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FORMATION AND PROPERTIES OF RADIOCOLLOIDS
IN AQUEOUS SOLUTION - A LITERATURE SURVEY

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CONTENTS

	page
1. INTRODUCTION	1
2. TRUE COLLOIDS AND PSEUDOCOLLOIDS	1
3. METHODS FOR CHARACTERIZING RADIOCOLLOID PARTICLES	5
3.1. Dialysis	5
3.2. Adsorption	7
3.3. Filtration and ultrafiltration	8
3.4. Centrifugation	8
3.5. Autoradiography	9
3.6. Diffusion	9
3.7. Electrophoresis	10
3.8. Electronmicroscopy	10
3.9. Quasielastic laser light scattering	11
4. PARAMETERS INFLUENCING THE FORMATION AND PROPERTIES OF RADIOCOLLOIDS	11
4.1. The pH of the solution	11
4.2. The salt medium	12
4.3. The concentration of the radioactive element	13
4.4. Time and temperature	14
5. SUMMARY OF RADIOCOLLOID PROPERTIES AND STUDIES	15
6. REFERENCES	19

1. INTRODUCTION

The sorption of radionuclides on various rocks and minerals has been studied within many national waste programs as a means of predicting the migration behaviour of radionuclides that might be released from e.g. an underground repository for radioactive waste. One major objection against the conclusions that can be drawn from laboratory sorption studies is that the possibility of a formation of small fractions of highly mobile particulates are usually not considered.

The elements, present in spent nuclear fuel, which are most likely to form colloid species would be hydrolyzable elements like the actinides and possibly Sr as well as Pb and Cu representing the encapsulation material.

Moreover the radionuclides would be present in aqueous solutions in very low concentrations and under these conditions other phenomena occurs than at macroconcentrations.

This literature survey is meant to be a basis for further studies on the formation and transport of radiocolloids in the groundwater-rock environment. The colloids will probably not be retarded by the same mechanisms as dissolved species in true solution, but may in some cases migrate with the same velocity as the groundwater. Neretnieks made some investigations of colloids¹ within the Swedish program, but further investigations are desirable.

2. TRUE COLLOIDS AND PSEUDOCOLLOIDS

The investigations of radiocolloids began with a work of Paneth² in 1913. In this work it was shown that bismuth and polonium have colloidal properties in solutions at concentrations about $10^{-10}M$.

Hahn³ stated that at such low concentrations of radioactive elements, colloids are formed as a result of the adsorption of the radionuclide on impurities in the solution. These colloids are called pseudocolloids. The impurities in the solution are in principle dust particles from the air or colloidal particles from the vessel walls.

The rate of pseudocolloid formation is basically determined by the sorption rate on colloidal impurities. There is a lot of data available on pseudocolloidal formation rate. An equilibrium distribution of tetravalent plutonium between colloidal impurities, the vessel surface and the solution is claimed to be attained within 3-5 hours.⁶

Paneth², Starik⁴ and Haissinsky⁵ suggested that true colloids may be formed even at very low concentration of the radioelement. True colloids are formed by condensation of the molecules or ions as a result of hydrolytic or precipitation processes. Thus they are mostly hydroxides or polymers of ions formed by hydrolysis of the radioelement.

Starik discovered that the pH-range of maximum adsorption on an exposed solid and the maximum concentration of colloidal species did not coincide for polonium. This fact was considered to be an evidence for the existence of true colloids.(Fig.1.)

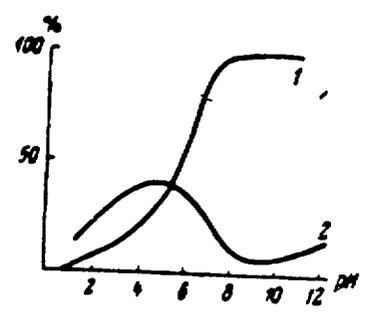


Figure 1. 1) Fraction of Po in colloidal state in solution
— 2) Percent of adsorption of Po on glass

The possibility of the formation of true colloids of radionuclides at very low concentrations was also confirmed by the radiochemical method of Skulskii⁹. Davydov⁶ stated that the problems of the nature of radiocolloids formed at very low concentrations in solution cannot be regarded as conclusively solved.

The following abbreviations were used:
TC - true colloids
PSC - pseudocolloids
NSC - presence of non-specified colloids proved
A - autoradiography
AD - adsorption

Generally the true colloids are very small, not larger than 10 nm in diameter. Both true colloids and pseudocolloids have different properties than species in true solution; they are subject to the force of gravity and they can be separated from the dispersing medium by a semipermeable membrane with small pores. These properties can be used for determination of colloidal forms of radio-nuclides.

Another important characteristic of radiocolloids is their charge, which affects their chemical behaviour and mobility in an electric field.

At the time when the radiocolloids were discovered, the colloidal behaviour of nonradioactive elements in such low concentrations was unknown. Thus many scientists thought that their formation was connected with the radioactivity. However, Ichikawa and Sato¹⁰ found in a study of europium that there is no difference in the colloidal behaviour between radioeuropium and stable europium. (Fig.2-3.)

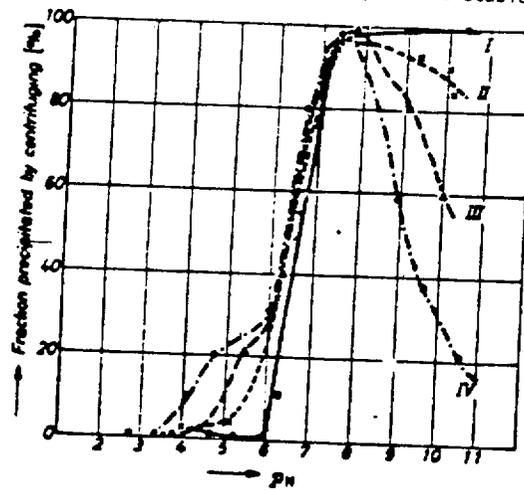


Figure 2. The dependency of the fraction precipitated by centrifuging of ^{155}Eu on the pH of the solution. Europium concentrations: I: 10^{-4} M, II: 10^{-6} M, III: 10^{-8} M and IV: 10^{-9} M (carrier-free)

References	
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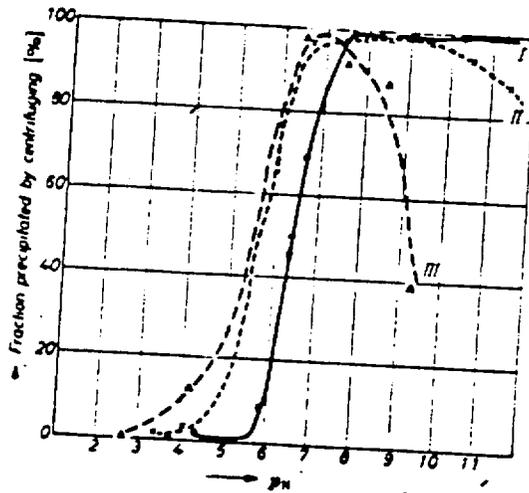


Figure 3. The dependency of the fraction precipitated by centrifuging of non-radioactive europium on the pH of the solution.

