THE GEOCHEMISTRY OF IODINE AKD ITS CIRCULATION IN NATURE1

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Since iodine was discovered by Courtois in 1811 the question of its occurrence and distribution in nature has been the subject of much research. In the older literature we find that a number of scientists claim to have found iodine in the majority of materials examined. The French pharmacologist, **A.** Chatin was especially interested in the question of the occurrence of iodine. In a number of works which were published about 1850, and afterwards, he established and demonstrated the presence of iodine in plants and animals, and in air, water and soil. He found this element everywhere, but in very small quantities. It appeared that all the iodine in the world was distributed everywhere, and that thus it was everywhere to be found only in very small concentrations.

I must not omit to mention another French scientist A. Gautier, who about 1900 published the results of his investigations on the occurrence of iodine in sea and air. Through him we find also the first mention of the occurrence of iodine in igneous rocks. He was the first, who with certainty demonstrated and established this fact.

Contemporary scientists doubted the correctness of Chatin's investigations however, and a committee, appointed by the Academy of Sciences in Paris to verify his results could only partly confirm their correctness on account of the defective analytical methods at their disposal.

Later 'nvestigations of the occurrence of iodine, however, showed that Chatin's results were quite correct. The Swiss

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Scientist, Th. von Fellenberg, also investigated the whole of nature, organic and inorganic, and in agreement with Chatin was everywhere able to demonstrate and determine quantitatively the occurrence of iodine in small quantities. The impression we get of the occurrence and distribution of iodine from these authors is thus almost as follows: Iodine seems to occur everywhere, in rocks, in the sea, in the air and in all organisms. But no conformity to law in regard to its occurrence and distribution was demonstrated. We note specially that iodine is always present in organisms. There is also comparatively more iodine in plants than in animals Whence comes this iodine in organisms'? Where was the iodine before the birth of organic life, when the earth was still a floating mass? In other words, it is the history of iodine that we wish to clear up.

The questions regarding the history of elements and the laws affecting their distribution on earth are the subjects with which the new modern branch of science, geochemistry, is concerned. The first object of geochemistry is to find out the laws for the distribution of the elements on the solidification of the earth. This branch of geochemistry has been specially worked out by V. M. Goldschmidt and H. S. Washington. As the earth cooled and organic life began, a whole range of elements were drawn into the organic world. Such elements as chiefly build up organisms are carbon, hydrogen, oxygen, nitrogen and a number of others, but also the great majority of the other elements are regularly found, in greater or lesser quantities, as component parts of organisms. The branch of geochemistry, which specially includes the history of the elements in the organic circulation, we can call bio-geochemistry and this has been particularly worked out by the Russian scientist W. Vernadsky.

We shall now first discuss the geochemistry of iodine, its history and its laws of distribution. I follow here the views propounded by V. M. Goldschmidt as to the geochemical distribution of the elements, according to which the chemical components of the earth, on solidifying, divided themselves, according to affinity and specific weight, into several concentric shells or phases. The innermost is a kernel, consisting of metallic iron; outside

this a sulfide phase, chiefly consisting of ferrous sulfide and, outside all, a molten silicate mass, whose outermost layer forms the crust of the earth. Besides these fluid phases we had a steam-phase that now forms the atmosphere and the hydrosphere. I cannot here go further into these interesting matters, but they have been published by V. M. Goldschmidt in a series of papers in "Skrifter utgitt av Videnskapsakademiet i Oslo." Goldschmidt has sought to find distribution laws for the chemical elements, and he has characterized the elements which are found concentrated in the iron kernel as siderophile, those that are concentrated in the sulfide shell as chalcophile, and those that are concentrated in the silicate mass as lithophile, and finally he called the elements that are present in the steam-phase, the atmophile.

In which of these phases was the iodine or the main portion of the iodine at the time of the earth's original separation into phases? In order to answer this question it was necessary to examine the various phases as to their iodine-content.

