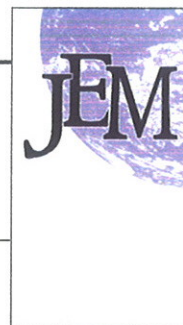


Cloud water and throughfall deposition of mercury and trace elements in a high elevation spruce–fir forest at Mt. Mansfield, Vermont



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As part of the Lake Champlain Basin watershed study of mercury (Hg) and pollutant deposition, cloud water and cloud throughfall collections were conducted at the south summit (1204 m) of Mt. Mansfield, Vermont between August 1 and October 31, 1998, for multi-element chemical analysis. A passive Teflon string collector was deployed during non-precipitating events to sample cloud/fog water at timberline, while three sets of paired funnels collected cloud throughfall under the red spruce–balsam fir canopy. Samples were analyzed for concentrations of Hg, major ions, and 10 trace elements. Ultra-clean sampling and analysis techniques were utilized throughout the study. Six events were sampled for cloud water alone and four events were sampled for both cloud water and cloud throughfall. Cloud throughfall chemistry showed substantial modification from incident cloud water. Much higher concentrations of Hg (2.3 ×), base cations (Ca²⁺, K⁺, Mg²⁺; 3–18 ×) and certain trace elements (Ni, Cu, Mn, Rb, Sr; 2–34 ×) were observed in throughfall than in cloud water. These results confirm that cloud water can leach a wide variety of elements from tree foliage and wash off dry deposited elements. Cloud water deposited an average of 0.42 ± 0.12 mm of water per hour. Estimated cloud water deposition of Hg was 7.4 µg m⁻² for the period August 1–October 31, approximately twice that deposited by rain during this period at a nearby low elevation Hg monitoring site. Our results indicate that cloud water and Hg deposition at Mt. Mansfield are likely to have considerable ecological effects.

Introduction

Cloud water has been shown to be ecologically significant and important in the deposition of water and atmospheric pollutants in high elevation forests.^{1–4} The deposition of cloud water droplets on exposed surfaces occurs during both precipitating and non-precipitating cloud events and represents a large portion (30–50%) of total water inputs (rain + snow + cloud) in ecosystems above 1000 m elevation.^{5–8} Research has consistently shown higher concentrations of major ions and other atmospheric pollutants in cloud water compared to precipitation collected at the same site.^{3,8–11} Previous studies in the northeastern USA have documented significant loading of toxic metals such as lead, cadmium, and copper in cloud water events.^{3,12–14} However, concentrations of mercury (Hg) in cloud water have not yet been reported in the literature.

Mercury is a naturally occurring element that is also considered a highly toxic environmental pollutant of global concern, due to additions of Hg to the atmosphere by anthropogenic emissions. In Vermont, elevated levels of Hg have been measured in lake water and tissues of fish species in the Lake Champlain Basin.¹⁵ Mercury contamination of the lake has been attributed to direct wet and dry atmospheric deposition and the transport of Hg in surface waters to the lake.¹⁶ Previous estimates of Hg loading to this watershed^{17,18} have not accounted for cloud water inputs of Hg to high elevations and likely underestimated total Hg deposition. The input of Hg to high elevation sites is likely much greater than low elevation sites due to orographically enhanced precipitation and the additional contribution from clouds.

A progressive decline of high elevation forests in the

northeastern US in recent decades has been well documented.^{19–23} In Vermont, high elevation red spruce forests have shown extensive mortality and significantly reduced density and basal area over the last 40 years.^{24,25} Acidic deposition has directly contributed to the observed decline of red spruce in the Northeast.^{23,26,27}

Cloud water is an important source for the loading of chemicals to mountain ecosystems and entire watersheds. This research aims to improve the quantification of pollutant inputs to the Lake Champlain watershed and provide measurements of Hg in cloud water and cloud throughfall. The objectives of this paper are to quantify the deposition of water, mercury, and trace elements by clouds and to provide an assessment of the forest canopy processing of cloud water by analyzing cloud throughfall. A companion paper²⁸ presents a second set of objectives, which are to quantify Hg in cloud water, characterize cloud chemistry at a high-elevation site, and understand the factors that control measured concentrations. Data for concentrations of Hg in clouds are needed, and this study makes a contribution to the limited number of observations of trace elements in cloud water.

Methods

Site description

The study site is the south summit of Mount Mansfield (1204 m), located along the spine of Green Mountains in north-central Vermont. Mt. Mansfield has been a site for meteorological measurements and long-term ecological study since 1954.