Europium concentrations: I: 10^{-4} M, II: 10^{-6} M and III: 10^{-8} M

The pseudocolloids are usually much larger than the true colloids, up to 500 nm. Pseudocolloids can be divided into two groups, reversible and irreversible. The former are produced by reversible surface adsorption and the radionuclide can easily be freed. In the latter case the element is either incorporated into the interior of the foreign particle or irreversibly sorbed on it.

A special case is the formation of pseudocolloids through interaction of the radioelement with organic particles of colloidal dimensions. Such organic particles, for example humus substances, are present in natural waters.

3. METHODS FOR CHARACTERIZING RADIOCOLLOID PARTICLES

Radiocolloids are largely defined by their size (number of atoms, molecular weight) and charge. Various methods have been used for the study of these properties. Some of them are shortly described here. It is usually not sufficient to use one of these methods. Starik recommended simultaneous study of at least the adsorption, as well as colloidal properties of the elements. Warbick et.al.¹¹ made a review of the methods available for determining the size of radiocolloids (Table 1), and made a comparison between three of these methods, ultrafiltration, centrifugation and electronmicroscopy.

3.1. Dialysis

The dialysis method was first used by Paneth² to study the colloidal state of nuclides in low concentrations. It is based on the fact that colloidal particles do not pass through membranes with pores smaller than the particle diameter. There are some complicating factors such as osmosis, membrane hydrolysis etc. and to avoid the interference of these, The experiments are usually carried out with two solutions of the same pH and composition, one of them being a reference solution without the radioelement. The concentration of the radioelement is very low compared with the electrolyte, and therefore the effects of membrane equilibria can be ignored.

Information on the chemical state of the radioelement in solution can be found from the dialysis rate. The diffusion of ions or molecules through the membrane will be rather rapid. If there are true colloids present, the concentration of the nuclide will remain higher in the starting solution due to slower diffusion rate.

Pseudocolloids formed by irreversible adsorption behave similarly to true colloids. If, however, the adsorption is partly reversible, the element can desorb during the dialysis and thus change the ratio between the colloidal and other states of the radioelement⁸. This principle has been used to determine whether there are colloidal particles in radionuclide solutions at low concentrations or not^{14,15}.

6. REFERENCES

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Table 1. A comparison of different sizing techniques

Method	Advantages	Limitations
Electron microscopy	<ol style="list-style-type: none"> 1. Resolution 0.2-0.3 nm. 2. Micrographs can be scanned with image-analysis computer to measure size distribution. 3. 3-dimensional: images can be obtained by shadowing. 4. Particle identification by x-ray fluorescence analysis. 	<ol style="list-style-type: none"> 1. Larger particles may be subject to heating by the electron beam and may change or sublime off. 2. Difficulty in analyzing preparations containing stabilizer and contaminants. 3. Difficulty in identifying exact chemical nature of components in polydisperse system without energy-dispersive analysis of x-rays.
Light microscopy	<ol style="list-style-type: none"> 1. Particle size greater than 200 nm. 2. Ease of sample preparation. 	<ol style="list-style-type: none"> 1. Visibility limit 120 nm. A halo effect occurs as the particle size approaches the wavelength of the incident light. 2. Inaccurate particle sizing below 2000 nm.
Coulter counter	<ol style="list-style-type: none"> 1. Particle size greater than 400 nm. 2. Useful for sizing spherical particles. 3. Fast technique. 	<ol style="list-style-type: none"> 1. Difficult to differentiate particles below 400 nm from background electrical noise. 2. Measures particle volume, not shape. 3. Colloids that are good conductors may cause increases in currents and present false results. 4. Requires preliminary calibration of equipment. 5. Requires dilute solution so that particles pass through the orifice one at a time.
Ultrafiltration		
(a) Nucleopore filter	<ol style="list-style-type: none"> 1. Practical particle size greater than 100 nm. 2. Membrane surfaces are not charged. 	<ol style="list-style-type: none"> 1. Does not measure particle shape. 2. Filtration through filters less than 100 nm is difficult.
(b) Millipore filter	<ol style="list-style-type: none"> 1. Practical particle size greater than 80 nm. 	<ol style="list-style-type: none"> 1. Charged surfaces can cause particle retention. 2. Does not measure particle shape. 3. Filtration through filters less than 100 nm is difficult.
Ultracentrifugation	<ol style="list-style-type: none"> 1. Particle size limit less than 100 nm. 	<ol style="list-style-type: none"> 1. Preliminary calibration of apparatus required. 2. Factors to be considered in evaluating results: <ol style="list-style-type: none"> (a) limiting size for Stokes' law (b) centrifuge tube shape (c) particle shape and weight (d) colloid stability (e) wall effects (f) initial particle movement (g) experimental errors caused by decantation, speed of rotation measurement, acceleration and imbalance. 3. With particle less than 100 nm, excessive time is required for determinations and inaccuracies occur due to Brownian motion. 4. Concentration must be kept low to avoid interference between particles. 5. Very time consuming.
Photometric determinations (Nephelometry, Turbidometry, Light scattering)	<ol style="list-style-type: none"> 1. Ease of sample preparation. 2. Particle size limit less than 100 nm. 	<ol style="list-style-type: none"> 1. Most measurements are based on the assumption of sphericity. 2. Factors to be considered are: <ol style="list-style-type: none"> (a) number of components in dispersed phase and medium (polydispersity) (b) all components as to: <ol style="list-style-type: none"> (i) material differences (ii) optical differences (c) aggregation, coagulation, flocculation, micelle formation, polymerization and the reverse phenomena of subdivision. 3. Interaction of physical electro-magnetic radiation with the dispersion may be relatively complex and the complexity of the system will contribute to the number or such interactions.

