

DRY AND WET ATMOSPHERIC DEPOSITION OF NITROGEN, PHOSPHORUS AND SILICON IN AN AGRICULTURAL REGION

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Abstract. We measured atmospheric nutrient deposition as wet deposition and dry deposition to dry and wet surfaces. Our analyses offer estimates of atmospheric transport of nitrogen (N), phosphorus (P) and silicon (Si) in an agricultural region. Annual dry and wet deposition ($\text{ha}^{-1} \text{ year}^{-1}$) was 0.3 kg of P, 7.7 kg of N, and 6.1 kg of Si; lower than or similar to values seen in other landscapes. N:P and Si:N imply that atmospheric deposition enhances P and Si limitation. Most P and soluble reactive P (SRP) deposition occurred as dryfall and most dry-deposited P was SRP so would be more readily assimilable by plant life than rainfall P. Dry deposition of N to wet surfaces was several times greater than to dry surfaces, suggesting that ammonia (NH_x) gas absorption by water associated with wet surfaces is an important N transport mechanism. Deposition of all nutrients peaked when agricultural planting and fertilization were active; ratios of NH_x :nitrate (NO_x) reflected the predominant use of NH_x fertilizer. Wet deposition estimates were consistent over hundreds of km, but dry deposition estimates were influenced by animal confinements and construction. Precipitation wash-out of atmospheric nutrients was substantial but larger rain events yielded higher rates of wet deposition. Methodological results showed that local dust contaminated wet deposition more than dry; insects, bird droppings and leaves may have biased past deposition estimates; and estimating dry deposition to dry plastic buckets may underestimate annual deposition of N, especially NH_x .

Keywords: atmospheric deposition, dry, nitrogen, phosphorus, precipitation, silicon

1. Introduction

Atmospheric deposition is among the least understood pathways of nutrient transport. Wet deposition occurs through rain and snowfall, while dry atmospheric deposition arises from gaseous and particulate transport from the air to the surfaces of aquatic and terrestrial landscapes. Wind, burning, planting and tillage can cause nitrogen- (N) and phosphorus- (P) bearing particles to become airborne. Nitrate/nitrite (NO_x) and ammonia (NH_x) can also enter the atmosphere as gases. Particulates and gases are then deposited by fallout or washed out of the atmosphere by precipitation. Atmospheric N and P may be important nutrient sources where land disturbances through agricultural activity are extreme (Ahn and James, 2001; Asman, 2002; Asman *et al.*, 1998; Pearson and Stewart, 1993; Winter *et al.*, 2002). The Midwestern U.S. has a long history of intensive agricultural activity,

so the characterization of atmospheric nutrient transport through space and time is important to an understanding of agricultural contributions of nutrients.

Atmospheric N transport has long been studied but P has been considered a minor constituent in rainfall and dryfall studies (Graham and Duce, 1979; NADP, 2004; Peters and Reese, 1995; Tabatabai *et al.*, 1981; Tabatabai and Laffen, 1976) so has been ignored in many studies of atmospheric nutrient deposition. For many ecosystems (e.g., lakes and reservoirs), P is a principal limiting nutrient. Past records of total P (TP) in wet precipitation from several locations in the U.S. suggest concentrations ranging from 5 to 19 $\mu\text{g L}^{-1}$, with deposition ranging from 0.01 to 0.1 $\text{kg ha}^{-2} \text{ year}^{-1}$ (Tabatabai, 1983). Although these levels of P have been considered insignificant when compared to other sources of P, such as agricultural runoff, recent studies suggest that atmospheric deposition can be a significant source (Chen and Fontaine, 1997; Guerzoni *et al.*, 1999; Jassby *et al.*, 1994; Peters and Reese, 1995; Shaw *et al.*, 1989; Winter *et al.*, 2002).

Although data on wet deposition are emerging, dry deposition can apparently contribute equal or greater amounts of nutrients (Ahn and James, 2001; Asman *et al.*, 1998; Caiazza *et al.*, 1978; Eisenreich *et al.*, 1977; Guerzoni *et al.*, 1999; Newman, 1995; Scheider *et al.*, 1979; Shaw *et al.*, 1989; Tarney *et al.*, 2001). Although many recent studies have estimated dry deposition using buckets and other passive samplers (Ahn, 1999; Cole *et al.*, 1990; Jassby *et al.*, 1994; Peters and Reese, 1995; Pollman *et al.*, 2002; Tamatamah *et al.*, 2005), techniques for measuring dry deposition of nutrients are not as well established. They are also hampered somewhat by the high variability observed in deposition to different types of surfaces, as well as the effects of variable turbulence and surficial properties of such samplers. Since dry deposition may be an important means of nutrient transport to water bodies and moist landscapes, dry deposition to wet surfaces may be more relevant than dry deposition to dry surfaces (the usual collection method) (Cole *et al.*, 1990; Gomolka, 1975; NADP, 2004; Peters and Reese, 1995). Boundary-layer resistance governs depositional velocities, but a dry surface imposes an artificially high aerodynamic resistance that could lower deposition rates (Jassby *et al.*, 1994). Therefore, estimations of dry deposition to wet surfaces might better mimic transport to waterbodies and moist landscapes.

Studies of dry deposition have some methodological unknowns. Comparisons of TP deposition to lake shorelines and at increasing distances from the shore on the lake surface show greater deposition near the shoreline (Cole *et al.*, 1990; Gomolka, 1975). This indicates the immediate environment of deposition samplers can influence apparent deposition estimates. Contamination of deposition samples by vegetation and insects has been found to be a frequent problem (Ahn and James, 2001; Cole *et al.*, 1990; Graham and Duce, 1979; Peters and Reese, 1995), although some contaminants might reasonably be considered local deposition. Some have recommended that samplers be located at heights greater than the standard ~ 2 m to minimize contamination by birds, insects, and vegetation (SFWMD, 1997) and thus distinguish locally recycled material from net system inputs.

Spatial and temporal variations may lead to uncertainties about the significance of atmospheric deposition as a nutrient source. Atmospheric deposition of N varies in space, as is shown by maps produced by the National Atmospheric Deposition Program (NADP) (NADP, 2004). The resolution of such maps is quite crude; i.e., there are only two monitoring sites in the state of Iowa, one of the most agricultural regions in the world. Atmospheric deposition maps of Denmark (Asman *et al.*, 1998) show great spatial variation in total N (TN) deposition attributed to dry deposition of NH_3 deposited close to the source. Nutrient deposition also varies in time (Linsey *et al.*, 1986; Shaw *et al.*, 1989). In studies in central Alberta, Canada, P deposition was greatest in May, and decreased throughout the summer. This pattern was attributed to agricultural processes, which disturb the soil and release P to the air.

