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Aerosol Scavenging by Falling Snow

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ABSTRACT

Removal of atmospheric aerosols by falling snowflakes was studied during several winters in Hanover, New Hampshire, using micrometer-sized particles. Experiments were performed in open air and within static and dynamic aerosol chambers. The primary scavenging mechanism for particles in this size range was inertial impaction. Scavenging efficiencies were determined by collecting and chemically analyzing snowflakes that were allowed to fall naturally through the static chamber containing a cloud of known aerosol concentration. The average scavenging efficiency (defined as the ratio of the mass of aerosol collected by the snowflake to the total mass of aerosol in the swept volume) of several different types of snowflakes and ice crystals was 0.11 ±0.08. Higher scavenging efficiencies were observed for three-dimensional spatial dendrites than for planar crystals, such as planar dendrites and hexagonal plates. Overall, snow was found to be four to five times more efficient than rain in scavenging 0.3- to 6-µm-sized particles. Laser attenuation measurements within a dynamic aerosol chamber indicated that particle scavenging can cause relative transmission increases of as much as 15% for each minute of exposure to snowfall. Model calculations predicted aerosol cloud half-lives of 2 to 20 minutes for snow precipitation rates of 2.5 to 0.5 cm/hr.

INTRODUCTION

Aerosols can be removed from the atmosphere either passively by gravitational sedimentation or actively by rainout and washout. Rainout includes all processes occurring within atmospheric water clouds, while washout refers to processes that occur below clouds. Washout mechanisms include Brownian diffusion, inertial interception and impaction, eddy turbulence, and electrostatic and phoretic attraction (Murakami et al. 1983, Cragin and Hewitt 1987a, Mitra et al. 1990).

While the largest weight fraction of atmospheric particles is removed by dry deposition (gravitational sedimentation), this process is important only for very large (>10-µm aerodynamic diameter) particles that have appreciable Stokes settling velocities. Coarse particles (2.5–10 µm) are of greater concern because they are ubiquitous in the urban atmosphere and remain suspended for longer periods of time. Such particles originate from industrial emissions, sea spray, plants (pollen) and wind-blown terrestrial dust (Lyons and Scott 1990). Industrial and commercial activities, such as refuse incinerators and coal- and oil-fired boilers, can contribute 20 to 40% of the urban coarse particulate atmospheric loading (Mamane and Dzubay 1988). These emissions are of particular concern because they are not only respirable but many are carcinogenic as well. Coarse particles, which represent the largest fraction of atmospheric particles either by number or cross-sectional area, are removed predominantly by washout, often called precipitation scavenging.

During the last several years we have conducted field studies of coarse aerosol scavenging by precipitation, primarily snow. Our main objective was to experimentally assess the effects of snow scavenging upon the visibility through and the longevity of aerosols used as military obscurants. Since the size distribution of the aerosol particles used is similar to that of coarse atmospheric aerosols, our findings are directly applicable to snow scavenging of aerosols in urban areas. Here we report scavenging efficiencies of snowflakes and different snow crystal types and the effect of scavenging upon aerosol cloud longevity and visibility.

EXPERIMENTAL

In evaluating the effectiveness of aerosol scavenging by falling snow, we used two separate approaches. First, we experimentally measured snowflake scavenging efficiency using a "static" aerosol chamber in which snow was allowed to fall downward through an aerosol cloud of known concentration and the individual snowflakes subsequently collected and chemically analyzed to determine the mass of aerosol removed. Second, to assess the rate of change in visibility (transmission) due to scavenging, we monitored the intensity of a laser beam through the aerosol cloud during snowfall using a "dynamic" cloud chamber. All experiments were conducted on aerosol clouds produced from dispersed brass powder with an average particle diameter of 1.7 µm (Wentsel et al. 1986). Chemical analysis showed its composition to be 59 ±1% Cu and 40 ±1% Zn. The average Stokes equivalent diameter of the aerosol was determined by cascade impaction (Anderson Model 20-800, 1 CFM Ambient) to be d_{50} = 3.3 µm. Using electrozone analysis (Particle Data, Inc., Model Elzone 180), often referred to as the Coulter principle, and an aqueous suspension of the bulk powder treated with the dispersant Triton X100, we established a number concentration of approximately 6 × 10^{10} particles/gram for the bulk material.