The high elevation forest at this site is predominately balsam fir [*Abies balsamea* (L.) Mill.] with a lesser component of red spruce [*Picea rubens* Sarg.]. The canopy is characterized by a highly variable mix of mature trees, regenerating clumps of

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young trees, and open gaps created by wind throw or forest dieback. The average canopy height is approximately 7–10 m which decreases rapidly near the elevation limit of this forest at about 1200 m. Meteorological measurements are made in an open area adjacent to several buildings which service radio and television transmitter towers at the summit. The site receives an annual average of 174 cm of precipitation (1955–1998) which is twice the annual average of 87 cm (1955–1994) at Burlington, Vermont, a low elevation site (101 m) located on the eastern shore of Lake Champlain.²⁹ The weather during our sampling period (August 1–October 31, 1998) was representative of average conditions during this time of the year at Mt. Mansfield.²⁹

Cloud water and throughfall collection

Cloud water was sampled using a passive Teflon string collector (Fig. 1) designed for this study.²⁸ The cloud water collector was installed just above the timberline on the western slope of Mt. Mansfield, near the summit, allowing direct interception of ambient clouds. Cloud throughfall was sampled with three replicate pairs of funnel collectors (each pair consisting of one glass and one polypropylene funnel; 182.4 and 167.55 cm² collection area, respectively) placed under the canopy of the mature spruce–fir forest spaced approximately 15–20 m apart in random directions with the funnel opening 0.5 m above the ground.¹⁷ The throughfall collection site is approximately 10 m lower in elevation and 100 m southwest of the cloud collector in an area with a gentle slope (<5%) and a western aspect. The methods for chemical analyses are described in Malcolm *et al.*²⁸

Throughfall collection was initiated during periods when regional non-precipitating clouds were present at the summit (not localized or “cap clouds”). Sampling commenced after a “loading-up” period of at least 2 h, which allowed the trees to become saturated with cloud water and begin to drip.⁵ Our

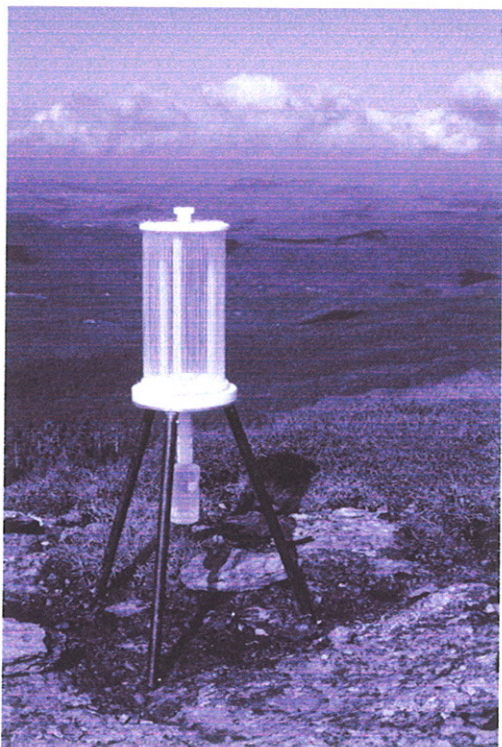


Fig. 1 Cloud collector at Mt. Mansfield, Vermont (photo courtesy of Sean T. Lawson).

observations at this site indicated that little or no volume of drip water could be collected during this loading period. However, initial wash-off is likely to be more concentrated, and our methodology may have the potential to bias TF concentrations downward. When the canopy is saturated, throughfall closely approximates net cloud deposition⁵ and the drip rate is sufficient to collect the volume required for sample analyses. This method slightly underestimates total deposition, since there is some evaporation of water from the canopy (interception loss) and some water moves to the forest floor as stemflow.⁵ However, interception loss is likely to be small under the high humidity conditions that were sampled, and stemflow has been found to be very low ($\leq 2\%$ of water deposited) in coniferous stands.³⁰

Ultra-clean sampling techniques were utilized in all phases of collection and handling of samples. Funnels and bottles were subjected to a rigorous, multi-day acid washing process and stored in triplicate zip-lock bags.²⁸ Particle-free gloves were worn at all times during handling of equipment and sample bottles. Polypropylene funnels and bottles were used to collect cloud water throughfall for major ion and trace element analyses, while glass funnels and Teflon bottles (1 l capacity with 20 ml 0.08 M HCL acid preservative) were used for mercury analysis. After collection, sample bottles were sealed with Teflon tape and triple bagged for transport to the laboratory.

