

## References and Notes

1. D. Q. Wark and D. T. Hilleary, *Science* **165**, 1256 (1969).
2. D. Q. Wark, *Appl. Opt.* **9**, 1761 (1970).
3. The measured radiance is represented by the integral form of the radiative transfer equation (1). By taking the horizontal ascendent of this equation and combining the result with the thermal wind equation, it can be shown that the horizontal radiance gradient measured by channel 4 of the SIRS reflects the difference between two weighted mean winds, one centered near 12 km, the other centered near 6 km. Details will be included in J. A. Woods and H. A. Panofsky, in the Proceedings of the 1972 IUCRM Colloquium on Waves and Turbulence in Stratified Layers and Their Effects on EM Propagation, 6-15 June, San Diego, to be published in *Boundary Layer Meteorology*, in press.
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5. I thank Dr. H. A. Panofsky for advice and guidance, and also Dr. D. Q. Wark of the Satellite Experiment Laboratory, A. Timchalk of the National Environmental Satellite Service, P. E. Kraght of American Airlines, J. Gardner of Trans World Airlines, and Lt. D. Trout for their assistance. I thank Dr. J. Winston and Dr. H. Tennekes for their careful reading and helpful criticism of the manuscript. Supported by Department of Commerce grant SAT 11 4939.

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## Mercury Content of Common Foods Determined by Neutron Activation Analysis

**Abstract.** *The mercury contents in samples of flour, sugar, nonfat dry milk, potatoes, hamburger, chicken breast, shrimp, liver, eggs, and whole milk were determined by neutron activation analysis. The mercury was separated by anion exchange chromatography and precipitated as the sulfide. The mercury concentrations for all these foods were below 50 parts per billion.*

Data accumulated by the U.S. Food and Drug Administration and other agencies have confirmed that fish can contribute mercury to the diet. Fish, however, represents only a small portion of the normal dietary components of the American public. Data reported by Jervis *et al.* (1) indicate that the occurrence in common foods of mercury at significant concentrations may be widespread. We therefore set out to determine if other foods besides fish contain significant amounts of mercury. The foods selected were those specific foods or food groups that have a high total nationwide consumption. We made exceptions to this in including liver and shrimp because these foods might be expected to be accumulators of mercury from their respective en-

vironments and therefore be useful secondary indicators of mercury. Representative nationwide samples of the following types of food were collected for analysis: flour (wheat); milk (nonfat dry and whole); sugar (cane); potatoes (white, raw, unpeeled); beef (raw, ground hamburger); chicken (raw, boned breast); shrimp (frozen, peeled, deveined); liver (beef); and eggs (shelled).

To avoid contamination, food samples were shipped in glass jars and handled in a "clean room" until they had been sealed into quartz irradiation vials. The contents of the jars were thoroughly mixed before shipment and before any sampling was done. Food samples which had a water content of more than 15 percent were freeze-dried in

order to lessen any pressure increase when the sample, enclosed in a sealed quartz vial, was exposed to the neutron flux. In practice, a sample of 1 to 10 g was withdrawn with a stainless steel spatula, transferred to a 25-ml Erlenmeyer flask, frozen in liquid nitrogen, and freeze-dried in a freeze-dry apparatus (VirTis). After the sample had reached ambient temperature it was kept under vacuum for two additional hours. The freeze-dried sample was finely pulverized, transferred to a small polyethylene vial, and placed in a desiccator.

For each analysis, approximately 200 mg of a dry food sample was accurately weighed directly into a clean quartz vial, which was then sealed with an oxygen-methane torch. The mercury standard consisted of 12.5  $\mu\text{g}$  of mercury (in the form of mercuric acetate in 1M acetic acid) which was adsorbed onto about 30 mg of powdered silicon dioxide and sealed in a quartz vial. The standards and samples were packaged and irradiated in a neutron flux of approximately  $6 \times 10^{13}$  neutron  $\text{cm}^{-2}$   $\text{sec}^{-1}$  for 4 hours in the 10-Mw research reactor at the National Bureau of Standards (NBS), Gaithersburg, Maryland. In order to avoid exposing the personnel to the radiation and to allow short-lived radionuclides some time to decay, the samples and standards were processed approximately 1 week later. The quartz vials were washed with aqua regia after irradiation to remove any contamination from the outside of the quartz vials, an important cause in the early phases of this work of very high and quite variable results. The importance of avoiding contamination cannot be overemphasized for mercury measurements at the parts per billion (ppb) level.

Because the mercury content of most of these samples was quite low, a chemical separation was needed to enhance the 279-keV gamma-ray peak of  $^{203}\text{Hg}$  with respect to the background. Mercury was separated by a procedure developed by Jervis *et al.* (1) involving anion exchange chromatography and sulfide precipitation. An alternative electrodeposition procedure of Sjöstrand (2) was found to give equivalent results at all concentrations of mercury. However, for the data reported here we used the procedure of Jervis *et al.* (1) because it was readily adaptable to processing a large number of samples and did not require any

Table 1. Analyses of mercury standards and of samples examined in several laboratories.

| Material   | Mercury (parts per million) |                                      | Reference   |
|--|-----------------------------|--------------------------------------|-------------|
|  | This work                   | Other work                           |             |
| NBS orchard leaves*                                | $0.148 \pm 0.010$           | $0.155 \pm 0.006$<br>$.162 \pm .010$ | (7)<br>(8)  |
| International Atomic Energy Agency standard flour* | $4.6 \pm .5$                | $4.87 \pm .06$<br>$4.9 \pm .3$       | (7)<br>(9)  |
| Food and Drug Directorate flour No. 32573          | $0.011 \pm .003$            | $0.011 \pm .004$<br>.007 to .02      | (7)<br>(10) |
| Swedish fish No. 410-30                            | $1.29 \pm .13$              | 1.14<br>1.17                         | (11)<br>(8) |
| Swedish fish No. 410-28                            | $2.16 \pm .22$              | 2.24<br>2.20                         | (8)<br>(11) |
| Bowen kale*  | $0.25 \pm .03$              | 0.23                                 | (9)         |

\* Standard material.

special equipment. Carrier mercury was used to determine the chemical yield, which averaged about 90 percent. Complete equilibrium was achieved by allowing the sample and carrier to digest together overnight in fuming nitric acid.

