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CONCENTRATIONS OF RADIONUCLIDES AND
SELECTED STABLE ELEMENTS IN FRUITS AND VEGETABLES*

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ABSTRACT

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Twenty-two types of fruits and vegetables collected from two commercial supermarkets have been analyzed for their radionuclidic and stable-element composition. A specific gamma-emitting isotope analysis was performed on each sample for ^{40}K , ^{60}Co , $^{95}\text{Zr-Nb}$, ^{106}Ru , ^{137}Cs , ^{226}Ra , and ^{232}Th . The concentration of the stable elements in each sample were determined using multi-element neutron-activation analysis (Al, Ag, Au, As, Ba, Br, Ca, Ce, Cl, Co, Cr, Cs, Cu, Eu, Fe, Hf, I, K, La, Mn, Mo, Mg, Na, Rb, Sb, Sc, Se, Sr, Ta, Te, Th, Ti, Zn, Zr) and atomic absorption (Cd, Ni, Pb). Information on the composition of a typical diet is used to estimate the radiological dose to man subsequent to ingestion of these fruits and vegetables. The stable-elemental compositions of the foodstuffs analyzed were compared with estimated values assuming foliar deposition and long-term buildup of effluents from a large modern coal-fired steam plant. It is tentatively concluded that for the general case of a precipitator-equipped, coal-fired steam plant, no toxic levels of trace elements in foodstuffs are expected as a result of the plant operation.

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INTRODUCTION

A joint program at the Oak Ridge National Laboratory (ORNL) between the Environmental Surveillance and Evaluation Section and the Health Effects Study Group has been undertaken to quantify one of the major sources of radiation exposure and potential exposure to toxic materials - that is, food. It is necessary that exposures to these materials be determined so that a proper perspective of the relative risks due to materials released from the Oak Ridge facilities can be developed. Such knowledge will be of invaluable aid to epidemiological studies designed to investigate possible biological effects due to low-level radiation. Naturally, for a comprehensive study, in addition to foodstuffs, other components of the radiation background, such as cosmic rays, external background from soils and building materials and airborne radon must be included. Studies to determine these sources are presently under way (16).

Natural radioactive materials in the environment are the major sources of radiation exposures to man. The two principal modes of entry into man are ingestion of foodstuffs and inhalation, with ingestion of foodstuffs playing the major role in contributing to radiation exposures (10, 14). For example, potassium-40, the principal naturally-occurring source of internal radiation, enters man mainly through foodstuffs (5). Other natural radioelements include thorium, uranium, and a few less abundant isotopes. In addition to the natural radioelements, synthetic radioelements have been added to the environment since the first nuclear detonation in 1945 up to as recently as September, 1976 (4).

Two sources also exist for potentially hazardous stable elements. Trace elements exist naturally at some level in all soils and waters, and some are present at extreme levels at highly localized sites. These trace elements can also be deposited from the air, once released from a source (e.g., coal combustion). Most trace elements which are deposited on the ground surface do not become evenly distributed through the soil, but are concentrated near the surface and are quite resistant to leaching to lower depths except in sandy or very acid soils. Thus, while they may not be a problem to the underground water supply in most areas, they may pose difficulties for the case of long-term-deposition-caused buildup in surface soils. This could be so, since many members of food chains (which feed on surface soils or on plants grown thereon) have poor discrimination factors for some elements and others have the ability to concentrate certain elements. Although substantial information exists in the agronomic literature on the uptake and metabolism of certain trace elements for some crops, this is not true for many elements found in atmospheric discharges from coal-combustion facilities. Inasmuch as high and low levels of trace materials are closely connected with numerous kinds of health disorders (although many have beneficial effects at optimum levels), it is clearly appropriate to monitor perhaps the most important pathway of trace elements to man, i.e., the food chain. Hence, as the beginning of a larger program, a survey was initiated to determine the trace and radioelement compositions of foodstuffs available at the local market in the Oak Ridge area.

MATERIALS AND METHODS

Several pieces of a variety of fruits and vegetables were purchased from two of the major supermarkets in the Oak Ridge area. Tomatoes,

eggplant, peppers, yellow squash, cabbage, potatoes, lettuce, green beans, turnips, carrots, celery, asparagus, cucumbers, broccoli, apples, bananas, tangelos, oranges, tangerines, pears, grapefruits, and raisins were chosen for analysis. The edible parts of the samples were placed in preweighed beakers, weighed, and covered with aluminum foil until dry ashed. The samples were put in a muffle furnace for 2 hours at 100°C and heated at 150°C for approximately 16 hours. Then the furnace temperature was raised in 50° increments, every hour, until 325°C was reached. At this point, the sample was allowed to ash for 16 to 24 hours at 325°C. The furnace temperature was then raised to 375°C for 1 hour and finally raised to 425°C for at least 24 hours. The cooled dry-ash sample was transferred to a preweighed polyethylene bottle, and the weight of the dry ash was determined.

A specific gamma-emitting isotope analysis was performed on each sample for ^{40}K , ^{60}Co , $^{95}\text{Zr-Nb}$, ^{106}Ru , ^{137}Cs , ^{232}Th , and ^{226}Ra using a 6 x 6-in. sodium-iodide (thallium) crystal. The detector output was then analyzed using a Nuclear Data Model 3300 multichannel analyzer. The Alpha-M computer program (17) was used to determine radioisotope concentration by means of a least-squares resolution of their gamma-ray spectra. Alpha-M takes into account corrections, such as counting time, decay time, background subtraction, sample size, dead time, and automatic compensation for gain and threshold shifts. The minimum sensitivities are those concentration values below which it can not be stated, at the 99.7% confidence level, that any amount of radioactivity above background is present in the sample. Concentrations with errors larger than the reported concentrations are given as "not detectable" (ND).