(26) Grebenshchikova V.I. and Davydov Yu.P.; Radiokhimiya 2, 165(1961).
 (27) Grebenshchikova V.I. and Davydov Yu.P.; Radiokhimiya 3, 155(1961).
 (28) Haissinsky M.N.; Radiokhimiya 11, 479 (1969).
 (29) Davydov Yu.P.; Radiokhimiya 9, 52 (1967).
 (30) Davydov Yu.P.; Radiokhimiya 10, 508 (1968).
 (31) S. I. ...



3.2. Adsorption

Starik suggested simultaneous investigation of the adsorption and abundance of colloidal forms of a radioelement in low concentration as a method for determination of the nature of radiocolloids⁴. The method is based on the assumption that the maximum formation of pseudocolloids should occur in solutions where the radioelement is strongly adsorbed on a suitable adsorbent. If the maximum abundance of the colloidal forms does not coincide with the maximum adsorption, the colloidal forms are predominantly true colloids.

This method has been used in many studies where pseudocolloids would exist^{4,6,16,17}. It appears that freshly prepared solutions are better than aged solutions for the study of the adsorption, since the former more closely resemble the conditions of pseudocolloid formations¹⁶.

It is important to know the nature of colloidal impurities in the solution. Starik suggests that the major source of impurities is the vessel material and he has in particular investigated the adsorption of radionuclides on glass. Starik's conclusion on the existence of true colloids in the pH region where the adsorption is low, has been criticized by some authors. Davydov¹⁶ states that true colloids cannot be distinguished from irreversibly formed pseudocolloids if the adsorption is studied in aged solutions.

Betenekov and co-workers^{18,19} used a mathematical adsorption model and came to the conclusion that the pH value corresponding to maximum adsorption should coincide with the pH value of maximum concentration of true colloids. This would be the pH of minimum solubility of the corresponding hydroxide and also its isoelectric point.

3.3. Filtration and ultrafiltration

Filtration is a direct method to decide if the radionuclide is in a colloidal state or not⁴. The colloid particles with sizes larger than the pore diameter, will be retained. In principle the sizes of the colloidal particles can be determined in this way, but other particles can also be retained by the filters, depending on adsorption and electrostatic phenomena.

True colloids and pseudocolloids behave in the same way if they are larger than the pore diameter and cannot be separated by this method.

Since the colloid particles are so small, membranes with very small pore diameters have to be used. There are some types of filters, for example millipore and nuclepore filters with pore diameters below 100 nm, but filtration through them requires high pressures¹¹.

3.4. Centrifugation

It is characteristic for the colloidal particles that they precipitate under the action of gravity. When centrifuging is used the force of gravity increases and sedimentation velocity also increases.

In an ordinary centrifuge with a rotation speed of 3000 - 6000 rpm a separation of colloid particles with radii of 30 - 40 nm and greater can usually be accomplished. Starik⁴ and others suggest that these particles would largely be pseudocolloids. True colloids are generally smaller and much greater centrifugation rates (≈ 20000 rpm) are necessary for their separation from the solution. Consequently centrifugation can be used to separate true and pseudocolloids.

The adsorption on the vessel walls must be studied simultaneously. This can best be done by carrying out a parallel experiment with the same solution and vessels, but without centrifuging^{4,8}.

The sizes of the particles can be calculated if their approximate density is known and if they are assumed to be spherical.

3.5. Autoradiography

Radiography was originally applied by Chamie^{20,21} for the investigation of radionuclides in solution. If the radionuclide is in ionic state there is a homogenous blackening of the photographic plate, but if the radionuclide forms colloidal particles, there are inhomogenous dark spots on the plates (radiocolloid "stars"). (Fig.4.)

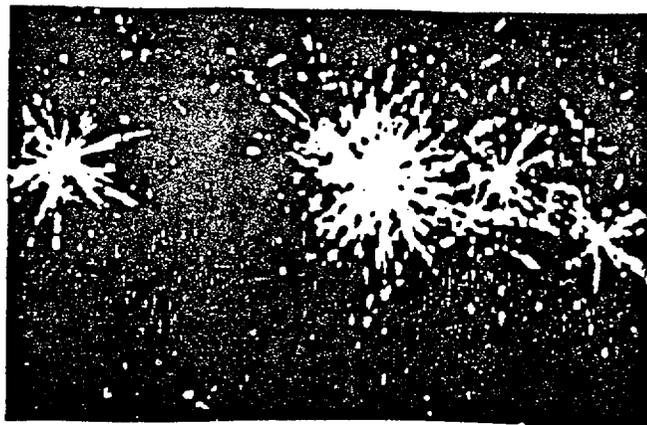


Figure 4. Colloid formation of Po in water

The radiographic method has also been used for differentiating true colloids and pseudocolloids, since the pseudocolloids tend to yield spots with less degree of darkening than true colloids. The size of the spots should be a measure of the dimensions of the colloidal particles⁸.

3.6. Diffusion

Information on the state of radionuclides in microquantities in solution can be obtained from diffusion rate measurements. From the diffusion rate the average size of species or particles in solution can be determined and thus indications of colloidal properties^{4,22-24}.

3.7. Electrophoresis

The mobility of colloidal particles does not necessarily depend on their dimensions. Thus electrophoresis cannot be employed to determine the absolute size of radiocolloids. However, the surface charge and the electrophoretic mobility are important quantities determining the behaviour of colloidal particles and thus electrophoresis is an important method for studies of radiocolloid properties⁸.

3.8. Electronmicroscopy

Electronmicroscopy can differentiate particles down to 0.2-0.3 nm¹¹ and it seems to be a good method to detect and characterize radiocolloids. (Fig.5.) It may be the best method available for determination of the particle size.



Figure 5. Electron micrograph of technetium-99m sulfide colloid

3.9. Quasielastic laser light scattering

Electrophoresis combined with diffusion measurements by quasielastic laser light scattering is a new method to measure the size and charge of radiocolloid particles. The apparatus has been constructed and described by Lim et.al.^{12,13}

This method enables measurements in the same solution of the electrophoretic mobility and the diffusion coefficient of dissolved macromolecules or particles. Both the radius and the charge of the particles can be calculated from the same measurement. This method seems to be very interesting for characterizing radiocolloids.

4. PARAMETERS INFLUENCING THE FORMATION AND PROPERTIES OF RADIOCOLLOIDS

4.1. The pH of the solution

The formation and properties of colloidal hydroxides are directly dependent on the pH of the solution, determining the degree of hydrolysis. At a certain pH value, the solubility product of the hydroxide is reached, and colloidal hydroxide particles can be formed. In alkaline solutions some hydroxides can form anionic hydroxocomplexes, or possibly polymeric products⁶. This could either lead to an increased solubility or an enhanced formation of colloidal anionic aggregates.

