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Determination by Neutron Analysis of Mercury Residues in Foodstuffs

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방사화분석법에 의한 식품중의 잔류수은의 정량

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Abstract

In order to find out the degree of mercury contamination of common foodstuffs a series of determination was carried out by a highly sensitive activation analysis and the following results were obtained.

1. Polished rice contained 0.050 ppm of mercury whereas rice bran had 0.095 ppm mercury which was found in other grain in lesser degree.
2. Vegetables and fruits also contained 0.035~0.190 ppm of mercury with relatively small variations from sample except persimmon which had a considerably higher amount of mercury.
3. Soybean sprout contained an unexpectantly high amount of mercury.
4. Of the animal products chicken and egg contained more mercury than the meat.

Introduction

Pesticides are detectable in many foods, in man and animal, and in our natural surroundings.[1~8] Because pesticides are formed to kill or injure some living organisms,[7,8] they are potentially dangerous to other living organisms.[9] It has become an important task to determine the levels of pesticides in biological materials. Radioactivation analysis is very useful for this purpose. Mercury is a toxic element whose determination in tissues and foodstuffs by neutron activation is quite sensitive. Trace mercury analysis is not only difficult but very necessary since the WHO-FAO recommended safe upper limit for mercury content in foodstuffs is 0.05 ppm.[10] As most mercury compounds are rather volatile, it seems natural to separate mercury from the dissolved sample by distillation.[11] Different methods of extracting mercury from the distillate might be applied,[12] precipitation as sulfide, precipitation by organic precipitants, or extraction by organic solvents, and electrolytic deposition.[13] Neutron activation analysis, exhibiting a very high sensitivity for the determination of mercury, has been extensively used in this methods in the literature.[14~19] Activation analysis of mercury in sample of various nature, with or without chemical separation has been studies by workers, as is apparent from the survey of Lutz.[20~21] Because of the volatility of various compound of mercury, special precaution must be taken during

the preparation sample, the irradiation step and the subsequent chemical separation procedures. Brune [22~24] report the liquid sample was irradiated in a frozen from and corrected the flux perturbation in the sample. For radiochemical separation of mercury Sjostrand[16] demonstrated the distillation technique and Kim[15] the isotopes exchange method. This method was found to be highly satisfactory because of the high radiochemical purity of the mercury and of the high chemical yield with enhances the good accuracy of the methods.

Materials and Methods

1. Sampling

Paldal variety, the sample rice, was harvested in 1969 Suwon region and hulled into polished rice and unpolished one by the common method adopted by the Agricultural Products Inspection Station of Korea. Samples of soybean sprout, chicken, egg, vegetables and various grain are obtained from commercial soure in Seoul and Kyung-gi province, fruits such as apples, orange, persimmon are purchased from different growing site in the period of February to November 1969.

2. Irradiation

Irradiation were performed in the Institute' TRIGA Mark II reactor. The sample in heat sealed in a vial and irradiated for 2~3 days in a thermal neutron flux 3.8×10^{12} n/cm sec.

The length of irradiation and sample weight adjusted according to the approximate mercury content, and were generally in the range 5~24 hours and 1.0 grams respectively.

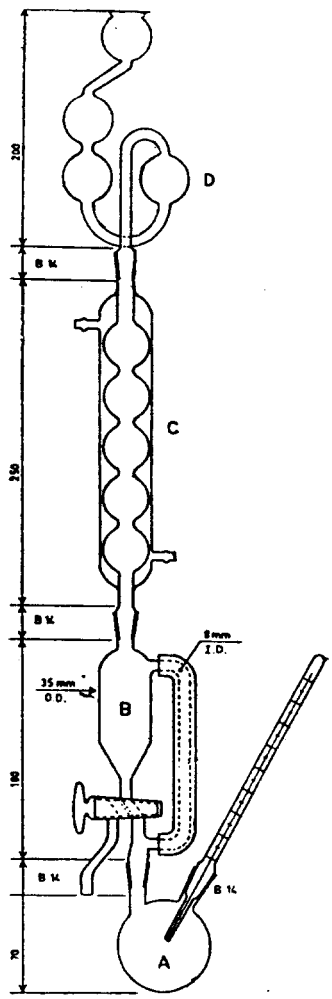

Fig. 1. Apparatus for controlled decomposition of organic materials, also used as distillation apparatus

