海水中に溶存する元素量の規則性について

及び海洋の年齢について (その二)

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On the Regularities of Quantities of Elements dissolving in Sea-water.<sup>1)</sup> And the Age of the Ocean  $(\Pi)$ 

> -Another New Improved Method for the Determination of the Age of the Ocean-

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# 旨

本研究は石橋及び協力者により遂行中の海洋化学に於ける基礎的研究に属するものである。元素の原子番号と Clark数との間に何等かの没則性の存すべき事を予想して、此等の間の相関々係を求めようとする試みは、W.D. Harkins 氏(米)により提唱されて以来多くの人々により行われた所であるが、著者等は海水溶存元素に関す る呆統的研究を遂行の途上、その溶存元素量と各原子番号との間に驚くべき規則性の存在するを見出すことがで きた。この規則性は本文中の各図に示された直線形によつて一目瞭然である。此等の各図に於て、元素の量を表 わすためにとつた著者等の特徴は、従来 Clark 等の行つた重量%表示法ではなく、その代りに原子数に比例す る量即ちミリモル(m.m.)を以てし、更に其の常用対数を取つて、之を縦軸とし、横軸には原子番号をとつた ことにある。何故なれば、斯くする方がすべての説明に有効適切なるのみならず、重量%法を用いるよりも一層 満足すべき結果を得たからである。

即ち著者等は、他の研究者により定批された分析数値と著者等の数室に於て測定し得た分析数値に基プき、重要にして興味ある規則性を認めることができたのであるが、それにより吾々は注目すべき次の事実を察知することができるであろう。

1. 稅上の規則性は只に海水に於てのみならず地球上の至水圈に於ても成立するであろう。何となれば全水圈の中、海水は約98%を占有し、他は合計しても約2%に過ぎないからである。更に岩石圏を加え、この地球全体に於ける此等の元素の存在量の規則性をも示唆するのではあるまいか。

2. Csの量の予測の際に述べたように、内挿又は外挿法により海水中の溶存元素量を、他の近接元素の量より 予知し得るであろう。それによれば週期表に於て未だ空位に残されている唯二つの元素 Eca Cs及びEca I の量は あまりにも超微量であつて分析が不可であろうと思われる。況んや夫等が放射崩壊をするに於ておやである。

3. 放射性元素の崩壊現象を種々の地球化学的年代の決定に利用することは、近年に至り醒々試みられる所で あり、此等の方法により比較的に確実らしい年齢が多数算定されている。この種の方法 (U→Ra→Pb) を、既 に石橋及び協力者は海洋年齢の新算定に適用し印刷公表ずみである (1938)。而して更に著者等の提唱する海洋 年齢の算定法は等しく放射性に関連する理論によるものであるが、其の諸想、内容ともに従来の夫とは全く趣を 異にする新方法である。

本文中の(3)式に於て明かなように、海洋の年齢 TがRaの崩壊恒数入に比して充分大なる時は(事実然りである)、海水中のRaの現存量Nrと、1年間を通じて海水へのRa搬入量Noとの間には、Tに無関係に

 $N_T = 2.27 \times 10^8 No$ 

なる式が成立する。又第3図のMg-Baを結ぶ直線とRaの原子番号88の横軸との交点をRとし、 Rの縦軸の指標 をNとすれば

#### $N = No \times T$

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なる関係が成立する。故に之を前式と組合すれば

$$T = \frac{N}{No} = \frac{N}{\frac{N_T}{2.27 \times 10^3}} = \frac{2.27 \times 10^3 N}{N_T}$$

となる。而して上式に於てN, Nrの数値は旣知であるので、T即ち海洋の年齡を求めることができる。

即ち N=1×10<sup>-6</sup>、 Nr=7×10<sup>-13</sup> を代入すれば

$$T = \frac{2.27 \times 10^3 \times 10^{-6}}{7 \times 10^{-13}} \doteq 3 \times 10^{9} \text{ years}$$

斯して海洋の年齡は大約30億年と云うことになる。この数字は、海洋の年齡は10~50億年の間にあるべしと云う 石橋の前報告の提唱を支持するものである。

4. なお海水中に溶存する元素量の規則性については、其の後石橋の主宰する海洋化学研究所に於て更に著し い研究成果を挙げつつある。稿を新たにして報告したいと思う。

## I. Introduction

Under presumption that there should lie some regularities between atomic numbers and Clark's numbers of the elements, several attem t; for finding some relation between these numbers have been made since Mr. W. D. Harkins<sup>2</sup>) first proposed it. He stated its general tendency in his well known rule named after him thus: "The element of even atomic number has, as a rule, larger Clark's number than the neighbouring element of old atomic number."

