POLLUTANT WET DEPOSITION MECHANISMS IN PRECIPITATION AND FOG WATER

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ABSTRACT. Wet deposition of acid-related substances takes place by two processes: precipitation scavenging and fog water impaction/sedimentation on natural surfaces. The relative importance of each deposition pathway depends on the frequency of occurrence of precipitation or fog, the magnitude of the event and the efficiency of pollutant removal by each mechanism. The latter, in turn, is governed by the type of cloud or fog, complex precipitation formation mechanisms and cloud-surface interactions. These factors are examined in the light of our current knowledge. Particular emphasis is placed on how cloud micro-physical as well as air and precipitation measurements, made aloft by aircraft and at the ground, have been used to further our knowledge of wet deposition mechanisms. Future research is needed to quantify the importance of the fog-water deposition pathway in eastern North America to better understand the interaction of gaseous pollutants with cloud and fog-water and to improve our knowledge of pollutant scavenging processes in mesoscale and synoptic weather systems.

1. INTRODUCTION

Particles and gases are removed from the atmosphere by precipitation and fog-water deposition (wet deposition) as well as by direct uptake at the earth's surface (dry deposition). The deposition of relatively clean dew or frost (Wisniewski, 1982) is expected to lead to little direct pollutant deposition. Indirectly, however, by altering surface roughness and wetness it may enhance dry deposition of particles and gases. The wet deposition process is a major pathway of acidic pollutants from the the atmosphere to the biosphere. In eastern North America, approximately 71% of the total S reaching the earth annually is wet deposited by precipitation (Galloway and Whelpdale, 1980). Four years of air and precipitation chemistry observations in the southern part of eastern Canada show that wet deposition by precipitation accounts for 70 to 80% of the total annual atmospheric deposition of S and N (Barrie and Sirios, 1986). The wet

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deposition process involves complex chemical transformation of SO_2 and NO_2 to H_2SO_4 and HNO_3 in a cloud, as well as the processes of cloud formation, precipitation formation and, in the case of fog water deposition, the interaction of fog droplets with the Earth's surface. In this paper, a brief review is given of the mechanisms of wet deposition and the methods that are used to gain a better understanding of them.

Precipitation at mid-latitudes is usually associated with extra-tropical cyclones (low pressure zones) which have cloud patterns similar to that shown by the satellite photo in Figure 1. These cyclones typically involve the intrusion of a tongue of warm southern air northward from the Gulf of Mexico or Caribbean that overrides colder northern air. This in turn is undercut from the west-northwest by colder northern air. A warm front marks the boundary between advancing warm air and the colder air over which it is rising. A cold front lies between the advancing cold air in the northwest and the

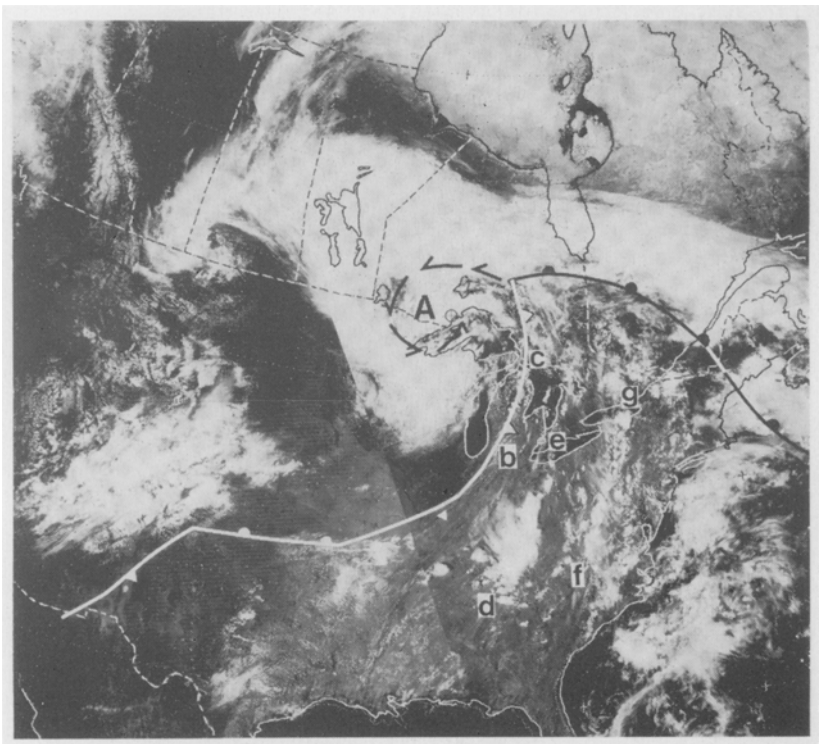

Figure 1: A satellite photo in the visible, illustrating the typical cloud and frontal structure associated with an extra-tropical cyclone occurring May 12, 1985. Δ marks a cold front, a warm front. Convective cloud bands are visible in the warm sector between b and c, e and d as well as g and f. Warm frontal cloud is present in central Quebec and northwestern Ontario.

