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Atmospheric Inputs of Nitrogen, Carbon, and Phosphorus across an Urban Area: Unaccounted Fluxes and Canopy Influences

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Key Points:
- Atmospheric deposition is a substantial source of organic nitrogen, organic carbon, and phosphorus to urban ecosystems
- Inorganic phosphorus inputs in bulk deposition and throughfall were greater than sewage effluent inputs during the study period
- The tree canopy amplifies inputs to the urban ground surface, potentially increasing nutrient fluxes in runoff to nearby waterways

Supporting Information:
- Supporting Information S1.

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Abstract Rates of atmospheric deposition are declining across the United States, yet urban areas remain hotspots of atmospheric deposition. While past studies show elevated rates of inorganic nitrogen (N) deposition in cities, less is known about atmospheric inputs of organic N, organic carbon (C), and organic and inorganic phosphorus (P), all of which can affect ecosystem processes, water quality, and air quality. Further, the effect of the tree canopy on amounts and forms of nutrients reaching urban ground surfaces is not well-characterized. We measured growing season rates of total N, organic C, and total P in bulk atmospheric inputs, throughfall, and soil solution around the greater Boston area. We found that organic N constitutes a third of total N inputs, organic C inputs are comparable to rural inputs, and inorganic P inputs are 1.2 times higher than those in sewage effluent. Atmospheric inputs are enhanced two-to-eight times in late spring and are elevated beneath tree canopies, suggesting that trees augment atmospheric inputs to ground surfaces. Additionally, throughfall inputs may directly enter runoff when trees extend above impervious surfaces, as is the case with 26.1% of Boston's tree canopy. Our results indicate that the urban atmosphere is a significant source of elemental inputs that may impact urban ecosystems and efforts to improve water quality, particularly in terms of P. Further, as cities create policies encouraging tree planting to provide ecosystem services, locating trees above permeable surfaces to reduce runoff nutrient loads may be essential to managing urban biogeochemical cycling and water quality.

1. Introduction

Rates of atmospheric deposition, particularly of nitrate (NO_3^-), have been declining around the United States (U.S.) over the last few decades (Li et al., 2016). Despite this broad-scale reduction, urban areas remain hotspots of atmospheric deposition; recent studies in urban areas demonstrate that rates of atmospheric deposition of inorganic nitrogen (N) are elevated more than twofold over rural rates, are highly spatially variable, and are positively correlated with impervious surface area and local vehicular N emissions (Decina et al., 2017; Fang et al., 2011; Rao et al., 2014; Templer & McCann, 2010). Elevated atmospheric inputs of inorganic N contribute to eutrophication in waterways (Valiela et al., 1997), a problem both within and near urban areas (Carpenter et al., 1998; Moore et al., 2003; Paerl, 1997). While national monitoring networks that measure inorganic N in deposition have generally avoided cities (Lohse et al., 2008), some, including the National Atmospheric Deposition Program (NADP) in the U.S., have begun to expand into urban areas such as New York City and Boston, Massachusetts (MA). However, in comparison to atmospheric deposition of inorganic N, less is known about the controls on and the effects of atmospheric inputs of other important elements in urban ecosystems, namely those of organic N, organic carbon (C), and phosphorus (P).

Studies dating back over 100 years have shown organic N to be a substantial component of N deposited in rainwater (Miller, 1905), and others have shown dry deposition to be an important source of organic N (González-Benitez et al., 2009; Matsumoto et al., 2014). Organic N includes a variety of molecules (e.g., urea, amines, and peptides) found in aqueous, particulate, and gaseous phases with a range of solubility and reactivity (Cape et al., 2011). The amount of organic N as a proportion of total N (inorganic + organic N) N deposition has been found to be about a third in mostly rural locations across 160 studies worldwide and

58 studies in North America (29% and 35%, respectively; Cornell, 2011). A similar proportion of organic N in total deposition has also been found in urban areas in studies around the world (Araujo et al., 2015; Chen et al., 2015; de Souza et al., 2015; González-Benitez et al., 2009; He et al., 2011; Izquieta-Rojano et al., 2016; Li et al., 2012; Matsumoto et al., 2014). There have been a handful of studies in the U.S. measuring organic N deposition in small towns in Connecticut and North Carolina, in winter precipitation in Salt Lake City, Utah and in Miami, Florida; four of these studies either do not report organic N deposition rates or do not report organic rates in the urban sites individually, and the others report the percentage of organic N in total N as below 20% (Brezonik et al., 1983; Hall et al., 2016; Keene et al., 2002; Luo et al., 2002; Nadim et al., 2001; Zamora et al., 2011). The rate of atmospheric deposition of organic N is thus currently unknown in many large urban areas throughout the U.S, and as the majority of urban studies only consider atmospheric inputs of inorganic N, estimates of total N inputs into these ecosystems are likely to be underestimated.

Like organic N, atmospheric deposition of organic C is a large input of organic C to ecosystems, delivering 430 Tg of dissolved organic C in rain to the surface of the Earth per year (Willey et al., 2000), an amount equivalent to about half of the C delivered to oceans by rivers annually (IPCC, 2014). This sizeable deposition flux has the potential to affect regional C cycling, radiative forcing, and human health through creation of toxic compounds such as polycyclic aromatic hydrocarbons (Yan & Kim, 2012). Organic C contains compounds that are varied in structure and chemical properties (Likens & Galloway, 1983). Sources of atmospheric organic C are both biogenic, such as emission of volatile organic compounds from plants and forest fires, as well as anthropogenic, such as fossil fuel combustion (Iavorivska et al., 2016). A recent review of studies of deposition of organic C in mostly rural sites reported a global mean wet deposition flux of 34 ± 33 kg C ha^{-1} a^{-1} across 40 sites and a mean wet deposition flux of 21 ± 19 kg C ha^{-1} a^{-1} across seven sites in North America (Iavorivska et al., 2016). Studies conducted in urban areas have linked atmospheric organic C to fossil fuel emissions (Huang et al., 2010; Santos et al., 2014; Siudek et al., 2015; Wang et al., 2016; Yan & Kim, 2012) and deposition of organic C has been shown in some studies to be higher in urban than nonurban sites (Likens & Galloway, 1983; Lohse et al., 2008; Siudek et al., 2015). To our knowledge, organic C in precipitation has only been measured in several cities of varying size and climate across the U.S. (Likens & Galloway, 1983; Velinsky et al., 1986; Sakugawa & Kaplan, 1993; Willey et al., 2000; Lohse et al., 2008) and at rates comparable to rural rates, but with so few studies, deposition of organic C in urban areas is still poorly understood.

An essential nutrient for biota, P is often the limiting element for net primary productivity in many ecosystems, with excess inputs causing freshwater eutrophication (Mahowald et al., 2008; Smil, 2000). Rates of total P (inorganic + organic P) in atmospheric deposition have been found to be minimal across mainly rural locations in 250 sites around the world and 38 sites in North America (mean rates = 0.43 ± 0.42 and 0.39 kg P ha^{-1} a^{-1}, respectively; Tipping et al., 2014). With many atmospheric sources in cities such as industrial activity, dust from roads and construction sites, combustion of fossil fuels, fertilizer, and biogenic particles, it is not surprising that studies in some urban locations report rates of total P deposition elevated above the mean value for rural sites (Eisenreich et al., 1977; Hou et al., 2012), and in some cases up to 2 kg P ha^{-1} a^{-1} (He et al., 2011; Pandey et al., 2016; Sun et al., 2014). These urban fluxes are comparable to or greater than mean rates of total P in runoff from a variety of urban land uses (0.7 kg P ha^{-1} a^{-1}; Burton Jr. & Pitt, 2002). Deposition of P in the U.S. has been measured in urban areas in several states (Brezonik et al., 1983; Eisenreich et al., 1977; Koelliker et al., 2004; Lohse et al., 2008; Pollman et al., 2002) with variable rates of atmospheric P deposition reported across studies. As many cities in the U.S. are required by the U.S. Environmental Protection Agency, as well as local regulatory agencies, to reduce the amount of P in their waterways (Boyer & Kieser, 2012), quantifying the magnitude of the atmospheric P flux is crucial for determining and mitigating urban P pollution.

