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Iodine in soil

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Abstract

A literature study of the migration and the appearance of iodine isotopes in the biosphere particularly in soil is presented. Some important papers in the field of iodine appearance in soil and the appearance of ^{129}I in the surroundings of reprocessing plants are discussed.

The most important conclusions are:

1. Iodine binds to organic matter in the soil and also to some oxides of aluminium and iron.

2. If the iodine is not bound to the soil a large fraction of added ^{129}I is volatilised after a rather short period.

3. The binding and also the volatilisation seems to be due to biological activity in the soil. It may take place within living microorganisms or by external enzymes excreted from microorganisms.

4. Due to variations in the composition of soil there may be a large variation in the distribution of ^{129}I in the vertical profile of soil – usually most of the ^{129}I in the upper layer – which also results in large variations in the ^{129}I uptake to plants.

Contents

1 Introduction

There is only one stable isotope of iodine, ^{127}I, but more than 20 radioactive isotopes. Most of them have short or even very short half-lives and therefore the risk of exposure of man after a release of radio nuclides from a nuclear accident is concentrated to the first weeks after the release. ^{131}I for example has 8 days half-life. The only iodine isotope with long half-life is ^{129}I which has a half-life of 15.7 million years. ^{129}I is also the only radioactive iodine isotope which is found as a measurable natural activity concentration. ^{129}I is produced by fission of uranium and thorium or by spallation reactions of xenon in the upper atmosphere or by neutron-induced reactions on tellurium. These production of ^{129}I would produce a steady state value of the ^{129}I/^{127}I ratio of about $3x10^{-14}$ in the hydrosphere, atmosphere and the biosphere /Edwards, 1962/. Natural iodine contain 99.99% ^{127}I and 0.01% ^{129}I. Most of the ^{129}I which can be found on the earth has been produced by atmospheric nuclear weapons testing or coming from nuclear reprocessing plants. The weapon testing has produced nearly 1 TBq of ^{129}I. It is also possible to detect ^{125}I, which has 60 days half-life, on many places but there are always possible to derive such activity to some sources for example hospitals or laboratories.

^{129}I disintegrates with a complex decay with emission of beta, gamma and roentgen radiation. The probability that the gamma energy is producing conversions electrons is high, resulting in emission of around 10 Auger and conversion electrons per decay. Due to the long half-life of ^{129}I the activity concentration will always be rather low and often supposed to give non significant radiation doses. It could be true if ^{129}I produced only sparse ionising radiation but due to the cascade of Auger electrons there will be dense ionising radiation close to the decay. Furthermore all iodine isotopes in a human body will, during a certain period, be located in close vicinity, a few nm, of DNA, the critical biomolecule for radiation and thus being a serious genotoxic agents. In the thyroid gland iodine is used for producing thyroid hormones which during a short period will be bond to a nuclear receptor which is bond to DNA in thyroid hormone responsive cells /Ludwikow, 1992/. The energy deposition in DNA could therefore locally be high and produce a high LET effect.

Before the use of nuclear energy the ^{129}I/^{127}I ratio was between $I0^{-14}$ and 10^{-15}. In AgCl findings in Australia, Srinivasan found a ratio of between 2.3 and 3.3 $x10^{-15}$ /Srinivasan et al, 1971/. It probably show the ratio which was on the earth for millions of years ago. In sea water collected before the nuclear age the ratio was found to be $2x10^{-12}$ /Kohman and Edwards, 1966/. The highest ratio found before the nuclear age was $4x10^{-11}$ which Brauer et al /1974/ found in thyroid glands from persons who died before 1936. This ratio has then changed and is now in reference areas around 10^{-7} to 10^{-9} /cf. Handl et al, 1990/. In a control area in Missouri, USA the ratio was for example in human thyroid glands $2.3x10^{-9}$, in thyroids from cows $5.9x10^{-9}$ and in thyroids from deer $1.8xl0^{-8}$ /Ballad et al, 1978/. Close to reprocessing plants up to 10^{-3} has been observed in thyroids /Magno et al, 1972/.

There has also been a discussion if stable iodine can have a negative effect on various organisms. Man as all mammals has a strict control on the levels of iodine in the body, at least when we increase the intake above the normal level. Such control has not been developed in some lower organism groups. For example there have been reported that

Daphnia magna populations is affected by high iodine levels sure /cf. Sheppard and Evenden, 1995/. Detrimental effects could be observed at 5 mg iodine per kg soil. Akagare reclamation disease of plants is a situation where naturally occurring stable iodine is released from the soil to the extent that it is chemically toxic for rice plants and may occur after a soil is flooded for rice production /Sheppard and Motycka, 1997/. Microbial population in iodine rich bog groundwater showed that the native wetland microbial population was sensitive to iodine concentrations in excess of 200 mg per litre and anaerobic microbes showed a threshold of 100 mg per litre /Sheppard and Hawkins, 1995/. If this could have any importance for populations living close to a nuclear waste site is not possible to know for sure /cf. Sheppard and Evenden, 1995/. But it could not be excluded that some species or populations of some species have been adapted to very low levels of iodine and then becoming sensitive for elevated levels. However, there is only a speculation which possibly should be investigated more properly.

The chemistry of iodine is very complex with many oxidation levels; -1, 0, +1, +3 and +5. Iodide (I^-) and iodate (IO_3^-) are the most important inorganic ions of iodine which can be found in the biosphere. Beside the inorganic iodine compounds there are also many organic iodine compounds and some of them have been synthesised by biological activity. Methyl iodide is one example on an organic iodine compound found in the nature. Radioactive iodine, mainly ^{125}I and ^{131}I, are used in many cellular biological laboratories to label biomolecules since iodine easily react with many biomolecules particularly those with unsaturated bonds or ring structure. The iodine atom radius is rather big – 2.20 Å – and can in many biomolecules replace a methyl group. One example is iododeoxyuridine where the methyl group in thymidine has been replaced by an iodine but still can be incorporated into DNA in our cells.

It is important to have a good knowledge about behaviour of radioactive iodine in various ecosystems both in the short period due to possible spread of ^{131}I but also in a very long perspective due to ^{129}I. This report is a summary of some important publications (in refereed journals) dealing with iodine and the appearance if iodine in the terrestrial ecosystems. Particularly important in the selection of publications has been studies of ^{129}I in close vicinity of reprocessing plants where release of substantial and controlled amounts of ^{129}I has occurred. I have also tried to find the reasons and the mechanisms for the distribution of ^{129}I which have been observed.

2 Natural occurrence of iodine

The concentrations of iodine varies very much between different media and also from place to place on the earth. In Table 2-1 the typical concentrations of iodine in various components of the environment are presented /Whitehead, 1984/. Normal levels of iodine are within the range from 0.3 ppm till 10 ppm. Coughtrey et al /1983/ suggested that the average concentration of about 0.5 ppm could be considered representative for most parent materials, other than coal and shales which may contain 2 to 5 ppm. In soils a concentration of 5 ppm has been adopted, however, it seems likely that this concentration is more applicable to organic fractions of soils than to the mineral fractions, which approach the concentrations associated with parent material /Cougthrey et al, 1983/.

Table 2-1. Typical concentrations of iodine in various components of the environment (on a dry weight basis for the solid material). /Whitehead, 1984/

	Iodine concentration	
Igneous rocks	0.08–0.50	mg per kg
Sedimentary rocks	0.2–10.0	mg per kg
Marine sediments	3–400	mg per kg
Soils	0.5–20	mg per kg
Seawater	45–60	µg per l
Rainwater	0.5–5.0	µg per l
River and lake water	0.5–20	µg per l
Atmosphere	0.01–0.02	ng per l
Higher plants	0.05–5	mg per kg
Marine algae	90–2500	mg per kg
Mammalian tissue	0.05–0.5	mg per kg
Marine fish (soft tissue)	0.5–6	mg per kg
Freshwater fish	0.06–0.2	mg per kg
Coal	1–15	mg per kg

Iodine is a rather rare element (Table 2-2) although iodine can be found a nearly all media on the earth. Iodine is an essential element for mammals and it is an important part of the thyroid hormones. Among plants iodine seems not to be an essential element.

Table 2-2. Some estimates relating to the global distribution of iodine /Whitehead, 1984/.