warm air. Different pollutant removal situations arise depending on location relative to these fronts in the cyclone. In the vicinity of the cold front and in the warm sector ahead of the cold front, convective clouds of 2 to 20 km diameter form in clusters. Each cloud draws air from the near-surface boundary layer below. Ahead of the surface warm front the situation is quite different. Clouds form over large areas as a result of widespread uplift. The air in which they form originates from the near-surface layer several hundreds of kilometres to the south. Precipitation falls from the overriding warm air into cooler air below.

The nature of precipitating clouds in extra-tropical cyclones has been extensively studied using satellite photography, radar, and aircraft (Parsons and Hobbs, 1983). Precipitation tends to occur from organized bands of convective cells (see examples in the warm sector of Figure 1). This is true even in the warm frontal sector. The nature of a precipitation event associated with the passage of an extra-tropical cyclone depends on the intensity of the cyclone, its speed and the location of the sampling site relative to it. A climatology of storm features in the northeastern United States (Thorp and Scott, 1982) shows that, in general, there is a marked difference in storm types between winter and summer. In winter, storms move twice as fast across the region (15 m s^{-1} versus 7 m s^{-1}) but their extent is such that the average duration is much longer (26 hr) than in summer (2.5 hr). Precipitation intensity is lower in winter than in summer (0.9 versus 2.5 mm hr^{-1}) . The fraction of storms that are isolated convective bands is higher in summer than in winter.

Fogs can be simply regarded as clouds in contact with the Earth's surface. In some cases, this is literally true, for example when a layer of cloud passes over a mountain. In other cases, the fog is formed by processes (different from those of cloud formation) that take place near the Earth's surface. If air is cooled sufficiently, the water vapour in it will condense into liquid drops. The cooling can be produced by several different meteorological situations leading to different types of fogs. At night, the earth loses heat through longwave radiation. Sufficient cooling leads to 'radiation fog'. Light winds ($<$ 10 km hr^{-1}) promote the development by mixing cool, moist, near-surface air to moderate heights. At higher wind speeds, mixing with the drier air aloft tends to inhibit fog formation. When warm moist air moves over a cold surface, the cooling that results can produce an 'advection fog'. Most coastal fogs are of this type. Air that is forced to rise over a topographical barrier experiences decreasing atmospheric pressure that produces expansion and cooling of the air. These conditions can generate 'upslope fog'. Other types of fog are less important vehicles of wet deposition. 'Steam fog' is produced by the condensation of water vapour from a relatively warm water surface in the cooler air above. 'Ice fog' is produced at temperatures below -30°C when water vapor sublimes directly onto ice nucleii (particles) thus forming tiny ice crystals. This illustrates a characteristic of fogs (and clouds) that is important in the deposition of pollutants, namely, they remain liquid (containing supercooled droplets) at temperatures as low as $\approx$ -20°C and

