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On the Determination of Total Deposition to Remote Areas

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Abstract

Discussion of "critical loads" and "total deposition" has focused attention on the importance of such concepts in the regulatory and policy processes, yet actual quantification of either is exceedingly demanding. Here, alternative measurement and modeling approaches will be explored, and the effects of complicating factors like terrain complexity and vegetation inhomogeneity will be considered. Patchy surfaces tend to receive more deposition than is indicated by a simple area-weighting, as can be easily demonstrated for patches of trees on a grassy countryside. The magnitude of the difference is greatest for substances that are transferred with low surface resistance. In general, for applications that demand consideration of target areas that are currently unstudied, an exploratory measurement program is desirable, coupled with modeling activity that is then benchmarked against the observational data base. Without models, the measurement programs can only address those specific areas that are studied. Without measurements, the models risk failing to allow for site-specific peculiarities that may be dominant. Thus, in general an integrated approach seems optimal, though perhaps difficult to attain in many practical situations.

1. INTRODUCTION

In contemporary society, the easy decisions about how to design and impose remedial emission controls have already been made. The remaining difficult decisions require more justification, and decision makers turn to models for this support. Modelers are faced with the need to compress complicated considerations into simple concepts, among which the ideas of "critical loads" and "total deposition" have achieved considerable favor.

A "critical load" is a level of pollution input into an ecosystem below which specific damage will not occur, and above which damage is expected. "Total deposition" is the sum of all input derived from the atmosphere. Both concepts are intended to be aids to the decision-making process, but the question arises as to how much the complexity of the subject jeopardizes the conclusions that are reached using such simple concepts.

Consider a lake that is potentially susceptible to acid deposition. The concentrations of pollution in the water itself are determined by the flow of pollutant into the lake and the flow out of it — the situation is dynamic, and the pollutants do not merely concentrate in the water as more and more input occurs. But it is not only the deposition to the water surface that defines the "load" of importance, but also the deposition to the surrounding catchment area. There are clearly two limits on what might constitute the quantity of importance — the total deposition (wet plus dry plus that due to fog) to the water surface (Fw) on the one extreme, and the total (Fc) to the entire catchment area on the opposite extreme. We must necessarily ask what input property is most relevant — Fw, Fc, or some intermediate value.

The important measure of deposition is surely that which correlates most directly with the effects to be avoided or alleviated. From the perspective considered above, it is not either of Fc or Fw, but the net effect of these as influences the lake itself. Part of the potential insult delivered to the catchment as Fc will be retained by the catchment and not delivered to the lake. That part which is delivered may well be delayed for years. How the net influence on the lake at any specific time relates to any index of deposition from the atmosphere constitutes a question that is generally unanswered. To obtain the answers would require site-specific research that has usually proved too expensive for replication. What is now available is enough understanding to recognize that SOME deposition to a catchment affects the water bodies in it, but is not yet adequate to enable confident prediction of the retention rate without on-site studies for guidance. There is need to specify the area that must be considered, and to determine (preferably on the basis of external information) the weighting factor to apply to the deposition occurring at different places.

All of the discussion above assumes that accurate atmospheric deposition rates can indeed be determined. In fact, the concept of "total deposition" carries different meanings in different communities. Atmospheric sciences are unified on meaning of "total deposition:" it is the sum of deposition from the atmosphere by wet and dry mechanisms, with a (usually small) additional component due to interception of fog droplets and cloud liquid water, all expressed as mass of pollutant per unit time and per unit horizontal area. In materials science, however, "total deposition" refers to the deposition (by all mechanisms) to a portion of an exposed structure; in this regard, it is not the net vertical flux from the atmosphere that is considered, but the rate of transfer to specific elements of the surface, such as the surroundings of a window, the arm of a statue, or the roof of an automobile. This concept extends to many modeling studies, such as in wind tunnels, where even deposition velocities are quantified as being appropriate for a specific leaf, for example, instead of for a layer of trees constituting the lower boundary of the atmosphere.

In some modern usage, distinctions between receptor element-specific and atmospheric areal average terminology are made by the use of lower-case symbols for the former and upper case for the latter. Thus, for a given coniferous forested landscape the deposition velocity appropriate to compute the flux of material to a single pine needle may be $v_d(n)$, to the trunk of a pine tree $v_d(t)$, and to the underlying forest floor $v_d(s)$. At the same time, the flux from the atmosphere to a unit horizontal area of the same pine forest is computed using a separate quantity V_d. A major challenge is to relate V_d to $v_d(n,t,s)$.