	Mass (kg)
Earth’s crust	$3.4x10^{15}$
Sedimentary rock	$2.9x10^{15}$
Hydrosphere	$7.0x10^{13}$
Atmosphere	$4.0xl0^{7}$
Biosphere	$9.0x10^{8}$
Annual transfer in rainfall	$4.2x10^{8}$

The most important source for iodine in the atmosphere seems to be sea water which contains about 50 µg iodine per litre which is 10 till 100 times higher concentration than in rain water /cf. Whitehead, 1984/. The fact that the ratio of iodine to chlorine in the atmosphere and rainfall about 1000 times greater than the ratio in seawater /Mason and Moore, 1982/ clearly indicates that not only evaporation of sea spray may contribute to atmospheric iodine but other mechanisms are important. Some photochemical oxidation or reaction with atmospheric ozone of iodine to elemental iodine may occur in sea water. Other evidence suggests that the biological production of methyl iodide is a major route of transfer from the sea to the atmosphere /Mason and Moore, 1982; cf. Whitehead, 1984/. There seems thus to be not only evaporation or sea spray containing iodine and chlorine but it also indicate some biological or may be chemical process are involved. Also in rain water and river water the iodine-chlorine ratio is higher than in sea water. In sea water there are also methyl iodide and Rasmussen et al /1982/ calculated that 1.3×10^9 kg methyl iodide might well be transferred from the sea to the atmosphere each year. They also considered that a large proportion of this total would be released from the 10% of the oceanic surface associated with the highest biological productivity, particularly reflected in the growth of marine algae. Even in the atmosphere chemical reactions both oxidation and reduction can affect the methyl iodide and thereby producing iodine with various oxidation levels. In Table 2-3 the distribution of different compounds of iodine in the atmosphere is shown for some places in USA. The process of oxidation and reduction of iodine may thus be important for the distribution of iodine.

Table 2-3. Concentrations (ng per m^{-3}) of gaseous and particulate iodine in the atmosphere /Whitehead, 1984/.

	Organic	Inorganic	Particulate
Arizona, U.S.A.	5	11	1.3
Bermuda	28	17	3.7
Kansas, U.S.A.	12	5	2.9
N. W. Territories, Canada	3	0.4	0.2

Other inputs of iodine to the atmosphere may occur from the land surface by processes including volatilisation from the soil. Combustion of fossil fuels also result in an input since the iodine content in both coal and petroleum are rather high. Whitehead discuss the concentrations of iodine in the atmosphere over sea areas and over land areas. His conclusion is that except for very closed area to the sea relatively similar concentrations of iodine is found over seas and land. Whitehead suggest that the mean concentration of iodine in the atmosphere is 10 to 20 ng per m^2 /Whitehead, 1984/.

An important factor influencing the transfer of iodine from the atmosphere to the land surface is the distribution of the iodine between gaseous and particulate forms and between the various chemical forms of the gaseous forms and the different sizes of the particles. Iodine is removed from the atmosphere by the process of either dry or wet deposition. Particulate iodine and gaseous elemental iodine are readily susceptibility to wet deposition but methyl iodide has a low susceptibility. Other gaseous compounds will be influenced by their chemical reactivity /cf. Whitehead, 1984/. It could be assumed that the closeness to the sea at least in the most common wind direction should determine iodine concentrations in rain water and also the levels in the soil and in the food stuffs produced in the area. The spread of iodine is, however, much more complex for example there is also a release of iodine from soil to the atmosphere. Large areas on the earth have very low concentration of iodine resulting in deficiency of iodine. Most of these area – but not all – are located far away from the sea. The concentration of iodine in rain water in UK has been reported to be 2.2 µg per litre and containing about 45% iodate and 55% iodide. Similar results have been reported by other. Investigation of iodine coming from Chernobyl in rain in Japan showed that about 2/3 vas iodide and 1/3 iodate /Seki et al, 1988/. They found no organic iodine or elementary iodine. In air samples also from Japan Nogochi and Murata /1988/ found, however, 19% particular iodine, 5% elementary iodine, 6% HOI and 70% organic iodine which seems to be rather contradictory what Seki et al reported. Alternatively chemical changes occurred in the rain.

Assuming a precipitation of 800 mm rain per year and a iodine concentration of 2.0 µg per litre there will be a deposition of 16 gram iodine per ha and year. Iodine concentration in the upper 15 cm of soil would increase with 7.1 mg per kg soil during a period of 1000 year /Whitehead, 1984/. Additionally there is a dry deposition and totally there will be 9.6 gram per ha and year /Nielsen, 1981/ or 4.3 mg per kg soil during 1000 year. Totally this will mean addition of 11.4 mg iodine per kg of soil.

The hypothesis that most of the iodine in soil has been coming from the sea has been questioned by B. L. Cohen who think that the iodine in the soil is coming from sedimentary rocks by weathering /Cohen, 1985/. This would mean that iodine in food which mainly is coming from land based production also can be derived from the ground and sedimentary rocks. This hypothesis seems, however, not be accepted by most of the scientists. If most of the iodine is coming from the sea, the deposition of iodine will occur at the surface and the following migration is from the upper layer to lower thus opposite to the possible situation for nuclear fuel repository.

3 Iodine in soil

The input of iodine to soil occurs predominantly through transfer from the atmosphere and to a certain extent from dead parts of plants and animals. Weathering of soil parent material may sometimes make significant contribution. A certain addition of iodine is also coming from fertiliser and some chemical biocides which contain iodine. Whitehead for example thinks that there is practically no addition of iodine due to weathering of soil parent material /Whitehead, 1984/. The iodine content of soil reflects the balance between the input which had occurred and the soil's ability to retain iodine against leaching and volatilisation. Loss of iodine can thus either occur by vertical transport down in deeper layers of the soil or that iodine is released to the atmosphere. In Table 3-1 the levels of iodine in various soils from Great Britain is shown. As can be seen the variation in the concentrations of iodine is rather large – from 0.5 mg per kg to 98.2. The highest levels were found in soil rich in organic content.

Table 3-1. Iodine content of U. K. soils (0–15 cm) derived from nine categories of parent material. /Whitehead, 1984/

Category of parent material	Interval	Mean value
	mg per kg dry soil	
Acid igneous rocks and associated till	4.4–15.7	10.4
Till associated with basic igneous rocks	3.4–16.3	10.9
Slate, shale and associated till	4.4–27.6	9.8
Sand and sandstone	1.7–5.4	3.7
Chalk, limestone	7.9–21.8	13.0
Clay	2.1–8.9	5.2
River, and river terrace, alluvium	0.5–7.1	3.8
Marine and estuarine alluvium	8.8–36.9	19.6
Peat	18.7–98.2	46.8

Iodine can be found in various chemical forms in soil /Fleming, 1980/. There seems more common that iodide is incorporated into silica structures rather than forming discrete iodide minerals although iodide of Cu, Ag and Hg can be found. Iodate can be found in combination with Cu and Ca usually in neutral or basic soils. Iodide can be adsorbed on sesquioxides at least if the pH is below 6.0. Iodide can also be bound to clay or to various organic matter in soils with pH lower that 6.9. At higher pH iodide seems to be exchanged by OH^-. Binding of iodide to organic material in the soil have been reported in several papers and the retention of iodine in soil are often interpreted as binding to these organic components. In soil profiles the highest levels of iodine is often found in the upper layers where also the organic contents is highest. The levels of iodine is often highest in peat soil. There have been suggestions that iodine is reacting with thyrosine, thiols and polyphenols in the organic part of the soil. Distribution of iodine in a soil profile will therefore show rather large variation due to the type of soil. These can be due to the distribution of binding molecules but also due to the fact that iodine can show various oxidation levels depending on for example pH and Eh. In a podsol soil iodine will accumulate in the organic material and in the B horizon where iodine are associated to iron and aluminium oxides. In peat soil the levels of iodine is highest in the organic layers and much lower in the underlying mineral soil. One complicating factor is that the bulk density of peat soil is about 20% of the bulk density of mineral soil. This will mean that if we express the iodine concentration as mg per dm^3 the differences will be much smaller. The chemical species which seems to be most efficient in binding iodine is thus organic matter and oxides of iron and aluminium.

