

RUJ-21

Application of Activation Analysis to the Determination of Trace-Element Concentrations in Meat^a

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SUMMARY

The concentrations of 27 trace elements were determined in four meats: beef, pork, ham, and chicken. Because the concentrations of most of these elements were expected to be very small, the extremely sensitive method of neutron activation analysis was used in this study. Qualitative analyses were performed for 8 of the elements, and the concentrations of 19 elements were determined quantitatively. The quantitatively measured concentrations varied from ~0.1% for phosphorus to ~10⁻³ ppm for cerium. Most data are estimated to be accurate to ±10%. The results demonstrate the applicability of this analytical method to determination of the inorganic constituents in foods and related substances at a constant level of accuracy throughout the concentration ranges of practical interest.

INTRODUCTION

A comprehensive program has been initiated to characterize the elemental constituents in bulk samples of four meats: beef, pork, ham, and chicken. In the first phase of this study, activation analysis has been applied to the determination of 27 trace elements in these meats. The analytical data demonstrate the applicability of this analytical method to determination of the inorganic constituents in foods at a constant level of accuracy throughout the concentration ranges of practical interest.

Many elements normally present as trace constituents in foods are necessary to the maintenance of life and health. For example, calcium and phosphorus are the main components of bone; sodium, potassium, and chlorine help maintain the osmotic pressure of body liquids; and a number of elements, such as iron and manganese, activate hormones. Other elements, such as cobalt, are components of vitamins or other substances essential to a balanced diet.

The recognition of the importance of trace elements in nutrition has led to increasingly stringent requirements for sensitivity and specificity in food analysis. Several analyti-

cal methods, such as emission spectroscopy, flame spectrophotometry, colorimetry, amperometric titrations, and activation analysis, are generally useful for trace-element analysis. A comparison of these methods (Meinke, 1955) shows that activation analysis has the best sensitivity for a majority of the elements. Activation analysis has been used for the determination of specific elements in animal tissue and vegetation (Bowen, 1956, 1959a,b; Gibbons, 1958; Goetté, 1955; Harrison, 1955; Kruger, 1962; Smales, 1952; Smith, 1959). The demonstrated versatility of activation analysis and its intrinsic sensitivity for most elements led to its choice for this application.

Activation analysis has been defined (Koch, 1960) as a method of determining the concentration of an element in a matrix by measuring the characteristic radiations emitted by a radioactive nuclide resulting from a specific nuclear reaction in the trace element. This radioactive reaction product, or activation product, possesses a unique combination of physical, chemical, and nuclear properties that provides the necessary specificity for its identification and measurement.

The general method of neutron activation analysis involves irradiation of the samples

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to be analyzed in a neutron flux, such as is found in a nuclear reactor, to produce the desired activation products in sufficient quantity for accurate measurement. After irradiation, the samples are analyzed for the radioisotopes formed in each element of interest. This analysis involves chemical separation and purification of the activation product and determination of its disintegration rate by measurement of its characteristic beta or gamma radiations.

During the irradiation, neutrons are absorbed by nuclei of some of the atoms in the sample, producing different nuclei. The neutron absorption is accompanied by emission of a second nuclear particle or a quantum of electromagnetic radiation. The most common reaction involving reactor neutrons is the (n, γ) reaction in which the neutron is absorbed in the nucleus, followed by emission of a high-energy gamma ray. This reaction produces an isotope of the same element as the reactant atom. The product isotope is often radioactive. Hence, it is termed an "activation product."

The production (Koch, 1960) of an activation product proceeds ideally according to equation 1:

$$D(t) = \phi n \sigma (1 - e^{-\lambda t}) \quad [1]$$

where $D(t)$ = the rate of decay of the radioactive species in the sample after irradiation time, t , disintegrations per second;

ϕ = the neutron flux, neutrons/cm²-sec

n = the number of atoms of the target isotope in the irradiation sample;

σ = the activation cross-section, cm²/atom;

λ = the decay constant of the nuclide, sec⁻¹; and

t = the irradiation time, sec.

Low-energy or thermal neutrons induce the (n, γ) reaction primarily. However, higher-energy, or fast, neutrons are also present in reactors. The net effect of the interactions of these neutrons may be the transmutation of an atom of one element to an atom of a different element, since charged

particles such as protons or alpha particles may be produced in these reactions. Thus, if an (n, p) reaction, in which a proton is produced, occurs in an atom having atomic number Z , an isotope of the element $Z-1$ is produced.

Activation analyses are usually performed by a comparative method. In this method, comparator samples containing known quantities of the trace elements to be determined, are irradiated and analyzed concurrently with the matrix sample. The concentration of a trace element in the matrix is then calculated using equation 2:

$$m_s = \frac{m_c D(t)_s}{D(t)_c} \quad [2]$$

where m_s and m_c are the respective masses of the trace element in the matrix sample and comparator, and $D(t)_s$ and $D(t)_c$ are the respective disintegration rates of the activation product.

The performance of a neutron activation analysis requires a set of six operations (Koch, 1960): 1) the selection of an appropriate neutron-induced activation reaction; 2) the specification of comparator samples; 3) the choice of a suitable irradiation facility; 4) the preparation of the samples for irradiation; and 5) the irradiation and 6) the post-irradiation analyses. Consideration was given to the requirements imposed by each of these operations in delineating the scheme of analysis used in this work. A description of these considerations and of their application in this program is presented.