That we today find iodine in the air and in the sea, that is in the atmosphere and in the hydrosphere, is no proof that there was iodine in the original steam-phase. It may have come there later on. This question might be settled by systematically determining the ratio between the quantity of chlorine and iodine in the igneous and the sedimentary rocks. Should the sedimentary rocks contain more iodine in proportion to chlorine than the igneous, then we could conclude that iodine must have occurred in the original steam-phase, and thus become a constituent part of the first hydrosphere.

Such investigations have not yet been carried out, but we shall subsequently see that we have been able in another manner to give a positive answer to this question. We note in the meantime that iodine is found in the atmosphere, and that therefore according to Goldschmidt's nomenclature, it shows atmophile characteristics.

The next question will be: Does iodine occur also in the other phases, for instance in the silicate mass, the lithosphere. In order to answer this, von Fellenberg of Bern and I have together

conducted a series of experiments to determine the iodine content of igneous rocks.

The result of these investigations is, in short, the following: Iodine could be demonstrated and quantitatively determined in all the igneous rocks examined. I cannot here go further into the

roame comem of theous found and minerale			
	IODINE		
	mam. per kam.		
Rocks:			
	0.20		
	0.30		
	0.23		
	0.32		
	0.31		
<i>Minerals:</i>			
	0.55		
	0.07		
	0.77		
Hypersthene, Soggendal, Ekersund-Territory	0.94		
Labradorite, Soggendal, Ekersund-Territory	0.44		
	0.50		
	0.16		
	0.23		
	0.90		
	0.63		
	0.18		
Perithitic-microcline, Halvorsrød, Råde, Østfold	0.37		
	0.15		
	0.69		
	0.55		
	0.33		

TABLE 1 *Iodine content* of *igneous rocks and minerals*

methods used in determining these small quantities, but will only refer to the bibliography.

The quantity we find is about **0.2** to **0.3** mgm. per kilogram. All igneous rocks appear to contain the same quantity, or at any rate quantities of the same order. Nor can we find any difference in the iodine content in the more basic and the more acid

rocks. In table 1, I have grouped some figures that illustrate this.

It might now be of interest to know if the iodine content in the rocks was united to a certain mineral or if it was evenly divided over the whole mass. In order to investigate this, a number of rock-forming minerals were examined as to their iodine content, and we find it noteworthy that all minerals, with some few exceptions, contain more iodine per kilogram than the rocks. (See table 1.) This fact is however, easily explained. For these investigations only large crystals were used, and these were probably all of them from pegmatitic veins. In the residual liquor of the magmatic differentiation out of which the pegmatitic minerals are crystallized, the easily soluble compounds are enriched, and as the alkali iodides belong to these easily soluble salts, we can expect to find an enrichment of iodine in just these minerals. The iodine has not, however, preferred any special mineral, if we except the fact that we find in mica minerals a fairly constant quantity of 0.6 to 0.7 mgm. iodine per kilogram. **A** number of minerals containing chlorine were examined, as it might be supposed that the iodine accompanied the other halogens, but this proved not to be the case. Some of the minerals were tested also as to their bromine content, but the proportion between bromine and iodine content was variable and without any distinct conformity to law.

That iodine besides its atmophile tendencies, has also a lithophile character is thus confirmed, but we know nothing as yet of the laws relating to its distribution among the various minerals.

In order to test whether iodine, besides its atmophile and lithophile characteristics, also is capable of appearing in the other phases, von Fellenberg, at the suggestion of Professor Goldschmidt tested some magmatic sulfides as to their iodine content. These also are iodine-bearing, but they contain still smaller quantities than the silicates. We arrive therefore at the interesting result that iodine displays also certain chalcophile tendencies.