On the way to systemait research<sup>3</sup>) on the elements dissolved in the sea-water, the writers noticed some regular relation between the quantities of dissolved elements and their atomic numbers, even in taking up only the h drosphere. Persuing further, the writers found surprising regularities in the case of the sea-water. As shown in Table 1, by

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the elements of the same family, the quantity of the elements dissolved in the sea-water decreases as the position of the elements in the periodic table becomes lower. This rate of decrease, as shown in Figure  $1 \sim 3$ ,



gives almost a straight line in the case of the already quantified akIali elements, alkali earth elements and halogen group elements. But by the elements of highest position in the same group, such as Li, F(Be is not yet found), they show quite exceptional quantities, just as they do in

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their chemical and physical property.

As seen in the above figures, the writers' method of indicating the quantity of the elements differs from weight % method which has been employed by Mr. Clark. Instead, the writers used the quantity proportional to the atom number, that is, millimol (m.m.). And moreover, they took its common logarithm as its vertical axis, and atomic number as its horizontal axis. In doing so further exposition was faciliated, and more satisfactory result was obtained than by the use of weight % method.

Owing to the enormous effort of chemical and physical researches of the past century, wek now that the atomic number represents all the essential qualities of atom itself, both chemical and physical. Considering that the quantity of elements dissolved in the sea-water is dependent on the degree of facility of their delivery from land by the water  $^{4)}$ , it is assumed from large point of view that the periodioal character of chemical and physical property of atom itself is found, as one of its manifestations between the quantity of the elements in the sea-water and their atomic numbers. Especially in dealing with such great source as the ocean, statistical law of great numbers (Gesetz der grossen Zahlen) would be applied rather satisfactorily. Moreover, in the lineage of strong uniformity such as the sea-water, totality of each individual analysis value is far superior to that of the cases on land. This fact, too, is the main cause of regurality recognized by the writers. Thus considering, in geochemical arguments, its fundamental quantity of the elements should rather be dealt from the stand point of the quantity rational to their atom numbers (m.m.) than from weight %. And the writers intend to maintain this view point henceforth in this kind of researches.

elements	atomic numbers	g/L	m.m./L	log m.m.
F	9	0.000822	0.043	-1.367 Carnot
		0.0014	0.74	-0.131 Wattenberg
Cl	17	18.42	519.6	2.71567
Br	35	Q. 063	0.79	-0.102
I	53	0.00005	0.00038	-3.420
Ecal	85	-	_	
Li	3	0.0002	0.029	-1.538
Na	11	10.16	432.0	2.635
к	19	0.37	9.4	0.973
Rb	37	0.0000035	0.0004	- 3. 398
Cs	55	n×10-9	n×10 <sup>-8</sup>	-7~-8
EcaCs	87	_		-
Be	4		- 1	-
Mg	12	1.24	51.0	1.708
Ca	20	0.40	10.0	1.000
Sr	38	0.013	0.15	-0.824
Ba	56	0.0002	0.0015	-2.824
Ra	88	$n \times 10^{-13}$	$n \times 10^{-13}$	-12~-13

Table	1
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II. Consideration on the Experimental Value

Since former ages there have been enormous number of researches made on the quantities of the elements dissolved in the sea-water. And there have by no means been a few cases where fairly

varied analysis values were obtained. However, the values referred in this thesis are taken from the generalizing thesis of Mr. T. G. Thompson<sup>5</sup>) and Mr. H. Wattenberg<sup>6</sup>) which are considered most reliable today, except by the elements Li, Rb, Cu, Au which have been determined at the writers' laboratory. But only in case of Ra, as Mr. C. S. Piggot and Mr. W. D. Urry7) recognized, the writers considered the most recent analysis value of Mrs. R. D. Evans, A. F. Kip and E. G. Moberg to be correct, and it was employed. Thus Table 1, Table 2 and Figurel  $\sim 4$  were made according to the above said values.

