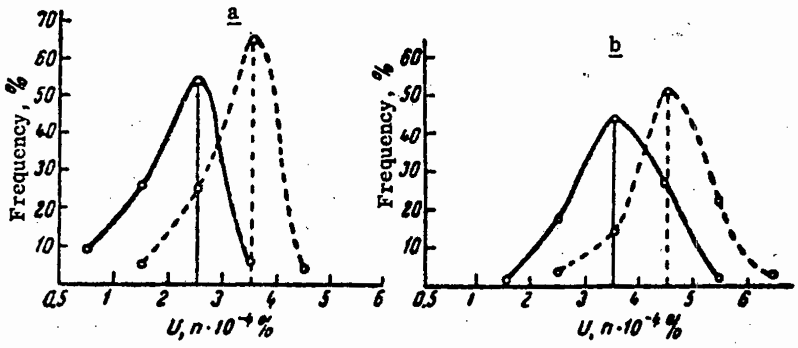

Fig. 6. Concentration of uranium in rocks and in the dry residue of waters circulating through these rocks.

—— uranium in rocks; ----- uranium in dry residue

a—Caucasus (granodiorites): 142 rock samples, 75 water samples;
b—Eastern Siberia (granites): 99 rock samples, 50 water samples

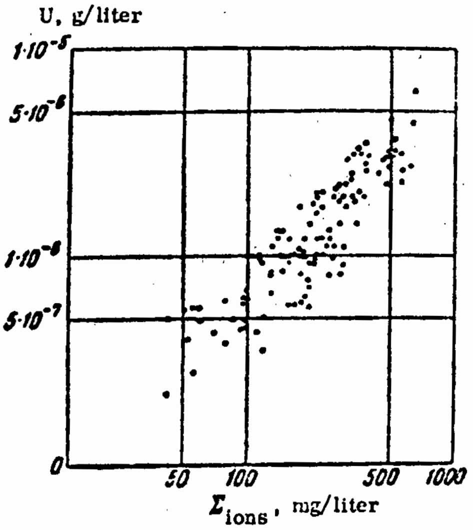

Fig. 3. Correlation between uranium content and salinity in the waters of the Caucasus.

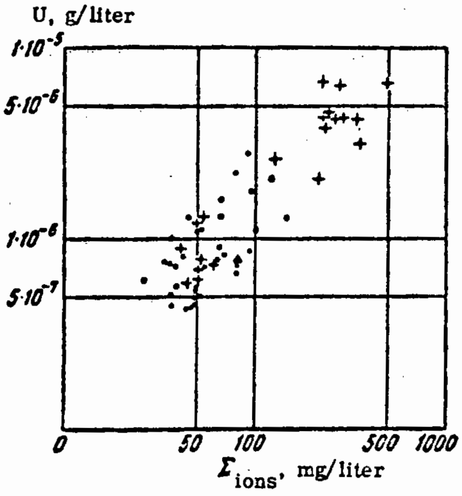

Fig. 4. Correlation between uranium content and salinity in the waters of the Caucasus and Eastern Siberia.

\+ Caucasus · Eastern Siberia

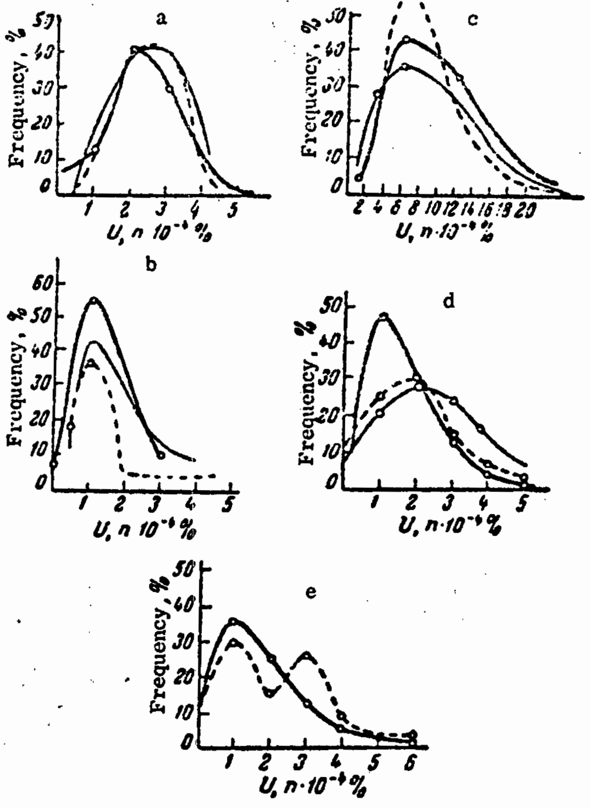

Fig. 7. Actual and calculated uranium contents in different rocks.

—— actual U content; ----- U content calculated by formula $n_U = \frac{m_U \cdot 100}{2a}$; ——— U content calculated by formula $n_U = \frac{m_U \cdot 100}{2.8\,HCO_3^-}$

a—Caucasus (granodiorites); 142 rock samples, 75 water samples; b—Caucasus (pyroclastic-sedimentary beds): 54 rock samples, 22 water samples; c—Eastern Siberia (granites): 99 rock samples, 50 water samples; d—Eastern Siberia (granites): 53 rock samples, 138 water samples; e—Eastern Siberia (sedimentary rocks): 389 rock samples, 25 water samples would be independent of any empirical knowledge of the abundance and distribution of uranium in the waters. Alternatively $[U]_{water}$ and [TDS] can be used to calculate $[U]_{rock}$, which value can then be compared to the known or postulated abundance of the metal in the rocks of the area of interest. In this manner the vagarious effects of climate and changing lithology are minimized.

In the examples given the author has used data from igneous rocks only but he stresses that these concepts "can be extended to the waters in continental sedimentary rocks formed as a result of the mechanical disintegration of igneous rocks". However, the patterns described above are disturbed in the vicinity of "salt beds". If the concentrations of sulphate and chloride are raised by briny materials then the $[U]_{water}/[TDS]$ ratio fails as a reliable index - "it is practically impossible to establish a relationship between it and the uranium content of the rocks .....". The same effect is observed in arid areas where evaporation increases "the absolute and relative concentrations of the stable ions, Na^+, Cl^-, $SO_4^=$, Mg^{2+} and uranium (and decreases) the concentrations of the unstable ions, Ca^{2+} and CO_3^- (*sic*, $CO_3^=$ or HCO_3^-)". Lopatkina adds rather pessimistically that "it is practically impossible to establish a quantitative relation between the uranium content in the waters and in rocks in the regions with arid climate".

A rather brief but interesting observation on the mobility of uranium in groundwaters was made by Serikov (1964/813/). He noted that the release of the metal into groundwaters in shales was very dependent on the grain size of the shale material and that "a slight increase in the content of sand "led to an increased rate of leaching of uranium. It follows that false uranium anomalies may be generated purely by subtle changes in the clastic composition of the host rocks.

Vovk (1975/1088/) put forward the radical proposal that the "intensity of geochemical migration" in groundwaters "depends on radioactivity of rocks and waters", i.e. is promoted by radiolysis in these waters. The principle mechanism of mobilization, he claims, is redox reaction between the products of water radiolysis and rocks. This effect can apparently be substantiated by regional field studies and laboratory experiments. Vovk concedes the importance of better known controlling effect such as time, "constants of reaction" and concentration of elements in the host rocks contacting the water.

Dankovtsev (1965/832/) summarized the program for groundwater hydrogeochemical prospecting for uranium in Siberia, where this technique is the most important form of geochemical surveying. The area is humid and, depending on pH and pCO_2, uranium may be mobilized as either the carbonate ion or as uranyl hydroxide. The latter complex is more readily precipitated from natural waters. Sulphate is not an important mobilizing ion in this region.

Kapkov (1969/833/) demonstrated the danger of assuming that the mobility of uranium will necessarily be the same in both the groundwater and surface water of a given district. Anomalous spring waters emerging from granites in mountain-taiga areas and containing 3 to 5 μg U/l rapidly lost a substantial proportion of the metal, which became incorporated into stream sediments.

Novikov and Kapkov (1965/882/) showed that the dispersion of uranium in groundwater was dependent on topographic slope. In mountainous terrain anomalies could be detected up to 400 m from the orebodies but in the "foot hills regions" this distance shrank to between 50 and 70 metres. Germanov (1963/886/) presented a short but excellent review of other Russian studies of the geochemistry of uranium in natural waters.