Chemical analysis

To determine the amount of aerosol collected by snowflakes, and hence their scavenging efficiency, individual or clusters of flakes were dissolved in dilute (2%) HNO_3. Copper concentrations in the acidified samples were determined after vigorously shaking the collection vessels to dislodge particles adhering to container walls and allowing them to digest for 24 hr. The analysis of Cu was performed by either Flame or Graphite Furnace Atomic Absorption (Cragin and Hewitt 1987a).

Field equipment

Aerosol clouds were produced by dispersing the brass powder with a commercially available backpack crop duster (Green Machine, Model 4600 LP with duster adapter Model 46700). Approximately 10 g of brass powder was used per experiment. Aerosol cloud concentrations within the chambers were originally determined by mass loading on high volume filters, then after calibration, read continuously by transmissometry. The correlation between filter mass loading and transmission was linear in the region of interest (0.5 to 2 mg/L). More detailed descriptions of the equipment and measurement protocols have been reported elsewhere (Cragin and Hewitt 1987b, Hewitt 1988, Hewitt et al. 1988).

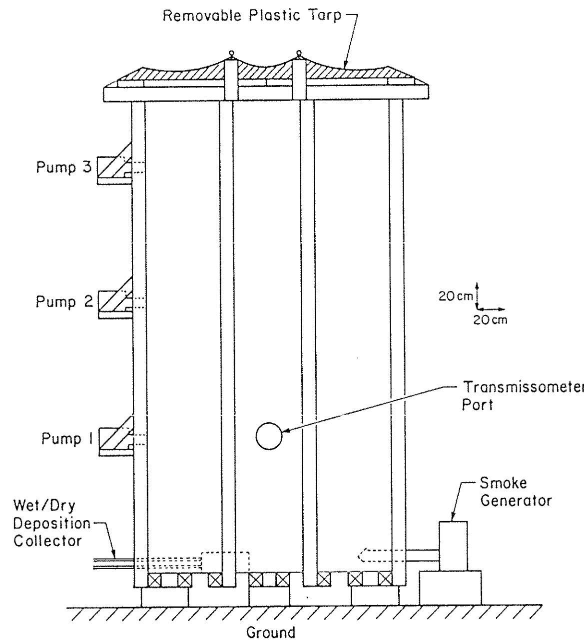


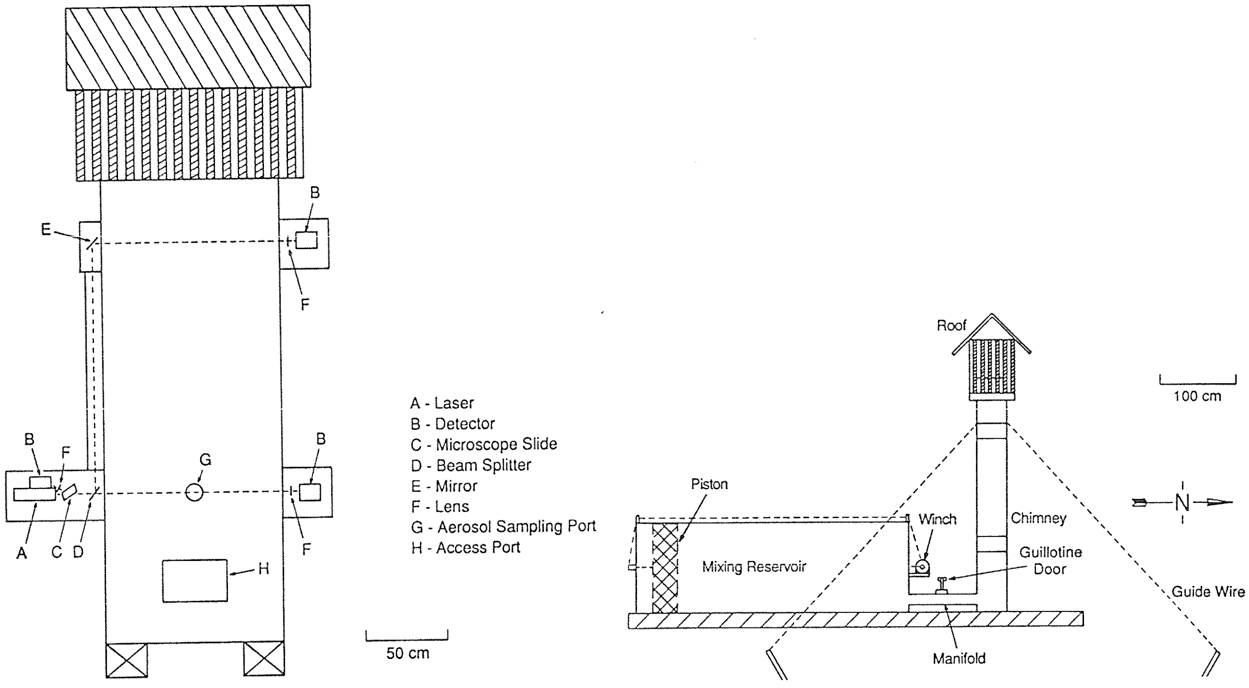
Figure 1. Static aerosol scavenging chamber.

Aerosol scavenged by falling snowflakes

Scavenging efficiencies were determined using a 12-m^3 static cloud chamber, 1.8 m × 1.8 m × 3.6 m high (Fig. 1). A 60-cm^2 opening in the middle of the roof with a hinged door served as the entrance port for naturally falling hydrometeors. Attached to the chamber's floor was a tunnel leading from the back wall to