Assuming that the salts in the sea-water have been carried in mainly by rain water and rivers and accumulated during a long period, it is easily presumed that especially in lithophile elements such as alkali metal, alkali earth metal and halogen group which are widely distributed near the earth surface mainly, relation between their (nantity and atomic numbers is regular; and that in cases of the elements not so well distributed on the earth surface such as chalcophile and siderophile elements from the third A group to the fourth B group..... the sixth A group in the periodic table, it is somewhat irregular.

As there lies great difficulty in this kind of research, analysis value of the elements existing in the sea-water is at present very insufficient and imperfect. Therefore it is impossible to ascertain the above stated presumption yet. Neverthless, as an example, the following Table 2 and Figure 4 will show the relation between dissolving concentration and atomic numbers of the elements such as copper group elements Cu, Ag, Au of which analysis is considered almost completed.

Table 9			10	Atomic number										
		Table	- 4			10	2	0	30	40	50	60	70	80
elements	atomic numbers	g/L	m.m./L	log m.m.	logmn	-1 -2			Cu					
Cu	29	$3 \times 10^{-5}$	0.0047	-2.328	۶. ا	-3					40			
Ag	47	$\int 2 \times 10^{-7}$	0.00028	-3.553Wattenberg	ł	-4				Y	1			
		3×10-7	0.00020	-3.699Thompson	ł	-5								
Au	79	4×10-9	$2 \times 10^{-8}$	-7.699	ł	-6						1		
					ł	-1							Ν	9

#### 1. On the Elements of Halogen Group.

As shown in Table 1, Figure 1, and in the above exposition, it is clear that the writers attempt has been fulfilled. The



analysis value of F varies by different people more or less, and its position on the figure, accordingly, is not decisive. However, it matters little in the present argument.

## 2. On the Elements of Alkali Group.

It explains from itself by Table 1, Figure 2, and the above stated consideration. Only concerning Cs, its existence, of course was anticipated, but its qualification and quantification have not been cuite sufficient. But it has become possible to presume the cuantity of Cs dissolved in the sea-water by extending the binding line of alkali elements and acquiring the crossing point with the vertical axis of the atomic number 55 of Cs, which is shown in Figure 2.

This value is thus acquired from Figure 2. If the required concentration of Cs is C millimol :

$$\log C = -7.5$$

 $\therefore$  C =  $3 \times 10^{-8}$  millimol

The writers invented a micro-analytical method enabling the identifying of Cs  $(4 \times 10^{-9} g/L)$  to the above said degree, and by applying it on the natural brine prepared from sea-water, the quantity of Cs in the sea water was indirectly measured. Thus the above stated presumption was proved correct. That is to say the quantity of Cs in the sea-water, according to the writers' experiment, is  $n \times 10^{-8}$ millimol ( $n \times 10^{-9}$ g/L), and within the limit of experimental error, it coincides well with the presumed value.

## 3. On the Elements of Alkali Earth Group.

On Table 1 and Figure 3, detailed consideration on the regularity of the second A Group, that is alkali earth group will be made. First it is noticeable that Ra which is dishibuted to high degree all over the earth surface, exists in the sea-water only as an exceptional case, remarkably deviated from the extended line of Mg-Ca-Sr-Ba. And while the cuantity of Ra obtained from outside by Figure 3 abould be  $1 \times 10^{-6}$  millimol, that is  $2 \times 10^{-7}$ g/L, the cuantity of the same in the sea-water obtained by actual measuring is far smaller,  $n \times 10^{-13}$  millimol, that is  $n \times 10^{-13}$  g/L.<sup>7)8)</sup>

The Writers thought out of the following two reasons as its main causes, and attempted quantitative explanation with them which brought satisfactory result. The lack of actual quantity of Ra, as stated above, comes from :

1. Decrease of the quantity by natural disintegration caused by radioactivity of Ra.

2. Decrease of the quantity by precipitation and deposition of Ra to the sea bottom.

And the writers calculated the first case to be far greater than the second (about 100 times as much). It will be stated further on this.

