Radionuclide Concentrations in Surface Air: Direct Relationship to Global Fallout

Abstract. *By relating the average monthly hemispheric concentrations of radionuclides in the surface air to the monthly hemispheric fallout of these nuclides, we have derived proportionality constants. For every disintegration per minute of a radioactive nuclide per 1000 cubic meters of the surface air in the Northern Hemisphere, 3.3 kilocuries of that nuclide will be deposited on the surface; in the Southern Hemisphere, 5.0 kilocuries will deposit for every disintegration per minute per 1000 cubic meters. On the basis of these factors, the global deposition of radionuclides can be estimated from a relatively small number of measurements of the radioactivity in the surface air.*

Using data obtained in two U.S. AEC Health and Safety Laboratory (HASL) sampling programs, we report here on an attempt to relate the concentration of strontium-90 in samples of surface air to the deposition of Sr^{90} on a worldwide scale. Since the beginning of 1963, HASL has been conducting measurements of the monthly concentrations of selected radionuclides in surface air (*1*) at about 25 stations, mostly along the 80th meridian ranging in latitude from Thule, Greenland, at about 76°N to the South Pole. Likewise, the monthly deposition of Sr^{90} has been measured since 1959 at more than 100 sites (*2, 3*).

If we assume that the concentration of a nuclide in surface air is directly proportional to the amount of that nuclide deposited on the surface of the earth, the following equation may be written:

$$D_n = X C_n \qquad (1)$$

where D_n is the deposition (in kilocuries) of a nuclide on the earth's surface during month n, X is a constant which relates the concentration of that nuclide in the surface air to its deposition, and C_n is the average concentration of that nuclide in surface air in disintegrations per minute per 1000 standard cubic meters (disintegrations per minute per 1000 m^3) during month n (calculated by weighting the concentrations measured in each 10° band of latitude according to the surface area of that latitude band).

This equation also assumes that, within each hemisphere, the troposphere is rapidly mixed longitudinally, and, therefore, that the sampling stations for the surface air are representative of their respective bands of latitude. Because the rate of interhemispheric exchange of tropospheric air is relatively slow (*4*), we consider the hemispheres to be completely insulated from each other, and separate equations are written for each.

If we substitute the measured monthly hemispheric depositions of Sr^{90} and the average monthly hemispheric concentrations of Sr^{90} in surface air into Eq. 1, the value of the constants in both the Northern and Southern hemispheres can be computed for each month in the period 1963 through 1967. The constants average 3.3 ± 0.7 kc per disintegration per minute per 1000 m^3 in the Northern Hemisphere and 5.0 ± 0.7 kc per disintegration per minute per 1000 m^3 in the Southern Hemisphere. Thus, for every disintegration per minute of Sr^{90} per 1000 m^3 of surface air in the Northern Hemisphere, 3.3 kc will be deposited on the surface of that hemisphere; similarly, for every disintegration per minute of Sr^{90} per 1000 m^3 of surface air in the Southern Hemisphere, 5.0 kc will be deposited.

The monthly values of X in each hemisphere exhibit seasonal variability. In general, it appears that the ratio of the deposition of a radionuclide to its concentration in surface air is lower in both hemispheres during the seasons of maximum fallout, the spring and winter. Because of this seasonal variability, deposition estimates for individual months may be in error by about 30 percent; however, estimates of annual deposition are not affected.

Table 1. Measured (M) and estimated (E) deposition of Sr^{90} (in kilocuries).

Year	Northern Hemisphere		Southern Hemisphere	
	M	E	M	E
1963	2620	3714	325	444
1964	1660	2083	436	426
1965	780	820	351	334
1966	330	376	208	282
1967	165	146	114	115
1968	190	181	90	120

Table 2. Worldwide deposition of Pu^{238} from a SNAP-9A power source (in kilocuries).

Year	Source of data		
	Measured deposition	Stratospheric depletion (*3, 4*)	Surface air
1967	4.2	5.1	5.7
1968	3.0	2.7	3.3

The two constants, X_N and X_S, are probably not the same value, because their averages, each based upon 60 individual monthly determinations, are not within one standard deviation. The significance of the difference is not immediately attributable to any particular meteorological phenomenon, although it is intriguing to speculate on the possible contributions from the relative areas of sea in the two hemispheres and from interhemispheric transfer in the troposphere.