Yet another meaning of "total deposition" arises in some ecological literature, where it refers to deposition arriving at a forest floor, as often measured collecting precipitation samples beneath any vegetation canopy. These samples are affected by (a) wet deposition to the canopy, (b) dry deposition intercepted by the canopy during the period since the previous "wash-off", (c) release of materials following transfer from the root systems to the foliage, and (d) deposition not due to atmospheric transport aloft but to local redistribution from one part of the surface to another. In practice, long-term throughfall measurements also include the leaf-fall component for deciduous vegetation. Measurements of this kind are well suited to quantify deposition as it may affect soils and aquatic ecosystems. The measurements are less well suited to considerations of atmospheric transport or of effects on vegetation. It is clear, however, that the two divergent definitions of total deposition given above both relate to the flux of material to the roughness elements that are exposed to air, and that the third definition given now refers to the deposition to the ground itself, with these surface elements viewed as an interruption.

There is no "right" or "wrong" definition, but there is definite need to recognize that different scientific communities have different working understandings of what is meant by "total deposition." There is no danger of confusion while communication is within specific communities. However, suppose that there is an attempt to refine regulations and policies on the basis of limnology and watershed-scale ecology. Then the critical "total deposition" that is likely to be relevant is that which corresponds to the third case discussed above -- the deposition, by all mechanisms, that arrives at a forest floor. If there are no measurements made at a specific location, then estimates can be produced by models, based on the computation of local air chemistry from assumptions about atmospheric dilution, reactions, and dispersion of emissions perhaps from far upwind. But these models consider "total deposition" to be something different; they contain no local surface-source component, they cannot address well the (sub-grid-scale) peculiarities of particular watersheds, and they do not consider the biological processes that affect the transfer and redistribution of pollutants after they leave the atmosphere and before they enter the soil ecosystem.

2. WET AND DRY DEPOSITION — SOME BASIC DIFFERENCES

The processes by which clouds remove pollutants from the atmosphere have been studied with considerable detail over the last forty years, with early attention mainly on the problems of particle removal, especially as they relate to the problem of radioactive fallout. The emphasis of studies over the last ten years has been more on gaseous pollutants. The studies have included consideration of the role of clouds as mechanisms for relocating pollutants throughout the troposphere, as well as on their part in the transformation and removal processes. Because of their recognized efficiency in removing pollutants from the atmosphere, convective storms have been a major focus for these studies. On an event basis, convective storms entrain large amounts of air, remove pollutants with comparative efficiency, transform these pollutants into other chemical species, and deposit the products in intense bursts over relatively small areas.

In contrast to convective systems, large scale synoptic precipitation systems scavenge pollutants from elevated layers of the atmosphere, and deposit them far more slowly, over longer time periods, and across larger areas. Different parts of each continent experience different proportions of these two mechanisms. For North America, for example, it is evident that the areas of the continent most affected by convective activity are in the southeastern U.S.A., and that the relative importance of severe storms decreases markedly to the north and the west. The part of the continent that receives the most acidic precipitation is an area where convective activity contributes substantially.

Not only does the precipitation process vary with geography, but so does the chemistry that affects wet deposition. Temperature plays an important role in the chemistry of clouds, regardless of whether they rain. As an obvious example, the rate of solution of even soluble gases into solid ice particles is much slower than for liquid droplets. Moreover, the effects on further absorption of dissolved gases such as sulfur dioxide is such that ice crystals quickly become inefficient receptors for transfer from the gas phase. Several repercussions are of practical relevance. First, high altitude cold clouds are likely to be less efficient scavengers of gaseous air pollutants than low altitude warmer clouds (even though solubility of gases increases as temperature decreases). Second, severe thunderstorms that have considerable activity at high altitudes may not be as efficient at removing atmospheric chemicals as the less intense convective cells that are confined to the lower levels of the atmosphere. Third, the temperature effects will be different for different chemical species, thus additionally complicating the patterns of deposition observed at the ground. In general, the deposition "footprint" left by a raining cloud is exceedingly complicated, and depends on the chemical species that is being measured.

The geographic and temporal variability of the wet deposition process is sufficiently great that there has been limited success in relating the observations made by any specific wet deposition collector to the chemical and meteorological characteristics of the surrounding atmosphere. Regardless of this difficulty, a major goal of precipitation chemistry research is to derive such a capability, for both event and long-term average situations. Most models represent ensemble average conditions, rather than any single precipitation event. In this regard, it is of special importance to note that deposition by precipitation is essentially an ergodic process in space. Thus, measurements made at any single location will be like those made at any neighboring location (but not, of course, over distances such that orographic effects and climatology become important), provided averages are constructed over a long enough time. The high spatial variability evident in event precipitation chemistry data is greatly reduced as longer time averages are constructed.