1.1. PROBLEM AND SIGNIFICANCE

To date, estimates of atmospheric nutrient transport have been deficient in several ways. First, although dry deposition is measured less often than wet deposition, it may contribute more nutrients than wet deposition. Therefore, studies of wet deposition alone, may not provide an accurate characterization of total nutrient deposition. Second, dry deposition to wet surfaces may be more relevant to aquatic studies or studies of moist landscapes, because wet surfaces may capture more nutrients than dry surfaces. Third, there has been little recent characterization of atmospheric P deposition in highly agricultural regions. In addition, few have monitored Si deposition, in spite of its important role as a plant nutrient. More complete knowledge of atmospheric nutrient transport is needed if the role of atmospheric deposition in anthropogenic nutrient enrichment is to be understood.

1.2. OBJECTIVES

Our goal was to examine several methods of measuring nutrient deposition, and to estimate annual loading of N, P, and Si in a manner relevant to understanding their impact on freshwater ecosystems and moist landscapes. Our specific objectives were: (1) to determine the relative contributions of dry deposition to a dry surface (DD), dry deposition to a wet surface (DW), and wet deposition (W) and to determine their roles in the sum of measured wet and dry deposition; (2) to determine the relative importance of the soluble inorganic constituents of P and N (i.e., SRP , $\text{NO}_x\text{-N}$, $\text{NH}_x\text{-N}$); (3) to determine differences of short- versus long-distance transport of nutrients; (4) to characterize the spatial and temporal patterns of both wet and dry atmospheric nutrient deposition across a highly agricultural landscape; and (5) to calculate the annual nutrient deposition and the average nutrient concentrations in rainfall.

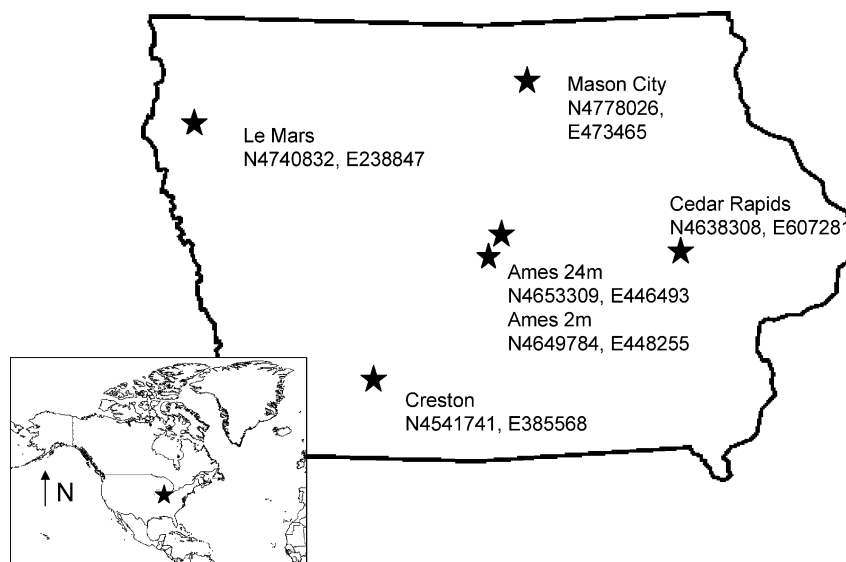


Figure 1. Map of Iowa, USA, showing sampling locations and geographic coordinates (UTM Zone 15 meters NAD83).

2. Materials and Methods

The overall objective of this study was to characterize wet and dry nutrient deposition in Iowa, which, with 91.3% of land in farms (ISU, 2004), is one of the most highly agricultural regions of the world (Arbuckle and Downing, 2001). The study included 12 automated samplers at 6 sites across the region (Figure 1). We collected samples every 7 days to be consistent with the NADP (2004). We collected nutrients as wet deposition (W), dry deposition to dry surfaces (DD), and dry deposition to wet surfaces (DW). The constituents of interest were soluble reactive phosphorus (SRP), total phosphorus (TP), nitrogen as ammonia + ammonium ($\text{NH}_x\text{-N}$), nitrogen as nitrate + nitrite ($\text{NO}_x\text{-N}$), total nitrogen (TN), and soluble reactive silicon as $\text{SiO}_2\text{-Si}$ (Si).

2.1. SAMPLING NETWORK

To monitor an annual cycle of atmospheric deposition, in December of 2002, we installed an automated sampler on the roof of a building on the Iowa State University campus (24 ± 1 m above the ground). This sampler was meant to sample ambient deposition, uninfluenced by local transport of dust and material from the ground. In the spring of 2002, we established a network of sampling stations across the region to characterize depositional patterns (Figure 1). This consisted of 12 automated samplers (LODA electronics, Loda IL), as used by the

NADP. Polyethylene buckets measured 0.0615 m² at the bucket opening, and were 35 cm high. Two automated samplers were deployed at each of six locations, five of which were situated at airports to facilitate rapid sample transport, and the sixth set was deployed on the roof of the same building as above at Iowa State University. Detailed descriptions of each site are given elsewhere (Anderson, 2004). Locations were evenly distributed across the state (Figure 1) to reflect the northwest-to-southeast precipitation gradient (SCAS, 2000). Sampler placement at each location followed the protocols established by the NADP (2004) except that the roof-top site (Ames 24m) was 24 m from the ground to avoid collecting dust and detritus. The Ames 24 m and Ames 2 m sites were closely spaced (3 km) to determine how local transport of particles (i.e., windblown dust) contributes to estimates of atmospheric deposition.

The automated samplers each consisted of two buckets on an elevated table with a moveable cover. Movement of the cover was initiated by a moisture sensor. During dry periods, the cover shielded the wet-deposition bucket, and when activated by rain or snow, the cover shielded the dry deposition bucket and exposed the wet deposition bucket. The sensor was heated to dry it following a precipitation event, to return the cover to the wet deposition bucket.

2.2. SAMPLE COLLECTION

Samples were collected at the Ames 24m site from January 28, 2003, to January 5, 2004, and June 17, 2003, to January 5, 2004, at the Ames 2m site. We collected samples at all locations from July 1 to September 30, 2003. Samples were collected approximately weekly from each location. Buckets were replaced with clean ones, and to determine DW and DD, one dry-deposition bucket at each site was pre-loaded with 3 L of distilled, deionized water, while the other dry-deposition bucket was left dry. The 3 L volume of water was chosen for the DW collector as a compromise between simulating a water surface in the context of this standard sampling device, while offering low evaporative loss and a sample volume small enough to yield detectable concentrations and deposition rates.

2.3. SAMPLE PROCESSING AND ANALYSIS

The tubs, funnels, bottles, buckets, and all equipment in contact with the samples were cleaned with phosphorus-free detergent, acid-washed with 10% HCl solution, and kept in plastic bags until they were used for sample collection. Each batch of distilled, deionized water added to samplers was analyzed along with the deposition samples and any detectable nutrients were subtracted from deposition estimates.

As noted by others (e.g., Cole *et al.*, 1990) sample contamination with foreign matter was frequent but all contaminated samples were removed from analysis. Contaminated samples were determined as: overlapped samples (DD buckets that collected some rain water), bulk deposition samples (wet + dry deposition, due to

sampler malfunction), samples with contaminants (dead insects, bird droppings, obvious algal growth) and samples that were collected longer than a week. We used only data from non-contaminated samples for all deposition calculations and concentration data.