Total cloud water deposition is defined as canopy throughfall (TF) + stemflow (water flowing along the main stem of trees) + canopy retention (the amount of water required to saturate the canopy) + evaporation. Net cloud water deposition is defined as the quantity of water that reaches the forest floor, measured as cloud throughfall, which slightly underestimates total deposition. Deposition of Hg, trace elements, and major ions by cloud water was calculated by multiplying volume-weighted mean cloud water concentration data from the passive string collector by the mean TF volume collected. Deposition of these elements by cloud TF was calculated by multiplying the volume-weighted mean TF concentration data by the total volume of TF. Net TF deposition was calculated as total TF deposition minus cloud deposition.

The passive collector used in this study does not exclude precipitation, thus sampling was only conducted during non-precipitating cloud periods. The cloud collector and funnels were deployed during appropriate cloud conditions and then dismantled and acid cleaned after each cloud event. Two to three sequential samples were obtained for cloud water in each event when sample volume was sufficient, while three replicates of cloud throughfall were obtained in each event. Sequential samples were volume-weighted to yield an event mean for cloud water concentrations. Ten events were sampled. Four events provided concurrent collection of both cloud water and TF, while six events were sampled for cloud water only.

Sample analyses

Samples were immediately transported to the Proctor Maple Research Center (PMRC) in Underhill Center, Vermont, where sub-samples were poured off in a portable HEPA filtered air chamber for major ion analysis. Bulk samples were shipped overnight to the University of Michigan Air Quality Laboratory (UMAQL) for trace element and mercury analyses. Sub-samples were transported to the University of Vermont Environmental Testing Laboratory in Burlington, Vermont, where they were stored frozen ($-20\text{ }^{\circ}\text{C}$) for 2 to 6 weeks until major ion analyses (Ca^{2+} , K^{+} , Mg^{2+} , Na^{+} , SO_4^{2-}) were performed.²⁸

A series of field blanks were collected by pouring ~ 500 ml of de-ionized water through a funnel, washing all sides, into a sample bottle ($n = 6$) following the protocol of Landis and

Keeler.³¹ Concentrations of Hg, trace elements, and major ions in blanks were below method detection limits. QA/QC procedures for all other data analyses are presented in Malcolm *et al.*²⁸

Results and discussion

Cloud water deposition

Cloud deposition rates were calculated by dividing the volume of water collected in the TF funnel collectors (ml) by the collecting surface area of the funnels (cm²) and the duration of collection (h). Three non-precipitating cloud events were sampled on 8/18, 8/26, and 9/10. The following rates were observed, respectively: 0.52 ± 0.15 , 0.29 ± 0.0 , and 0.33 ± 0.14 mm H₂O h⁻¹. The mean deposition rate for the three events was 0.42 ± 0.12 mm H₂O h⁻¹. These results agree closely with the mean deposition rate (0.45 mm h⁻¹) measured at Whitetop Mountain,³² and are slightly higher than the rates observed and modeled (0.1–0.4 mm h⁻¹) at Mt. Moosilauke, NH.⁵ Miller *et al.*⁸ used a modified version of Lovett's model to calculate cloud water deposition at Whiteface Mountain, NY. They reported an estimated annual cloud water deposition of 81.1 cm at 1225 m elevation and an observed cloud frequency of 23.2%. These results yield an average deposition rate of 0.39 mm h⁻¹ during cloud immersion, which is very close to our short-term mean. Based on visual observations and meteorological records,²⁹ cloud immersion at the summit of Mt. Mansfield is estimated at 25% of the year. Table 1 shows estimated annual deposition rates of cloud water at similar sites in the northeastern US.

Cloud water interception in high elevation ecosystems varies considerably over short distances as a function of micrometeorology, wind speed, liquid water content, droplet size distribution, and forest canopy structure.^{5,33} Exposed forest edges and individual trees that protrude from the canopy will collect much greater quantities (up to 10×) of cloud water than a homogeneous canopy.^{14,34} Because high-elevation forests are quite heterogeneous and cloud immersion changes dramatically with elevation,⁸ cloud deposition rates will vary widely across different elevations and years in these ecosystems.

Throughfall chemistry

Throughfall samples ($n = 10$) in four events ranged in volume from 12 to 124 ml, with a mean of 71 ml. In the event on 8/26, only one replicate contained enough volume (12 ml) to perform analyses. Due to the small volume of this sample, only trace element analysis was performed. Therefore, in comparisons of incident cloud water and throughfall chemistry, trace elements have four paired collections, while major ions and Hg have three paired collections.