In contrast to wet deposition, dry deposition is not a spatially ergodic process. In other words, there is no guarantee that values obtained at a particular measurement site will be representative of a neighboring area, even if exposed to much the same air concentrations. Dry deposition is a product of several processes working in combination. First, concentrations of the pollutant in question must be present in the air to which surfaces are exposed. Second, the surfaces in question must be such that the material in the air can be transferred to them. This second factor is what causes dry deposition to be so markedly different from wet, since the nature of the surface itself influences (and sometimes controls) the efficiency of transfer from the air to the ground. In the case of wet deposition, the control is not as related to the surface (excepting, of course, the obvious effects of climatology and topography), but rather to processes within the cloud.

On the average, the chemistry of wet deposition does not differ greatly over small spatial scales (*i.e.*, over distances of tens of kilometers). Dry deposition varies from place to place more than wet deposition. The air chemistry affecting wet deposition is mainly that at cloud level, whereas the air chemistry affecting dry deposition is mainly that in air in contact with the surface. Since dry deposition is controlled by the detailed nature of the surface as well as by air concentrations, spatial detail will not be diminished by time averaging. In contrast, for wet deposition any specific event must be expected to have great spatial variability, but this detail will be lost as time averaging proceeds. Thus, the concept of routine monitoring of wet deposition is substantially different from that associated with routine monitoring of dry deposition. For wet deposition, a few locations can be used to represent regional values, which are then representative of the wet deposition of all areas near the stations where measurements are obtained. This is fundamentally not the case for dry deposition.

3. THE DRY TO WET RATIO

A direct repercussion of the lack of spatial ergodicity in dry deposition is that the concept of a "dry to wet ratio" is poorly founded. Even though it is possible to use data obtained at any particular location to define and quantify such a ratio, the value obtained may well be inappropriate even for a neighboring location. It should be noted, however, that spatial averaging serves to smooth much of the variability that characterizes local quantifications of the dry/wet deposition ratio, and hence the ratio approach may well have more validity as the target area increases in size.

Data on the relationship between dry and wet deposition can be derived from several US sources. Here, data obtained in the Atmospheric Integrated Research Monitoring Network (AIRMoN) program of NOAA will be used. In AIRMoN, wet deposition is derived from daily measurements at a small array of stations, supporting weekly measurements elsewhere. Dry deposition is derived using the Dry Deposition Inferential Model (see Hicks *et al.*, 1987) at AIRMoN-Dry sites, some of which make considerably more detailed measurements designed to benchmark the inferential methods. Tables of the resulting wet and dry deposition rates are presented elsewhere (e.g. Sisterson, 1990). Table 1 presents a summary of the results for sulfur species (wet deposited as sulfate and dry as the sum of deposits as sulfur dioxide and particulate sulfate) and for a subset of the nitrogen species (nitric acid vapor for the dry component, and nitrate for the wet; hence, ammonia and ammonium are not considered).

Figure 1 shows how the ratio of dry to wet deposition varies with the amount of rainfall, across the array of sites of the AIRMoN-Dry network. For purposes of clarity, winter months are excluded from these plots of seasonal data. It is clear that a strong dependence on the amount of precipitation exists. This is anticipated on first principles — clearly, dry deposition will dominate when there is little rainfall. The overall behavior appears identical for both chemical species; regression yields a slope such that the dry/wet ratio varies according to the inverse square root of the rainfall (exponent = -0.55 ± 0.04 for sulfur; -0.54 ± 0.05 for nitrogen).

Table 1. Seasonal ratios of dry to wet deposition of sulfur (S) and nitrate (N), for sites of the NOAA AIRMoN network. Contributions associated with ammonium are not included. Mean seasonal values are given. The means are computed geometrically, and the standard errors on these means ($\sigma(\pm)$) are quoted as percentages. WM — Whiteface Mountain, NY; WP — West Point, NY; SC — State College, PA; AR — Argonne, IL; BV — Bondville, IL; OR — Oak Ridge, TN; PA — Panola, GA; PG — Pawnee Grasslands, CO.