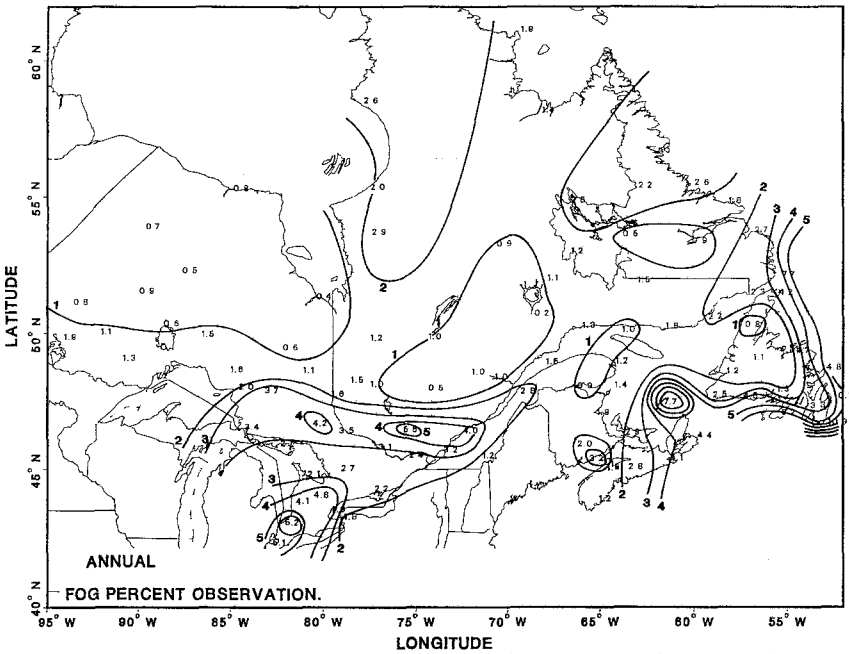

Figure 2a: The percentage of hourly observations reporting fog in eastern Canada. The data represent a 30 yr annual average (1953-1982) (Joe, 1985).

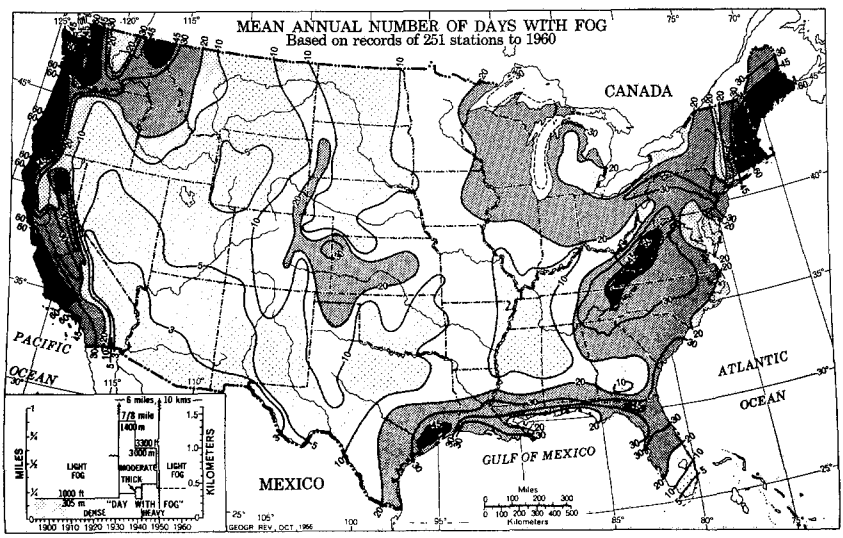

Figure 2b: The mean annual number of days with fog in the United States (Court and Gerston, 1966).

occasionally to much colder temperatures.

At most North American locations fog occurs on 0.5 to 5 day mo^{-1}. Figure 2 shows the spatial distribution of fog frequency in the United States and eastern Canada, respectively.

2. MECHANISMS OF POLLUTANT WET DEPOSITION

2.1 Precipitation Scavenging

At mid-latitudes, pollutants are scavenged from the atmosphere by precipitation via the sequence of processes depicted in Figure 3.