the center, allowing for the exposure of a sliding tray. The tunnel could be opened and closed, and the tray moved permitting precise control of exposure time and the sample retrieval without entering the chamber. Aerosol clouds generated for these tests ranged in concentration from 0.5 to 1.6 mg/L. Frozen hydrometeors were collected for 10 to 15 s on a clean velvet cloth that covered the bottom half of a 23-cm square petri dish which rested on the floor of the sliding tray. After exposure the drawer was closed, the dish removed and covered, then transferred to an on site cold room at −10°C. Without delay, one to as many as four hydrometeors were transferred to a 7.5 mL CPE bottle (Nalgene) with a fine needle, and the number, size, and type of the crystals was recorded. Background (blank) concentrations inherent to gravitational aerosol sedimentation in the absence of falling hydrometeors were determined to be less than 5% of the concentrations attributed to scavenging (Hewitt et al. 1988).

Aerosol cloud depletion and visibility

A dynamic cloud chamber (Fig. 2) was constructed to observe aerosol cloud depletion. This apparatus consists of two main functional chambers, the first being a



a. Front view. *b. Side view.*

Figure 2. Dynamic aerosol scavenging chamber.

mixing reservoir where the brass aerosol cloud was originally dispersed and homogenized, and the second, a vertical tower or chimney through which the aerosol cloud was slowly pushed upward. This dynamic aerosol tower was modeled after the wind tunnel of Blanchard (1951). A more complete description of the design, construction and operation of the aerosol chamber can be found elsewhere (Hewitt 1988).