	Winter		Spring		Summer		Autumn	
	S	N	S	N	S	N	S	N
WM	0.66	0.36	0.33	0.54	0.19	0.41	0.33	0.52
WP	0.61	0.33	0.68	1.05	0.37	0.42	0.43	0.46
SC	0.95	0.73	0.37	0.66	0.50	0.53	0.59	0.79
AR	2.45	1.11	1.38	1.00	1.28	0.56	1.13	0.83
BV	1.49	3.03	0.80	1.41	0.67	0.62	0.57	0.92
PA	1.19	1.46	1.28	1.44	0.65	0.64	0.71	0.69
OR	1.00	0.63	1.39	1.14	0.89	0.41	1.12	0.67
PG	1.08	4.56	0.61	2.33	0.29	1.32	0.56	2.85
Means	1.08	1.02	0.75	1.09	0.52	0.57	0.63	0.81
$\sigma(\pm)$	16%	37%	21%	16%	23%	13%	15%	20%

The information presented here is not new, and is presently being updated with considerably better analyses for a much longer observational period (ten years of data will soon be available, versus the three years used to construct the present tables and figures). However, even the present limited data set demonstrates that the common assumption that dry deposition equals wet is flawed in a consistent fashion that will inject biases. The ratio of wet to dry deposition is a function of location, of rainfall amount, and of the chemical species in question. Using wet deposition as a basis for estimating dry is likely to be misleading unless a site has been calibrated for this purpose. As yet there is no alternative to on-site measurements if accurate dry deposition data are desired.

4. DRY DEPOSITION — EFFECTS OF SURFACE HETEROGENEITY

There is a further aspect that deserves attention — the consistent underestimation of dry deposition due to omission of edge effects in contemporary models. Consider an area A containing portions A_1 of grass and A_2 of forest. The forested area is composed of a number of n patches, that therefore have a characteristic length scale $Ls = (A_2/n)^{0.5}$ As a first approximation, we may consider the exchange of the area as a whole as the sum of exchanges for the two contributing components, so that