Table. 1. Comparison of methods for mercury neutron analysis

Material	Quantity	Time for analysis	Chemical treatment	Reference
Biol. mat. muscles	05. g—1g dry	Activ. 2—3d	Wet ashing, carrier, distillation	(16)
Biol. mat.	20 mg	Activ. 1 week	Wet ashing, preciptat. separat. 3 steps, gravimetr yield determin.	(17)
Urine	200 ml	Chem. tr. 17—20 min	Extraction with APDTC	(33)
Urine	5~10 ml	1 min	Dilute with H_2O	(34)
Blood	0.1 ml	Ashing 1.5hr Evapor. 1hr	Wet ashing with H_2SO_4 and $KMnO_4$, extraction into dithizone, liberation of Hg by heating	(35,36)

3. Experimental procedure

The important step in the mercury analysis is destruction of sample, which causes ususlly difficulties because the mercury compounds are easily volatilized. Destruction of sample was therefore carried out, with nitric acid, in a closed system, specially designed for this purpose. As shown in Fig. 1, this apparatus consits of a 100 ml distillation flask (A), a receiver flask (B), a reflux conderser (C), a secondary conderser (D), a reagent input flask (E), a heating mantle. The apparatus combines a refluxing and distillation function in order to prevent mercury distillation and to diminish the solution volume for a subsequent procedure.

Add oxidation mixture 7 ml of concentrated sulfuric acid and 7 ml of conc. nitric acid to make 14 ml.

Connect the flask to the Bethge apparatus. Pour a few milliter of water into the splash head. Close the tap and heat the flask until the nitric acid has distilled into the reservoir. Allow the temperature to rise to over 300°C and boil the sample in the sulfuric acid remaining. When cold, run the distillate back into the flask. Repeat the heating cycle until the solution in completely cleared and colorless when boiling under reflux.

Table 2. Nuclear data

Target nuclide	Abundance(%)	Act (b)	Isotope produced	Half life	Radiation and energy (MeV)
^{196}Hg	0.146	880[a]	^{197}Hg	65hr	r: 0.077 (20%), 0.19 (0.5%) 0.068 AuK-X ray
		420[a]	^{197m}Hg	24hr	r: 1.33 (31%), 1.64 (4.5%) 0.071 Hg K-X ray
^{198}Hg	10.02	0.018[a]	^{199m}Hg	4.2min	r: 0.159, 0.368
^{202}Hg	29.80	3.8	^{203}Hg	46.9d	β: 0.208 r: 0.279 (83%) Tl K-X ray (0.073)
^{205}Hg	6.8	0.43	^{205}Hg	5.5min	β: 1.8 r: 0.203

[a] : Quoted from (32)

4. Mercury distillation

A mixture of 5ml. of 70% perchloric acid and 0.5 gram of glycine added to the dissolved sample and heat is applied to the flask. When the solution is boiling under reflux, close the tap and collect the mercury fraction in the reservoir. The distillation when the temperature exceeds 250°C should be interrupted. The distillate was neutralized by adding concentrated ammonium hydroxide and made acid by adding 5 drop of concentrated hydrochloric acid. The solution was gently heated and Mercury was preciptated as sulfide by introducing hydrogen sulfide. The precipitate was filter though a glass filter, washed with distilled water, dissolved in hot aqua regia and reprecipited as sulfide. The chemical yield obtained, after correcting for absorption of γray in the precipitates.

5. Radioactivity measurement

The radioactivites were measured with a TMC Model (102) 100 channel pluse height analyzer with 2 inch ×2 inch in Nal (TI) crystal. The ^{203}Hg activity was determined by measuring the area of its 0.279 Mev photopeck. After neutron radioactivation of the food, characteristic mercury nuclides are produced; ^{197}Hg, 65hr half-life, 0.77 MeV gamma rays. ^{203}Hg, 47d half-life 0.28-MeV gamma rays.

Table 3. Comparison of several methods of mercury trace analysis

Qualitative	1. Reinsch method	(25)
	2. Aluminium amalgam method	(26)
Quantitative	1. Colorimetric method (a) dithizone	(27)
	(b) Di-β-napholthiocarbalzine	(28)
	2. Polarographic method	(13)

Table 4. Comparison of detection limit for mercury

Method	Unit:10^{-9}g	References
1. Light absorption photometry	5	(29)
2. Atom absorption	30	(13, 30)
3. Flame spectrometry	100	(13, 30)
4. Spark-source mass spectrometry	0.6	(13, 30)
5. D.C. arc emission spectrography	100	(13, 30)
6. Copper spark spectrography	500	(13, 30)
7. Graphite arc spectrography	10	(13, 30)
8. Gas chromatography	10	(36)
9. Nuclear activation	5	(16, 17)

Table 4. Mercury containing pesticides.