The concentration of the stable elements in each sample were determined using multi-element neutron activation analysis (Al, Ag, As, Au, Ba, Br, Ca, Ce, Cl, Co, Cr, Cs, Cu, Eu, Fe, Hf, I, K, La, Mo, Mg, Mn, Na, Rb, Sb, Sc, Se, Sr, Ta, Te, Th, Ti, Zn, Zr). After homogenization and drying, a portion of the sample was weighed and sealed inside a plastic vial. The samples were irradiated in the Oak Ridge Research Reactor for a period varying from a few seconds to 30 minutes. Counting was performed at various decay times using a Ge(Li) detector and a Nuclear Data PDP-15 analyzer computer system (15).

Concentrations of Cd, Ni and Pb in each sample were determined by flameless atomic absorption spectroscopy using a Perkin-Elmer Model 503 Spectrophotometer with a Model 2100 Heated Graphite Atomizer System (19).

RESULTS AND DISCUSSION

Radiological Content

The results of the radionuclide content in the combined samples of fruits and vegetables from the commercial stores are presented in Table 1. Most of the data were reported from the laboratory analysis as "less than or equal to" numbers, except ^{40}K with all the numbers being absolute. The ^{40}K values range from 0.56 (for apples) to 6.1 pCi/g (for raisins). Our ^{40}K values were noted to be in good agreement with the literature. Klement (9) states that Burton et al. (2) found ^{40}K concentrations of 3.5 pCi/g for bananas; 1.4 pCi/g for oranges; 3.4 pCi/g for carrots; 3.4 pCi/g for potatoes; 1.9 pCi/g for cabbage; 1.2 pCi/g for lettuce; and 1.9 pCi/g for tomatoes. Our respective values for these same foodstuffs are: 2.3 pCi/g, 1.5 pCi/g, 2.6 pCi/g, 3.3 pCi/g, 2.4 pCi/g, 1.3 pCi/g, and 1.8 pCi/g. Also Chhabra and Hukkoo (3) analyzed food in India and found ^{40}K concentrations of 2.7 pCi/g

in carrots, 3.5 pCi/g in potatoes, and 1.8 pCi/g for cabbage. These values are also in excellent agreement with our data. Klement (9) states that ^{40}K is relatively uniformly distributed and that the levels in foods and water do not vary appreciably with agricultural practice.

Radium-226 is another natural radionuclide of great abundance. In comparing values, we find that Hallden and Fisenne (6) found 2.0×10^{-3} pCi of ^{226}Ra /g of potatoes, and Muth et al. (13) observed ^{226}Ra values of 1.0×10^{-3} pCi/g in potatoes. Both are in good agreement with our value of 3.9×10^{-3} pCi/g. Muth et al. (13) also noted the ^{226}Ra in carrots varied from $(1.6-6.1) \times 10^{-3}$ pCi/g and cabbage from $(1.0-2.4) \times 10^{-3}$ pCi/g. Both of these values are a factor of 10 less than our data indicated.

Most of the man-made radionuclides (i.e., ^{60}Co , ^{95}Zr , ^{106}Ru , ^{125}Sb , and ^{137}Cs) were found in only extremely small amounts. Most of the data in the literature is reported as non-detectable for these nuclides. Chhabra and Hukkoo (3), however, report 1.5×10^{-3} pCi of ^{95}Zr /g of carrots, and this corresponds exactly with our value.

Radiological-Dose Commitment

The radiological-dose commitment from consumption of the fresh fruits and vegetables are presented in Table 2. All dose values in Table 2 were calculated using data from the United States Department of Agriculture (21) on the g/yr person for each of the particular fruits and vegetables; these are listed in the first column of Table 2. The dose-conversion factors used were the latest values obtainable (7, 12, 20).

Radium-226 is the largest critical-organ dose contribution; this is due to its high conversion factor for bone. It should be remembered, however, that most of the ^{226}Ra values in Table 1 are "less than or equal to" values

implying that the dose values in Table 2 should be taken as maximum values. Again, the man-made gamma emitters analyzed contributed insignificantly to dose in comparison to the natural radionuclides. The total maximum ^{40}K whole-body dose from consumption of all the fruits and vegetables was 3.6 mrem/yr. The other radionuclides contributed at the maximum 13.4 mrem/yr from consumption of all the fruits and vegetables.

Stable Elements

Values of elemental concentrations in foods (Tables 3 and 4) were compared to available literature data. The more common essential elements Ca, I, Mg, and Na were measured to be somewhat greater, in the order of 10 times, than typical literature values while the more common contaminant elements Cd, Ni, V, and Pb were found to be nearly 10 times lower than typical values. The majority of elements were found to be present within the range of literature values; these include Al, As, Ba, Co, Cr, Cs, Cu, Fe, K, Mo, Mn, Rb, Se, Sr, Te, Th, Ti, Zn, and Zr. The remaining elements, Ag, Au, Br, Ce, Cl, Eu, Hf, La, Sb, Sc, and Ta were either not found reported for other than total diets or not at all. It is clear from the data that none of the elements are approaching toxic levels in the foods examined and that enormous quantities would be required to enter the soil prior to the plant's approaching levels supportive of chronic toxic intakes.

