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SNOW AND RAIN WASHOUT COEFFICIENTS FOR INORGANIC IODINE VAPOR

R. J. Engelmann and R. W. Perkins*

ABSTRACT

Washout coefficients for gaseous iodine in snowfall and an estimate of the coefficient for rainfall have been experimentally determined for the first time. The coefficient for rain is in agreement with theory. The coefficient for snow appears to depend upon crystal type and size as well as upon snowfall rate.

Woefully little is known about the effectiveness of snow in scrubbing the atmosphere, and in the past there have been no reported measurements of the efficiencies with which atmospheric constituents are removed by rain or snow. Theoretical calculations of the collection by raindrops of soluble gases have been reported by Chamberlain⁽¹⁾; however, similar calculations for snow would require a knowledge of the electrostatic capacitance of the snow crystals, their surface area, and their number per volume in air. A knowledge of the removal efficiencies of atmospheric constituents by precipitation is essential in the study of many basic problems in atmospheric physics, and the washout coefficients of both gaseous and particulate materials by both rain and snow are being studied. This information is of particular importance in explaining the fall-out rates and mechanisms of airborne radionuclides. Several experiments to determine the washout coefficients of radioiodine vapor by snow, and preliminary experiments to determine its washout by rain have been conducted during the past year using the minute amounts of I^{131} which are present in some stack gas effluents at the Hanford Atomic Plant.

This radioiodine in these effluents is all in the vapor state and is about one half inorganic (HI and I_2) and half in the form of organic iodides (apparently alkyl iodides)⁽²⁾.

Experiments were designed to determine the removal of the I^{131} from the stack plume by comparing the I^{131} carried down by precipitation in a sampling arc which subtended the plume with the total I^{131} leaving the stack. The data from these studies were treated in a manner similar to that employed by May⁽³⁾ in his study of the washout of lycopodium spores.

The quantity of I^{131} washed out is used to determine the washout coefficient (fraction scavenged per time) for that rain or snowfall, using the following formula:

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$$A = V \Delta y \Sigma c / Q t A$$

where A = washout coefficient

V = wind speed at plume height

Δy = distance between samplers

Σc = total material collected in all samplers, less background

Q = source release rate

t = sampling time

A = area of one sampler

The sample collectors are one square meter saucer-like dishes formed by attaching polyethelene sheets to large hoops made of plastic hose. Fifty to seventy of these are exposed on a line or arc transverse to the stack plume overhead. The collected rain or snow is gathered after a few hundredths inch has fallen. There appear to be no splash residue problems with these simple basins.

The organic and inorganic radioiodine was separated from the melted snow or rain by standard chemical techniques which were modified to accommodate large volumes⁽⁴⁾ and it was counted in a high sensitivity gamma-ray spectrometer⁽⁵⁾.

Six useful washout tests were conducted this winter. The first, which provided an estimate of the washout of inorganic iodine by rain, we call PUREX-1. It was conducted November 3, 1964, in a light natural rain of 0.37 mm/hr. The sampling arc of hoops was limited by construction activity on the south end, and the wind at stack height occasionally carried the stack plume past the end of the arc. We can only estimate what fraction of the time the plume was not sampled, perhaps a third, and, therefore, can only estimate the washout coefficient for inorganic gaseous iodine. The estimated value from this test was $2.8 \times 10^{-5} \text{ sec}^{-1}$ V which is not significantly different from that theoretically predicted by Chamberlain for this rainfall rate of 0.37 mm/hr. We found that organic iodine was washed out with a distinctly lower coefficient, but detection efficiency of the counter was insufficient to provide a quantitative result.

Tests PUREX-2 and PUREX-3 were actually the successive samples in a single snowfall of December 14. This snow consisted of spatial dendrites,

much of it in agglomerates up to 3/4 inch in diameter. It was a "wet" snow, following a short rain at the start of the sampling. The source strength changed by nearly a factor of two during the test, as determined by two consecutive stack gas samples. The washout for the entire snowfall of 1.0 mm/hr (water equivalent) was $4.5 \times 10^{-5} \text{ sec}^{-1}$ for inorganic gaseous iodine. This was the result of a coefficient of 4.2×10^{-5} in the first part of the snow, and 4.7×10^{-5} in the last of the snow, at precipitation rates of 1.2 and 0.79 mm/hr, respectively.

It is interesting that the higher coefficient was measured with the lighter snow. This may well be due to sampling errors in these first tests, but the authors instead feel that it is due to the smaller fall velocity of the flakes in the lighter snow. Whereas in the case of rain, one may assume that the gaseous washout coefficient varies with about the 0.6 power of the precipitation rate, in the case of snow one must consider the type and size of flake.

Tests REDOX-4 of January 10, 1965, and REDOX-6 and 7 of January 23, 1965, were conducted in snows composed almost entirely of columnar crystals. These tests were conducted using a different stack source but the same techniques. Tests 4 and 7 yielded essentially the same washout coefficient for about the same precipitation rate (see Figure 1). The washout coefficient observed in Test 6 suggests a linearly increase in the washout coefficient with the snowfall rate.

Lacking theoretical predictions and additional measurements, but still requiring a basis for prediction, we have speculated that the washout coefficient of gaseous inorganic iodine varies with snowfall rate as indicated in Figure 1. We assume that there is a parallel line for each type of crystal, the position of which is determined by the snow crystal's terminal velocity and electrostatic capacitance.

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Figure 1 Washout coefficients of inorganic iodine vapor by snow as a function of snowfall rate.

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