USE OF SURROGATE SNOW SAMPLES
TO MEASURE DRY DEPOSITION ON SNOWPACKS IN FORESTS

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Abstract

Snowpack dry deposition of calcium, magnesium, nitrate, and sulfate was monitored by conducting a mass balance on surrogate snow samples. Surrogate snow samples, which consisted of shaved deionized ice, were exposed in petri dishes at the snowpack surface. The effects of windblown snow and organic debris contamination on dry deposition estimates were considered. Preliminary results have shown that the number of samples needed to estimate snowpack dry deposition by using surrogate snow samples is much lower (n ≤ 45) than for direct repetitive sampling of the natural snowpack. Dry deposition rates for 24 hour periods can be resolved. Preliminary data indicated that snowpack dry deposition was significantly reduced by the forest canopy.

Introduction

The major complication for estimating dry deposition is that this process is surface dependent for small particles and gases, where deposition is not strongly controlled by gravity (Hicks, 1986; Hicks, 1984). In the United States and Canada, snow is an important surface for several months of the year. Unfortunately, few studies have looked at dry deposition on snowpacks, especially dry deposition on snowpacks in forests.

Various techniques have been used to calculate dry deposition rates. Micrometeorological techniques such as, eddy correlation and the gradient approach, are fairly complicated; hence, they are not suitable for routine monitoring (Hicks,
Other limitations are that both techniques require stringent site criterion and sensors of high resolution, which are only available for a few chemical species (Hicks, 1986; Hicks and McMillen, 1986). In addition to these limitations, the micrometeorological methods do not directly measure the quantity of accumulated materials to a natural surface (Lindberg and Lovett, 1985). Numerous surrogate surfaces have been used to estimate dry deposition. The major problem associated with surrogate surfaces is that it is difficult to extrapolate the data collected from various synthetic surfaces to natural surfaces (VandenBerg and Knoerr, 1985; Hicks and McMillen, 1986; Lindberg and Lovett, 1985). Sequential snow sampling from an existing snowpack is desirable because a natural surface is used. However, high variability in natural snowpack chemistry makes it difficult to reliably detect small concentration increases for a short exposure period (DeWalle, Sharpe, and Halverson, 1986). The use of a wet/dry precipitation sampler with natural snow in the dry bucket has also been employed (Cadle, Dasch, and Mulawa, 1985). Natural snow samples were placed in the dry position of the sampler. However, the high chemical variability of natural snow and the inability of the wet/dry sensor to accurately detect light precipitation also present some difficulties with this technique (Cadle, Dasch, and Mulawa, 1985).

In order to avoid the problems with chemical variability of the natural snowpack, the surrogate snow method was developed to measure snowpack dry deposition. In this surrogate snow technique, petri dishes, which are filled with shaved deionized ice, are exposed during non-precipitation periods. These dishes are set into the natural snowpack so that the rim of each dish is flush with the surface of the snowpack. The blank (initial) concentrations are very low and uniform; therefore, this technique holds promise for detecting small concentration increases over a short exposure period.

The objectives of this paper are to describe the methodology for measuring snowpack dry deposition using the surrogate snow technique and to present preliminary data demonstrating that this technique appears to be sensitive in detecting significant differences in dry deposition rates between open and forest sites.

**Study Site**

Black Moshannon State Park (latitude = 40° 53' 30" N, longitude = 78° 4' 0" W) in Centre County, PA was used as a field test site. The snowpack on Black Moshannon Lake was selected as the open site, which was adjacent to the deciduous and conifer sites. The elevation of the open, deciduous, and coniferous sites was 569, 570, and 571 m, respectively. The deciduous forest site consisted of black cherry (*Prunus serotina*), red maple (*Acer rubrum*), northern red oak (*Quercus rubra*), white oak (*Quercus alba*), and hornbeam (*Carpinus caroliniana*) trees; and, the conifer site predominately consisted of red pine (*Pinus resinosa*) trees. Dry deposition rates for calcium, magnesium, nitrate, and sulfate plus total vapor losses from the surrogate snow samples were measured for one 24 hour exposure period. For all three sites, surrogate snow samples were set out between 1100 EST to 1230 EST on 3/21/88 and were collected between 1100 EST to 1230 EST 3/22/88.