Since the elements listed in Tables 3 and 4 are virtually identical to those listed by Klein et al. (8) as trace element discharges from coal combustion for power production, it is instructive to determine whether or not one could expect to find large scale contamination of foods by deposition of these discharges onto agricultural soil. According to Bolton et al. (1), the annual average dilution factor is a minimum at 5 km

from a 122 m stack and is near 1.6×10^{-9} sec/cm³. For a 1 g/sec release, a 1 cm/sec deposition velocity, and assuming no washout, the ground concentration is added at 5×10^{-4} g/m²-yr. If crop plants are assumed to take in their trace elements from the first 10 cm of soil, the soil concentration of "contaminants" increases by 3.2 ppb/yr/g/sec of released material. The discharge rates of the elements reported for the 870 MWe Allen Steam Plant^{**} (8) were compared to average soil concentration near that plant (1) and to world average figures (11, 22). At the reported rates (8), selenium, mercury, lead, and uranium are discharged at the highest rates relative to their soil concentrations. For the above-stated conditions, it would take some 5,000 years of continuous discharge to double the soil concentrations of selenium and mercury; for the majority of other elements, the time would be greater by 10 to 10³. This calculation is for the point where the plume intersects the ground 5 km from the source (1), and the build-up rate would be reduced by several orders of magnitude with increasing distance from the source, although for some locations closer, the rate may be slightly higher. The inclusion of washout might raise the concentration levels near the source (and hence lower the doubling time) by up to a factor of 5 or so.

Most fruits and vegetables have lower concentrations of trace elements than the soil on which they grow, usually by a factor of 10 to 100. This is true for selenium, mercury, lead, and uranium (except for selenium in selected foodstuffs) (18), and it is clear from Tables 3 and 4 that the fruits and vegetables have extremely low levels of potentially toxic elements.

^{**} The Allen Steam Plant in Memphis, Tennessee has a peak capacity of 290 MWe from each of three-cyclone-fed boilers. Ash produced in boilers is distributed about equally between slag and fly ash, in contrast to the more common pulverized coal boilers, where as much as 90% of the total ash is fly ash. The study (8) was conducted on one of the three units on which a new high efficiency electrostatic precipitator was recently installed.

While the soils where the foods were grown are unknown and were not analyzed, they certainly represent a wide variation in soil types, and yet, most foods show quite similar levels for a given element. Hence, for the general situation, releases from coal-fired plants would not be expected to appreciably raise levels of trace elements in foods as a result of uptake from the soil.

Uptake through foliar absorption is another method by which plants can accumulate materials released to the atmosphere. If fruit and vegetable production over a four-month period is roughly taken to be 1 kg/m^2 of crop plant, half of which is food bearing and 20% of the materials settling on the foliage is assumed to be incorporated into the food parts, then elemental enrichment of fruits and vegetables relative to the values in Tables 3 and 4 arising from releases similar to those from the Allen Steam Plant (8) (using the assumptions above) would occur only for aluminum, antimony, and scandium. These increased levels would be in the order of those in Tables 3 and 4 would perhaps double the values, causing essentially no change as to the toxic potential of the foods analyzed. Arsenic and iron could possibly be present at the extreme lower levels found in Table 3 and the remainder would be substantially lower.

SUMMARY

This paper is the first-phase of a long-term trace-element study being conducted at Oak Ridge National Laboratory, and data is presented on fresh fruits and vegetables, which comprise approximately 14% of a typical Southern diet. In the study, it was observed that many elements could be analyzed accurately through neutron-activation techniques; thus, providing data that is otherwise not available in the literature. Potassium-40, as expected, was

the radionuclide of highest concentration in all of the fruits and vegetables. Foodstuffs were shown to not accumulate toxic levels of trace materials as a results of exposure to the stack effluents of a large modern coal-fired steam plant. The baseline data presented can be used as a basis for comparison with similar foodstuffs grown in the vicinity of nuclear and nonnuclear energy facilities.

LITERATURE CITED

1. Bolten, N. E. *et al.* 1974. *Trace Element Measurements at the Coal-Fired Allen Steam Plant - Progress Report February 1973 - July 1973*. ORNL-NSF-EP-62.
2. Burton, B. T. *et al.* 1958. *Nutritional Data*, H. J. Heinz Co., Heinz Nutritional Research Inst., Melton Inst., Pittsburgh.
3. Chhabra, A. S., and R. K. Hukkoo. 1962. Natural and fallout γ -activity in foodstuffs, *Health Phys.* 7:151-156.
4. Eldridge, J. S., and T. W. Oakes. 1977. Environmental surveillance for radionuclide contamination utilizing high-resolution gamma-ray spectroscopy, in *Proceedings of the Eleventh Annual Conference on Trace Substances in Environmental Health*, University of Missouri, Columbia, Missouri.
5. Garner, John R. 1972. *Transfer of Radioactive Materials from the Terrestrial Environment to Animals and Man*, Chemical Rubber Co., Cleveland, Ohio.
6. Hallden, H. A., and I. M. Fisenne. 1961. Radium-226 in the diet in three U.S. cities, in *Fallout Program Quarterly Summary Report*. E. P. Hardy, Jr. *et al.* (eds.). HASL-113.
7. International Commission on Radiological Commission. 1960. Report of Committee II on Permissible Dose for International Radiation (1959), *Health Phys.* 3:1-380.
8. Klein, D. H. *et al.* 1975. Pathways of thirty-seven trace elements through coal-fired power plant, *Env. Sc. Tech.* 9:973-979.
9. Klement, A. W. 1965. Natural radionuclides in foods and food source materials, in *Radioactive Fallout, Soils, Plants, Foods, Man*, E. B. Fowler (ed.), Elsevier Publishing Co., New York.
10. Klement, A. W., C. R. Miller, R. P. Minx, and B. Shleien. 1972. *Estimates of Ionizing Radiation Doses in the United States 1960-2000*. ORP/CSD 72-1
11. Lisk, D. J. 1972. Trace metals in soils, plants, and animals, *Advan. Agron.* 24:267-325.
12. Medical Physics and Internal Dosimetry Section, 1977. Oak Ridge National Laboratory, Personal Communication.
13. Muth, H., B. Rajewsky, H. J. Hantke, and K. Aurand. 1960. The normal radium content and the $^{226}\text{Ra}/\text{Ca}$ ratio of various foods, drinking water and different organs and tissues of the human body, *Health Phys.* 2:239-245.