1. Ethylmercuric urea (E.M.U.)
2. Methoxymercuric chloride (M.M.C.)
3. N-Tolymercuri-p-toluenesulfonanilide (T.T.S.)
4. Phenylmercuric acetate (P.M.C.)
5. Ethylmercuric chloride (E.M.C.)
6. Phenylmercuric chloride (P.M.C.)
7. Ethylmercuric phosphate (E.M.P.)
8. Chlorophenyl mercury (usplun) (C.P.M.)
9. Phenylmercuric iodide (P.M.I.)
10. Ethylmercuric-p-toluene sulfonanilide (E.M.T.S.)
11. Phenylmercuric-p-toluene sulfonanilide (P.M.T.S.)
12. Phenylmercuric dinaphthylmethanie disulfonic acid (P.M.D.D.) (Quated from. 35)

Results

They provide evidence that radioactivation analysis can play an important role for mercury in the environment.

Mercury was detected in all samples tested in the present experiments as is seen in Table 7~12. Activation analysis an important part in obtaining these analytical data. This technique, which is commonly used for biological material facilitates complete dissolution of the organic fraction but leaves an undissolved silicate residue. Negligible loss of mercury will occur by this process. The method is some what time-consuming, however, and only a limited number of samples can be processed simultaneously.

Table 5. Volatilization of mercury during decomposition and distillation

Heating cycle	Percentage of ^{203}Hg activity detected		
	Flask	Reservoir	Splash head
1. Decomposition of tobacco	38	62	not detectable
2. Decomposition of apple	44	56	not detectable
3. Decomposition of rice	41	59	not detectable
4. Mercury distillation	not detectable	99	not detectable

Table 5 shows that about 60% of the mercury was transferred to the reservoir during each of the three heating cycles, representing the percentage of the mercury that would have been lost if the distillate had not been collected. No mercury was detected in the splash head. Mercury lesses during the destruction process were followed by losses of activity of ^{203}Hg, and result are given in Table 6.

Table 6. Mercury losses during digestion of milk

Sample	Activity added counts/min	Activity recovered counts/min	Hg lost %
1	4,850	4,845	0.1
2	4,801	4,790	0.2
3	5,118	5,104	0.2
4	5,247	5,242	0.1
5	5,145	5,135	0.1

The fact that the mercury was found in an amount exceeding the tolerance in rice, quantitative the most important in our life, indicates that the rice plant uptakes a considerable amount of mercury contained in agricultural fungicides applied and may accumulate it. Experimental data on the mercury in whole rice, dehulled rice and polished rice showed that the polished rice contained 0.65 to 0.80 ppm of mercury, an amount several times higher than that of sample dehulled rice which contained only 0.09 to 0.128 ppm.

Table 7. Results from analysis of mercury in Korean grain and corn. Data on these reservoirs are essential before realistic standards can be developed for essential food supplies

Sample	μg/g
1. Rice (Paldal)	0.075
Rice (Honam)	0.250
	0.080
Edible rice	0.065
Unpolished rice	0.095
Unpolished rice	0.028
2. Barley	0.117
3. Wheat	0.215
4. Soybean	0.041
5. Pea	0.033

Table 8. Results from analysis of mercury in Korean persimmons

Sample No.	Area	ppm
1.	Kun San	0.125
2.	Chun Joo	0.110
3.	Ei Ri	0.126
4.	Kwang Joo	0.132
5.	Ann Dong	0.130
6.	Tae Ku	0.140
7.	Kyung Joo	0.125
8.	Chung Do	0.128
9.	Mil Yang	0.134
10.	Sam Rang Jin	0.131
	Average	0.125

A neutron activation study of environmental contamination and distribution of mercury in foodstuffs. Since the vegetable samples tested in the present experiment were collected from common markets the application of the said agricultural fungicides could not be verified. Nevertheless, an average of 0.123 ppm of mercury was detected. Application of agricultural fungicides to fruit trees has been a common practice ever the drugs were invented yet the amount of mercury found in various fruits marketed is sample astonishing and it certainly exceeds by 0.012-0.190 ppm that found in other country.

Table 9. Result from analyses of mercury in Korean fruits

Species of fruit	μg/g
Apple	0.085
Orange	0.130
Pear	0.140
Peach	0.130
Persimmon	0.113
Grape	0.073
Mandarin orange	0.145

Apple Species	Peel μg/g	Flesh μg/g
Golden	0.120	0.08
India	0.190	0.07
Starking	0.170	0.05
Hong Ok	0.135	0.04
Kuk Kaung	0.147	0.06

Fruit species	μg/g	μg/g
Orange	0.130	0.03
Pear	0.140	0.03
Peach	0.310	0.09

The behaviour of animals we have investigated does not indicate any ill effects attributable to mercury intoxication. Although occasional intoxications do occur among the human population, these are mainly in newcomers to the area. We tentatively suggest that there is some accommodation effect to increased levels of mercury. Further studies are in progress to collect more evidence on this question. Again, a study of the distribution of mercury in various vegetable was made, the results of which are presented in Table 10.