Selection of activation reactions. The first operation in performing activation analysis is the selection of the optimum activation reaction. This selection involves an evaluation of: 1) the suitability of the activation product, including the feasibility of performing the post-irradiation assays; and 2) the extent to which interfering reactions in the matrix may produce or consume the activation product to be assayed. Since alternative neutron activation reactions provide analytical sensitivity sufficient for most applications, sensitivity is seldom an important factor in this selection.

The basic requirement for a useful activation product is that it be radioactive, with

a half-life long enough to permit the post-irradiation measurements. The minimum useful half-life is dependent primarily on the complexity of the chemical separations. In special cases where direct measurement of the activation product can be made in the sample, a half-life of the order of minutes may be acceptable.

Activation products that are gases or may be present in volatile forms should be avoided. If such species must be used, precautions are required to prevent their release from the sample during irradiation or as a result of chemical processing.

An interfering reaction has been defined (Koch, 1960) as a nuclear reaction in a constituent of the matrix, other than a stable isotope of the trace element to be determined, that produces or consumes the activation product to be measured. An interfering reaction may significantly affect the measured quantity of the activation product and produce an erroneous result. There are several possible sources of interference, but only two general types warranted serious consideration in this work.

The first type of interference involves nuclear reactions resulting in the production of charged particles. For example, if the trace element having atomic number Z is to be determined, interference may result from an (n,p) reaction with element $Z + 1$ or from an (n, α) reaction with element $Z + 2$, either of which produces the desired activation product. The effective cross-sections for the (n, p) and (n, α) reactions are generally several orders of magnitude less than those for the (n, γ) reactions. Therefore, this interference is usually very small if the two elements have similar concentrations. It may be important, however, if the trace element is present only in exceptionally small concentrations.

The second type of interference involves the production of the desired activation product by neutron-induced fission reactions, primarily in uranium. During the fission process, the uranium nucleus is split into two fragments, producing radioactive isotopes of the entire spectrum of elements from zinc to terbium. Since many of the useful activation products of these elements are also fission products, this type of interference is poten-

tially widespread. Uranium may interfere with the determination of many of these elements, even when it is present in only trace concentrations.

Since the concentrations of many of the elements in the meats to be analyzed in this investigation were expected to be very small, special attention was directed to avoiding or compensating for potential interfering reactions. For some elements, alternative isotopes are available for which interference does not occur. For example, the 65-day Sr^{85} and 11.6-day Ba^{131} isotopes were chosen for use in analysis for strontium and barium, rather than the 54-day Sr^{89} and 85-min Ba^{139} , because the latter two isotopes are formed in uranium fission.

For some of the elements of interest, interfering reactions could not be avoided. Therefore, data were required to determine the extent of the interference and to correct for it. The necessary data are obtained by analysis for the sought activation product in a comparator sample of the interfering element, and by determination of the interfering element in the matrix. The quantity of the activation product in the matrix that is due to the interfering reaction is then calculated using equation 2.

An important example of this type of interference occurs in analysis for phosphorus and sulfur. The pertinent section of the Chart of the Nuclides (Stehn and Clancy, 1956), presented in Fig. 1, shows the several possible neutron-induced reactions in these elements and the modes of interference with sulfur and phosphorus analyses. From Fig. 1 it is seen that (n, γ) reactions in chlorine yield the 3×10^5 -year Cl^{36} and the 37-min Cl^{38} , and the (n, p) reaction in Cl^{35} yields

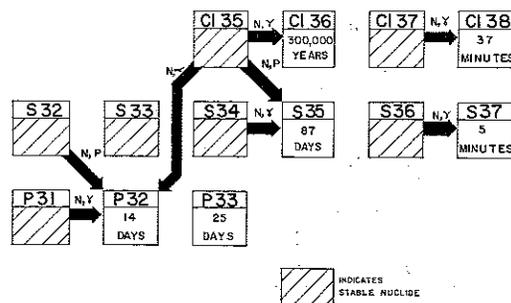


Fig. 1. Chlorine, sulfur, and phosphorus activation.

DETERMINING TRACE ELEMENTS IN MEAT

Table 1. Data for activation products and interfering reactions.