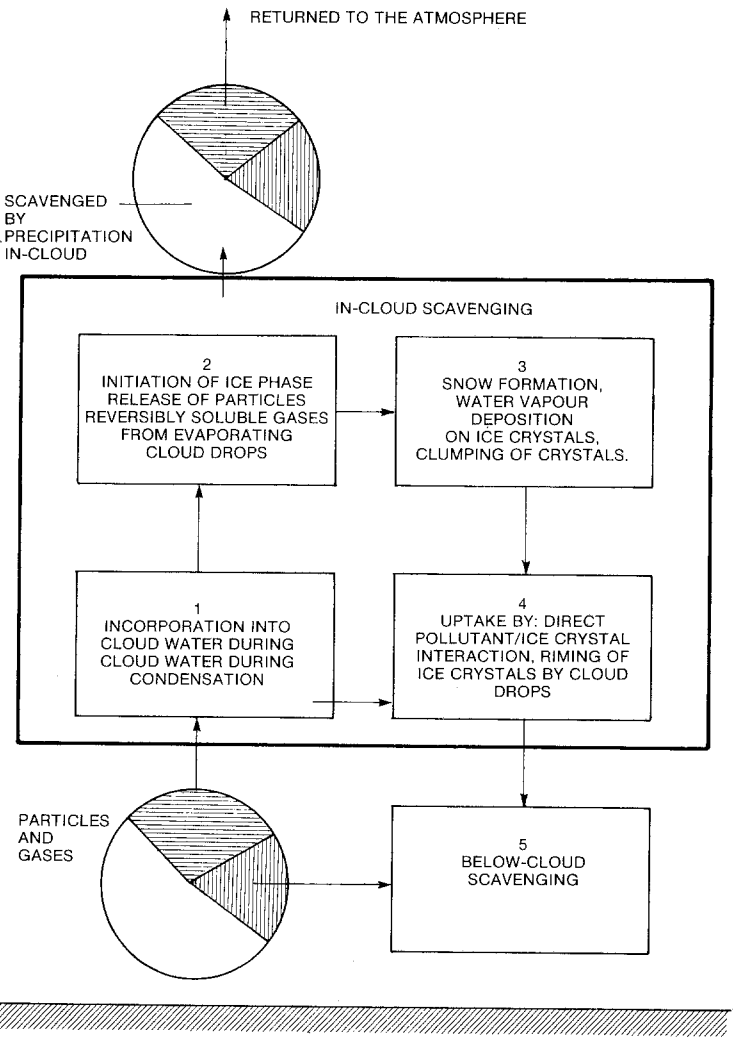

Figure 3: The precipitation scavenging process.

Condensation of water on hygroscopic aerosols entering a cloud as well as dissolution of soluble gases such as HNO_3 and SO_2 (step 1) leads to supercooled cloud drops containing inorganic ions, insoluble particulate matter and organic constituents. Typically in precipitating clouds, condensation takes place on particles of diameter greater than 0.2 μm (Pruppacher and Klett, 1978). The greater the supersaturation in a cloud the lower the minimum particle size. In precipitating clouds 50 to 100% of the particle mass is usually incorporated into cloud water at this stage. A fraction of SO_2 and NO_2 may also be converted to $SO_4^=$ and NO_3^- at this point or shortly thereafter. Indirect evidence based on an analysis of ground level observations (Barrie, 1985) indicates that 20 to 100% of SO_2 entering rain storms in eastern Canada is oxidized to $SO_4^=$ and enters cloud water while a much smaller fraction is converted in snow storms. There is also evidence that considerable amounts of cloud water NO_3^- are formed by NO_2 oxidation in both winter and summer. This is confirmed by aircraft observations (Leaitch et al., 1985).

Two things can happen to the cloud water: (i) if ice is present in-cloud, cloud water can evaporate. Water vapor, driven by the difference in saturation vapor pressure of water and ice, diffuses from the drops to ice crystals (steps 2 and 3). The gaseous and particulate matter in cloud water is then returned to the atmosphere in a form that is not necessarily the same as when it entered the water (ii) cloud water can be captured by large snow crystals (diameter > 200 μm). This process is called riming. It is the collection by snow of supercooled cloud drops 2 to 50 μm diameter by inertial impaction and interception as the heavier snow crystals fall past the small drops. The cloud drops freeze upon contact. Pollutants trapped in cloud water thereby enter precipitation and fall from the cloud.

Particles and gases that do not enter cloud water during condensation (step 1) or that are released back into the air during vapor growth of the ice phase (step 2) can be incorporated into ice crystals (steps 3 and 4). This occurs by Brownian diffusion, electrical attraction and inertial impaction. Gases may react with ice surfaces. HNO_3 is more reactive with ice than SO_2. Thus precipitation leaving cloud base can consist of rimed ice, unrimed ice or if the temperature at cloud base is above 0°C, water drops. Scott (1981) has found that snow storms with mostly unrimed snow crystals (i.e. scavenging steps 1-2-3-4) remove SO_x from the atmosphere much less efficiently than storms with rimed snow crystals (i.e. scavenging steps 1-2-3-4 and 1-4). Particles and gases that are not scavenged from the cloud by the precipitation processes discussed above leave the cloud at the evaporating edges. As a result of chemical reactions in cloud water, the chemical composition of air at the outflowing edges can be different than that at the inflow. Furthermore, the size distribution of particulate matter can be changed as a result of in-cloud chemical and physical transformations.

