Comparison of Atmospheric Transport Model Calculations with Observations of Radioactive Debris

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A semi-empirical box model of atmospheric transport that permits the calculation of stratospheric inventories, surface air concentrations, and deposition of debris injected into the stratosphere, mesosphere, or higher levels has been developed. The model divides the atmosphere of each hemisphere into three compartments: the atmosphere above 21 km, the stratosphere below 21 km, and the troposphere. The transfer between compartments follows first-order kinetics, although the season and height of injection regulate the onset of the transfer. The model adequately computed the fallout parameters of the specific injections of ^{102}Rh, ^{109}Cd, ^{238}Pu, and ^{90}Sr from the 1961–1962 tests, and ^{90}Sr from the sixth Chinese nuclear test in June 1967. It also predicted the 1969 fallout from the recent atmospheric tests.

INTRODUCTION

The reentry burnup of Snap-9A, the ^{238}Pu fueled nuclear space generator, and the controversy over the antiballistic missle system emphasize the need for a method that can predict the ultimate fate of a high-altitude injection of radioactivity in the atmosphere. In addition to the Snap-9A, several high-altitude injections have already occurred. In August 1958, ^{102}Rh was produced at 17°N at 43 km; ^{109}Cd was injected at 17°N at 400 km on July 19, 1962 [*List et al.*, 1966]. No formalized technique had been developed to predict accurately the surface air concentrations and deposition from such injections, however. To fill this need, we attempted a nonmeteorological approach to the problem and developed a semi-empirical box model of atmospheric transport. This report compares the model's calculations with the observations from a number of radioactive injections and presents the model's prediction of the fallout in 1969 to demonstrate the model's usefulness in estimating fallout parameters.

DESCRIPTION OF MODEL

This model, which is an extension of the concepts developed by *Volchok* [1966], *Peirson* [1967], and *Fabian* [1968], divides the atmosphere of each hemisphere into three compartments, as illustrated in Figure 1: the atmosphere above 21 km, the stratosphere below 21 km, and the troposphere. The model specifies that the transfer between compartments follows first-order kinetics and that the only dynamic interchange between hemispheres occurs between the stratospheres, as indicated by the constant k. For an injection above 21 km, however, the model assumes an initial division of debris between the hemispheres. This division may be the result of a mesospheric circulation (a theory first proposed by *Murgatroyd and Singleton* [1961]); this theory involves an ascent of air over the summer pole, a meridional flow from the northern to the southern hemisphere, and a descent of air over the winter pole. Other investigators have also proposed as a cause the exchange of air across the equator at altitudes in excess of 40 km [*Kellog and Schilling*, 1951; *Leovy*, 1964; *Martell*, 1968; *List and Telegadas*, 1969; *Newell et al.*, 1969]. Other phenomena, such as electrostatic interactions of the ionized debris with geomagnetic fields, may also affect this distribution between hemispheres.

The model assumes that the debris from an injection well above 21 km will arrive below 21 km during the first winter or spring season of each hemisphere after a 1-year delay from the time of injection. This assumption may also have some substantiation in Murgatroyd and Singleton's proposed circulation, but here it suffices to apply the assumption merely as a condition to the model. For a stratospheric

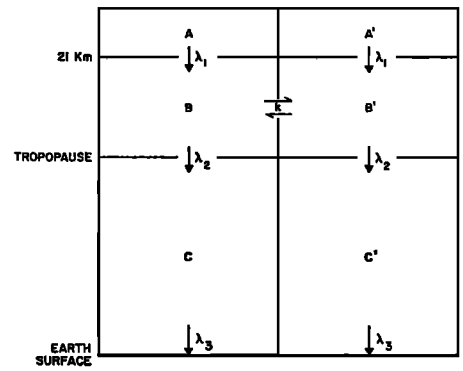

Fig. 1. Box model of atmospheric transport. The transfer between compartments follows first-order kinetics: $\lambda_1 = 0.116$ month^{-1}, $\lambda_2 = 0.0693$ month^{-1}, $\lambda_3 = 0.693$ month^{-1}, $k = 0.0116$ month^{-1}.

injection in the neighborhood of 21 km or below, in most cases it is assumed that transfer between compartments will begin during the first winter or spring season of each hemisphere after the time of injection, as was evidenced by the fallout behavior after the 1961–1962 tests [*Volchok*, 1968] and the Chinese test of June 1967 [*Krey et al.*, 1969].