Szalay and Samsoni (1969/921-2/) described a large-scale investigation, involving 4000 samples, of the causes of variations in the background concentration of uranium in the groundwaters of northeastern Hungary. No uranium deposits are known here, yet [U] varied from 0,01 to 100 μg/1. The average uranium content "of large surface areas" here is only 4-5 μg/g. The authors found that [U] tended to be statistically higher in waters associated with rocks of finer grain size but that this factor alone could not account for the irregular distribution of the metal. They considered that these irregularities must be due to "processes which control the leaching of uranium from rocks" and set up laboratory experiments to investigate these processes. Samples of gabbros, granites, andesites and rhyolites were exposed to simulated natural waters. As expected, they found that U^{6+} is mobile whereas U^{4+} is not and that bicarbonate ion is a very effective complexing agent for uranium. The leaching of the metal was initially rapid but equilibrium was soon established between the water and the crushed rock samples. The equilibrium concentration in the water depended on rock type and was highest for granite. However, this value seldom exceeded one percent of the total mass of uranium in the rock. Crushing the samples to smaller fragments increased the equilibrium value of [U] only slightly. When the equilibrated water was replaced by uranium-free water, more of the metal dissolved and a new equilibrium became established after a few hours. When an excess of dissolved uranium was added to the system it was rapidly absorbed onto the rock powder. These phenomena are best explained by postulating a dynamic equilibrium:

$$\text{rock} \underset{\text{adsorption}}{\overset{\text{dissolution}}{\longleftrightarrow}} \text{water}$$

The results may explain why the concentration of uranium in groundwaters varies from one lithological unit to another. The absolute mass of uranium in the rock may not be as important as the equilibrium constant

$$K = [U]_{rock}/[U]_{water}$$

for each rock type. The results may also explain why a decrease in grain size does not always result in an increase in $[U]_{water}$, since the greater surface area may also result in greatly increased absorption of the uranyl ion. An interesting corollary of these observations is that uranium anomalies in groundwater passing through a uraniferous ore ought, as a result of adsorption, to "decay" to background values within a few hours once the water is clear of the source of the metal. It would follow that true uranium anomalies may be very localized features - a proposition certain to raise the eyebrow of many an exploration man.

The authors suggest that the principles of water/rock equilibrium that they described for uranium may apply equally well to other metals also.

The recent Western literature of groundwater hydrogeochemical exploration for uranium includes a number of excellent general summaries as well as case histories from various parts of the world.

Wenrich-Verbeek *et al.* (1976/1079/) discussed some recent developments in hydrogeochemical techniques for uranium exploration. It has been noted that some springs yield mixtures of young and old waters and that this can complicate interpretation. Groundwater appears to be a better "geochemical evaluator of subsurface terrane" than surface water is, but is more costly to sample. There is still a very strong interest in the use of radioactivity in spring waters as a guide to ore. The authors give examples of the use of factor analysis in U-Ra prospecting by means of spring sampling. In one study the "unmeasured geochemical variables" such as "the degree of uranium mineralization" controlled "almost half the variance of occurrence of uranium and radium" in the spring water samples. A second study of spring and well waters showed that the concentrations of HCO_3^-, Ra, U and TDS tended to increase sympathetically. The authors noted that some association exists between [U] and [NO_3^-] in certain areas and suggest that this may be due to the presence of uranium in fertilizers. They note the increasing use of the fission track method for uranium analysis.

Gabelman (1970/1052/) stated that uranium was leached predominantly from those orebodies undergoing oxidative destruction. Deep-seated, reduced rocks do not yield appreciable concentrations of uranium to groundwaters. He points out that false anomalies and suppressed anomalies are rather commonly encountered in groundwater hydrogeochemical exploration for uranium. Several waters in uraniferous sandstones from different parts of the U.S.A. do not have better-than-average uranium contents, while in many areas high concentrations of the metal occur in lithological units that are not known to carry uranium ore.

Saum and Link (1969/932/) provided a useful review of aspects of the geochemistry of uranium. This publication is notable in that it contains some claims that would probably be disputed by many hydrogeochemists. For example, the authors state that uranium may form soluble complexes with "hydroxides and sulphides". Their claim that urananite will only precipitate when [U] reaches 500 µg/1 might also be challenged (cf. the work of Dall'Aglio *et al.* /904/). However, these are minor shortcomings in an otherwise meritorious review.

Figure 5 is a good idealized portrayal of the relationships that exist between source areas, uranium deposits and the gravitational movement of groundwater. Note that in this model all the uranium may be retained at the roll front, so that waters sampled down-dip could be very poor in uranium. The authors stress that orebodies of this sort can be severely leached of radioactive species and will therefore be detectable only by geochemical

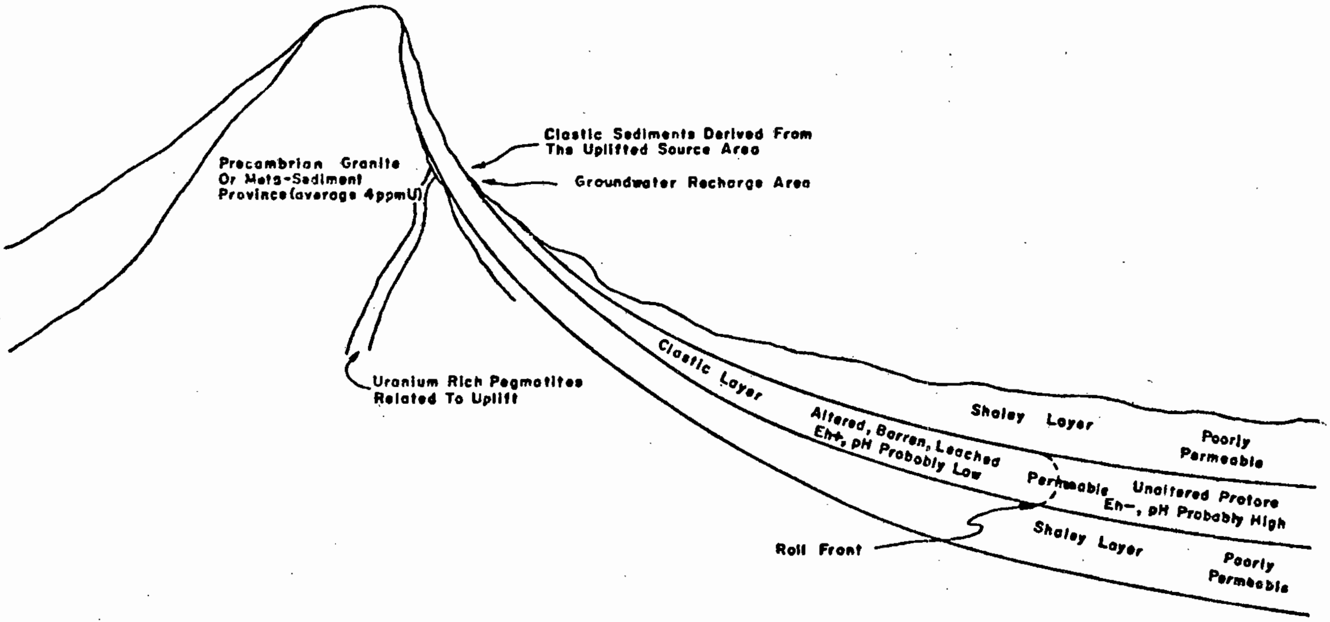

FIGURE 5—Idealized cross section of solution front deposit.

means and not by radiometric methods. Conversely, the "hot" daughter elements may accumulate elsewhere to give a displaced radioactive anomaly. Saum and Link also make the important point that, in the U.S.A. at least, different types of uranium orebodies are characterized by different element assemblages and that these various assemblages may be reflected in the composition of associated groundwaters. For example, the Palaeocene/Late Cretaceous lignitic ores of Montana and the Dakotas are accompanied by subsurface waters rich in U, As, Cu, P, V and Mo.

Some of the concepts stated by Saum and Link were recast by Bowie *et al.* (1971/851/) in a slightly different form. They emphasized an additional point that may perhaps be overlooked by uranium prospectors, viz, that the uranium in *transit* in subsurface waters can be detected only by chemical and not by radiometric methods, since the latter technique is dependent upon the activity of associated decay products rather than upon uranium itself. The authors showed that in moorland areas (Dartmoor) hydrogeochemical methods (springs) were superior* to stream sediment methods of prospecting for uranium. They remained enthusiastic about the potential for using additional indicator elements for uranium in certain cases. Zinc and Mo "have been shown to be particularly successful .....".

It is clear from the foregoing, and from other examples in the literature that the question of the use of additional pathfinders for uranium remains an inviting possibility. The topic has been relatively little studied, even in recent years, simply because the determination of uranium itself usually works so well. It is certainly true that few present-day hydrogeochemists

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* Note the effect of regional influences. By way of contrast, Morse (1971/858/) concluded that "as a geochemical prospecting medium (for uranium), sediments are clearly more useful than water" in his study area in Ontario.

are prepared to make dogmatic pronouncements one way or the other about the latent possibilities of other indicator elements. The list of candidates is certainly quite long; Michelson (1966/820/) gives the following list of elements "that are geochemically associated with uranium": Ca, Fe^{++}, Zn, Y, Pb, Sb, Si, V, Co, As, Zr, Ba, P, Cr, Ni, Se, Mo, Re, Yb, S, Mn, Cu, Sr, Ag, Th and Bi.

According to Wyman (1970/788/) uranium in the Colorado Plateau ores is accompanied principally by V, Cu, Fe, Mo, Se, Ag and Re and the movement of these elements is linked to the mobilization of uranium. Vanadium, selenium and uranium tend to be precipitated under the same reducing conditions, whereas the iron present remains dissolved and is removed. The mobilities of the other elements listed are either poorly known or rather complicated.

Davy (1974/942/) gave one of the best recent summaries of the geochemical relationships between uranium and the other radio-elements such as Th, Ra and Rn. He shows why even the geophysicist cannot afford to entirely ignore the hydrogeochemistry of these elements. Solution chemistry will determine the nature of the elemental assemblage of redeposited radioactive minerals and will thus have a profound effect on the character of the ray spectrum of the deposit. Uranium is very mobile in solution but can be immobilized by evaporation, reduction, adsorption, co-precipitation, heating or loss of carbon dioxide. Thorium is very immobile and "is left behind when uranium is removed by leaching". Radium is immobile under most conditions and "can only travel freely in sulphate-free, neutral or acid solutions". Radon is mobile and can be detected in groundwaters near uranium deposits. Davy emphasizes the importance of the water table in geochemical prospecting for uranium. Almost everywhere it is noted that deep, oxygen-starved uranium orebodies have weak aqueous haloes.