Rain and snow falling below the cloud base scavenge particles and reactive gases from the atmosphere by the same mechanisms as they do within cloud with the exception of the riming process and with the

addition of phoretic forces caused by evaporation in the sub-saturated air beneath the cloud. A quantitative description of the interaction between drops or ice crystals and pollutant gases or particles is a complex one. Even in the relatively simple case of a plate-like ice crystal scavenging particles, the collision efficiency depends on crystal and aerosol size distributions, on the degree of charging and on evaporation (Martin et al., 1980). Add to this an extremely variable ice crystal shape and one is left with a complicated situation to handle theoretically, experimentally or observationally.

2.2 Fog Deposition

The means by which gases and particles are incorporated into fog droplets are the same as those discussed above for cloud droplets. The ice phase is usually less important in fogs than in precipitating clouds and the riming process is essentially non-existent due to the small sizes of any ice crystals. Therefore, a simplified version of Figure 3 applies to pollutant deposition mechanisms in fogs. Pollutant-bearing fog droplets can be: deposited on the Earth's surface by sedimentation, impacted on surface roughness elements such as tree needles or leaves, and evaporated either at the edges of the cloud or internally. The first two processes constitute pollutant deposition pathways. They are dependent on the physical/chemical properties of a fog, namely: the size distribution of fog drops; the chemical composition of fog drops as a function of drop size; as well as on characteristics of the earth's surface and meteorological factors. Juisto (1981) conducted an extensive review of the current state of knowledge of drop size distributions and of the visual range in fogs. All these properties are extremely variable in time and in space as is seen below. Mean drop diameters of surface generated fogs are typically 10 (inland) to 20 μm (coastal). Mean number concentrations range from 10 to 200 drops cm^{-3} and the maximum liquid water content is approximately 0.3 g m^{-3}. With the exception of mountain top fogs, the number concentrations and liquid water contents of most fogs are lower than one typically finds in clouds.

The microstructure of fogs is strongly influenced by the characteristics of the condensation nucleii (Hudson, 1980). Thus, polluted air masses with high concentrations of condensation nucleii generally produce fogs with high cloud drop number concentrations and smaller drop sizes. Sea fogs generally have lower total droplet concentrations with larger particles. Knowledge of the chemical composition of fog water as a function of size is important because the collection efficiency of foliage and other objects is dependent on droplet size. Thus, wet deposition to an object may include only droplets in a specific size range and not the entire droplet spectrum. Unfortunately, reliable size dependent chemical composition data is not available for fogs (nor for clouds).

Aspects of the fogwater deposition process other than net pollutant deposition are also important in determining the impact on vegetation. Figure 4 is a schematic showing the deposition processes on a conifer needle. The fog droplets contain particulates such as

soil derived dust, pollen, fly ash, $SO_4^=$ and NO_3^- as well as H^+, SO_2, oxidants (O_3, H_2O_2, etc.) and other dissolved gases. When a droplet contacts the needle, damage can result directly from its acidity. But even if only the waxy surface of the needle is destroyed, this will leave it prone to subsequent damage. Evaporation of the deposited droplet can lead to even higher droplet acidity, and hence, even greater damage. Conversely, the small supersaturations that exist in fogs can cause condensation and hence dilution of the drop. The deposited droplet can also migrate over the needle surface encountering material that has been dry deposited. Like evaporation, this increases the chemical concentration of trace substances in the drop.

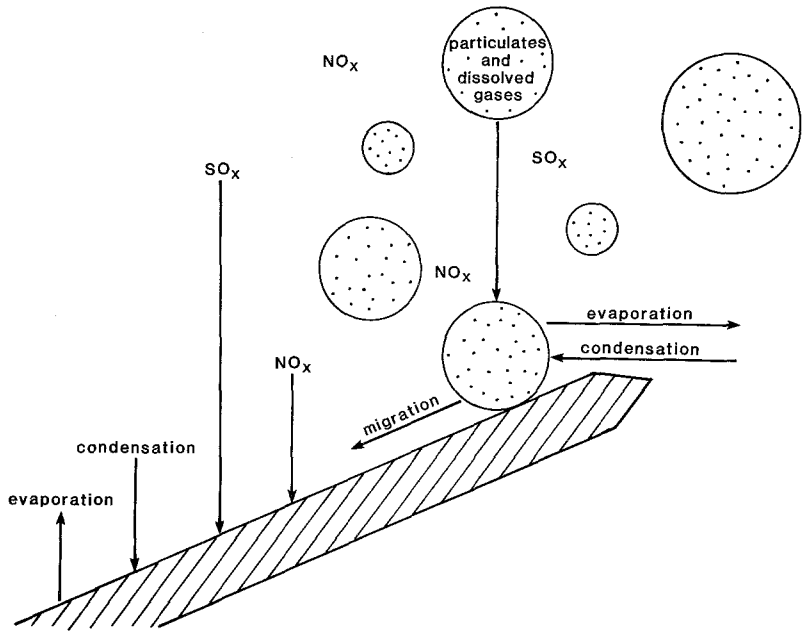

Figure 4: A schematic of the deposition of fog water drops on a conifer needle and factors that influence their composition after deposition.