The samples of separated mercury were counted in a 7.6 by 7.6 cm (3 inch by 3 inch) NaI(Tl) well detector (Harschaw) connected to a multichannel pulse-height analyzer (Nuclear Data model 2200). Each sample was counted for 800 seconds and each standard for 80 seconds. The 279-keV  $^{203}\text{Hg}$  gamma-ray peak was used for the analyses. The samples were checked regularly for purity with a high-resolution Ge(Li) detector (Ortec) (resolution, 2.3 keV at 1.33 MeV; efficiency, 10.8 percent). No interfering peaks were detected in the 279-keV region.

Since the  $^{203}\text{Hg}$  gamma-ray peak is much less pronounced at low concentrations of Hg (about 1 ppb), a good method for subtracting the background must be available. To find the net peak area, 24 channels centered on the peak were summed, and then the background, determined by summing 12 additional channels on either wing of the peak, was subtracted. In cases where the peak was just slightly higher than the background, this method of background subtraction gave substantially smaller peak area deviation than background subtraction methods which involve fewer data points. We tried other methods of background subtraction but we found these to be either equivalent to this method or to give variable results. The concentration of mercury was calculated from both  $^{203}\text{Hg}$  and the shorter-lived  $^{197}\text{Hg}$  ( $t_{1/2} = 65$  hours); the results in each case were basically the same. As time went on, the interval between the end of irradiation and the counting time became long enough so that only the longer-lived  $^{203}\text{Hg}$  remained. In addition,  $^{203}\text{Hg}$  was considered a better indicator nuclide because of the possible contribution of x-rays in the 0.077-MeV region, the danger from radiation to personnel of working with samples which had been out of the reactor only a short time, and the poor resolution of the NaI(Tl) detector in the x-ray region.

Several standards and collaboratively studied materials were analyzed during the course of this work to check the validity of the procedure (Table 1). The mercury separation procedure (1) used

Table 2. Concentrations of mercury in common foods. All results are given for material as received.

| Food        | Samples analyzed (No.) | Minimum detectable amount of mercury (ppb) | Number of samples with mercury content below the detection limit | Mercury (ppb) |        |
|-------------|------------------------|--|--|---------------|--------|
|             |                        |  |  | Range         | Median |
| Flour*      | 28                     | 3  | 25   | < 3-6         | < 3    |
| Milk, dry*  | 33                     | 4  | 13   | < 4-27        | 10     |
| Milk, whole | 32                     | 1  | 23   | < 1-9         | < 1    |
| Sugar*      | 22                     | 3  | 17   | < 3-10        | < 3    |
| Potatoes    | 33                     | 1  | 14   | < 1-15        | 3      |
| Beef        | 23                     | 2  | 9  | < 2-7         | 3      |
| Chicken     | 24                     | 1  | 9  | < 1-7         | 3      |
| Shrimp      | 32                     | 2  | 0  | 5-43          | 14     |
| Liver       | 22                     | 2  | 11   | < 2-8         | 3      |
| Eggs        | 33                     | 2  | 30   | < 2-5         | < 2    |

\* Not freeze-dried.

here gave consistently reliable results over a wide range of mercury concentrations (3). For brevity, only the most recent results from other laboratories are listed.

Pillay *et al.* (4) have recently reported that mercury is lost when biological samples are freeze-dried. However, in view of the work by Bate (5), it is uncertain whether the mercury is lost during freeze-drying or by escaping through the polyethylene sample vial during reactor irradiation. Using methylmercury, phenylmercury, and inorganic mercury, LaFleur (6) has shown that for rat and guinea pig tissues the mean mercury loss during freeze-drying is less than 3 percent. An extrapolation of these results to the foods used here (for example, experimental animal tissues and hamburger containing mercury would probably lose mercury to the same extent when freeze-dried) would indicate that losses of mercury during freeze-drying were not a problem in this work. The close agreement between our results on freeze-dried portions of Swedish fish and the results from other work, in which the fish was not freeze-dried, shows that mercury was not lost from fish under our experimental conditions (Table 1). In addition, we analyzed flour samples containing known amounts of mercury before and after freeze-drying; the results in each case were equivalent.

A summary of the results of this survey is shown in Table 2. Approximately 30 samples of each food were analyzed for mercury; these samples from all areas of the United States constituted a representative cross section for the mercury content of such basic

foods. None exceeded 50 ppb. With the exception of certain fish, the major foods in the United States are essentially free of mercury.

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12. We thank the personnel of the District Laboratories of the Food and Drug Administration for collecting the samples and R. E. Simpson, Food and Drug Administration, Washington, D.C., and H. Yule, P. D. LaFleur, and the personnel of the Reactor Radiation Division of the National Bureau of Standards for their contributions to various phases of this work. Mention of commercial products does not imply endorsement by the Food and Drug Administration.

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