It is observed that in both the U.S.A. and Australia 5 µg/l is a typical background concentration for uranium in groundwaters. Background values of 30 to 100 µg/l are known but are rare. In these and other situations it is possible that Rn/U ratios in water may help to distinguish between truly anomalous and non-significant uranium occurrences.

During the 1960's it became generally apparent that the mobility and hydrogeochemistry of uranium was in some instances affected by biological processes. Several writers have shown that the concentration of uranium in natural waters is strongly influenced by the activities of the sulphide and iron oxidizing bacterium *Thiobacillus ferrioxidans* /38, 947-52/. The effect is indirect in that the bacteria continuously produce Fe^{3+}, which oxidizes U^{4+} to U^{6+}:

$$UO_2 + Fe_2(SO_4)_3 + 2H_2SO_4 \rightarrow UO_2(SO_4)_3^{4-} + 2FeSO_4^- + 4H^+ .$$

The bacteria also regenerate the sulphuric acid. They operate in the pH range 1,3 to 3,5. Oxygen, CO_2, NH_4^+ and PO_4^{3-} must be present. Some strains of the organism are inhibited by the presence of anionic uranium compounds and by anions other than sulphate, but none of them appear to be influenced by most cations. Little is known of the importance of these phenomena outside of uranium mines.

Dall'Aglio, who has been one of the most prolific authors within the discipline of hydrogeochemistry, has given us some comprehensive overviews of many of the principles and practices of groundwater sampling in uranium exploration. A good example is his discussion of the fundamentals of planning and interpretation in hydrogeochemical prospecting (1972-1973/1078, 1084/) in which he has turned a philosophical eye on some tenets that are taken for granted but are seldom so clearly stated.

Dall'Aglio offered two interesting diagrammatic representations of the relative mobilities of the elements. Figure 1 shows that the mobility of uranium is intermediate between the extremes represented by, for example, Cl and Ti. Figure 2 shows that the mobility of uranium is anomalous for an element with an intermediate ionic potential. This is due to the stability of the uranyl ion, as opposed to the simple U^{6+} ion. Uranium is much more mobile than either copper or zinc, and in anomalous waters it shows peak-to--background ratios that are far higher than those exhibited by any other element. Dall'Aglio indicates that hydrogeochemical prospecting for uranium is most likely to be useful in the temperate zones and that the method suffers from certain intrinsic disadvantages at very low or high latitudes and in very arid regions. The method ought to be used for reconnaissance surveys rather than for detailed work since water is too mobile for the latter purpose. It is suggested that careful multiple regression "between the concentration of uranium and that of other constituents dissolved in water" will eliminate "most of the (background) variability attributable to environmental factors". Generally speaking the frequency distribution is not a good method of detecting hydrogeochemical anomalies, because water samples consist of "mixtures" that have undergone natural processes of different kinds and intensities, depending on the local environment from which they were collected. These processes can change the concentration of the element being sought even in the absence of a mineral deposit containing it. Thus, for example, "the content of the mobile trace elements increases as the water proceeds along the hydrological cycle". Because of the stability and mobility of uranium in water the raw [U] data can readily be adjusted for this increase through the use of U/TDS ratios. Changes in the slope of the cumulative plot of U/TDS for many samples may reflect changes in the lithology of the bedrock. For instance, TDS may

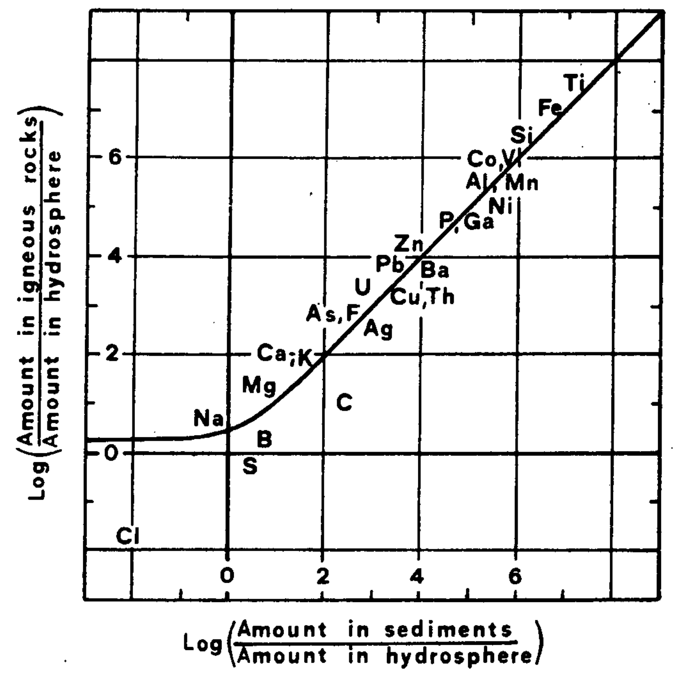

Fig. 1 Geochemical mobility of the elements. Position of an element on the graph gives a measure of its geochemical mobility in the supergene environment. From Dall'Aglio and Tonani[5]

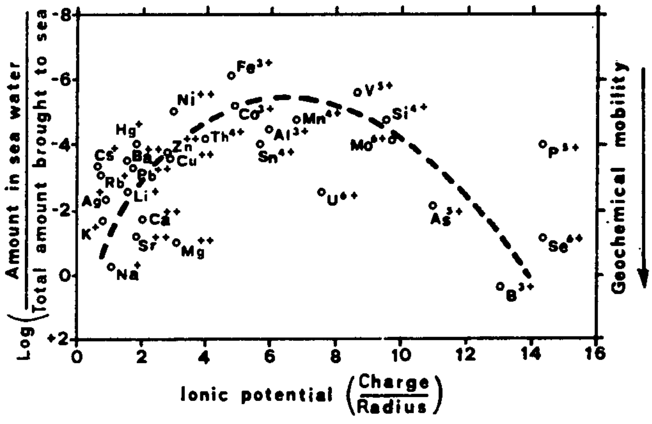

Fig. 2 Geochemical mobility: uranium is in an anomalous position because it shows a higher value of geochemical mobility than that corresponding to its ionic potential. From Garrels and Christ[6]

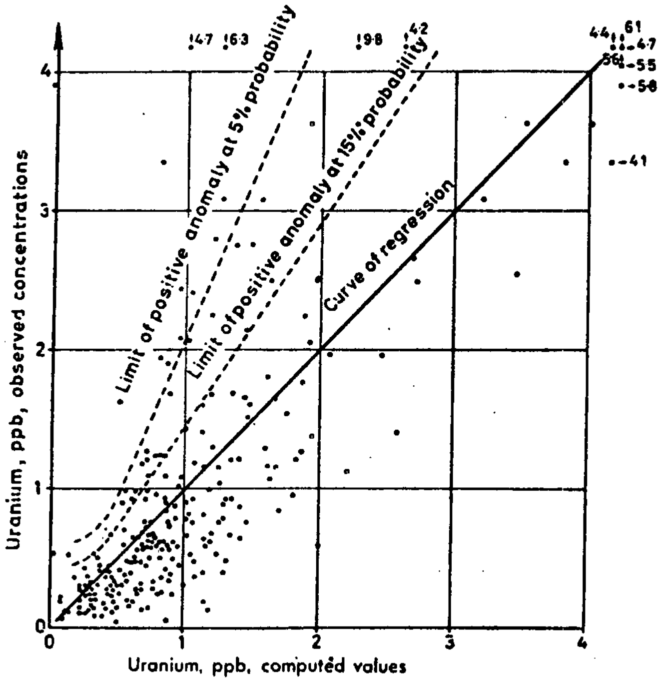

Fig. 5 Multiple-regression analysis (see Table 3) of uranium on the major constituents. Stream-water samples from Sicily

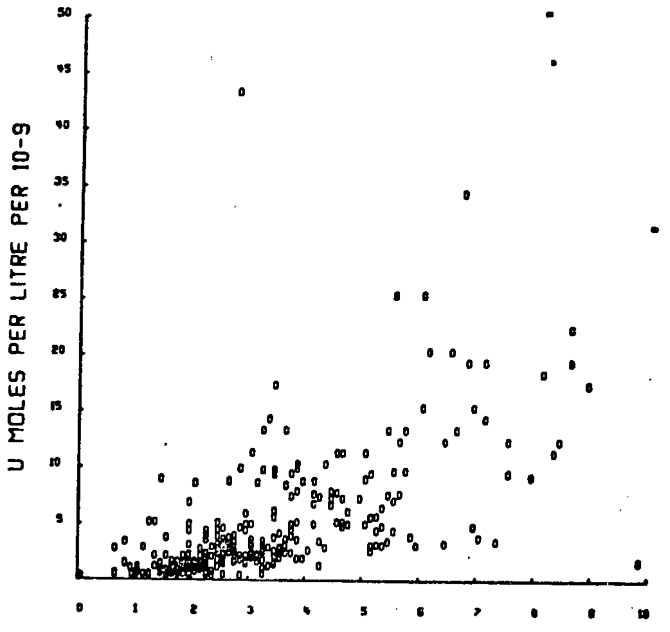

Fig. 6 Uranium–fluorine diagram plotted directly by computer. Stream-water samples from eastern Sicily

increase somewhat faster than [U] in water traversing saline marine beds while the opposite might be true for waters passing through some rhyolites. "All the processes to which a water sample has been subjected, including those due to the various lithological, environmental and hydrodynamic conditions, leave an indication of their effect on the concentration that each elements achieves. Considering the major constituents as independent parameters and the uranium as the dependent parameter, random variations in the latter can be considerably reduced". Dall'Aglio gives examples of computerized statistical data handling procedures (regression analysis) that greatly speed and clarify the interpretation of the raw uranium data. Anomalies can by this means be defined relative to the curve of regression, which is the locus of the calculated [U] "predicted" by the concentrations of the major components (Figure 5, page 200). The regression need not necessarily be made between [U] and the concentration of the major components only. In some areas a correlation between [U] and the contents of certain trace elements may reveal systematic, non-significant increases in the abundance of uranium that are related to lithological factors (Figure 6).