3. THEORETICAL AND OBSERVATIONAL APPROACHES TO UNDERSTANDING WET DEPOSITION MECHANISMS

The ultimate goal of wet deposition research is to develop the capability of understanding the influence of a precipitation or fog system on atmospheric trace constituents in its vicinity. Thus, from meteorological observations of storm or fog characteristics and models, one would like to know what fraction of a pollutant entering a system is removed, the redistribution of remaining pollutant in the atmosphere and the physical/chemical transformation it has undergone.

An important step in attaining this goal is to understand wet deposition mechanisms. This involves three simultaneous activities: making in situ atmospheric observations of processes, developing theoretical descriptions of them and whenever possible testing them in the laboratory. The mechanisms of interest include the following: (i) nucleation scavenging of particles (ii) the oxidation steps associated with $SO_4^=$ and NO_3^- formation in cloud or fog water from SO_2 and NO_2 (iii) the in-cloud transfer of cloud water pollutants to precipitation (iv) below-cloud scavenging of pollutants by precipitation and (v) the interaction of fogs with the Earth's surface.

The nucleation scavenging efficiency (F) is defined as the fraction of total particulate mass incorporated into cloud (or fog) water during the condensation growth stage. F can be measured from aircraft by measuring the concentration of particulate matter in air entering a cloud and the air equivalent concentration in cloud water just inside the cloud (Leaitch et al., 1985; Hegg et al., 1984a). The latter measurement involves the cloud liquid water content (L in kg m^{-3} of air). An alternate approach is to measure the partitioning of mass between the gas and liquid phase just after condensation (Daum et al., 1984). Hegg et al. (1984b) report an F-value of 0.8 ± 0.2 for $SO_4^=$ bearing particles in non-precipitating cumulus, stratocumulus and stratus clouds. On the basis of measurements in stratus and stratocumulus clouds, Daum et al. (1984) conclude that the mass of aerosol in the air between the droplets is generally low but not negligible compared to the mass in cloud water. Theoretical calculations of F can be made if the composition and mass size distribution of the condensation nuclei as well as cloud supersaturation are known (e.g. Jensen and Charlson, 1984). These tend to agree with observations.

Measurements of SO_2 and NO_2 oxidation in-cloud are done from aircraft by one of two methods: (i) a mass balancing method (Hegg et al., 1984a; Lazrus et al., 1983; Leaitch et al., 1985) (ii) detection of changes in the relative composition of trace constituents in air just outside the cloud and in cloud water just inside the cloud (Daum et al., 1984). In the mass balancing method, measurement of an increase in the total amount of $SO_4^=$ or NO_3^- in air entering a cloud is attempted. It has proven to be an effective method in detecting large amounts of conversion (60% of existing $SO_4^=$ or more). Its limitation is the large uncertainty involved in calculating small differences in large numbers. The other method involves measuring the ratio of $SO_4^=$ or NO_3^- to another particulate trace constituent that does not have a gaseous precursor inside and outside the cloud. To date this method has not been widely used. Theoretical models of in-cloud oxidation of SO_x and NO_x have proven useful in guiding and explaining observations (e.g. Chameides, 1984).