The soil has nearly always higher concentrations of iodine compared with the underlying parent material. According to Fleming /1980/ soil formed from sediments has lower iodine concentration than the soil formed from bed rocks. In parent material the iodine concentrations was 0.521 ppm and in corresponding soil 9.339 ppm. For sedimentary rocks the iodine concentration was 1.545 and in corresponding soil 3.850 ppm. This seems to be opposing the hypothesis suggested by Cohen that the source of iodine is the sedimentary rocks. In a review from 1980 Fleming stated that atmospheric precipitation has already been discussed and its contribution is no longer in doubt. The correlation between the iodine concentrations in soils and the distance to the sea has previously been pointed out as a prove of the role of sea in the balance of iodine. However, there are many exceptions from the “role” which often is depending on differences in the soil capacity to bind iodine. A sandy soil should be expected to hold lower amount of iodine compared to a peat soil and this phenomena can explain that in some cases low iodine concentrations is found rather close to the sea (Table 3-2). Iodine can be transferred from soil to atmosphere both by chemical and biological processes, The chemical processes often gives elementary iodine or HI and the biological processes often produce organic iodine for example methyl iodide. The differences can be high for example Whitehead /1981/ showed the iodide added to a sandy soil already after 30 days had disappeared from the soil but in other soil no loss could be seen at all. The highest amount of iodine loss was observed in soils with pH below 5 and organic content below 3%. Release of organic iodine can occur from wet soils since there are biological processes. Methyl iodine is very little dissolved in water and is nearly not bound to soil components. Release of iodine from podsol type soils have been reported /Oosterveld, 1976; Reiniger, 1977/.

Table 3-2. Iodine in soils from various parts of the world /Fleming, 1980/.

Country or state	Range (ppm)
Argentina	0.14–2.8
Britain	0.06–37
Czechoslovakia	0.2–6.5
France	1.6–16
Germany	0.19–28
Ireland	0.7–83
Japan	0.49–63
New Zealand	0–70
South Africa	0.19–7.5
Spain	0.4–52
USA (Texas)	1.7–11

An important type of binding in soil is electrostatic binding. The colloidal fraction of soil is containing clay and organic material and holds quite large amount of mostly negative charges and therefore we could expect that negative charged iodine ions should not be bound particularly strongly. Instead we could expect repulsion. Still there is binding either unspecific or specific binding to free hydro oxides by iron and aluminium or free surface of aluminium or silicates where oxygen atoms is not fully coordinated with aluminium or silica. Specific binding of negative ions such as iodide or iodate can occur if they can bind strong bonds with aluminium, iron (III) and other cations in hydrooxides and aluminium silicate in the clay. Preparation of iron(III) oxides can bind iodide at pH below 5.5 and aluminium oxides at pH above 5.5. Iodide and iodate can of course also bind to organic components in the soil. Iodine can react spontaneously with thiols and polyphenols which both are relatively common in organic soil. Treatment of the soil with hydro peroxides destroy a large part of the organic substances and also reduce the binding of iodine to the soil. Sorption of iodine to soil is drastically reduced after heating of the soil which indicate that biological processes are involved. Also biological material from a compost has a significant binding of iodine which is dependent on pH in the same way as soil. However, many of these bindings are reversible so it is possible to get a desorption with the following magnification and plant uptake. Desorption seems to increase with pH and being dependent on several anions and also show variation between various soil types. This will probably mean that it is necessary to use site-specific parameters to obtain relevant parameters to be used in modelling.

Migration of iodine in a soil profile shows large variation depending on soil types. Often there is a slow transport downward in the soil. It has for example been observed near reprocessing plants where ^{129}I has been released. The ratio ^{129}I/^{127}I is a sensitive index on the migration in the soil. Brauer and Strebin /1982/ found that the ratio changed from 10^{-5} in the surface to 10^{-8} in the layer 10 cm down. Only small changes occurred within a time period of 8 years. Kantelo et al /1982/ reported a retention of ^{129}I in surface soil close to the Savannah River reprocessing plant. Also close to the Karlsruhe reprocessing plant ^{129}I was found in the surface layer of the soil /Schüttelkopf and Pimpl, 1982/. The most efficient retention seems thus to occur in the upper soil layer possible in the root mat.

The iodine uptake to plants can occur either by root uptake or by other parts of the plants from the atmosphere. The direct interception of atmospheric iodine on leaves or a direct inflow through the stomata seems to be rather efficient. At first this way seems to rather marginal but since iodine both can be bound to soil but also released from soil this interception could have a certain importance. The plants are considered to not need iodine but it has been shown that during the growing of for example barley seeds iodine is transferred to diiodotyrosin which is a compound close to triiodothyronine the more active thyroid hormone.

The fact that plant can take up gaseous iodine mainly by the stomata means that the weather conditions affect the uptake. When the stomata are opened the uptake of iodine is higher than when the stomata is closed. Calculations have shown that 60% of the uptake of elementary iodine is dependent on the stomata and the rest 40% by sorption from the outer part of the plants. Other gaseous iodine compounds like methyl iodide is taken up less. The type of vegetation also affect the uptake. Clover for example take up more iodine that grass.

Although iodine is not an essential element it is taken up rather efficiently by the plant roots. Experiments using aqua cultures show that the uptake of iodine is linearly dependent of the iodine concentrations in the water media at least in the range from 10^{-3} to 10^{-4} M of iodide. At higher concentrations iodide has an inhibitory effect on the root growth. Relatively large variation in the root uptake has been reported and seems to be dependent on the type of soil. Tikhomirov showed that the uptake of ^{125}I to oat plants showed a pronounced variation depending on the soil being highest in sierozem soil and lowest in chernosem soils. He also showed that the uptake is decreasing during a time period of 10 days after deposition. This phenomena has also been shown by Schüttelkopf and Pimpl /1982/. They showed that ^{129}I in milk decreased with a factor of 100 after that the ^{129}I emission from Karlsruhe reprocessing plant had decreased. In soils with high organic content the root uptake of iodine seems to be rather low and the uptake will be rather high in sandy soils or other light soils. The concentration ratio – Bq per kg of plant material divided by Bq per kg soil – varied from 0.001 to 1.5 with the most common ratios between 0.01 and 0.1. A site specific investigation of these parameters will thus be necessary after determination of the site of a repository.

Iodine is en essential element for all mammals and we need a intake of 150 to 200 µg per day. For cows a daily uptake of 0.5 mg iodine is recommended. Milk and other milk products are the most important sources of iodine at least close to the sea. The concentration of iodine in the human body is under homeostatic control. In the thyroid gland iodine is converted to the thyroid hormone, first thyroxine which then is stored extra cellularely in the thyroid gland. After the storage period the thyroid hormone will be distributed to target cells in various organs and converted to triiodothyronine which is involve din regulatory processes at the gene level. This means that triiodoithyronine is bound via a specific receptor to DNA and that iodine is within a few nm from DNA. If ^{129}I disintegrate when triiodothyronine is bound to DNA a large part of the energy released at the decay will be deposited in DNA with a risk of serious DNA damages and possible unrepaired lesions.

Oktay et al /2000/ have studied iodine as well as some other radionuclides in Mississippi River delta sediments and demonstrate that significant amounts of anthropogenic ^{129}I are buried in these sediment with a well-resolved bomb pulse. Close relationship between profiles of ^{129}I/^{127}I ratios and those of ^{240,239}Pu and ^{137}Cs (decay corrected to the time of deposition) strongly suggest that the source for the elevated concentrations is bomb fallout deposited to the drainage basin of the Mississippi River. The peak in iodine ratios was sharp which suggests post-depositional immobility of bomb produced ^{129}I.

4 Whitehead 1973–1984

Whitehead has performed a large investigation resulted in 8 publications where he presented results about various aspects of iodine in soil. In a work from 1973 /Whitehead, 1973a/ he collected soils from twenty-three widely separated sites in the UK and found that the total iodine concentrations ranged from 2.7 to 36.9 mg per kg. The total iodine concentrations were not closely correlated with the distance of the site from the coast or with the rainfall (r = 0.23). When two soils derived from marine alluvial were excluded, a positively correlation were found between total iodine concentrations and aluminium oxide extracted by Tamm's reagent – "free" aluminium – (r = 0.88), with ferric oxide extracted with citrate-dithionite – "free" iron – (r = 0.64) and with soil organic matter (r = 0.59). The correlation between concentrations of iodine and the levels of clay in soil was not so good (r = 0.23) as well as between iodine levels and pH (r = 0.33). The conclusion of the work is that soil iodine is in part associated with sesquioxides and in part with soil organic matter.