Dall'Aglio draws attention to the significance of negative uranium hydrogeochemical anomalies, which may indicate areas in which orebodies are presently forming. He claims that "the application of this concept has led to the discovery of new uranium orebodies" in the Latium region of Italy.

In a separate publication (1974/904/) Dall'Aglio *et al.* examined the conditions under which uranium may be removed from groundwater to form secondary uranium minerals such as autunite and carnotite. It is observed that waters of varying TDS can all lose uranium by precipitation, fresh waters in wet, mountainous areas not excepted. In arid areas evaporation is the principle mechanism causing precipitation of carnotite. Some of the problems that have been encountered in attempts to calculate equilibrium solubility data for uranium have stemmed from a disregard for the importance of activity coefficients. Dall'Aglio takes care to distinguish clearly between the activity of uranium in solution and its concentration. These may differ by orders of magnitude in the presence of the carbonate-uranyl complex, for which the equilibrium dissociation constant is very low ($K=10^{-24}$). Even with good theoretical thermodynamic data for the uranium minerals it is difficult to make accurate predictions about the behaviour of uranium in real natural systems. We know too little about the kinetics of these reactions and the catalytic/absorptive effects of clays and organic matter. For example, the observed concentrations of uranium in certain Italian spring waters, which are definitely depositing autunite, are extremely low and "are in considerable disagreement with the results given by Hostetler and Garrels (/905/) and by Muto and co-workers (/906/), in the sense that the samples analysed show a much lower uranium content than might be expected on the sole basis of the solubility of carnotite and autunite". Moreover, the ratios of element concentrations (e.g. U/V) in waters do not always correspond to the proportions of these minerals in the supposed source mineral (e.g. carnotite). "The theoretical equilibria with the secondary minerals are not the only processes conditioning the U and V contents in waters. The equilibria of each of the two elements with clay minerals, organic matter, etc. are more important in conditioning the U and V content in solution".

The importance of the activity coefficient in quantitative uranium hydrogeochemistry was demonstrated in a study undertaken by the authors on the Sila Plateau in Italy. They noted the distribution of uranium in spring waters and other waters and found that there was generally a clear relationship between high [U] on the one hand and high [HCO_3] and low TDS values on the other. This phenomenon was attributed partly to the influence of lithology and partly to structural/erosional controls related to differential uplift. It appears that those areas that have undergone relatively strong uplift have a more rapid hydrological cycle and that this in turn affects the rate of leaching of uranium. The uranium activity in these samples was calculated from the formula

$$\log(([UO_2(CO_3)_2(H_2O)_2^{2-}])/([UO_2^{2+}])) = 6,9 + 3 \log [HCO_3^-] + 3pH.$$

Anomalies defined by the calculated uranium activities showed a much closer correspondence with known autunite deposits than the simple µg/l concentrations of metal did. It could also be shown that all autunite deposits above the present water table were losing uranium to groundwaters whereas those below the water table were apparently still actively growing.

In this paper Dall'Aglio gives some information about the behaviour of uranium in the saline waters that occur in very arid regions and which are probably responsible for the uranium mineralization found within the calcrete mantle in certain of these areas. The example he uses is the calcrete* carnotite deposits of the Yeelirrie Basin in Australia. Uranium concentrates strongly in evaporating solutions (Figure 5, page 204) and is not removed unless organic matter or clay minerals are present to cause reduction or absorption. Organic matter is often scarce under these extreme desert conditions because of the small supportable biomass. It is notable that vanadium is not as mobile as uranium under these conditions and that the accumulation of V in the evaporation sequence is not a certainty.

The author's work on evaporating brines (Dall'Aglio and Casentini, 1970/907/) has confirmed that this process of uranium accumulation proceeds,

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* Sanders /908/ gives an account of the hydrogeology of calcrete formations in Australia.

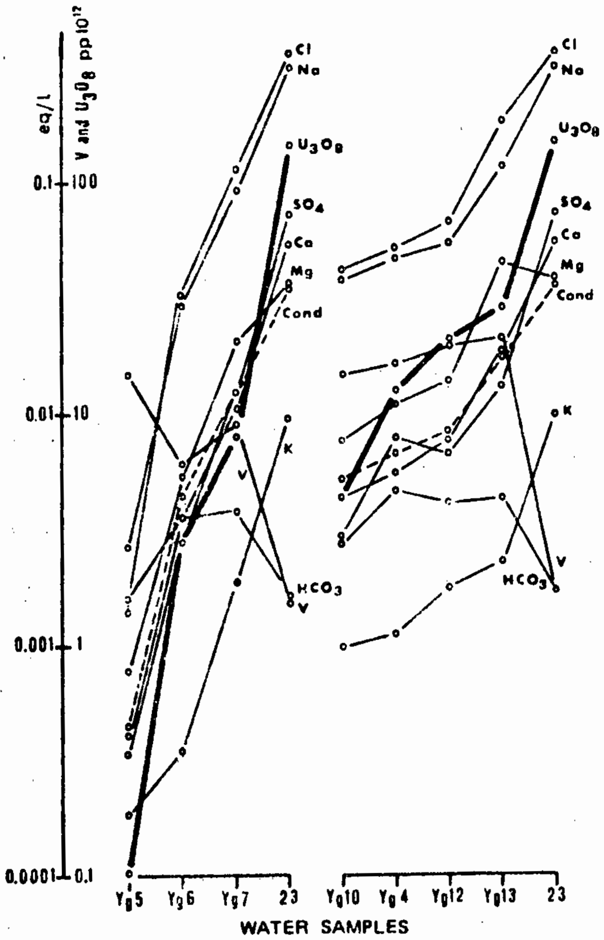

FIG.5. U and V contents, and chemical composition of water samples from a calcreted basin from Australia. Samples arranged according to the increasing maturity of the waters.

literally, to the bitter end. The metal is concentrated in the final bitterns of salt ponds and is not carried down by the precipitation of either gypsum or halite. However, it will be readily removed from brines by either clay minerals or organic substances.

Despite the known importance of seasonal fluctuations in the chemical composition of groundwaters the number of research publications on this problem is rather small. Dall'Aglio and Giannotti provide a useful discussion of the subject in their paper dealing with the distribution of certain elements in springwaters near the Casaccia Nuclear Centre in Italy (1967/910/). Measurements made three years apart showed significant changes in TDS and in the relative proportions of the major components of the springwaters. The concentration of uranium also varied (Figure 6) but the overall distribution of U/TDS ratios did not show too much scatter (Figure 7). The abundances of B and F also changed during the three year period. The authors noted that the mobility of boron is very high and that it is not greatly affected by changes in Eh. The concentration of fluorine appeared to be inversely proportional to $[Ca^{++}]$.

In another publication (1971/888/) Dall'Aglio examined the causes of false uranium hydrogeochemical anomalies that occur in metamorphic basement rocks in the Italian Alps. Here the abundance of the metal in rocks averages 5 μg/g and no deposits are known to occur, yet pronounced concentrations of uranium are encountered in some spring waters (10 μg/l, equivalent to 100 μg/g in the dry residue). These springs have a shallow, rapid circulation and are cold, alkaline (pH 8), oxidizing, very dilute (TDS about 100 mg/l) and relatively rich in $CaHCO_3$. Some sulphate is contributed by the weathering of pyrite but there is generally no large-scale hydrogeochemical attack on rock minerals. The U/TDS ratios of these northern Italian waters are therefore extremely high (Figure 3). This is a marked exception to the rule that

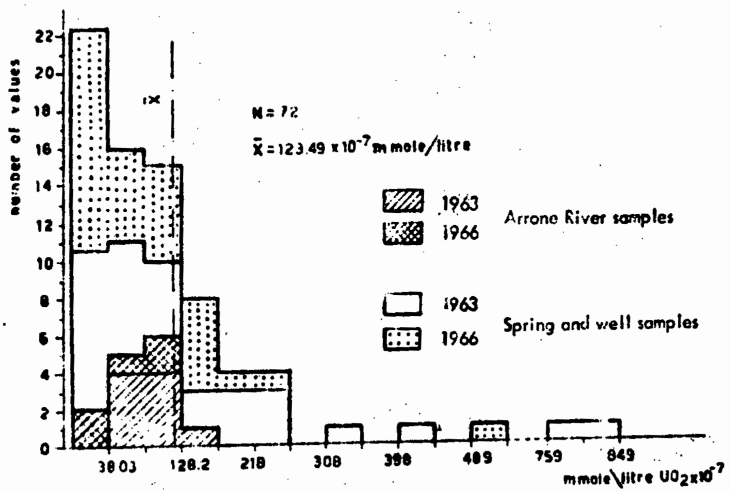

FIG. 6. Histogram of the uranium content of the waters.