But we may ask, when iodine is found in all these three phases, why then is it not to be found also in the iron kernel of the earth? At first it appears quite out of the question that iodine should be

found in some form or other dissolved in the iron, but our investigations have proved that this really is the case. If iodine is found in the earth's kernel, it must in some way or other be soluble in iron, and as iodine is found in all materials that are used in iron smelting works, we might expect to find iodine in all technical

NUMBER	MARK	ORIGIN	IODINE
			mgm, per kam.
1	Hematite	Germany	0.35
$\overline{2}$	Hematite	Luxemburg	0.10
3	Hematite	Luxemburg	0.18
$\overline{\mathbf{4}}$	Hematite	France	0.28
5	Hematite	Longwy	0.69
6	Hematite	Holland	0.50
7	Hematite	England	0.28
8	Grey cast iron	Geneva	0.43
9	Wrought iron	Geneva	0.62
10	Electro steel	Geneva	0.40
11	Cast steel	Geneva	0.55
12	Steely iron	Switzerland	0.22
13	Steel $8N_2C_2$	Sweden	0.25
14	Steel Ti _s W	Tcheckoslovaky	0.38
15	Steel 3Ni ₃	Sweden	0.70
16	Steel CNS	Tcheckoslovaky	0.25
17	$Steel S_{10}$	Sweden	0.40
18	Steel 1Niso	Switzerland	0.70
19	Steel 3Niso	Switzerland	0.31
20	Steel 1 Ni ₄₀ Cr ₁₀	Switzerland	0.26
21	Steel M_{40}	Germany	0.26
22	Steel C ₁	Switzerland	0.63
23	Steel E 60/70	Switzerland	0.17
24	Nave steel	Germany	0.19
25	Nave steel E.	Germany	0.56
26	Nave steel E_{45}	Germany	1.10

TABLE **²** *Iodine content* of *various irons and steels*

iron. In the smelting of iron, coke is used, an article which is particularly rich in iodine, and an admixture of lime, which is also generally rich in iodine. All the conditions favorable to such a result are thus present.

Following up this idea, von Fellenberg and I set to work to test

a number of varieties of technical iron and steel as to their iodine content. We examined, in all, **26** different specimens and found in all of them iodine varying from 0.1 up to 1.1 mgm. iodine per kilogram of iron.

We found, however, no relation between the iodine content, and the chemical composition of the iron. The quantity of iodine present in the iron is dependent only on the iodine content of the materials used in the working of it.

It was now proved that iodine is a constant associate of all iron, but still it was not proved that iodine appears in the earth's iron kernel, though our investigations showed that it was very probable.

A proof of the siderophile character of iodine was however forthcoming when von Fellenberg succeeded in determining iodine in iron-meteorites. The iodine content in the ironmeteorites tested, varied from 0.14 to 1.5 mgm. per kilogram. As we must assume that meteorites come from heavenly bodies that have approximately the same chemical composition as our own planet, we may take it for granted that iodine must also be found in the earth's iron phase, and thus is also siderophile.

Regarding the concentration of iodine in the earth's iron kernel, however, we still know nothing. The meteorites examined give, of course, an indication of the order of size, but we can come to no certain conclusion. On the original division of the earth into phases iodine must however, have been distributed between the various molten masses according to its affinity to every single one of them, and there must be found a proportion for this distribution, conformable to a fixed law. This proportion von Fellenberg endeavored to obtain by testing the proportion between iodine concentration in the two different phases in a socalled compound-meteorite, which contains both iron and a solidified silicate molten mass separated from this. In the case of a compound meteorite from Bomba there was found a distribution quotient between silicate and iron of 100:15. There was, however, as von Fellenberg personally has informed me so little material for such a test that these figures cannot form any basis for broader conclusions.

I accordingly endeavored to find the distribution quotient in another manner. We have seen that iodine is found in all varieties of technical iron. It is therefore natural to suppose that iodine must also occur in the slag formed in iron smelting. In the technical blast furnaces we find, however, both the fluid iron mass and the fluid silicate slag present at the same time, and the iodine will be distributed according to its affinity between the two fluid phases. The distribution quotient between iron and silicate molten mass can thus be found directly.

TABLE 3 *Analysis of iron furnace slags*

A: Blast furnace slag, Steiermark, Austria.