In addition to measuring unaccounted fluxes of organic N, organic C, and organic and inorganic P, it is important to quantify the effect of the urban tree canopy on rates of urban deposition and nutrient budgets. Extensive work in rural areas has shown that processes within the tree canopy can transform ammonium (NH_4^+) and NO_3^- through a variety of biological and chemical mechanisms (Balestrini & Tagliaferri, 2001; Fenn et al., 2013; Kopáček et al., 2009; Lovett & Lindberg, 1993; Ponette-González et al., 2015; Templer et al., 2015a; Zimmermann et al., 2006), such as foliar uptake and leaching (Garten Jr. et al., 1998), microbial transformations (Ferm, 1993), the action of epiphytic lichen (Reiners & Olson, 1984), and cation exchange (Tukey Jr., 1970). The tree canopy has also been suggested to produce P from foliar leaching and to be an important source of organic C to the forest floor (Qualls et al., 1991). Moreover, the tree canopy is known to

augment rates of nutrient inputs to the ground, as well as to waterways (Ormerod et al., 1989; Reynolds et al., 1986), in nonurban areas through capturing dry deposition (Lovett & Lindberg, 1993) and the production of pollen (Kopáček et al., 2011), insect frass (le Mellec et al., 2011), and other biological material (Osono et al., 2006; Pedersen & Bille-Hansen, 1995). Studies in urban areas have reported elevated rates of throughfall (inputs beneath tree canopy) compared to bulk (inputs beneath open sky) deposition (Juknys et al., 2007; Michopoulos et al., 2007; Kimura et al., 2009; Fang et al., 2011; Bettez & Groffman, 2013; Tulloss & Cadenasso, 2015; Izquieta-Rojano et al., 2016, Ponette-González et al., 2017). In many studies, however, throughfall has been measured at a limited number of urban sites and/or has not been collocated with bulk measurements for direct comparisons. As numerous cities have undertaken urban tree planting efforts to provide ecosystem services such as heat island and pollution reduction (Bealey et al., 2007; Bowler et al., 2010; Brack, 2002; Tallis et al., 2011), though with questionable efficacy in achieving the latter goal (Vos et al., 2013), it is essential to determine the potential effects of the tree canopy on delivery of atmospheric inputs to soils and nearby waterways.

In 2015, we measured organic and inorganic N, organic C, and organic and inorganic P in bulk and throughfall atmospheric inputs at nine sites throughout the greater Boston area, five of which had collocated bulk and throughfall measurements. To quantify the effect of urban atmospheric inputs on the loss of organic and inorganic N, organic C, and organic and inorganic P in bulk in urban soils, we also measured these compounds in soil solution at the five sites with soil present. We expected to find that (1) organic N constitutes about a third of total atmospheric N inputs, (2) rates of organic C input are comparable or elevated with respect to rural rates, (3) rates of atmospheric P input are negligible, and (4) inputs of all elements beneath tree canopies are amplified with respect to atmospheric inputs in open areas.

2. Materials and Methods

2.1. Study Area and Site Selection

The Boston, MA metropolitan area has 4.5 million inhabitants and is the most populous metropolitan area in New England (U.S. Census Bureau, 2013). The area experiences about 115 cm of precipitation per year, with a mean pH of 5.3 in rainfall during the time period of the study (data from NADP site MA22), and mean summer and winter air temperatures of 22.2 and 0.2°C, respectively (National Climatic Data Center, 2016). The state of MA is primarily covered with mixed deciduous forest (Homer et al., 2015), with oak trees (*Quercus spp*.) among the most common canopy species. We sampled at nine sites. Three sites (sites 1, 6, and 7) were collocated with MA Department of Environmental Protection air quality monitoring stations, three sites were collocated with current (sites 4 and 9; site 4 was not operational until 3 months after this study ended) and former (site 3) NADP National Trends Network (NTN) sites, and two were located on rooftops at Boston University (sites 8 and 9). Ground site 1 and roof site 8 were collocated, with site 1 a distance of 75 m from site 8. Site details are reported in Table 1. Results from a roof-ground comparison of fluxes, as well as a comparison of NADP NTN wet deposition measurements with bulk ion exchange resin measurements, are discussed in the Supporting Information S1, including Figures S1 and S2.

2.2. Bulk and Throughfall Total N, Organic C, and Total P

We measured organic and inorganic N, organic C, and organic and inorganic P in both bulk deposition and throughfall. Bulk measurements were made by placing three ion exchange resin collectors beneath open sky (no tree canopy) at each site ($n = 27$ resin collectors total). For sites with throughfall measurements (sites 1–5), three additional ion exchange resin collectors were placed beneath the tree canopy ($n = 15$ resin collectors total). Bulk measurements capture mostly wet (ions dissolved in precipitation) and some dry (particulate and gaseous) atmospheric inputs (Lovett, 1994). Throughfall measurements have been in common use for the last three decades to estimate wet and dry N inputs from the atmosphere to the surface of the Earth (Lovett & Lindberg, 1993; Bettez & Groffman, 2013; and many others). In throughfall, particles and gases adsorbed or dissolved on leaf cuticle surfaces in the tree canopy wash off during precipitation events, and the precipitation containing these species as well as ions already dissolved in the precipitation enters the throughfall collector. As the tree canopy can alter throughfall chemistry by uptake of ions and conversion of ions by microbes and leaf surfaces (Fenn et al., 2013; Lovett & Lindberg, 1993; Sparks, 2009; Templer et al., 2015a, 2015b), this method provides an approximation, rather than a direct measurement, of total wet and dry atmospheric inputs. Throughfall measurements do however represent the total nutrient flux reaching the ground surface, regardless of whether these inputs are atmospheric in nature or are canopy derived.

Table 1.
Site Details

Site	Location	Measurement type	Rooftop	Monitoring site	Dominant canopy species
1	42°20′56.20′′N, 71° 5′52.32′′W	Bulk, Throughfall	No	Mass DEP	*Malus sp.*
2	42°21′5.40′′N, 71° 6′40.19′′W	Bulk, Throughfall	No	None	*Quercus sp.*
3	42°23′0.34′′N, 71°12′46.52′′W	Bulk, Throughfall	No	NADP (inactive)	*Acer sp.*
4	42°17′44.79′′N, 71° 7′51.74′′W	Bulk, Throughfall	No	NADP (active)	*Quercus sp.*
5	42°17′50.35′′N, 71° 7′10.28′′W	Bulk, Throughfall	No	None	*Prunus sp.*
6	42°19′30.50′′N, 71° 3′21.62′′W	Bulk	No	Mass DEP	No canopy
7	42°19′46.41′′N, 71° 4′57.69′′W	Bulk	No	Mass DEP	No canopy
8	42°20′59.12′′N, 71° 5′51.37′′W	Bulk	Yes	None	No canopy
9	42°21′0.60′′N, 71° 6′15.63′′W	Bulk	Yes	NADP (active)	No canopy

All sites located in the City of Boston, with the exception of site 3, located in Waltham, MA. NADP = site collocated with a currently active or inactive National Atmospheric Deposition Program National Trends Network site; Mass DEP = site collocated with current Massachusetts Department of Environmental Protection air quality monitoring stations. Sites 1 and 8 were collocated roof and ground sites.

Ion exchange resin collectors collected bulk and throughfall inputs between 12–13 May and 23–24 October 2015 (roughly corresponding to the period in which most deciduous trees had leaves in Boston, MA, hereafter referred to as the "growing season") and were replaced three times after initial installation for a total of four measurement periods. These measurement periods are referred to in the text as T1 (12–13 May – 22 June), T2 (22 June–3 August), T3 (3 August–14 September), and T4 (14 September–23–24 October). Each ion exchange resin collector consisted of a polypropylene funnel attached to a 20-mL polypropylene chromatography column (BioRad, Hercules, CA) filled with mixed anion-cation exchange resin (Dow Chemical, Midland, MI). A ball of polyester fiberfill was inserted into the opening of the funnel to filter large particles, and the funnel/column setup was mounted atop a 1-m tall piece of PVC (method described in greater detail in Templer & McCann, 2010). Charged ions such as NH_4^+ and NO_3^-, organic C compounds, and phosphate (PO_4^{3-}) were adsorbed onto the ion exchange resin as rainwater percolated through the chromatography column.

In the laboratory, resin from each ion exchange resin collector was extracted with a total of 150 mL 2 M potassium chloride (KCl) and shaken on a shaker table for three 30-min intervals over 1.5 h. The extract solution was analyzed for NH_4^+, NO_3^-, and PO_4^{3-} via the colorimetric microplate method (Sims et al., 1995). Briefly, samples were pipetted into 96-well microplates with two standard curves per plate (one at the beginning and one at the end) with an external quality control (QC) solution (NH_4^+ ion ERA no. 985; NO_3^- ion ERA no. 991; PO_4^{3-} ion ERA no. 993; ERA, Golden, CO) for each curve and a QC and standard check solution in the middle (after nine samples). Procedures for making and adding reagents to microplates, as well as analysis of microplates on a microplate reader (VersaMax microplate reader, Molecular Devices, Sunnyvale, CA) followed Sims et al. (1995) and Doane and Horwath (2003) for NH_4^+ and NO_3^- and D'Angelo et al. (2001) for inorganic P.