In addition, the model assumes that the surface air concentration of a particular nuclide is directly proportional to the tropospheric inventory of that nuclide. The variables are defined as follows:

- t_0, time when major influx of debris from stratosphere to troposphere begins (if injection occurs above 21 km, t_0 is the time when such debris first enters the stratosphere below 21 km).
- t, time after t_0.
- A, hemispheric inventory above 21 km.
- A_0, amount of tracer injected into hemisphere above 21 km.
- B_0, amount of tracer injected into hemispheric stratosphere below 21 km.
- C, hemispheric inventory in the troposphere.
- D, cumulative hemispheric deposition on earth's surface from t_0 to t.
- S, mean hemispheric surface air concentration.
- F, proportionality constant relating surface air concentration to tropospheric inventory.

Primed terms, A', A_0', B', B_0', C', D', and S' represent the respective values in the opposite hemisphere.

From these definitions, we have

$$dA/dt = \lambda_1 A \tag{1}$$

$$dB/dt = \lambda_1 A - \lambda_2 B - kB + kB' \tag{2}$$

$$dC/dt = \lambda_2 B - \lambda_3 C \tag{3}$$

$$D = \int_{t_0}^{t} \lambda_3 C \tag{4}$$

$$S = FC \tag{5}$$

The solutions to these equations and the detailed formulation of the model are discussed elsewhere [*Krey and Krajewski*, 1969]. The proportionality factors for both hemispheres were calculated from the Health and Safety Laboratory (HASL) surface air and deposition data. The constants λ_1, λ_2, and k were evaluated by fitting the equations to the stratospheric inventories from the ^{109}Cd injection and to the stratospheric inventories and deposition of ^{90}Sr from 1961–1962 tests. The value λ_1 reflects a half-residence time of 6 months in the upper atmospheric compartment; λ_2 reflects 9 months in the stratosphere below 21 km; λ_3 reflects 30 days in the troposphere; and k reflects 60 months between the hemispheric stratospheres.

Comparisons with Previous Injections

Stratospheric inventories. It is instructive to observe how well the model estimates the fallout from earlier specific injections; we must remember that partial agreement is to be expected for the ^{109}Cd and 1961–1962 ^{90}Sr debris because of the way the constants were evaluated. Figure 2 shows good agreement between the measured [*Telegadas*, 1968] and the calculated inventories of ^{109}Cd in the northern and southern hemisphere stratospheres below 21 km.

To obtain this agreement, it was necessary to have estimates of two of the model's input parameters. The first estimate was the actual ^{109}Cd production in the detonation and its availability to each hemisphere. *Salter* [1964] reported that about 250 kc was produced, whereas *Feely* [1966] could only account for 75 kc by his stratospheric inventories. If we use our model as we inspect the stratospheric data, an initial available inventory of only 124 kc, with 90 kc in the southern hemisphere and 34 kc in the northern hemisphere is suggested. Because the injection occurred in July 1962, the meri-

dional flow in the mesosphere was from the northern to the southern hemisphere, which could explain the disproportionation between hemispheres. Another factor that may have had some bearing on the hemispheric distribution was the reported transport of ionized debris from this detonation to the southern magnetic conjugate point by an interaction with the earth's geomagnetic field [*D'Arcy and Colgate*, 1965]. Consequently, of the estimated 250 kc produced, either half escaped the atmosphere at the time of the detonation or there was an error in that estimate.

The second estimate was the adjustment of the initial appearance of ^{109}Cd below 21 km. According to the model, the initial appearance should have occurred in July 1963 in the southern hemisphere and in January 1964 in the northern hemisphere. Although the appearance did occur in the prescribed months, the fit as shown between the measured and estimated inventories for both hemispheres was markedly improved by delaying the predicted time of initial appearance by a relatively short period of 3 months.

The inventories of ^{90}Sr from the 1961–1962 tests in the model's atmospheric compartments were calculated for January 1963 from Telegadas' data (private communciation, 1968). Figure 3 shows that the model then adequately estimated the measured stratospheric inventories below 21 km in the northern and southern hemispheres from 1963 through early 1966 [*Telegadas*, 1968]. Because most of the ^{90}Sr in the stratosphere in January 1963 was produced by the 1962 tests, it is assumed in these computations that the fallout began in January 1963 in the northern hemisphere and in July 1963 in the southern hemisphere.