**Weather Conditions**

The weather conditions for State College, PA, which is approximately 5 km from Black Moshannon State Park, are shown in Table 1 for 0700 EST 3/21/88 through 0700 EST 3/23/88. On 3/21/88, 5.6 cm of snow fell; therefore, a continuous snowpack existed at all three sites on 3/22/88. A high pressure system dominated the dates of 3/22/88 and 3/23/88 with the barometric pressure peaking at 1100 EST on 3/22/88. On average, the air temperature at Black Moshannon State Park is approximately 2 to 5 °C lower than State College, PA. The maximum air temperatures for 3/22/88 and 3/23/88
occurred at 1400 EST and 1730 EST, respectively. The fraction of cloud cover for 3/22/88 and 3/23/88 was 0/10 and 1/10, respectively. Therefore, this one 24 hour exposure period was dominated by a high pressure system with cold air temperatures and clear skies.

<table>
<thead>
<tr>
<th>Date</th>
<th>Precipitation (mm)</th>
<th>Wind Direction</th>
<th>Wind Speed (m s⁻¹)</th>
<th>Max/Min Air Temperature (°C)</th>
</tr>
</thead>
<tbody>
<tr>
<td>3/22/88</td>
<td>0.0</td>
<td>WNW</td>
<td>3.6 - 4.5</td>
<td>0.6/-11.7</td>
</tr>
<tr>
<td>3/23/88</td>
<td>0.0</td>
<td>SSW</td>
<td>1.3 - 3.1</td>
<td>5.0/-10.6</td>
</tr>
</tbody>
</table>

3/21/88: 0700 EST 3/20/88 through 0700 EST 3/21/88
3/22/88: 0700 EST 3/21/88 through 0700 EST 3/22/88

**Methods**

**Surrogate Snow Sample Preparation**

Surrogate snow was prepared from frozen deionized water. A plastic bucket was filled with approximately 10 L of deionized water, sealed with a plastic lid, and placed in a walk-in type freezer unit. Once frozen, this ice was shaved using an electric ice shaver (Gold Medal Products, Pittsburgh, PA: model 29014). Shaved ice was sieved to a particle size less than 4.0 mm, thereby producing "surrogate snow" (average density = 0.50 g/cm³). The surrogate snow was then placed into petri dishes (diameter = 14.0 cm, depth = 1.3 cm, volume = 0.2 L, area = 1.54 x 10⁻² m²), which were previously rinsed with deionized water. After being filled with snow, the dishes were covered with plastic lids. Each dish was then weighed to the nearest 0.05 g with a top loading analytical balance and placed in petri dish racks. These racks were placed into ice chests and these ice chests were transported to the study site.

**Grid Layout and Field Procedures**

At each site, 25 dishes were exposed in a rectangular grid at 3 m spacing for a total grid size of 12 x 12 m. One grid was located at each of the three sites; and, the grids, which were located along a transect, were approximately 150 m from one another. Of the 25 dishes, twenty dishes contained sieved surrogate snow, while, the other five dishes were empty and were used as windblown snow/organic debris traps. All dishes were pushed into the existing snowpack so that the rim of the dish was level with the surface of the snowpack. After the dishes were placed into the snowpack, all the lids were removed from the dishes and returned to the laboratory where they were rinsed with deionized water. After a 24 hour period, the lids were replaced on the dishes; and, these dishes were once again stacked in petri dish racks, the racks were placed in ice chests, and these ice chests were transported back to the laboratory. Preliminary studies indicated that an exposure time of 24 hours was sufficient to calculate dry deposition rates.