Table 10. Result from analysis of mercury in Korean vegetables

Sample	μg/g
Spinach	0.012
Carrot	0.035
Radish	0.39
Chinese cabbage	0.190
Kale	0.183
Potatoes	0.083

Table 11. Results from analyses of mercury in Korean soybean sprout

Sample No.	Area name	p.p.m.
1.	Myung Ryun	0.092
2.	Rak Won	0.082
3.	Dong Dai Moon	0.115
4.	Nam Dai Moon	0.068
5.	Fee Kyung Dong	0.096
6.	Jung Ang	0.126
7.	Chin Chon	0.129
8.	Chung Pa Dong	0.123
9.	Suk Kwan Dong	0.115
10.	Jeang Jeung	0.158
11.	Shin Seal Dong	0.097
12.	Won Hyo Ro	0.152
13.	In Chun	0.132
14.	Su Won	0.117
15.	Dong Du Chun	0.129
	Average	0.123

Table 12. Result from analysis of mercury in Korean egg

Sample	μg/g
Egg	0.103
Egg white	0.051~0.059
Egg yolk	0.165~0.185
Hen muscle	0.04~0.05
Lamb, liver	0.06~0.07

Another thing noteworthy is that the mercury content found in chicken, 0.05 ppm was several times higher than the amount found in meat, 0.05 ppm. In the case of egg 0.103 ppm found in yolk 0.165 ppm is considerably higher than that found in white, 0.056 ppm.

The neutron activation method used to determine the mercury content of samples, which lie between 0.1 and 200 μg/g, is described.

The distribution in organs was determined so far from only two animals, but eggs have been analysed white and yolk separately. Variations in diet, does not seem to influence the results essentially, where as concentrations appear to increase with the age of the hen.

Table 12 shows the mercury content of organs in freshly killed hens from the in Seoul.

Except for one sample a considerably higher concentration of mercury was found in kidney than in liver, the two organs with the highest burden. This exceeds 0.15 μg/g in some kidney samples. We have also looked one step backwards in the biological chain and analysed some of the food. The most striking feature is the large ratio of yolk to content in both areas and generally higher levels (up to 0.1 μg/g) in Seoul. This parallels the uptake of mercury in certain plants as discussed else-where. A sample of egg shell from each area gave values similar to that in egg white. The mercury concentrations in muscle inner organs, eggs are comparatively high. In muscle tissue and eggs, 59~189mg/g are normal values. Feathers, which contain no water, for this reason have higher mercury concentrations, about threefold that of muscle.

Discussion

Ever since the application of mercury containing agricultural fungicides and pesticides to vegetations the mercury contamination of foodstuffs as well as surroundings has become as increasing concern to human health since excessive corporal accumulation of this substance was found to be harmful. The usefulness of activation analysis is mainly strictable to its high sensitivity, but its specificity, general reliability and ability to accept virtually all types of sample materials are also essential. From this point of view toxic metals in the environment present a better-than-average opportunity for the application of nuclear techniques, and particularly for neutron activ-

ation analysis. Mercury toxicity has been recongized as a hazard in industry for several decades, the victims of the effects often being referred to as the Mad Hatters. Potential hazards to man have been hinted at in the preceding sections. The provisional highest acceptable limit for mercury in soy sprout. Further statements are expected to be issued at the end of 1970 or begining of 1971, following a detailed toxicological review.[37][38] Smith[39] reported forensic-inverstigations of mercury poisonings by activation technique in dentists. A concentration of 0.005 ppm has been considered as an operating standard for drinking water. This, as well as the basic properties of mercury, suggests that a reservoir of toxic material has been deposited in some bodies of water. The establishment of standard of 0.5 ppm in fish by the Food and Drug Administration was reported in 1970. In view of the long range nature of this problem, the essential need to trace elemental mercury from source to man and the requirement. Under idealized conditions the enrichment can be related to the intake of food, its content of mercury and the excretion kinetics by means of a simple mathematical model.[40]