Working on the ^{102}Rh problem, *Machta* [1964] inventoried the stratosphere for this nuclide in May 1961 and found about 1 Mc below 30 km, with equal distribution between hemispheres. Although ^{102}Rh production had been estimated at 3 Mc [*List et al.*, 1966], Machta's findings suggest that only 1 Mc was available to fallout. Because the injection occurred in August during the late winter season in the southern hemisphere, the mesospheric meridional flow toward the southern hemisphere was apparently on the wane; this phenomenon allowed for an equal split of the ^{102}Rh between

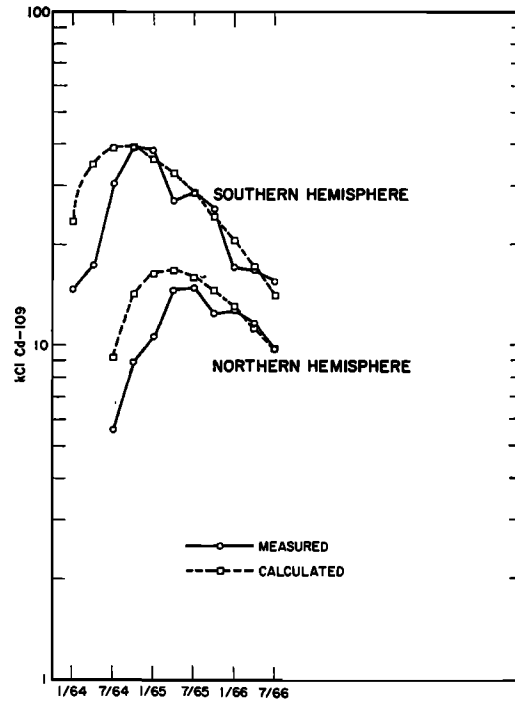

Fig. 2. The ^{109}Cd stratospheric inventory below 21 km. The inventories are decay corrected to July 9, 1962, the date of the ^{109}Cd injection at 400 km over 17°N.

the hemispheres. On the basis of these two assumptions, the model calculated the stratospheric inventories of ^{102}Rh below 21 km, from early 1960 to late 1961, fairly well, as shown in Figure 4.

The reentry burnup of the Snap-9A generator offered an ideal test for the model. The disintegration of this generator released 17 kc of ^{238}Pu at 46 km over the Indian Ocean in April 1964 [*Krey*, 1967]. At that time, the meridional flow of the upper altitudes from the northern to the southern hemispheres might have already begun or was about to begin. Therefore, it was assumed that the disproportionation between hemispheres was the same as that for the ^{109}Cd (73% in the southern and 27% in the northern hemisphere). Figure 5 shows the good agreement between the measured [*Telegadas*, 1969, and personal communication, 1969] and calculated stratospheric inventories of ^{238}Pu below 21 in both hemispheres.

On June 17, 1967, Communist China detonated a 3-MT nuclear explosion at Lop Nor

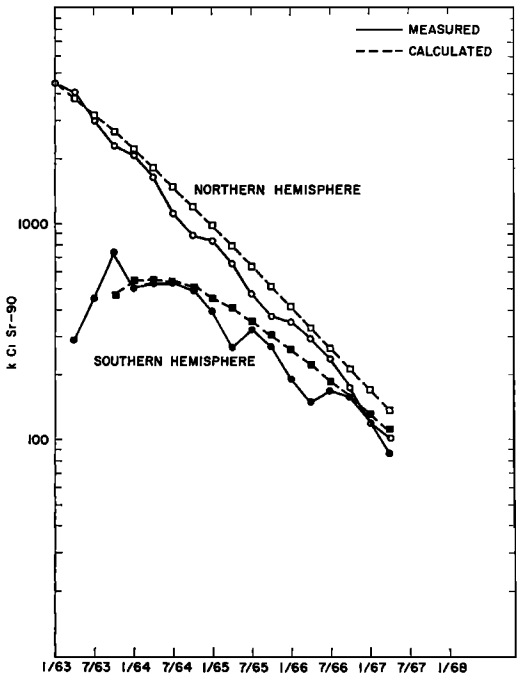

Fig. 3. The ^{90}Sr stratospheric inventory below 21 km. This debris is from the 1961–1962 nuclear weapons tests and is decay corrected to collection time.

44°N and injected 170 kc of ^{90}Sr into the stratosphere [*Krey et al.*, 1969]. This was the first large yield nuclear detonation in the midlatitudes of the northern hemisphere and is an interesting case to test our model. Almost all the debris was injected into the lower stratosphere below 21 km. The solid line in Figure 6 illustrates the trend of the ^{90}Sr burden of the northern hemisphere stratosphere from the sixth Chinese test. It is clear that fallout of this debris from the stratosphere began in November 1967 and not in January 1968 as was predicted by the model. If the model is adjusted for this 2-month period, the agreement between the measured [*Krey et al.*, 1969] and calculated inventories in the northern hemisphere, as shown in Figure 6 is good.

