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Dry deposition of nitrate to a deciduous forest

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Abstract. Because dry-deposition inputs are difficult to measure, they are often ignored in biogeochemical studies. In this study three separate methods were used to estimate dry deposition of nitrate to a deciduous forest (Walker Branch Watershed) in eastern Tennessee. The range of estimates of dry-deposition flux was from 1.8 to 9.1 kg NO_3^--N ha^{-1} yr^{-1}. Using a hybrid approach that combines some aspects of all three methods, a best estimate of 4.8 kg NO_3^--N ha^{-1} yr^{-1} was derived. About 75% of this flux is attributable to deposition of HNO_3 vapor with large particles contributing most of the remainder; the contribution from small particles is negligible. The range of estimates obtained from the three techniques suggest that dry-deposition measurements should be interpreted with caution.

Our best estimate indicates that dry deposition of NO_3^- is the largest single form of inorganic nitrogen (N) deposition to this forest, contributing almost half of the 10.1 kg N ha^{-1} total annual input. All of the enhancement of NO_3^- deposition in stemflow and throughfall relative to incident precipitation can be explained by washoff of dry-deposited NO_3^-, and some canopy uptake of dry-deposited NO_3^- is suggested. This uptake occurs primarily during the growing season and contributes from 0.2 to 7.5 kg N ha^{-1} yr^{-1} to the N requirements of the ecosystem, with a best estimate of 3.2. Despite the uncertainties, the magnitude of the potential input fluxes to forested ecosystems necessitates consideration of nitrate dry deposition in ecosystem nitrogen cycling studies.

Introduction

Atmospheric deposition of nitrate is important both in ecosystem nitrogen cycling and as a component of the acidic deposition phenomenon. Several studies have shown wet deposition (i.e., deposition via precipitation) of nitrate to be a rapidly increasing form of acidic input (Galloway and Likens, 1981; Brimblecombe and Stedman, 1982; Horvath, 1983). In addition, Friedland et al. (1984) suggest that elevated N inputs may be linked to forest decline phenomena observed in the high elevation forests of the eastern U.S.

Nitrate exists in the atmosphere not only dissolved in droplets, but also in the form of particulate matter (such as NH_4NO_3) and as HNO_3 vapor. Other nitrogen oxides such as NO and NO_2 can be converted to NO_3^- in the atmosphere or after deposition to an ecosystem. Nutrient cycling studies frequently measure atmospheric deposition with open funnels ("bulk" collectors) that

collect only wet deposition and some unknown portion of the total dry deposition. These funnels underestimate total deposition because they collect most particles and vapors with an efficiency much less than that of a full canopy.

Methods for measuring dry deposition (direct deposition of particles and gases) are not well developed (Hicks, 1983; Hicks et al., 1980). For nitrogen oxides, deposition estimates have been made using micrometeorological techniques (Huebert, 1983; Wesely et al., 1982) and measurements of accumulation on natural and artificial surfaces (Dasch, 1983; Lindberg and Lovett, 1985; Sickles et al., 1983; Lovett and Lindberg, 1984). In this paper, we describe three methods for estimating dry deposition of NO_3^-, derive from these a range of possible values and a best estimate, and compare the range of estimates to other fluxes of inorganic N in the ecosystem.

Estimates of dry deposition

All of the measurements described below for estimating NO_3^- dry deposition were made during the period July 1981 to July 1983 at the Walker Branch Watershed in eastern Tennessee. The site of the measurements was a ridgetop deciduous forest dominated by chestnut oak (*Quercus prinus* L.), white oak (*Q. alba* L.), and several species of hickory (*Carya*). The climate is of the humid mesothermal type, with an average annual temperature of 14.3 °C and an average annual precipitation of 140 cm, falling almost entirely as rain. Winds are light and come predominantly from the southwest in the summer and northwest in the winter. The site is in a rural area but is within 30 km of three coal-burning power plants, a small city (Oak Ridge, population about 30,000), and a larger city (Knoxville, population about 175,000).