For a typical test, an aerosol cloud was first produced within the mixing reservoir by dispensing about 10 g of brass powder; the aerosol cloud was then forced by a piston, slowly (2.5 cm/s) through the chimney. Light energy from a 633-nm He-Ne laser (Oriel, model 79255) was monitored using silicon detectors (Ealing Electro-Optics, model 28-7821) at two horizontal levels, 1.5 m apart. Our earlier studies predicted that snow falling at rates of 80 to 120 cm/s (Locattelli and Hobbs 1974) with concomitant particle scavenging could be detected by monitoring transmission through the cloud over a 1-minute period. Thus, the difference in transmissions between the two laser paths is proportional to the rate of aerosol cloud depletion.

The following snow characterization parameters were also monitored: snow air mass concentration (Lacombe 1982), gravimetric snow mass precipitation rate (Cragin and Hewitt 1987a), optical snowflake number concentration (Koh 1988), and snowflake area by Formvar replication.

RESULTS AND DISCUSSION

Scavenging can occur by Brownian diffusion, electrostatic attraction, eddy turbulence, diffusiophoresis, thermophoresis, and inertial impaction. The efficiency of any given mechanism depends primarily upon the size of the particle being scavenged. Table 1 compares the effectiveness of each of these mechanisms for scavenging of coarse aerosol particles of the size range used in this study. In the absence of electrostatic charges, inertial impaction is the primary scavenging mechanism for this aerosol. Inertial impaction is the mechanism whereby a falling hydrometeors collide with an airborne particles. For this process to occur the particle in the path of the falling hydrometeor must possess sufficient inertia to cross the air streamlines.

Initial aerosol clouds were generated in open air both during and in the absence of precipitation. Surface snow samples collected over a 300-m^2 downwind grid showed order-of-magnitude higher aerosol concentrations of deposited aerosol during snowfall (as high as 3.6%) than during clear-air conditions (<0.3%). Some additional experiments were then conducted to determine the effect of precipitation rate and type upon aerosol scavenging. A plot (Fig. 3) of the percent of released aerosol recovered vs. mass precipitation rate shows two distinct linear relationships (significantly different at the 99% confidence level), one for rain/hail

Table 1. Particle scavenging mechanisms.

Mechanism	*Particle diameter over which effective (μm)*	*Estimated importance for scavenging of 0.5—5 μm aerosol*
Brownian diffusion	< 0.1	None
Inertial impaction	≥ 1	Very
Electrostatic	0.01–2	Possibly very for opposite charges
Eddy turbulence	All	Minor
Phoresis		
Thermophoresis	0.1–1	Some for colder flake
Diffusiophoresis*	0.1–1	None for subliming flake

*Requires that hydrometeor be growing or shrinking.

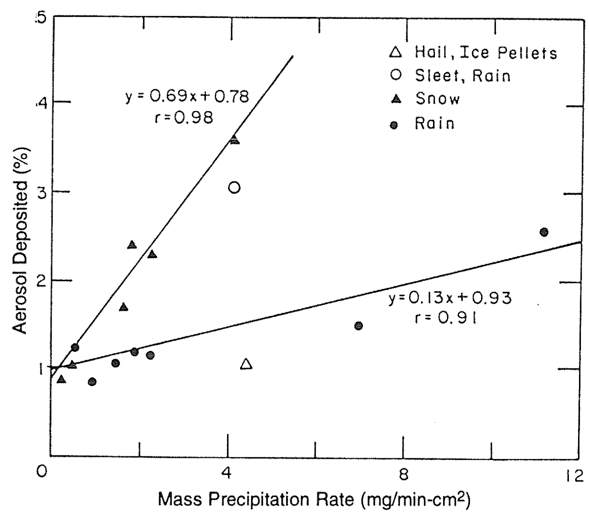


Figure 3. Percent of released aerosol deposited within sampling area during precipitation events. For a given mass precipitation rate, snowflakes scavenge coarse aerosols as much as five times more efficiently than raindrops.

and another for snow. The slopes of the lines in this plot show that on a mass basis snow is five times more efficient than rain in scavenging coarse aerosol particles. This is in agreement with previous work of others (Georgii 1965, Graedel and Francy 1975, Murakami et al. 1983, Raynor and Hayes 1983).