14. National Council on Radiation Protection and Measurements. 1975. *Natural Background Radiation in the United States*, NCRP Report No. 45.
15. Northcutt, K. J. 1977. Oak Ridge National Laboratory, Personal Communication.
16. Oakes, T. W., K. E. Shank, and C. E. Easterly, 1976. Natural and man-made radionuclide concentrations in Tennessee soils, in *Proceedings of the Tenth Midyear Topical Symposium of the Health Physics Society*, Saratoga Springs, New York.
17. Schonfeld, E. 1966. *Alpha-M an Improved Computer Program for Determining Radioisotopes by Least-Squares Resolution of the Gamma-Ray Spectra*, ORNL-3975.
18. Schroeder, H. A., D. V. Frost, and J. J. Balassa. 1967. Essential trace metals in man: selenium, *J. Chron. Dis.* 23:227-243.
19. Turner, R. R., and S. E. Lindberg. 1977. *Interlaboratory Comparison of Trace Metal Analyses by Graphite Furnace (Flameless) Atomic Absorption Spectroscopy*, ORNL-TM-5422.
20. United States Atomic Energy Commission. 1974. *Environmental Statement of Liquid Metal Fast Breeder Reactor Program*. WASH-1535.
21. United States Department of Agriculture. 1968. Food consumption of households in the south, Spring 1965, in *Household Food Consumption Survey 1964-1966*, U. S. Government Printing Office, Washington, D. C.
22. Vinogradov, A. P. 1959. *The Geochemistry of Rare and Dispersed Chemical Elements in Soils*, Chapman and Hall, London.

Table 1. Radionuclide content in fresh fruits and vegetables

(pCi/g)

	$^{40}\text{K}^a$	^{60}Co	$^{95}\text{Zr-Nb}$	^{106}Ru	^{125}Sb	^{137}Cs	^{226}Ra	^{232}Th
FRUITS								
Apples (Golden Delicious)	5.6(-1) ^b	7.1(-3)	2.4(-3)	3.5(-2) ^{a,c}	9.8(-3)	9.0(-3)	1.1(-2)	6.9(-3)
Apples (Red Delicious)	1.0	4.8(-3)	1.2(-3)	1.9(-2)	6.0(-3)	4.2(-3)	8.9(-3)	4.3(-3)
Apples (Winesap)	7.7(-1)	2.3(-3)	7.9(-4)	1.3(-2)	4.1(-3)	5.4(-3) ^a	3.9(-3)	2.8(-3)
Bananas	2.3	1.0(-2)	2.5(-3)	2.9(-2)	8.3(-3)	9.6(-3)	1.3(-2)	8.2(-3)
Grapes ^c	9.3(-1)	6.4(-3)	1.7(-3)	3.4(-2)	9.5(-3)	1.2(-2)	2.1(-2) ^a	9.8(-3)
Oranges	1.5	5.4(-3)	9.6(-4)	2.0(-2)	6.4(-3)	4.2(-3)	1.2(-2)	4.1(-3)
Pears	2.3(-1)	6.5(-3)	1.6(-3)	3.2(-2)	1.3(-2)	7.1(-3)	9.9(-3)	8.5(-3)
Raisins ^c	6.1	1.4(-2)	1.4(-2)	5.5(-2)	1.4(-2)	1.2(-2)	1.9(-2)	1.2(-2)
Watermelons	7.1(-1)	4.5(-3)	1.1(-2)	2.3(-2)	4.7(-3)	5.2(-3)	1.3(-2)	4.7(-3) ^{a,c}
Watermelons	1.2	4.8(-3)	1.2(-3)	1.1(-2)	2.8(-3)	8.1(-3) ^a	5.8(-3)	2.3(-3)
Tomatoes	1.8	6.5(-3)	2.1(-3)	3.1(-2)	7.3(-3)	6.4(-3)	1.4(-2)	1.1(-2)
VEGETABLES								
Asparagus ^c	2.2	8.6(-3)	5.1(-3)	3.0(-2)	8.9(-3)	7.8(-3)	2.1(-2)	9.8(-3)
Broccoli ^c	2.8	4.8(-3)	9.5(-3)	1.5(-1)	8.8(-3)	3.5(-3)	1.6(-2) ^a	3.6(-3)
Cabbage	2.4	5.2(-3)	1.5(-3)	2.0(-2)	5.3(-3)	1.8(-3)	1.2(-2)	3.2(-3)
Carrots	2.6	5.5(-3)	1.5(-3)	3.0(-2)	7.4(-3)	5.9(-3)	1.9(-2) ^{a,c}	4.2(-3)
Celery	2.2	7.1(-3)	1.9(-3)	3.0(-2)	7.1(-3)	6.9(-3)	1.9(-2)	9.1(-3)
Cucumbers ^c	1.5	5.5(-3)	1.1(-3)	1.2(-2)	3.7(-3)	2.1(-3)	6.5(-3)	2.9(-3)
Egg Plant	1.8	4.2(-3)	1.2(-3)	1.5(-2)	4.0(-3)	4.2(-3)	1.1(-2) ^{a,c}	3.3(-3)
Green Beans	1.7	6.8(-3)	2.9(-3)	2.0(-2)	6.5(-2)	7.0(-3)	1.0(-2)	4.9(-3)
Lettuce ^c	1.3	3.7(-3)	1.1(-3)	1.8(-2)	4.4(-3)	4.0(-3)	7.2(-3)	2.8(-3)
Peppers (Bell)	1.7	7.9(-3)	2.3(-3)	2.5(-1)	9.4(-3)	9.2(-3)	2.1(-2)	6.7(-3)
Potatoes (Irish)	3.3	9.8(-3)	1.5(-3)	2.8(-2)	6.8(-3)	6.3(-3)	3.9(-3)	3.9(-3)
Potatoes (Sweet)	2.9	7.2(-3)	1.9(-3)	1.4(-1)	2.0(-2)	8.1(-3)	1.3(-2) ^{a,c}	7.5(-3)
Pumpkins	1.5	3.3(-3)	5.0(-3)	2.2(-2)	6.3(-3)	4.3(-2)	1.1(-2)	2.6(-3)
Yellow Squash	2.1	6.9(-3)	1.6(-3)	2.4(-2)	2.3(-2)	3.7(-3)	1.4(-2)	3.9(-3)