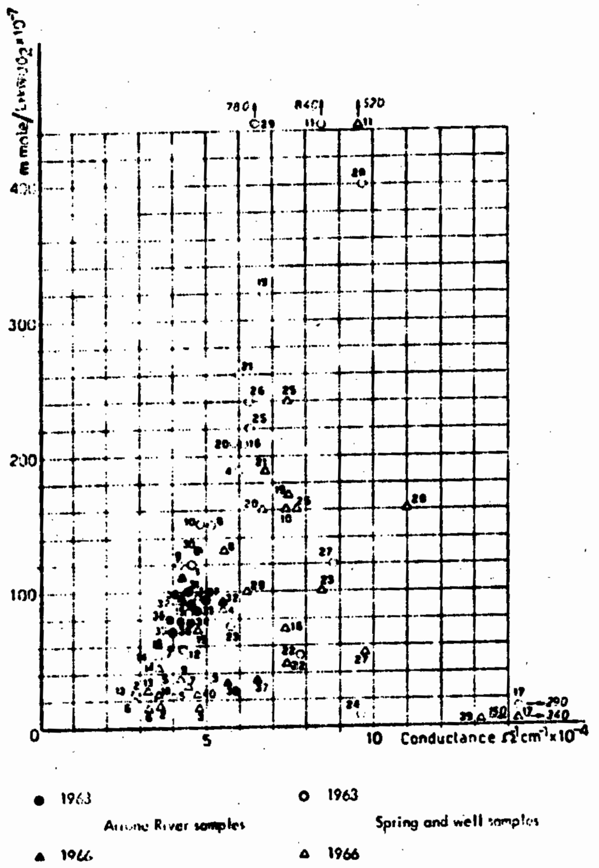

FIG. 7. Diagram of the uranium content versus electrical conductance.

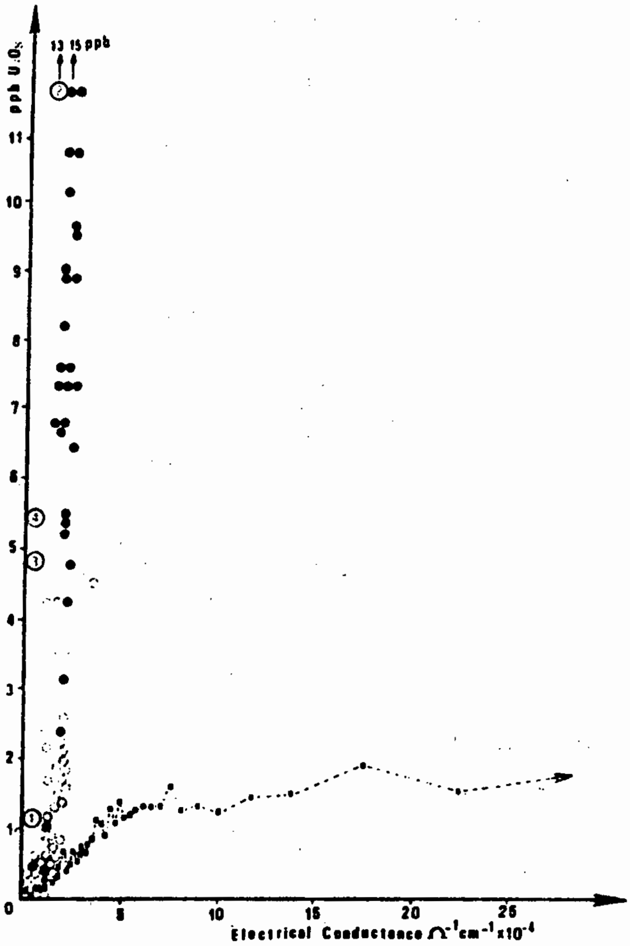

Fig. 3. Uranium contents vs. electrical conductance values. ○ represents water samples from the Venosta Valley, ● represents water samples from the Novale di Soi Creek. Numbers in circles refer to leaching tests carried out on various types of rock samples, as follows: 50 g of rock, ground to minus 1 mm were continuously stirred with 500 ml of distilled water. After 33 hr of mixing, the waters were filtered and analyzed. The dashed line represents the regression analysis of uranium contents on the electrical conductance values. This calculation was carried out by means of an especially prepared computer program (see text). This regression analysis is based on more than 4000 water samples from many Italian regions. Each point of the regression curve is the mean value of 100 analyzed water samples. This curve represents the "normal" processes of uranium dissolution in the environmental conditions of Italy. These data show that the ratio uranium/conductance is much higher than normal in the water samples from the Novale di Soi Creek, and that the leaching tests furnished the highest values of the above ratio.

"uranium contents in natural waters tend to increase as the waters mature in the hydrological cycle", i.e. as TDS and electrical conductance rise with progressive rock weathering and dissolution. Leaching experiments showed that the uranium in the country rock occurs in a very soluble form. Dall'Aglio implies that the rapid dissolution of uranium from these rocks may be in some way linked to the concomitant release of sulphate.

Rose and Keith (1976/1110/) reported an investigation of the stability of uranium in hydrogeochemical samples. A small degree of adsorption onto the walls of plastic vials was noted. Filtration caused a 10 - 20% drop in [U]. They noticed severe seasonal effects on the concentration of the metal in natural waters. Within a year the amount of uranium in a spring near a uranium occurrence at Penn Haven Junction, Pennsylvania, dropped from 14 to 0,2 μg/l. However, the value did not rise again and "so the effect is not a simple seasonal one. The temporal decrease of U in water is not related to any decrease in conductivity of the water, so the decreases do not appear to be a simple dilution ..... An obvious interpretation of this pattern is that U is appreciably leached from the ores only during dry periods" (not necessarily occurring every year) "when groundwater levels are relatively low".

Butler (1969/926/) reviewed the relationship between subsurface waters and the sandstone uranium deposits of Wyoming. It is this author's opinion that the most powerful single factor controlling the large-scale movement of dissolved species in groundwater is gravity. Figure 1 (reproduced from Domenico *et al.* /927/ is a useful idealized representation of the broad patterns of groundwater movement in a river valley in an arid area (Las Vegas). Butler observes that "there were flowing springs near the centre of Las Vegas valley, although surface runoff rarely reached that far. The water discharged by these springs moved upwards from hundreds of feet in depth transversely to the stratification". Clearly, one would have to be very careful about the interpretation of chemical data from such springs.

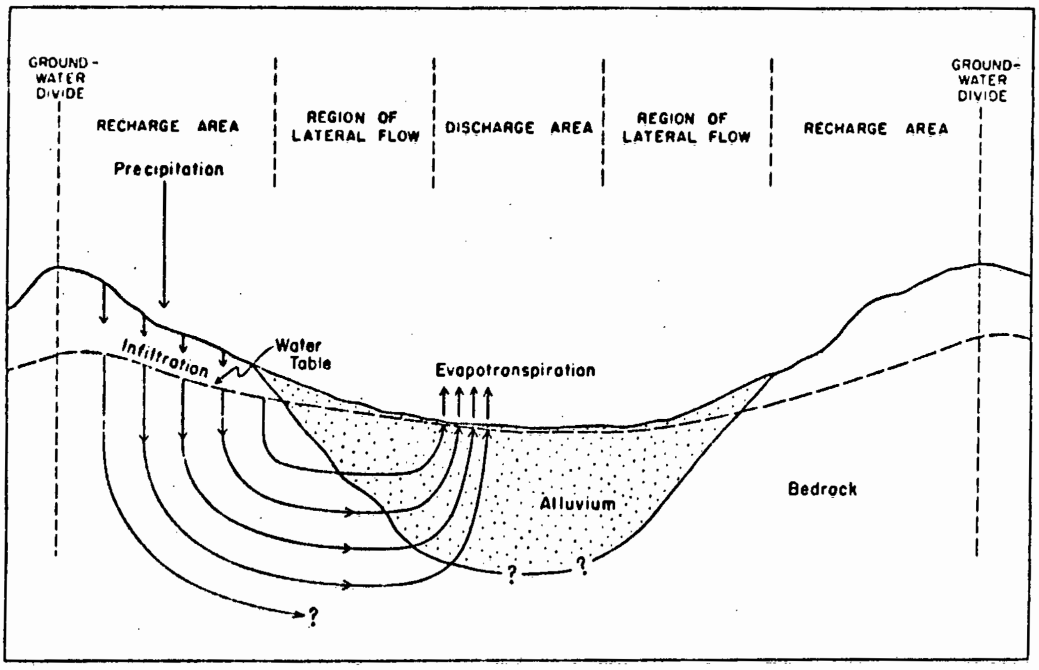

Figure 1. Idealized flow system of ground water, Las Vegas Valley, Nevada. From Domenico, Stephenson, and Maxey (1964, fig. 9).

It is Butler's view - and one shared by many other writers (e.g. /541, 928/) - that uranium in groundwater "may serve as a general guide to an area in which deposits occur ..... (but) is, however, a very uncertain guide to the deposits themselves". One of the principle problems in groundwater hydrogeochemical exploration is the variation in the background value, which may change both laterally and vertically. For example, water in the vicinity of deep uranium orebodies in the Shirley Basin of Wyoming have no more uranium than the waters from barren overlying formations that have suffered a higher degree of oxidation. It is impossible to say whether the uranium in the water of the barren formations represents an anomaly related to the deeper ore, or whether the metal has merely been leached from disseminated uraniferous matter in the zone of oxidation. Despite these problems, the author believes that the background value for uranium in groundwaters in Wyoming may be set at 5 µg/l, and the anomalous threshold at 10 µg/l. He notes a proposal that had been made by Cheney and Jensen (1966/929/), to the effect that sulphate anomalies, at a threshold value of 450 mg/l, were a possible ore guide in Wyoming. However, he presents data that show that this hypothesis is invalid and that it is statistically impossible to relate areas of high sulphate and high uranium with any confidence.

