



Investigation of iodine concentration in salt, water and soil along the coast of Zhejiang, China*

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Abstract: Objective: We aim to describe the environment iodine concentration in salt, water and soil along Zhejiang Province coast in the China foreland. It will be helpful for us to judge whether this area is insufficient in iodine and universal iodized salt is necessary or not. Methods: We collected iodized salt samples, drinking water samples (tap water in the towns, and well water or spring water in the villages), water samples from different sources (ditches, lakes, rivers) and soil samples through random sampling in June, 2005. Salt, water and soil iodine was detected by arsenic-cerium redox method. Statistical analysis was expressed as mean \pm SEM by Windows SPSS 13.0. Results: (1) The iodine concentration in salt was 27.9 \pm 4.33 mg/kg ($n=108$). (2) Seventy-five water samples were collected. The water iodine value was 0.6~84.8 μ g/L (mean of 11.66 μ g/L). The watershed along the Qiantang River has significantly higher iodine content than the water in Lin'an in mountain area ($P<0.01$). The iodine content and mean iodine content of tap water, well or spring water and natural water sources were 4.30 \pm 2.43 μ g/L ($n=34$), 23.59 \pm 27.74 μ g/L ($n=19$) and 12.72 \pm 10.72 μ g/L ($n=22$) respectively. This indicated that among environmental water sources, the ditch iodine content was the highest with river water iodine being the lowest ($P<0.01$). (3) Soil iodine value was 0.11~2.93 mg/kg (mean of 1.32 mg/kg). Though there was no statistical difference of soil iodine in different districts ($P=0.131$), soil iodine content correlated positively with water iodine content. Conclusion: Iodine concentration in salt accords with national policy of adding iodine in salt. Foreland has more iodine in water than mountain area. The data reflected that water and soil iodine in foreland area was not high, which suggests universal iodized salt should be necessary. Environment iodine has relatively close association with pollution.

Key words: Iodine, Salt, Water, Soil, Coast

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INTRODUCTION

Significantly close relationship exists among environment, health and disease such as thyroid disease associated with iodine metabolism. Iodine has long been recognized as an essential micronutrient for humans and livestock (Kelly and Snedden, 1960). Thyroid disease is closely associated with absorption of iodine in food. Its deficiency causes goiter and different forms of physical and mental retardation

(WHO, 2002). Although the number of iodine deficiency disorders affecting people is declining, on a global scale the number of affected people is still over 740 million which is 13% of the world's population with 30% of the remainder being at risk (WHO, 2002). Human iodine intake is closely related to iodine concentration of water, soil and salt. Iodine concentration in water and soil reflects the environmental iodine distribution, and is also an important index of human's natural iodine intake and an indirect index of environmental pollution. In recent years argument has always existed whether the amount of iodine intake has impact or not on the incidence rate of thyroid

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disease. Hangzhou famous for Westlake in Zhejiang Province is located in the China foreland. In order to get the statistics on proper iodine addition in salt, and on prevention and cure of iodine related disease, in June 2005 we conducted for the first time survey of iodine concentration of salt, water and soil in the Hangzhou area.

MATERIALS AND METHODS

Sources of materials

(1) Iodized salt in different supermarkets, (2) drinking water (tap water in towns, well water or spring water in villages) and water samples from ditches, lakes, rivers, (3) soil. All the samples mentioned above were collected randomly from the Hangzhou area.

Sample collection methods

We chose an ecosphere in the south, west, north, east and center of the study area as shown in Fig.1 and collected samples including water from ditches, lakes, rivers, well water, tap and superficial soil. The research team members were mainly from the Department of Endocrinology of Sir Run Run Shaw Hospital, and received unified training to guarantee the quality control. All the sampling bottles were soaked in diluted hydrochloric acid, cleaned and heated dry.