The mercury levels reported here are obviously of total mercury content. We are at present introducing GLC and TLC techniques and hope soon to be also to determine this ratio for all types of sample. This possibility was rejected because of difficulty in obtaining reagents and resist of sufficiently low mercury content and in ascertaining quantitative uptake of all mercury present in the sample, especially as the chemical state of this mercury would generally be unknown. Neutron activation analysis is a very important technique for the study of problems arising in environmental the sensitivity of this method allows detection limit of a nanogram. Since the chemical separation procedure used is very specific with regard to mercury, the absolute detection limit of 0.1 to 0.3 μg of usually maintained, even when large samples are processed. Considence in the values obtained by activation analysis is, however, strengthened by the comparative studies organized and reported by Bowen[41]. The accuracy of the various versions of methods used for activation analysis of mercury and can therefore be claimed to approach their precision. Bowen rejects as erroneous one set of colorimetric analysis for each element (differing by as much as a factor of 10 from the activation analysis values). The error of the method, as defined by the coefficient of variation, is therefore 6% at most.

Comparing them with those of Swensson[42] on rats and poultry[43] injected with various mercury compounds, it appears that often the initial rapid decline due to diet switching, elimination proceeds at a similar rate to that for inorganic or phenylmercury compounds. This conclusion is supported by the pattern of distribution of mercury in the organs, which in the case of methylmercury has been found to be much more uniform.[42~45] In an experiment in which phenylmercuric-nitrite, 8-phenylmercurioxyquinoline and other organic mercury substances were sprayed to an orchard in 10 times between April and July and the apples harvested 10 to 17 weeks after the application were found to contain 0.01 to 0.07 ppm of this element miller[46] pointed out that the decrease of said substance when applied to apple tree was due to the decay and volatilization of this substance by the combined action of high temperature, abundant sun light and washing down by rain. However, in the case of rice grown on the paddy with abound water the situation differs, there will be neither volatilization of mercury nor diminution by rain even washed down by rain it will remain in the paddy and will be taken up by rice plant through the root. Therefore it can well be anticipated that rice and its byproduct will contain a substantially large amount of this element. A rather similar line of reasoning can be applied to the animal products; chicken raised and egg laid on high grain feeds containing a large amount of rice-bran should have more chance to accumulate this element than the meat of cattle raised on the pasture. Nevertheless the fact that egg yolk contained more mercury than egg white is very interesting and it must further be elucidated. The initial samples consisted of hens' eggs and cows' milk were also analyses at different periods of the year. Residual mercury in various organs was finally determined and compared egg chicken from the original area analysed immediately. The mercury content of various organs of hens taken from the in Seoul was also determined. Finally comparison is made with work previously published on the content and accumulation of mercury in animals some tentative

conclusions are drawn on the possible ability of certain species to accommodate to considerably increased levels. The excellent trace element studies which have been made on hair, mainly by activation analysis, for forensic purpose are of great value in establishing normal values. Material from more than 750 different persons was studies with respect to the content of mereury by Perkons.[47] The amount of mercury found in the soybean sprout is not surprising since certain commercial producers apply a considerable amount of mercury containing fungicides in cleaning the vessels in which soybean is sprouted but is alarming because a very large quantity of this sprout is consumed throughout the year and particularly in the winter when green vegetables are relatively expensive. While the contribution of mercury containing pesticides is important in the accumulation of this element in the environmental.

The gradual increase in environmental pollution makes it important to study the natural abundance and regional distribution of chemical elements and compounds that can be considered as potential contaminates.

초 록

유기수은제 농약은 도열병 방제의 목적으로 다량 살포되는데 그 결과로 식품중의 수은잔류량증가로 그 피해가 예견되어 수은함량을 검색할 필요성이 요구된다.

그러나 종래 분석방법인 dithizone 비색법은 시료 분해시 수은 화합물의 손실, 유출율의 변동으로 인하여 정확도를 기대하기 어려운점이 있으나 본 연구에서 시도한 방사화 분석법에 의하면 극미량도 고감도로 정량한 수가 있다.

본 실험에서는 곡류, 야채, 육류, 과일, 계란을 산지별로 시료를 수집하여 건조, 포장하여 vial에 넣고 열중성자속 3.8×10^{12} n/cm sec에 15시간 조사하였다.

이 시료를 Bethge 장치로 분해시켜 수은을 증류하여 TMC 100 Channel pluse height analyzer로 ^{203}Hg 방사능을 0.279 MeV Photopeck로 측정하였다.

시료별로 수은 함량은 곡류 0.033~0.250 ppm, 야채 0.012 ~ 0.190 ppm, 닭고기 0.04 ~ 0.07 ppm, 과일 0.085~0.145 ppm, 계란 0.051~0.165 ppm, 콩나물 0.123 ppm 됨을 알게 되었다.

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