In the next paper /Whitehead, 1973b/ he showed that the sorption of iodine was reduced when soil was dried before equilibration with iodine solution. With undried soils, sorption continued for more than 48 hours and the maximum sorption occurred at pH below 5 but a secondary peak occurred at pH 8.5 to 9.0, particularly with soil containing a high level of organic matter. In soil profiles with 10 cm layers the amount of iodine sorbed were closely related to the contents of organic matter in the soils but not to iron and aluminium oxides or of clay. Treatment of surface soils with hydrogen peroxide to destroy organic matter greatly reduced the sorption of iodine at pH of about 5.5 – from about 80% to about 20%. Removing of iron and aluminium oxides with Tamms reagent also resulted in a marked reduction of sorption at pH less than 5. The results indicate that sorption was due in part to soil organic matter and in part to iron and/or aluminium oxides. At pH above 6, organic matter appeared to be the major sorbing constituent but under more acid conditions the oxides appeared to be increasingly important.

The sorption of iodide by several soil component materials was examined over a range of pH values and compared with the sorption by a surface soil. A compost, prepared from grass roots and decomposed for 24 weeks sorbed substantial amounts of iodide from solution and showed changes in sorption due to pH and drying similar to those shown by the soil /Whitehead, 1974a/. Freshly precipitated hydrated ferric oxide also sorbed substantial amounts of iodide from solutions of pH below 5.5 but the amount decreased to zero as the pH approached 7. Freshly precipitated hydrated aluminium oxide could not be assessed at pH below 5 but at pH 5.5 to 7.5 its sorption was similar to that of ferric oxide. No sorption of iodide was detected by kaolinite, montmorillinite, chalk or lime stone.

In /Whitehead, 1974b/ he studied sorption of iodine in the form of iodide, elemental iodine and iodate to a sandy loam or to a mixture of the soil with composted grass roots, chalk and sesquixoides and its solubility determined after various periods of incubation. With iodide, solubility in both 0.01 M $CaCl_2$ and l.0 M NH_4^+ acetate declined rapidly over a period of 0 to 3 days and reached an equilibrium levels of 2.8% solubility in $CaCl_2$ and 7.8% in NH_4 acetate (mean of the results obtained after 48, 103 and 160 days) Five percent replacement of the soil by composted grass roots had no appreciable effect on the solubility of added iodide, while 5% chalk depressed the solubility of iodide in $CaCl_2$ to 1.8% but caused a slight increase in solubility in NH_4 acetate. Incorporation of 2% of ferric oxide or aluminium oxide reduced the solubility in both solutions to 0.1 and 0.3% respectively. Elemental iodine was similar to iodide in its solubility. Iodate, however, differed considerable from the other two forms of iodine. In soil alone and with soil/chalk mixture its decline in solubility with increasing incubation time was relatively slow but after 160 days its solubility was similar to that of iodide and elemental iodine. The incorporation of composted grass roots caused a rapid reduction of iodate solubility suggesting that the organic matter accelerated the reduction of iodate to elemental iodine or iodate.

The uptake of various forms of iodine to perennial ray grass was studied /Whitehead, 1975/. A pot experiment using sandy loam soil into which 20 mg I per litre was added. The uptake of iodine was much greater from iodate than from the other two forms. Replacement of 5% of the soil with manure reduced uptake from all three forms of iodine more than ten-fold. Similar replacement by chalk reduced the uptake from iodide but increased the uptake of iodate.

In a work from 1978, /Whitehead, 1978/ the iodine content in successive 10 cm horizons of eighteen soil profiles from England and Wales was determined and correlated with contents of “free” aluminium and iron oxides and organic matter. In the rendzina soils, the content of iodine was relatively high in the surface 10 cm but declined markedly with depth. In the podsols, iodine was low at the surface but increased in the B horizon. In one of these soils an iron pan of about 1 cm thickness had an iodine content as high as 37.6 mg per kg. In a peat soil the iodine content was relatively high in the horizons above 60 cm but was low in the underlying clay. In soil derived from river alluvium and from lowland clay, the iodine content were usually low and showed little variation with depth in the profile. In all 154 samples from the eighteen sites iodine content correlated with oxalate-soluble aluminium (r= 0.834) but not with oxalate soluble iron (r=O.35) or organic matter (r= 0.37). However, in the five most acidic soils the iodine content was more closely correlated with the iron than with aluminium.

In a work from 1981, /Whitehead, 1981/ he studied the volatilisation from soils and mixtures of soil components after adding potassium iodide. The extent of volatilisation was measured after 30 days exposure in a well ventilated room. With acid sandy podsol 57% of the added iodide was released. Particularly with sand alone having a pH of 5.7 volatilisation amounted to 100%. With many of the surface soils the volatilisation was negligible except for the acid sandy soil. Organic matter reduced volatilisation probably by retaining iodine in bound form. Montmorillonit, kaolinit and ferric oxides also reduced volatilisation in comparison with sand alone but had less effect than did organic matter.

Whitehead has also presented an excellent review about the distribution and transformation of iodine in the environment, /Whitehead, 1984/.

5 Tikhomirov and some other Russian scientists

A number of Soviet works dealing with iodine have been translated to English and generally they show rather similar results as Whitehead and have rather similar conclusions. In one experiment iodine was mixed with humus horizon of a sod-podzolic soil to give a homogenous vertical distribution and then incubated in situ for some months /Aleksandrovskaya, 1979/. At the beginning the iodine concentrations was similar down to 20 cm but after incubation for 2 months the vertical distribution had changed. There was a decrease of iodine in nearly all layers and a heterogeneous distribution of the iodine had developed with most of iodine found in the upper part of the profile. One practical conclusion was that under normal conditions iodine is strongly fixed to peat but when conditions are disturbed peat begins to release iodine into the atmosphere and ground waters. The type of distribution resulting in such release is, however, not mentioned in the work. In the introduction there is a discussion about the spreading of iodine and the importance of the sea. There seems to be two possibilities for spreading of iodine from deeper layers. One possibility is that iodine is spread from oil deposits and another from coal deposits. High iodine concentration in the ground is suggested to be an indication of oil or coal deposits. At burning of coal iodine is released.

Tikhomirov et al have studied the appearance and sorption of iodine in Russian soils by adding ^{131}I or ^{125}I to soil samples and then after various incubation times extract iodine with different methods /Tikhomirov et al, 1980/. Using a sequential extraction with organic solvents, water and ending up with extraction of humic acids in 0.1 M NaOH they found that in sod-podzolic soil, leached chernozem soil and lowmoor peat between 37 and 55% of added ^{131}I was bound to the humic acids fractions. Precipitation of humic acids with strong acids also precipitate 12 to 25% of the ^{131}I activity and in the fulvic acids fraction – not precipitated by strong acids – was between 22 and 34% of the total ^{131}I activity. In the water soluble fraction between 7 and 18% of the ^{131}I activity was found. Besides there was found between 2 and 18% in fractions extracted by organic solvents, alcohol, bensen and carbontetrachlorid. Seven days after the addition of ^{131}I to soil all the iodine in the water extracts becomes incorporated into low-molecular-weight fractions with a molecular weight of less than 5 000. In humic acids a near-equilibrium state becomes established between natural iodine and radioactive iodine within 12 to 18 days.

In another paper, Tikhomirov discuss the two possible uptake mechanisms which could be important for plant uptake of iodine – by roots or by direct deposition on leaves and uptake through the stomatas /Tikhomirov, 1984/. His conclusion is that both these routes are significant but the absolute value of the contribution of each of them varies by no less than 10-fold. In early development of plants is root uptake of iodine the most important mechanism for uptake but at the end of the vegetation period uptake of iodine via air is more important at least for grass and legymenous plants. (Table 5-1)

Table 5-1. Iodine content in dry aboveground biomass and fractions of its entry from soil /Tikhomirov, 1984/.

Soil	Species	Phase of plant development					
		Tillering		Flovering		Maturation	
		$\mu g g^{-1}$	%	$\mu g g^{-1}$	%	$\mu g g^{-1}$	%
Soddy-podzolic	Timothy	0.23	100	0.21	65	0.18	5
	Clover	0.30	100	0.29	85	0.27	50
Soddy-carbonate	Timothy	0.23	100	0.82	40	1.21	4
	Clover	0.28	100	1.05	73	1.17	22

There are quite large regional variation in iodine concentration in the atmosphere and also in the soil. One important factor is according to Tikhomirov the content and qualitative state of the humus, with which the bulk of the iodine in soils is bound. With the passage of time a state of dynamic equilibrium is established at which the entry of iodine into the soil from the air is compensated for by the reverse process and the leaching of the element with surface and ground water. The time for establishment of this equilibrium and the other quantitative characteristics of these processes are according to Tikhomirov unknown. The role of the various groups of humus and other soil factors in the fixation of iodine by the soil and its biological availability to plants remain unclear. The biological availability for plants of iodine in the soil is mainly provided by its water-soluble form, the iodine-humus compounds are among the unavailable forms and function as an iodine depot in the soil.