Element	Activation product	Radiation measured	Interfering reaction
Phosphorus	14.2-d P ³²	1.707 Mev β ⁻	S ³² (n, p), Cl ³⁵ (n, α)
Sulfur	87-d S ³⁵	0.1617 Mev β ⁻	Cl ³⁵ (n, p)
Chlorine	3.0 × 10 ³ -y Cl ³⁶	0.714 Mev β ⁻	None
Scandium	83.9-d Sc ⁴⁶	0.885 Mev γ	Ti ⁴⁶ (n, p)
Iron	45.1-d Fe ⁵⁹	1.098 Mev γ	Co ⁵⁸ (n, p)
Cobalt	5.24-y Co ⁶⁰	1.1728 Mev γ	Ni ⁶⁰ (n, p)
Zinc	245-d Zn ⁶⁵	1.119 Mev γ	None
Selenium	127-d Se ⁷⁵	0.402 Mev γ	None
Rubidium	18.66-d Rb ⁸³	1.079 Mev γ	Sr ⁸³ (n, p)
Strontium	64-d Sr ⁸⁵	0.513 Mev γ	None
Zirconium	65-d Zr ⁸⁵	0.723, 0.756 Mev γ	U, Th fission
Ruthenium	39.8-d Ru ¹⁰³	Multiple β ⁻	U, Th fission
Palladium	17-d Pd ¹⁰³	Multiple γ	Cd ¹⁰⁶ (n, α)
Indium	49-d In ^{114m}	0.190 Mev γ	None
Tin	119-d Sn ¹¹³	0.393 Mev γ	Sb ¹²³ (n, p)
Tellurium	104-d Te ^{123m}	0.158 Mev γ	None
Cesium	2.07-y Cs ¹³⁴	0.796 Mev γ	Ba ¹³⁴ (n, p)
Barium	11.5-d Ba ¹³¹	0.214 Mev γ	None
Cerium	32.5-d Ce ¹⁴¹	Multiple γ	Pr ¹⁴¹ (n, p) U, Th fission
Hafnium	70-d Hf ¹⁷⁵	0.089 Mev γ	None
Iridium	74.4-d Ir ¹⁹²	Multiple β ⁻	Pt ¹⁹² (n, p)
Platinum	4.3-d Pt ^{193m}	0.136 Mev γ	None
Uranium-238	2.346-d Np ²³⁹	Multiple β ⁻	None
Uranium-235	12.8-d Ba ¹⁴⁰	Multiple β ⁻ (La ¹⁴⁰)	None

Table 2. Data for comparator samples.

Trace element	Primary comparator	Secondary comparator	Typical wt. of element in prim. comp. (mg)
Phosphorus	Mg ₂ P ₂ O ₇	S, NaCl	2.8
Sulfur	S	NaCl	23
Chlorine	NaCl	10.4
Scandium	Sc(NO ₃) ₃	Ti	0.001
Iron	Fe	Al-0.1% Co alloy	9.8
Cobalt	Al-0.1% Co alloy	Ni	0.01
Zinc	Zn	11
Selenium	Se	1.0
Rubidium	RbCl	Sr(NO ₃) ₂	1.0
Strontium	Sr(NO ₃) ₂	20
Zirconium	ZrO(NO ₃) ₂ · 2H ₂ O	10.3
Indium	In	Sn	0.12
Tin	Sn	Sb	22
Tellurium	Te	10
Cesium	CsNO ₃	Ba(NO ₃) ₂	0.08
Barium	Ba(NO ₃) ₂	11
Hafnium	Hf	0.12
Uranium-235	Enriched U	0.01
Uranium-238	Natural U ₃ O ₈	1.0

87-day S^{35} . The latter nuclide is also formed by the (n, γ) reaction in S^{34} . Since the only other (n, γ) activation reaction in sulfur yields the very short-lived S^{37} , measurement of S^{35} is required for neutron activation analysis for sulfur in meat ash.

Similarly, the only (n, γ) activation product in phosphorus is 14.2-day P^{32} , which is also produced by the (n, p) reaction in S^{32} . Therefore, the mutual interference of these elements is unavoidable, and an experimental study was required to determine the extent of the interference. This problem is discussed in detail in Appendix I.

A review of the above criteria for the nineteen elements to be analyzed quantitatively led to selection of the activation products shown in column 2 of Table 1. Column 3 of the table presents the radiation that was measured, and column 4 shows the potential interference for the respective elements.

Selection of comparators. Comparator samples are required for each trace element to be determined. If interfering reactions may occur, secondary comparators, containing the interfering element, are also utilized. In each case, the comparators are prepared from known amounts of the respective elements or of their compounds. If a compound is used, it must exhibit stoichiometric and radiolytic stability. In addition, care must be taken to avoid local neutron flux disturbances by comparators for elements having large cross-sections for neutron absorption.

Table 2 summarizes the chemical form and typical weights of the comparator samples used in this program. The weights were chosen to limit the attenuation of the neutron flux by the sample to 1%.

EXPERIMENTAL PROGRAM

Sampling and sample preparation. Since the bulk meat samples were to be representative of portions consumed by humans, all bones, cartilage, glands, and other inedible matter were removed. To obtain statistically significant samples, each meat was ground through a $\frac{1}{2}$ -in. plate, followed by a second grinding through a $\frac{1}{8}$ - $\frac{1}{4}$ -in. plate. The meat was then mixed by machine into a homogeneous mass, packaged, and kept frozen until used.

Weighed portions of the homogenized meats were ashed to concentrate the inorganic constituents into samples convenient for irradiation. The

samples were partially ashed by slow combustion in large stainless-steel vessels and then ignited in large porcelain crucibles at 800°C. In removing the partially ashed residues from the steel vessels, care was taken to avoid their contamination with traces of metal. The ratios of ash to bulk weights varied from approximately 0.5% to 2%, as shown in Table 3.

Table 3. Weights of bulk and ashed meats.

Food	Bulk wt (kg)	Ashed wt (g)	Percent ash
Chicken	11.55	71.2	0.62
Beef	11.47	69.9	0.70
Pork	11.41	52.0	0.46
Ham	11.10	220.6	2.0

Samples of the ashed meats and comparators were then prepared and packaged for irradiation. Two-gram portions of each ash were packed in aluminum-foil cylinders. These cylinders were tightly crimped and sealed in quartz ampoules.

Three primary comparator samples were prepared for each element, along with secondary comparators for their respective interfering elements. The comparators were also doubly-contained in aluminum foil and quartz. The quartz ampoules were inserted in an aluminum cylinder for irradiation in the Brookhaven Graphite Reactor.