Ten blanks were used to establish the mean initial concentration due to any impurities in the deionized water and any background contamination due to handling. A blank is a surrogate snow sample in a petri dish, which was transported to and from the study site, but was never exposed to the atmosphere.
Laboratory Procedures

At the laboratory, exposed snow samples and windblown snow/organic debris (WBS/OD) traps were reweighed and then all samples (exposed snow samples, blanks, and WBS/OD traps) were allowed to completely melt. It should be noted that for the designated exposure period, no windblown snow was collected in any of the empty dishes; hence, these dishes were essentially debris traps. After melting, all samples were transferred to 125 mL plastic bottles. Due to the small amount of organic material collected in the debris traps, all of the organic material from the forest debris traps was composited. A leaching test was conducted for the accumulated debris. This debris and approximately 80 mL of deionized water were placed into a 125 mL plastic bottle; and, this sample was allowed to leach for a time equivalent to the holding time for the exposed snow samples. This leaching test sample was then analyzed for the same constituents as the other samples (see below). Therefore, the final concentrations for an exposed surrogate snow sample were corrected for any contamination due to organic debris leaching. This correction factor for the organic debris contamination is further discussed in the Calculation and Data Analyses Section.

All samples in 125 mL bottles were analyzed for total suspended solids (TSS), calcium, magnesium, nitrate, and sulfate. TSS were determined to 0.01 mg L\(^{-1}\) by passing the sample through a 0.45 μm membrane filter, which was dried at 103-105 °C (EPA Method 160.2, EPA 1983). Calcium and magnesium concentrations were determined to 0.005 mg L\(^{-1}\) by atomic absorption spectrophotometry (EPA AA Direct Aspiration Method 215.1 and EPA AA Direct Aspiration Method 242.1, respectively, EPA 1983). Nitrate concentrations were determined to 0.001 mg L\(^{-1}\) by colorimetry using a Technicon™ Auto Analyzer (EPA Cadmium Reduction Method 353.2, EPA 1983). Sulfate concentrations were determined to 0.01 mg L\(^{-1}\) by ion chromatography (EPA Method 300.0, EPA 1984). Analyses were all completed within the recommended holding times and were conducted at the Environmental Resources Research Institute (ERRI), at The Pennsylvania State University.

Calculations and Data Analyses

For each exposed snow sample, the correction for organic debris contamination, which was based on the leaching test (accumulated debris plus deionized water), was calculated in the following manner (i.e., for calcium):

\[
\text{Ca in mg L}^{-1}/\text{TSS in mg L}^{-1} \times \text{TSS in mg L}^{-1} \\
\times [0.08 \text{ L/final sample volume in L}] = \text{Ca in mg L}^{-1}
\]

where, Ca/TSS is the ratio of the calcium concentration to the total suspended solids concentration in the leached sample, TSS is the concentration of total suspended solids in the exposed snow sample, and 0.08 L/final sample volume is the ratio of the amount of deionized water used in the leaching test to the final volume of the exposed snow sample. The product of the above equation represents the the calcium contamination due to organic debris in mg L\(^{-1}\). This value was then subtracted from the final calcium concentration of the exposed snow sample, therefore resulting in the corrected final calcium concentration. All the final concentrations for exposed snow samples were corrected for any organic debris leaching. It should be noted that the final concentrations and the corrected final concentrations for calcium, magnesium, nitrate-N, and sulfate were essentially identical; therefore, it appears that small amounts of organic debris do not greatly influence the final concentrations for the exposed snow samples.
Snowpack dry deposition rates (DDR) for calcium, magnesium, nitrate-N, and sulfate-S were determined by using the following mass balance equation:

\[
DDR = \frac{[C_f \cdot V_f - C_1 \cdot V_1]}{[\Delta t \cdot A]} = \text{mg m}^{-2} \text{ d}^{-1}
\]

where, \( C_1 \) is the initial or mean blank concentration (mg L\(^{-1}\)), \( C_f \) is the corrected final concentration (mg L\(^{-1}\)) for the sample after the exposure period, \( V_1 \) is the initial volume of the sample (L), \( V_f \) is the final volume of the sample (L) after the exposure period, \( \Delta t \) is the exposure period (days), and \( A \) is the area of petri dish (m\(^2\)).

The total vapor losses (TVL) were determined by the following equation:

\[
TVL = \frac{(V_1 - V_f)}{[\Delta t \cdot A]} = \text{mm d}^{-1}
\]

For both the dry deposition rates and total vapor losses, the number of samples \( n \) needed to estimate the mean value to within \( \pm 10\% \) of the mean at a 95\% confidence level was calculated by the following equation:

\[
n = \left( \frac{t^2 \cdot s^2}{d^2} \right)
\]

where, \( t \) is the student-\( t \) value from the \( t \) distribution initially set equal to 2.00 with an alpha value equal to 0.05, \( s \) is the standard deviation, and \( d \) is the desired confidence interval which was arbitrarily set equal to 0.10 times the mean (Payandeh and Beihartitz, 1979). Since \( t \)-values vary with "n", a minimum of three \( t \)-value iterations were used in obtaining "n" estimates.