Method 1: *Atmospheric concentrations and deposition velocities*

The most common method for routine estimation of dry deposition is monitoring of atmospheric concentrations of the chemical species of interest and application of a deposition velocity (ratio of deposition flux to atmospheric concentration at some reference height) derived from limited on-site measurements or from the literature. During the two-year experimental period, we measured atmospheric NO_3^- concentrations for approximately 4000 h (distributed evenly throughout the four seasons) at a height of 19.8 m on a tower extending through the forest canopy. This height is 1–1.5 m above the average height of dominant trees at this site. Measurements were made using a filterpack consisting of an inert Nuclepore prefilter (0.4 μm pore size) for particles and a nylon backup filter for HNO_3 vapor. Ambient air was drawn through these filters at rates of 2-3 L min^{-1} for periods of several days to a week. Details of sampling and extraction procedures are given elsewhere (Lovett and Lindberg, 1984; Lindberg et al., 1984). Sampling artifacts are a potential problem with the filterpack sampling arrangement,

Table 1. Concentrations and deposition velocities used in computing dry deposition of NO_3^- by Method 1

	Season	Concentration ($\mu g\,N\,m^{-3}$)	Deposition velocity ($cm\,s^{-1}$)	Flux ($kg\,NO_3^-\text{-}N\,ha^{-1}$)
HNO_3 vapor	Growing	0.82	2.0	3.0
	Dormant	0.92	0.5	0.60
Particulate NO_3^-	Growing	0.07	0.3	0.04
	Dormant	0.25	0.1	0.03

especially transfer of NO_3^- from the prefilter to the nylon filter by volatilization of NH_4NO_3 particles (Appel et al., 1981). This artifact may cause an overestimate of HNO_3 vapor and a corresponding underestimate of particle NO_3^-. In addition, air filtration methods are known to undersample the large ($>5\,\mu m$) particles in the atmosphere (Wedding et al., 1977).

Table 1 shows the results of the air sampling, along with the deposition velocities used and the calculated fluxes. Deposition velocities for HNO_3 vapor have rarely been determined elsewhere. Huebert and Robert (1985) reported a deposition velocity of $2.5 \pm 0.9\,cm\,s^{-1}$ for HNO_3 vapor to a grassy field in Illinois, and Huebert (1983) suggested that comparable values are appropriate for our forested site in Tennessee. We choose a value of $2\,cm\,s^{-1}$ in our calculation for the growing season. Because HNO_3 vapor is expected to deposit to forests more efficiently than to grass (Fowler, 1980), this is probably a conservative estimate. For the dormant season, we assumed the dormant: growing season ratio of deposition velocities equaled the ratio of total surface area available for deposition in the two seasons (leaf and branch area in the growing season, branch and ground area in the dormant season). This surface area ratio is 0.25 (Hutchinson et al., in press), so our estimated dormant season deposition velocity is $0.5\,cm\,s^{-1}$. Appropriate deposition velocities for particles are a matter of current debate, with estimates ranging from 0.01 to $0.5\,cm\,s^{-1}$ (Hicks and Garland, 1983). We chose values of 0.1 $cm\,s^{-1}$ for the dormant season and $0.3\,cm\,s^{-1}$ for the growing season as representative of the range of values reported in the literature. These values are intended to be averages across the range of ambient particle sizes. The total annual flux estimated from this method is $3.6\,kg\,NO_3^-\text{-}N\,ha^{-1}\,yr^{-1}$, of which about 98% is attributed to HNO_3 vapor. Uncertainties in this estimate will be discussed further below.

Method 2: *Artificial surfaces*

We performed 7 determinations of the flux of NO_3^- to 9-cm-diam. Whatman 42 filter papers which were supported by narrow, polycarbonate, cruciform frames as shown in Fig. 1. The filters were deployed on a tower within the canopy for largely rain-free periods of several days at a time. At the end of an exposure period, the filters were carefully removed from the holders using