Scavenging efficiency

The scavenging efficiency of a hydrometeor passing through an aerosol cloud can be expressed as the fraction or percent of particles collected in its fall path,

$$E = N_s / N_p$$

where N_s and N_p are the number of aerosol particles scavenged and the number present, respectively. The number of particles in the hydrometeor's path being

$$N_p = C\,A\,P$$

where C = aerosol concentration in particles/m^3
A = hydrometeor cross-sectional area
P = vertical length of the fall through the smoke cloud.

If we assume that the particle size distribution collected by the falling hydrometeor is representative of the aerosol cloud, scavenging efficiency can be calculated from the ratio of aerosol mass on the hydrometeor to the aerosol mass in its fall path:

$$E = \text{mass on snowflake} / C_m\,A\,P$$

where C_m is the aerosol mass/volume concentration.

Table 2 contains the determined scavenging efficiency for various types of falling snow crystals. Crystals that fell as aggregates of clusters were treated as individual flakes since they had actually passed through the aerosol cloud as a single hydrometeor.

The overall mean scavenging efficiency for several types of snow crystals and flakes for this aerosol was 11 ±8% (n = 27). Scavenging efficiencies of planar dendrites (7.3 ±4.6%) and irregular crystal clusters (18 ±8%) were statistically significantly different at the 95% confidence interval. This agrees with our earlier findings (Cragin and Hewitt 1987b) and those of others (Redkin 1973, Lew et al. 1986) that showed stellar snowflakes and hexagonal plates scavenge aerosols less efficiently than irregular crystal clusters and spatial dendrites. The greater efficiency of the latter two snowflake habits is due to their three-dimensional structure in contrast to the flat, planar structure of hexagonal plates and planar dendrites. Structured, three-dimensional flakes can act as depth filters as they fall, thus more efficiently removing aerosol particles by the dominant scavenging mechanism, inertial impaction.

Mitra et al. (1990) plotted the scavenging efficiency of snow crystals and flakes vs. $^1/_2$ Stokes number for results of their experiments as well as those of others. Including the average of our results on such a plot (Fig. 4) shows that the log/log relationship is linear for at least another decade for coarse aerosols.

Table 2. Scavenging efficiency of different types of snow crystals and flakes.

Type of crystal	*No. of flakes collected*	*Flake area (mm²)*	*Scavenging efficiency (%)*
Irregular crystal	3	9.6	10
Irregular crystal	2	7.1	14
Irregular crystal	1	20	28
Irregular crystal and wet needle clusters	1	20	12
Irregular crystal and wet needle clusters	1	20	14
Irregular crystal and wet needle clusters	1	20	12
Irregular crystal and wet needle clusters	1	20	19
Irregular crystal	1	1.8	17
Irregular crystal	1	1.8	26
Irregular crystal	1	1.8	32
			Avg. 18±8
Planar dendrite rimed	1	20	6.6
Planar dendrite plate cluster	1	79	5.4
Planar dendrite plate cluster	1	79	6.5
Planar dendrite plate cluster	1	79	4.1
Planar dendrite plate cluster	1	79	4.9
Planar dendrite plate cluster	1	79	6.5
Planar dendrite rimed	1	3.1	6.0
Planar dendrite rimed	1	7.1	4.4
Planar dendrite rimed	1	38	5.0
Planar dendrite heavily rimed	1	3.1	14
Planar dendrite heavily rimed cluster	1	13	12
Planar dendrite plate	1	7.1	3.7
Planar dendrite plate	1	1.8	17
Planar dendrite plate	1	1.8	13
Planar dendrite cluster	4	20	12
Planar dendrite	2	14	3.1
Planar dendrite cluster	1	180	6.0
			Avg. 7.3±4.6
			Overall Average **11±8**