Surface air concentrations and deposition. These comparisons suggest that the model can satisfactorily describe the stratospheric history of an injection into any of its upper atmospheric compartments. To test the accuracy of the model for predicting surface air concentration and deposition, reference is made to these same injections. For ^{90}Sr from the 1961–1962 tests, good agreement was obtained between the measured [*Kleinman and Volchok*, 1969] and calculated mean annual surface air concentrations in both hemispheres, as shown in Figure 7. Figure 8 demonstrates that the model also adequately calculated the annual deposition of ^{90}Sr in each hemisphere from these tests [*Kleinman and Volchok*, 1969]. In these computations, as in those for the stratospheric inventories for the 1961–1962 ^{90}Sr debris, it is assumed that the fallout in the southern hemisphere began in July 1963. Consequently, the 1963 values for the southern hemisphere in Figures 7 and 8 are representative of only the last half of the year.

Unfortunately, the fallout measurements of all the other nuclides studied were far less complete, thereby weakening the comparisons with the model's calculations. Nevertheless, Table 1 presents a summary of these comparisons of surface air concentration and deposition in the form of ratios of the calculated values to the measured values. A ratio of 1 represents perfect

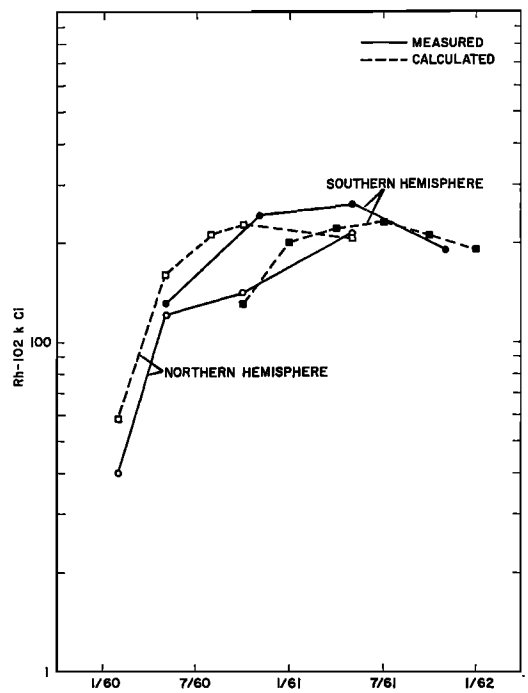

Fig. 4. The ^{102}Rh stratospheric inventory below 21 km. This tracer was injected high in the equatorial stratosphere on August 12, 1958; the inventories are decay corrected to that date.

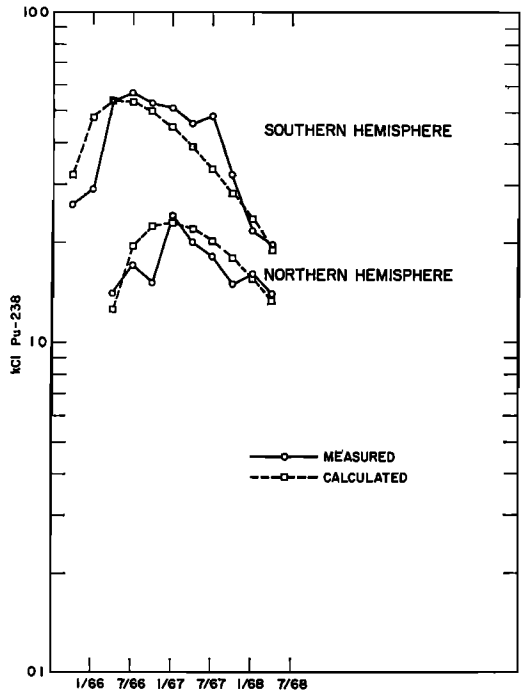

Fig. 5. The ^{238}Pu stratospheric inventory below 21 km. This debris was produced by the atmospheric reentry burnup of the Snap-9A nuclear fueled space generator on April 21, 1964.

agreement. For ^{109}Cd during the years 1964–1966, the data show that the model calculated the mean annual hemispheric surface air concentrations in both hemispheres [*Volchok and Kleinman*, 1969] to within a factor of about 2. These measured concentrations of ^{109}Cd are hardly precise, are spotty, and in many cases are inconsistent. Because of this uncertainty, the agreement is considered acceptable.