At first, the explanation will be made on the decrease by disintegration. On Figure 3, the meaning of the crossing point R of the straight line binding Mg-Ba and the horizontal axis of the atomic number, 83, of Ra is considered. This indicates the concentration of Ra with assumption that Ra, delivered from various sources<sup>0</sup>) into the sea-water ever since pirmeval ages, has, like other stable elements, been accumulated without being disintegrated. As the theory of disintegration of Ra is wellknown, by its application, R is acquired theoretically from the actual quantity of Ra in the sea-water. Thus :

If quantity of Ra delivered into 1 L of sea-water per year be No millimol, the age of the ocean be T year, the actual quantity of Ra in sea-water of 1 L be  $N_T$  millimol, and the disintegration coefficient of Ra be. $\lambda$  (year unit ), then

The above formula is geometrical progression of  $e^{-\lambda T}$  as its first term and  $e^{-\lambda}$  as its common ratio.

And T is the age of the ocean, to which various different values have been given, and is considered to be between 1 to 100 hundred million years. Formerly it has been taken as around 1 hundred million years, but recently its value tends to increase, and especially the value acquired by radioactivity is considered to be from over 10 hundred million years to several 10 hundred million years.

Therefore in the formula (2)  $e^{-\lambda} \cdot e^{-\lambda}$  may be ignored against  $e^{-\lambda}$ . Thus

$$\mathbf{N}_{\boldsymbol{T}} = \mathbf{N}_{0} \left\langle \frac{-\mathbf{e}^{-\lambda}}{\mathbf{e}^{-\lambda} - 1} \right\rangle = \mathbf{N}_{0} \left\langle \frac{\mathbf{e}^{-\lambda}}{1 - \mathbf{e}^{-\lambda}} \right\rangle$$

When e = 2.71828,  $\lambda = 4.38 \times 10^{-4}$  is substituted in the above formula

$$N_{T} = N_{0} \left\langle \frac{e^{-4 \cdot 38} \times 10^{-4}}{1 - e^{-4 \cdot 38} \times 10^{-4}} \right\rangle = N_{0} \times \frac{0.99956}{0.00044} = 2.27 \times 10^{3} N_{0} \dots (3)$$

On N<sub>T</sub>, that is the actual quantity of Ra in the sea-water, there have been great number of determination made following the research of Mr. Joly, Mr. Eve. And also it has been known that its quantity varies somewhat according to the depth of the sea-water. However, the writers take 1.5  $\times 10^{-13}$ g/L, that is  $7 \times 10^{-13}$  millimol, as the quantity of Ra representing the whole ocean. The rea-

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son of which will be stated later.

Thus if  $N_T = 7 \times 10^{-13}$ 

according to the formula (3),

∴  $N_0 = 3 \times 10^{-13} = 2.27 \times 10^3 N_0$ 

Therefore N<sub>0</sub>, the quantity of Ra delivered into 1 L of sea-water in a year, is  $3 \times 10^{-10}$  millimol. And if the concentration of Ra on the imaginary point R is N millimol :

 $N = N_0 \times T = 3 \times 10^{-16} T$ 

Therefore if  $T=10^{\circ}$  (that is 10 hundred million years)

 $N = 3 \times 10^{-16} \times 10^9 = 3 \times 10^{-7}$  millimol

And the characteristic is  $\log 3 \times 10^{-7} = -6.523$ 

Also if  $T=5\times 10^9$  (that is 50 hundred million years)

 $N = 3 \times 10^{-16} \times 5 \times 10^{9} = 1.5 \times 10^{-6}$  millumol

And the characteristic is  $\log 1.5 \times 10^{-6} = 5.824$ 

As the characteristic acquired by Figure 3 is -6, the above stated value becomes slightly greater if **T** is taken as 10 hundred million years, and slightly smaller if it is taken as 50 hundred million years. Even today it still is hard to know the exact age, **T**, of the ocean, and it is very hard to make accurate calculation. However, as stated above, if the age of the ocean is assumed to be between 20 to 30 hundred million years, concentration of Ra at the point R would be well explained. Although correction is necessary for the decrease of Ra by its precipitation and deposition to the sea botton, it seems so small avalue that it could be ignored, comparing with the decrease by disintegration on which will be stated later.