In the lab, we added 1 L of distilled, deionized water to the DD buckets. Small brushes were used to scrub the sides of the dry deposition buckets, and the water swirled to ensure that the contents and particles were removed. If there was no precipitation, then W buckets were processed like DD buckets. We used all W bucket data in calculations of deposition rates, even if there was no precipitation, to account for small rain events that may have evaporated. If needed, sample volume was brought up to 1 L to yield sufficient analytical volume. DW buckets were processed like DD buckets, but without water addition.

NO_x-N and TN were analyzed using second derivative spectroscopy (Crumpton *et al.*, 1992), and Si using the silicomolybdate/heteropoly blue method (APHA, 1998). Analysis for NH_x-N (Nessler method), and both TP and SRP (ascorbic acid method, with persulfate digestion) were performed according to Standard Methods (APHA, 1998). All concentrations and deposition values were expressed as nutrient mass (e.g., NO_x-N, NH_x-N).

2.4. CALCULATIONS AND STATISTICAL METHODS

Atmospheric nutrient loading rates ($\mu\text{g m}^{-2} \text{d}^{-1}$) were calculated from the concentration of nutrient in the sample ($\mu\text{g L}^{-1}$) multiplied by the volume of sample (L) to yield μg of nutrient deposited. Deposition rates were adjusted to unit area and time by dividing by the surface area of the sampler (m^2) and the time the sample represented (d).

Comparisons between types of deposition measures (DD, DW, and W) for all analytes, and between the Ames 2m and 24m sites, were made using the Wilcoxon signed-rank test (Ramsey and Schafer, 2002). Data from all sites from July to September 2003 were combined for each deposition estimate. Ratios of DD:DW, DD:W, and DW:W were calculated. We fitted a first-order linear model with TP versus SRP, and TN versus NO_x + NH_x-N, to determine the dissolved inorganic proportion of TP and TN, respectively. Data from all sites and dates in 2003 were used for these plots.

Weekly samples from all sites (except Ames 24m and Ames 2m) did not always cover identical time periods, due to the time needed to fly to four of the sites to pick up samples. The majority of each time period for all sites overlapped, however, so we used these data for analyses of site differences. Site comparisons for DD, DW, and W, for all analytes, were made using PROC MIXED in SAS[®] software, Version 9.1 of the SAS System for the PC.¹ Site and date were included as class variables. The error used to test these effects was the residual error, which in this case was the interaction between site and date. The errors on all observations were treated as independent, because correlation of errors for the repeated measurements on the same

site would have been confounded with the site effect. If differences were detected, we used the Tukey-Kramer adjusted p -values (2-sided) for comparing the means of each of the pairs of sites for each analyte and deposition type. We back-calculated the means of log-transformed data to estimate geometric means for all sites, for W, DW, and DD. Geometric means and 95% confidence intervals of concentrations in precipitation were also calculated. Correlations of deposition among sites and dates were calculated to determine the degree of temporal synchrony in deposition rates.

Annual deposition from the Ames 24m site was calculated by summing W and DD weekly deposition estimates. We had a full year of data for DD and W, but only six months of DW data, which would be more relevant than DD as an estimate of annual dry deposition to a moist surface. To remedy this, we used ratios of DD:DW to extrapolate DD values to DW for all analytes. DW was not measured because, for the first six months in 2003, we only had one sampler. All annual estimates are missing three weeks of data in January 2003 due to a power failure to the automated sampler. Dry deposition during the winter, however, was low, and there was no precipitation in January (NOAA, 2003), so this missing fraction would not substantially alter annual estimates.

3. Results and Discussion

3.1. COMPARISONS BETWEEN TYPES OF DEPOSITION

We calculated differences in DD vs. DW, DD vs. W, and DW vs. W by pooling data from each week of collection (July to September 2003) at all sites for each analyte. We first calculated means, medians and standard errors for each deposition type (Table I), and then calculated and tested for differences between pairs of deposition types (Table II) determining significance with the Z -statistic of the Wilcoxon signed-rank test. Dry deposition to dry surfaces vs. dry deposition to wet surfaces differed for TP, $\text{NH}_x\text{-N}$, and TN. DW yielded slightly lower deposition estimates than DD for TP, and DW yielded substantially higher deposition estimates than DD for $\text{NH}_x\text{-N}$ and TN (Tables I and II). Dry deposition differed from W for all analytes except $\text{NO}_x\text{-N}$ and Si. Dry deposition of P was an average of 2.8-times higher than that received in precipitation while dry SRP deposition was up to 12-times higher. Wet deposition and DW were substantially higher than DD for $\text{NH}_x\text{-N}$ and TN. Deposition of $\text{NH}_x\text{-N}$ to wet surfaces was 2.4-fold greater than that in precipitation, while DW and wet deposition of TN were similar (Table II).

The relative size of DD and DW for different nutrients may help to elucidate pathways of deposition. Differences between DW and DD for SRP departed from the findings of Jassby *et al.* (1994), who found greater deposition of SRP to wet surfaces. DD was significantly higher than DW for TP, which was surprising because Cole *et al.* (1990) and Gomolka (1975) found more dry deposition of TP to

TABLE I

Sample size, means, medians, and standard errors for rates dry deposition to dry surfaces (DD), dry deposition to wet surfaces (DW), and wet deposition (W) for all analytes

Analyte	Deposition type	<i>n</i>	Mean ($\mu\text{g m}^{-2}\text{ day}^{-1}$)	Median ($\mu\text{g m}^{-2}\text{ day}^{-1}$)	Standard Error
SRP	DD	55	39	34	2.5
	DW	40	43	36	7.1
	W	52	8	3	2.4
TP	DD	59	94	89	4.5
	DW	41	76	61	8.8
	W	58	31	22	4.1
NH _x -N	DD	49	74	68	6.6
	DW	40	2165	2141	168.6
	W	51	1419	903	225.0
NO _x -N	DD	54	410	400	30
	DW	36	340	320	40
	W	54	740	360	130
TN	DD	54	670	640	70
	DW	41	1950	1700	170
	W	55	2090	1350	320
Si	DD	53	1160	1010	110
	DW	36	1540	1070	320
	W	50	1790	1150	320

Note: Statistics were calculated across all sites (July–September 2003).

wet surfaces than to dry surfaces. Dry surfaces may attract more particles than wet surfaces due to roughness from previously deposited particles or different electrostatic charge. If DD vs. DW differences were simply due to particle attachment, we might have expected the same pattern to be seen in silica (Table II). However, there were no significant differences between DD and DW for silica. Since small particles adhere more tightly to dry polyethylene than large particles (Liu *et al.*, 1994), our TP and Si results are consistent with each other if phosphorus is transported on finer particles than silicious dust. The ratios of DD:DW (Table II) show the greatest differences for NH_x-N and TN (DW > DD). NH₃ dry-deposits up to 50 km away from the source (Asman, 2002); all sites were near NH₃-releasing sources (e.g., large animal confinements); and NH₃ passes rapidly into water from an atmospheric gaseous phase (Larsen *et al.*, 2001). There were no significant differences in NO_x-N deposition between DD and DW, unlike Peters and Reese (1995) and Jassby *et al.* (1994) who found higher NO_x-N dry deposition to wet surfaces. Peters and Reese (1995) suggest that the highly soluble gaseous forms of NO_x (such as HNO₃) may have diffused into wet surfaces at their sites, thus yielding higher deposition.