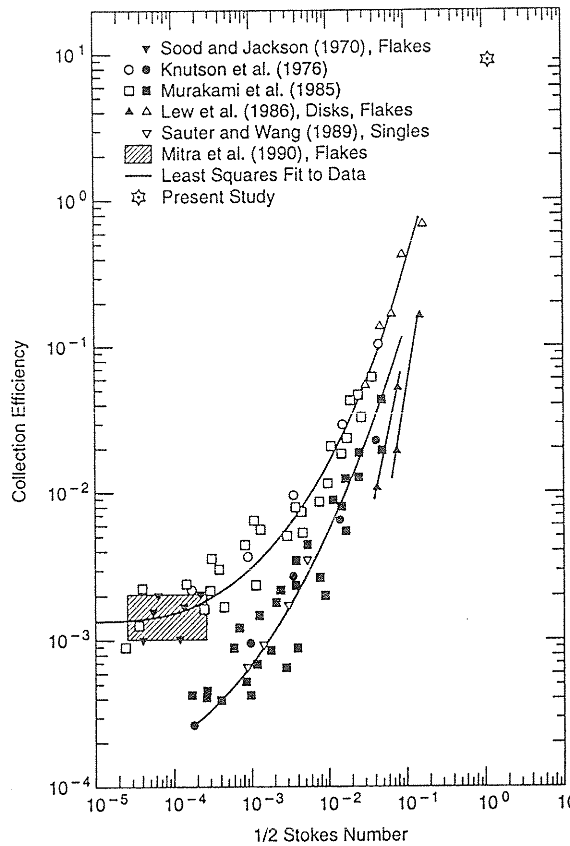


Figure 4. Efficiency of aerosol scavenging by snowflakes as a function of the Stokes number. (Original plot from Mitra et al. 1990.)

Aerosol cloud depletion rate and visibility changes

Transmission measurements through aerosol clouds were measured during clear air and snowfall under instrumentally identical conditions. Transmission through the aerosol cloud under clear-air conditions served as a reference (control) for those with snowfall. A typical real-time transmission tracing for a test is shown in Figure 5. As the aerosol cloud rises within the tower it

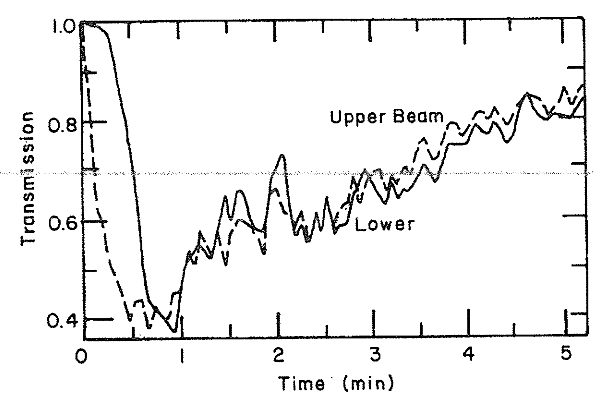
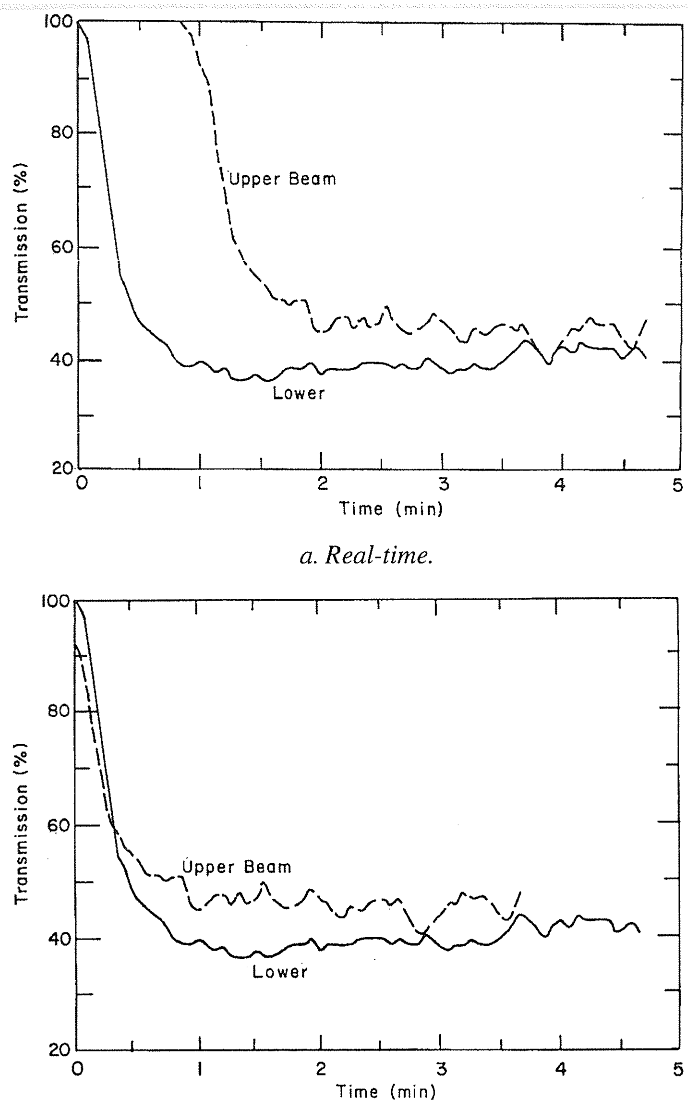