A major uranium exploration survey (National Uranium Resources Exploration or NURE program) has been in progress in the U.S.A. since 1973 and some of the groundwater hydrogeochemical data is becoming available in the form of preliminary reports /874-8/. The aims of the project have been described by Ferguson and Price (1976/1109/). Little of the data has been thoroughly interpreted yet by the workers responsible for the program but the project obviously has great potential because of the vast scale on which it is being planned. Neutron activation techniques have made possible, for the first time, the determination of a very wide range of unusual elements in groundwaters at concentrations as low as a few nanograms per litre. Some representative data are compiled below for spring and well waters from the Winnemucca Dry Lake Area:

U	0,1 - 57 μg/l	Mo	6 - 120 μg/l
$SO_4^=$	19 - 40 mg/l	Sr	15 - 600 μg/l
Sb	30 - 200 ng/l	Ta	about 70 μg/l
As	2 - 20 μg/l	Th	10 - 100 ng/l
Ba	3 - 60 μg/l	W	0,7 - 2 μg/l
Br	2 - 40 μg/l	Y	about 25 μg/l
Cr	0,6 - 2 μg/l	Zr	about 280 μg/l
Co	16 - 354 ng/l	Fe	50 - 260 μg/l
Au	about 0,7 ng/l		

The NURE program includes special wellwater projects, with seasonal variation studies. The data thus far collected indicate that in some areas at least (for instance the eastern U.S.A.) "ground water is an excellent indicator of regional trends in uranium mineralisation". In some regions with high background values uranium anomalies were in fact detectable only by hydrogeochemical methods /878/. It is estimated that in some areas the composition of a groundwater sample is representative of about one square kilometre of rock. The program has scheduled 100 000 wellwater samples by 1981.

Some of the NURE publications (e.g. /878/) include useful appendixes on sampling procedures, sample handling, equipment etc.

Hydrogeochemical exploration for uranium in Canada became important in the early 1960's and has subsequently grown rapidly /821, 824-5, 828/. Chamberlain (1964/821/) recorded the results of the first major Canadian hydrogeochemical study of uranium, which took place in the Bancroft-Haliburton area and included some sampling of groundwaters. Here the uranium occurrences are related to syenitic rocks and there is a fair but not foolproof association between zones of syenite and bodies of uraniferous waters. Waters emerging from uraniferous ground contained 200 to 500 μg U/litre.

Dyck is one of the leading Canadian writers in this discipline and he has recently produced a fine summary of the role of groundwater analysis in uranium prospecting (1975/879/). He believes that uranium is perhaps the most mobile of all trace elements. Dissolved uranium complexes strongly with carbonate, sulphate and silicate in basic, acidic and neutral waters respectively and is immobilized by organic matter. It may be that organic matter is the principle cause of the concentration of the metal in sedimentary rocks.

Dyck has summarized the most important practical findings of several major Canadian hydrogeochemical projects. In the Beaverlodge area the hydrogeochemical data had to be corrected for the effect of organic matter and uranium background values were observed to be far higher than normal in tracts underlain by carbonate rocks. Dyck reported that "uranium may disappear into the wall of bottles upon long storage". He notes that the following criteria will be of help in assessing hydrogeochemical anomalies: (i) An increase in [U] to three times background or more. (ii) An increase in the concentrations of elements "such as Mo, Pb, Cu, Zn, As, P and V". (iii) "A sharp rise in concentration (of U) after a rain ..... up to ten times" (normal). It is useful to know the rate of turnover of groundwaters (i.e. surface to ground recycling) and both TDS and $[O_2]$ can be used to estimate this rate.

Dyck *et al.* (1976/897, 1142/) described the results of wellwater reconnaissance orientation surveys in southeastern Saskatchewan. They explain that this method of exploration is relatively new in Canada because surface waters are abundant over much of this nation. However, in Saskatchewan "extensive overburden has made it difficult or impossible to detect uranium mineralization in bedrock by the conventional ..... tests". Fortunately "wells penetrate the overburden in many places and therefore can serve as windows to see deeper". About 1000 groundwater samples were collected from an area of 17900 km^2 and the aim was to keep the sample density at one per 13 km^2. Uranium, Rn, F, O_2, Eh, pH, alkalinity and conductivity were determined or measured. Uranium concentrations of up to 240 μg/l were encountered but were not regarded as representative of "ore grade environments". The waters are alkaline and the uranium anomalies were attributable to the weathering of radioactive coal seams. The distribution of [U] in the samples was quite regular and it was possible to contour the data without too much trouble. Background values varied in different lithological formations and the chemistry of the spring samples and the well waters could be equated. It is important to note that the composition of the groundwater changes with depth. This is in accordance with the established principle of decreasing Eh with depth, which leads to reduction of U^{6+} to U^{4+}.

Dyck *et al.* (1976/1111/) recorded the results of additional regional wellwater reconnaissance surveys in eastern Canada. They found that U, Rn, He, F, conductance and alkalinity show systematic patterns indicating broad belts of regional enrichment of uranium. The distribution of Zn, Cu, Pb, Mn and Fe "is more spotty than that of the uranium elements". Most of the observed anomalies can be correlated with known mineralization. They noted that in eastern Canada, as in Saskatchewan (*vide supra*), changes in the depth of wells was an "important cause of data distortion. One should in principle produce three-dimensional maps. In practice that would require much greater sample densities ..... and would involve the expense of drilling new holes". The sampling density here, as in the Saskatchewan project, was one per 13 square kilometres. The authors observed that the contrast between anomalies and background levels was higher in groundwaters than in stream sediments in this region. This was especially true for uranium in those areas with waters of high alkalinity.

Jonasson and Gleeson (1976/893/) used spring waters (*inter alia*) in a low density, helicopter supported reconnaissance survey of uranium potential in parts of Yukon Territory. The elements determined were U, Zn, Cu, Pb, Fe, Mn, F, Cl, Si and sulphur. The best pathfinders appeared to be U, F and Zn, with Cu and sulphate ion showing promise in certain areas. Lead was not useful. Waters from old adits were very metalliferous if the pH was below 6. The authors found that springs only 100 ft. apart yielded waters that were radically different in trace element composition. They noted that uranium in spring waters was more stable than Zn or Fe but that iron oxides or hydroxides had a definite scavenging effect on the dissolved uranium.

Darnley (1975/887/) *et al.* chose subsurface waters as the principle sampling medium for uranium prospecting in areas of thick overburden and flat-lying sediments in the Federal Canadian Uranium Reconnaissance Program. They proposed to use Dyck's methods /879/. Several other publications /894-6/ deal briefly with the preliminary results of the Canadian wellwater sampling project.

Fauth (1973/1085/) described a large-scale, low-cost hydrogeochemical operation in the Black Forest of Germany during which 5000 spring-, well- and surface waters were sampled and analysed for uranium by polarography. Data were fed to a computer via a digitizing table and interpreted by means of frequency distribution diagrams and trend surface analysis up to the seventh degree. The latter approach yielded poor results, whereas simple inspection often revealed important anomalies. There was in fact little problem with the interpretation of the data from these relatively fresh waters, in which the background value for uranium was about 0,5 µg/l and the threshold was 3,0 µg/l. All known uranium occurrences were delineated by the hydrogeochemical data and some new anomalies were revealed but this information was inadequate for a detailed assessment of the size or grade of the new finds. Nevertheless these positive indications were important, as they had been missed by radiometric surveys.

Southern Africa is one of the principle uranium provinces of the world and so it is not surprising to find that large-scale hydrogeochemical exploration programs have been in operation here for the last few years. The subcontinent is generally rather arid, with few perennial streams and so sampling is restricted almost entirely to groundwaters. Over the years a number of publications had appeared (e.g. /228, 1012-3, 1017/ that dealt with relevant aspects of the chemistry of groundwater in this region but true hydrogeochemical prospecting studies were unknown before 1968 or 1969. Almost all of the effort made in the past ten years has been directed towards the search for uranium. Most of the data accumulated to date is unpublished but some reports have appeared in Geological Survey publications and elsewhere (e.g. /993-4, 1000, 1005, 1015-6.

Hambleton-Jones (1976/990/) studied the geochemistry of some subsurface waters of the Namib desert in a uraniferous area near the Swakop River. The deposits here occur in duricrusts and were apparently formed as the result of precipitation of uranium from groundwater. The authors' aims were, *inter alia*, to establish hydrogeochemical prospecting criteria for such ores and to elucidate the mechanisms of uranium migration in very arid environments. Some typical data are compiled in the following Table (page 216). Some

MEANS AND RANGE OF CONCENTRATIONS OF IONS IN SUBSURFACE WATERS FROM THE NAMIB DESERT FOR 31 SAMPLES (AFTER HAMBLETON-JONES /990/)

	MEAN	MINIMUM	MAXIMUM
pH	7,7	7,0	9,2
Cond (ppm)	5 933	1 800	18 500
TDS (ppm)	4 306	1 130	15 285
Na (ppm)	721	136	3 480
K (ppm)	49	6	340
Ca (ppm)	512	118	1 638
Mg (ppm)	142	49	431
U (ppb)	81	6	342
V (ppb)	27	3	79
Sr (ppb)	5 754	826	47 772
Ba (ppb)	271	31	766
SO_4 (ppm)	585	89	1 596
NO_3 (ppm)	54	2	165
SiO_2 (ppm)	28	10	40
F (ppm)	1,2	0,4	3,5
Cl (ppm)	1 639	340	7 700
CO_3 (ppm)	130	54	219

analyses of Pb, Cu, Ni, Ag and Zn were performed but these values all fell in the lower part of the µg range and did not correlate with [U].