The uptake of ^{125}I to plants was studied in pot experiments using various soil types Moiseyev et al /1984/. The plant uptake of iodine after 30 days incubation were highest (30%) in the soils with course texture (the alluvial-stratified sandy soil) and in soils that are low in humus. The coefficient of accumulation is highest in the alluvial-stratified soil because of the high content of water-soluble ^{125}I in the soil. The variation could be up to 20 times. The uptake varied also in the various organs of crops studied. The difference could be several orders of magnitude. 1n pot experiments using various soils and cultivating peas and oat the ^{125}I activity was mostly found in the vegetative parts and the concentrations in the seeds of oat and peas was below 10% of that found in straw. Organic addition as well as addition of fertilisers to the pots reduce the uptake of ^{125}I with 40% or even more depending on the soil type. (Table 5-2)

Table 5-2. Influence of fertilisers on the coefficient of ^{125}I accumulation by oat sprouts /Moiseyev et al, 1984/.

	Soiltype		
Variant	Sod-podzolic soil	Yellow earth	Sierozem
Control	0.68	0.89	2.8
+5 g peat	0.24	0.56	0.71
+ l0g peat	0.18	0.62	0.50
+NPK 90 kg/ha	0.33	2.58	4.27
+NPK 135 kg/ha	0.23	1.8	5.31
+ $CaCO_3$	0.51	0.97	–

Perevenzentsev et al have studied the migration of ^{125}I down in soil profiles /Perevezentsev et al, 1984/. In Table 5-3 the distribution of ^{125}I after 73 days diffusion is shown. The data show that most of the iodine (95 to 97%) remained in the upper 0 – 1 cm layer. In the layer 0-1 cm, 8.2% of the iodine content was extracted by water in the leached chernozem and 15% in sod-podzolic soil. The corresponding value for deeper layers, 6–7 cm, were 40% and 38% respectively is extracted. They also cut out various layers and studied how much of the iodine was evaporated to the air. It was shown that in the upper layers only small amounts of iodine was released and in the deeper layers 3 to 4 order of magnitude higher activity of ^{125}I was released to the air.

Table 5-3. Vertical distribution of ^{125}I in soil 73 days after application to the surface (Perevezentsev et al, 1984).

Depth, cm	Relative concentration in soil layer (% of total soil content)	
	Leached chernozem	Sod-podzolic soil
0–1	97.2	95.4
1–2	2.3	3.8
2–3	0.36	0.36
3–4	0.06	0.07
4–5	0.01	0.02
5–6	0.007	0.01

6 Robens and Aumann

Since 1983, Robens and Aumann have conducted a measurement program in the environment of the small Karlsruhe nuclear fuel reprocessing plant (WAK) to determine the levels of ^{129}I and ^{127}I in soil, herbage, vegetables, fruits, grains, milk, eggs, water, rain and air. The objectives of the study was to establish the current levels of ^{129}I and its distribution in the environment of the WAK and to evaluate the environmental transport process of ^{129}I in terrestrial ecosystems and its pathways in the human food chain. The reprocessing plants has been in operation since 1971 and about 121 t of the fuel have been reprocessed. The main source of ^{129}I emission was through the 60 m high exhaust air stack of WAK. Since 1971, about 7.3 x 10^9 Bq have been released to the environment mainly through the exhaust air stack /Schüttelkopf and Pimpl, 1982/. The installation of an iodine filter in 1975 considerably reduced the amount of ^{129}I released the environment and about 90% of the total ^{129}I released was probably emitted prior to installation of the iodine filter. Sampling has occurred about 3 till 4 km from the plants on arable land.

The concentrations of ^{129}I and ^{127}I in soils, food crops and animal products collected in the environment, within 3 to 4 km, of the small Karlsruhe nuclear fuel reprocessing plants were determined by neutron activation analysis /Robens and Auman, 1988a/. Compared to a current average biospheric background value in regions which are remote from sites where ^{129}I is being released of 10^{-9}–10^{-8} it is clearly demonstrated that the levels of ^{129}I as well as the ratio ^{129}I/^{127}I are elevated in soils, food crops and animal products collected in the environment of WAK. The ratio in surface soils was enhanced to about 10^{-6}. The ratio in foods are in the range 10^{-7}–10^{-6} thus several orders of magnitude higher than the background levels.

The potential dose to humans was calculated to adult and children assuming that all food consumed was produced in the near-environment of WAK. Radiation dose and about 1.3x10^{-7} Sv per year for adults and infants were estimated. The most important contributor to ^{129}I intake were wheat and rye flour, leek and hen's egg, for children the most important contributor are vegetables, hen's egg and milk and milk products.

In a second paper /Robens et al, 1988b/ the concentrations of ^{129}I and ^{127}I in soils, forage plants and deer thyroids collected in the environment of WAK were determined by neutron activation analysis. The results presented demonstrate that the levels of ^{129}I and the corresponding ratio ^{129}I/^{127}I in soils and forage plants are several orders of magnitude above current biospheric background values. The results for grass and forage plants indicate a correlation between the ^{129}I and natural ^{127}I contents in plants. These indicate that the two iodine isotopes react similar in the soil. Deer thyroids collected in February in the vicinity of WAK were found to have very high ^{129}I levels and corresponding high ratios – 0.9 x 10^{-5} in roe deer and 1.5 x 10^{-5} in fallow deer. The high ^{129}I content were probably caused by the high ^{129}I contents in tree bark on which deer feed in winter.

In a third paper, /Robens et al, 1988c/ they reported data on soil-to-plant concentration factor for field-grown food crops and forage plants. Information about these concentration factors can thus be found in this publication. The geometric mean of the soil-to-plant concentration factors for ^{129}I in food crops was found to be 3 x 10^{-2} thus rather similar as the default estimates used in most assessment models. A soil-to-plant concentration factor for ^{129}I in forage plants of 6.2 x 10^{-1} was observed. The default values used in ABG and NRC assessment models appears to be too small by about an order of magnitude. Transfer coefficient to animal products of 2 x 10^{-1} (days x litre^{-1}) to cow's milk, 12.8 (days x kg^{-1}) to hen's egg and 1.5 x 10^{-2} (days x kg^{-1}) to pork were determined for ^{129}I. The data for the transfer coefficients to cow's milk and to pork are within the range of the values measured by other authors while the value for hen's egg is higher by a factor of around three than the highest value reported previously in the literature. Tables in which comparison of results obtained by other authors can be found in the publication.

Concentration profiles of ^{129}I and ^{127}I soils from the environment of WAK were measured at sites in the predominant downwind direction between 1 100 m and 18 000 m from the exhaust stack /Robens et al, 1989/. The residence half-times in surface soils was estimated using a compartment model. The residence half-time within the uppermost 5 cm of soil was 9.0 years. In the top 30 cm of soil the residence half-times were found to average 28.4 years. The concentrations of ^{129}I seem to decrease exponentially with increasing depth. Characteristic of the exponential decrease is the half-depth , i.e. the depth at which the concentration is half that at the soil surface. Half-depths of ^{129}I in soil were ranged from 4.2 to 10.9 cm with a mean of 7.6 cm. The small values for the ^{129}I half-depth indicate slow migration of ^{129}I into the soil. The retention of ^{129}I near the soil surface may be a function of soil characteristics and organic matter content. In contrast, ^{127}I clearly indicate that natural ^{127}I shows a uniform distribution with depth and may support the assumption by Cohen /1985/ that most iodine in soil is derived from weathering from iodine in rocks and not, as widely believed from marine iodine. The findings that ^{129}I is found in the upper layer of the soil are in agreement with several other authors.

A field investigation of the transfer of ^{127}I and ^{127}I in the soil-pasture-cow-milk/meat pathway was carried out at a diary farm situated 5 400 m to the north of WAK /Hauschild and Aumann, 1989/. From the concentration of ^{129}I and ^{127}I in soil and pasture vegetation, soil-to-plant concentration factors for ^{129}I and ^{127}I in pasture vegetation could be calculated and the geometric means for the concentration factors for ^{129}I and ^{127}I for pasture vegetation were 8.5 x 10^{-1} and 7 x 10^{-2} (Bq per kg dry grass/Bq per kg dry soil) respectively. For the grazing season the transfer coefficients for ^{129}I and ^{127}I from feed to milk were determined F_m (day per litre) and found to be 1.1 x 10^{-3} (geometric mean) and 2.6 x 10^{-3} respectively. One cow and one calf were slaughtered during the field investigation and the transfer coefficients from feed to beef or veal were determined and found to be in beef 11 x 10^{-3} and 3.9 x 10^{-3} day kg^{-1} for ^{129}I and ^{127}I respectively. In veal the corresponding values were 3 x 10^{-3} and 0.2 x 10^{-3}. In this paper there are also a summary of various transfer factors from their own work and from other authors works.