Total N and organic C were analyzed on a Total N-Total Organic Carbon Analyzer (Shimadzu, Kyoto, Japan) in the Weintraub laboratory at the University of Toledo. For analysis of total P, samples underwent an alkaline persulfate digestion in an autoclave at 120°C for 90 min to convert all P in the sample to inorganic PO_4^{3-} following De Borba et al. (2014). As a digestion efficiency check, samples of a known concentration of organic P (0.5 ppm glycerophosphate) were digested along with field samples (De Borba et al., 2014). Based on recovery from this digestion efficiency check, values for total P in samples were adjusted upward 4.9% (Doyle et al., 2004). Organic N and P were determined as the difference between inorganic N or P and total N or P, respectively. For samples where the total N or P were found to be less than inorganic N or P (33 out of 304 samples, average negative difference = 9.8%), we set organic N or P to zero, resulting in total N or P being equivalent to inorganic N or P.

Concentrations in ion exchange extract solutions were converted to per area rates by multiplying concentration by the total volume of KCl used to extract samples (150 mL) and dividing by the surface area of the funnel top (20.2 cm diameter). Values for bulk and throughfall N, organic C, and total P measurements are presented three ways: per-day units (kg ha^{-1} d^{-1}) in order to examine temporal variation across the sampling dates, per-year units (kg ha^{-1} a^{-1}) with growing season flux extrapolated over the year in order to compare the values from this study to those in other studies, and the measured cumulative growing season rates for the entire study period (kg ha^{-1} growing $season^{-1}$; number of days unique to each site, Table S1 in Supporting Information S1). Extrapolating partial year rates to the entire year has been done in multiple studies (Hou et al., 2012; Rao et al., 2014; Tulloss & Cadenasso, 2015), but as deciduous trees do not have the same amount of adsorptive canopy surface area in winter as trees with leaves, this extrapolation provides only a rough estimate, rather than an absolute quantification, of potential annual inputs, particularly for throughfall measurements.

Estimates of atmospheric input rates were based on the mean flux values from the three replicates at each site for each type of measurement (bulk or throughfall), and summed over the four time periods of the growing season. Daily rates were obtained by dividing the summed growing season rate by the total number of days in the field, with annual values obtained by multiplying the daily value by 365 days. For throughfall, we additionally calculated annual fluxes based only on the final three time periods, following the procedure above, but excluding the first time period, which occurred during late spring (T1: 12–13 May –22 June 2015). This additional calculation was made to compare extrapolation to the entire year from all four time periods versus the last three time periods only, since the late spring sampling period (T1) was elevated compared to the other three sampling periods and represents a seasonal peak (more detail provided below).

2.3. Soil Solution Total N, Organic C and Total P

Soil solution total N, organic C, and total P were measured at the five sites with available soil (sites 2, 3, 4, and 5, and underneath the throughfall collectors only at site 1). Two porous nylon bags ("resin bags"; $n = 6$ per each bulk or throughfall site; $n = 54$ total per measurement period) filled with 10 g of ion exchange resin were buried within 25 cm of each ion exchange resin collector at a depth of 10–15 cm beneath the soil surface to measure mobile total N, organic C, and total P in soil solution. The amount of total N, organic C, and total P accumulated in resin bags provides a proxy for soil total N, organic C, and total P that may be potentially lost in leachate and is referred to throughout the paper as "soil solution." Bags were replaced once after installation for a total of two measurement periods spanning the entire length of the study from 12 May to 24 October 2015. Upon collection from the field, ion exchange resin from each resin bag was extracted and analyzed in the same manner as ion exchange resin in atmospheric collectors. Concentrations of total N, organic C, and total P in extract solution were converted to daily rates of total N, organic C, and total P per gram resin by multiplying the solution concentration by the total volume of KCl used to extract samples (150 mL) and dividing by the amount of resin in each resin bag (10 g). Values for soil solution total N, organic C, and total P were averaged across the six replicates at each site, summed over the season, and divided by days in the field (unique to each site, Table S1 in Supporting Information S1) to provide a daily rate per site. For samples where the total N or P were found to be less than inorganic N or P (12 out of 162 samples, average negative difference = 6.8%), we set organic N or P to zero, resulting in total N or P being equivalent to inorganic N or P.

2.4. Statistical Analyses

All statistical analyses were conducted in RStudio (The R Project for Statistical Computing, version 1.0.136). All error values reported are given in standard error (SE). Shapiro–Wilk tests and quantile–quantile plots were used to test for normal distribution of data and equal variance. Linear mixed effects model ANOVAs were used to examine time period differences in NH_4^+, NO_3^-, organic N, organic C, and inorganic and organic P in atmospheric inputs and in soil solution with time as the fixed effect and site as the random effect. A Tukey's Honestly Significant Difference post hoc test was conducted after the mixed effects model to determine which fluxes were statistically significantly different from one another across time periods. For normally distributed data, a Welch's *t*-test was conducted to test the difference between growing season throughfall and bulk inputs and the difference in soil solution beneath bulk and throughfall collectors for all analytes. For non-normally distributed data, a Wilcoxon rank sum test was conducted to test the

difference between annualized throughfall and bulk inputs and the difference in soil solution beneath bulk and throughfall collectors. Statistical significance was determined at $\alpha \leq 0.05$. As there were four different tree genera across each of the five throughfall sites, the sample size for each genus was too small to note species effects on throughfall; however, Decina et al. (2017) found no tree species effect on rates of throughfall for sites around the greater Boston area. Finally, to remove outliers, 9 out of 1165 values for atmospheric inputs and soil solution greater than four standard deviations from the mean for that sample period were replaced with average values for the site at which they were measured.

3. Results and Discussion

3.1. Atmospheric Bulk and Throughfall Inputs

Our results demonstrate that atmospheric deposition is an important source of organic N, organic C, and total P to urban ecosystems during the growing season. Supporting our first hypothesis, organic N makes up 38.4 ± 1.9% of total (inorganic + organic) bulk N inputs across study sites (calculated as the mean of organic N:total N ratios from individual sites; individual site data provided in the Table S1 in Supporting Information S1); taking into account inputs of organic N increases estimates of bulk inputs of total N by 61.5 ± 1.9% across study sites (Table S1 in Supporting Information S1). This substantive fraction of organic N as a percentage of total N agrees with mean literature values of 35% across 58 studies in mainly rural sites in North America (Cornell, 2011), but is considerably larger than percentages in the few U.S. cities in which it has been reported (range: 7%–17.2%; Nadim et al., 2001, Zamora et al., 2011). Additionally, bulk inputs of inorganic N (5.23 ± 1.39 kg N ha^{-1} a^{-1}; Table 2) are similar to values of bulk inputs in New York City (5.53 kg N ha^{-1} a^{-1}; Lovett et al., 2000) and near Baltimore, MD (roughly 6.5 kg N ha^{-1} a^{-1}; Bettez & Groffman, 2013). Corroborating our second hypothesis, the annualized rate of organic C inputs in bulk deposition (18.32 ± 2.98 kg C ha^{-1} a^{-1}; Table 2) is comparable to values reported in seven studies in mainly rural sites in North America (21 ± 19 kg C ha^{-1} a^{-1}; Iavorivska et al., 2016) and is in the middle of the range of values from four studies measuring organic C deposition in U.S. cities (range: 8–42 kg C ha^{-1} a^{-1}; Likens & Galloway, 1983; Velinsky et al., 1986; Willey et al., 2000; Lohse et al., 2008). Contrary to our third hypothesis, the annualized rate of bulk atmospheric inputs of total P (0.77 ± 0.26 kg P ha^{-1} a^{-1}, 86% inorganic P, 14% organic P; Table 2) is higher than mean bulk input rates of total P reported across 38 studies from mostly rural sites in North America (0.42 kg ± 0.39 P ha^{-1} a^{-1}; Tipping et al., 2014) and higher than total P measured in precipitation in the few U.S. cities in which it has been reported (range: 0.05–0.56 kg P ha^{-1} a^{-1}; Eisenreich et al., 1977; Koelliker et al., 2004).