B: Blast furnace slag, Choindez, Berner Jura, Switzerland.

C: Cupola furnace slag, Geneva, Switzerland.

In pursuance of this idea I started an investigation of blastfurnace slags and iron, with the object of determining their iodine content. So as to be able to make a direct comparison with the crust of the earth the slags were analyzed.

Tables **3** and **4** show the iodine content of the slags and of the iron. We see that the proportion is about 100:50 or **2:** 1. The composition of these slags is, however, not identical with the composition of the earth's crust, nor with that of any existing rocks. In the first place these blast-furnace slags do not contain alkalis; likewise their calcium content in proportion to the magnesium is generally greater than in rocks. They are also more basic than the average composition of all igneous rocks. We therefore tested a cupola furnace slag and the cast iron obtained from it at the same time. This slag is considerably richer in silicic acid, and here we find the proportion, somewhat displaced in favor of the iodine content of the iron. The order of magnitude is, however, the same, and as we know that the iodine content of silicate rocks is 0.2 to 0.3 mgm. per kilogram, we can place the iodine content of the iron kernel at 0.1 to 0.15 mgm. per kilogram.

Our studies in connection with the blast-furnace process made another matter clear for us. In the dust which was found left in the flue, the iodine was many times enriched as compared with the content in the slag and in the raw materials used. From this

	IODINE		PROPORTION
	Slag	Iron	
	mgm. per kgm.	mgm. per kgm.	
Α	0.80	0.32	100:40
в	0.85	0.32	100:59
	0.60	0.43	100:72

TABLE ⁴ *Iodine content* of *slag and* of *corresponding iron*

we may conclude that the iodine escaped in its elementary form during the process and was caught by the particles of dust in the flue. Von Fellenberg had also previously found a high concentration of iodine in fireplace-soot and also in cement dust. We have thus the direct proofs also of the atmophile character of iodine. We can sum up the results arrived at in the following law regarding the geochemical distribution of iodine: *Iodine is an element, as far as we hitherto know the only one, which on the earth's division into phases did not show remarkable afinity to any* of *the phases. It became part of them all, but showed, however, more conspicuous atmophile and lithophile than chalcophile and siderophile characteristics.*

Besides the four inorganic phases into which Goldschmidt divides the earth in his geochemical survey, we have still a fifth one, the biosphere. The biosphere is bounded by the atmosphere and the hydrosphere on the one side, and the rocks on the other, and is the seat of all the organic life on earth. As I have already mentioned it was Vernadsky in particular, who urged the importance of the biosphere in all geochemical surveys.

When an element enters into organisms and is thus drawn into the biosphere, its geochemical history is changed in an essential degree. Iodine is in this case specially interesting because the concentrations in which most often we find it in the biosphere are greater than in the rocks. A concentration of iodine must therefore have taken place in the biosphere. Iodine has, so to speak, been pursued by organisms.

Another characteristic of iodine is that it is found in specific combinations with other elements, and this also only in the biosphere. The iodine minerals that we know are formed and exist only in the upper layers of the earth's crust, and onlyin deposits of organic origin. These minerals are, however, all of them, relatively rare. We must assume that the iodine content here has been enriched by organisms capable of accumulating iodine and that through the decay of these organisms it was withdrawn from the organic circulation. We know the compounds iodargyrite, AgI, marshite, CUI, and miersite, 4AgI.CuI. This last must be regarded as a mixed crystal of AgI and CuI. Further, we know iodembolite Ag(Br,I), a mixed crystal of AgBr and AgI. In the saltpeter deposits in Chile we find the minerals lautarite, $Ca(10₃)₂$, and dietzeite, $7Ca (10₃)₂ \cdot SCaCrO₄$. All these, however, except the silver minerals are mineralogical rarities. We cannot say this of the occurrence of iodine in the saltpeter deposits above mentioned, for there we find iodine in very great quantities. The annual production in Chile is about **70** per cent of the total world production, nevertheless only 10 per cent of the total iodine content of the minerals is utilized as iodine. Nevertheless these great quantities of iodine are practically negligible compared with the enormous reserve of iodine in the rocks.