To obtain measurable ^{109}Cd deposition data, reference must be made to the special large-area collectors employed by the USSR at 40°N latitude in 1964 and 1965 [*Fedorov et al.*, 1967]. The annual northern hemispheric deposit of ^{109}Cd was estimated from these measurements by means of the relationships derived from the ^{90}Sr distributions with latitude [*Hardy*, 1968]. Whereas the model's calculations are high by as much as a factor of 2.5, the agreement is again considered reasonable because of the paucity of the ^{109}Cd measurements.

For ^{102}Rh, global or even hemispheric measurements of surface air concentrations have not been made. However, Gustafson analyzed monthly surface air collections at Argonne National Laboratory in 1960 and part of 1961 [*Gustafson*, 1962]. The mean annual hemispheric concentration in surface air was estimated from Gustafson's measurements by comparing them with ^{90}Sr measurements in surface air. The model's calculations, as shown in Table 1, agreed with these estimates to within a factor of 2.

Similarly, global deposition of ^{102}Rh was not measured, although Volchok has estimated worldwide values from measurements in New York City [*Volchok*, 1966]. Table 1 demonstrates good agreement between the model's calculations and Volchok's estimates, except for a factor of almost 2 in 1961. This deposition agreement held in 1963, despite the report that additional ^{102}Rh was injected into the lower stratosphere by the 1962 United States test series [*Feely et al.*, 1963].

For ^{238}Pu, Table 1 demonstrates that the model

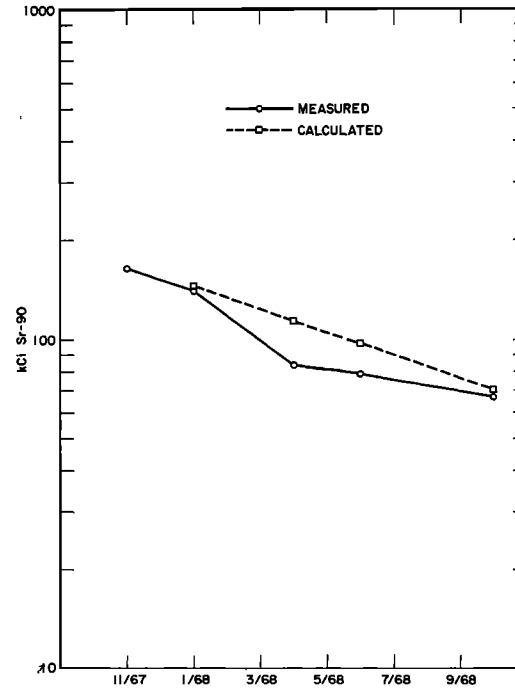

Fig. 6. The ^{90}Sr stratospheric inventory below 21 km in the northern hemisphere from the sixth Chinese nuclear weapons test of June 17, 1967. This was the first megaton detonation in the atmosphere of the midlatitudes of the northern hemisphere.

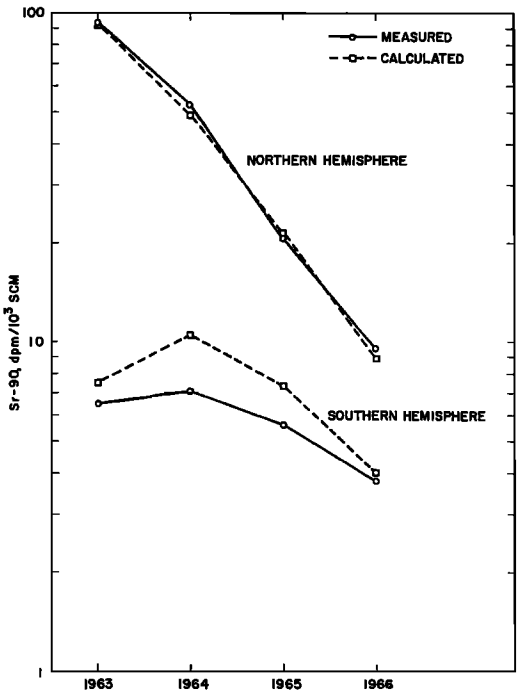

Fig. 7. The mean annual surface air concentration of ^{90}Sr from the 1961–1962 nuclear weapons tests. The 1963 values in the southern hemisphere are representative only of the last half of the year, because the model assumes that the debris observed in the stratosphere in January 1963, from which the calculations are made, will not begin to fallout significantly in the southern hemisphere until July 1963.

calculated the mean annual hemispheric concentrations of ^{238}Pu in surface air [*Volchok and Kleinman*, 1969] quite well in the southern hemisphere and within a factor of 2 in the northern hemisphere. *Volchok* [1969] has estimated the hemispheric deposition of ^{238}Pu from measurements in New York City and Melbourne, Australia. The ^{238}Pu ratios in Table 1 also indicate that the model calculated these hemispheric depositions satisfactorily, with the exception of the northern hemisphere in 1966.