TABLE II

Comparisons using the Wilcoxon signed-rank test for rates of dry deposition to dry surfaces (DD), dry deposition to wet surfaces (DW), and wet deposition (W), for all analytes

Analyte	Comparison	n	Z-statistic	p-value	Ratio
SRP	DD/DW	34	0.46	0.6455	0.9
	DD/W	45	5.84	<0.0001*	11.3
	DW/W	31	3.99	<0.0001*	12.0
TP	DD/DW	34	2.74	0.0061*	1.5
	DD/W	50	5.82	<0.0001*	4.0
	DW/W	34	3.40	0.0007*	2.8
NH _x -N	DD/DW	32	-4.95	<0.0001*	<0.1
	DD/W	43	-5.19	<0.0001*	0.1
	DW/W	35	2.94	0.0033*	2.4
NO _x -N	DD/DW	31	0.28	0.7794	1.3
	DD/W	49	-1.80	0.0719	1.1
	DW/W	31	-1.50	0.1336	0.9
TN	DD/DW	33	-4.54	<0.0001*	0.4
	DD/W	46	-3.93	<0.0001*	0.5
	DW/W	33	0.98	0.3271	1.3
Si	DD/DW	31	-0.13	0.8966	0.9
	DD/W	43	-0.53	0.5961	0.9
	DW/W	25	-0.89	0.3735	0.9

Note: Associated sample size, Z-statistic, p-value (two-sided), and ratio of the medians of the two deposition types used in the comparison are also shown. Ratios were calculated using the left-hand measurement type under "Comparison" as the numerator and the right-hand measurement type as the denominator. A "*" after the p-value indicates a significant difference within the deposition pair. Because statistics were calculated across all sites (July–September 2003) where pair-wise comparisons were possible, the ratios shown here diverge slightly from those in Table I.

Our data indicated that dry deposition can be more important than wet deposition, especially for P and NH_x (Table II). Both DD and DW were significantly greater than W for phosphorus. Several studies support the finding that D > W for P (Ahn and James, 2001; Cole *et al.*, 1990; Eisenreich *et al.*, 1977; Guerzoni *et al.*, 1999; Jassby *et al.*, 1994; Kleusener, 1972), because P deposition originates from soil and does not become incorporated into rainfall to a great degree. Deposition of NO_x was comparable to the results of Kleusener (1972) who found similar deposition rates of NO_x for wet and dry deposition in central Wisconsin. Asman *et al.* (1998) and Asman (2002) indicated that most NH_x in the form of NH₃ is deposited within 50 km, whereas NH₄⁺ can travel farther from the source, and almost all NH_x is deposited within 1000 km of the source. All sites we sampled were well within

50 km of ammonia sources (e.g., animal feeding operations, fertilized fields), and because NH_3 is highly water soluble (Larsen *et al.*, 2001), we expected DW to be much higher than DD. NH_x and TN indeed showed $\text{DW} > \text{W} > \text{DD}$, with all differences being significant. This indicates that solution of NH_x gas in water on wet surfaces may be a significant pathway of N transport. Nutrient budgets calculated using wet N and P deposition, alone, might greatly underestimate total atmospheric nutrient deposition to moist surfaces.

3.2. PROPORTION OF DISSOLVED INORGANIC NUTRIENTS

Approximately 41% of TP occurred as SRP when considering all deposition types (see Anderson, 2004). Approximately 32% of the TP in DD occurred as SRP, while 62% of the TP in DW occurred as SRP, and only 7% of the TP in wet deposition occurred as SRP. Most or all of the TN occurred as $\text{NO}_x + \text{NH}_x\text{-N}$ (DIN) when considering all deposition types, while 55% of the TN in DD occurred as DIN, 79% of the TN in DW occurred as DIN, and most or all of TN in wet deposition occurred as DIN.

Because only 7% of the P in wet deposition appeared as SRP, wet deposited P was supplied in either the organic or particulate form. Shaw *et al.* (1989) also found that SRP was only 15% of TP wet deposition whereas Jassby *et al.* (1994) found that SRP constituted 44% of TP wet deposition near Lake Tahoe (California-Nevada, USA). The phosphorus dry-deposited to wet surfaces was predominantly in the soluble reactive form (62%), similar to results obtained by Peters and Reese (1995; SRP was 75% of wet P deposition) and Jassby *et al.* (1994; SRP was 47% of wet TP deposition). Phosphorus dry-deposited to dry surfaces was intermediate in inorganic content between W and DW deposition. Peters and Reese (1995) suggest that P in dry deposition may be quite readily available to biota (such as algae and Cyanobacteria) compared to wet deposition, as there is a greater percentage of SRP in dry deposition. Thus, P in dry deposition should receive more emphasis in nutrient budgets because it is more abundant than rainfall P and is readily assimilable. Most of the N deposited as wet or dry deposition occurred as DIN. In comparison, Jassby *et al.* (1994) found dissolved inorganic nitrogen (DIN) was 76% of TN in wet deposition, and only 24% of TN in dry deposition to a dry surface. Although their wet deposition results were similar to ours, we found that inorganic N was a greater fraction of dry-deposited N. This may be due to differences in nutrient sources between the regions under study.

The atomic ratio of $\text{NH}_x:\text{NO}_x$ in wet deposition was 5.7 for all sites and dates, much greater than the ratio of 1.1 found by Tabatabai and Lafen (1976) in the same region. This may indicate that NH_x is increasing in importance over NO_x for total N deposition. This is plausible because the principal nutrient fertilizer applied in this region has recently shifted from NH_4NO_3 to NH_3 (Turner and Rabalais, 1991; Follett, 1995).

TABLE III

Correlation coefficients between Ames 2m and Ames 24m were calculated from July–December 2003 data for each analyte

Analyte	SRP	TP	NH _x -N	NO _x -N	TN	Si
DD (<i>n</i> = 19)	0.78*	0.86*	0.49*	0.68*	0.74*	0.82*
DW (<i>n</i> = 21)	0.38	0.51*	0.70*	0.36	0.70*	0.29
W (<i>n</i> = 28)	0.90*	0.93*	0.99*	0.98*	0.96*	0.98*

Note: A “**” indicates a significant correlation ($p < 0.05$).