The present-day waters in this area are Na-K-Cl or Na-Ca-Cl-SO_4 brines in which the concentrations of some components such as K and U vary widely. These waters are saturated with respect to calcite but are depleted in magnesium. The concentrations of most components increase with TDS. Samples were collected over a wide area and from bedrock formations of several different lithologies. The data were interpreted with the aid of a multivariate discriminant analysis program (BMD07M). This showed that the samples were readily separated into four groups on the basis of the concentrations of U, Na, carbonate and sulphate. However, the samples in these groups were not rationally distributed within the study area - there seemed "to be no uniformity in the distribution of the samples in their groups". The author believed this to be due to differences in the depths of the boreholes that were sampled, since adjacent holes did not necessarily tap the same body of water. In this manner the true rational relationship that exists between water chemistry and rock lithology is disguised. Calculations showed that most of the mass of dissolved ions present could have been derived from the destruction of rock minerals, especially feldspars. The author believed that this destruction was promoted by the repeated passage of ephemeral, meteoric waters, which are rich in carbon dioxide and chemically reactive. On the other hand, the relationship between dissolved uranium and the content of the metal in calcretes and soils is unambiguous. They are generally very well correlated - although there are naturally some exceptions - and the threshold value for waters associated with "zones of potentially high uranium concentration" is 30 µg/l.

The concentration of uranium correlated closely with [$CO_3^=$] in all waters and in one group it was also clearly related to pH. For these particular samples the relationship

$$\log [U] = c_1 + c_2 \log [CO_3^{=}] + c_3 pH \quad (c_1 \text{ to } c_3 \text{ are constants})$$

was obeyed very well. Why this was not true for all the waters is not clear. The author discussed some of the reasons for the precipitation of uranium and the formation of the duricrust ores. It was his opinion that the vanadate ion is a very important cause of the dissociation of the uranylcarbonate complex in these waters.

In his discussion of the possible application of these observations in regional groundwater hydrogeochemical reconnaissance, Hambleton-Jones stressed that uranium analyses and other chemical data will be practically useless without some geological and/or hydrological controls. Mass or "blanket" sampling of vast areas is no substitute for these controls and will not prove to be the exploration geochemist's panacea. He notes, for example, that it is now conceded that the vast ERDA program, conducted on the "blanket" principle in the U.S.A. in 1975-6, was somewhat less than successful and was being revised and scaled down. It is preferable to work from the specific (i.e. small integral units) to the general, larger unit.

Hambleton-Jones also furnishes a lengthy discussion of the use of uranium isotopic ratios in water as a guide to mineralization.

Beeson (1977/991/) correlated most of the available data on seasonal variations in the uranium content of groundwater from the Beaufort West area* of South Africa. A set of fifty-one boreholes in this area was sampled on three separate occasions. Sample set A was collected "at the end of a relatively dry summer, when ground water levels were quite low" (April-May 1975).

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* This area is widely referred to as "the Karoo".

"Little rain fell between (the collection of sets) A and B and the ground water table remained fairly stable" (November 1975). "The period from December 1975 to March 1976 was one of the wettest in recent years, and ground water levels rose sharply and remained high ..... until the final sampling project C" (Dec. 1976 to Jan. 1977). It was Beeson's opinion that these sample sets "encompassed the likely extremes of seasonal variation, and as heavy rain fell during each sampling project, it must include all of the possible short term variations that will effect (*sic*) the trace element concentration of the ground water". The three sets of data also include the extremes of probable analytical variation, because the analytical method was changed before the samples in sets B and C were processed. The results of this study are best illustrated by example (Table 3). The data are quite encouraging in that there are no radical temporal changes in [U]. However, some individual changes are significant and could adversely influence the reliability of sets of data collected over a long period.

In a separate study in the same region, Beeson (1976/992/) made the important observation that the background concentration of dissolved uranium was dependent upon topography. In the mountainous escarpment areas of active recharge and young waters the value is about 5 µg/l, but in the lower--lying, flat areas below the escarpment, where well-travelled, older waters occur, the background [U] is as high as 30 µg/l.

Additional reports on the distribution of uranium in groundwaters at Beaufort West were prepared by Murphy (1977-8/995-6/). Over 600 samples were collected from farm windpumps within a large area around this town, and the data show three major regional populations of uranium values. In Murphy's opinion the patterns of distribution of data reflect the influences of "topography, geology, hydrology and groundwater chemistry". The effect

TABLE 3 : URANIUM VALUES OBTAINED DURING REPEATED SAMPLING OF BOREHOLES IN PPB

Date of Sampling Sample No.	April 1975	Nov. 1975	Dec. 1976	Mean
MC 61	2,0	3,8	3,5	3,1
MC 4	4,0	2,6	5,4	4,0
MC 64	4,0	5,5	6,1	5,2
MC 75	8,0	9,3	5,0	7,4
MC 65	2,0	1,8	3,2	2,3
MC 66	2,0	1,9	3,5	2,5
MC 67	2,0	2,2	6,4	3,5
MC 5	4,0	7,1	6,7	5,9
MC 7	2,0	1,6	3,5	2,4
MC 74	4,0	3,1	5,6	4,2
MG 84	40,0	22,0	19,1	27,0
MG 101	14,0	15,9	17,7	15,9
MG 122	8,0	10,9	13,8	10,9
MG 118	33,0	31,2	27,0	30,4
MG 115	34,0	40,3	50,0	41,4
MG 125	2,0	0,8	1,1	1,3
MG 4	24,0	14,3	12,2	16,8
MG 1	10,0	16,4	10,9	12,4
MG 106	18,0	23,1	19,6	20,2
MG 25	32,0	31,9	37,0	33,6
MG 22	20,0	45,4	54,0	39,8
MK 8	15,0	24,2	18,5	19,2
MK 9	13,0	17,4	19,8	16,7
MK 6	4,0	5,7	4,9	4,9
MK 7	18,0	17,2	19,6	18,3
MK 5	2,0	7,5	8,6	6,0
MK 4	4,0	8,5	8,6	7,0
MK 79	24,0	19,8	20,9	21,6
MK 77	10,0	12,1	12,3	11,5
MK 76	12,0	16,4	15,7	14,7
MK 75	18,0	19,3	13,1	16,8
MK 74	7,0	11,9	13,1	10,7
MK 122	8,0	13,5	6,5	9,3
MK 110	9,0	15,0	14,5	12,8
MK 108	3,0	12,3	4,0	6,4
MK 111	3,0	9,0	8,4	6,8

of topography is paramount and the concept of a "sliding scale" background value must be applied to data from regions of differing relief (Table 2). There are naturally some areas that do not fit the general pattern, and variations* and trends occur on a small scale within all three major zones. Geohydrological studies have helped to explain this phenomenon, which is attributable to the ageing of waters moving down from the mountainous escarpment and into the lower plains. "Young meteoric water from the high relief areas (have) a short residence time in these rocks, and there is little time for salts and minerals to be dissolved. On entering very low relief areas ..... because of the lower pressure head and gradient ..... they are in contact with salts and minerals for a much longer period. Consequently the amount of dissolved salts normally increases with the age of the groundwater". A complicating factor is the presence in this region of dolerite dykes that form compartments that sharply separate groundwaters of different quality and therefore different background concentrations of uranium. Murphy feels that the interpretation of the data is hindered by a lack of knowledge of the drilled depth of boreholes and the position within them of major aquifers.

Upon taking all of these facts into account (as far as possible) and interpreting the data subjectively by plotting them on 1:50 000 topographic sheets, several anomalies were apparent. Some are related to known uranium mineralization but others are not and are still to be investigated. The data were also examined statistically by plotting logarithmic cumulative frequency curves for the various sample subgroups (Figure 2). These curves show that the distribution of uranium is indeed controlled by topography and that [U] is approximately lognormally distributed within each topographic group.