For China's sixth nuclear weapons test, only 1 year of fallout data are now available for comparison. On the basis of ^{144}Ce measurements in deposition and surface air samples [*Volchok and Kleinman*, 1969], the contributions of Chinese debris to each type of sample could be made. Table 1 demonstrates that the model's calculations are in good agreement with these estimates.

The averages of the respective data in Table 1 indicate that in general there is no bias in the model's calculations of annual hemispheric surface air concentrations and deposition (ratios of 1.03 and 1.21), although individual variations of a factor of 2 were common. The model actually computes monthly deposition and surface air concentrations. These monthly estimates are only approximate, however, because in its present form the model does not account for seasonal variations in the transfer constants. Nevertheless, the annual means are sufficiently accurate. Modifications are in progress to allow for accurate monthly or seasonal computation.

Predictions

The model permits forecasting the fallout from the 1968 atmospheric tests, but no measurements are yet available for a reasonable comparison. According to our measurements, the two thermonuclear tests conducted by the French in the southern hemisphere on August 24 and September 9, 1968, injected a total ^{90}Sr input of 130 kc into the stratosphere. The

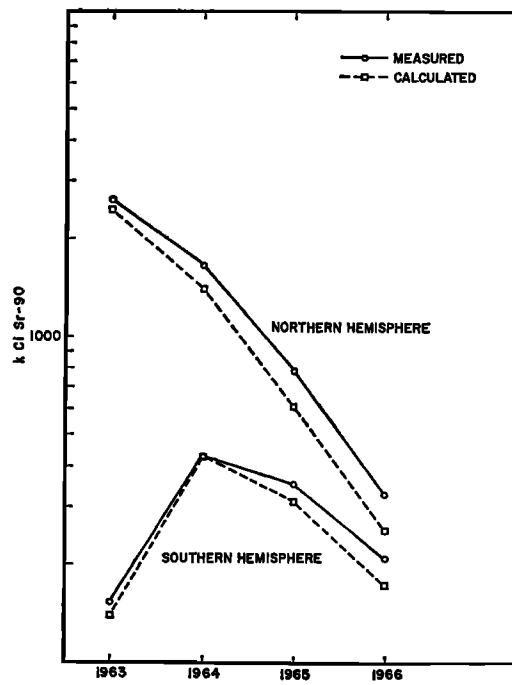

Fig. 8. The annual deposition of ^{90}Sr from the 1961–1962 nuclear weapons tests. The deposition in the southern hemisphere in 1963 is the accumulation from only July through December 1963.

TABLE 1. Ratio of Predicted/Measured Fallout

Nuclide	Year	Surface Air Concentration N. Hemisphere	Surface Air Concentration S. Hemisphere	Deposition N. Hemisphere	Deposition S. Hemisphere	Deposition World
^{109}Cd	1964	0.4	0.5	1.1		
	1965	1.1	0.9	2.6		
	1966	0.4	0.3			
^{102}Rh	1960	2.3				
	1961	1.2				1.7
	1962					1.1
	1963					0.8
^{238}Pu	1966	2.0	1.4	4.0*	1.6	
	1967	1.8	1.0	0.8	1.2	
	1968	1.2	0.9	0.7	0.8	
^{90}Sr from China's 6th test	1968	0.7		0.9		
	Ave.	1.03		Ave.	1.21	

* Omitted from average.

eighth Chinese test of December 28, 1968, injected 115 kc of ^{90}Sr into the northern hemisphere stratosphere. By allowing for immediate fallout of the French debris, we estimate that the stratospheric burden of ^{90}Sr in January 1969 was 200 and 132 kc in the northern and southern hemispheres, respectively, and essentially all was contained below 21 km. In the northern hemisphere in 1969, the model predicts a mean annual hemispheric surface air concentration of 3.8 dpm/10^3 SCM and an annual deposition of 100 kc of ^{90}Sr. In the southern hemisphere in 1969, the fallout predictions are 1.7 dpm/10^3 SCM and 70 kc of ^{90}Sr for air concentration and deposition, respectively.

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