3.3. LOCAL VERSUS LONG-DISTANCE TRANSPORT

Comparison of the Ames 24m site with the Ames 2m site with data from July–December 2003 was meant to give an estimate of the importance of local aeolian transport of ground dust into deposition samplers. We conducted temporal correlation analyses of the Ames 2 m and 24 m (Table III); the sites were significantly correlated ($p < 0.05$) for most deposition types and analytes (except SRP, NO_x, and Si for dry deposition to a wet surface). Wilcoxon signed-rank tests were also used to evaluate differences between these sites for DD, DW, and W (see Anderson, 2004). The samplers closer to the ground (Ames 2m) yielded estimates that were significantly higher ($p < 0.05$) than the Ames 24m site only for wet deposition, but for all analytes (Anderson, 2004). No differences were seen between high and low samplers for dry deposition (DD or DW) of any analyte (Anderson, 2004).

The lack of significant differences between DD or DW measured at the high and low sites suggests that dust from the ground contributes little bias to dry deposition estimates. However, weak temporal correlations of DW estimates between Ames 24m and Ames 2m sites for SRP, NO_x, and Si (Table III) suggest that sampler height or other site differences contributed divergent variability to dry deposition at these two sites.

Although the lower sampler collected wet deposition with significantly higher concentrations of all analytes, the Wilcoxon signed-rank test is not sensitive to the size of the differences. Therefore, these differences were frequently quite modest (Table IV) and were small enough that they were not detectable with a Tukey-Kramer test (Table IV). The slight divergence between W estimates at the high and low site may be due to small, site-specific differences in the amount of nutrient entrained with rainfall. Temporal correlations of deposition at these sites were significant ($p < 0.05$) for all analytes for both DD and W, and for TP, NH_x and TN for DW (Table III). This supports the concept that deposition may show regional continuity, at least at the scale of a few km, for W and DD. Sites with higher levels of disturbance may have higher levels of local dust transport than we observed here, however. Alternatively, the Ames 24m site may not have been

TABLE IV
Rates of deposition ($\mu\text{g m}^{-2} \text{ day}^{-1}$) from July through September 2003 at six sites in the state of Iowa, USA

Site	Type	Geometric mean (95% C.I.)					
		SRP	TP	NH _x -N	NO _x -N	TN	Si
Ames 2m	DD	30*	78	30	120	380	670
Ames 24m		30*	69*	40	220	380	690
Cedar Rapids		41	101*	37	210	830	1480
Creston		35	94	27	180	610	920
Le Mars		51*	111*	35	140	590	730
Mason City		34	94	18	90	620	920
Ames 2m	DW	21	66	1638	50	1030	590
Ames 24m		10	29	1705	60	1000	560*
Cedar Rapids		18	67	870*	50	1030	3100*
Creston		16	40	1124	60	1050	350*
Le Mars		51	107	2553*	30	2130	940
Mason City		44	124	1348	40	980	670
Ames 2m	W	4	19	478	360	750	990
Ames 24m		2	15	377	320	510	690
Cedar Rapids		3	11	265	210	550	680
Creston		2	16	272	180	510	620
Le Mars		6	15	234	140	590	850
Mason City		4	18	182	120	570	1000

Note: If global analysis within an analyte-deposition type combination using a mixed effects model indicated one or more statistical difference, each pair of sampling sites was compared using a Tukey-Kramer test. “*” indicates high and low values for an analyte-deposition type combination that differed significantly from each other ($p < 0.05$). Differences between site pairs are discussed in the text.

high enough to differentiate between local and regional signals. Replication of near-ground and elevated samplers would clarify this.

3.4. SITE COMPARISONS

When analyzed using a global, parametric, mixed-effects method (Table IV) wet deposition (July-September 2003) did not differ significantly among sites for any analyte. Estimates of dry deposition showed a few site-to-site differences, however, and all sites diverging significantly from any other for a given analyte and deposition type are highlighted in Table IV. DW of NH_x was significantly higher at Le Mars than Cedar Rapids ($p = 0.0357$). DW of Si was significantly higher at Cedar Rapids than Ames 24m and Creston ($p = 0.0276$; $p = 0.0047$). DD of SRP was significantly higher at Le Mars than Ames 2m and Ames 24m ($p = 0.0074$; $p = 0.0038$). DD

of TP was significantly higher at Cedar Rapids and Le Mars than Ames 24m ($p = 0.0078$; $p = 0.001$). Precipitation concentration statistics (Table V) were also calculated from July-September 2003 data for each site. Concentration geometric means for various sites ranged from 6.4 to 12.1 $\mu\text{g L}^{-1}$ for TP, 400 to 600 $\mu\text{g L}^{-1}$ for TN, and 230 to 590 $\mu\text{g L}^{-1}$ for Si.

The fact that wet deposition was very similar among sites (Table IV) agreed well with another study in the same region (Tabatabai and Lafen, 1976). The few significant differences among dry deposition measures (Table IV) can generally be attributed to site differences such as proximity to confined animal feeding operations (CAFOs) and construction activity during sample collection (Anderson, 2004).

Dry deposition appears to be driven by more localized processes than wet deposition. Elsewhere, we report correlation analyses of weekly measures of all analytes made at all sites. These correlations show that, for DD and DW, only a few sites were significantly positively correlated (see Anderson, 2004). The few temporal correlations in dry deposition among sites may be due to the similarity of local processes at these sites, or simply to chance. More correlations among sites were seen for wet deposition, indicating that wet deposition is generally driven by large-scale processes. Wet deposition at the Ames 24m site and the Ames 2m site were significantly correlated for all analytes (Table III). Creston and Le Mars were separated by 352 km, and yet wet deposition was significantly correlated for all analytes except NO_x . Temporal correlations between samplers separated by large distances suggest that wet deposition data can be extrapolated to sites 10s to 100s of km away. This may be due to the uniformity of land use in our agricultural region since Hendry *et al.* (1981) found that high wet deposition of P in Florida were found near agricultural areas and phosphate mines, and low P deposition rates were found near coastal and forested regions.

3.5. NUTRIENT CONCENTRATIONS

Estimates of nutrient concentrations in wet deposition have varied widely among studies. Chan and Kuntz (1982) collected rainfall near Lake Ontario, and found annual average rainfall TP of 20–300 $\mu\text{g L}^{-1}$, much higher than our geometric means (6.4 to 12.1) (Table V). Likewise, their wet deposition concentrations for NO_x -N and Si (1000 to over 2500 $\mu\text{g L}^{-1}$, and 200–3000 $\mu\text{g L}^{-1}$, respectively) were greater than ours (Table VI). We compared our data to the NADP's 2003 data for two sites in Iowa (NADP, 2004). When converted to units of nitrogen mass, their NO_3 -N volume-weighted means were very close to our geometric means (250–270 $\mu\text{g L}^{-1}$ versus 220–310 $\mu\text{g L}^{-1}$), and their values for NH_x -N are overlapped by our 95% confidence intervals (380–467 $\mu\text{g L}^{-1}$ versus 104–400 $\mu\text{g L}^{-1}$, respectively) (Table V). Peters and Reese (1995) found higher SRP and TP concentrations than our data (10 ± 16 versus 0.9–3.3 $\mu\text{g L}^{-1}$, and 52 ± 89 versus 6.4–12.1 $\mu\text{g L}^{-1}$, respectively). Their NO_x -N data are, however, very similar to ours