Major ions. Major ion concentrations for three paired cloud water and throughfall collections are shown in Table 2. Due to the small size of the data set, a rigorous statistical analysis is not possible and our interpretations are limited. Base cation

Table 2 Mean (volume-weighted) concentrations of major ions in three paired cloud water and throughfall collections at Mt. Mansfield, Vermont

Ion/mg L ⁻¹	Ca ²⁺	K ⁺	Mg ²⁺	Na ⁺	SO ₄ ²⁻
<i>Cloud water</i>					
Mean	0.17	0.26	0.009	0.08	0.86
Standard deviation	0.12	0.12	0.007	0.04	0.53
Median	0.15	0.29	0.014	0.10	0.56
Maximum	0.32	0.52	0.020	0.12	1.77
Minimum ^b	<0.005	<0.05	<0.005	<0.005	0.29
<i>Throughfall</i>					
Mean	0.57	1.10	0.168	0.11	1.33
Standard deviation	0.62	0.66	0.196	0.05	1.39
Median	0.21	1.00	0.040	0.14	0.57
Maximum	2.04	2.56	0.557	0.26	4.52
Minimum ^b	0.09	0.50	0.010	<0.005	0.33
Net throughfall	0.40	0.84	0.16	0.03	0.47
TF Enrichment factor ^a	3.4	4.2	18.1	1.4	1.5

^aEnrichment factor calculated as mean throughfall divided by mean cloud water. ^bSome sample concentrations were below instrument detection limits.

concentrations were much higher in throughfall than in incident cloud water, with an increase of 3.4× for Ca²⁺, 4.2× for K⁺, and 18.1× for Mg²⁺. Enrichment of these cations can be due to leaching from foliage, wash-off of dry deposition, or evaporation in the forest canopy, although evaporation is generally low during cloud immersion.^{5,35}

It is well known that foliar leaching of cations increases significantly as the pH of input water decreases.^{36–38} Joslin *et al.*³⁷ found large increases of base cations in cloud TF in a spruce forest, with Ca²⁺, Mg⁺ and K⁺ concentrations 8×, 4×, and 7× higher than in cloud water, respectively. Foliar Ca²⁺ and Mg⁺ losses were markedly intensified by increased acidity in this study. DeHayes *et al.*³⁸ demonstrated that leaching by acidic clouds causes significant loss of Ca²⁺ from red spruce needles, leads to foliar membrane instability, and decreases foliar cold tolerance by 3–10 °C. Foliar calcium depletion is linked to the observed decline in high elevation red spruce in Vermont over the past 40 years, and is now found to cause similar disruptions in other tree species, posing a potential threat to forest and ecosystem health.^{38,39}

Mercury and trace elements. Throughfall concentrations of mercury and certain trace elements were considerably higher than in cloud water (Table 3). The mean TF concentration of Hg (17.5 ± 6.6 ng L⁻¹) was 2.3× higher than in cloud water (7.5 ± 2.7 ng L⁻¹) collected concurrently. This increase is similar to that observed in precipitation TF (1.85×) by Rea *et al.*¹⁷ in a lower elevation (400 m) hardwood forest on the western slope of Mt. Mansfield. Increased Hg concentrations in TF have been attributed to wash-off of dry deposited Hg from tree foliage.^{17,40} We observed that increases in concentrations of Hg in TF vs. cloud water at Mt. Mansfield were larger for two events (8/18 and 10/1) preceded by a dry period of >48 h

Table 1 Estimated annual cloud water deposition at mountain locations in the northeastern US

Site	Elevation/m	Year	Cloud deposition/ cm per year	Cloud frequency (%)	Reference
Whiteface Mountain, NY	1050	1986–89	28.4	10	Miller <i>et al.</i> (1993b)
Whiteface Mountain, NY	1225	1986–90	81.1	23	Miller <i>et al.</i> (1993a)
Whiteface Mountain, NY	1350	1986–89	153.5	36	Miller <i>et al.</i> (1993a)
Whiteface Mountain, NY	1483	1987	127.0	42	Mohren (1988)
Mt. Moosilauke, NH	1220	1990	40.5	—	Schaefer and Reiners (1990)
Mt. Moosilauke, NH	1220	1980–81	84.0	40	Lovett <i>et al.</i> (1982)
Madonna Mt., VT	1110	1980–81	154.0	—	Scherbatsky and Bliss (1984)
Camels Hump, VT	1110	1970	76.0	—	Leedy (1972)