The pH value also have an effect on the charge of colloidal or precipitated hydroxides. This charge is generally positive at low pH values, decreases in magnitude with increasing pH, changes sign and again increases. The pH value at which the hydroxide surface is uncharged is called the isoelectric point⁸.

The existance and properties of other true colloids depend on the pH only indirectly.

The formation of pseudocolloids is to a great extent related to pH, due to the pH dependence of the adsorption of the radionuclide on foreign colloidal impurities. Changes in the properties of pseudocolloids can be related to changes in the properties of the colloidal impurities. These can be very different and their nature and properties are often unknown.

In many experimental studies of the behaviour of metals at trace concentration levels a formation of pseudocolloids with material dissolved from the equipment seems to be unavoidable. This pseudocolloid formation would occur in the same pH-region where significant sorption at vessel walls is observed^{6,17,25-27}. This is particularly the case for experiments in glass vessels. In addition, experimental results suggest that the hydrolysis of the radioelement in solution enhances this formation of pseudocolloids.

4.2. The salt medium

It is known from the macrochemistry that the ionic strength of the electrolyte affects the solubility of many substances⁸. By addition of an electrolyte colloid coagulation can occur. When a complex formation between the trace element and the ionic medium occurs similar effects would be expected for all true colloids.

Ichikawa and Sato¹⁰ added some electrolytes to a solution of carrier free europium before centrifuging. The concentration of

the electrolytes was varied. There were no influence of the electrolyte at pH values less than 7, but in alkaline solutions an enhanced coagulation was observed with increasing ionic strength. Fig.6. shows the effects of the electrolytes at pH = 8.0.

The effects of the electrolyte on pseudocolloid formation can be related to both changes in the properties of the carrier particles and changes in the degree of sorption of the radioelement on these particles.

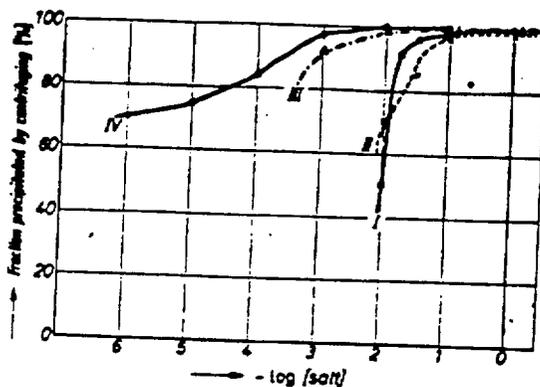


Figure 6. A comparison of the effects of electrolytes on the centrifugal behavior of carrier-free europium. pH:8.0 electrolytes: I: NaCl, II: NaNO₃, III: Na₂SO₄ and IV: BaCl₂

Impurities in the solution will of course affect the formation and properties of pseudocolloids. By proper purification of the initial solutions the amounts of impurities can be decreased. Ichikawa and Sato¹⁰ found that the best method for purifying solutions was centrifuging, but there still are impurities in the solution which come from the equipment, e.g. silicic acid from glass vessels.

4.3. The concentration of the radioactive element

The concentration of the radioelement is one of the determining factors in the formation and properties of radiocolloids. One basic condition for the existence of a separate solid phase in solution is that the solubility product of some compound of the radioelement is exceeded.

The structure and composition of radiocolloids can depend on the concentration of the radioelement and on the amount of impurities in the solution. So the question arises as to whether the solubility

product values obtained for macroconcentrations can be used for trace concentrations and whether the solubility product makes any sense at extreme dilutions. This question is particularly important for hydroxides. Haissinsky²⁸ and other authors²⁹ means that the solubility product determined from experiments with radiocolloids cannot be taken as thermodynamic constants. Davydov⁶ analyzed the variability of the solubility product of trace hydroxides and suggested the following reasons: a) polymeric forms are not in equilibrium with monomers (hydroxocomplexes) because polymerization is an irreversible reaction; b) at various concentrations of a metallic element in solution, and at various pH values precipitates of different compositions may be formed. Other factors are precipitate ageing and effect of impurities.

Moreover, in the presence of solid sorbents a precipitation process may occur on the surface, possibly since the properties of the solution in the interphase are different from the properties in the bulk (giving a decreased solubility product in the surface layer).

Starik and co-workers have suggested that the the solubility product corresponds to the beginning of the polymerization of the hydrolysis products. If the solubility product is not reached then a separate solid phase of the radioelement in solution cannot be formed, which means that any colloidal particles produced would be of a pseudocolloidal nature. If the solubility product is exceeded, then true colloids can be formed, although this cannot be regarded as a proof of the existence of true colloids^{4,6}.

4.4. Time and temperature

The formation of true colloids is probably a relatively rapid process⁸. However there are time effects affecting the formation and in particular the properties of radiocolloids. Spontaneous processes leading to changes in the amount and properties of existing colloids are often termed "ageing". These processes may take more time than the colloid formation itself and will not have any well defined starting point.

The rate of pseudocolloid formation is basically determined by the sorption rate on colloidal impurities. There is a lot of data available on pseudocolloidal formation rate. An equilibrium distribution of tetravalent plutonium between colloidal impurities, the vessel surface and the solution is claimed to be attained within 3-5 hours⁶.

Examples of ageing processes are recrystallization of microcrystals and coagulation of colloids. A slowly increasing concentration of pseudocolloids corresponding to an increasing amount of impurities in the system (e.g. from the vessel walls) is sometimes also considered as an ageing process⁴.

The temperature has a significant effect on the dispersity of some radiocolloids⁹. Cooling neutral solutions of chromium, iron, zirconium and niobium leads to an increase in the dimensions of radiocolloids of these elements.

5. SUMMARY OF RADIOCOLLOID PROPERTIES AND STUDIES

A few reviews on the colloidal behaviour of radionuclides in extremely low concentration in solution are available. Starik gives a detailed discussion of the colloidal behaviour of individual radioelements in his monograph⁴. Kepák gives a review of results obtained up to 1969⁷. The latest review is given by Benes and Majer in their book "Trace Chemistry of Aqueous Solutions"⁸, based on data published up to 1975. By this review the pH influence on the formation and nature of radiocolloids is recognized and emphasized, because it is the most important single factor influencing the colloid formation.

Data for actinides and some other elements of interest, largely from ref.8, are presented in table 2. The second column gives the concentration of the radionuclide. The note c.f. (carrier free) means that the radionuclide with the highest available specific radioactivity was used without any addition of its stable isotope. The fourth column shows the pH region where more than 10% of the radioelement is present in colloidal form. In the fifth column are listed the experimental methods used for the analysis.