TABLE V
 Rainfall concentrations ($\mu\text{g L}^{-1}$) of six analytes over a three-month period (July to September 2003) at six sites in Iowa. This period is representative of the warmest and driest part of the summer. Geometric means and confidence intervals were back-calculated from log-transformed data

Site	Geometric mean (95% C.I.)					
	SRP	TP	NH ₄ -N	NO _x -N	TN	Si
Ames 2m	1.3 (0.4–4.1)	8.9 (3.6–22)	169 (25–1150)	280 (150–510)	560 (230–1310)	420 (190–920)
Ames 24m	0.9 (0.2–3.7)	9.4 (3.7–23.7)	177 (26–1221)	330 (170–620)	480 (190–1210)	330 (100–1060)
Cedar Rapids	1.1 (0.3–4.4)	6.4 (1.1–36)	135 (12–1465)	330 (150–720)	600 (180–1910)	260 (60–960)
Creston	1.2 (0.2–7.4)	12.1 (4.3–33.7)	145 (14–1517)	300 (90–940)	440 (100–1810)	230 (40–1130)
Le Mars	3.3 (0.4–25.7)	11 (1.8–67.4)	400 (98–1629)	310 (90–940)	350 (70–1550)	590 (130–2590)
Mason City	1.1 (0.2–5)	11 (3.4–35.7)	104 (8–1430)	220 (80–590)	480 (150–1540)	350 (120–1010)

TABLE VI

Studies of total deposition ($\text{kg ha}^{-1} \text{ year}^{-1}$). Values represent total dry deposition to a dry surface + wet deposition, except where noted

Study site	Reference	P	N	Si
This study		0.30	7.71	6.09
Florida	(Hendry <i>et al.</i> , 1981)	0.24–0.96	7.70–11.30	
Lake Michigan	(Eisenreich <i>et al.</i> , 1977)	0.29		1.38
Lake Michigan	(Murphy and Doskey, 1976)	0.17 [§]		
Lake Simcoe, Ontario, Canada	(Winter <i>et al.</i> , 2002)	0.56	9.20	
Lake Superior	(Johnson and Eisenreich, 1979)			3.17
Many locations	(Newman, 1995)	0.07–17.00		
Narrow Lake, Alberta, Canada	(Shaw <i>et al.</i> , 1989)al. 1989	0.20	4.24	
Northeast Minnesota	(Munger, 1982)	0.09–0.15 [§]		
South Florida	(Ahn and James, 2001)	0.41 ± 0.33		

[§] wet deposition only.

($300 \pm 170 \mu\text{g L}^{-1}$ versus $220\text{--}310 \mu\text{g L}^{-1}$). Our concentrations were averaged from July–September 2003, whereas Peters and Reese (1995) showed only data from May–June 1992. Further, their data were collected in Florida, which receives P aerosols from the ocean and fertilizer application (Peters and Reese, 1995).

3.6. ANNUAL DEPOSITION

The data from the Ames 24m site illustrates the annual trend in DD and W. Most analytes showed a seasonal pattern of high deposition in spring, and lower deposition in winter (Figure 2). Phosphorus deposition was dominated by dryfall; deposition was maximal in the spring and declined through the summer (Figure 2). Both dry and wet P deposition declined to low levels in winter. Deposition of nitrogen and silica was dominated by wet deposition, with high rates in early spring, and sporadic peaks throughout the summer. Dry and wet N deposition also declined to low levels in winter (Figure 2).

Annual trends in atmospheric deposition agree with those seen by several others (Delumyea and Petel, 1978; Linsey *et al.*, 1986; Scheider *et al.*, 1979; Shaw *et al.*, 1989) who attributed this trend to agricultural activities. In Iowa, in 2003, planting and fertilization of maize and soybeans began on April 13 and was complete by mid-June (USDA/NASS, 2004). Harvest began in September and was complete by November 23, with substantial post-harvest fertilizer application. Planting and fertilization coincided with the peak deposition rates. Because ratios of DW to DD for nitrogen deposition were high (Table II), annual dry N deposition to a wet surface would likely exceed DD. This is due to the large deposition of NH_x to wet surfaces. Dry deposition of P was greater than wet deposition from spring through autumn (Figure 2), but dry and wet deposition of P declined to similar, low levels

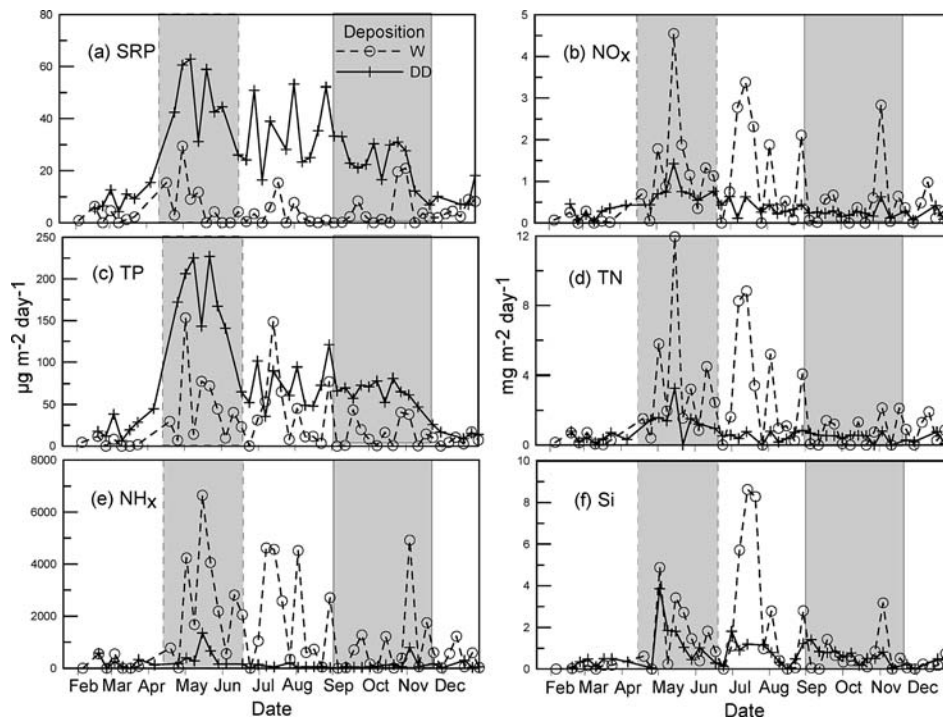


Figure 2. Ames 24m annual wet deposition and dry deposition to a dry surface (2003) were plotted for the following analytes: (a) Soluble reactive phosphorus (SRP), (b) Total phosphorus (TP), and (c) Ammonia/ammonium nitrogen (NH_x), (d) Nitrate/nitrite nitrogen (NO_x), (e) Total nitrogen (TN), and (f) Silica (Si). Deposition rates of all nutrients are expressed as masses of N, P or Si. Shaded areas with dotted and solid lines indicate planting and harvesting periods, respectively.

in winter. Wet and dry deposition of silica were similar, with a few departures in mid-summer.