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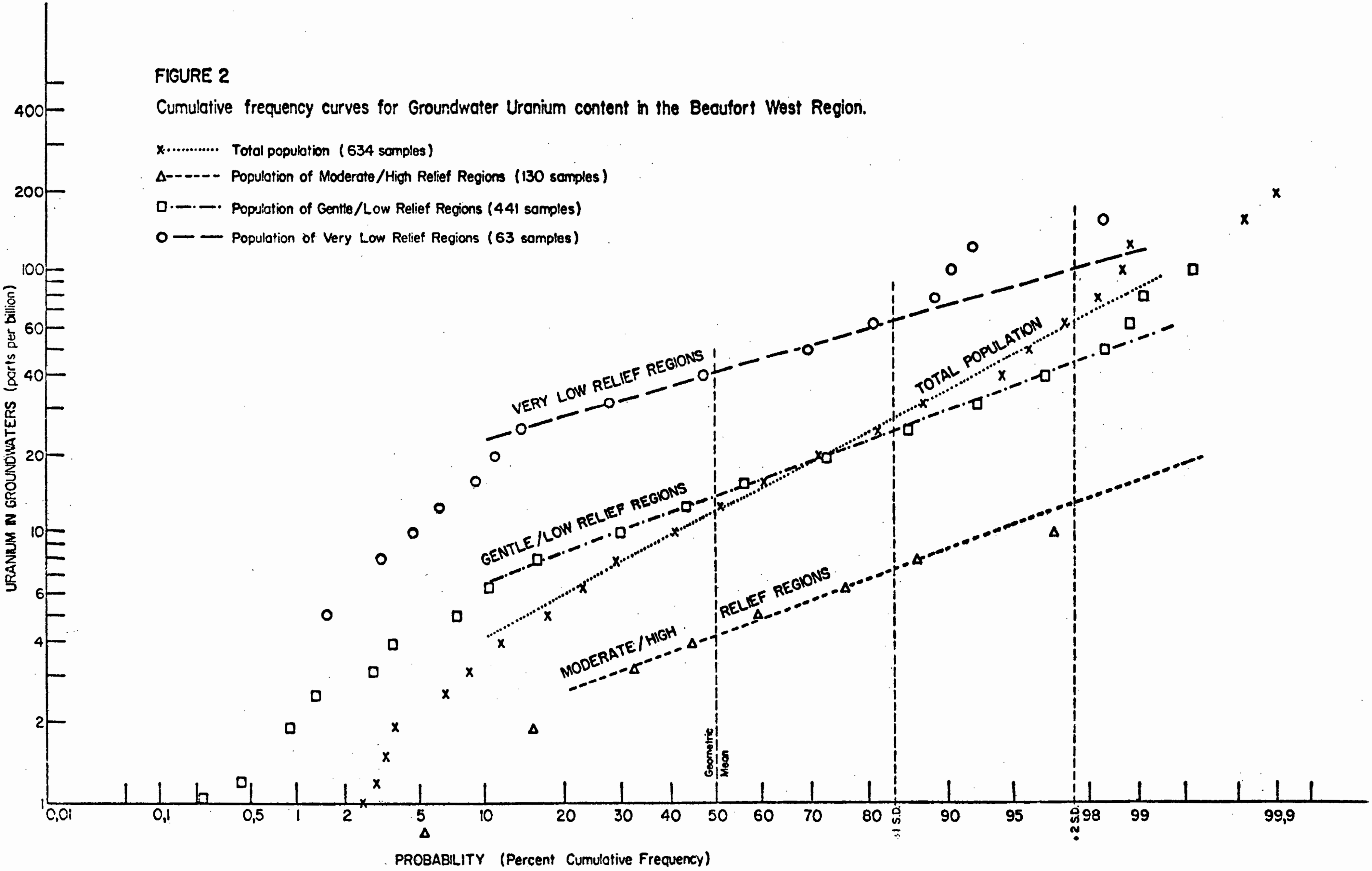
* For example, U background values appear to be enhanced in areas covered by calcrete.

TABLE 2 : SUMMARY STATISTICS OF URANIUM CONCENTRATIONS IN GROUNDWATERS IN THE THREE TOPOGRAPHIC REGIONS

	No of samples	Geometric Mean ppb	$\bar{X}$ + 1 S.D. ppb	$\bar{X}$ + 2 S.D. ppb
REGION I Moderate to high relief	130	4,8	7,4	13
REGION 2 Gentle to Low relief	441	14	25	44
REGION 3 Very low relief	63	41	64	100
TOTAL POPULATION	634	12	28	64

FIGURE 2

Cumulative frequency curves for Groundwater Uranium content in the Beaufort West Region.



Murphy feels that the very high mobility of uranium in the Karoo will make it rather difficult to track anomalies, and suggests that the routine chemical surveys might be coupled with measurements of Rn or Ra in order to give the method more focus. This possibility has not yet been investigated. The author also points out that highly reducing waters have occasionally been encountered in the Karoo - although the bulk of the shallow groundwater here is undoubtedly well oxygenated - and that the significance of these "black waters" ought to be very carefully assessed when they are encountered during hydrogeochemical sampling. Marchant (1978/997/) showed that these "black waters" consist of suspensions of reduced Fe-Mn compounds, which are poor in organic matter but relatively rich in Ti, Cr, Ni, Cu, Zn, Sr and tin.

Brunke (1977/1007/) conducted groundwater hydrogeochemical orientation studies around uranium deposits in the Karoo. His thesis is a document of nearly two hundred pages and it cannot be adequately reviewed here. His principle conclusions may be summarized as follows: (i) Of the many elements examined in these waters, only U and Mo appear to hold any promise as indicators of uranium mineralization. (ii) On the local scale the correlation between the groundwater anomalies and known deposits is sporadic. The reasons are not yet fully understood. (iii) [Mo] is not as dependent upon [TDS] as [U] is, but is nevertheless the less reliable of the two indicators. (iv) [U] is highly dependent upon TDS, and [U]/electrical conductance ratios must be used to normalize all raw [U] data. In addition, waters must be subdivided on the basis of topographic factors, and each subgroup must be assessed independently. (v) It is necessary to stretch the sample coverage to the maximum by making use of every single groundwater source. Single-hole anomalies are common. (vi) It appears that [U] in anomalous waters is more subject to seasonal variation than [U] in background waters. This phenomenon is well-known in other parts of the world. "A rise in water level may initially leach substantial amounts of oxidized ore ..... before the U content tails off to an equilibrium abundance level". Thus [U] in water in unmineralized rock may vary between 5 and 10 µg/l over one year, but in an anomalous water it could vary from 7 to 50 µg/l during the same period. It will obviously be difficult to make practical use of this fact for purposes of regional reconnaissance but it could be used to "zero in" on groups of boreholes that are suspected of yielding anomalous waters. (vii) Water migrating down-slope changed significantly in composition within a few hundred metres, but was remarkably homogeneous over a profile depth of up to 80 m at any given point along the traverse.

Some hydrogeochemical prospecting for uranium in the Northern Cape has also been reported /998-9/. Wilkenson (1977/998/) described a project in which 216 wellwater samples were collected from this granitic, desert area and analysed for uranium by delayed neutron counting /993/. The survey was part of an attempt to follow up airborne radiometric surveys, and was adopted because it was clear that certain secondary radioactive anomalies must have been produced by the movement of subterranean water. The concentration of uranium varied from 0 to 356 µg/l, with a mean value of 53 µg/l. Ten samples had concentrations in excess of 150 µg/l. The high values appeared to be randomly scattered and boreholes within a short distance of one another yielded widely varying concentrations. There appeared to be no correlation between the hydrogeochemical anomalies and the radiometric highs. There was also no simple relationship between the uraniferous waters and local lithology or known occurrences of mineralization. There was, however, some evidence of a complex relationship between topography and [U]. It is clear that the hydrogeological phenomena in operation here are highly complex and will not be unravelled without substantial additional work in the region.

4.6 THE FUTURE

The most important advances in the immediate future are most likely to be in the field of data interpretation. There is little doubt that the next few years will witness a "quantitative revolution" in hydrogeochemical exploration. Very sophisticated computer techniques will be used to perform statistical analyses of regional data and to examine background and anomalous values in terms of complex, simultaneous solution/mineral equilibrium phenomena. Several recent publications /841-3/ indicate that a start has already been made in this direction. The observations of Hitchon (1976/843/) are particularly relevant: "The subsurface situation is one of potential continuous fluid-rock interaction as the moving fluid is chemically and physically equilibrated and then re-equilibrated as it passes rocks and environments of differing chemical and physical properties ..... As we become more sophisticated in our approach to fluid-rock interaction it will be possible to consider solid, liquid and gaseous organic phases reacting among themselves and with the inorganic phases. In both the inorganic and organic systems it should be possible, ultimately, to take into account stable and radioactive isotope transfers. ..... There exist the possibilities of equilibrium reactions incorporating irreversible changes, such as may occur in weathering, diagenesis and evaporative concentration, osmotic and ion exchange phenomena with clay minerals, the non-ideal behavior of the gas phase, incongruent reactions, chromatographic effects, volume changes in the fluid phases or due to the precipitation or solution of reactants, and of course, the possibility of partial or local equilibrium ..... The computed equilibrium is independent of time and represents a static case, that is, given sufficient time, the equilibrium state ..... will be reached eventually. However, as previously noted, the subsurface circuit is a system in dynamic equilibrium, both with respect to fluid flow and fluid chemistry ..... A start has been made ..... to link hydrodynamics and hydrochemistry by developing a partial equilibrium simulation model of the chemical state of a regional groundwater flow system that incorporates mass transfer rates and reaction kinetics. These valuable initial steps will undoubtedly lead, before the end of this decade, to comprehensive, powerful computer programs that can completely simulate the hydrodynamic and hydrochemical equilibria present in the subsurface circuit, both for inorganic and organic parameters ..... (There is an) extreme need for a new approach to the sampling and analysis of waters so that they are compatible to the sophistication of the present computer programs for mineral solution equilibria. It seems highly likely, in my opinion, that the most relevant approach to exploration for mineral deposits through examination of fluids might be statistical analysis of regional mineral solution equilibria data - for example, a trend surface analysis or map of factor scores using mineral saturation equilibria data as the input. This should be regarded as a minimal effort, which can be supplemented later, when the more advanced hydrodynamic--kinetic reaction models ..... become available, as they surely will".

CONTENTS

NOTES ON THE REFERENCE LISTS

(A) The abbreviations used in the reference lists are, by and large, those defined in the following publication:

BROWN, P. and STRATTON, G.B. (1963) World list of scientific periodicals /282/.

(B) The lists of references have been produced by a line printer and consist of two parts:

1. Sequential listing of publications according to the reference tag number used in the text.

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(C) The line printer used cannot reproduce quotation marks ("). This appears in the reference lists as the symbol _ .

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