The following abbreviations were used:

TC - true colloids
PSC - pseudocolloids
NSC - presence of non-specified colloids proved
A - autoradiography
AD - adsorption
CF - centrifugation
DF - diffusion
DIA - dialysis
EF - electrophoresis
UF - ultrafiltration
ED - electrodialysis
R - radiochemical method

Table 2

Trace element	Concentration (M)	Composition and age of the solution	Existence and nature of trace colloids	Experimental methods	References
^{241}Am	10^{-9}	$\text{HNO}_3\text{-KOH}$	pH 4.5-11	UF	4, 25
^{241}Am	10^{-9}	$\text{HNO}_3\text{-KOH}$	pH 5-8 PSC		
^{134}Cs	$\sim 10^{-7}$	$\text{HCl-NH}_4\text{OH}$	pH 9-12 TC(?) No evidence of colloids at pH 0-10	CF, EF	4, 27
^{137}Cs	c.f.	0.5-1 M NaNO_3 , 0.005-1 M sodium tetraphenylborate	TC of Cs tetraphenylborate	CF R	31 32
^{155}Eu	10^{-9}	$\text{HCl, NaOH, 0.001-1 M NaCl}$	pH 4-11 NSC	CF	10
^{155}Eu , Eu stable	10^{-8}	$\text{NaNO}_3, \text{Na}_2\text{SO}_4, \text{ or BaCl}_2$ HCl or NaOH	pH 5-11 NSC, probably TC at pH ≥ 7	CF	10
^{155}Eu , Eu stable	10^{-6}	HCl or NaOH	pH 5.5-11 NSC, probably TC at pH ≥ 7	CF	10
Eu	$1.4 \cdot 10^{-7}$	$\text{HNO}_3 \text{ or } \text{NH}_4\text{OH (NaOH)-}$ $\text{CO}_2\text{-free}$	pH 5.8-11.5 PSC	UF, EF, AD	33
^{155}Eu	c.f.	$\text{HCl, NaOH, 0.01 M or}$ 0.1 M NaCl	pH 12-13 TC pH 5-11 NSC	DF	34
^{239}Np	c.f.	0.0001-0.021 M HNO_3 , $8 \cdot 10^{-6}$ - $8 \cdot 10^{-5}$ M La	No evidence of colloids	CF(860g)	35
^{233}Pa	10^{-11} - 10^{-13}	0-14 M $\text{HNO}_3, \text{NH}_4\text{OH or}$ KOH	pH 2-7, 11-14 PSC	AD, UF, EF, E	4, 6, 36, 44
^{233}Pa	$7 \cdot 10^{-1}$	0.015-0.5 M H_2SO_4 , $\text{NH}_4\text{OH } T=0-13 \text{ d}$	pH 5-11 TC pH 1.5-5 TC (hydrolytic polymers)	CF(25000g)	37

231 _{Pa}	10 ⁻⁶	0.015-0.15 M H ₂ SO ₄ T=0-13 d	0.015-0.05 M H ₂ SO ₄ TC (hydrolytic polymers)	CF(25000g)	37
212,210 _{Pb}	c.f. (~10 ⁻¹¹)	HCl-NH ₄ OH or KOH	≥10 ⁻³ M OH ⁻ NSC	DIA, DF, ED, E	2
212 _{Pb}	c.f.	HCl, satd. H ₂ S	pH 2-6 TC (Pbs)	AD	4
238 _{Pu}	10 ⁻⁹	0.0001-.021 M HNO ₃ , 8·10 ⁻⁶ -8·10 ⁻⁵ M La, various electrolytes	pH 2-4 PSC	CF, AD	35
Pu(IV)	7·10 ⁻⁸	HNO ₃ -KOH T=0-10 d	pH 2.8-7.5 PSC, pH 7.5-12 TC	UF, CF, AD, EF	26, 27
Pu(IV)	(1-2)·10 ⁻⁵	HNO ₃ -KOH	pH 1.4-12 TC	EF, UF, CF, AD	26, 38
239 _{Pu(IV)}	5·10 ⁻¹⁰ -10 ⁻⁸	HNO ₃ -NH ₄ OH T=0-3 d	pH > 2.8-3.6 TC(?)	AD	39
238,239 _{Pu(IV)}	10 ⁻¹⁰ -10 ⁻⁸	HNO ₃ -NH ₄ OH T=24 h	pH 2.5-6 PSC, pH > 3 TC(?)	CF, EF, AD	40
238,239 _{Pu(IV)}	5·10 ⁻⁷ -10 ⁻⁶	HNO ₃ -NH ₄ OH T=24 h	pH 3-6 PSC, pH 5.3-9(10) TC	CF, EF, AD	40
238 _{Pu(IV)}	~10 ⁻¹¹	HNO ₃ -NaOH, I=0.01	pH 2-5 PSC	AD, CF, A	41
234 _{Th}	2.4·10 ⁻⁸	HClO ₄ -NaOH, various electrolytes	Probably TC at pH > 4	AD	42
228,234 _{Th}	c.f.	HCl-NH ₄ OH	pH 5-9.5 TC 0.03-1 M NH ₄ OH TC + PSC	UF, CF	4
233 _{U(VI)}	10 ⁻⁸	HCl-Na ₂ CO ₃	pH 2-8 PSC	EF, AD, UF	2
233 _{U(VI)}	4·10 ⁻⁷	HCl-NH ₄ OH	pH 5-11 PSC	UF, AD, EF	43
233 _{U(VI)}	10 ⁻⁶ -10 ⁻⁵	HCl(HClO ₄)-NH ₄ OH	pH 3-9 PSC(?), pH > 9 TC	UF, AD, CF, EF	16