Fig.1 Geographic instruction

Hangzhou including Yuhang, Jianggan, Xiacheng, Shangcheng and Xihu districts is along the Qiantang River. Fuyang is a plain region of the Fuchun River's drainage basin upstream the Qiantang River flowing into Hangzhou Bay in the East China Sea. Lin'an is in mountain area

Iodine assay

Salt, water and soil iodine was detected by arsenic-cerium redox method. Reaction indicator and seconds-counter were used to indicate reaction end point and time. As there was a linear correlation between the logarithm of iodine concentration and the logarithm of reaction time, iodine concentration in solution could be calculated by double logarithmic regression equation. Staff in the Center of Disease Control of Zhejiang Province did the detection of iodine content. Water iodine-detection kit was produced by Wuhan Zhongsheng Biochemical Technique Limited Company in China. The laboratory procedures are described below: (1) Salt samples were dissolved in distilled water and water samples were detected directly. Soil samples were incinerated at 550 °C for 4 h with mixed alkaline compounding chemicals including potassium carbonate, zinc sulfate, potassium chlorate and sodium chloride. The ash left was dissolved in distilled water and the supernatant was removed for detection; (2) A pipette was used to accurately transfer 1.0 ml of standard iodine-detection solution and water or solution sample respectively into test tube (There were two kinds of standard iodine-detection solution. One for low water iodine concentration is 2~10 µg/L and the other for high concentration of 10~80 µg/L); (3) Reducer of 0.5 ml was added into each tube and joggled; (4) Indicator of 0.5 ml was added into each tube and joggled; (5) One tube was chosen. Sample injector was used to rapidly add 0.5 ml oxidant into the tube while the seconds-counter was started. After the tube was joggled, the solution color turned blue. White paper was used as background to observe the changing of color. With continuing reaction, the color turned from blue to amethyst and finally to red. The seconds-counter was stopped when the amethyst just turned red and the time was recorded; (6) Iodine concentration was calculated. As there was a linear correlation between the logarithm of iodine concentration C (µg/L) $[\lg C]$ and logarithm of reaction time S (s) $[\lg S]$, the regression equation of standard curve $[\lg C = a + b \lg S]$ could be obtained by computer. With this equation and sample reaction time, sample iodine content (µg/L) could be calculated. Unit (µg/L) of salt and soil was converted to another unit (mg/kg).

Statistical analysis

Comparison within groups was made by Windows SPSS 13.0 software.

RESULTS

Iodine content in salt

We randomly obtained 108 salt samples from different supermarkets of different districts. The distribution of iodine value was 20.0~37.8 mg/kg and iodine concentration in salt was 27.9±4.33 mg/kg which accorded with Chinese official policy published in 1995.

Iodine concentration of tap water sample (Table 1)

Thirty-four 0.6~9.9 µg/L (mean of 4.30 µg/L) tap water samples were collected. Tap water sample iodine concentration was highest in Yuhang district and lowest in Lin'an City, there was significant difference ($P<0.001$) between the two values.

Iodine concentration of well or spring drinking water sample (Table 2)

The concentration range and mean of 19 samples were 0.6~84.8 µg/L and 23.59 µg/L respectively. Yuhang and Lin'an had the highest and lowest iodine content respectively. There was significant difference ($P<0.001$) among these districts.

Iodine concentration of natural water source (Table 3)

Twenty-two samples were gathered. The concentration range and mean were 1.4~39.9 µg/L and 12.72 µg/L respectively. The natural water source iodine concentration in natural ditches, lakes and river (Fuchun River) differed significantly ($P=0.001$). The muddier the water was, the higher was the iodine content.