Similar to bulk deposition, we found that organic N makes up 35.3 ± 2.9% of total throughfall N inputs across study sites (Table S1 in Supporting Information S1); taking into account organic N increases estimates of total N inputs by 64.7 ± 2.9% across study sites (Table S1 in Supporting Information S1), a result similar to two urban studies in Brazil and Spain (de Souza et al., 2015; Izquieta-Rojano et al., 2016). We did not find any studies conducted in U.S. cities that measured organic N in throughfall to which we could compare our results. Throughfall inputs of inorganic N (Table 2; 14.90 ± 2.40 and 9.96 ± 3.70 kg N ha^{-1} a^{-1}, including and excluding T1, respectively) are similar to values of throughfall inputs measured in three previous Boston-area studies (14.9, 14.8 ± 3.48, and 8.70 ± 0.68 kg N ha^{-1} a^{-1}; Templer & McCann, 2010; Rao et al., 2014; Decina et al., 2017). Our annualized rates of throughfall organic C (Table 2; 94.46 ± 10.39 and 63.31 ± 15.43 kg C ha^{-1} a^{-1}, including and excluding T1, respectively) are in the middle of the range of values found across numerous rural studies (range: 29–161 kg C ha^{-1} a^{-1}: Michalzik et al., 2001; Schmidt et al., 2010; Pitman et al., 2010; Fujii et al., 2011; Arisci et al., 2012), but to our knowledge, no study reports organic C in throughfall in cities around the world. Annualized rates of throughfall total P (5.21 ± 0.79 and 2.60 ± 0.67 kg P ha^{-1} a^{-1}, including and excluding T1, respectively, 93–94% inorganic P; Table 2) are elevated compared to throughfall rates found in nonurban studies (range 0.34–2.7 kg P ha^{-1} a^{-1}: Kopáček et al., 2011; Parron et al., 2011; Coble & Hart, 2013; Runyan et al., 2013; Lequy et al., 2014; Du et al., 2016; Salehi et al., 2016), but as with organic C, we did not find any past urban studies measuring P in throughfall worldwide. Taken as a whole, these results suggest that atmospheric deposition can be an important nonpoint source of organic N, organic C, and total P during the growing season in urban areas.

Additionally, we find that our measured rates of bulk and throughfall inputs of inorganic P are higher than inorganic P in sewage effluent for the same period as our study. Details of the following calculations are provided in the Supporting Information S1, but we explain them here briefly. Using our measured input

Table 2.
Rates of bulk and throughfall N, P, and C inputs and N, P, and C in soil solution beneath bulk and throughfall collectors measured throughout the 2015 growing season

	Growing season inputs (kg ha^{-1})		Annualized inputs (kg ha^{-1} a^{-1})			Soil solution (μg g resin^{-1} d^{-1})	
	Bulk	Throughfall	Bulk	Throughfall including T1	Throughfall excluding T1	Bulk	Throughfall
NH_4^+-N	1.80 ± 0.62	5.29 ± 1.04	4.01 ± 1.39	11.77 ± 2.29	7.11 ± 3.37	0.58 ± 0.18	0.62 ± 0.14
NO_3^--N	0.55 ± 0.01	1.40 ± 0.49	1.22 ± 0.02	3.13 ± 1.08	2.85 ± 0.81	0.98 ± 0.12	1.45 ± 0.52
Inorganic N	2.35 ± 0.62	6.69 ± 1.10	5.23 ± 1.39	14.90 ± 2.40	9.96 ± 3.70	1.56 ± 0.27	2.07 ± 0.42
Organic N	1.64 ± 0.50	3.59 ± 0.51	3.65 ± 1.12	7.99 ± 1.13	4.72 ± 1.71	0.49 ± 0.11	0.32 ± 0.10
Total N	3.99 ± 1.11	10.28 ± 1.48	8.88 ± 2.49	22.89 ± 3.25	14.68 ± 5.31	2.05 ± 0.37	2.39 ± 0.36
Inorganic P	0.30 ± 0.11	2.17 ± 0.36	0.66 ± 0.24	4.83 ± 0.80	2.45 ± 0.65	0.03 ± 0.01	0.03 ± 0.01
Organic P	0.05 ± 0.01	0.17 ± 0.03	0.11 ± 0.02	0.38 ± 0.06	0.14 ± 0.03	0.07 ± 0.05	0.05 ± 0.02
Total P	0.35 ± 0.11	2.34 ± 0.36	0.77 ± 0.26	5.21 ± 0.79	2.60 ± 0.67	0.10 ± 0.06	0.07 ± 0.03
Organic C	8.22 ± 1.33	42.41 ± 4.72	18.32 ± 2.98	94.46 ± 10.39	63.31 ± 15.43	5.24 ± 1.38	5.29 ± 0.93

Values are means with standard error. Input values are presented as cumulative inputs measured across the growing season and yearly inputs, which extrapolate growing season rates across the entire year. Soil solution values are presented in mean daily values measured across the growing season. Inorganic N is the sum of NH_4^+-N and NO_3^--N, total N is the sum of inorganic N and organic N, and total P is the sum of inorganic P and organic P.

rates and a land area for the City of Boston of 125 km^2 with 25.5% canopy cover (data from 2006 to 2007, Raciti et al., 2014), we calculate that the growing season flux of inorganic P from bulk + throughfall inputs to the City of Boston is 10,719 kg P over the 166 days of the study period (9711 kg inorganic P and 1008 kg organic P). This value for inorganic P represents 123% of the inorganic P input to Boston Harbor (7895 kg P) derived from sewage effluent during our study period (http://www.mwra.com/02org/html/whatis.htm; Maguire & Fulweiler, 2017). Sewage effluent is responsible for the majority of P inputs to surface waters in the continental U.S. (Mainstone & Parr, 2002; Maupin & Ivahnenko, 2011), and inorganic P has been shown to compose between 85% and 95% of total P in sewage effluent in nearby Narragansett Bay, Rhode Island (Nixon et al., 1995; Nixon et al., 2008). By comparing our bulk and throughfall inputs of P to this known, important source of P, we demonstrate that bulk deposition and throughfall are also important sources of P to nearby aquatic ecosystems.

3.2. Canopy Influence on Inputs to Urban Ecosystems

Substantiating our fourth hypothesis, comparing bulk and throughfall inputs indicates that the urban tree canopy enhances growing season nutrient inputs to urban ecosystems. Rates of throughfall inputs were statistically significantly greater than bulk inputs for all analytes (growing season values, $p < 0.05$), likely due to a combination of the capture of dry deposition on canopy surfaces (Lovett & Lindberg, 1993), numerous canopy processes (Reiners & Olson, 1984; Ferm, 1993; Garten et al., 1998), and production of biological matter (Kopáček et al., 2011; Osono et al., 2006; Pedersen & Bille-Hansen, 1995). In urban areas with extensive canopy cover, elevated throughfall inputs may thus represent a substantial, concentrated input of nutrients to the ground surface in these ecosystems. In the case of N, these rates of throughfall inputs (22.89 ± 3.25 and 14.68 ± 5.31 kg N ha^{-1} a^{-1}, including and excluding T1, respectively; Table 2) are well above the critical load threshold of 3 kg N ha^{-1} a^{-1} in eastern temperate and northern forests of the Northeastern U.S. (Pardo et al., 2011). Considering these rates and Boston's 25.5% canopy cover (Raciti et al., 2014), inputs through the tree canopy roughly double the amount of total N and triple the amount of total P inputs to the 125 km^2 area of the City of Boston over the study period as compared to bulk inputs alone. Though we do not have direct canopy density or canopy cover measurements for each of our study sites, the City of Boston's canopy

cover is similar to mean canopy cover in urban areas across the U.S. (27.1%; Nowak & Crane, 2002). With significant urban tree cover and efforts in many cities to enhance canopy cover for a variety of management goals, including mitigation of the urban heat island effect, C capture, improved water quality, and beautification (Young, 2011), our results show that it is important for cities to take into account how tree canopies magnify inputs to ground surfaces and potentially nearby waterways.

Past studies have shown that urban soils have the capacity to process a significant portion of nutrient inputs (Groffman et al., 2004; McPhillips et al., 2016; Wollheim et al., 2005), thus decreasing export of nutrients into nearby waterways via internal cycling and uptake, as well as gas loss to the atmosphere. However, unlike in forests, tree canopy in urban areas is often located above pavement, rather than soils. By overlaying a high-resolution impervious surface area data layer (1-m resolution; Massachusetts Office of Geographic Information (MassGIS), 2009) with a tree canopy data layer (~1-m resolution; Raciti et al., 2014) for the City of Boston and summing areas where impervious surface and tree canopy intersect, we calculated that 26.1% of the tree canopy in Boston is located above impervious surfaces. Considering our mass flux calculation of growing season throughfall inputs described above, approximately 1805 kg of inorganic P in throughfall was deposited directly onto impervious surfaces underneath tree canopy in the City of Boston over the 166 days of our study, an amount equivalent to 23% of inorganic P in sewage effluent (details of this calculation are provided in the Supporting Information S1). Impervious surfaces increase the amount of runoff (Arnold & Gibbons, 1996); thus inputs above impervious surfaces are more susceptible to flowing directly into waterways without being processed in soils, representing a substantial input of nutrients, particularly of P, to urban waterways (Hobbie et al., 2017). The urban tree canopy therefore not only amplifies rates of atmospheric inputs, but because a significant proportion of it is located above paved ground, may also represent a large flux of nutrients to urban waterways through runoff if nutrients are not retained in other permeable surfaces.