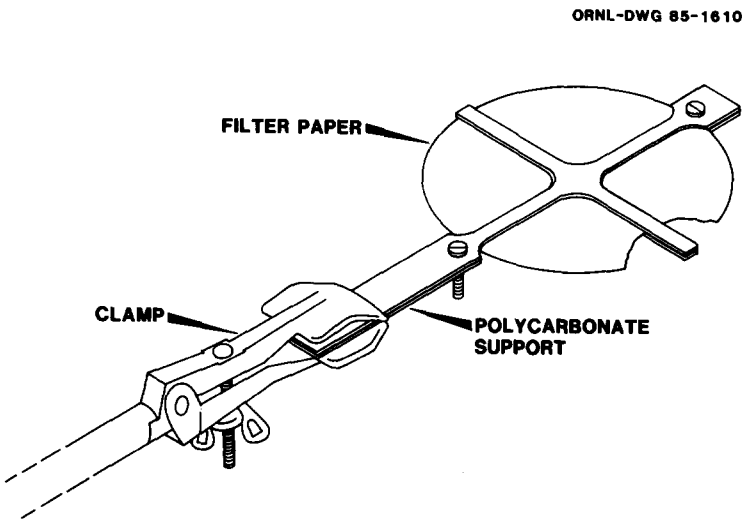

Figure 1. Diagram of apparatus used for field exposures of Whatman filters. Filter paper is 9 cm diam.; support structure is clear polycarbonate < 1 mm thick. Width of support arms is about 3 mm. A portion of the filter is cut away in the diagram to show top and bottom support arms.

plastic gloves and forceps, sealed in a plastic bottle, and returned to the laboratory. Extraction of NO_3^- was accomplished by adding 50 mL of distilled water to the bottle, vigorously shaking the bottle and filter for 2 h, then centrifuging to remove the shreds of paper. The extraction solution was analyzed using an ion chromatography. A total to 661 h of sampling were performed, distributed throughout the seasons. The filters were protected from unpredicted rain by automatic covers (Lindberg and Lovett, 1985). These Whatman filters should collect both deposited particle NO_3^- and absorbed HNO_3 vapor, and thus give an estimate of total NO_3^- dry deposition. It is often assumed that artificial surfaces are poor surrogates for assessing vapor deposition because they lack the stomatal control exerted by plant leaves. However, HNO_3 is an extremely reactive vapor and may deposit so readily to external plant surfaces that stomatal control is unimportant. Hicks and Garland (1983) and Huebert and Robert (1985) indicate that the surface resistance to HNO_3 vapor deposition is essentially zero, thus the deposition is under aerodynamic rather than surface control. In this situation, an absorbing surface exposed to in-canopy aerodynamic conditions could provide an adequate surrogate for leaves. For this study, samples taken at 8.8, 12.8, and 18.3 m were averaged to give a mean flux within the canopy. The mean flux was computed per unit surface area of the filters and multiplied by the total surface area available for deposition to estimate the flux to the forest (Table 2). Estimated NO_3^- deposition (in kg NO_3^--N ha^{-1} yr^{-1}) using this method was 6.6 for the growing season and 2.5 for the dormant season, for a total of 9.1.

Table 2. Calculation of total NO_3^- fluxes to the forest from artificial surface measurements (standard errors in parentheses)

	Season	
	Growing	Dormant
Flux to filter papers[a] ($\mu g\, NO_3^-\text{-}N\, m^{-2}\, h^{-1}$)	11 (1.3)	24 (2.2)
Days per season	213	152
Plant area index[b]	11.7	2.9
Deposition to forest ($kg\, NO_3^-\text{-}N\, ha^{-1}$)	6.6	2.5
Flux to inert plates	1.2 (0.40)	3.0 (0.32)

[a]Based on two-sided area of filters
[b]Total surface area, calculated from Hutchison et al. (in press). Dormant season value includes ground surface

Deposition to similarly exposed polycarbonate petri dishes, which should collect particles but not vapors (Spicer, 1979), indicated that particles could account for about 12% of the flux to the filter papers (Table 2). However, this may be an overestimate because the rimmed petri dishes may retain deposited particles more efficiently than nonrimmed surfaces like filter paper or leaves (Davidson et al. 1985). It should be noted that NO and NO_2 could deposit to these filters and then be converted to NO_3^-. Deposition of NO_2 is definitely controlled by stomatal opening (Grennfelt et al., 1983), so these artificial surfaces cannot be used to quantify its deposition.

We note in Table 2 that deposition to both filters and petri dishes is lower in the growing season than in the dormant season. This indicates the effectiveness of the foliage in removing both gaseous and particulate NO_3^- from the air, and suggests that branch surfaces are exposed to higher concentrations of atmospheric NO_3^- in the winter than in the summer.