Table 1 Tap water iodine concentration

(I) District	N	Iodine conc. (µg/L)	Sig. (I-J)						
			(J) District						
			L-A	F-Y	X-C	S-C	X-H	J-G	Y-H
L-A	4	1.45±0.93	–	0.503	0.007	0.007	0.006	0.000	0.000
F-Y	5	2.01±0.16	0.503	–	0.026	0.024	0.022	0.000	0.000
Hangzhou1 X-C	5	3.80±0.35	0.007	0.026	–	0.979	0.938	0.001	0.000
Hangzhou2 S-C	5	3.82±0.19	0.007	0.024	0.979	–	0.959	0.001	0.000
Hangzhou3 X-H	5	3.86±0.22	0.006	0.022	0.938	0.959	–	0.001	0.000
Hangzhou4 J-G	5	6.74±2.96	0.000	0.000	0.001	0.001	0.001	–	0.147
Hangzhou5 Y-H	5	7.88±0.44	0.000	0.000	0.000	0.000	0.000	0.147	–
Total	34	4.30±2.43	$F=17.858, P<0.001$						

Lin'an=L-A, Fuyang=F-Y, Yuhang=Y-H, Jianggan=J-G, Xiacheng=X-C, Shangcheng=S-C, Xihu=X-H; Statistics showed that all the paired groups had significant difference except Xihu and Xiacheng district ($P=0.938$), Xihu and Shangcheng district ($P=0.959$), Yuhang and Jianggan district ($P=0.147$), Xiacheng and Shangcheng ($P=0.979$)

Table 2 Well (or spring) water iodine concentration

(I) District	N	Iodine conc. (µg/L)	Sig. (I-J)			
			(J) District			
			L-A	F-Y	J-G	Y-H
L-A	4	1.45±0.93	–	0.660	0.041	0.000
F-Y	5	5.20±2.30	0.660	–	0.077	0.000
Hangzhou4 J-G	5	20.18±17.38	0.041	0.077	–	0.000
Hangzhou5 Y-H	5	63.10±16.58	0.000	0.000	0.000	–
Total	19	23.59±27.74	$F=24.693, P<0.001$			

Comparison between every two groups showed that there was significant between Lin'an and Yuhang district ($P=0.000$), Jianggan and Yuhang ($P=0.000$), Fuyang and Yuhang ($P=0.000$). No statistical difference was found to exist between Lin'an and Fuyang district ($P=0.660$), Fuyang and Jianggan ($P=0.077$)

Table 3 Physical environment water iodine concentration

(I) Water source	N	Iodine conc. (µg/L)	Sig. (I-J)		
			(J) Water source		
			Rivers	Lakes	Ditches
Rivers	5	5.20±2.30	–	0.684	0.001
Lakes	8	6.99±4.10	0.684	–	0.001
Ditches	9	22.00±10.91	0.001	0.001	–
Total	22	12.72±10.72	$F=11.505, P=0.001$		

No statistical difference existed between water of rivers and lakes ($P=0.684$). Significant difference existed between water of lakes and natural ditches ($P=0.001$), water of rivers and natural ditches ($P=0.001$)

Result of comparing iodine content among tap water, well or spring water and natural water source sample (Fig.2)

We collected 75 water samples with iodine concentration of 0.6~84.8 µg/L (mean of 11.66 µg/L). Well or spring water had highest iodine content of drinking water samples, followed by natural water source, with tap water having the lowest content. Significant difference ($P<0.01$) existed. Underground water had more iodine than surface water.

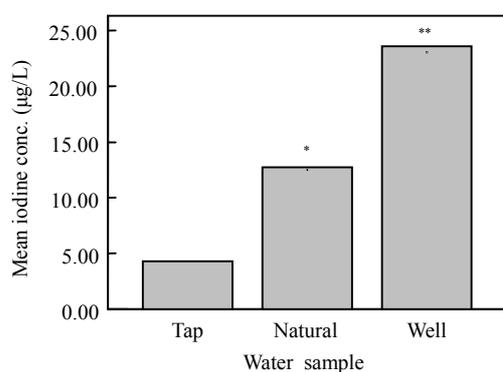


Fig.2 Water iodine compared in Hangzhou

Tap: Tap water sample; Natural: Natural water source sample; Well: Well or spring drinking water sample, Conc.: Concentration; *Represents values statistically higher than tap water ($P=0.045$); **Represents values statistically higher than tap water ($P<0.001$) and natural water source ($P=0.025$)

Iodine content in soil of Hangzhou area (Table 4)

Forty-six soil samples were collected in which iodine concentration was 0.11~2.93 mg/kg (mean of 1.32 mg/kg). The statistics indicated there was highest iodine content in Xihu district and lowest in Lin'an City.