Using the derived hemispheric constants and the measured concentrations of Sr^{90} in surface air for the period from 1963 through 1968, we calculated the annual Sr^{90} deposition. These estimated deposition values are compared with the measured values in Table 1. The agreement is reasonably good, especially in the most recent years. Although the relatively large deviation for 1963 and 1964 might suggest that X varies with the age of debris, there is no evidence of such a variation in 1967 and 1968 when the French and Chinese atmospheric tests occurred and most of the deposited radionuclides were "fresh."

This method of computing fallout from measured concentrations of Sr^{90} in surface air should be applicable to all materials dispersed like Sr^{90} in the atmosphere. The only other radionuclide whose fallout we could verify with deposition measurements was Pu^{238} which was disseminated during the reentry and burnup of a SNAP-9A (Systems for Nuclear Auxiliary Power) power source on a weather satellite (*5*). The estimates of plutonium-238 deposition for 1967 and 1968 are shown in Table 2, along with the measured values (*6*) and values derived from the annual depletion of the stratospheric reservoir extrapolated to the end of 1968 (*7*). Unfortunately, the data on Pu^{238} in surface air prior to 1967 are not reliable, and comparisons with early deposition measurements could not be made. The estimated values are in good agreement with the measured values from 1967 and 1968.

It appears, therefore, that the annual deposition of a nuclide can be accurately calculated from average monthly hemispheric concentrations of that nuclide in surface air. The significance of this relationship between the concentration of a radionuclide in surface air and its corresponding fallout lies in the relative simplicity of surface air sampling and the sensitivity of the existing system. If we can make worldwide estimates of fallout with acceptable accuracy, based simply upon a limited number of measurements of radionuclides in surface air, we can effect substantial savings in both time and funds. Further refinement of this relationship to smaller specific geographic areas should make possible estimates of local deposition and may lead to a greater understanding of hemispheric differences. In addition, the possibility of estimating the worldwide contamination from pollutants dispersed in a manner similar to that of global fallout is of great value.

MICHAEL T. KLEINMAN
HERBERT L. VOLCHOK
Health and Safety Laboratory,
U.S. Atomic Energy Commission,
New York, New York 10014

References

1. H. L. Volchok, *Nature* **206**, 4988 (1965); H. L. Volchok and M. T. Kleinman, *U.S. At. Energy Comm. Rep. HASL 204* (January 1969), p. I-17; *U.S. At. Energy Comm. Rep. HASL 207* (April 1969), p. C-1; H. L. Volchok, *U.S. At. Energy Comm. Rep. HASL 210* (July 1969), p. I-2.
2. H. L. Volchok and P. W. Krey, *J. Geophys. Res.* **72**, 20 (1967).
3. H. L. Volchok, *U.S. At. Energy Comm. Rep. HASL 200* (October 1968), p. I-2.
4. D. H. Pierson and R. S. Cambray, *Nature* **216**, 5117 (1967).
5. J. H. Harley, *U.S. At. Energy Comm. Rep. HASL 149* (October 1964), pp. 138–141.
6. H. L. Volchok, *U.S. At. Energy Comm. Rep. HASL 207* (April 1969).
7. K. Telegadas, *U.S. At. Energy Comm. Rep. HASL 204* (January 1969), p. I-2; personal communication.

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One Thousand Centuries of Climatic Record from Camp Century on the Greenland Ice Sheet

Abstract. *A correlation of time with depth has been evaluated for the Camp Century, Greenland, 1390 meter deep ice core. Oxygen isotopes in approximately 1600 samples throughout the core have been analyzed. Long-term variations in the isotopic composition of the ice reflect the climatic changes during the past nearly 100,000 years. Climatic oscillations with periods of 120, 940, and 13,000 years are observed.*

Use of the oxygen-18 concentration in glacier ice as an indicator of past climatic conditions was proposed in 1954 (*1*). The concentration of O^{18} in precipitation, particularly at high latitudes, is determined mainly by its temperature of formation. Decreasing temperature of formation leads to decreasing content of O^{18} in rain or snow. Analysis of stable isotopes has become an important method in studies of ice flow patterns in glaciers (*2–4*) and for determining past rates of accumulation in the upper layers (*3–5*).