Analytical procedures. Two irradiation experiments were performed. Samples from the first irradiation were used for qualitative analysis for some rare earths and platinum metals. Quantitative determinations of 19 additional elements were made on samples from the second irradiation.

After each irradiation, the ash and comparator samples were dissolved. Aliquots of the solutions were taken for radiochemical separation and purification of the respective activation products, and for measurement of their radiations.

The ash samples were normally dissolved in boiling aqua regia. Any insoluble residue was dissolved with hydrofluoric acid. Metallic comparators were dissolved in an appropriate acid. Closed systems were used to dissolve comparators for volatile elements and ash samples in which chlorine analyses were to be performed.

Known aliquots of each solution were taken for the specific elemental analyses. Since only tracer quantities of the activation products are produced in such irradiations, known quantities of each element were added to the respective aliquots to serve as carriers. The carriers facilitate chemical separations and also indicate the fractional recovery, or chemical yield, at the completion of the purification procedure, upon analysis by conventional techniques.

DETERMINING TRACE ELEMENTS IN MEAT

Each activation product was identified by measurement of its half-life and the energies of its beta or gamma radiations. The energies of gamma radiations were measured with a flat 3×3-in. NaI(Tl) crystal and a 256-channel pulse-height analyzer. End-window, methane-flow proportional counters were used to measure beta radiations. Beta energies were derived from aluminum absorption curves. As an additional check of sample purity, the decay of each sample was followed for several half-lives, when possible, to verify the half-life.

The procedures for the qualitative analyses involved group separations of the rare earths and platinum metals, followed by separations within each group. The separations of the platinum metals were based on the system described by Noyes and Bray (1943). The rare earths were separated into the yttrium and lanthanum groups by successive extractions of the yttrium group into tributyl phosphate. Many of the quantitative analysis procedures were modifications of standard radiochemistry procedures (Kleinberg, 1958; Meinke, 1949). For some elements, essentially new procedures were developed.

EXPERIMENTAL RESULTS

Qualitative analyses. The presence of several platinum metals and of several rare earths in meat ash was established by qualitative neutron activation analysis.

The radionuclides 40-day Ru¹⁰³, 17-day Pd¹⁰⁵, 74-day Ir¹⁹², and 3.4-day Pt^{195m} were identified by the energies of their beta or gamma radiations and their half-lives. Estimates were made of the order of magnitude of the concentrations of these elements in each food sample. The values, which range from ~ 10⁻⁴ to 10⁻⁶ ppm in the bulk meats, are shown in Table 4.

Certain rare earth elements were identified from sequential gamma spectrum analyses of the yttrium and lanthanum groups. Typical gamma spectra

of the yttrium fraction from the beef ash sample are presented in Fig. 2. Spectrum A was taken approximately two months after the irradiation, and spectrum B was taken after an additional two-month decay. Spectra are essentially the same for the yttrium fractions from the other meat ash samples. The most prominent photopeaks appear in channels 89, 110, and 200. These peaks correspond to gamma energies of 0.88 Mev, 1.12 Mev, and 2.0 Mev, and arise from the decay of 85-day Sc⁴⁶. Although scandium follows yttrium group chemistry, its rather high concentration in these samples was unexpected. Therefore, a quantitative analysis for scandium was performed in a subsequent irradiation.

Because of the comparatively large amount of scandium activity in these spectra, the sensitivity for the detection of other constituents was decreased substantially. However, it was possible to identify photopeaks due to the 84-Kev photon of 129-day Tm¹⁷⁰ and to several photons of 32-day Yb¹⁶⁹ and 6.8-day Lu¹⁷⁷.

Gamma spectra for the lanthanum fraction from the beef ash sample are presented in Fig. 3. These spectra were taken at the same time as those for the yttrium fraction. Spectra for the other ash samples were nearly identical. The most prominent photopeaks, corresponding to photon energies of 0.120 Mev, 0.34 Mev, 0.86 Mev, and 1.09 Mev, indicate the presence of 13-year Eu¹⁵² and 16-year Eu¹⁵⁴. There is also some evidence for a 20-Kev photon, which may indicate the presence of 80-year Sm¹⁵¹.

Estimates were made of the concentrations of these rare earth elements in each meat. The values, as shown in Table 4, are of the order of 10⁻³ to 10⁻⁵ ppm.

Quantitative analyses. The concentrations of 19 elements in each meat ash were determined in samples from the second irradiation. The analytical data are shown in Table 5, along with the concentrations in the bulk meats, which were cal-

Table 4. Concentrations (ppm) of trace elements in meat (qualitative analyses).