Table 4 Soil iodine

(I) District	N	Iodine conc. (µg/L)	Sig. (I-J) (J) District				
			L-A	F-Y	J-G	Y-H	X-H
L-A	8	0.76±0.33	-	0.525	0.123	0.036	0.040
F-Y	10	1.02±0.11	0.525	-	0.316	0.102	0.109
Hangzhou4 J-G	10	1.41±0.25	0.123	0.316	-	0.498	0.485
Hangzhou5 Y-H	10	1.68±0.21	0.036	0.102	0.498	-	0.952
Hangzhou3 X-H	8	1.71±0.48	0.040	0.109	0.485	0.952	-
Total	46	1.32±0.14	$F=2.047, P=0.131$				

Statistics of all every two groups left had no significant difference excluding Lin'an City and Xihu district ($P=0.040$), Lin'an City and Yuhang district ($P=0.036$)

Comparison of iodine concentration between water and soil sample (Fig.3)

Fig.3 shows that the curves of concentration of iodine in water and soil have similar rising trend in direction. The higher the water iodine content was, the higher was the soil iodine content. Iodine content in the environment was found to decrease with increasing distance from the sea.

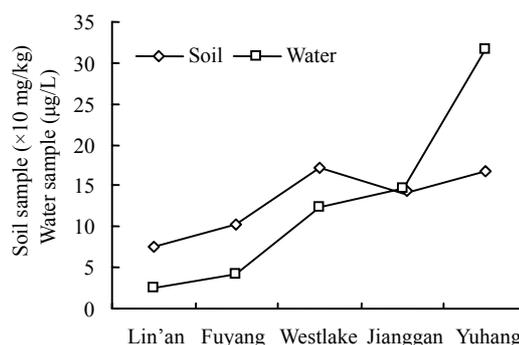


Fig.3 The trend of iodine concentration in soil and water

DISCUSSION AND CONCLUSION

Human health including character development and mental change is generally associated with physical environment. Air, soil and water are necessary for all life. Some ingredients in soil and water play an important role in human life's development. As an important endocrine gland maintaining life activity, the thyroid secretes thyroxine which promotes growth and development of bone, muscle, height and weight, and maintains the stabilization of energy and material metabolism (Fuge and Johnson, 1986). It is known that iodine is necessary for the synthesis of thyroxine. Iodine content in soil and water is directly associated with the content of human food iodine and affects survival rate and living quality. Both extremely high and low iodine intake has obvious damage to human: (1) hyperthyroidism, (2) hypothyroidism and young children's mental retardation due to hypothyroidism, (3) thyroid carcinoma, (4) autoimmune thyroid diseases. A body of clinical and epidemiologic evidence points to excessive ingestion of iodine as an environmental agent (Rose et al., 2002). As inducers of autoimmune thyroid diseases, environmental factors could also aggravate

already existing autoimmune thyroid diseases (Marrack *et al.*, 2001).

This survey revealed that water iodine content was highest in Hangzhou, that Fuyang was the second highest, and that Lin'an water iodine content was the lowest. The data indicate that water sources of foreland have higher iodine content than those of inland. Although statistical difference did not exist in soil iodine concentrations of different districts, their absolute values were similar to those of water samples. So we considered that iodine content in the environment decreases from high to low level with increasing distance from the sea, probably because during the process of forming land plain area continuously battered and infiltrated by seawater, the iodine content of soil and freshwater in this area is high. Foreland residents have more iodine intake than inland residents due to the water they drink.