Next, the writers will make consideration on the above said second cause, the decrease of Ra by precipitation. The experiment value of Mr. Evans and others (1930) seems to show most clearly the manner of precipitation and deposition of Ra in the sea-water. Referring it from his thesis, it is shown in Table 3 as follows :

depth (m)	quantity of Ra (g/L)					
0	$(0.3\pm0.1)\times10^{-13}g$					
0	$(0.5\pm0.2)\times$ "					
600	$(1.5\pm0.1)\times$ "					
600	$(1.3\pm0.3)\times$ "					
1, 300	$(3.2\pm0.2)\times "$					
1, 300	$(2.6\pm 0.3) \times "$					
sea bottom deposit (1, 300m)	$(3.00\pm0.05)\times10^{-12}g/g$					
11	$(3.17 \pm 0.05) \times "$					

Table 3 Ra content in the sea-water at various depths

It is found there that Ra content in the sea-water increases rational to the depth fairly straight lineally, and that in the sea-bottom soil deposit on the same place, enormous amount of Ra is deposited and condensed (comparatively speaking, of course). Here, the quantity of Ra is  $3 \times 10^{-12}$ g Ra/g, in other word  $3 \times 10^{-9}$ g Ra/kg. It will not be mentioned here of various statements <sup>11</sup>) on the subject of the mechanism of this condensation. It is found here by the experiment that Ra alone, unlike othere elements, is precipitated and concentrated to unusually great quantity. Of course all the elements dissolved in the sea-water are also precipitated more or less by various causes, but comparing with that of Ra they are too far smaller degree. The writers have taken this up quantitatively

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as follows. Table 4 shows the quantity of the elements of each alkali earth group in the sea-water and in the red clay <sup>12</sup>) of the deep sea bottom, and the ratio of the quantity of these. As for the quantity, the ratio of the atom number, that is m.m. is taken.

#### Table 4.

The quantity of the elements of alkali earth group in the sea-water and in the deep sea bottom.

sea-water			red clay					
	millimol ratio, the quantit of Mg as 1		weight %	%/atomic weight	ratio, the qua- ntity of Mg as1			
Mg	51.0	1	1.99	8.2×10 <sup>-2</sup>	1			
Ca	10.0	$200 \times 10^{-1}$	1.42	$3.5 \times 10^{-2}$	4.3×10-1			
Sr	0.15	$2.9 \times 10^{-3}$	0.045	5.1×10-4	6.2×10-3			
Ba	0.002	$3.8 \times 10^{-5}$	0.18	$1.3 \times 10^{-3}$	1.6×10-2			
Ra	7×10-13	1.4×10 <sup>-14</sup>	1×10-9	4.4×10 <sup>-1 2</sup>	5.4×10 <sup>-11</sup>			

As seen in Table 4, 51.0 millimol of Mg existing in the sea-water is 1.99 % in the red clay. Therefore, if the rate of its precipitation and condensation in the red clay from the sea-water is taken as 1, Ca should be deposited as much as

$$1 \times \frac{4.3 \times 10^{-1}}{2 \times 10^{-1}} \Rightarrow 2$$
 multiple of Mg

And as to Sr, similarly

 $1 \times \frac{6.2 \times 10^{-3}}{2.9 \times 10^{-3}} \approx 2 \text{ multiple of Mg}$ 

Similarly this relation whth Ba and Ra are indicated as follows.

#### Table 5.

The ratio of the quantity of the elements of alkali earth group in the sea-water precipitated and condensed to the sea bottom.

	the ratio of the quantity precipitated to the sea bottom
Mg	1
Ca	2
Sr	2
Ba	400
Ra	4,000

According to this, Ba must be precipitated to the sea bottom about 400 times as much as Mg, and as to Ra, about 4,000 times as much. Therefore the decrease of Ra by this phenomena could also be considered as one of the causes of the afore said deviation from the straight line. The writers calculated the quantity of Ra deposited by this cause as follows. The result show, as mentioned before, the influence of this

cause to be comparatively small. The value employed in this calculation is by no means without doubt, and the reliability of the acquired value is not always decisive. However, trusting this mothod to be logical, it suffices to know a approximate value.

As shown in Table 3, according to the afore said experiment of Mr. Evans, Ra content in the surface soil deposit of the sea bottom of 1,300m depth is  $3 \times 10^{-12}$ g Ra/g. Assuming a water pillar to be made on 10cm square of this soil, its water volume will be 1,300×10=13,000L. Therefore, considering that Ra contained in this 10cm square of the sea bottom soil is precipitated and condensed from 13,000L of the sea-water above, the loss of Ra from 1 L of the sea-water per yeas by precipitation is acquired as follows.