Table 3 Mean concentrations (volume-weighted) of trace elements in four^b paired cloud water and throughfall collections at Mt. Mansfield, Vermont

Element/ $\mu\text{g L}^{-1}$	As	Cd	Cr	Cu	Hg ^b	Mn	Ni	Pb	Rb	Sr	Zn
<i>Cloud water</i>											
Mean	0.13	0.03	0.09	0.51	7.5	1.7	0.24	0.58	0.07	0.30	5.5
Standard deviation	0.68	0.16	0.19	1.11	2.7	5.3	0.58	2.56	0.34	1.42	12.1
Median	0.18	0.03	0.10	0.56	6.1	1.5	0.24	0.93	0.13	0.51	4.0
Maximum	2.12	0.52	0.63	3.64	11.8	17.3	1.90	8.17	1.08	4.61	30.3
Minimum	0.04	0.01	0.01	0.12	3.7	0.3	0.05	0.11	0.03	0.04	0.7
<i>Throughfall</i>											
Mean	0.18	0.07	0.11	0.99	17.5	40.9	0.64	0.76	2.53	1.18	6.9
Standard deviation	0.12	0.09	0.08	0.36	6.6	52.3	0.57	0.49	3.14	1.73	6.8
Median	0.13	0.04	0.13	1.03	16.5	50.7	0.64	0.62	2.06	0.53	3.7
Maximum	0.39	0.27	0.23	1.61	33.3	172.1	2.11	1.54	10.26	4.62	23.9
Minimum	0.07	0.01	0.00	0.54	9.9	5.0	0.26	0.27	0.52	0.17	2.9
Net throughfall	0.05	0.05	0.02	0.48	10.0	39.2	0.40	0.18	2.46	0.88	1.4
Enrichment factor ^a	1.4	3.0	1.2	1.9	2.3	23.7	2.7	1.3	38.3	4.0	1.3

^aEnrichment factor calculated as mean throughfall divided by mean cloud water. ^bHg units are ng L^{-1} in three paired collections of cloud water and throughfall.

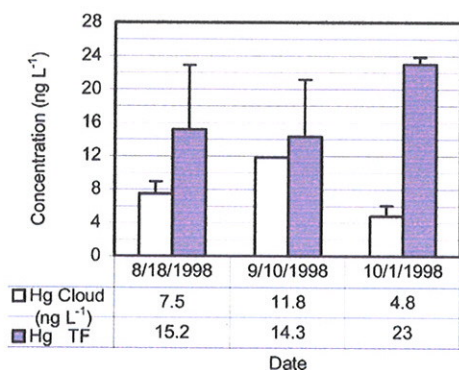


Fig. 2 Concentrations of mercury (Hg) in cloud water compared to throughfall (TF) during two cloud events preceded by >48 h dry period (8/18, 10/1) and one event preceded by >48 h cloud immersion and precipitation (9/10) at Mt. Mansfield, Vermont.

than in an event (9/10) preceded by 48 h of cloud immersion and intermittent rain showers (Fig. 2). Many other studies have shown that wash-off concentrations of elements increase with the length of dry period preceding a canopy wetting.

Remarkably similar increases in Hg concentrations for TF vs. precipitation have been observed across different forest types and years. At Walker Branch, a deciduous forest in Tennessee, precipitation TF had a mean concentration of Hg about 2 \times higher than rainwater.⁴⁰ In a coniferous forest in Sweden, annual mean TF concentration of Hg was 1.8 \times to 2.3 \times higher than precipitation over three study years.⁴¹ These studies suggest that similar processes (e.g., wash-off of Hg on foliar surfaces) operate for TF deposition of Hg across different geographic regions and tree species.