When the earth cooled, the first steam-atmosphere was condensed and became the hydrosphere and an atmosphere which

contained less moisture. Disintegration of the rocks took place, and in consequence part of the iodine which was found in the rocks was released, The greater part of the iodine of the disintegrating rocks accompanies them when they become soil. Plants take their iodine from the soils, animals get their iodine by feeding on plants. Figure 1 shows the circulation of iodine in nature. Plants and animals on decaying set free their iodine again. But plants do not obtain their iodine exclusively from the soil; they also absorb iodine direct from the atmosphere. They "inhale" iodine. When they decay, a good deal of the iodine set free escapes again into the air. During certain processes some iodine is set free from the soil and escapes into the atmosphere. The atmosphere returns iodine to the soil again; the

FIG. 1. CIRCULATION OF IODINE IN NATURE

iodine falls to the earth dissolved in rain-water. Also snow, hail and rain etc., contain iodine. Plants are always richer in iodine than animals.

A part of the iodine which is set free by disintegration is carried away by surface water. Fresh-water alge absorb great quantities of iodine and are considerably richer in iodine than land-plants. Fresh-water fish again get their iodine from these algæ.

The iodine which is not captured by organisms is carried by the surface water into rivers and thus into the sea. The sea has obtained its iodine-content, partly in this way, and partly from the first steam-atmosphere which surrounded the earth. The iodine concentration in the sea is considerably greater than in fresh water. Many scientists have fixed this at about 0.02 mgm. per liter. Marine alga are therefore also richer in iodine than

fresh-water plants and for the same reason marine animals are also richer in iodine than land-animals and freshwater ones. Figure 1 shows how plants obtain iodine from the sea, seaanimals again from plants, On the decay of organisms a part of the iodine is again set free, but large quantities are withdrawn from the organic circulation and pass over into sediments. In later geological periods, these sediments may be forced up above the sea-level, they may again be exposed to disintegration and the iodine be set free. It will then be drawn into the biosphere, and

we have the iodine circulation completed. In these remarks regarding the circulation of iodine I have chiefly confined myself to publications by von Fellenberg and myself, but I must not fail to call attention to important papers by J. F. McClendon on the iodine content in various fresh waters in the United States and weighty investigations of the iodine content of various foodstuffs from goiterous and non-goiterous regions in the United States.

During the past year I have made a special study of the iodine content of marine organisms, for the purpose of making clear the circulation of iodine in the sea. As far as the iodine content in sea-organisms is concerned, if we omit the determinations governing iodine content in marine algee, we have only some older investigations on fishes by the French scientist, P. Bourcet, as well as more recent investigations by D. K. Tressler and **A.** W. Wells of the United States Bureau of Fisheries in Washington.

In cooperation with Karl Closs, Helmer Haaland and Sigurd Opstad Madsen, some representatives of the commonest Norwegian teleosts were examined. The results are to be found in table *5.*

Compared with the figures found by Bourcet, Tressler and Wells, our own are much higher. It is therefore probable that the iodine content is dependent on the location of the fishing grounds.

IODINE IN WATERFREE SUBSTANCE	
Muscles	Intestinal canal
mgm. per kam.	mam. per kam.
29.4	42.3
24.6	41.8
1.75	1.40
0.90	4.08
0.50	1.65
0.60	2.24

TABLE 6 *Iodine content of the intestinal canal of Norwegian teleosts*

The specimens examined by us were caught in spring 1928, at Moere on the west coast of Norway.