By another method, 13) the speed of precipitation of the deposition in the deep sea bottom is calc-

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ulated as approximately 0.07cm rer 1,000 years.<sup>14</sup>) Therefore, by obtaining the number of years for the production of 1g of the deposition on 10cm square of the sea bottom, and by dividing  $3 \times 10^{-12}$ g Ra with'it, the quantity of Ra precipitated from the sea-water in a year is acquired. If 1g of the deposition is spread evenly on 10cm square, its thickness h would be as follows. The specific gravity of the deposition is considered here as 1.5, temporarily.

$$1.5h \times 10^3 = 1$$

: h = 0.007 cm

2×10-

As it takes 1,000 years for 0.07cm of deposition, for 0.007cm of it, is should take

$$1,000 \times \frac{0.007}{0.07} = 100$$
 years

And from 13,000L of the sea-water pillar,  $3 \times 10^{-12}$  g of Ra is precipitated in 100 years. Therefore the quantity of the deposition from 1L of it in a year will be

$$\frac{3 \times 10^{-13}}{100 \times 13,000} \approx 2 \times 10^{-18} \text{g Ra/I}$$

And its sum total from the beginning of the ocean (assumed as 10 hundred million years) is :

$$^{18}$$
g  $\times 10^9 = 2 \times 10^{-9}$  g Ra/L

$$=1 \times 10^{-8}$$
 millimol Ra

Comparing this with the afore said decrease  $10^{-6}$  millimol by disintegration, it is very small (about 1/100). Therefore the loss of Ra from the sea-water by precipitation and deposition, comparing with that by disintegration, may well be ignored when the writers take up the problem of deviation of Ra from the straight line.

The following fact should de noted. At seen in Table 5, deposition of Ba is greater than that of Mg, Ca and Sr and accordingly its value should be somewhat below the straight line. But in reality it is as above. It is not known yet whether this comes from insufficiency of analysis value or from some other reason.

4. On the Elements of Copper Group.

In Table 2 and Figure 4, the relation between concentration of Cu, Ag, Au content in the seawater and their atomic numbers is snown, which approves the writers' proposition well. As the writers' view on chalcophile elements and siderophile elements is already outlined in chapter II, consideration on experimental value, it will be omitted here. Further research on this subject is expected in the future.

# III. Another New Improved Method for the Determination of the Age of the Ocean

By the above exposition it ir known that the elements distributed all over land, when they are in the same family, keep straight lineal relation between their quantity dissolved in the sea-water and their atomic numbers, unless there is some special cause to hinder it. By the application of this relationship, the writers propose a new method of determination of the age of the ocean.

The application of the phenomena of disintegration of radio-elements to the determination of geochemical age has been attempted often lately. And various ages have been determined which are comparatively accurate. In the writers' laboratory this sort of method was already applied to the ocean, and a new method has been proposed.<sup>15</sup>Now, although it is based on the theory of radioactivity, it differs entirely from the already known methods in its conception and content.

As already stated in Chapter I, formula(3), when the age of the ocean T is large enough comparing with  $\lambda$  the fixed number of disintegration of Ra (and in fact it is so), between N<sub>T</sub>, the actual quantity of Ra in the sea-water, and N<sub>0</sub> the quantity of Ra delivered in a year, the relation

#### $N_T = 2.27 \times 10^8 N_0$

is established, with no connection with T. As shown in Figure 3, when the crossing point of the straight line binding Mg-Ba and the horizontal axis of the atomic number 88 of Ra is indicated as R, and the characteristic of the vertical axis of R as N, the relation

$$N = N_0 T$$

is established. Consequently combining this with the former formula it becomes

$$T = \frac{N}{N_0} = \frac{N}{\frac{N_T}{2.27 \times 10^8}} = \frac{2.27 \times 10^3 N}{N_T}$$

And as N,  $N_T$  are known in the above formula, T, the age of the ocean is obtainable. That is, by substituting  $N=1 \times 10^{-6}$ ,  $N_T = 7 \times 10^{-13}$ 

$$T = \frac{2.27 \times 10^3 \times 10^{-6}}{7 \times 10^{-13}} \approx 3 \times 10^9 \text{ years}$$