Method 3: *Statistical analysis of throughfall*

Lovett and Lindberg (1984) described a method for determining dry-deposition rates by regression analysis of net deposition (deposition in throughfall minus deposition in rain). In this technique, net deposition for individual storm events is regressed against the length of the antecedent period (the dry period preceding the event) and the amount of rainfall in the storm. Because the dry-deposition flux is correlated with the length of the antecedent period, and the internal leaching is correlated with the amount of precipitation, this technique can separate these two components of net deposition. The coefficient of the antecedent period term in this regression is a full-canopy dry-deposition rate, which can be multiplied by the total non-raining hours in the year to calculate an annual dry-deposition flux. In the data set used for this study, the dry-deposition coefficients were significant at the $p < 0.01$ level (Lovett and Lindberg, 1984). However, there are several

caveats to this approach, the most important of which in this case is that the canopy may irreversibly absorb dry-deposited nitrate. This absorption renders the deposited NO_3^- unavailable to washoff by subsequent rain events, and results in an underestimate of the dry-deposition rate. Using this technique for the data collected at our experimental site resulted in dry-depositon estimates (again in $kg\,NO_3^-\text{-}N\,ha^{-1}\,yr^{-1}$) of 0.90 for the growing season and 0.88 for the dormant season, for a total of 1.8.

Uncertainties and a best estimate

The estimates from the three methods are summarized in Table 3 and range from 1.8 to 9.1 $kg\,NO_3^-\text{-}N\,ha^{-1}\,yr^{-1}$. All three estimates have uncertainties and biases, some of which we can identify. Method 1 has considerable uncertainty associated with the deposition velocities. As described above, standard air-filtration methods are known to undersample large particles. In addition, the artifact of particle volatilization from the prefilter would result in an overestimate of HNO_3 vapor concentrations and an additional underestimate of particle concentrations. Because of the relative magnitude of the deposition velocities, this would lead to an overestimate of the total dry deposition. Appel et al. (1981) indicate that up to 50% of the particles can be lost from the prefilter by volatilization. If this worst case were true, the data in Table 1 indicate that the result would be overestimates of about 20% for HNO_3 vapor deposition and about 13% for total NO_3^- dry deposition. However, Kelly et al. (1984) showed this artifact to be insignificant in their sampling of atmospheric nitrate at Whiteface Mt., New York.

The results of Method 2 will be biased if the filter papers absorb HNO_3 vapor differently than do plant surfaces. Because HNO_3 is very water soluble, its deposition characteristics may be strongly influenced by hydration of the receiving surface. We expect that the filter papers would hydrate more readily than would the wax-coated plant surfaces during the episodes of high humidity that are common in eastern Tennessee. This may result in a systematically high estimate of NO_3^- deposition by Method 2. However, we could find no significant correlation between nitrate deposition to these surfaces and the average relative humidity during the corresponding sample periods. Another possible bias in this method is that the artificial surfaces, which were located within the canopy, may have received deposition of insect frass and other debris not representing a true atmospheric input.

There is good reason to believe the throughfall estimate (Method 3) is low because of canopy absorption of deposited NO_3^-. Canopies are known to take up NO_3^- from incident precipitation (Verry and Timmons, 1977; Olson et al., 1981; Lovett and Lindberg, 1984), so it seems reasonable to expect uptake of dry-deposited NO_3^- as well. Note in Table 3 that the growing season and dormant season deposition estimates from Method 3 are very similar, despite the significant differences in canopy surface area in the two periods. This could indicate a very active uptake of NO_3^- by the canopy in the growing

Table 3. Summary of estimates of NO_3^- dry deposition (all data in kg NO_3^--N ha^{-1} yr^{-1})

Method	Growing season	Dormant season	Annual total
1. Atmospheric concentrations and deposition velocities	3.0	0.63	3.6
2. Artificial surfaces	6.6	2.5	9.1
3. Statistical analysis of throughfall	0.90	0.88	1.8
4. Best estimate (hybrid approach)			
HNO_3 vapor	3.0	0.60	3.6
small particles	0.01	0.004	0.01
large particles	0.79	0.38	1.2
Total	3.8	0.98	4.8

season, as would be expected. In addition to this bias, there is a statistical uncertainty associated with the deposition rates estimated from the regressions. The standard errors of the deposition rates were 10 and 21% of the mean values for the dormant season and growing season, respectively. However, the regression statistics showed these rates to be highly significant, as mentioned above.