Figure 5. Real-time transmission through aerosol cloud during clear-air conditions.



a. Real-time.

b. 60-second phase-shifted.

Figure 6. Transmission through aerosol cloud during snowfall. The vertical displacement of transmission between the two beams is proportional to the rate of aerosol particle scavenging by falling snow.

impedes the lower light beam, causing a decrease in transmission. Approximately one minute later, the rising cloud impedes the upper beam path, producing a similar reduction in transmission. The transmission through both beams is then partially reduced for the remainder of the test.

The light intensity detected at the two elevations in the tower cannot be compared simultaneously because at any point in time the beams are passing through different portions of the aerosol cloud. To correct for this, the response of the upper detector was phase shifted by the time required for the cloud to travel the distance between the two beams (60 seconds). Phase-shifted transmission results for a clear-air test follow each other quite closely.

Figure 6 shows real-time and phase-adjusted (60 seconds) transmission tracings obtained during a typical snowfall experiment. The most salient feature of these plots is the significantly higher transmission level for the upper beam than for the lower beam. This increased transmission is a direct result of particle scavenging by falling snow. The particle concentration is lower and the transmission is higher for the upper beam because the rising aerosol cloud has been exposed to falling snow for an additional minute. The increased transmission reflects the rate of aerosol particle removal by snowfall. Snow falling through the light beams has little effect upon transmission over the 1-m horizontal path length. Based upon measurements of airborne snow concentration and its mass extinction coefficient, snow within the chamber caused less than 1% loss in transmission, and attenuates both beams equally.

Twenty-seven tests were conducted during snow storms to evaluate the effect of scavenging upon transmission or visibility through an aerosol cloud. The transmission increased 5 to 15% (average 9.3 ±7.9%) per minute of aerosol exposure to falling snow, suggesting that coarse urban aerosols would be depleted to negligible levels within tens of minutes during snowfall.