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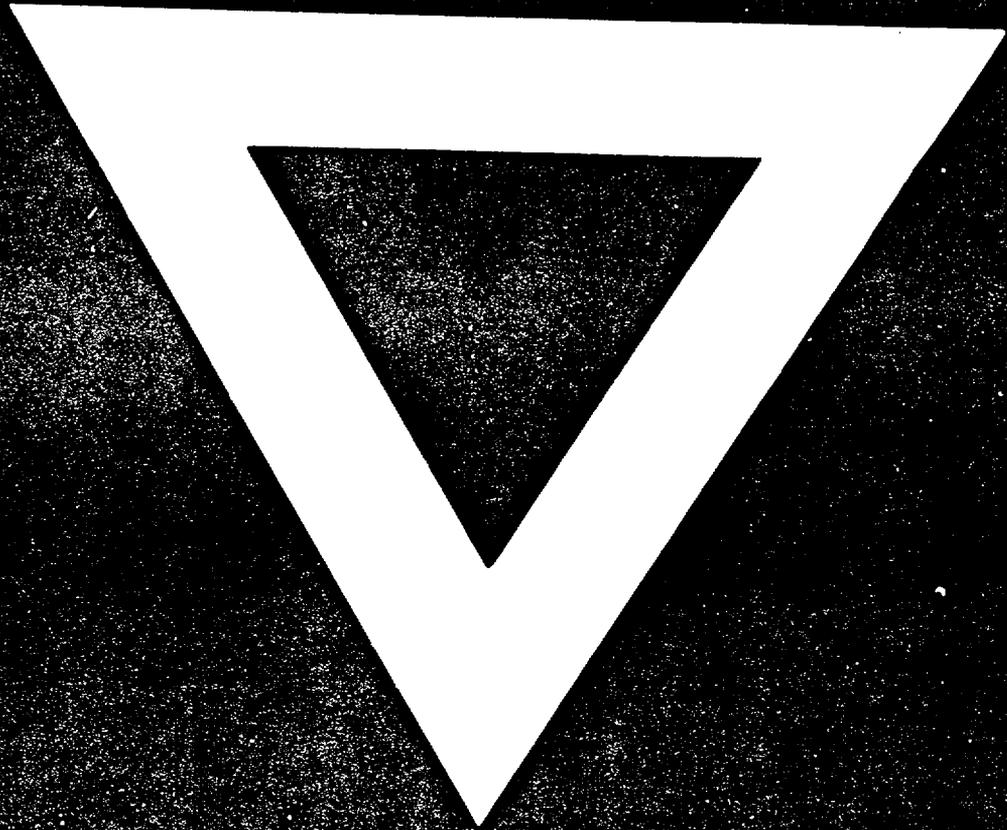
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FIGURE 12. LOG-SCALE CONCENTRATION PROFILES FOR ⁹⁹Tc ALONG A MODERATE MIGRATION PATH AT 20,000 YRS, RECHARGE = 0.5 MM/YR, DISPERSIVITY = 1M, 0.1M.

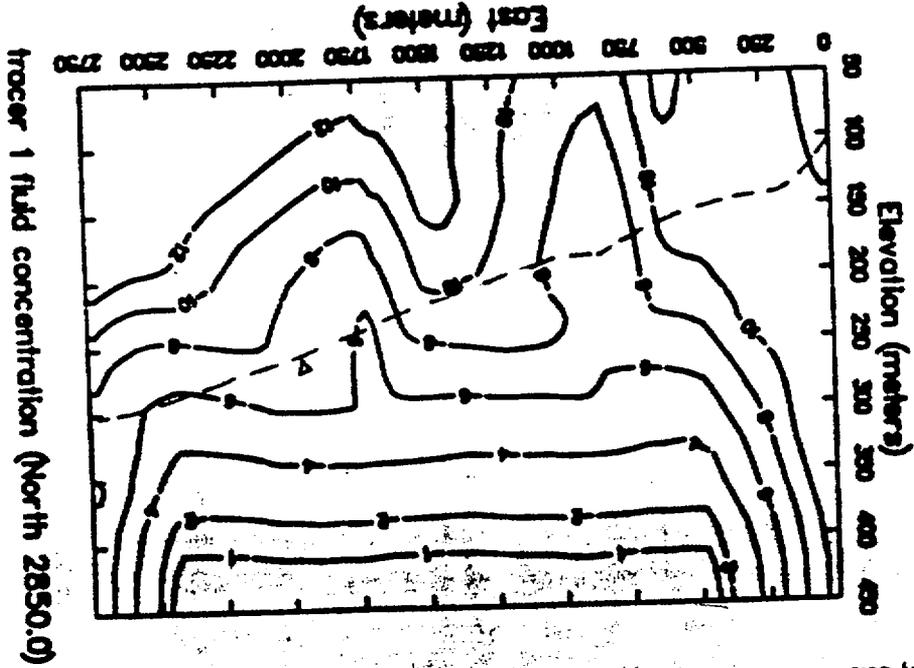
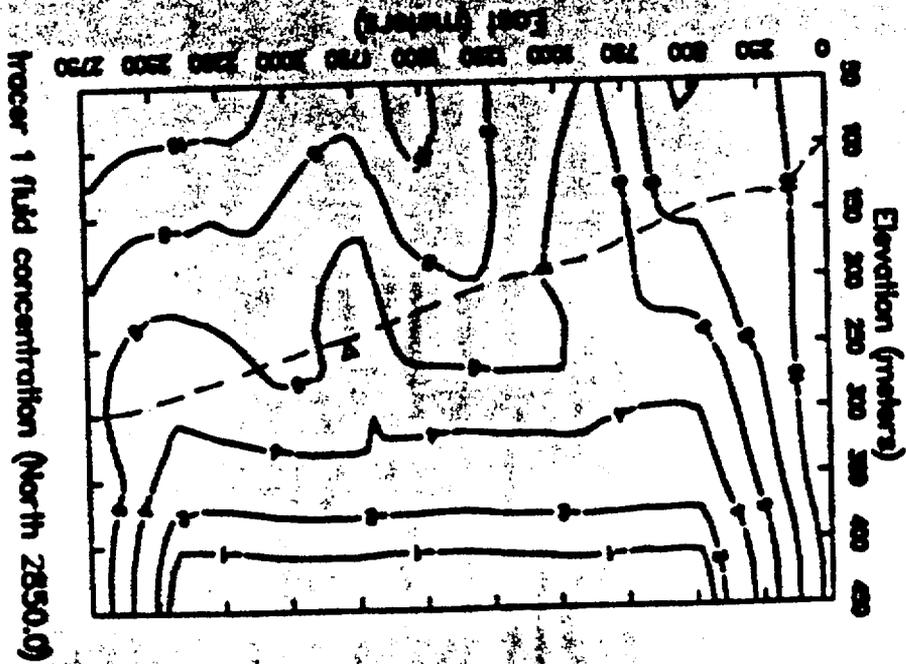
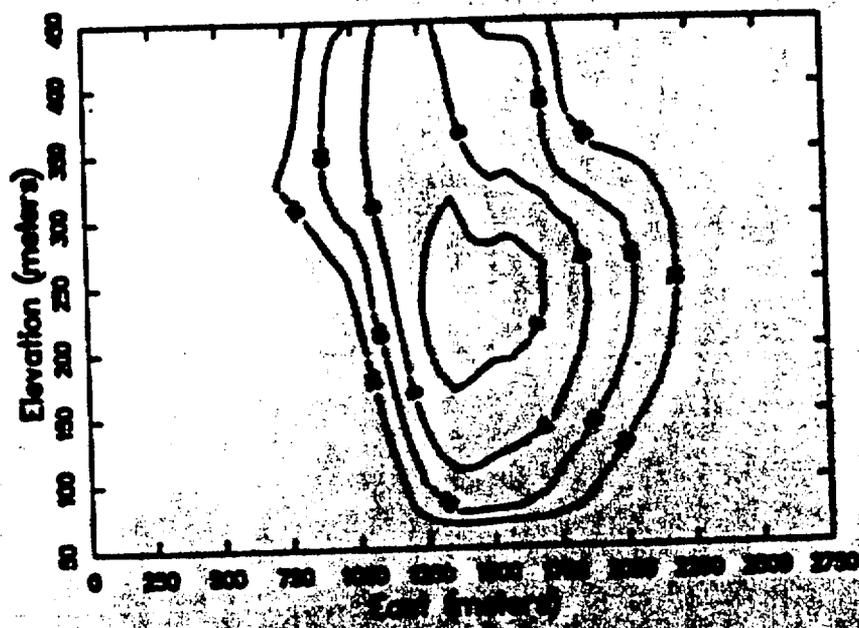