To avoid some of these problems, we developed an alternative, hybrid approach that uses some aspects of the previous three methods. For HNO_3 vapor, our best estimate of deposition is the concentration and deposition velocity technique discussed in Method 1. However, a more accurate estimate of particle deposition can be made by considering large and small particles independently. For small particles, the concentration and deposition velocity approach is practical. Using an Andersen cascade impactor, we determined the fraction of particulate atmospheric NO_3^- associated with $< 2\,\mu m$ particles at this site to be 20% in both the growing and dormant seasons, based on seven measurements of several days' duration each. This is consistent with small particle fractions reported for other areas in the Eastern U.S. by Wolff (1984). We estimated deposition velocities for the small particles to be 0.05 $cm\,s^{-1}$ for the dormant season and 0.2 $cm\,s^{-1}$ for the growing season (Hicks, 1984; Fowler, 1980; Wesely et al., 1983).

Several recent papers (Davidson et al., 1983; Lindberg and Lovett, 1985; Hicks and Garland, 1983) indicate that large particles, though difficult to sample and numerically relatively rare in the atmosphere, can contribute a disproportionately large share of the deposition of some elements. This occurs because large particles can be efficiently deposited by inertial impaction and gravitational sedimentation, while small particles cannot. The micrometeorological methods commonly used to measure deposition velocities do not accurately measure the deposition of large particles, so using the concentration/deposition velocity approach is difficult (Hicks and Garland, 1983). An alternative means to assess deposition of large particles is direct collection on artificial surfaces. Use of surfaces positioned above the canopy minimizes

contamination by frass and debris liberated within the canopy, but presents the problem of scaling the measurements to the full canopy.

We approached this scaling problem by using airbourne calcium (Ca), which exists mainly on large particles at this site (Lindberg et al., 1984), as a "tracer" of large-particle behaviour. By comparing the dry-deposition flux of Ca to the full canopy, as evaluated by the throughfall technique (Method 3, Lovett and Lindberg, 1984), with the flux measured by above-canopy deposition plates, we calculated large-particle scaling factors of 2.7 in the growing season and 1 in the dormant season. These factors represent the enhancement of large-particle deposition that can be expected from the canopy relative to a deposition plate positioned above it. The canopy does not absorb dry-deposited Ca; therefore the estimate of dry deposition from the throughfall technique is not likely to be an underestimate, as it is for NO_3^-. The canopy does release a small amount of Ca by foliar leaching during rain events, but this is explicitly accounted for in the regression analysis (Lovett and Lindberg, 1984), and the dry-deposition estimate used to compute these scaling factors does not include the leaching component. While we expect these scaling factors to be generic to large particles at this site, they may be particularly appropriate for NO_3^-, which has, like Ca, been shown to be associated with soil dust in the atmosphere (Wolff, 1984). This association probably results from the absorption of HNO_3 vapor by airborne, alkaline soil particles.

We multiplied the measured nitrate deposition to the above-canopy plates by the scaling factors to calculate the flux of large-particle nitrate to this ecosystem. This flux is shown in Table 3 along with the estimated fluxes of gaseous and small-particle nitrate.

Total annual dry deposition as estimated by this hybrid approach is 4.8 kg NO_3^--N ha^{-1} yr^{-1}. This is our best estimate of nitrate-dry depositon to this forest. Deposition of HNO_3 vapor contributes about 75% of this flux, with the remainder due mainly to large particles. Small particles contribute less than 1% of the total flux.

Significant uncertainties remain in the deposition velocities and scaling factors, but the biases which we were aware of in the other techniques probably do not apply, with the exception of the possible sampling artifact in collection of HNO_3 vapor.

Discussion

For comparison to these dry-depositional estimates, precipitation delivers an average of 2.8 kg NO_3^--N ha^{-1} yr^{-1} to the canopy at this site (Lovett and Lindberg, 1984), while throughfall and stemflow deliver 4.4 kg NO_3^--N ha^{-1} yr^{-1} to the forest floor (Lindberg et al., 1984). Wet and dry deposition of NH_4^+-N are 1.7 and 0.8 kg ha^{-1} yr^{-1}, respectively (Lindberg et al., 1984). Dry deposition of NO and NO_2 may also contribute to N inputs. Kelly et al. (in

press) report deposition of these gases totalling 2.4 kg N ha^{-1} yr^{-1}, but their measurements of the gas concentrations were probably influenced by HNO_3 vapor, which would lead to an overestimate of the NO and NO_2 deposition. Dry deposition of NH_3 has not been measured at this site, but is expected to be insignificant (Tjepkema et al., 1981). Our best estimate of total inorganic N deposition is 10.1 kg ha^{-1} yr^{-1} for this ecosystem. This may be low if dry deposition of NH_3, NO, or NO_2 is important, but is nonetheless more than twice the wet-deposition input.