Comparison of the measurements since 1982 of the ^{129}I in soil, food crops and forage plants was compared with the predictions of the radiological assessment model used in the Federal Republic of Germany /Robens-Palavinskas et al, 1989/. The model prediction of the I concentrations seems to be in reasonable agreement with the measured ^{129}I concentrations in soil, at least to a distance of about 10 km from source. The predicted ^{129}I concentrations in food crops were overestimated by the model, in some cases considerable. The ^{129}I concentrations in meadow grass and pasture grass were underestimated by the model by factors of between 1.2 and 11. The results indicate that some improvement in the model were needed to reduce the uncertainties.

Gaseous and particulate iodine samples were collected simultaneously from the atmosphere around WAK /Wershofen and Aumannn, 1989/. Iodine were separated into three fractions; (a) associated with aerosol particles, (b) gaseous inorganic and (c) gaseous organic. In 1986, the aerosol fraction range between 5.2 to 30.8% for ^{129}I and 12.9 to 28.0% for ^{127}I. The gaseous inorganic fraction varied between 26.8 and 35.2%. The gaseous organic fraction of ^{129}I accounted for 34.0 – 65.9%. The found values were compared with values obtained by models /Wershofen et al, 1991/.

In a field experiment the small amount of fallout ^{129}I and natural ^{127}I were measured in soil and vegetation of permanent pasture in a region remote from the sites where ^{129}I is being released /Deitermann et al, 1989/. The data for the soil-to-plant transfer factor for ^{129}I in pasture vegetation generated in this study show that most of the default value used in the most frequent assessment models are too low. The transfer factors for pasture vegetation obtained in this study were of about the same magnitude as those determined in the near-environment of WAK /Hauschild and Aumann, 1989/. The geometric mean of 15 measurement gave a transfer factor (Bq kg^{-1} d.w. in grass/ Bq kg^{-1} d.w in soil) of 0.402 for ^{129}I and 0.114 for ^{127}I. The transfer factor for natural ^{127}I is thus lower than those determined for ^{129}I in both pasture vegetation and meadow grass indicating different plant availability of the two iodine isotopes.

The concentrations of ^{129}I in rain, total deposition of ^{129}I and ^{129}I/^{127}I isotope ratios at five sampling sites distributed in West Germany were determined in 1994 and 1995. The average ^{129}I/^{127}I isotope ratio vary between 9.7 x 10^{-7} to 6.0 x 10^{-7} during 1995. The content of the inorganic fraction were the highest at most sample sites. Values of the inorganic ^{129}I were found to range from 43.8 to 79.1% /Krupp and Aumann, 1999/.

The data point to the North Sea as a huge ^{129}I source when most of the ^{129}I emission of the nuclear reprocessing facilities at la Hague (France) and Sellafield end up.

7 Iodine and biological activity

In the mentioned papers by Whitehead and Tikhomirov they showed that iodine can be bound to various components of the soil. Often the conclusion is that iodine will be bound to organic substances mainly humic and fulvic acids. These have been studied in a number of experimental investigations and some of them have already been presented. Already in the 1960ies studies which could lead to these conclusions /Raja and Babcock, 1961/. They treated soils in various ways and incubated then the soil with ^{131}I and then studied the extractability of ^{131}I. The results of pretreatments by autoclaving, oxidation with peroxide and digestion with alcohol, as well as extraction of ^{131}I with various salt solutions, all indicate that the large fraction of ^{131}I retained by the soils is due to reaction with organic matter. Based on this studies of the kinetics in the fixation of ^{131}I, the fixation to soil occurred during 8 days, they concluded that the organic fraction of the soil was involved in the process. They autoclaved the soil and studied the fixation of iodine and found that the fixation was reduced from 85 to 90% of added activity to 26 to 34%. A similar effect was also observed after treatment with hydro peroxide.

In a study performed by Dertinger et al a similar investigation was done but with some more modern design /Dertinger et al, 1986/. ^{125}I was added to soil from the humic horizons in a small column and the water extractable iodine fraction was determined after different periods of time (0.5–550 h). In non-treated soil more than 90% was fixed within the first few hours and then the fixation increased slowly up to 99%. When the experiment was done with sterilised soil (autoclaved for 20 min at 121°C) the initial rapid immobilisation process was not observed rather than a continuous decrease of soluble iodine. If the sterile soil was re-inoculated with an extract of soil organisms the resulting curve was similar to the “sterile” curve for the first 50 hours. Thereafter, a considerable acceleration of the fixation kinetics took place leading to approximately the same equilibrium level of fixation as with the untreated soil. Based on the results, they postulated a chemical conversion of the iodide into another soluble form during a transient phase of immobilisation. This converted form is then fixed to the solid soil components. The model is, according to Dertinger et al, in satisfactory agreement with biogeochemical findings.

Knälmann /1972/ showed that the sorption of iodine increased with increasing levels of organic material in the soil. This was mostly seen in soil with pH above 5.5 where the correlation coefficient often were between 0.81 and 0.91. In soils with lower pH the correlation coefficient was not above 0.61.

Bors and Martens /1992/ investigated the effects of the biomass of the microorganisms in the soil on the sorption of iodine and found interesting correlation. They studied two types of soil – chernocem soil and a podsol soil. They treated the soil in different ways in, order to change the biomass of the soil. Fumigation with chloroform reduce the biomass drastically, drying reduced the biomass less drastically, anaerobic conditions or various oxygen concentrations also affected the biomass of the soil. The binding of iodine was determined after addition of ^{125}I, incubation, centrifugation and determination of the distribution coefficient (K_d). In Table 7-1 the effects of various treatment on the K_d values.

Table 7-1. Effect of increased or decreased biomass on K_d value /Bors and Martens, 1992/.

Treatment	**Chernozem**		**Podsol**	
	Biomass µg C/g soil	**K_d**	**Biomass µg C/g soil**	**K_d**
Control	317	276	142	31
Chloroform	27	25	11	14
Change of C:N:P	431	375	199	36
Change of C:N:P	546	385	232	37
Change of C:N:P	716	376	292	45

A pronounced decrease of the K_d value was seen after sterilisation of the soil (chernozem) using chloroform treatment or gamma irradiation – control value 317.3, chloroform treatment 25 and for gamma irradiation still more. Also changing the temperature had a pronounced effect on the K_d value – at 5°C the K_d was close to 300 for the chernozem soil and decreased to less than 50 at temperature above 30°C.

Fukai /1996/ studied the binding of iodide and iodate to soil by determination of K_d after incubation under various conditions. He showed that an increase in temperature in ambient air or during drying or heating of solid samples markedly decreased K_d values. Grain size had an inverse relationship to K_d of IO^- whereas for I^-, K_d was independent of grain size. The K_d of both radioiodine species for insoluble humic substances were one order of magnitude larger than those for soil.

Bird and Schwartz /1997/ showed that K_d for organic lake sediment were 50 l kg^{-1} under oxic conditions but decreased to 1 under anoxic conditions. This indicate that sorption to compacted organic sediment may not be an effective barrier for iodine released from underground repository to the surface environment. Treating the organic sediment with fungicide, bactericide, gamma irradiation or heat treatment decreased the K_d values by a factor of 1.1, 2.3, 7.5 and 22 respectively. Thus bacteria are important in the sorption of iodine to sediment, although chemical and physical changes may also be important. These results are in agreement with many other results indicating that fixation of iodine in many soils and in lake sediments is dependent on biological processes.

There seems thus that soil organisms are involved in the fixation of iodine. The bacteria seems to be more important than the fungi. Similar results have also been reported by other authors for example; Behrens /1982/ studying aquatic environments, Bunzl and Schimmack /1988/ and El-Kekly and Johanson /1985/ studying soils. In the latter work experiments in order to characterise the ^{125}I labelled compounds which was produced. Still we do not know if iodine is taken up by the soil organisms or if they release enzymes involved in the fixation of iodine. Both possibilities can be an interpretation of available results. The most probably interpretation of the results obtained by Bors and Martens is, however, that the process is going on within the cells of micro organisms but is can not be seen to be a final proof. There are many extracellular enzymes in the soil and if their life time is rather short the level of them will be dependent on the biomass of the enzyme producing cells. A possible explanation could be that there are similar processes as we have in the thyroid gland when producing thyroid hormones for example peroxidase which react with iodine which then react with cyclic compounds such as found in humic and fulvic acids in the soils. Kinetic studies done by the Karlsruhe group indicate such a process /Dertinger et al, 1986/.