^a Absolute number, others are less than or equal to.^b Values in parentheses are powers of ten.^c Only one sample.

Table 2. Radiological-dose commitment from consumption of fresh fruits and vegetables

(mrem/year)

Critical-Organ Dose

	g/yr	(% of diet)	⁴⁰ K	⁶⁰ Co	⁹⁵ Zr-Nb	Radionuclide ¹⁰⁶ Ru	¹²⁵ Sb	¹³⁷ Cs	²²⁶ Ra	²³² Th	Whole-Body Dose from ⁴⁰ K
			Muscle	G. I.	G. I.	Critical Organ G. I.	G. I.	Liver	Bone	Bone	
FRUITS											
Apples ^a	7,978	(1.2)	1.9(-1)	1.7(-3)	4.5(-4)	4.0(-2)	1.1(-3)	6.0(-3)	9.5	7.9(-2)	1.0(-1)
Bananas	9,782	(1.5)	7.0(-1)	3.8(-3)	7.8(-4)	5.5(-2)	1.6(-3)	1.1(-2)	1.8(+1)	1.5(-1)	3.9(-1)
Grapefruit	4,926	(0.74)	1.4(-1)	1.2(-3)	2.7(-4)	3.2(-2)	1.4(-3)	6.5(-3)	1.9(+1)	1.2(-1)	7.7(-2)
Oranges ^a	7,215	(1.1)	3.4(-1)	1.5(-3)	2.1(-4)	2.8(-2)	8.7(-4)	3.3(-3)	1.2(+1)	5.6(-2)	1.9(-1)
Pears	347	(0.05)	8.9(-3)	8.2(-5)	1.8(-5)	1.2(-3)	8.5(-5)	2.7(-4)	4.7(-1)	5.6(-3)	4.9(-3)
Raisins	277	(0.04)	5.2(-2)	1.5(-4)	1.2(-4)	3.0(-3)	7.5(-5)	3.6(-4)	7.5(-1)	6.1(-3)	2.8(-2)
Tangerines+ Tangelos	208	(0.03)	6.2(-3)	3.8(-5)	7.7(-6)	6.8(-4)	1.5(-5)	1.5(-4)	2.7(-1)	1.4(-3)	3.4(-3)
Tomatoes	9,019	(1.3)	5.0(-1)	2.1(-3)	4.1(-4)	4.1(-2)	1.3(-3)	5.3(-3)	1.9(+1)	9.0(-2)	2.2(-1)
VEGETABLES											
Asparagus ^b	347	(0.05)	2.4(-2)	1.1(-4)	5.6(-5)	2.0(-3)	5.9(-5)	3.0(-4)	1.0	6.2(-2)	1.3(-2)
Broccoli ^b	139	(0.02)	1.2(-2)	2.6(-5)	4.3(-5)	3.9(-3)	2.4(-5)	6.3(-5)	3.1(-1)	1.1(-3)	6.5(-3)
Cabbage	6,174	(0.92)	4.5(-1)	1.2(-3)	2.9(-4)	2.5(-2)	6.2(-4)	3.3(-3)	1.0(+1)	3.7(-2)	2.4(-1)
Carrots	2,497	(0.37)	2.0(-1)	5.4(-4)	1.2(-4)	9.5(-3)	3.6(-4)	1.6(-3)	6.6	2.0(-2)	1.1(-1)
Celery	1,734	(0.26)	1.2(-1)	4.8(-4)	3.0(-4)	1.0(-2)	2.4(-4)	1.3(-3)	3.0	3.0(-2)	6.4(-2)
Cucumbers ^b	2,012	(0.29)	9.1(-2)	4.2(-4)	7.2(-5)	4.6(-3)	1.4(-4)	6.8(-4)	1.8	1.1(-2)	5.2(-2)
Green Beans	5,272	(0.79)	2.8(-1)	1.4(-3)	5.1(-4)	2.0(-2)	6.6(-4)	4.1(-3)	7.7	4.8(-2)	1.5(-1)
Lettuce ^b	7,007	(1.0)	2.8(-1)	9.2(-4)	2.5(-4)	2.5(-2)	6.0(-4)	3.1(-3)	7.0	3.6(-2)	1.5(-1)
Peppers (Bell)	486	(0.07)	2.6(-2)	1.5(-4)	3.6(-5)	2.3(-2)	5.8(-4)	4.9(-4)	1.5	6.1(-3)	1.4(-2)
Potatoes (Irish)	29,692	(4.4)	3.0	1.1(-2)	1.4(-3)	1.6(-1)	4.1(-3)	2.1(-2)	1.6(+1)	3.1(-1)	1.7
Potatoes (Sweet)	1,179	(0.18)	1.1(-1)	3.3(-4)	7.3(-5)	3.2(-2)	4.5(-4)	1.1(-3)	2.1	1.7(-2)	5.9(-2)
Turnips	208	(0.03)	9.7(-3)	2.6(-5)	3.4(-5)	9.6(-4)	2.0(-5)	1.0(-4)	3.1(-1)	9.9(-4)	5.4(-3)
Total		14.3									3.6

^a Winesap, red and golden delicious.^b One sample.