Trace element	Chicken		Beef		Pork		Ham	
	Ash	Bulk	Ash	Bulk	Ash	Bulk	Ash	Bulk
Ruthenium	0.1	6 × 10 ⁻⁴	0.1	7 × 10 ⁻⁴	10 ⁻²	5 × 10 ⁻⁴	0.1	2 × 10 ⁻³
Palladium	0.1	6 × 10 ⁻⁴	0.1	7 × 10 ⁻⁴	0.1	5 × 10 ⁻⁴	0.1	2 × 10 ⁻³
Iridium	10 ⁻³	6 × 10 ⁻⁶	10 ⁻²	7 × 10 ⁻⁶	10 ⁻³	5 × 10 ⁻⁶	10 ⁻³	2 × 10 ⁻⁵
Platinum	0.1	6 × 10 ⁻⁴	0.1	7 × 10 ⁻⁴	0.1	5 × 10 ⁻⁴	0.1	2 × 10 ⁻³
Ytterbium	10 ⁻²	6 × 10 ⁻⁵	10 ⁻²	7 × 10 ⁻⁵	10 ⁻³	5 × 10 ⁻⁵	10 ⁻³	2 × 10 ⁻⁵
Thulium	10 ⁻³	6 × 10 ⁻⁶	10 ⁻³	7 × 10 ⁻⁶	10 ⁻³	5 × 10 ⁻⁶	10 ⁻³	2 × 10 ⁻⁵
Lutetium	10 ⁻²	6 × 10 ⁻⁵	10 ⁻²	7 × 10 ⁻⁵	10 ⁻²	5 × 10 ⁻⁵	10 ⁻³	2 × 10 ⁻⁵
Europium	10 ⁻²	6 × 10 ⁻⁵	10 ⁻²	7 × 10 ⁻⁵	10 ⁻³	5 × 10 ⁻⁵	10 ⁻³	2 × 10 ⁻⁵

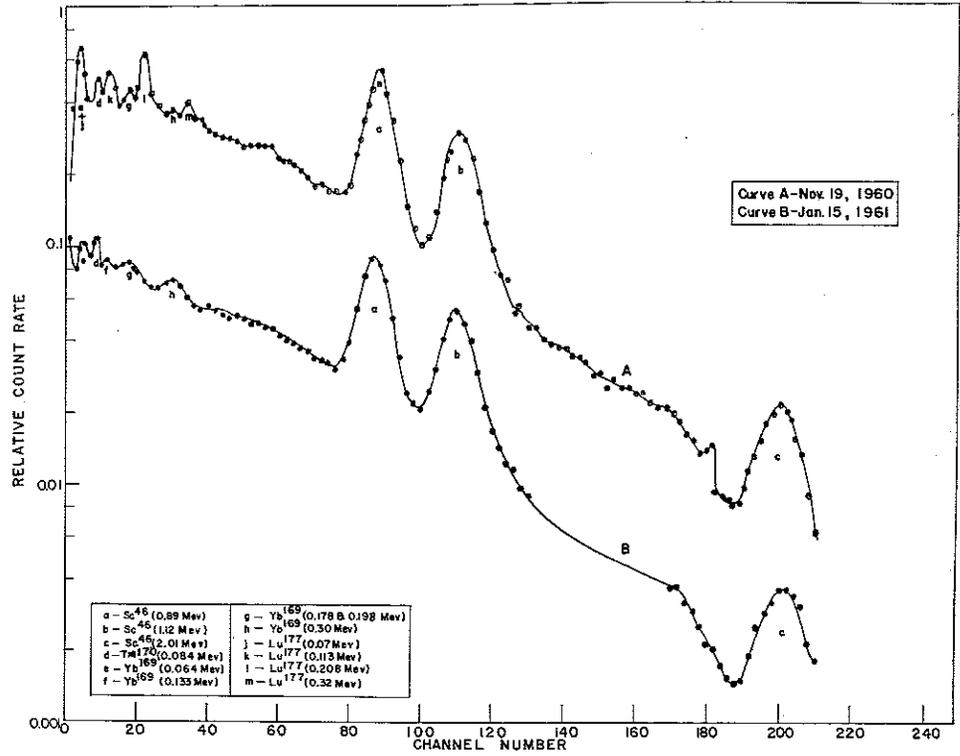


Fig. 2. Gamma spectra for yttrium group in irradiated beef ash.

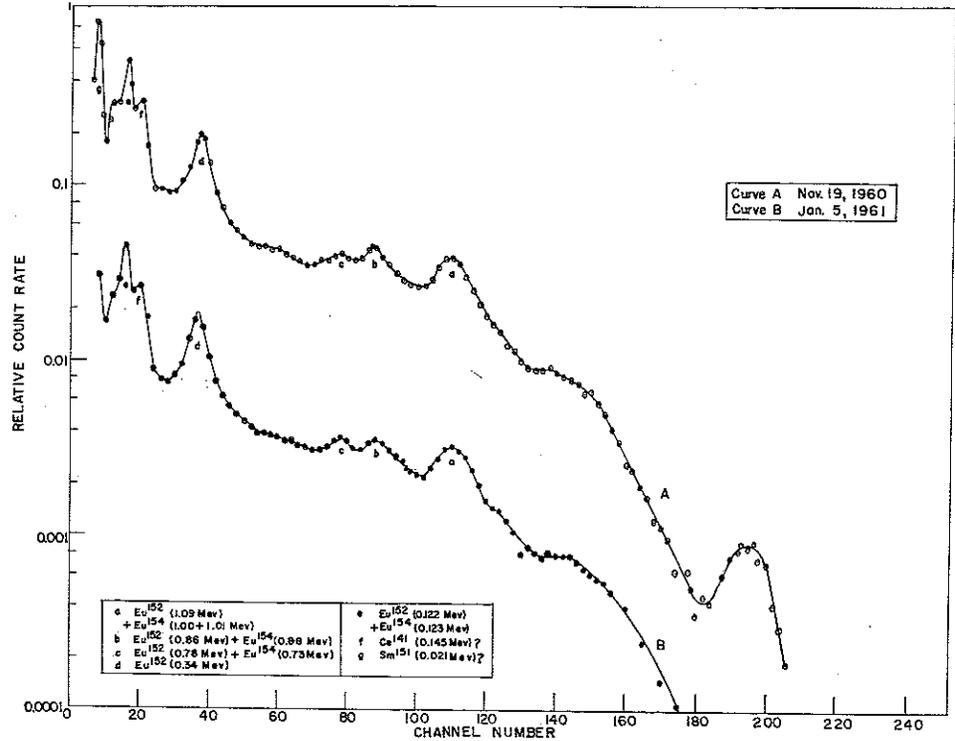


Fig. 3. Gamma spectra for lanthanum group in irradiated beef ash.