We present a conceptual diagram (Figure 2) of the potential fates of elements deposited beneath the urban tree canopy located above permeable and impervious surfaces. When inputs of elements are deposited through a tree canopy onto permeable surfaces, runoff likely decreases and the residence time of the element increases, providing the opportunity for an element to be recycled, taken up by vegetation or microbes, and retained within the ecosystem (Wollheim et al., 2005). Additionally, these inputs are able to remain in the soil long enough to be lost in gaseous form, and a smaller fraction is lost via leaching to waterways (McPhillips et al., 2016). Conversely, inputs deposited from a tree onto an impervious surface like pavement may be quickly lost to runoff, providing little opportunity or time for internal cycling or gaseous loss (Wollheim et al., 2005). While vegetation provides numerous ecological (Bowler et al., 2010; Nowak et al., 2013) and social (Alcock et al., 2014; Branas et al., 2011) benefits in urban areas (though see Vos et al., 2013 and Lahr et al., 2015 for a different perspective), our results highlight the complex tradeoffs of planting urban trees (Bodnaruk et al., 2017) and suggest that trees should be planted above soils, rather than pavement in order to take advantage of the potential nutrient retention capabilities of urban soils (Groffman et al., 2004; Wollheim et al., 2005) and to minimize the loading of nutrients to waterways that come from throughfall.

3.3. Elevated Throughfall Inputs in Late Spring

Throughfall inputs during the late spring sampling period (T1: 12–13 May –22 June 2015) were two to eight times greater than throughfall during the rest of the growing season, depending on the analyte (all but NO_3^-; Figure 1), a result sustained even after correcting for precipitation differences across the four time periods. The pattern of higher late spring throughfall inputs has been found in other urban studies (Ayars & Gao, 2007; Varenik et al., 2015; Izquieta-Rojano et al., 2016; Decina et al., 2017), and is possibly due to a combination of factors that may be elevated in the late spring, such as pollen deposits, microbial activity, deposition of aphid honeydew and insect frass, as well as fertilizer that has aerosolized or volatilized and deposited onto canopy surfaces (Bettez & Groffman, 2013; Kopáček et al., 2011; le Mellec et al., 2011; Stadler & Michalzik, 1998; Templer et al., 2015a, 2015b). This spring pulse of inputs is likely to come from the aforementioned sources rather than foliar leaching itself, as it has been shown that mature leaves (those present later in the growing season) are more susceptible to nutrient loss than are young leaves (Hutchinson & Havas, 1980; Potter, 1991). However, as many tree canopies in rural areas are net sinks for (or take up) nutrients (Piirainen et al., 1998; Zimmermann et al., 2006), the precise processes controlling elevated spring

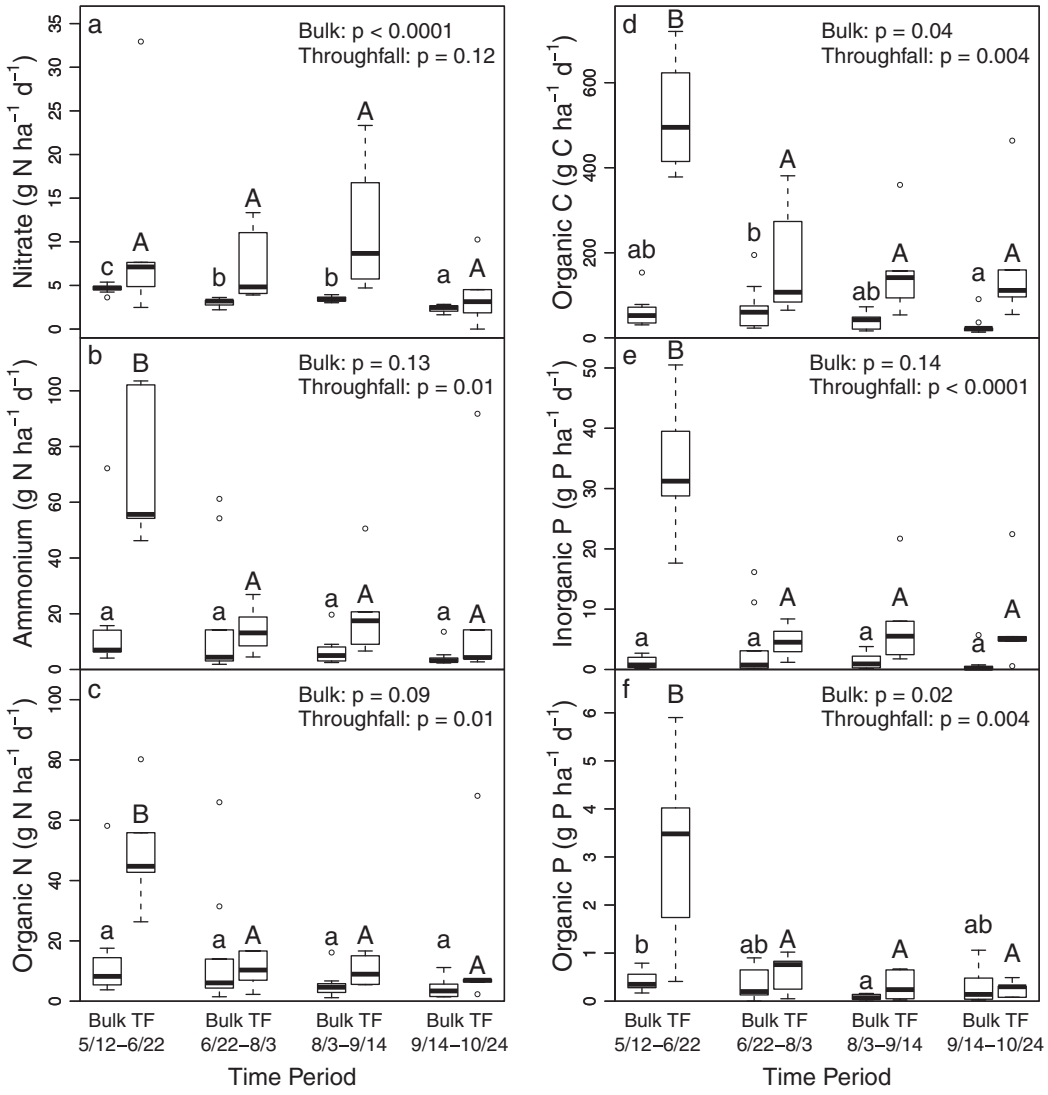

Figure 1. Daily rates of bulk and throughfall (TF) inputs of (a) nitrate, (b) ammonium, (c) organic nitrogen, (d) organic carbon, (e) inorganic phosphorus, and (f) organic phosphorus over the four time periods of the study. Boxes represent interquartile range (IQR) of the data, black line within boxes represents median values for each group, whiskers represent IQR × 1.5, open circles represent individual values that lie without the IQR × 1.5. Distinct letters represent statistically significant differences among rates of bulk deposition (lowercase letters) across the four time periods, and among TF inputs (uppercase letters) across the four time periods with associated *p* values in the upper right corner of each plot (linear mixed effects model analysis of variance, Tukey's Honestly Significant Difference posthoc test).

inputs in urban areas is unknown and requires further study. Regardless of source, however, this late spring pulse of nutrients represents a large flux of N, C, and P to the urban ground surface.

Considering that the late spring pulse may not be representative of the entire growing season, we calculated annual rates of deposition both with and without this sampling period; excluding the late spring throughfall value reduces annualized rates of throughfall inputs from 33% to 63% for each analyte with the exception of NO_3^-, which only is reduced about 9% by excluding the late spring period (Table 2). Excluding the late spring sampling period, throughfall remained greater than bulk inputs for all analytes, but was significantly higher during the T1 sampling for inorganic P and organic C only ($p = .04$ for organic C and $p = .02$ for inorganic P).