The trace elements selected for our study were chosen due to their toxicity (Hg, Cr, Ni, As, Cd, Pb), importance as plant nutrients (Zn, Mn, Cu) or behavior similar to essential plant

Table 4 Cloud water and throughfall deposition of major ions to the forest floor during three cloud events and estimated deposition by cloud water for the three month period (August 1–October 31, 1998) at Mt. Mansfield, Vermont

Deposition/ mg m^{-2}	Ca ²⁺	K ⁺	Mg ²⁺	Na ⁺	SO ₄ ²⁻
Cloud water	2.1	2.7	0.15	0.65	9.2
Total throughfall	8.0	15.5	2.50	1.47	18.3
Net throughfall	5.9	12.8	2.35	0.82	9.1
Estimated deposition/ mg m^{-2} August 1– October 31, 1998	90	100	40	50	640

nutrients (Rb mimics Mn, Sr mimics Ca²⁺). Some of the trace elements (Cu, Mn, Rb, Sr) are subject to leaching from the leaves of plants.^{42–44} In our study at Mt. Mansfield, mean concentrations of Mn and Rb were 24 \times and 38 \times higher, respectively, in throughfall than in cloud water which likely indicates a strong influence of foliar leaching on these elements. Sr and Ca²⁺ have similar behavior in plants. Sr was enriched 4 \times in throughfall, which is close to the observed 3.5 \times increase in Ca²⁺.

The toxic group of elements (Hg, Cr, Ni, As, Cd, Pb) have known emission sources to the atmosphere such as combustion of fossil fuels, smelters, and incineration⁴⁵ are deposited by both wet and dry processes, and are generally not leached from foliage.⁴² Cloud events which follow periods without precipitation can wash dry deposited materials from foliar surfaces, as observed for Hg and Pb in precipitation throughfall.^{17,46} Similar increases in throughfall concentrations were also observed for other dry deposited metals at Mt. Mansfield (Cr, Ni, Cd).

Mercury and trace element deposition

Major ion deposition for four events is shown in Table 4, while mercury and trace elements are shown in Table 5. Note that the

Table 5 Cloud water and throughfall deposition of trace elements to the forest floor during four cloud events and estimated deposition by cloud water for the three-month period (August 1–October 31, 1998) at Mt. Mansfield, Vermont

Deposition/ $\mu\text{g m}^{-2}$	As	Cd	Cr	Cu	Hg ^a	Mn	Ni	Pb	Rb	Sr	Zn
Cloud water	4.4	0.54	1.49	8.2	8.0	30.6	2.9	9.9	1.6	6.5	90.5
Total throughfall	2.8	1.14	1.84	15.3	16.0	693.5	10.4	11.7	41.9	19.0	94.7
Net throughfall	-1.6	0.60	0.35	7.1	8.0	662.9	7.5	1.8	40.3	12.5	4.2
Estimated cloud deposition/ mg m^{-2} August 1–October 31, 1998	0.19	0.05	0.05	0.55	7.4 ^b	1.0	0.20	0.82	0.07	0.27	5.5

^aHg units are ng L^{-1} ^bHg units are $\mu\text{g m}^{-2}$

cloud water, throughfall, and net deposition displayed are for four events only. Estimated deposition for the three-month sample period (August 1–October 31, 1998) is shown on the last line, and was calculated using the mean deposition rate of cloud water (0.42 mm h^{-1}), the mean incident cloud concentration from all events measured, and an estimated cloud frequency of 35% during this period of time.²⁹ Mercury deposition by cloud water was estimated at $7.4 \mu\text{g m}^{-2}$ for the three month period. This result is nearly twice the measured deposition of Hg in precipitation ($3.9 \mu\text{g m}^{-2}$) for the same period, and is comparable to annual precipitation Hg deposition at the low elevation monitoring site (PMRC $\sim 400 \text{ m}$) on Mt. Mansfield.¹⁶ Precipitation deposition of Hg shows a strong seasonal trend and is highest in the summer.^{18,41}

Deposition of Hg by precipitation to low elevation forests in Vermont has been reported to be 7 to $9 \mu\text{g m}^{-2}$ per year.¹⁶ Annual precipitation at the summit of Mt. Mansfield is $2 \times$ that of nearby low elevation sites.²⁹ With increased precipitation and added cloud deposition, high elevations in the Green Mountains are subject to a wet loading of Hg that is at least $3 \times$ higher than in the surrounding lowlands. Thus, we estimate that wet deposition of Hg is in excess of $24 \mu\text{g m}^{-2}$ per year at high elevation sites in Vermont. This estimate does not consider dry deposition and suggests that total Hg loading to high elevation sites may be much higher.

Increased wet deposition of pollutants at high elevations has been well documented in other studies. Forests in the Adirondack Mountains, NY, where precipitation and cloud water account for over 90% of the annual deposition of N and S, receive up to five times more N and four times more S than surrounding low-elevation forests.⁸ Our data for trace elements are among the few reported for TF in this region or in North America. It is important to emphasize that the contribution by cloud water of major ions, toxic metals, and other atmospheric pollutants are enhancements in addition to precipitation.