We calculated annual estimates of total (dry plus wet) deposition (Table VII) by summing weekly estimates. We also estimated annual deposition of DW plus W, by extrapolating DW rates from DD assuming ratios of DD:DW found through the July-September portion of the season. Annual deposition rates of NH_x -N and TN deposition were much higher when considering DW, whereas the other analytes showed similar values.

The annual rate that is most relevant to moist aquatic and moist terrestrial systems is $\text{DW} + \text{W}$. Most literature values, however, calculate $\text{DD} + \text{W}$ or only W as an annual estimate of atmospheric deposition. Estimates excluding dry deposition to wet surfaces would greatly underestimate annual deposition of N. In our study, TP deposition calculated as $\text{DD} + \text{W}$ was $0.3 \text{ kg ha}^{-1} \text{ year}^{-1}$ which is well within the review of annual deposition given by Newman (1995) (Table VI). However, Newman states that the majority of studies reviewed showed P deposition in the

TABLE VII

Estimates of annual total deposition ($\text{kg ha}^{-1} \text{ year}^{-1}$) (defined as dry deposition to a dry surface plus wet deposition, and dry deposition to wet surfaces plus wet deposition) from the Ames 24m site for January 28, 2003, to January 5, 2004, for all analytes

Analyte	Annual estimate (DD + W)	Annual estimate (DW* + W)	Ratios	
			DD:W	DW:W
SRP	0.094	0.078	5.2	4.2
TP	0.299	0.254	2.6	2.2
NH _x -N	4.812	10.290	0.1	1.6
NO _x -N	3.67	3.26	0.4	0.4
TN	7.71	10.51	0.3	1
Si	6.09	5.52	0.5	0.5

Note: Note these estimates are missing a few weeks of winter data (see text). Ratios of DD:W and DW*:W are also given from the Ames 24m site for January 28, 2003, to January 5, 2004, for all analytes. The asterisk indicates that January to June DW values were extrapolated from DD values using ratios of DD:DW observed from July 2003 to January 2004.

range of $0.1\text{--}10 \text{ kg ha}^{-1} \text{ year}^{-1}$, and any source that provides much less than $0.1 \text{ kg ha}^{-1} \text{ year}^{-1}$ would be of little significance (Newman, 1995). Studies in the U.S. state of Florida (Hendry *et al.*, 1981) and on Lake Michigan (Eisenreich *et al.*, 1977) (Table VI) yielded annual P deposition rates that also exhibited a large dryfall component. Lake Simcoe in Ontario (Winter *et al.*, 2002) is surrounded by one of Canada's most agricultural and urbanized regions which could contribute greatly to higher atmospheric P deposition rates (Table VI). Past data in Iowa showed wet deposition of $0.32 \text{ kg ha}^{-1} \text{ year}^{-1}$ for P (Jones, 1974). Other studies for which only annual wet deposition was calculated yielded estimates of annual deposition of P of $0.09\text{--}0.15 \text{ kg ha}^{-1} \text{ year}^{-1}$ for north central Minnesota (Munger, 1982), and annual deposition of TP of $0.17 \text{ kg ha}^{-1} \text{ year}^{-1}$ for Lake Michigan (Murphy and Doskey, 1976). Annual P deposition seen in this agricultural region was greater than some of these but did not depart substantially from those observed elsewhere. Although it is surprising that the values derived for this highly agricultural region are not extreme, this may be due to our strict elimination of samples containing contaminants that can inflate deposition estimates and apparent variability.

Ranges of annual N deposition in other studies were less variable than those of P deposition. When converted to nitrogen mass deposition, NADP (2004) estimates of NO_x-N and NH_x-N wet deposition in Iowa in 2003 were lower for NH_x-N ($3.0\text{--}3.5$ versus $4.8 \text{ kg ha}^{-1} \text{ year}^{-1}$), and NO_x-N ($1.8\text{--}2$ versus $3.7 \text{ kg ha}^{-1} \text{ year}^{-1}$) than our estimates of total deposition. This is likely because NADP only measures

nutrients in rainfall (NADP, 2004), so these values do not include the dry component of atmospheric nutrient deposition. $\text{NO}_3\text{-N}$ deposition in Wisconsin (Andraski and Bundy, 1990) varied between 4.2 and 8.4 $\text{kg NO}_3\text{-N ha}^{-1} \text{ year}^{-1}$. $\text{NH}_4\text{-N}$ deposition in Wisconsin (Andraski and Bundy, 1990) averaged 8.7 $\text{kg ha}^{-1} \text{ year}^{-1}$, which was twice as high as our values. Wisconsin is known to have substantial point sources of NO_x emissions (WDNR-ADTF, 1983) and all Wisconsin sites were located in “important agricultural areas” (Andraski and Bundy, 1990), which may explain the high annual deposition rates. Hendry *et al.* (1981) reported annual deposition of TN with a great wet deposition component (Table VI). Shaw *et al.* (1989) report lower annual TN deposition than our values (Table VI), but this may be attributed to differences in study locations (boreal forest versus agricultural landscape). Past wet deposition data for our region showed $\text{NH}_3 + \text{NO}_3\text{-N}$ deposition of 8.3 $\text{kg ha}^{-1} \text{ year}^{-1}$ (Jones, 1974). Thus, it appears that landscape characteristics lead to variability in the deposition of N but that N deposition in this region is not extreme with respect to other reported values.

In addition, the N:P ratio in total atmospheric deposition for this study (as atoms) was 60:1, which is higher than both the 24.2 found in regional aquatic ecosystems in the mid-1970's (Bachmann and Jones, 1976), and the average of 53 found in 1991–1992 (Arbuckle and Downing, 2001). Since phytoplankton tend toward average N:P of 16:1 (Redfield ratio), anything above this ratio being added to a system, i.e., nutrients from the atmosphere, will drive ecosystems toward P-limitation (Downing and McCauley, 1992). This would also suggest that both aquatic and terrestrial ecosystems fed principally by these atmospheric nutrients would tend toward P limitation.

Our estimate of annual silica deposition is among the few published values and averaged 6.09 $\text{kg Si ha}^{-1} \text{ year}^{-1}$. Past data from the Great Lakes region show lower deposition rates (Table VI). Additional recent data show that Lake Michigan received 3.62 $\text{kg Si ha}^{-1} \text{ year}^{-1}$ in urban areas from dry deposition alone (Holsen *et al.*, 1993). Diatom phytoplankton, which require silica for growth, are important components of the biological community and food web and are sensitive to changes in water quality. Diatom phytoplankton in lakes require Si and DIN in a minimum atomic (molar) ratio of 1:1, and may be replaced by harmful algae if Si is low in relation to DIN (Officer and Ryther, 1980). Annual atmospheric deposition of DW + W in our study would not favor diatom growth, as the atomic ratio of Si:DIN is 1:3, and so may therefore contribute to harmful algal blooms.