Data were statistically analyzed using SAS (SAS Institute, Inc., 1985) software. The following statistical analyses were conducted for the dry deposition rates of calcium, magnesium, nitrate-N, and sulfate-S and the total vapor losses at each site: parametric test (F Test for analysis of variance) and nonparametric test (Kruskal-Wallis Test). Both the F Test and the Kruskal-Wallis Test were conducted at a confidence level = 0.95 (\( \alpha = 0.05 \)). Pairwise comparisons using the Tukey Method were conducted at a confidence level = 0.95 (\( \alpha = 0.05 \)) for both the F Test and the Kruskal-Wallis Test. For the data presented, parametric and nonparametric tests produced the same significant differences for the pairwise comparisons; therefore, the parametric statistical results are shown in the Results and Discussion Section.

Results and Discussion

Initial vs. Final Concentrations

For the one 24 hour exposure period at the open site, the mean initial (blank) and corrected final concentrations for calcium, magnesium, nitrate-N, and sulfate-S are shown in Table 2. At the open site, the corrected final concentrations for all constituents were significantly greater than the initial concentrations. Therefore, these data indicate that a 24-hour exposure period is sufficient to obtain significant increases in the final concentrations over the mean initial concentrations for calcium, magnesium, nitrate-N, and sulfate-S. One exception is that the nitrate-N final concentrations at the forested sites were not significantly greater than the mean initial concentration. This exception will be discussed in greater detail in the Dry Deposition Rates and Number of Samples Required Sections.

Dry Deposition Rates

For each site, the mean dry deposition rates of calcium, magnesium, nitrate-N, and sulfate-S are shown in Table 3. The mean dry deposition rates for both calcium
and magnesium at the open and deciduous sites were significantly greater than at the coniferous site. Calcium and magnesium are both deposited to the snowpack as large particles (Cadle et al., 1985). The low calcium and magnesium snowpack dry deposition rates at the conifer site is probably related to its dense canopy, which trapped some of these large particles.

On the other hand, both the nitrate-N and sulfate-S mean dry deposition rates at the open site were significantly greater than at the deciduous and coniferous sites. Sulfate is deposited to the snowpack as sulfur dioxide gas and particulate sulfate. Nitrate is deposited as particulate nitrate, nitric acid vapor, nitrogen dioxide, nitrogen oxide, PAN (peroxyacetyl nitrate), and other nitrogen species (Cadle et al., 1985; Hicks, 1984). Nitric acid vapor is fairly reactive with most surfaces; hence, most surfaces act as near perfect sinks for this vapor (Hicks, 1984). Cadle, Dasch, and Mulawa (1985) state that in rural areas, the high deposition velocity for nitric acid vapor suggests this vapor will dominate the nitrate dry deposition process (Cadle et al., 1985). The low sulfate-S and nitrate-N dry deposition rates at the forested sites are probably based on the capacity of canopies to effectively absorb gases and capture particles.

Table 2: Mean Initial and Corrected Final Concentrations in Surrogate Snow Samples at the Open Site for March 22-23, 1988

<table>
<thead>
<tr>
<th>Ion</th>
<th>Mean Concentrations (mg L⁻¹)</th>
<th>Number of Samples</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Initial</td>
<td>Final</td>
</tr>
<tr>
<td>Calcium</td>
<td>0.015 a</td>
<td>0.070 b*</td>
</tr>
<tr>
<td>Magnesium</td>
<td>0.004 a</td>
<td>0.015 b</td>
</tr>
<tr>
<td>Nitrate-N</td>
<td>0.019 a</td>
<td>0.061 b</td>
</tr>
<tr>
<td>Sulfate-S</td>
<td>0.01 a</td>
<td>0.04 b</td>
</tr>
</tbody>
</table>

* Mean concentrations values in the same row followed by different letters are significantly different (alpha = 0.05).