4. Conclusions

While a number of studies have reported large enhancements in inorganic N deposition in urban areas compared to nearby rural areas (Fang et al., 2011; Lovett et al., 2000; Rao et al., 2014; Templer & McCann,

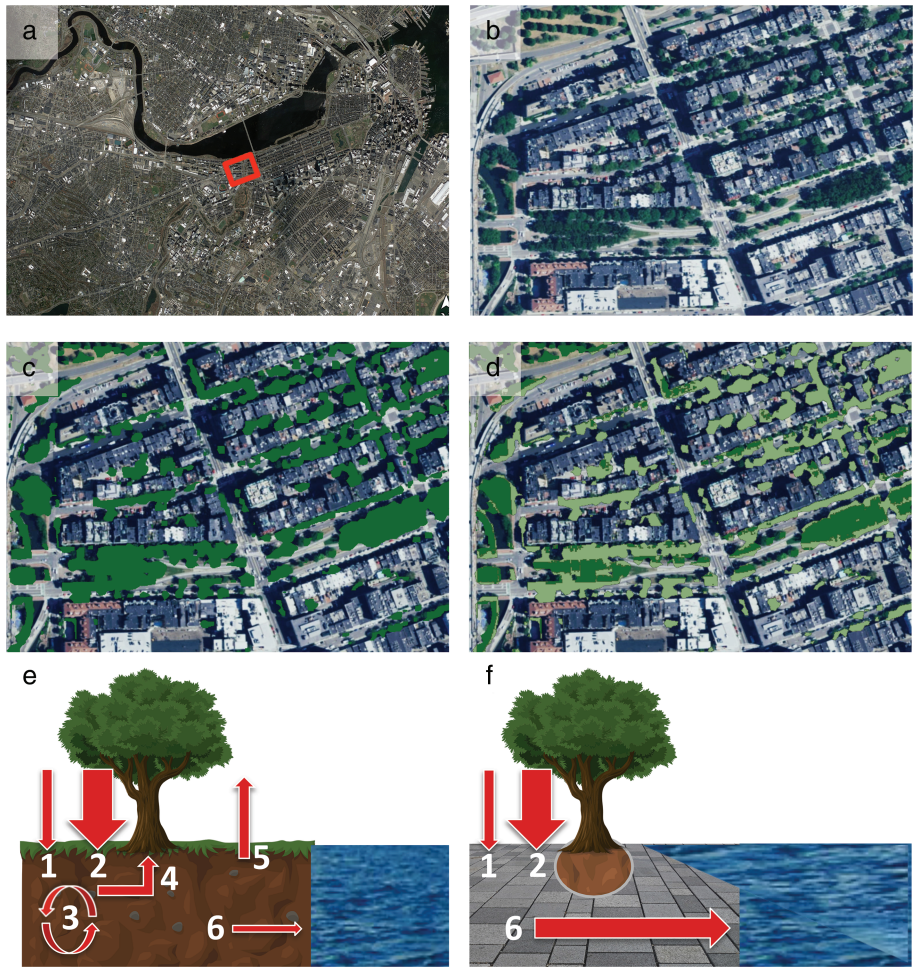

Figure 2. Conceptual diagram hypothesizing the relationship between atmospheric inputs, urban tree canopy, impervious surface area, and nutrient fluxes to waterways and the atmosphere. (a) Image of Boston with red box around the area whose aerial image is shown in panels (b), (c), and (d). (b) Aerial image of a section of the neighborhood containing sites 1, 2, 8, and 9. (c) The same aerial image from panel (b) with all canopy cover colored dark green. (d) The same aerial image from panel (c) with canopy cover above soil colored dark green and above impervious surface colored light green. (e) The potential fate of atmospheric inputs above soil. (f) The potential fate of atmospheric inputs above pavement. Arrows represent the following fluxes: (1) bulk deposition, (2) throughfall inputs, (3) nutrient cycling in soils, (4) uptake by vegetation, (5) gaseous losses to the atmosphere, and (6) losses to waterways through runoff. Inputs received over impervious surface have a higher potential to runoff directly into nearby waterways, while a portion of inputs received over soils are cycled within soil and vegetation and/or lost as gas, with a much smaller fraction likely lost to nearby waterways.

2010), there are no studies to our knowledge that simultaneously measure atmospheric bulk and throughfall inputs of organic and inorganic N, organic C, and organic and inorganic P. This study demonstrates that organic N makes up a substantial component of urban atmospheric N inputs, that atmospheric inputs of organic C are on par with inputs in rural areas, and that rates of urban inorganic P inputs are higher than rates in rural areas. Further, the input of inorganic P in bulk deposition and throughfall is 1.2 times higher than that in sewage effluent , a major source of P, for the City of Boston over the study period. We also find that throughfall inputs of total N, organic C, and total P are higher than bulk inputs, suggesting that the tree canopy amplifies inputs to urban ground surfaces and, as urban tree canopy is often located above impervious surfaces, potentially to waterways through runoff. Together, these findings suggest that atmospheric inputs in cities contribute substantially to urban biogeochemical budgets and that cities should consider planting trees above surfaces where precipitation can infiltrate soils to minimize loading of nutrient inputs to waterways.

As the urban population continues to grow both around the world (United Nations, Department of Economic and Social Affairs, Population Division, 2015) and in the U.S. (U.S. Environmental Protection Agency,

2014), finding effective ways of monitoring and mitigating anthropogenic pollution and providing ecosystem services in cities is essential. Our results indicate that long-term monitoring of inorganic and organic inputs of N, C, and P to urban systems is crucial for providing clean air and clean water for urban residents. As most of the world's population resides in urban areas, municipal policies that address and accurately manage pollutant inputs have an ever-increasing ability to improve the health and well-being of the majority of the world's citizens. Finally, as cities pursue greening efforts in a push to make their environments more livable, we suggest that they consider the suite of trade-offs associated with planting urban trees in order to pursue these efforts in ways that maximize the services provided by urban vegetation.

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References

Alcock, I., White, M. P., Wheeler, B. W., Fleming, L. E., & Depledge, M. H. (2014). Longitudinal effects on mental health of moving to greener and less green urban areas. *Environmental Science & Technology*, *48*, 1247–1255. https://doi.org/10.1021/es403688w

Araujo, T. G., Souza, M. F. L., De Mello, W. Z., & Da Silva, D. M. L. (2015). Bulk atmospheric deposition of major ions and dissolved organic nitrogen in the lower course of a tropical river basin, southern Bahia, Brazil. *Journal of the Brazilian Chemical Society*, *26*(8), 1692–1701. https://doi.org/10.5935/0103-5053.20150143.

Arisci, S., Rogora, M., Marchetto, A., & Dichiaro, F. (2012). The role of forest type in the variability of DOC in atmospheric deposition at forest plots in Italy. *Environmental Monitoring and Assessment*, *184*, 3415–3425. https://doi.org/10.1007/s10661-011-2196-2

Arnold Jr., C. L., & Gibbons, C. J. (1996). Impervious surface coverage: The emergence of a key environmental indicator. *Journal of the American Planning Association*, *62*(2), 243–258. https://doi.org/10.1080/01944369608975688

Ayars, J., & Gao, Y. (1997). Atmospheric nitrogen deposition to the Mullica River-Great Bay Estuary. *Marine Environmental Research*, *64*, 590–600. https://doi.org/10.1016/j.marenvres.2007.06.004

Balestrini, R., & Tagliaferri, A. (2001). Atmospheric deposition and canopy exchange processes in alpine forest ecosystems (northern Italy). *Atmospheric Environment*, *35*(36), 6421–6433. https://doi.org/10.1016/S1352-2310(01)00350-8

Bealey, W. J., McDonald, A. G., Nemitz, E., Donovan, R., & Dragosits, U. (2007). Estimating the reduction of urban PM10 concentrations by trees within an environmental information system for planners. *Journal of Environmental Management*, *85*, 44–58. https://doi.org/10.1016/j.jenvman.2006.07.007

Bettez, N. D., & Groffman, P. M. (2013). Nitrogen deposition in and near an urban ecosystem. *Environmental Science & Technology*, *47*, 6047–6051. https://doi.org/10.1021/es400664b

Bodnaruk, E. W., Kroll, C. N., Yang, Y., Hirabayashi, S., Nowak, D. J., & Endreny, D. T. A. (2017). Where to plant urban trees? A spatially explicit methodology to explore ecosystem service tradeoffs. *Landscape and Urban Planning*, *157*, 457–467. https://doi.org/10.1016/j.landurbplan.2016.08.016

Bowler, D. E., Buyung-ali, L., Knight, T. M., & Pullin, A. S. (2010). Urban greening to cool towns and cities: A systematic review of the empirical evidence. *Landscape and Urban Planning*, *97*(3), 147–155. https://doi.org/10.1016/j.landurbplan.2010.05.006

Boyer, K. B., & Kieser, M. S. (2012). Urban stormwater management – An MS4 success story for Western Michigan University. *Journal of Green Building*, *7*(1), 28–39. https://doi.org/10.3992/jgb.7.1.28

Brack, C. L. (2002). Pollution mitigation and carbon sequestration by an urban forest. *Environmental Pollution*, *116*, S195–S200. https://doi.org/10.1016/S0269-7491(01)00251-2.