Table 5. Concentrations (ppm)^a of trace elements in meat (quantitative analyses).

Element	Chicken		Beef		Pork		Ham	
	Ash	Bulk	Ash	Bulk	Ash	Bulk	Ash	Bulk
Phosphorus	2.0×10^5	1.2×10^3	1.9×10^5	1.3×10^3	2.0×10^5	2.0×10^3	5.2×10^4	1.0×10^3
Sulfur	$<2 \times 10^4$	$<1 \times 10^2$	$<8 \times 10^4$	$<6 \times 10^2$	$<4 \times 10^4$	$<2 \times 10^2$	$<5 \times 10^5$	$<1 \times 10^4$
Chlorine	3.2×10^3	1.8×10^4	9.2×10^3	9.9×10^4
Scandium	1.2	7.4×10^{-3}	0.79	5.5×10^{-3}	0.57	2.6×10^{-3}	0.12	2.4×10^{-3}
Iron	2.1×10^4	1.3×10^2	5.8×10^3	41	4.2×10^3	19	3.9×10^3	78
Cobalt	17	0.11	4.5	3.2×10^{-2}	4.0	1.8×10^{-2}	3.0	5.9×10^{-2}
Zinc	1.7×10^3	11	2.9×10^3	20	3.0×10^3	14	4.8×10^2	9.5
Selenium	6.6×10^{-2}	4.1×10^{-4}	3.9×10^{-2}	2.7×10^{-4}	1.7×10^{-3}	7.8×10^{-5}	1.3×10^{-2}	2.6×10^{-4}
Rubidium	3.9×10^3	24	1.8×10^3	1.3	2.8×10^2	1.3	7.5×10^2	15
Strontium	35	0.22	25	0.18	20	9.2×10^{-3}	19	0.38
Zirconium	<16	<0.10	<24	<0.2	<30	<0.1	<5	<0.1
Indium	0.75	4.7×10^{-3}	1.3	9.2×10^{-3}	0.47	2.2×10^{-3}	0.55	1.1×10^{-2}
Tin	17	0.11	77	0.54	14	6.4×10^{-2}	3.2	6.4×10^{-2}
Tellurium	1.0 ± 0.5	$(6 \pm 3) \times 10^{-3}$	0.7 ± 0.14	$(5 \pm 1) \times 10^{-3}$	0.7 ± 0.14	$(3 \pm 0.6) \times 10^{-3}$	1.0 ± 0.2	$(2.0 \pm 0.4) \times 10^{-2}$
Cesium	1.7	1.1×10^{-2}	1.3	9.2×10^{-3}	1.3	6.0×10^{-3}	0.33	6.6×10^{-3}
Barium	53	0.33	50	0.35	38	0.18	19	0.38
Cerium	$(1.7 \pm 0.4) \times 10^{-2}$	$(1.1 \pm 0.3) \times 10^{-5}$	$(5.4 \pm 1.4) \times 10^{-4}$	$(3.8 \pm 1.0) \times 10^{-6}$	$(3.9 \pm 1.0) \times 10^{-3}$	$(1.8 \pm 0.4) \times 10^{-5}$	$(7.5 \pm 1.9) \times 10^{-4}$	$(1.5 \pm 0.4) \times 10^{-5}$
Hafnium	3.1	1.9×10^{-2}	3.9	2.0×10^{-3}	4.2	1.9×10^{-2}	0.60	1.2×10^{-2}
Uranium	1.1 ± 0.2	$(6.8 \pm 1.0) \times 10^{-3}$	0.50 ± 0.07	$(3.5 \pm 0.5) \times 10^{-3}$	0.48 ± 0.07	$(3.4 \pm 0.5) \times 10^{-3}$	0.19 ± 0.03	$(3.8 \pm 0.6) \times 10^{-3}$

^a Errors are estimated to be less than or equal to $\pm 10\%$ except where indicated.

culated from the ratios of ash to bulk weights given in Table 3. Definitive values were obtained for all of the elements listed except zirconium and sulfur.

Zirconium was determined in a purified mixture of zirconium and hafnium by gamma spectrometry techniques. However, the hafnium photo-peaks in the spectrum were sufficiently predominant to mask those of Zr^{95} . The values of the upper limits for zirconium concentrations were calculated from estimates of the minimum quantity of Zr^{95} that could be detected in the presence of the hafnium activity in each sample. The limiting values for zirconium concentrations are large relative to the observed hafnium concentrations because the cross-section of zirconium for thermal neutron activation is much smaller than that of hafnium. If more definitive data for zirconium concentrations had been desired, specific chemical separations could have been performed to permit measurement of the radioactivity due to zirconium.