Snowpack dry deposition rates, computed from data presented by Cadle et al. (1985), agree quite well with dry deposition rates for the Black Mosshannon open site. For this Northern Michigan open site, average atmospheric concentrations in 1983-84 and corrected deposition velocities, provided by Cadle et al. (1985), were used to calculate the dry deposition rates for calcium, magnesium, sulfate-S, and nitrate-N, which were 0.27, 0.047, 0.20, and 0.19 mg m⁻² d⁻¹, respectively. In Table 3, the dry deposition rates for calcium, magnesium, sulfate-S, and nitrate-N were 0.290, 0.059, 0.16, and 0.210 mg m⁻² d⁻¹, respectively. Therefore, the Black Mosshannon values are remarkably similar to the Northern Michigan values. Sulfate-S and nitrate-N deposition rates were calculated for this comparison by assuming that sulfate is primarily deposited as sulfur dioxide and nitrate is predominately deposited as nitric acid vapor (Cadle et al., 1985). It should also be noted that the sulfur dioxide deposition velocity of 0.06 cm s⁻¹ for ambient temperatures below -30 °C was used in calculating the sulfate-S dry deposition rate (Cadle et al., 1985).

At the open site, the mean sulfate-sulfur deposition rate of 0.16 mg m⁻² d⁻¹ is low, but within the published range of other data for snow. Studies by Barrie and Walmsley (1978), Dovland and Eliassen (1977), and Forland and Gjessing (1975) give snowpack sulfate-sulfur deposition rates ranging between 0.13 and 1.77 mg m⁻² d⁻¹.

Total Vapor Losses

In Table 3 for each site, the mean total vapor losses are shown. The mean total vapor losses at each site were significantly different. These differences in total vapor losses can be explained by the differences in solar radiation and wind speed at the
Table 3: Mean Surrogate Snow Total Vapor Losses and Mean Dry Deposition Rates for Three Sites on March 22-23, 1988

<table>
<thead>
<tr>
<th>Site</th>
<th>Mean Dry Deposition Rates (mg m^-2 d^-1)</th>
<th>Mean Total Vapor Losses (mm d^-1)</th>
<th>Number of Samples</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Ca</td>
<td>Mg</td>
<td>NO3-N</td>
</tr>
<tr>
<td>Open</td>
<td>0.290 a</td>
<td>0.059 a</td>
<td>0.210 a</td>
</tr>
<tr>
<td>Deciduous</td>
<td>0.283 a</td>
<td>0.065 a</td>
<td>0.053 b</td>
</tr>
<tr>
<td>Coniferous</td>
<td>0.184 b*</td>
<td>0.051 b</td>
<td>0.032 b</td>
</tr>
</tbody>
</table>

* Mean Dry Deposition Rate and Mean Vapor Loss values in the same column followed by different letters are significantly different (alpha = 0.05).
surface of the snowpack. Increasing the magnitude of the solar radiation and wind speed will increase both the evaporation and sublimation processes. The wind speed and solar radiation were most likely the highest at the open site and the lowest at the coniferous site. The low mean total vapor losses at the conifer stand is probably related to its dense canopy, which effectively blocked a large portion of the incoming solar radiation and reduced the wind speeds at the snow surface. At the open site, the total vapor loss value is in general agreement with the values for an open site in Ottawa, Canada. Gold and Williams (1961) and Boyd, Gold, and Williams (1962) reported values of 2.4 mm d\(^{-1}\) for March 17–30, 1959 and 1.11 mm d\(^{-1}\) for March 14–31, 1960, respectively.

**Number of Samples Required**

For both the dry deposition rates and the total vapor losses, the number of samples needed to estimate the mean value to within ±10% of the mean at a 95% confidence level are shown in Table 4. In general, the calcium and magnesium mean dry deposition rates plus the total vapor losses required the lowest number of samples. The sulfate-S mean dry deposition rate at the coniferous site and nitrate-N mean dry deposition rate at the open site required a slightly greater number of samples.