$$F = (A_1.F_1 + A_2.F_2)/(A_1 + A_2) \qquad (1)$$

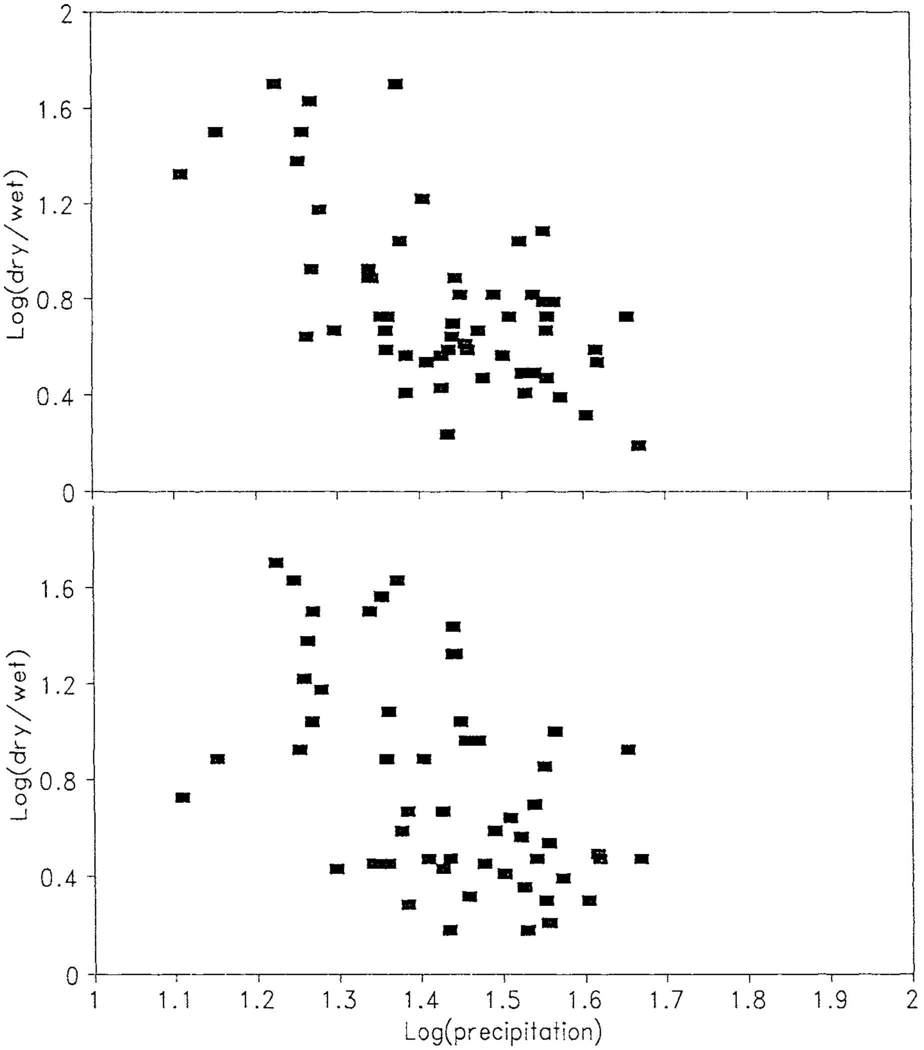

Figure 1. The relationship between the seasonal ratio of dry to wet deposition, for sulfur (upper) and nitrate-nitrogen (lower) stations of the NOAA AIRMoN array, in the eastern U.S.A.

In most numerical models, each of the individual fluxes F_1 and F_2 is approximated using a resistance model or a surface-specific deposition velocity approach. Land use characteristics are used to determine appropriate values of surface resistances, and of the factors that determine the aerodynamic resistance and/or deposition velocity. Consider, however, the effects of edges. Wind blowing across the grass penetrates into each stand of trees for a distance that scales according to the wind speed, the density of the vegetation, and the height of the trees (h). As this air passes through the canopy, there is interaction between it and the components of the foliage that involve a "filtration efficiency" ϵ. The edge-effect flux can then be written as

$$\delta F = \epsilon.n.h.Ls.u.(C_{02} - C_{a2})/(A_1 + A_2) \tag{2}$$

To a first order, we can consider the exchange by the horizontal edge-effect and filtration mechanism to be additive to the conventional vertically diffusive flux, and hence we can write the total exchange rate for the heterogeneous area as

$$F(new) = [(A_1.k_1 + A_2.k_2 + \epsilon.n.h.Ls.u)/(A_1 + A_2)].(C_{02} - C_{a2}) \tag{3}$$

where k_1 and k_2 are the appropriate total conductivities ($k_i = 1/(R_{ai} + R_{si})$, and where it has been assumed that concentrations in air and at the surface are constant across the area under consideration (a rather questionable assumption).

Additional insight can be derived from studies of throughfall under forest canopies, downwind of edges (e.g. see Beier, 1991), which indicate that deposition at the edge can be doubled, but falls off exponentially to the "infinite surface" value with an exponential distance constant that scales with the height of the trees. (Presumably, there will eventually be need to include a factor associated with canopy density and scavenging efficiency here, as indicated in the derivations above, but so far the experimental science does not permit this degree of sophistication. For further guidance, see Slinn, 1982, for the case of particles.) At distance x from a forest edge the flux Fx might be simplified as

$$Fx = F_2[1 + C.\exp(-x/(c.h))] \tag{4}$$

where the edge effect is generalized by introducing an "amplification factor" C. Integration of (4) and comparison with (3) leads to an areal flux amplification that depends on a quantity like $h\sqrt{n}$ — the higher the trees and the patchier the surface the greater the dry deposition.

5. AN EXAMPLE — THE CHESAPEAKE BAY

Several recent studies have emphasized that coastal ecosystems are vulnerable to effects of atmospheric pollution. The focus of much of the concern is nitrogen compounds, deposition of which adds to the input from rivers and groundwater and eventually leads to eutrophication. In general, the most vulnerable ecosystems are biologically productive shallow-water embayments, affected by and downwind of population centers. Most of the Atlantic coast of the U.S.A. is potentially vulnerable (see Fogel and Paerl, 1991, for example), potentially extending as far as Bermuda (see Owens *et al.*, 1992; Michaels *et al.*, 1993). Regulators are currently faced with a difficult problem — how to include air pollution in the complicated equations that underpin regulatory and control decisions.

Table 2 summarizes some contemporary assessments for the Chesapeake Bay, the large estuary serving the major population centers in the U.S. mid-Atlantic region. The values tabulated refer to assumptions about the retention of deposited nitrogen by the terrestrial surfaces through which the pollution must migrate on its way to the Bay. The land area is so large and the water body so shallow that a small error in the assumption about retention can have large consequences; as is seen, the assumptions vary considerably.

Table 2. Watershed retention values (in % of nitrogen loading) used in Bay loading studies to date (numbers in parenthesis indicate range tested for sensitivity studies).