In 1966, the U.S. Army Cold Regions Research and Engineering Laboratory (CRREL, now the U.S. Army Terrestrial Sciences Center) succeeded in drilling a core through the entire Greenland ice sheet at Camp Century (*6*). Certain physical and chemical studies have since been made over the upper 300 m of the 1390 m long, 12 cm in diameter, ice core (*7*).

We now show how a chronology can be obtained over nearly all of the vertical core profile by simple theoretical considerations; in particular, we show that climatic changes during the last 70,000 or 100,000 years are reflected as variations in the content of heavy isotopes in the core. The data represent nearly 1600 samples from 218 different core increments measured by mass spectrometry.

Few methods exist for dating a deep ice core. Some advances have been made with C^{14} dating (*8*) in which the carbon dioxide of occluded atmospheric air contained in huge blocks of glacier ice is measured for radiocarbon. Recently a technique for sampling C^{14} from ice *in situ* was tested in Greenland and Antarctica (*9*). These studies required a minimum of 1 ton of glacier ice.

Another way of dating an ice core is to determine the annual layers by measuring the seasonal oscillations in the O^{18} content in small vertical increments containing about 10 cm^3 of the core and then to count the summer maxima continuously from the surface downward. Unfortunately, various processes tend to diminish the isotopic gradients in snow and ice. For example, molecular diffusion in the firn and solid ice, hastened by the progressive plastic thinning of the layers with depth, gradually obliterates the oscillations that remain after firnification. This process alone establishes a limit of a few thousand years on the applicability of the stable isotope dating method on the Camp Century core (see *10*).

The approximate age of different sections of the core can be obtained by (i) considering the generally accepted glacier flow pattern outlined in Fig. 1, and (ii) making certain assumptions concerning the parameters that influence it. Correlation of the time as a function of depth (time scale) discussed here was calculated (*11*) on the basis of simple assumptions such as unchanged rate of accumulation a [35 cm of ice per year (*12*)], unchanged thickness of the ice sheet, H, and unchanged flow pattern back in time. Furthermore, any section of the core was assumed to originate from a location with essentially the same a and H as the Camp Century area. The following approximation to the horizontal velocity profile was used (see Fig. 1):

V_x proportional to $x \cdot y$ from
$y = 0$ to $y = h\ (= 400\ \text{m})$,

V_x proportional to x, but independent of
y, from $y = h$ to $y = H\ (= 1367\ \text{m})$,

x being the distance from the ice divide, y the distance from the bottom, and H the thickness of the ice sheet in meters of ice. Here H equals 1367 m of ice, instead of 1390 m of core, to correct for the low firn densities in the upper layers.

Basic mass balance considerations led to the following expressions for the vertical velocity components:

$$V_y = -\frac{a}{2H - h}(2y - h), \quad H \geqq y \geqq h$$

$$V_y = -\frac{a}{h(2H - h)}y^2, \quad h \geqq y \geqq 0$$

and, thereby, to the age of the ice at a distance y from the bottom:

$$t = \int_H^y \frac{dy}{V_y}$$

The results of the calculations are shown in Fig. 2, where the age of the core in years (before the present) is given together with the depth in meters from the 1966 surface.

The flow pattern discussed has recently been substantiated (*13*) insofar as it has explained the measured temperature profile (*7*) along the Camp Century borehole.

In the following, the isotopic composition of snow and ice will be denoted by the relative deviation $\delta(O^{18})$ in per mille, of the O^{18}/O^{16} ratio from that of Standard Mean Ocean Water (*14*). Unlike seasonal variations in $\delta(O^{18})$ of snow which are gradually obliterated in the ice by molecular diffusion, long-term variations are preserved for many tens of thousands of years. The mean isotopic composition of a section or long vertical increment of a core therefore reflects the average climatic conditions existing at the surface during the period of time when