Table 3. Stable element composition of fresh fruits and vegetables

Foodstuff	Ash wt.	$\mu\text{g/g}$ (wet wt.)																		
	wet wt.	Ag	Al	As	Au	Ba	Br	Ca	Ce	Cd	Cl	Co	Cr	Cs	Cu	Eu	Fe	Hf	I	K
Vegetables																				
Cucumbers	5.8(-3)	<2.9(-3)	2.6(-1)	6.4(-3)	4.1(-5)	<5.8(-1)	8.7(-2)	34	<5.8(-3)	4.5(-4)	1.3(2)	8.7(-4)	6.4(-3)	<4.1(-4)	<4.4(-1)	<2.9(-4)	9.5(-1)	<5.8(-4)	<2.3(-2)	5.9(+2)
Turnips	6.7(-5)	<6.2(-3)	1.2	<1.0(-2)	7.6(-5)	4.3(-1)	1.6(-1)	1.1(+2)	3.0(-3)	9.0(-4)	1.2(1)	5.6(-3)	9.9(-3)	3.3(-3)	3.3(-1)	<3.1(-4)	1.0	3.7(-4)	<2.0(-2)	5.7(2)
Cabbage	8.0(-3)	<8.0(-3)	1.5	<1.6(-2)	4.0(-4)	1.3	3.7(-1)	40	6.4(-3)	2.1(-3)	2.0(-)	3.6(-3)	1.3(-3)	<4.2(-3)	<4.0(-1)	<4.0(-4)	2.4	<4.8(-4)	<3.2(-2)	2.0(2)
Yellow squash	7.3(-3)	<7.3(-3)	1.0	<7.3(-3)	2.4(-5)	<7.3(-1)	1.6(-1)	47	<7.3(-3)	4.3(-4)	1.1(2)	7.9(-4)	8.0(-3)	<4.7(-4)	1.9(-1)	<3.7(-4)	8.0(-1)	<4.0(-4)	1.8(-2)	4.5(+2)
Carrots	8.9(-3)	<7.5(-3)	4.0(-1)	<1.3(-2)	<1.0(-4)	1.4(-1)	8.5(-2)	25	<7.5(-3)	1.9(-3)	1.8(2)	3.0(-3)	1.8(-2)	<4.0(-4)	2.2(-1)	<3.8(-4)	7.6(-1)	2.4(-1)	2.5(-2)	5.9(2)
Sweet potatoes	1.2(-2)	<1.2(-2)	2.7	<3.6(-2)	ND	1.4	6.4(-1)	1.3(2)	1.2(-2)	1.0(-3)	2.3(2)	2.4(-3)	1.8(-2)	2.9(-3)	8.8(-1)	<1.2(-3)	4.3	1.1(-3)	<4.8(-2)	1.9(3)
Green peppers	6.4(-3)	<6.4(-3)	1.2(-1)	<6.4(-3)	ND	1.9(-1)	6.6(-2)	12	<3.2(-3)	3.8(-4)	20	2.6(-3)	6.4(-3)	2.6(-4)	7.0(-2)	<3.2(-4)	4.3(-1)	<3.2(-4)	<6.4(-3)	1.8(2)
Green beans	6.7(-3)	<3.4(-3)	3.6(-1)	<6.7(-3)	ND	<4.0(-1)	6.2(-2)	68	<6.7(-3)	1.5(-4)	36	1.0(-3)	8.7(-3)	<3.4(-4)	1.5(-1)	<3.4(-4)	7.4(-4)	<3.4(-4)	<1.3(-2)	2.7(2)
Irish potatoes	9.9(-3)	<5.0(-3)	3.8	<9.9(-3)	ND	<6.9(-1)	6.0(-2)	18	<9.9(-3)	5.2(-3)	49	2.5(-3)	1.8(-2)	2.0(-3)	3.8(-1)	<5.0(-4)	2.8	5.0(-4)	<2.0(-2)	6.9(2)