Two points are quite clear from these data. First, dry deposition is an important, and probably the dominant, form of NO_3^- deposition to this ecosystem. Our best estimate suggests that dry deposition supplies about 60% of the atmospheric input of NO_3^-, with the other estimates ranging from 40% (throughfall method) to over 70% (artificial surfaces). Certainly, ignoring dry deposition of NO_3^- would result in a serious underestimate of N inputs to this ecosystem.

Comparative data are not available for topographic positions other than the ridgetop location of this study. Because deposition of HNO_3 vapor should respond to aerodynamic factors such as windspeed, we suspect that more sheltered slope positions may receive less dry deposition of NO_3^-. Our measured concentrations of NO_3^- in the air are near those cited as typical for nonurban areas in eastern North America (EPA, 1984; Kelly et al., 1984), so we feel that these results may be generally applicable to other geographic areas. A case in point is the Hubbard Brook Experimental Forest in central New Hampshire, where the N balance (= bulk precipitation inputs − ecosystem storage − streamwater outputs) shows a deficit of 14 kg ha^{-1} (Likens et al., 1977). This imbalance could be partially rectified by considering dry deposition of NO_3^- (Lovett, 1983). Using the simplifying assumption that no canopy leaching or uptake of inorganic N occurred, the throughfall data of Eaton et al. (1973) suggest a dry deposition of 6.6 kg N ha^{-1} for the 1969 growing season at Hubbard Brook (Lovett, 1983). This would be an overestimate if canopy leaching occurred, or an underestimate if canopy uptake occurred, as discussed below.

The second key point to emerge from our own data concerns the behavior of NO_3^- in the canopy. The wet-deposition and throughfall-plus-stemflow fluxes cited above indicate a net release of 1.6 kg NO_3^--N ha^{-1} yr^{-1} from the canopy. This would usually be interpreted as NO_3^- leaching from the canopy; however our results show that regardless of which estimation method is used, dry deposition can account for all of the net release. The difference between total (wet plus dry) atmospheric deposition and stemflow-plus-throughfall flux indicates that the canopy absorbs 3.1 kg NO_3^--N ha^{-1} during the growing season and 0.1 kg NO_3^--N ha^{-1} during the dormant season. On an annual basis, the behavior of the canopy is uptake, not release, of deposited N if dry deposition is considered. Our best estimate of the net amount of canopy uptake is 3.2 kg NO_3^--N ha^{-1} yr^{-1}, with the estimates ranging between

0.2 and 7.5 kg NO_3^--N ha^{-1} yr^{-1}, depending on which dry-deposition estimate is used.

Canopy uptake of deposited nitrate appears to be a general phenomenon in forest ecosystems. In the large data set assembled by Parker (1983), canopies which do not show uptake, i.e., which show greater fluxes of NO_3^- in the throughfall than in rainfall, are generally in areas of air pollution (e.g., McColl and Bush, 1978; Lemee, 1974), probably resulting in elevated levels of dry deposition. If the dry deposition to these forests were measured, it may account for the apparent net release as it does in the present study. The coniferous canopies studied by Olson et al. (1981) showed no apparent net release of NO_3^- despite copious non-rain inputs, probably because of the large population of epiphytic lichens which are vigourous NO_3^- absorbers (Lang et al., 1976).

The fate of this absorbed nitrate is unclear. Assuming nitrate reductase to be present or inducible in the canopy, the NO_3^- may be immediately assimilated by the plant cells. Another possibility is incorporation into the biomass of the microbes of the phyllosphere, with possible later release of the N in dissolved or particulate organic matter. Release of organic N from plant canopies has often been noted (e.g., Carlisle et al., 1967; Olson et al., 1981).

In conclusion, we note that the state of measurement of dry deposition is such that all estimates must be treated with caution. Use of multiple methods can provide a range of possible values, but understanding the limitations of each method is crucial for choosing a best estimate. Hybrid approaches such as the one used in this study probably provide the best opportunity for reducing inherent uncertainties.

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