FIGURE 11. LOG-SCALE CONCENTRATION PROFILES FOR ⁹⁹Tc ALONG A MODERATE MIGRATION PATH AT 20,000 YRS, RECHARGE = 0.5 MM/YR, DISPERSIVITY = 10M, 1M.

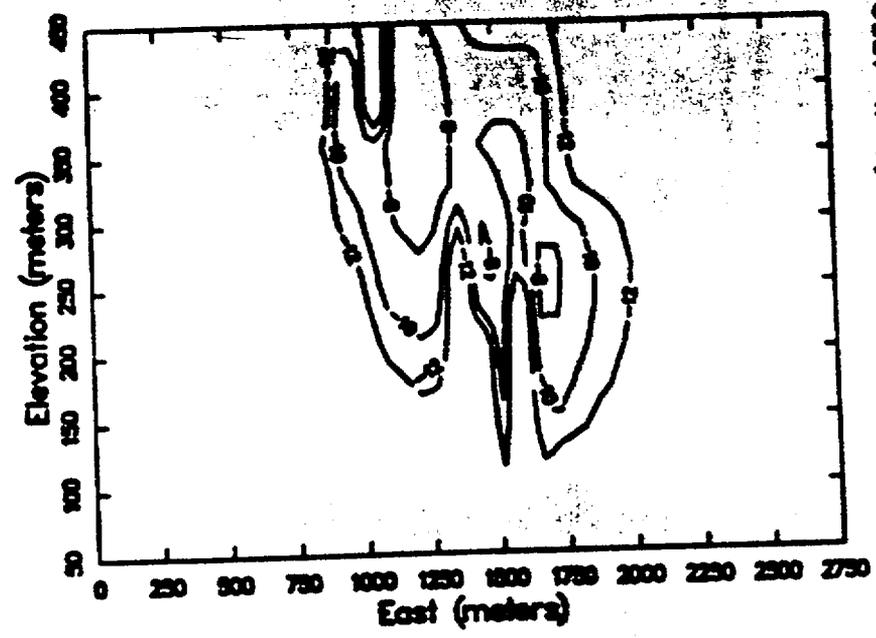


8 6 0 2 0 0 2 7



tracer 1 fluid concentration (North 4350.0)

FIGURE 13. LOG-SCALE CONCENTRATION PROFILES FOR ^{99}Tc AT 5,000 YRS USING THE IMPLICIT SOLUTION FOR THE TRANSPORT EQUATION, RECHARGE = 4.5 MM/YR.



tracer 1 fluid concentration (North 4350.0)

FIGURE 14. LOG-SCALE CONCENTRATION PROFILES FOR ^{99}Tc AT 5,000 YRS USING THE EXPLICIT SOLUTION FOR THE TRANSPORT EQUATION, RECHARGE = 4.5 MM/YR.

8 6 0 2 0 0 2 8

8 5 0 2 0 0 2 9

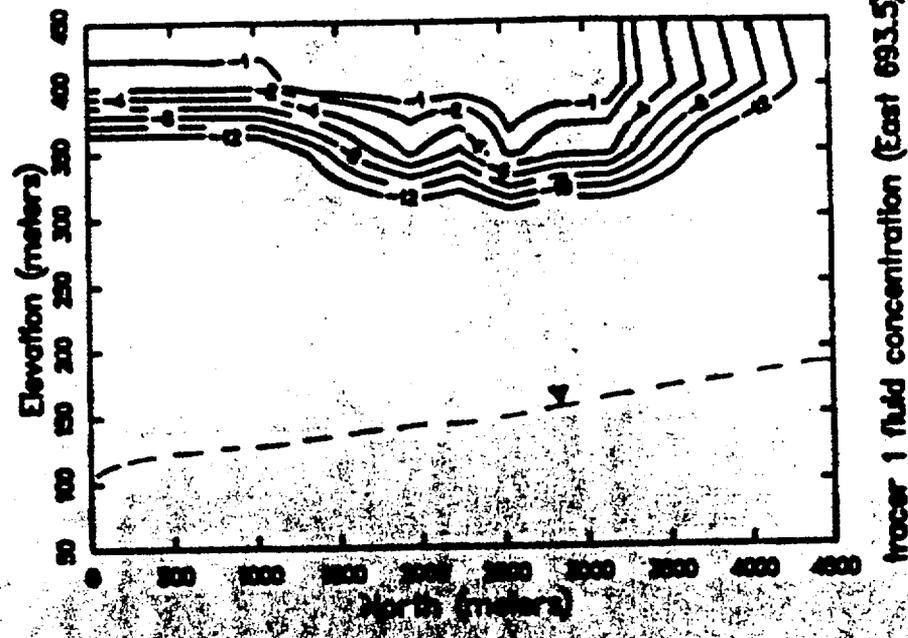


FIGURE 15. LOG-SCALE CONCENTRATION PROFILES FOR ^{138}C , ALONG A FAST MIGRATION PATH AT 20,000 YRS, RECHARGE = 4.5 MM/YR, DISPERSIVITY = 10M, 1M.