Egg plant	6.8(-3)	ND	6.4(-1)	<6.7(-3)	<5.1(-5)	<6.7(-1)	2.1(-1)	26	ND	4.2(-3)	88	3.7(-4)	1.5(-2)	<1.7(-4)	1.7(-1)	ND	6.8(-1)	ND	<1.7(-2)	6.1(2)
Lettuce	5.3(-3)	ND	1.0(-1)	<5.3(-3)	<5.8(-6)	<5.3(-1)	1.6(-2)	17	ND	1.2(-3)	44	5.3(-4)	8.0(-3)	1.6(-4)	<1.1(-1)	ND	3.4(-1)	ND	<1.1(-2)	1.7(2)
Broccoli	1.1(-2)	ND	8.1(-1)	<1.1(-2)	<5.5(-5)	<1.1	2.8(-1)	1.2(2)	ND	2.2(-3)	99	6.8(-3)	1.4(-2)	<6.6(-4)	5.7(-1)	ND	1.6	ND	<3.3(-2)	7.0(2)
Fruits																				
Winecup apples	5.4(-3)	ND	4.4(-2)	<5.4(-4)	4.9(-6)	5.9(-2)	1.5(-3)	7.2	ND	2.2(-5)	4.9(-1)	3.2(-4)	7.6(-3)	<1.1(-4)	2.3(-2)	ND	5.5(-1)	ND	<2.7(-3)	1.0(2)
Sunkist oranges	6.7(-3)	ND	1.4	<5.2(-3)	<6.7(-5)	<1.7(-1)	4.0(-2)	98	ND	6.6(-5)	22	4.3(-4)	1.5(-2)	<4.6(-4)	1.1(-1)	ND	2.2	ND	<1.0(-2)	5.9(2)
Bananas	1.4(-2)	ND	1.4	<1.4(-4)	<1.4(-4)	<1.4(-1)	1.1(-1)	15	ND	8.4(-5)	92	8.4(-4)	3.5(-2)	<5.6(-4)	4.5(-1)	ND	1.9	ND	<2.8(-2)	92
Tangelos	1.2(-2)	ND	5.0(-1)	<1.2(-2)	<1.9(-5)	<1.2(-1)	1.1(-1)	1.4(2)	ND	2.3(-4)	19	8.4(-4)	4.3(-2)	2.9(-3)	2.5(-1)	ND	1.2	ND	<2.4(-2)	1.2(3)
Raisins	2.6(-2)	ND	15	<5.2(-2)	<2.6(-4)	<2.6	1.1	3.6(2)	ND	2.4(-3)	1.1(2)	7.0(-3)	9.9(-2)	1.6(-2)	1.3	ND	10.4	ND	<7.8(-2)	3.9(3)
Grapefruits	4.4(-3)	ND	3.1(-2)	<8.8(-4)	2.2(-6)	<8.8(-2)	1.5(-2)	15	ND	1.4(-3)	3.5	4.4(-5)	5.3(-3)	<8.8(-5)	2.8(-2)	ND	3.2(-1)	ND	<4.4(-3)	1.1(2)
Pears	5.7(-3)	ND	9.1(-2)	<1.1(-3)	1.1(-5)	9.1(-2)	4.0(-3)	20	ND	2.8(-6)	1.0	3.4(-3)	1.0(-2)	<2.3(-4)	1.5(-1)	ND	1.8	ND	<5.7(-3)	3.0(2)
Golden Delicious Apples	4.2(-3)	ND	9.2(-2)	<2.1(-4)	3.8(-6)	2.4(-2)	5.7(-4)	3.4	ND	7.6(-5)	2.9(-1)	8.4(-5)	5.0(-3)	1.7(-4)	3.2(-2)	ND	3.6(-1)	ND	<4.2(-3)	64
Red Delicious Apples	2.0(-3)	ND	1.5(-2)	<1.0(-4)	1.2(-6)	<8.0(-3)	1.2(-4)	9.8(-1)	ND	5.4(-5)	6.2(-2)	4.0(-5)	3.2(-3)	<2.0(-5)	5.0(-3)	ND	8.4(-2)	ND	<1.0(-3)	20