3.1 Precipitation Scavenging

Aircraft studies of the transfer of cloud water pollutants to

precipitation and of below-cloud scavenging have been relatively scarce. It is particularly hard to conduct observations within precipitating convective bands because of the difficult flying conditions. Considering the complexity of the microphysical processes involved in precipitation scavenging, the variability in macroscopic features of storms and practical difficulties in flying in precipitating regions of extra-tropical cyclones, it is not surprising that other ways of using ground level observations to gain insight into scavenging processes have been sought (Slinn, 1984; Hidy and Countess, 1984; Barrie, 1985). For instance, for modelling purposes or in cases where air concentrations but no precipitation measurements are available, one often needs to estimate a substance's concentration in precipitation knowing its concentration in the atmosphere, the precipitation amount and microphysical features such as whether the riming process was active in the storm. The scavenging ratio (W) defined as the time-average concentration of substances in rain (C_R) divided by the time-average concentration in air entering a cloud (C_A) is one parameter which has proven useful in this regard. Its advantages and disadvantages are discussed by Barrie (1985). Ground level daily W values measured in Canada were used to gain insight into SO_2 oxidation in precipitating systems. Results indicate that W which is calculated from airborne particulate $SO_4^=$ concentrations is, on average, lower on days with relatively low SO_2 (SO_2 < 40% total S) than on days with relatively high SO_2 (SO_2 > 70% total S). This suggests that SO_2 oxidation is taking place. From a fit of a physical model to data, it was concluded that, on average, in eastern Canada, in-cloud oxidation accounts for approximately 60% of the $SO_4^=$ observed in rain and for almost none of the $SO_4^=$ observed in snow. A similar analysis for NO_3^- predicts that much of the NO_3^- observed in precipitation originates from in-cloud NO_2 oxidation regardless of precipitation type. This is consistent with recent aircraft observations (Leaitch et al., 1985) and with the observation that the molar ratio of $SO_4^=$ to NO_3^- is much higher in precipitation than in air (Summers and Barrie, 1986).

Theoretical simulations of pollutant removal by storm systems associated with extra-tropical cyclones can be used to understand scavenging processes. There are two types of models: diagnostic ones and simulative ones. The first type relies heavily upon observations in addition to a theoretical formulation of cloud processes to yield insight into the net scavenging of pollutants by a 'real' storm (e.g. Easter and Hales, 1983; Hegg et al., 1984b). The second type, is a dynamic storm model with chemistry incorporated (e.g. Tremblay and Leighton, 1986). Provided storm dynamics and microphysics resemble those of actual storms insight can be gained into scavenging processes.

3.2 Fog Deposition

In some areas, the amount of water deposited in fogs can be a substantial fraction of the total liquid water deposited (Nagel, 1956; Lovett, 1984; Lovett and Reiners, 1986). Values of fog-water

deposition rates ranged from ≈ 0.1 to 4 mm hr^{-1} and are supported by other observations (e.g. Merriam, 1973). These are comparable to precipitation rates. Falconer and Falconer (1980) report cloud water pH measured on Whiteface Mountain, N.Y. and review the existing