We also found tap water from factory had less iodine than that of the main drinking water source in the countryside. Water management including precipitation, filtration and disinfection when part of iodine is absorbed and volatilized probably results in this phenomenon. Because well water directly permeates through the soil and soil iodine content (mean=1.32 mg/kg) was higher than water iodine (mean=11.66 µg/L), underground has even more iodine. Most drinking water sources in the countryside are well supplying mainly underground water. This caused the distinction of iodine intake through water. The amount of town residents' water iodine intake may be smaller than that of countryside residents' on the basis of the analysis above.

Iodine content indirectly reflects environmental pollution especially of the water. When clear water is polluted, iodine cannot be easily precipitated from it, infiltrated into it and volatilized from it, leading to more solutes in the water. This survey revealed that ditches and Fuchun River have the highest and lowest iodine concentrations respectively and significant difference ($P<0.01$) existed between them. The muddier the water was, the higher was the iodine content, especially near large factories. The pollution of ditches and lakes may be the worst, although it is possible that Fuchun River which is one of the most important water systems in Hangzhou is still less polluted. That makes us think highly of environmental protection to decrease the incidence of ecolo-

gic disease due to water pollution and promote sustainable development of our natural resources and society.

On the basis of our standards that define water iodine value <10 µg/L as iodine-deficient district and water iodine value >200 µg/L as iodine-abundant district, China is an iodine-deficient country whose endemic area involves 29 provinces, including more than 7 000 000 iodine-deficient patients compromising more than half of the patients in the world (Zheng *et al.*, 2002). Distribution of soil iodine values in China (A level) is 0.39~14.71 mg/kg (mean of 3.76 mg/kg). Because mean of iodine content in water and soil samples is 11.66 µg/L and 1.32 mg/kg respectively, generally the Hangzhou area still lacks iodine although it is in the China foreland. Normal adults and children need 150 µg and 200 µg per day respectively (Chen, 2001). According to normal amount of drinking water per day iodine intake from water plus intake from food still cannot reach the standard. In 1995, our country reformed the policy on iodized salt in order to ameliorate the iodine-deficient condition. Iodine values in salt samples accords with national standard. If we take 3~5 g salt everyday routinely (Chen, 2001), we can have 100~150 µg iodine intake which mainly satisfies human requirement for iodine.

The relationship between the iodine intake level of a population and the occurrence of thyroid diseases in the population is U-shaped with an increase in risk from both low and high iodine intakes (Chen *et al.*, 2002; Laurberg *et al.*, 2001). Endocrinologists now have complete understanding of the relation between iodine and incidence rate of goiter by combining epidemiological investigations and experiments with animal models. Severe iodine deficiency with median 24-h urinary iodine excretion below 25 mg needs immediate attention and correction. Less severe iodine deficiency with median urinary iodine excretion below 120 mg per 24 h is associated with multinodular autonomous growth and function of the thyroid gland leading to goiter and hyperthyroidism in middle aged and elderly subjects. The lower the iodine intake, the earlier and more prominent are the abnormalities. At the other end of the spectrum, severely excessive iodine intake starting at median urinary iodine excretion levels of around 800 mg per 24 h is associated with a higher prevalence of thyroid hypofunction and goiter in children. A number of

studies indicated that moderate and mild iodine excess (median urinary iodine >220 mg per 24 h) are associated with a more frequent occurrence of hypothyroidism, especially in elderly subjects (Laurberg *et al.*, 2001). Finally the bottom represents sporadic goiter which could not be a public health problem. In short, extremely high or low iodine intake harms human health, so the mistake of taking universal iodized salt regardless of iodine content in the environment should be corrected. Some factories now producing food with excessive iodine will cause severe overflow of iodized food.

Finally, research about incidence rate of thyroid disease along the Zhejiang Province coast has not yet finished. The results of relationship between the environment iodine and prevalence of thyroid disease will be presented.

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