Table 4. Stable element composition of fresh fruits and vegetables

Foodstuff	$\mu\text{g/g (wet wt.)}$																		
	La	Mg	Mo	Mn	Na	Ni	Pb	Rb	Sb	Sc	Se	Sr	Ta	Te	Th	Ti	V	Zn	Zr
Vegetables																			
Cucumbers	1.2(-2)	35	<8.7(-2)	3.2(-1)	9.2	1.3(-2)	2.4(-2)	7.0(-2)	<3.5(-4)	1.6(-5)	<5.8(-3)	<1.2	<2.9(-4)	1.3(-1)	4.7(-3)	<4.6(-1)	<5.8(-3)	5.1(-1)	<2.3(-1)
Turnips	4.7(-3)	36	6.5(-2)	3.0(-1)	22	2.9(-2)	5.9(-3)	4.7(-1)	7.4(-4)	1.6(-4)	5.2(-3)	1.3	<2.6(-4)	4.6(-1)	4.9(-3)	5.1(-1)	4.8(-3)	4.2(-1)	<3.1(-1)
Cabbage	<5.6(-3)	22	<9.9(-2)	1.2	38	4.9(-2)	1.6(-2)	4.6(-1)	<4.3(-3)	<8.0(-5)	<8.0(-3)	<2.2	<4.8(-4)	2.8(-1)	<2.4(-3)	<6.8(-1)	8.0(-3)	5.6(-1)	<4.0(-1)
Yellow squash	<7.3(-3)	35	<9.8(-2)	2.4(-1)	4.0	9.0(-3)	9.7(-3)	7.0(-2)	<4.7(-4)	3.9(-5)	<7.3(-3)	<8.7(-1)	<5.5(-4)	2.6(-1)	2.2(-3)	<3.7(-1)	<7.3(-3)	4.1(-1)	<2.2(-1)
Carrots	<4.3(-3)	20	<8.0(-2)	1.5(-1)	28	2.0(-2)	1.2(-2)	1.1(-1)	<4.0(-4)	5.1(-5)	<5.3(-3)	<1.33	<2.4(-4)	2.8(-1)	<2.3(-3)	<5.3(-1)	<8.9(-3)	4.6(-1)	<3.2(-1)
Sweet potatoes	<2.4(-2)	1.1(2)	<4.8(-1)	1.0	87	3.2(-2)	3.2(-2)	9.2(-1)	4.2(-3)	3.8(-4)	<2.4(-2)	<4.8	<7.2(-4)	5.0(-1)	<4.8(-3)	<1.2	5.2(-3)	1.3	<6.0(-1)
Green peppers	<3.6(-3)	9.4	3.5(-2)	9.1(-2)	2.4	8.1(-3)	1.3(-3)	<5.7(-2)	3.0(-3)	1.2(-5)	<3.8(-3)	1.8(-1)	<1.9(-4)	3.8(-2)	<1.9(-3)	<1.2(-1)	<1.6(-3)	1.0(-1)	<1.3(-1)
Green beans	<6.7(-3)	33	1.0(-1)	2.8(-1)	6.5	2.3(-2)	1.9(-2)	4.7(-2)	<3.4(-4)	<3.4(-5)	<6.7(-3)	4.0(-1)	<3.4(-4)	<3.4(-1)	<2.0(-3)	<2.0(-1)	<3.4(-3)	2.6(-1)	<1.3(-1)
Irish potatoes	<9.9(-3)	45	<1.5(-1)	3.0(-1)	18	4.0(-2)	2.6(-2)	1.1	<6.9(-4)	5.9(-4)	<9.9(-3)	<2.0	<5.0(-4)	4.5(-2)	<4.0(-3)	2.1(-1)	9.3(-3)	2.7(-1)	<3.0(-1)
Egg plant	<6.8(-4)	37	1.3(-2)	2.8(-1)	9.5	2.3(-1)	4.3(-2)	1.2(-1)	<1.4(-4)	1.0(-4)	2.4(-3)	<1.5	<1.0(-4)	<6.8(-2)	ND	<3.4(-1)	6.7(-3)	3.5(-1)	ND
Lettuce	<5.3(-4)	5.8	5.3(-3)	9.9(-2)	4.6	1.2(-2)	1.4(-3)	<2.8(-2)	<1.1(-4)	5.3(-6)	<1.6(-3)	<2.1(-1)	<1.1(-4)	ND	ND	<2.7(-1)	5.3(-3)	1.3(-1)	ND
Broccoli	<2.2(-3)	42	2.1(-2)	9.9(-1)	61	4.6(-2)	1.6(-2)	3.7(-1)	<1.1(-3)	9.9(-5)	<5.5(-3)	<2.2	<2.2(-4)	ND	ND	<5.5(-1)	5.5(-3)	8.1(-1)	ND
Fruits																			
Winesap Apples	<5.4(-4)	4.9	1.1(-3)	2.6(-2)	9.2(-1)	2.5(-3)	1.4(-3)	3.1(-2)	1.6(-4)	5.4(-6)	<1.1(-3)	5.9(-2)	<5.4(-5)	ND	ND	<5.4(-2)	<5.4(-4)	3.2(-2)	ND
Sunkist Oranges	<6.7(-4)	29	<1.4(-2)	5.3(-2)	3.1	7.7(-3)	2.4(-3)	1.6(-1)	<2.7(-4)	1.0(-4)	<3.0(-3)	5.4(-1)	<1.2(-4)	<1.7(-1)	ND	<1.2(-1)	<1.2(-3)	8.7(-2)	ND
Bananas	<1.4(-3)	1.4(2)	2.8(-2)	2.8(-1)	12	6.3(-2)	2.1(-3)	2.4(-1)	1.4(-3)	5.6(-5)	<7.0(-3)	<2.8	<1.4(-4)	ND	ND	<7.0(-1)	<7.0(-3)	7.0(-3)	ND
Tangelos	<1.2(-3)	85	2.4(-3)	2.5(-1)	13	3.0(-2)	2.6(-3)	4.2(-1)	6.0(-4)	1.2(-5)	<2.4(-3)	1.1	<1.2(-4)	<3.6(-1)	ND	<6.0(-1)	<6.0(-3)	4.1(-1)	ND
Raisins	5.7(-3)	1.8(-2)	<1.3(-1)	1.3	7.8	1.7(-1)	3.6(-2)	6.1	<1.3(-2)	1.6(-3)	<5.2(-2)	1.8	<1.0(-3)	ND	ND	<1.3	1.5(-2)	ND	ND
Grape- fruit	<4.4(-4)	7.9	<2.2(-3)	2.1(-2)	1.4	2.2(-2)	6.3(-3)	1.8(-2)	<1.8(-4)	<2.2(-5)	<8.8(-4)	5.9(-1)	<8.8(-5)	ND	ND	<4.4(-2)	<2.2(-4)	4.4(-2)	ND
Pears	<5.7(-4)	16	<5.7(-3)	1.7(-1)	2.3	2.9(-3)	1.9(-3)	1.1(-1)	2.1(-4)	<2.3(-5)	<1.1(-3)	<4.0(-1)	<1.1(-4)	ND	ND	<1.1(-4)	<2.9(-4)	1.9(-1)	ND
Golden Delicious Apples	<2.1(-4)	3.0	<2.1(-3)	1.9(-2)	1.3	1.9(-3)	2.0(-3)	6.3(-2)	8.4(-5)	1.3(-5)	<1.7(-4)	<8.4(-2)	<1.3(-5)	<1.3(-2)	ND	9.2(-3)	8.4(-5)	2.0(-2)	ND
Red Delicious Apples	<1.0(-4)	8.6(-1)	<4.0(-4)	5.2(-3)	4.2(-1)	4.7(-3)	9.8(-4)	1.4(-2)	2.0(-5)	2.6(-6)	<1.8(-4)	<2.0(-2)	<1.0(-5)	ND	ND	<4.0(-3)	3.0(-4)	ND	ND