In an attempt to correlate the observed transmission increases with meteorological parameters, the snow characterization measurements made during several of the tests were evaluated. Mean snowflake fall speed (V) was calculated from the ratio of mass precipitation rate (M) and the air mass concentration (C):

$$V\,(\mathrm{m/s}) = M\,(\mathrm{g/m^2\,s}) \,/\, C\,(\mathrm{g/m^3})$$

From the mean fall speed, average snowflake area, and the snowflake number concentration, the volume of air swept out per second within the chamber tower was calculated. Figure 7 is a plot of the percent increase in transmission vs. the cloud volume sweep rate (VSR). The only parameter the VSR does not include is the scavenging efficiency of different crystal types. The previous experiment showed that scavenging efficiency is dependent on the snow crystal type. Including this parameter would result in a series of curves dependent

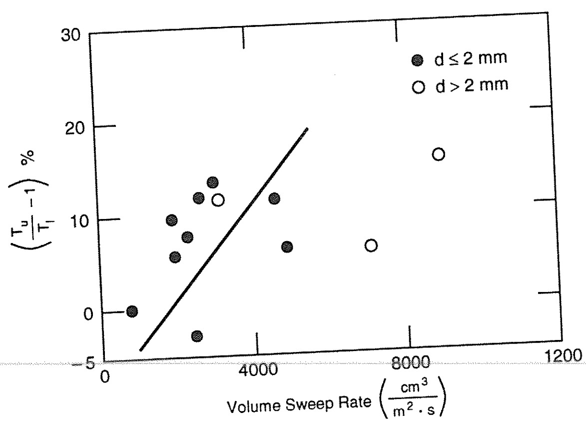


Figure 7. Average percentage increase in transmission vs. volume sweep rate for 12 scavenging experiments for which snow characterization data were taken. Swept volume, calculated from snowflake area and fall speed measurements, is the volume of air that snowflakes sweep out per unit horizontal area (1 m^2) per unit time (1 s).

on crystal type. For these tests the snow mass precipitation rates ranged from 0.3 to 4.2 mg/cm^2-min (0.18–2.27 mm/hr water equivalent), and usually the snow crystal type was mixed with a wide size distribution. Thus, an even more intensive snow characterization may be required before a strong quantitative relationship can be obtained.

The VSR and corresponding transmission increase functions can also provide an independent measure of scavenging efficiency. For the above experiments, the mean VSR and corresponding transmission increase were 1100 cm^3/s and 8%, respectively. Taking into account the total aerosol cloud volume (4.5×10^5 cm^3) and scavenging time (60 s), the scavenging efficiency for the cloud depletion tests averaged 8.8 ±5.5%. This average scavenging efficiency is not statistically different at the 95% confidence level from average value determined from chemical mesurements of 11 ±8% (Table 2).

Cloud half-life

Assuming that snowflake paths do not overlap, cloud half-life can be calculated using the relationship

$$C = C_0\,(1 - F)^t$$

where C is the smoke mass concentration at time t, C_0 is the initial mass concentration, and F is the fractional change per unit time (9.3%/min).

$$\ln C/C_0 = (t)\ln(1 - F)$$

$$t = 7.1\ \mathrm{min}$$

The terminal settling velocity of 10- to 1-µm unit density aerosol particles ranges between 0.31 and 0.0035 cm/s. If released from a 100-m-tall stack under clear conditions, they would require 9 hours to 33 days to reach the ground due to gravity alone. During snowfall, however, the airborne life span of such particles would be reduced significantly. The distances these particles travel from their source would also be reduced: at an average wind speed of 2 m/s, even a moderate rate of snowfall would remove and deposit 50% of the aerosol within one kilometer of its source.

CONCLUSIONS

- One to two orders of magnitude more aerosol is deposited on the ground surface during snowfall events than under clear-air conditions.
- For a given mass precipitation rate, snowflakes and ice crystals scavenge aerosols as much as five times more efficiently than raindrops.

- Scavenging efficiencies are higher for three-dimensional snowflakes and clusters than for planar crystals.
- Decreases in aerosol concentration due to scavenging by falling snow increase visibility within minutes.
- The rate at which visibility increases is a function of snow mass precipitation rate, the volume swept and scavenging efficiency of a snowflake crystal type.
- Experimental results verify models predicting aerosol cloud half-lives on the order of several to tens of minutes during snowfall.

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