Land Use	Tyler (1988)	Hinga *et al.*, (1991)	Fisher and Oppenheimer (1991)
Forest	95.2-100.0	80.0 (25.0-95.0)	80.0 (51.0-100.0)
Pasture	93.7-99.96	80.0 (25.0-95.0)	70.0 (51.0-90.0)
Cropland	76.0-99.97	60.0 (45.0-75.0)	70.0
Residential	62.0-95.3	25.0 (10.0-50.0)	35.0 (0.0-70.0)

In addition, every assessment assumes that dry deposition of nitrogen equals wet. One assessment omits consideration of ammonium. All assessments assume that deposition to the water itself equals that to the surrounding land. All of these assumptions are areas of potentially large error. In fact, it is not until the last year that even one dry and wet deposition monitoring station has been set up to test whether model predictions for the Chesapeake Bay watershed resemble reality. A recent workshop (see STAC, 1994) concluded that the major scientific need was for on-site data to verify model predictions.

Although much of the discussion about uncertainty given above relates to the matter of dry deposition, even the wet deposition component remains somewhat unclear. Although wet deposition is highly variable, the spatial distribution of wet deposition ought to have characteristics similar to those of precipitation. Experience has shown that if averaged over a sufficiently long time, the value obtained at a single location is a fairly good approximation of averages at surrounding locations. Average and representative values of wet deposition can be interpolated from isopleth maps derived from independent estimates of precipitation amounts and concentrations. Interpolation requires accounting for the consequences of correlations between the two estimates. Existing estimates appear to overlook the effects of this correlation, and so it is possible that current wet deposition data are slightly biased to overestimate deposition. Although the magnitude of the bias has not been determined, estimates of the bias range from 10% to 25% depending on location.

Estimates of wet deposition to the Chesapeake Bay surface range from 3.45 to 4.2 Gg NO_3^- N yr^{-1} (Fisher and Oppenhiemer, 1991; Hinga *et al.*, 1991; Tyler, 1988). These estimates omit consideration of the effects of the correlation mentioned above, and fail to take episodicity into account; in practice it may well eventuate that the key consideration is not the annual delivery of nitrogen to the catchment area but the number of severe inundations that cause atmospheric nitrogen to be wet-delivered in large quantities and which cause nutrients to be scavenged from the landscape as water floods over it.

6. CONCLUSIONS

There is accumulating evidence to demonstrate that reliance on a cardinal value for a "dry to wet deposition ratio" will be misleading. In some places, this will cause loadings to be overestimated; in others they will be underestimated. The patchiness of the surface has an influence that can be described in broad terms, but cannot yet be quantified in detail — the patchier the surface, the greater the underestimation of dry deposition.

If the goal is to protect a large area in which endangered ecosystems reside, then it is quite likely that contemporary knowledge will provide adequate guidance. However, if a particularly sensitive ecosystem is singled out, then particular attention should be paid to its specific circumstances. The spatial variability of dry deposition means that it cannot be isoplethed and interpolated like wet deposition, and hence interpolating loading estimates to a specific target area is potentially misleading. The likely biases that will result will not be unidirectional, so that it must be expected that control measures based on use of mapping and isopleths will not be wholly successful in protecting all sensitive regions.

The way in which the critical loads concept is applied in a regulatory environment risks asking atmospheric transport models to predict deposition of the kind measured by throughfall studies. This may well be an excellent goal for research of the future, but so far there has been no atmospheric transport and deposition model whose output has been tested in this context. In fact, few models have been tested for their predictions of deposition from the atmosphere. Most tests have focussed on the ability to simulate air chemistry, and some have addressed wet deposition. However, there has been no direct test of predictions of dry deposition. The nearest such test has been in North America, where dry deposition rates computed from model predictions of air chemistry have been shown to agree with estimates of dry deposition derived from field observations of air concentrations of the same chemicals. The sub-models used to derive the dry deposition rates in the two applications are essentially the same. In essence, this is little more than a test of air chemistry predictions.

Finally, an obvious question remains about what better can be done. There is no substitute for good on-site data. If the driving goals are related to the fate of pollutants emitted into the atmosphere, then large-scale models and statistically-derived arrays of deposition stations may provide good answers. However, if the intent is to help protect sensitive areas then monitoring should be at those areas, first emphasizing the locations that are best suited for high quality deposition measurements. This conclusion is emphasized by the demonstrations presented here that the smoothed outputs of contemporary models cannot be accurately interpolated to predict the deposition to small (i.e. greatly sub-grid-cell) ecosystems where there are no data. The practical importance of these conclusions will vary from one chemical species to another, in a manner that is not yet understood.

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