Conclusions

Cloud water is a significant source of Hg and pollutant deposition to high-elevation ecosystems. The toxicity of Hg is well known for humans and certain fauna, but the effects of Hg deposition on high elevation forests and other alpine biota are largely unknown. Cloud and precipitation TF washes off dry deposited Hg from tree foliage, increasing Hg and pollutant deposition on the forest floor. Total atmospheric deposition of Hg and other pollutants at Mt. Mansfield may be $2 \times$ to $5 \times$ higher than at surrounding low elevation sites. Mt. Mansfield likely receives a high proportion of the total annual water budget directly from clouds, as observed at other high elevation sites, resulting in enhanced inputs of both water and pollutants to this ecosystem. As such, interception of cloud water by mountains must be included in estimating pollutant deposition to the Lake Champlain Basin and other watersheds that contain landscapes subject to cloud immersion.

The fate of large mercury deposits in high elevation ecosystems remains unknown. In a study of Hg transport in a forested catchment on the lower slopes of Mt. Mansfield, Scherbatskoy *et al.*¹⁶ found net retention of 92% and 95% of total Hg deposition in two study years. These results are based upon export of Hg in surface waters. While a small fraction of deposited Hg may be released from soil as Hg vapor, the strong association of Hg with soil organic matter is evident.¹⁶ Therefore, we conclude that the vast majority of Hg deposited each year is retained in the ecosystem, accumulating in the soil. Further research should focus on Hg accumulation rates in mountain soils, terrestrial food webs, and sensitive species, as Hg deposition may pose a serious long-term toxicity threat to the biota of alpine ecosystems.

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References

- 1 H. W. Vogelmann, T. Siccama, D. Leedy and D. C. Ovit, *Ecology*, 1968, **49**, 1205.
- 2 G. M. Lovett, W. A. Reinert and R. K. Olson, *Science*, 1982, **218**, 1303.
- 3 T. Scherbatskoy and M. Bliss, in *The Meteorology of Acid Deposition*, ed. P. J. Samson, Air Pollution Control Association, Pittsburg, 1984, pp. 449–456.
- 4 G. M. Lovett and J. D. Kinsman, *Atmos. Environ., Part A*, 1990, **24**, 2767.
- 5 G. M. Lovett, *Atmos. Environ.*, 1984, **18**, 361.
- 6 D. A. Leedy, *MS Thesis*, University of Vermont, Burlington, 1972.
- 7 R. J. Vong, J. T. Sigmon and S. F. Mueller, *Environ. Sci. Technol.*, 1991, **25**, 1014.
- 8 E. K. Miller, A. J. Friedland, E. A. Arons, V. A. Mohonen, J. J. Battles, J. A. Panek, J. Kadlecek and A. H. Johnson, *Atmos. Environ., Part A*, 1993, **27**, 2121.
- 9 H. Mrose, *Tellus*, 1966, **18**, 266.
- 10 R. E. Falconer and P. D. Falconer, *J. Geophys. Res.*, 1980, **85**, 7465.
- 11 K. C. Weathers, G. E. Likens, F. H. Bormann, S. H. Bicknell, B. T. Bormann, B. C. Daube, Jr., J. S. Eaton, J. N. Galloway, W. C. Keene, K. D. Kimball, W. H. McDowell, T. G. Siccama, D. Smiley and R. A. Tarrant, *Environ. Sci. Technol.*, 1988, **22**, 1018.
- 12 V. A. Mohonen and R. J. Vong, *Environ. Rev.*, 1993, **1**, 38.
- 13 E. K. Miller, J. A. Panek, A. J. Friedland, J. Kadlecek and V. A. Mohonen, *Tellus Ser. B*, 1993, **45**, 209.
- 14 K. C. Weathers, G. M. Lovett and G. E. Likens, *Atmos. Environ.*, 1995, **29**, 665.
- 15 M. C. Watzin, *A research and monitoring agenda for Lake Champlain: Proceedings of a workshop*, Lake Champlain Basin Program Technical Report No. 1, US EPA, Boston, 1992.
- 16 T. Scherbatskoy, J. B. Shanley and G. J. Keeler, *Water, Air, Soil Pollut.*, 1998, **105**, 427.
- 17 A. W. Rea, G. J. Keeler and T. Scherbatskoy, *Atmos. Environ.*, 1996, **30**, 3257.
- 18 T. Scherbatskoy, J. M. Burke, A. W. Rea and G. J. Keeler, in *Atmospheric Deposition of Contaminants to Great Lakes and Coastal Waters*, ed. J. E. Baker, SETAC Press, Pensacola, 1997, pp. 245–257.
- 19 A. H. Johnson and T. G. Siccama, *Environ. Sci. Technol.*, 1983, **17**, 294A.
- 20 J. T. Scott, T. G. Siccama, A. H. Johnson and A. R. Breisch, *Bull. Torrey Bot. Club*, 1984, **111**, 438.
- 21 H. W. Vogelmann, G. J. Badger, M. Bliss and R. M. Klein, *Bull. Torrey Bot. Club*, 1985, **112**, 274.
- 22 *Ecology and Decline of Red Spruce in the Eastern United States*, ed. C. Eager and M. B. Adams, Springer-Verlag, London, 1992.
- 23 *National Acid Precipitation Assessment Program Report to Congress*, US Government Printing Office, Pittsburgh, 1992.
- 24 R. M. Klein and T. D. Perkins, in *ACS Symposium Series 483*, ed. D. A. Dunnette and R. J. O'Brien, American Chemical Society, Washington DC, 1992.
- 25 A. H. Johnson, D. H. DeHayes and T. G. Siccama, in *Forest Trees and Palms: Disease and Control*, ed. S. P. Raychudhuri and K. Maramorosch, Oxford University Press and IBH Publishers, New Delhi, India, 1996, pp. 49–71.