In a Japanese study the chemical forms of iodine in soil solutions were studied under non-flooded oxidising and flooded reducing soil conditions /Yuita, 1992/. The results showed that iodate (IO_3^-) was the dominant chemical form under non-flooded soil conditions (86% of water soluble iodine) although most of the iodine was combined with the soil in an insoluble form. Under the flooded soil condition a considerable portion of the insoluble iodine was transformed to water soluble iodine and the iodide was the dominant chemical form (87% of water soluble iodine). The redox potential seems thus to be an important factor affecting the concentration of soluble iodine and also the ratio between iodate and iodide in the soil solution. High concentrations of iodine might reduce the sorption to soil due to the toxic effect of iodine on particularly anoxic bacteria /Sheppard and Hawkins, 1995/.

8 Humic chemistry

The soil is one of the big carbon reservoirs on our earth. Only in the surface layer of the earth there are 7 x10^{14} kg carbon in the atmosphere, 4.8 x10^{14} in the biomass, 2.5 x10^{14} in lakes, 5 to 8 x10^{14} in the upper layer of the sea and 30 to 50 x10^{14} kg carbon in the organic content of the soil. In deeper layers such as sediments in sea, coal and petroleum layers there are much more carbon. The organic material in the soil has an important function to bind iodine to organic material. The organic material is coming from decomposition of plants and micro organisms. It is possible to get some estimate about the amount by determination of the litter fall from plants which is from 10 kg N per ha and year up to 170 kg N per ha and year i tropical forests. Additionally there are dead roots and micro organisms within the soil and their contribution could be several 1000 kg per ha and year /Burns and Martin, 1986/.

Soil includes a broad spectrum of organic constituents, many of which have their counterpart in biological tissues. Two types of compounds can be distinguished. First, non-humic substances consisting of compounds belonging to the well-known classes of biochemistry such as amino acids, carbohydrates and lipids. Second, humic substances, a series of high molecular-weight, brown to black substances formed by secondary synthesis reactions. Compounds in this group are distinctive to the soil or sediment environment and they have characteristics which are dissimilar to polymers from other natural sources. However, the two groups are not easily separated because some non-humic substances such as carbohydrates may be bound covalently to humic matter /Stevenson, 1982/. Stevenson define some of the terms as follows; **Organic residues** is undecayed plant and animal tissues and their partial decomposition products. **Soil biomass** is organic matter present as live microbial tissue. **Humus** is the total of the organic compounds in soil exclusive of undecacayed plant and animal tissues, the partial decomposition products and the soil biomass. **Soil organic matter** is the same as humus. **Humic substances** are a series of relatively high-molecular weight, brown to black coloured substances formed by secondary synthesis reactions. The term is used as a generic name to describe the coloured material or its fractions obtained on the basis of solubility characteristics. These materials are distinctive to the soil (or sediment) environment in that they are dissimilar to the biopolymers of higher plant and micro organisms (including lignin**). Non-humic substances** are compounds to known classes of biochemistry such as amino acids, carbohydrates etc.. **Humin** is the alkali insoluble fraction of soil organic matter or humus**. Humic acid** is the dark-coloured organic material which can be extracted from soil by various reagents for example alkali and is insoluble in dilute acids. **Fulvic acids** is the coloured material which remains in solution after removal of humic acids by acidification. **Hymatomelanic acid** is the alcohol soluble portion of humic acids. Both humic and fulvic acids are heterogeneous, random polymers of a variety of aromatic and aliphatic components.

Humic substances can be subdivided into some components often based on the solubility using some extraction procedure. The extraction often starts with extraction with dilute HCl and possible also some organic solvents. Then the residue is extracted with alkali for example 0.5 M NaOH. When extracted humic substances with alkali a insoluble fraction called humin and a soluble fraction is obtained. Adding acids to the soluble fraction gives a precipitation of humic acid and left in soluble form is fulvic acids. Humic acid fraction can then be extracted by alcohol and a soluble fraction called hymatomelenic acid is found. In the unsolved fraction it is possible to define a grey and a brown humic acids. The humic and fulvic acids are thus mainly defined by their solubility in alkali and acid solutions.

Humic substances are thus build up by secondary synthesis of substances in litter, leaves, needles and other dead material, which has fallen down to the surface. When it has been mixed with the upper soil layer the litter fraction is sometimes called the light fraction which can form up to 30% of the organic material in the soil. In the soil there are a large number of micro organisms. The number of bacteria can reach many billions per gram soil and several hundred millions of Actinomycetes and fungi can also be found. The micro organisms have at least 2 important functions in the soil, either as litter decomposers or as a labile pool of mineral nutrients. There are a pool of water soluble organic compounds in the soil and also a large amount of free enzymes particularly in soil with high productivity. The number of micro organisms and the enzyme activity varies depending on the season for example due to temperature, humidity etc.. From the agronomic point of view we can define two pools, an active or labile pool composed by litter, light fraction, biomass and nonhumic substances. This pool is functioning as a fast source of nutrients for plants and is important for a sustainable agriculture. For the binding of iodine the second pool seems to be more important and composed of the humus and the enzymes in the soil, possible together with the living organisms.

What is humic substances from a chemical point of view? It is not defined from the chemical classification but from the colour and from the solubility properties. The non-humic substances is used and is defined as compounds which can be classified using the terminology of organic chemistry. In contrast, humic substances have from the chemical point of view a very heterogeneous composition which is to some extent unknown. Of the organic material which is added to the soil by litter about 30 to 60% is decomposed already during the first year and the there will be a fraction left with a residence time of up to more than 1000 years and this is what we call humic substances. These fraction contains high molecular compounds formed by secondary synthesis. The humic substances are characterised by many functional group such as COOH, phenols and enol-OH, alcohol-OH-, and quinons all formed by biological processes in the soil. Both humic and fulvic acids show a large diversity of the biological compounds associated with the humic and fulvic acids. This fact together with the limited amount of energy obtained by catabolism of humic and fulvic acids are probably the major reasons that they show such long biological half-life.

For iodine chemistry the most important are probably the aromatic compounds such as phenols or polyphenols and quinons – cyclic compounds which have double bond where iodine relatively easy can react. There are thus certain similarities with the starting compounds in the thyroid gland when thyroid hormones are synthesised. Humic substances can also occur in combination with two and the charged ions such as Ca^{2+}, Fe^{3+} and Al^{3+} as well as combined with clay minerals.

Humic substances are formed by decomposition and changes of biomolecules in plant rests coming from the litter. The exact reaction for this formation is still quite unclear but there are 4 hypothesis;

1. For many years the common opinion has been that humic substances is formed by decomposition and changes of lignin. It will mean for example that transfer of

metoxygroups in thereby forming hydroxyphenoles and oxidation of aliphatic chains to COOH-groups. According to this hypothesis humin is first formed and then by oxidation humic and fulvic acids.

2. In this hypothesis humic substances is also coming by transformation of lignin but the reaction is different. Phenolderivates are released from lignin and then by specific enzyme reactions transformed to quinones which then is polymerised together with amino acids under the formation of humus.

3. The hypothesis 3 has many similarities with 2. Polyphenols synthesised by micro organisms from non lignin compounds for example cellulose. The polyphenols are then oxidised to quinons and transformed to humic substances.

4. In hypothesis 4 the starting compounds are carbohydrates and amino acids which are transferred to polymers by microbial metabolism.

There seems probable that the formation of humic substances is a combination of all these reactions. The result will, however, be the very complex molecules containing many more or less complex ring structure for example polyphenols and quinons. There seems to give many positions where iodine easily can react and form molecular bonds.

In Sweden the most common composition of the forest soil is an upper humus rich layer called mor layer in which there is practically no mixing of the surface organic matter with the mineral portion of the soil. Under this layer usually a leached, grey layer can be found and still deeper a layer with precipitated humic substances and sesquioxides. The thickness of the humus rich layer often show large variations, from a few cm to more than 10 cm. The organic content of this layer vary but as role more than 50% and sometimes up to 80–90%.

