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SCAVENGING IN PERSPECTIVE

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SCAVENGING IN PERSPECTIVE

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Thank you, Mr. Chairman. I also would like to welcome you to this workshop on precipitation scavenging. We do have some lay people and persons from adjacent disciplines here with us, so I would like to start with some perspective on the earlier work on this subject. During this workshop we will be brought up-to-date on many of the items which I will mention.

We have generally thought that the precipitation scavenging process has three major divisions: (1) delivery or transport of the material to the scavenging site, (2) in-cloud scavenging by the cloud elements and precipitation, usually called rainout and snowout, and (3) below-cloud scavenging by the precipitation, usually called washout. Material from low-elevation sources will be transported with the low-elevation winds and diffused upward by low-level turbulence. Once in the vicinity of clouds it may be carried into those clouds by organized vertical motions. When the material is from high elevations (the stratosphere), the general circulation of the earth's atmosphere and the exchange mechanism at the tropopause have control of the delivery.

There are several good starting points for the study of scavenging. Junge (1963) and Facy (1962) have looked at in-cloud scavenging, but with differing conclusions. Fletcher (1962) and

Mason (1957) have published texts on precipitation and cloud physics. Chamberlain (1953) appears to have attempted the first broad application of scavenging equations, and his approach is still used. Fuchs (1964) has presented an extensive volume on the mechanics of aerosols. Reiter (1964) and Danielsen (1964) serve as introductions to the controversy on stratosphere-troposphere exchange important to our fallout studies and now to the potential problems in jet transport.

Chamberlain used Langmuir's scavenging efficiencies and Best's (1950) fitted raindrop spectra to calculate washout coefficients for various rainfall rates and cloud droplet sizes. Similar calculations have been made by May (1958) using the somewhat higher efficiencies of Mason, which gave scavenging coefficients about 10% greater than Chamberlain's.

A different result is obtained using the efficiencies of Kinzer and Cobb (1958) with Washington State and Indian rain spectra reported by Engelmann (1963). Even though the Kinzer and Cobb efficiencies are greater than Mason's for the more numerous small drops, the calculated washout coefficients are lower. This is due to the greater contribution to the cross-sectional scavenging area made by the larger drops, where Mason's efficiencies are larger than Kinzer and Cobb's.

In about 1957 it appeared that the coefficients calculated from the Langmuir or Mason efficiencies might be regarded as slight over-estimates in the absence of significant electrical charges on the rain. Both the measurement of lower coefficients by Kinzer and Cobb and by Engelmann and the possibility of retention efficiencies less than one supported that view.

For particles less than about one micron in diameter, the calculations based only on inertial effects in idealized flow patterns showed negligible washout. They could not be considered at all reliable, however, owing to the presence of electrical charges, which increased the washout. Solutions for selected particle and drop charges had been made by several workers, but there was insufficient information for safely extrapolating these predictions to other charge values. In addition, there was insufficient information on the normal charges existent in rain and clouds.

There were also no measurements on natural washout of these smaller particles, although some research plans did include them. It was apparent that the problems of small-particle measurement, low collection efficiencies, and non-uniform raindrop charge would cause difficulty in the performance of these experiments.

Scavenging by snow offered additional complexities. The large numbers of frozen cloud droplets attached to snowflakes established that snow was an effective scavenger for at least large particulates. This effectiveness varied with types of crystal, its shape, fall speed, and electrical charge.

For the same rate of precipitation, we expected a greater scavenging of smaller particles by the slower falling snowflake, with its larger area and probable electrostatic charge, than by its equivalent waterdrop. Large numbers of Aitken particles (diameter <0.02 microns) had been observed on snowflakes. A few washout coefficients in rain and snow for submicron particles of silver iodide, scandium, and cesium salts were published (PNL, 1967).

Scavenging of gases was predicted on the basis of molecular diffusion to the drop or droplet in accordance with the vapor pressures and solubilities of the free and collected gases. Theoretically, trace quantities of insoluble gases were treated as soluble in the scavenging processes.

Measurements of the washout of bromine gas by Engelmann, Perkins, Hagen, and Haller (1966) verified the theoretical predictions in rain. However, the measurements for iodine gas yielded washout coefficients one and two orders of magnitude lower than those predicted and showed no relationship to precipitation rate. They proposed that the reaction rate with water, rather than the total solubility, determined the washout rates.