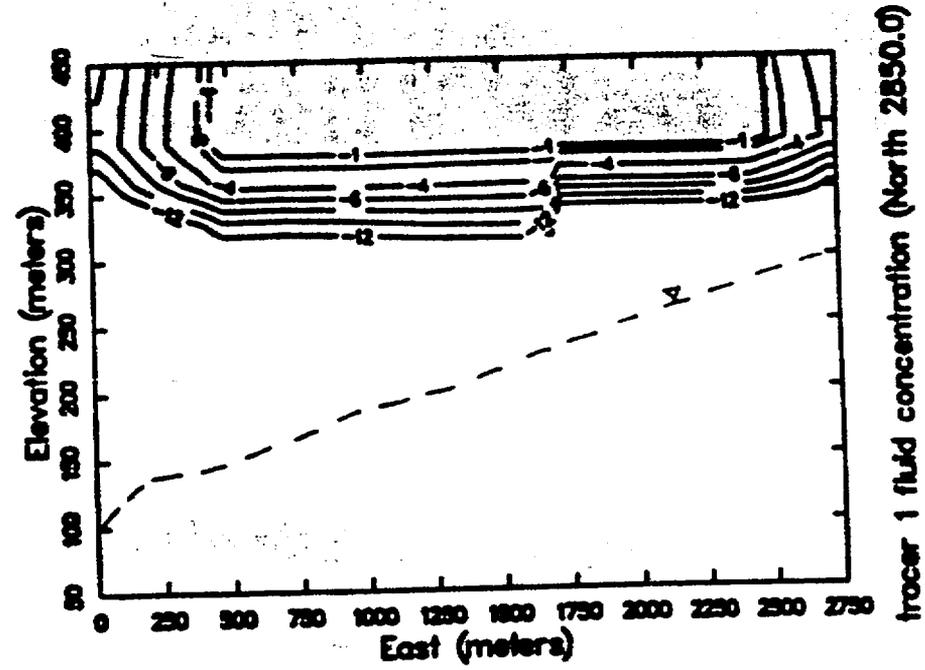


FIGURE 16. LOG-SCALE CONCENTRATION PROFILES FOR ^{138}C , ALONG A MODERATE MIGRATION PATH AT 20,000 YRS, RECHARGE = 4.5 MM/YR, DISPERSIVITY = 10M, 1M.

tracer 1 solid concentration (East 693.5)

8 6 0 2 0 0 3 0

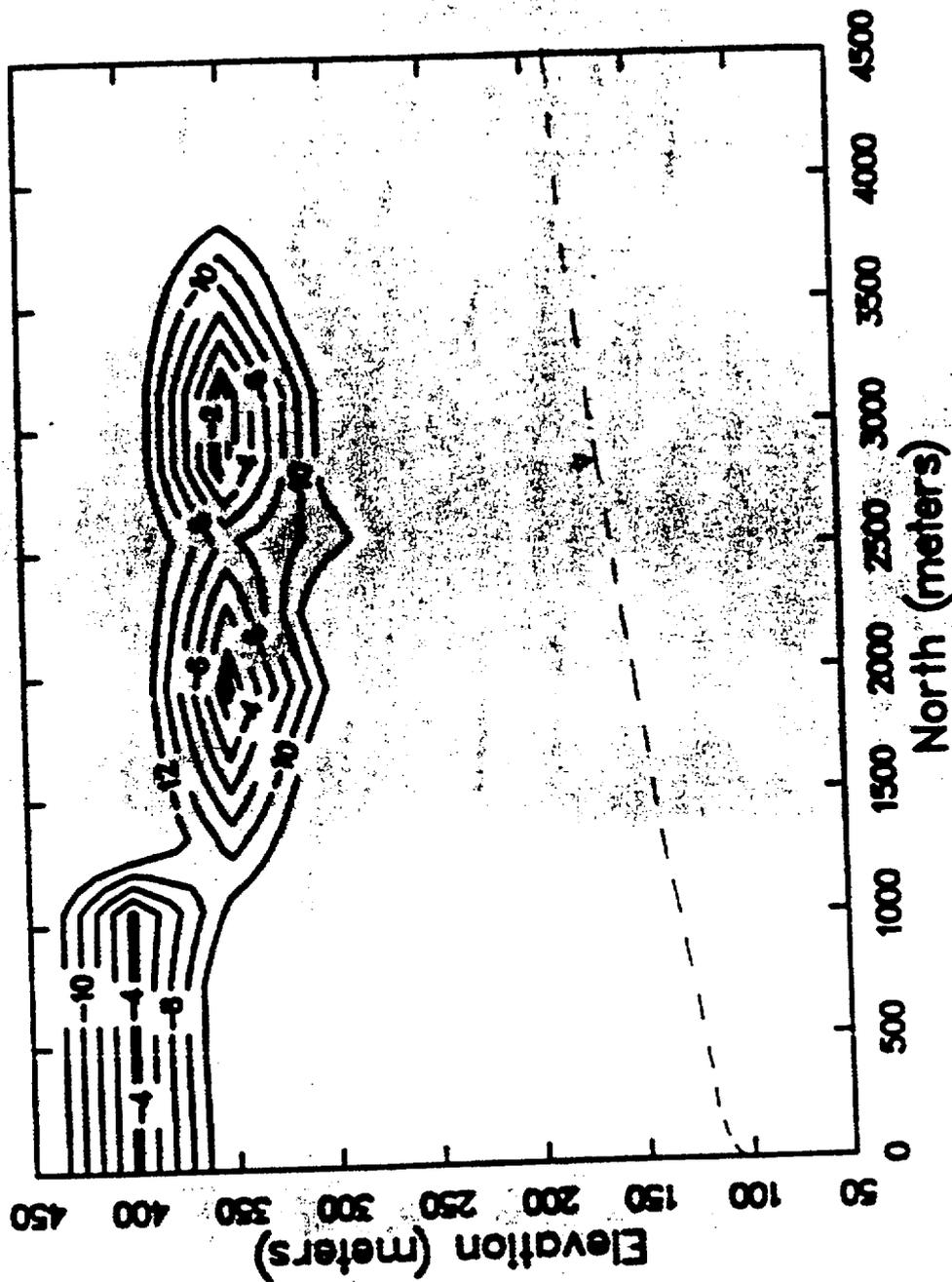


FIGURE 17. LOG-SCALE SOLID CONCENTRATION PROFILES FOR ^{134}Cs ALONG A FAST MIGRATION PATH AT 20,000 YRS, RECHARGE = 4.5 MM/YR, DISPERSIVITY = 10M, 1M. CORRESPONDS TO LIQUID CONCENTRATION, FIG. 15.