The special problems associated with the determination of phosphorus and sulfur are discussed in detail in Appendix I. The very large upper limits for the concentrations of sulfur are indicative of the serious interference due to the $Cl^{35}(n,p)S^{35}$ reaction in chlorine. The reported value of the upper limit is the sulfur concentration, which corresponds to the minimum quantity of sulfur-produced S^{35} that is detectable in the presence of the chlorine-produced S^{35} in each sample. For chicken, pork, and beef, the total S^{35} found was identical, within experimental error, to the chlorine-produced S^{35} . In the case of ham ash, the minimum detectable quantity of sulfur was unrealistically high. Therefore, for each case, it is concluded that chlorine interference precludes determination of sulfur in these samples by neutron activation analysis.

In general, the value of the sulfur concentration is a prerequisite to determination of phosphorus, to permit estimation of the effects due to the reaction $S^{32}(n,p)P^{32}$. However, the observed phosphorus concentrations and the concentrations of P^{32} in the sulfur comparators were used to estimate the sulfur concentration that would have been required to introduce a 10% error in each phosphorus analysis. The estimated sulfur concentration was 63% in ham ash and over 100% for the other ashes. Therefore, the maximum error introduced in the phosphorus analyses by neglecting the effects of sulfur interference was of the order of a few percent.

The interference due to fast neutron-induced reactions in analysis for other elements was also determined. However, for all experimentally determined elements, correction factors were neg-

ligibly small. Therefore, no significant errors were introduced by these effects.

In general, precisions of replicate determinations of elemental concentrations were maintained to better than $\pm 5\%$. On the basis of these precisions and of the evaluation of possible consistent errors in sample preparation and analysis, it is estimated that the values of the concentrations of most of the trace elements were determined with an accuracy of $\pm 10\%$. All results in Table 5 are judged to have this accuracy except where larger errors are given.

DISCUSSION

The analyses yielded definite values for the concentrations of seventeen of the elements. Their concentrations in the bulk meats ranged from values of $\sim 1,000$ ppm, for phosphorus, to $\sim 10^{-6}$ ppm, for cerium. Interferences precluded the determination of the concentrations of two elements, sulfur and zirconium. Therefore, maximum values for their concentrations, based on the limits of sensitivity of the methods employed, are reported.

Qualitative neutron activation analyses resulted in detection of four platinum metals and four rare earth elements. Estimated values for the concentrations of these elements range from $\sim 10^{-3}$ ppm to $\sim 10^{-6}$ ppm in the bulk meats.

The reported concentrations for chlorine in the ash samples are considered accurate to $\pm 10\%$. However, calculation of chlorine concentrations in the bulk meats from the ash analyses is not deemed valid, because of possible loss of chlorine through volatilization during the ashing operation. Direct analysis of the bulk meat for chlorine is probably required to determine its fractional loss, if any, during ashing. Therefore, no value is reported for chlorine concentration in the bulk meats.

The extent to which the concentration of an individual element is constant in the four meats is indicated by equation 3:

$$\Delta c = \frac{\text{maximum observed concentration of an element in a meat}}{\text{minimum observed concentration of an element in a meat}} \quad [3]$$

The distribution of the elements for varying values of Δc is summarized in Table

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Table 6. Variations of elemental concentrations in ashed and bulk meat.

Concentration variation (Δc)	Number of elements	
	Ashed meat	Bulk meat
$\Delta c < 2$	2	6
$2 < \Delta c < 4$	2	3
$4 < \Delta c < 8$	9	5
$8 < \Delta c$	4	2
Total	17 ^a	16 ^a

^a Chlorine data for ash only.

6. The values of the respective concentrations of a majority of the elements in the four meats were constant within a factor of eight. Furthermore, the concentrations of 9 elements were consistent within a factor of four in the bulk meats, and in the ash samples the range of 13 elemental concentrations is greater than a factor of four. These variations seem to indicate that the concentrations of the majority of these elements in these meat samples are related to the organic content of the samples and do not maintain a constant relationship with the principal inorganic constituents.

In view of the significant variations in elemental concentrations observed in different samples of the same food (Winton and Winton, 1949), it does not appear valid to assume that the concentration data for these samples typify the four meats. However, the results may be indicative of the order of magnitude of the concentration of each element.

APPENDIX

Activation Analysis for Phosphorus and Sulfur

Neutron activation analysis for phosphorus, us-

ing the activation reaction $P^{31}(n, \gamma)P^{32}$, is complicated by the possible interference of sulfur and/or chlorine in the matrix sample from the reactions $S^{32}(n, p)P^{32}$ and $Cl^{35}(n, \alpha)P^{32}$ as indicated in Fig. 1. Similarly, activation analysis for sulfur, using the reaction $S^{34}(n, \gamma)S^{35}$, is subject to interference from the reaction $Cl^{35}(n, p)S^{35}$. Since 14.2-day P^{32} is the only neutron activation product available for phosphorus, and 87-day S^{35} is the only sulfur activation product with a half-life of useful magnitude for many applications, neutron activation analysis techniques for these two elements must take into account the need for corrections in the analytical data for the effects of the interfering elements.