3.7. ATMOSPHERIC WASHOUT

Several authors (Andraski and Bundy, 1990; Gomolka, 1975; Shaw *et al.*, 1989) have suggested that rainfall washes atmospheric dust from the atmosphere and that atmospheric moisture does not carry nutrients *per se*. If this were so, the concentrations of nutrients found in precipitation would be negatively related to the amount of rainfall delivered per unit time. Figure 3 supports this hypothesis,

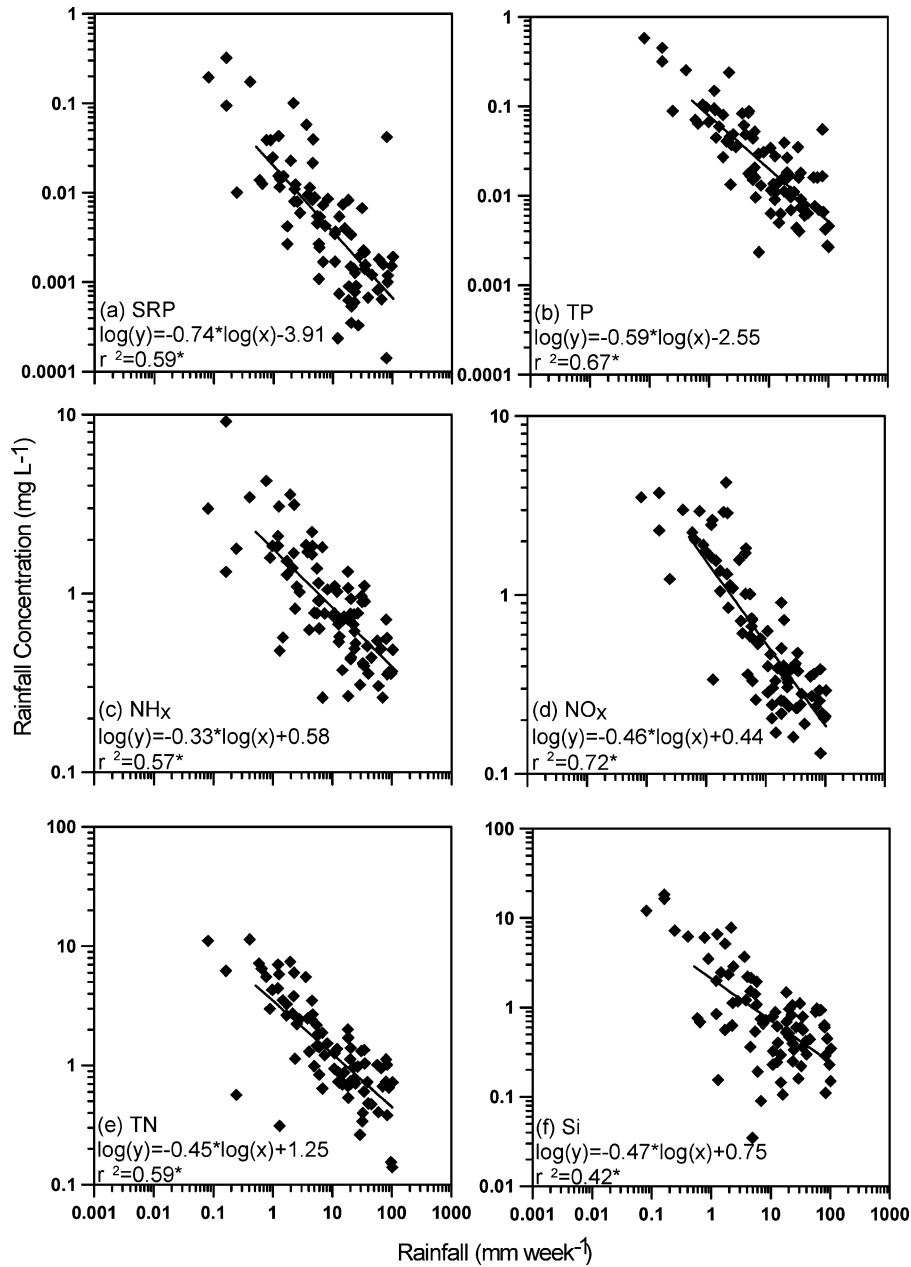


Figure 3. Washout relationships estimated from rainfall concentrations plotted against rainfall rates using data from all sites and dates for all analytes. Concentrations of all nutrients are expressed as masses of N, P or Si. Statistical analysis of the fit of the points to linear regression lines (equation and R² values; an “*” indicates $p < 0.001$ for F for mean squares for regression) is included in each panel.

as concentrations of each analyte in rainfall decreased with increasing rainfall rates. It is likely, therefore, that the source of wet deposited nutrients is airborne particulates and gases. In addition, even though large rainfall events lower nutrient concentrations, increased rainfall still results in increased nutrient deposition for all analytes because the exponents of the relationships in Figure 3 were all > -1 . 2003 was a relatively dry year with 75.5 cm of precipitation, whereas the average precipitation for this region from 1971–2000 was 88.0 cm (NOAA, 2002). Given that increased rainfall resulted in increased rates of deposition, our annual deposition rates may have been underestimates of long-term means. Because dry and wet deposition contribute different forms of nutrients at different rates, the availability, stoichiometry, and nutrient load from atmospheric sources will vary greatly with the rate and timing of precipitation.

3.8. CONTAMINATION

Our deposition estimates are not extreme despite the great mobility of nutrients in agricultural landscapes. This may be due to our strict use of uncontaminated samples. Many studies have, however, reported or included contaminants such as plant parts, insects, and bird droppings in estimates of atmospheric deposition (Ahn, 1999; Newman, 1995; Peters and Reese, 1995). Contamination in deposition studies could be cast as local recycling (Ahn and James, 2001), because birds and insects use and excrete nutrients on land. These sources of contamination may not, however, be part of the true atmospheric deposition signal. We did not use samples contaminated with dead insects or bird droppings in our analyses. This may explain why our measurements were lower than some of the relatively high values found in reviews of atmospheric deposition.

4. Conclusions

Our findings indicate that atmospheric deposition can be a significant nutrient source to ecosystems. Dry deposition of P represents the bulk of seasonal deposition and the most important fraction of total atmospheric P deposition. Annual wet deposition and dry deposition of N to wet surfaces were approximately equal, owing to very large NH_x -N deposition derived from gas transport. Si deposition from atmospheric sources was substantial, with dry deposition comparable to wet. Thus, it is essential to measure dry deposition and wet deposition when estimating budgets of N, P, or Si.

Atmospheric nutrient deposition varies seasonally and may be related to tillage and fertilization schedules in agricultural areas as well as other activities in the airshed. Temporal variability indicated that the highest rates of atmospheric deposition of nutrients in agricultural regions occurred during the spring when soils were tilled and fertilized prior to planting. Dry deposited nutrients appear to be driven somewhat by more local disturbance, and extrapolation to areas of divergent

land use may be inaccurate. Remediation of nutrient enrichment problems resulting from dry atmospheric deposition should therefore focus on methods that limit dust and particulate nutrient suspension. Nutrient deposition via rainfall may be less site-specific, however, and may result from more constant processes integrated over larger areas.

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Note

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