- 26 *Acidic Deposition at High Elevation sites*, ed. M. H. Unsworth and D. Fowler, Kluwer Academics, Dordrecht, The Netherlands, 1988.
- 27 D. H. DeHayes, in *Ecology and Decline of Red Spruce in the Eastern United States*, ed. C. Egar and M. B. Adams, Springer-Verlag, New York, 1992, pp. 295-337.
- 28 E. G. Malcolm, G. J. Keeler, S. T. Lawson and T. D. Scherbatskoy, *J. Environ. Monit.*, 10.1039/b210124f.
- 29 *Cooperative Summary of the Day (TD3200)*, National Climatic Data Center, National Oceanic and Atmospheric Administration, US Department of Commerce, Asheville, 1999.
- 30 S. E. Lindberg and J. G. Owens, *Biogeochem.*, 1993, **19**, 173.
- 31 M. S. Landis and G. J. Keeler, *Environ. Sci. Technol.*, 1997, **31**, 2610.
- 32 S. F. Mueller, J. D. Joslin, Jr and M. H. Wolfe, *Atmos. Environ. Part A*, 1991, **25**, 1105.
- 33 G. M. Lovett, *Ecol. App.*, 1994, **4**, 629.
- 34 J. M. Dasch, *Atmos. Environ.*, 1988, **22**, 2255.
- 35 W. J. Shuttleworth, *Boundary-Layer Meteor.*, 1977, **12**, 463.
- 36 T. Scherbatskoy and R. M. Klein, *J. Environ. Qual.*, 1983, **12**, 189.
- 37 J. D. Joslin, C. McDuffie and P. F. Brewer, *Water, Air, Soil Pollut.*, 1988, **39**, 355.
- 38 D. H. DeHayes, P. G. Schaberg, G. J. Hawley and G. R. Strimbeck, *BioScience*, 1999, **49**, 1.
- 39 P. G. Schaberg, D. H. DeHayes and G. J. Hawley, *Eco. Health*, 2001, **7**, 214.
- 40 S. E. Lindberg, J. G. Owens and W. J. Stratton, in *Mercury Pollution: Integration and Synthesis*, ed. C. J. Watras and J. W. Huckabee, Lewish Publishers, Ann Arbor, 1994, pp. 261-270.
- 41 J. Munthe, H. Hultberg and A. Iverfeldt, *Water, Air, Soil Pollut.*, 1995, **80**, 363.
- 42 A. W. Rea, *PhD Dissertation*, University of Michigan, Ann Arbor, 1998.
- 43 H. Heinrichs and R. Mayer, *J. Environ. Qual.*, 1980, **9**, 111.
- 44 S. E. Lindberg, in *Control and Fate of Atmospheric Trace Metals*, ed. J. M. Pacyna and B. Ottar, Kluwer Academic Publishers, Dordrecht, The Netherlands, 1989.
- 45 J. B. Milford and C. I. Davidson, *JAPCA*, 1985, **35**, 1249.
- 46 A. Iverfeldt, *Water, Air, Soil Pollut.*, 1991, **56**, 553.