The washout of bromine and inorganic iodine was also measured in snow. The lower coefficients in snow compared to rain supported the importance of reaction rates. It appeared that, although the large surface area and small terminal velocity of snow crystals increased the scavenging of gases, these factors were not able to balance the lower adsorption rates on ice crystals. Washout coefficients for other gases with similar reaction rates should have compared to those of bromine and iodine as the ratio of their diffusivities in air.

However, when process-plant inorganic iodine was used as a tracer, much higher washout rates were observed. In explanation of the high washout, it was noted that there was sufficient water vapor in the plant stack exhausts to produce a cloud of water droplets on a rainy day. If all of the radioiodine (or other gas) in the stack exhausts were swept up by the cloud droplets during formation, subsequent washout of iodine would have been at the rate appropriate to cloud drops. Similar washout rates were expected for reactive gases in other stack exhausts with higher water content. Again, reaction rates influenced the takeup during the condensation. In these tests, washout coefficients for organic forms of iodine were about 1% as large as for inorganic forms.

Scavenging of materials from the air by hydrometeors of all types is particularly intriguing because it represents perhaps the most significant natural mechanism for cleansing the air. However, cleansing of the air results in deposition of the pollutants on-or-in people, animals, vegetation, land, water, and structures. The impact of deposition processes on the biosphere poses a host of new "formidables".

Let us take lead as an example. Lazrus, et al (1970), reported metal was found in precipitation over the United States during six months of 1966 and 1967. Measurements included lead, zinc, copper, iron, manganese, and nickel. Their results suggested that the contaminant concentration in the atmosphere was relatively independent of the total amount of precipitation. It was, however, not completely independent of amount of rainfall, since frequent showers kept the air cleaner and contaminant concentration tended to decrease with duration of rainfall.

They point out certain generalizations applicable to all metals. The northwest portion of the United States was conspicuously low in contamination. The northeast was relatively high. The southeast and southwest varied from low to moderate in contamination, depending upon the metal. The analysis suggested that the concentration patterns of these minor trace contaminants could be engendered primarily by human activity. Concentrations tended to increase in areas of mining and manufacturing.

Because of the biological activity of several of these metals, there has been increasing concern over their allowable concentration limits in drinking water. One source of contamination is the atmosphere, from which aerosols may be deposited in surface water supplies by dry fallout or by entrapment in precipitation. Comparing average concentrations found in precipitation with those in surface water supplies before treatment indicated that there was about twice as much lead in atmospheric precipitation as in water supplies. They pointed out that this ratio implied the existencc of a process whereby lead is depleted after precipitation reaches the surface. In the case of copper, the quantity found in precipitation could account for the average value found in surface water. Nickel and manganese, however, must have had other sources in addition to atmospheric precipitation. The correlation relating lead in rainwater to sale of gasoline in the area was found to be significant when stations unduly affected by industrial activities were excluded.

Tatsumoto and Patterson (1963) reported lead concentrations in snow four orders of magnitude greater than the amount attributable to naturally formed dust in a remote area 500 miles east of the Los Angeles Basin at an altitude of 7000 feet. They also estimated that the lead concentration in the surface layer of the Pacific Ocean off the coast of southern California had increased tenfold during the past thirty to forty years. Recent ice samples taken in the Tatra Mountains contained 16 times more lead than did samples dating a century ago. Greenland snow exhibited about the same increase.

The impact on the biosphere of lead released to the atmosphere and brought to the ground through the various scavenging mechanism requires further elucidation. Lead was selected as an example because the concentrations in some localities are significantly high with respect to the allowable concentrations in drinking water. There are a myriad of trace elements and pollutants to consider.

We are hearing more and more about precipitation displaying a weekly cycle reflecting the consequence of man's activities. Recent reports of increased precipitation downwind of urban and industriallized areas offer some intriguing speculation. Advertent and inadvertent weather modification may become equally significant research activities when one considers the full impact on the ecosystems. The behavior of oxides of nitrogen from their source as pollutants or products of lightning discharges to their ultimate incorporation in the life systems holds fascinating prospects for understanding these systems. Many other examples are evident.

During our deliberations on some of the sophisticated intricacies of the scavenging process, I hope we keep in mind some of the greater problems to be solved using this technology. Thank you very much.

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