9 ^{129}I from nuclear centres

The most extensive and latest investigation of ^{129}I in the surroundings of a reprocessing plants have been performed by a group in Karlsruhe which is summarised in a separate part. In these works the many of the current problems with ^{129}I are discussed. There are of course a number of investigations performed around other reprocessing plants but it seems to me that the Karlsruhe studies are the most relevant and up to date studies. The other studies often came to the same results or may be the opposite that the Karlsruhe studies came to the same results. I have not tried to find the first work in the field but often the opposite the latest using the latest techniques. For these reasons I only discuss other studies relatively short.

^{129}I has been released from a number of reprocessing facilities. Sellafield in UK and la Hague in France has released more than 1000 kg ^{129}I which has been used to study main currents in North Atlantic and Arctic Oceans. In North America the Hanford Reservation and Savannah Reactor facilities have released substantial activities of ^{129}I. In Russia ^{129}I has been released from the Mayak industrial complex near Chelyabinsk. The most pronounced spread of ^{129}I seems to have occurred from Sellafield and la Hague due to the release in sea. In Hanford, the ^{129}I can locally be rather high with the highest ^{129}I/^{127}I ratio of 1.2×10^{-2} found in the thyroid of a gold fish in Gable Mountain Pond in the area /Kilius et al, 1994/. In contrast the ^{129}I/^{127}I ratios at the mouth of Colombia River were significantly lower than those measured from European nuclear fuel reprocessing facilities.

Large investigations about the appearance and transports of ^{129}I within the “Hanford Project“ has been performed /cf. Brauer and Strebin Jr, 1982/. Investigations in Richland, USA during the period 1976–1979, after that the Hanford Purex plant ceased in 1972, showed that the atmospheric ^{129}I activity still was elevated in the Hanford area. Resuspension of ^{129}I deposited in the environment is postulated as a source for the elevated ^{129}I currently observed near Hanford. The ^{129}I was found to accumulate in the top soil and litter layer. The surface concentrations of ^{129}I in forest communities were found to be several times higher than for nearby grass communities. There have been suggestions that these depends on the larger air filtering volume of the forest ecosystems (Table 9-1). These data showed that ^{129}I accumulates in the surface layers of the soil, an observation supported by several investigations. In Table 9-2 the ^{129}I concentrations relative to the surface soils on several sites in USA /Brauer and Strebin Jr, 1982/. A minor loss of ^{129}I to the ground water of soluble iodine seems to occur but a substantial part seems to be bond to the surface layer of soil during many years. These observations have also been reported from studies performed around for example Savannah River and West Valley. In grass collected in the Hanford area, the ^{129}I/^{127}I ratios were between 0.4×10^{-5} and 18×10^{-5} and in the surface soil between 0.3×10^{-5} and 4×10^{-5} /Brauer and Strebin Jr, 1982/. The levels in the top soils were relatively stable after that the reprocessing plants was shut down. ^{129}I levels in food staffs produced within the area has as a role lower levels than in the soil and a ^{129}I/^{127}I ratio from 0.2×10^{-6} (eggs) to 10×10^{-6} (beef) were reported. The surface concentrations of ^{129}I in forest land were much higher than in grass land which might be dependent on the higher interception in the forests. A small transfer of ^{129}I to the ground water could be seen around Hanford.

Table 9-1. ^{129}I levels observed in adjacent grass and forest communities near the West Valley, New York reprocessing site, /Brauer and Strebin Jr, 1982/.

Community	**^{129}I concentration (pg per g)**	
	Litter	**Soil**
Grass	8–60	50
Forest	400–1600	200–300

Table 9-2. Comparison of ^{129}I concentrations for litter and soil relative to surface soil expressed as fractions of surface soil values, /Brauer and Strebin Jr, 1982/.

Place	**^{129}I concentration (relative to 0–10 cm)**		
	Litter	**10–15 cm**	**0.5–l m**
Olympic Peninsula	–	0.07–0.4	0.3
Eastern Oregon	2	0.1–0.5	0.1–0.2
Illinois	0.4–2.1	0.1–0.4	<0.04
Hanford	0.8–2	0.01–0.02	0.002–0.01
Savannah River	1–10	0.03–0.4	
West Valley	0.8–5	0.01–0.04	0.004

In Hanford there have been about 1.900 GBq of ^{129}I generated in the fuel that was discharged from the production reactors and reprocessed between 1944 and 1986.This amount is used as a source term and assumed to be released to the air. The maximum thyroid dose accumulated up to 1995 of over 9 mSv occurred for individuals born in 1954 and 1955 /Robkin and Shleien, 1995/.

Calculations based on studies of the vertical distribution in soil of ^{129}I around Savannah River in USA show an effective residence half-times in the top 30-cm soil depth interval averaged of 30 years /Boone et al, 1985/. In another study of ^{129}I in the surroundings of Savannah River /Kantelo et al, 1982/ an exponentially decreased activity concentrations of ^{129}I in the soil was found with a mean half depth of 7 cm indicating that the deposited ^{129}I is efficiently retained near the surface of the soil. They found no clear correlation between ^{129}I concentration profiles and different soil parameters such as clay, sand or organic matter content. Schüttelkopf and Pimpl have investigated the surroundings of Karlsruhe and found that the ^{129}I was transported very slowly down in the soil.

Investigations in Japan around the reprocessing plants in Tokaimura/Ibaraki /Muramatsu and Ohmomo, 1986/ which had been used since 1972 showed also that the highest activity concentrations of ^{129}I were found in the surface soil (0–5 cm) where 33 mBq per kg was observed. The ^{129}I/^{127}I ratio was in the surface soil, 0–5 cm layers, $3.0x10^{-7}$ the ratio in the layer 20–25 cm vas more than 100 times lower. The corresponding ratios far away from the reprocessing plants was 10^{-9}. The highest ^{129}I levels seen was found in surface soil and in pine needles where 32 mBq per kg fresh weight was observed. The ^{129}I/ ^{127}I ratio in pine needles was $1.8x10^{-5}$. This ratio may well reflect the atmospheric level of ^{129}I. In rain water the ratio in the vicinity of the reprocessing plant was 5.0x10^{-6} compared to 1.1x10^{-8} in rain water from Chiba which was supposed to give background levels /Murumatsu and Ohinomo, 1986/. In algae collected 8 km south of the reprocessing plant 17 mBq of ^{129}I per kg was measured. In spite of the relatively high activity level the corresponding ratio was only 9.9x10^{-8} showing that the algae had a high levels of stable iodine.

10 Conclusions

1. The concentrations of stable iodine in soil is often low. There are two hypothesis about iodine in soil. The largest pool of iodine on our earth is sea water and many scientists suppose that iodine is spread from the sea to the atmosphere and fallout by rain. The other hypothesis says that the iodine in soil is coming from parent material of the soil.

2. From a $^{129}I/^{127}I$ ratio of about 10^{-14} this ratio has reduced by fission produced ^{129}I to about $l0^{-9}$. High ratios in environmental samples are about 10^{-5} and the highest ration is $1.2x10^{-2}$ found in thyroid of a gold fish in Hanford area.

3. In a soil profile the ^{129}I is usually found in the upper layers showing that iodine is bound to soil components. In soils with pH above 6 organic material is the most important soil component in binding of iodine. In more acid soils oxides of aluminium and iron are the most important.

4. In soils there are free enzymes which can transfer iodine to organic molecules in the soil or transfer iodide to elementary iodine or some volatile organic compounds. Release of iodine to the atmosphere from soil seems to be most important in soils with low content of organic material.

5. An effective residence half life in the upper 30 cm layers of soil have been reported to be about 30 years. It seems most probable that this value will have very large variation due to varied organic content in the soil.

6. Transfer factors for transfer from soil to plant (Bq per kg plant material (fresh weight) / Bq per kg soil (dry weight) varied from 0.01 to about 2.

7. Transfer factors for transfer to cow milk (Bq per kg in milk / daily intake Bq) have been determined to 0.002 and to pork 0.015.

8. Binding of iodine to organic matter can be heavily reduced by treated the soil with heat, chloroform, gamma irradiation or other sterilisation methods. There are a correlation between the iodine binding capacity and the biomass in different soils.

9. Large variation either in the distribution of ^{129}I in soil profiles, or in the soil-plant uptake due to soil parameters such as organic matter content or other iodine binding compounds in the soil or to biomass has been found.

10. Even if the iodine is coming from below up in the soil there will probably be a migration by air transport due to resuspension and thereby contaminated the surface soil.

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