The great difference in the iodine contents of the flesh of the species examined made it appear probable that the iodine content of the flesh depended on the character of the food. In order to prove this the intestinal canal of some of the most typical species were examined. The result of these investigations which are shown in table 6, show that the teleosts with the high iodine content of the flesh also always show a very high iodine content of the intestinal canal. The iodine-poor fishes, however, always give much smaller figures for the iodine content of the intestinal canal. Another interesting fact is that the species of teleosts which we found to be rich in iodine always are poor in fat and

rich in phosphorus. We further see that fishes rich in iodine are what we can call typical bottom fishes which chiefly feed on invertebrates of the bottom fauna. In collaboration with Closs and Madsen some typical specimens of the bottom fauna were examined, and in accordance with our supposition we found large quantities of iodine. Table **7** gives a review of some of our results.

In order to get a complete picture of the cycle of iodine in the sea, it was, however, first necessary to examine the plankton. The lower types of sea-animals and a great number of fish derive their chief supply of food from plankton. Table **7** contains our results for the invertebrates examined.

In cooperation with Karl Closs, I examined a large supply of phytoplankton, chiefly consisting of diatoms taken from the Oslo Fjord in March. The moist plankton contained 38.09 mgm. of iodine per kilogram, or 230.91 mgm. per kilogram waterfree substance, that is not only a larger quantity than in all other organisms previously investigated, but also of a quite different order of magnitude. The iodine content determined, is of a like order of magnitude as the iodine content in the various species of sea-weed (Fucus, Laminaria).

The plankton is thus able to concentrate the marine iodine. The iodine content of the ocean is about 0.02 mgm. per liter. Various scientists have arrived at various values; the quantity

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appears also to vary greatly with the locality, but the order of magnitude is always the same. According to von Fellenberg's investigations up to **25** per cent of this iodine is united to insoluble filterable particles (plankton). It is clear that these figures must vary considerably as the production of plankton for many reasons may be very great or very small. We see, however, that iodine is about 10,000 times as concentrated in water-free plankton as in the sea water. This gives a good idea of the extent to which plankton is capable of concentrating iodine. Samples of zooplankton also were examined and were found to be rich in iodine. Thus we found for the most common crustaceans the figures given in table 8.

<i>Ioarne content of crustaceans</i>	
	IODINE IN WATER- FREE SUBSTANCE
	mgm. per kgm.
	$11.6 - 21.9$ 7.1
Amphipods (Euthemisto compressa et E, bispinosa)	13.0

TABLE 8 *Iodine content* of *crustaceans*

On the basis of the investigations which we now have at our disposal we can form an idea of the circulation of iodine in the sea, or in other words the iodine metabolism of the sea, roughly as follows: (see fig. **2).** The iodine that is released from weathered rocks is carried by the rivers out to sea. Here it is absorbed by the plankton just like other nutritive salts from the land (especially phosphorus and calcium compounds). The production of plankton is periodical and depends at the Norwegian coast chiefly on the supply of nutriments from land. It is especially rich during the melting of snow in spring; at that time the great quantities of iodine, which have been released by the erosion of rocks in the course of the winter, are carried more or less suddenly out into the sea. During and immediately after the flowering of plankton in the upper layers of the ocean, only a very small quantity of it is utilized as food for the lower or higher species of animals (especially zooplankton). The great mass

sinks to the bottom, and some of this plankton serves there as food for the invertebrates of the bottom fauna. The bottom fishes, as we have seen before, obtain their iodine mostly from these animals. On the decay of the various plants and animals a part of iodine is, as mentioned before, once more set free and can then be absorbed anew by plants. But large quantities of iodine are withdrawn from the circulation by the fact that they sink to the bottom with the remains of plarits and animals and pass over into the sediments.

There is also a constant reciprocal action between the surface of the sea and the atmosphere. By certain processes iodine is

FIG. 2. CIRCULATION OF IODINE IN THE SEA

set free at the surface, it escapes and passes over into the atmosphere. It is carried back into the sea again by precipitation. As the concentration of iodine in the sea is so much higher than in fresh water, it is clear that sea-air will be considerably richer in iodine than inland air. And this fact is of no little importance for both men and animals in districts along the sea-coast. For experiments have shown a higher concentration of iodine in the surrounding nature and therefore in all organisms in coastal districts compared with corresponding inland organisms.

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