Thus the age of the ocean becomes approximately 30 hundred million years. In this calculation, the decrease of Ra by precipitation and deposition to the sea bottom is ignored for the afore said reason. However, the value employed there is not entirely reliable in the present situation, and if the amount of decrease is found actually great, it then would no more be ignored. In such case, the value of N of the above formula might become smaller, and accordingly the age of the jocean, T, might also become smaller. However, the above said value could be considered expressing the approximate age of the ocean, and the age acquired by this mothod coincides with that of the other methods based on the phenomena of radioactivity. And also it supports the former proposition of Ishibashi, that the age of the ocean should be between 10 to 50 hundred million years.<sup>15</sup>)

## IV. Conclusion and Suggestion for the Future

Based on the analysis value already quantified by other writers and on that determined by the writers' research, regularity of the relation between the quantity of the elements dissolved in the seawater and their atomic numbers has been stated hereto. Recognizing such regularity, the following notable facts are found.

1. The above said regularity should exist not only in the sea-water but also in the hydrosphere of the earth, because the sea-water occupies 98% of the hydrosphere, while the rest occupies merely 2% of it. Moreover, it seems to suggest the regularity of the existing elements on the whole earth.

2. As stated in the paragraph of the estimation of the quantity of Cs, the quantity of the elements dissolved in the sea-water either from within or without can be estimated by that of the neighbouring elements. Consequently, the quantity of Eka Cs and Eka I, the only two elements which at present in nature are left blank in the periodic table can be presumed approximately.

On Figure 1 and 2, concentration of No. 85, Eka I and No. 87, Eka Cs are, from without,  $10^{-8}$  m.m/L and  $10^{-14}$  m.m/L respectively. However, as these elements seem to possess radioactivity, similar to Ra in Figure 3, even if they existed in the sea-water, their quantity must be much less than the value stated above. If so, it seems almost impossible to obtain these two elements from the sea-water, as it is too small a quantity for the present analytical method.

3. Consideration has hereto been made on the elements dissolved in the sea-water. In enlarging this view point and applying it to the elements of the whole earth, the following facts are found. When the graph is drawn by taking common logarithm of the number which is Clark's number divided by its atomic weight as vertical axis, and by taking atomic number as horizontal axis, regularity is not very well found in all the families of ordinary elements. But in rare gas elements, as

				Atomic number
		Table 6.		10 220 30 40 50 60 10 00 90
e!ements	atomic number	Clark's number atomic weight	Clark's number atomic weight	log Ne Kr
He	2	2.00×10-7	- 6.699	3 +0 Xe
Ne	10	2.47 × 10 <sup>-8</sup>	- 7.607	-12
Ar	18	8.77×10-6	- 5.057	- 23
Kr	36	2.41×10 <sup>-10</sup>	- 9.618	100
Xe	54	2.32×10-11	-10.635	
Rn	86	4.54 × 10 <sup>-18</sup>	-17.343	Rn b
	<u>.</u>			10

shown in Table 6 and Figure 5, excepting Ar and Rn, fairly regular relation is found between the two.

Figure 5. Rare Gass Elements

In this case, the reason for the deviation of A1 is not known. But marked deviation of the quantity of Rn could be, just as in the case of Ra, explained as the decrease by disintegration.

4. As shown hereto, by the homogeneous system such as hydrosphere and atmosphere, relation of straight line is established between the quantity of the elements and their atomic numbers; and by the heterogeneous system such as the elements of the earth crust, no regularity is found <sup>16</sup>). While each analysis value of the homogeneous system is sufficiently reliable for the argument of the whole, the average Clarks' numbers have insufficient analysis value for their basis, and also the object of analysis is limited to a portion of the earth in this case, that is, only 15 km underground. Hence comes such declination to the argument of very general prospective as above.

From this point of view, the writers intend in the future to make correction of Clark's number itself and apply the hereto stated consideration on it.

In conclusion, the writers express their hearty thanks to the Department of Education for bestowing the fund for the scientific research for a part of this research.

# Literature

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In this research, Dr. Ishibashi estimated the age of the ocean to be between  $10 \sim 50$  hundred million years.

16) At Clark's number also, by the well distributed elements such as alkali and halogen, the relation approaches the straight line, but by siderophile and ahalcophile elements the degree of deviation becomes higher.

(昭和26年10月受理)

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