Sampling natural snowpacks over time requires more samples than needed for the surrogate snow method. DeWalle et al. (1986) found that the number of samples needed to determine the snowpack dry deposition of sulfate over a four-day period ranged from 66 to 2500 samples per day. Thus, the surrogate snow method, which required 26–45 samples, greatly reduces the sample size needed to estimate the mean surrogate snow dry deposition rates to within ±10% of the mean.

However, nitrate-N dry deposition rates at the forested sites required a much greater number of samples. For the forested sites, nitrate-N was the only species which did not show a significant increase in the mean final concentrations over the initial mean concentration. The final nitrate-N concentrations were 0.061, 0.029, and 0.026 mg L\(^{-1}\) for the open, deciduous, and coniferous sites, respectively; and, the mean initial concentration was 0.019 mg L\(^{-1}\). These very low final concentrations at the forested sites suggest that the forest is effectively reducing nitrate-N dry deposition to the snowpack. When the final concentrations and the mean initial concentration are near the detection limit, small amounts of background contamination can greatly increase the standard deviation, therefore increasing the number to samples needed to estimate the mean to within ±10% of the mean. One possible solution is that the dry deposition rates for nitrate-N at the forest sites may require a longer exposure period so that the final concentrations will be significantly greater than the mean initial concentration.

**Evaluation of the Method**

The three weaknesses of the surrogate snow technique are windblown snow contamination, organic debris contamination, and the melting of snow samples; but, these limitations can be avoided. Windblown snow contamination may occur from any intercepted snow in the forest canopy and from the natural snowpack. No snow was stored in the canopy during tests reported herein. Windblown snow from the snowpack does not appear to be a problem for relatively calm exposure periods. From preliminary data, organic debris contamination has not been a problem for relatively calm exposure periods. Corrections for sample contamination due to leaching of organic or soil debris are possible. The surrogate snow samples may melt more readily than the natural snowpack because these samples are in plastic petri dishes. If the samples are melted, they become very difficult to transport back to the laboratory and the unnatural presence of liquid water will increase dry deposition rates of at least.
sulfur dioxide gas and nitric acid vapor (Johansson and Granat, 1983; Johansson and Granat, 1986). Therefore, the method is best adapted to cold periods when melting can not occur.

Table 4: Number of Samples Required to Estimate the Mean 24-Hour Surrogate Snow Dry Deposition Rates and Total Vapor Losses to within ±10% of the Mean Value for an Alpha = 0.05

<table>
<thead>
<tr>
<th>Site</th>
<th>Open</th>
<th>Deciduous</th>
<th>Coniferous</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ca DDR *</td>
<td>29</td>
<td>11</td>
<td>19</td>
</tr>
<tr>
<td>Mg DDR</td>
<td>20</td>
<td>12</td>
<td>14</td>
</tr>
<tr>
<td>NO3-N DDR</td>
<td>46</td>
<td>622</td>
<td>345</td>
</tr>
<tr>
<td>SO4-S DDR</td>
<td>28</td>
<td>26</td>
<td>45</td>
</tr>
<tr>
<td>TVL</td>
<td>10</td>
<td>30</td>
<td>15</td>
</tr>
</tbody>
</table>

* DDR = Dry Deposition Rate, TVL = Total Vapor Loss

The three strengths of this method are that a relatively short exposure period is needed, a low number of samples are required, and this technique appears to be sensitive in resolving differences in dry deposition rates between open and forested sites. From the data presented, it appears that a 24 hour exposure period is sufficient to calculate dry deposition rates for calcium, magnesium, sulfate-S, and nitrate-N (only at the open site). A relatively low number of samples appear to be required to estimate the mean dry deposition rates to within ±10% of the mean value at a confidence level equal to 0.95 (except for nitrate-N at forested sites). From the presented data, 45 samples seems to be adequate. Also, the surrogate snow technique appears to be fairly sensitive in resolving differences in dry deposition rates between forested and open sites.

Dry deposition measured on surrogate snow samples should only be considered as an index to dry deposition on natural snowpacks. Preliminary tests have indicated that grain size and snow chemistry can affect dry deposition rates. Grain size can be controlled via sieving the shaved ice and methods to control initial snow chemistry are currently under development. Regardless, initial results from the use of surrogate snow samples to monitor snowpack dry deposition are encouraging.

Acknowledgements

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