Branas, C. C., Cheney, R. A., MacDonald, J. M., Tam, V. W., Jackson, T. D., & Ten Have, T. R. (2011). A difference-in-differences analysis of health, safety, and greening vacant urban space. *American Journal of Epidemiology*, *174*(11), 1296–1306. https://doi.org/10.1093/aje/kwr273

Brezonik, P. L., Hendry Jr. C. D., Edgerton, E. S., Schulze, R. L., & Crisman, T. L. (1983). Acidity, nutrients and minerals in atmospheric precipitation over Florida: Deposition patterns, mechanisms and ecological effects. (Rep. EPA-600/S3-83-004). United States Environmental Protection Agency Project Summary.

Burton Jr., G. A., & Pitt, R. E. (2002). *Stormwater Effects Handbook*. Boca Raton, FL: Lewis Publishers.

Cape, J. N., Cornell, S. E., Jickells, T. D., & Nemitz, E. (2011). Organic nitrogen in the atmosphere – Where does it come from? A review of sources and methods. *Atmospheric Research*, *102*(1–2), 30–48. https://doi.org/10.1016/j.atmosres.2011.07.009.

Carpenter, S. R., Caraco, N. F., Correll, D. L., Howarth, R. W., Sharpley, A. N., & Smith, V. H. (1998). Nonpoint pollution of surface waters with phosphorus and nitrogen. *Ecological Applications*, *8*(3), 559–568. https://doi.org/10.1890/1051-0761(1998)008[0559:NPOSWW]2.0.CO;2

Chen, Y. X., Chen, H. Y., Wang, W., Yeh, J. X., Chou, W. C., Gong, G. C., … Lin, C. T. (2015). Dissolved organic nitrogen in wet deposition in a coastal city (Keelung) of the southern East China Sea: Origin, molecular composition and flux. *Atmospheric Environment*, *112*, 20–31. https://doi.org/10.1016/j.atmosenv.2015.04.023

Coble, A. A., & Hart, S. C. (2013). The significance of atmospheric nutrient inputs and canopy interception of precipitation during ecosystem development in piñon-juniper woodlands of the southwestern USA. *Journal of Arid Environments*, *98*, 79–87. https://doi.org/10.1016/j.jaridenv.2013.08.002

Cornell, S. E. (2011). Atmospheric nitrogen deposition: Revisiting the question of the importance of the organic component. *Environmental Pollution*, *159*(10), 2214–2222. https://doi.org/10.1016/j.envpol.2010.11.014

D'Angelo, E., Crutchfield, J., & Vandiviere, M. (2001). Rapid, sensitive, microscale determination of phosphate in water and soil. *Journal of Environmental Quality*, *30*(6), 2206–2209. https://doi.org/10.2134/jeq2001.2206

De Borba, B. M., Jack, R. F., Rohrer, J. S., Wirt, J., & Wang, D. (2014). Simultaneous determination of total nitrogen and total phosphorus in environmental waters using alkaline persulfate digestion and ion chromatography. *Journal of Chromatography. A*, *1369*, 131–137. https://doi.org/10.1016/j.chroma.2014.10.027

de Souza, P. A., Ponette-González, A. G., de Mello, W. Z., Weathers, K. C., & Santos, I. A. (2015). Atmospheric organic and inorganic nitrogen inputs to coastal urban and montane Atlantic Forest sites in southeastern Brazil. *Atmospheric Research*, *160*, 126–137. https://doi.org/10.1016/j.atmosres.2015.03.011

Decina, S. M., Templer, P. H., & Hutyra, L. R. (2017). Variability, drivers, and effects of atmospheric nitrogen inputs across an urban area: Emerging patterns among human activities, the atmosphere and soils. *Science of the Total Environment*, *609*, 1524–1534. https://doi.org/10.1016/j.scitotenv.2017.07.166

Doane, T. A., & Horwath, W. R. (2003). Spectrophotometric determination of nitrate with a single reagent. *Analytical Letters*, *36*(12), 2713–2722. https://doi.org/10.1081/AL-120024647

Doyle, A., Weintraub, M. N., & Schimel, J. P. (2004). Persulfate digestion and simultaneous colorimetric analysis of carbon and nitrogen in soil extracts. *Soil Science Society of America Journal*, *68*, 669–676. https://doi.org/10.2136/sssaj2004.6690

Du, E., de Vries, W., Han, W., Liu, X., Yan, Z., & Jiang, Y. (2016). Imbalanced phosphorus and nitrogen deposition in China's forests. *Atmospheric Chemistry and Physics*, *16*, 8571–8579. https://doi.org/10.5194/acp-16-8571-2016

Eisenreich, S. J., Emmling, P. J., & Beeton, A. M. (1977). Atmospheric loading of phosphorus and other chemicals to Lake Michigan. *Journal of Great Lakes Research*, *3*(3–4, 304), 291–304. https://doi.org/10.1016/S0380-1330(77)72261-0.

Fang, Y., et al. (2011). Nitrogen deposition and forest nitrogen cycling along an urban-rural transect in southern China. *Global Change Biology*, *17*, 872–885. https://doi.org/10.1111/j.1365-2486.2010.02283.x

Fenn, M. E., Ross, C. S., Schilling, S. L., Baccus, W. D., Larrabee, M. A., & Lofgren, R. A. (2013). Atmospheric deposition of nitrogen and sulfur and preferential canopy consumption of nitrate in forests of the Pacific Northwest, USA. *Forest Ecology and Management*, *302*, 240–253. https://doi.org/10.1016/j.foreco.2013.03.042

Ferm, M. (1993). Throughfall measurements of nitrogen and sulphur compounds. *International Journal of Analytical Chemistry*, *50*, 29–43. https://doi.org/10.1080/03067319308027581

Fujii, K., Hartono, A., Funakawa, S., Uemura, M., & Kosaki, T. (2011). Fluxes of dissolved organic carbon in three tropical secondary forests developed on serpentine and mudstone. *Geoderma*, *163*(1–2), 119–126. https://doi.org/10.1016/j.geoderma.2011.04.012.

Garten Jr., C. T., Schwab, A. B., & Shirshac, T. L. (1998). Foliar retention of ^{15}N tracers: Implications for net canopy exchange in low- and high-elevation forest ecosystems. *Forest Ecology and Management*, *103*(2–3), 211–216. https://doi.org/10.1016/S0378-1127(97)00185-0.

González-Benitez, J. M., Cape, J. N., Heal, M. R., Van Dijk, N., & Vidal Diez, A. (2009). Atmospheric nitrogen deposition in south-east Scotland: Quantification of the organic nitrogen fraction in wet, dry and bulk deposition. *Atmospheric Environment*, *43*, 4087–4094. https://doi.org/10.1016/j.atmosenv.2009.04.061

Groffman, P. M., Law, N. L., Belt, K. T., Band, L. E., & Fisher, G. T. (2004). Nitrogen fluxes and retention in urban watershed ecosystems. *Ecosystems*, *7*, 393–403. https://doi.org/10.1007/s10021-003-0039-x

Hall, S. J., Ogata, E. M., Weintraub, S. R., Baker, M. A., Ehleringer, J. R., Czimczik, C. I., & Bowling, D. R. (2016). Convergence in nitrogen deposition and cryptic isotopic variation across urban and agricultural valleys in northern Utah. *Journal of Geophysical Research – Biogeosciences*, *121*(9), 2340–2355. https://doi.org/10.1002/2016JG003354

He, J., Balasubramanian, R., Burger, D. F., Hicks, K., Kuylenstierna, J. C. I., & Palani, S. (2011). Dry and wet atmospheric deposition of nitrogen and phosphorus in Singapore. *Atmospheric Environment*, *45*(16), 2760–2768. https://doi.org/10.1016/j.atmosenv.2011.02.036

Hobbie, S. E., Finlay, J. C., Janke, B. D., Nidzgorski, D. A., Millet, D. B., & Baker, L. A. (2017). Contrasting nitrogen and phosphorus budgets in urban watersheds and implications for managing urban water pollution. *Proceedings of the National Academy of Sciences USA*, *114*(16), 4177–4182. https://doi.org/10.1073/pnas.1618536114

Homer, C., et al. (2015). Completion of the 2011 national land cover database for the conterminous United States – Representing a decade of land cover change information. *Photogrammetric Engineering and Remote Sensing*, *81*(5), 345–354. https://doi.org/10.14358/PERS.81.5.345

Hou, P., Ren, Y., Zhang, Q., Lu, F., Ouyang, Z., & Wang, X. (2012). Nitrogen and phosphorus in atmospheric deposition and roof runoff. *Polish Journal of Environmental Studies*, *21*(6), 1621–1627.