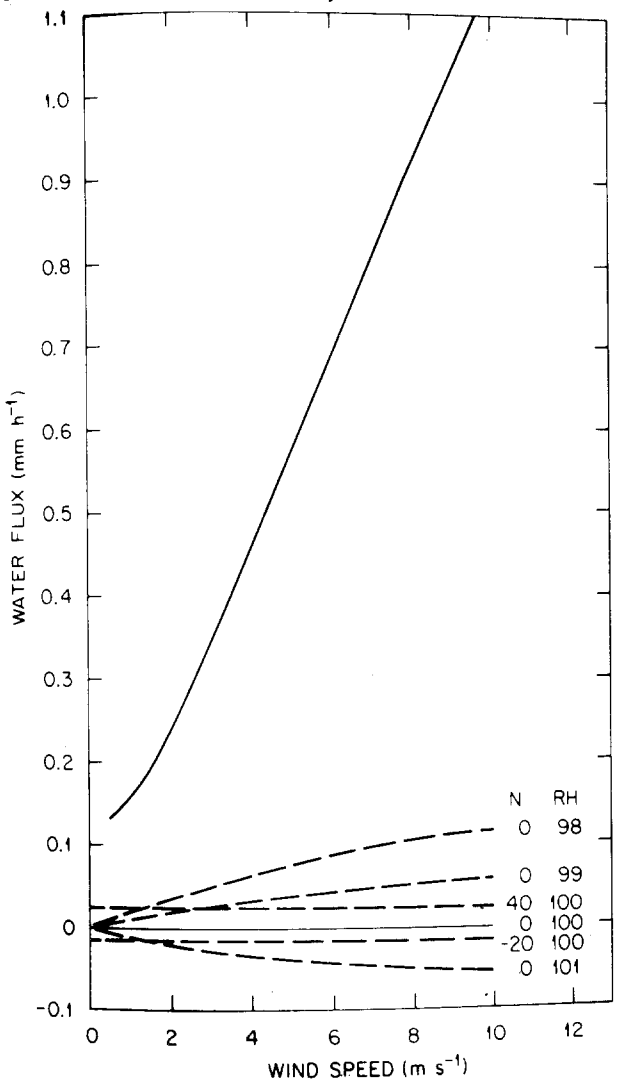

Figure 5: Simulated cloud water deposition (——) and evaporation (- - -) rates plotted against canopy-top wind speed. Evaporation rates are given for several combinations of net radiation (N) in W m^{-2} and relative humidity (R.H.) in per cent. Cloud liquid water content = 0.4 g m^{-3} modal droplet diameter = 10 μm and temperature = 10°C. Net cloud deposition is the difference between gross cloud deposition and evaporation (from Lovett, 1984).

literature. Nearly 90% of all Whiteface fog water (cloud moving over the mountain) pH observations lie in the range 2.7 to 4.7. In contrast, the most acidic rain on the mountain seldom has a pH < 4. Low fog water pH values have also been documented recently by Wisniewski (1982), Fuzzi et al. (1983) and Hoffmann (1985). In areas where fog frequency is high (coastlines or mountainous areas), the H^+ deposition from fog is comparable to that from precipitation. The same is likely true for $SO_4^=$ and NO_3^-.

Several models of vegetation-fog interactions have been developed. Shuttleworth (1977) looked at fogwater capture and the evaporation-condensation process. His theoretical description was quite restricted applying only to wind driven fog in areas of extensive uniform vegetation (i.e. mountain forests rather than lowlands with radiation fogs). Lovett (1984) used a more complex model to study a similar situation. The deposition of cloud droplets to a balsam fir forest whose canopy was divided into 1 m strata each containing seven canopy component types was modelled. Three drop-size classes were used. Theory and observations at a sub-alpine site in New England agreed reasonably well with fogwater deposition of the order of a few tenth's of millimetres per hour. The deposition velocity of fog water ranged from 1 to 80 cm s^{-1}. Figure 5 shows the marked effect on the water flux resulting from an increase in canopy top wind speed.

The mass flux of fog water to a forest canopy can be calculated with some confidence provided fog microphysical and surface boundary layer meteorological characteristics are known. In principle, if the distribution of pollutant mass as a function of drop size is known, the net deposition of pollutant by fog can be calculated. In practice, however, the measurement of composition as a function of size is extremely difficult. Field programs have concentrated on developing passive and active (fan driven) fog water samplers designed to obtain samples over as large a droplet size range as possible. This in itself is a major challenge given the wide range of wind, temperature and fog conditions encountered.

4. CONCLUSION

Wet deposition of pollutants takes place in complex precipitating cloud and fog systems. In some mountainous and coastal areas, fog water deposition is at least as important as deposition in precipitation. Thus, previous estimates of the impact of acidic deposition which did not take into account fog water (and to a lesser extent dew) are actually underestimates. Insight into precipitation scavenging processes is gained by making measurements quite different from those used to understand fog water deposition. Precipitation scavenging is studied using ground level air and precipitation chemistry measurements and cloud microphysical and chemical observations from aircraft. Deposition in fogs requires a knowledge of the fog microphysical and chemical properties, details of the structure of the Earth's surface and the micrometeorology of the

boundary layer. From available evidence, one can conclude that the acidity of cloud or fog water originates mainly from particulate sulphate and nitrate as well as gaseous SO_2, HNO_3 and NO_2. Furthermore, the acidity of deposited fog water is generally higher than that of precipitation, whereas the amount of fog water deposited is generally less than that of precipitation. The relative contribution of precipitation and fog water to wet deposition depends on the nature and frequency of fog, total precipitation amount and the nature of the Earth's surface. These factors are so variable spatially that an accurate assessment of the role of fog water deposition has not been made on a regional basis.

Models of wet deposition due to fog water, although relatively primitive, show basic agreement with observations. Improvements are needed in: the characterization of fog droplet chemistry as a function of size; the description of wind flow around mountains and in the parameterization of deposition to a variety of vegetative surfaces. Instrumentation and techniques to measure fog water deposition at the Earth's surface are still in a developmental stage. In contrast, the methodology of making a precipitation deposition measurement is well developed and relatively easily used. Precipitation scavenging has been modelled for a much longer time than fog water deposition. Despite this, a capability to understand the pathways that pollutants travel through storms has not been sufficiently developed. Consequently, much more work is needed before one has a non-empirical formulation of precipitation scavenging to use in chemical transport models.

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