Since the total quantity of S^{35} produced during the irradiation of the matrix is the sum of the contributions from the $S^{34}(n, \gamma)S^{35}$ and $Cl^{35}(n, p)S^{35}$ reactions, it is necessary to differentiate between the S^{35} produced by these two reactions. This can be accomplished by measuring the total S^{35} content of the matrix and the S^{35} produced from the chlorine in the matrix. The S^{35} produced from the sulfur in the matrix is the difference between these two quantities.

To make these measurements, the concentration of chlorine in the matrix must be determined by some method. Activation analysis, using the reaction $Cl^{35}(n, \gamma)Cl^{36}$, was the method chosen for this program.

In a similar manner, in phosphorus analyses, the total quantity of P^{32} must be determined along with the quantities produced by the $S^{32}(n, p)P^{32}$ and the $Cl^{35}(n, \alpha)P^{32}$ reactions. The amount of P^{32} due to activation of phosphorus is the balance remaining after subtraction of the contributions of the two interfering reactions from the total P^{32} found. Therefore, the concentrations of both sulfur and chlorine must be known in order to compute the concentration of phosphorus. The methods used for these determinations are detailed below.

Table 7. Method of analysis for sulfur.

Data required	Experimental operation
1. Cl concentration in ash ($\mu\text{g Cl/g ash}$)	1a Determine Cl^{36} in ash 1b Determine Cl^{36} in Cl comparator 1c Calculate Cl concentration
2. S^{35} production from Cl (dpm $S^{35}/\mu\text{g Cl}$)	2a Determine S^{35} in Cl comparator
3. S^{35} production from Cl in ash (dpm $S^{35}(\text{Cl})/\text{g ash}$)	3a Calculate: item (2) \times item (1)
4. Total S^{35} production in ash (dpm $S^{35}/\text{g ash}$)	4a Determine S^{35} in ash
5. S^{35} production from S in ash (dpm $S^{35}(\text{S})/\text{g ash}$)	5a Calculate: item (4) - item (3)
6. S^{35} production from S (dpm $S^{35}/\mu\text{g S}$)	6a Determine S^{35} in sulfur comparator
7. S concentration in ash ($\mu\text{g S/g ash}$)	7a Calculate: item (5) \div item (6)

Table 8. Method of analysis for phosphorus.

Data required	Experimental operations
1. Cl concentration in ash ($\mu\text{g Cl/g ash}$)	1a See operation no. 1c, Table 7
2. S concentration in ash ($\mu\text{g S/g ash}$)	2a See operation no. 7a, Table 7
3. P^{32} production from Cl (dpm $\text{P}^{32}/\mu\text{g Cl}$)	3a Determine P^{32} in Cl comparator
4. P^{32} production from Cl in ash (dpm $\text{P}^{32}(\text{Cl})/\text{g ash}$)	4a Calculate: item (3) \times item (1)
5. P^{32} production from S (dpm $\text{P}^{32}/\mu\text{g S}$)	5a Determine P^{32} in S comparator
6. P^{32} production from S in ash (dpm $\text{P}^{32}(\text{S})/\text{g ash}$)	6a Calculate: item (5) \times item (2)
7. Total P^{32} production in ash (dpm $\text{P}^{32}/\text{g ash}$)	7a Determine P^{32} in ash
8. P^{32} production from P in ash (dpm $\text{P}^{32}(\text{P})/\text{g ash}$)	8a Calculate: item (7) - [item (6) + item (4)]
9. P^{32} production from P (dpm $\text{P}^{32}/\mu\text{g P}$)	9a Determine P^{32} in P comparator
10. P concentration in ash ($\mu\text{g P/g ash}$)	10a Calculate: item (8) \div item (9)

SULFUR ANALYSES

Sulfur analyses require the irradiation of comparator samples for both sulfur and chlorine along with the ash samples. The data required and the method of obtaining them are shown in Table 7.

If the ratio of the concentration of chlorine to sulfur in the ash is relatively large, the calculation performed in Operation No. 5a in Table 7 may result in a small difference term of two large numbers. The smallest statistically significant value of this difference represents the minimum quantity of sulfur-produced S^{35} /gram ash that is detectable in the sample. In this case, the calculation in Operation No. 7a yields a value for the upper limit of the sulfur concentration in the ash.

PHOSPHORUS ANALYSES

The analysis for phosphorus requires that comparators for sulfur and chlorine be irradiated along with the matrix and the phosphorus comparators. The data required and the corresponding experimental operations are shown in Table 8.

The extent to which sulfur or chlorine may interfere in the phosphorus analysis is a function of the relative concentrations of the three elements. If the concentration of either sulfur or chlorine is much higher than that of phosphorus, the difference term in Operation No. 8a may be less than the statistical error of the other terms. In this case, analysis for phosphorus would be precluded. The limit of sensitivity for a given sample would then be determined in a manner analogous to that described for sulfur.

A special type of interference due to a second-order neutron reaction may also interfere in analyses for phosphorus. If large quantities of silicon are present, the reaction (Stehn and Clancy, 1956) $\text{Si}^{30}(n, \gamma)\text{Si}^{31} \beta^- \text{P}^{31}(n, \gamma)\text{P}^{32}$ may enhance the production of P^{32} and yield apparent phosphorus con-

centrations that are too large. However, it was shown (Kruger and Gruverman, 1962) that this reaction was not important in samples containing very high silicon concentrations that were irradiated under similar conditions at the Brookhaven Graphite Reactor. Therefore, it is estimated that this effect was unimportant in these samples.

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