Huang, X., Li, X., He, L., Feng, N., Hu, M., Niu, Y., & Zeng, L. (2010). 5-year study of rainwater chemistry in a coastal mega-city in South China. *Atmospheric Research*, *97*(1–2), 185–193. https://doi.org/10.1016/j.atmosres.2010.03.027

Hutchinson, T. C., & Havas, M. (1980). *Effects of Acid Precipitation on Terrestrial Ecosystems*. New York, NY: Plenum Press. https://doi.org/10.1007/978-1-4613-3033-2

Iavorivska, L., Boyer, E. W., & Dewalle, D. R. (2016). Atmospheric deposition of organic carbon via precipitation. *Atmospheric Environment*, *146*, 153–163. https://doi.org/10.1016/j.atmosenv.2016.06.006

IPCC (2014). In R. K. Pachauri & L. A. Meyer (Eds.), *Climate Change 2014: Synthesis Report. Contribution of Working Groups I, II and III to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change* [Core Writing Team. Geneva, Switzerland: IPCC 151 pp.

Izquieta-Rojano, S., García-Gomez, H., Aguillaume, L., Santamaría, J. M., Tang, Y. S., Santamaría, C., … Elustondo, D. (2016). Throughfall and bulk deposition of dissolved organic nitrogen to holm oak forests in the Iberian Peninsula: Flux estimation and identification of potential sources. *Environmental Pollution*, *210*, 104–112. https://doi.org/10.1016/j.envpol.2015.12.002

Juknys, R., Zaltauskaite, J., & Stakenas, V. (2007). Ion fluxes with bulk and throughfall deposition along an urban-suburban-rural gradient. *Water, Air, and Soil Pollution*, *178*(1–4), 363–372. https://doi.org/10.1007/s11270-006-9204-0

Keene, W. C., Montag, J. A., Maben, J. R., Southwell, M., Leonard, J., Church, T. M., … Galloway, J. N. (2002). Organic nitrogen in precipitation over eastern North America. *Atmospheric Environment*, *36*, 4529–4540. https://doi.org/10.1016/S1352-2310(02)00403-X

Kimura, S. D., Saito, M., Hara, H., & Xu, Y. H. (2009). Comparison of nitrogen dry deposition on cedar and oak leaves in the Tama Hills using foliar rinsing method. *Water, Air, and Soil Pollution*, *202*, 369–377. https://doi.org/10.1007/s11270-009-9983-1

Koelliker, Y., Totten, L. A., Gigliotti, C. L., Offenberg, J. H., Reinfelder, J. R., Zhuang, Y. A. N., & Eisenreich, S. J. (2004). Atmospheric wet deposition of total phosphorus in New Jersey. *Water, Air, and Soil Pollution*, *154*, 139–150. https://doi.org/10.1023/B:WATE.0000022952.12577.c5

Kopáček, J., Turek, J., Hejzlar, J., & Porcal, P. (2011). Bulk deposition and throughfall fluxes of elements in the Bohemian Forest (Central Europe) from 1998 to 2009. *Boreal Environmental Research*, *16*, 495–508.

Kopáček, J., Turek, J., Hejzlar, J., & Šantručková, H. (2009). Canopy leaching of nutrients and metals in a mountain spruce forest. *Atmospheric Environment*, *43*, 5443–5453. https://doi.org/10.1016/j.atmosenv.2009.07.031.

Lahr, E. C., Schade, G. W., Crossett, C. C., & Watson, M. R. (2015). Photosynthesis and isoprene emission from trees along an urban – Rural gradient in Texas. *Global Change Biology*, *21*, 4221–4236. https://doi.org/10.1111/gcb.13010

le Mellec, A., Gerold, G., & Michalzik, B. (2011). Insect herbivory, organic matter deposition and effects on belowground organic matter fluxes in a central European oak forest. *Plant and Soil*, *342*, 393–403. https://doi.org/10.1007/s11104-010-0704-8

Lequy, E., Calvaruso, C., Conil, S., & Turpault, M. (2014). Atmospheric particulate deposition in temperate deciduous forest ecosystems: Interactions with the canopy and nutrient inputs in two beech stands of northeastern France. *Science of the Total Environment*, *487*, 206–215. https://doi.org/10.1016/j.scitotenv.2014.04.028

Li, J., Fang, Y., Yoh, M., Wang, X., Wu, Z., Kuang, Y., & Wen, D. (2012). Organic nitrogen deposition in precipitation in metropolitan Guangzhou city of southern China. *Atmospheric Research*, *113*, 57–67. https://doi.org/10.1016/j.atmosres.2012.04.019

Li, Y., Schichtel, B. A., Walker, J. T., Schwede, D. B., Chen, X., Lehmann, C. M. B., … Collett Jr., J. L. (2016). Increasing importance of deposition of reduced nitrogen in the United States. *Proceedings of the National Academy of Sciences USA*, *113*(21), 5874–5879. https://doi.org/10.1073/pnas.1525736113

Likens, G. E., & Galloway, J. N. (1983). The composition and deposition of organic carbon in precipitation. *Tellus*, *35B*, 16–24. https://doi.org/10.1111/j.1600-0889.1983.tb00003.x

Lohse, K. A., Hope, D., Sponseller, R., Allen, J. O., & Grimm, N. B. (2008). Atmospheric deposition of carbon and nutrients across an arid metropolitan area. *Science of the Total Environment*, *402*(1), 95–105. https://doi.org/10.1016/j.scitotenv.2008.04.044

Lovett, G. M. (1994). Atmospheric deposition of nutrients and pollutants in North America: An ecological perspective. *Ecological Applications*, *4*(4), 629–650. https://doi.org/10.2307/1941997

Lovett, G. M., & Lindberg, S. E. (1993). Atmospheric deposition and canopy interactions of nitrogen in forests. *Canadian Journal of Forest Research*, *23*, 1603–1616. https://doi.org/10.1139/x93-200

Lovett, G. M., Traynor, M. M., Pouyat, R. V., Carreiro, M. M., Zhu, W. X., & Baxter, J. W. (2000). Atmospheric deposition to oak forests along an urban-rural gradient. *Environmental Science & Technology*, *34*, 4294–4300. https://doi.org/10.1021/es001077q

Luo, Y., Yang, X., Carley, R. J., & Perkins, C. (2002). Atmospheric deposition of nitrogen along the Connecticut coastline of Long Island sound: A decade of measurements. *Atmospheric Environment*, *36*(28), 4517–4528. https://doi.org/10.1016/S1352-2310(02)00421-1

Maguire, T. J., & Fulweiler, R. W. (2017). Fate and effect of dissolved silicon within wastewater treatment effluent. *Environmental Science & Technology*, *51*(13), 7403–7411. https://doi.org/10.1021/acs.est.7b01276

Mahowald, N., Jickells, T. D., Baker, A. R., Artaxo, P., Benitez-Nelson, C. R., Bergametti, G., … Tsukuda, S. (2008). Global distribution of atmospheric phosphorus sources, concentrations and deposition rates, and anthropogenic impacts. *Global Biogeochemical Cycles*, *22*, 1–19. https://doi.org/10.1029/2008GB003240

Mainstone, C. P., & Parr, W. (2002). Phosphorus in rivers – Ecology and management. *Science of the Total Environment*, *282–283*, 25–47. https://doi.org/10.1016/S0048-9697(01)00937-8

Massachusetts Office of Geographic Information (MassGIS). (2009). Land use, impervious surface, roads datalayers. http://www.mass.gov/anf/research-and-tech/it-serv-and-support/application-serv/office-of-geographic-information-massgis/.

Matsumoto, K., Yamamoto, Y., Kobayashi, H., Kaneyasu, N., & Nakano, T. (2014). Water-soluble organic nitrogen in the ambient aerosols and its contribution to the dry deposition of fixed nitrogen species in Japan. *Atmospheric Environment*, *95*, 334–343. https://doi.org/10.1016/j.atmosenv.2014.06.037

Maupin, M. A., & Ivahnenko, T. (2011). Nutrient loadings to streams of the continental United States from municipal and industrial effluent. *Journal of the American Water Resources Association*, *47*(5), 950–964. https://doi.org/10.1111/j.1752-1688.2011.00576.x.

McPhillips, L. E., Groffman, P. M., Schneider, R. L., & Walter, M. T. (2016). Nutrient cycling in grassed roadside ditches and lawns in a suburban watershed. *Journal of Environmental Quality*, *45*, 1901–1909. https://doi.org/10.2134/jeq2016.05.0178

Michalzik, B., Kalbitz, K., Park, J., & Solinger, S. (2001). Fluxes and concentrations of dissolved organic carbon and nitrogen – A synthesis for temperate forests. *Biogeochemistry*, *52*, 173–205. https://doi.org/10.1023/A:1006441620810.

Michopoulos, P., Baloutsos, G., Economou, A., Voulala, M., & Bourletsikas, A. (2007). Bulk and throughfall deposition chemistry in three different forest ecosystems. *Fresenius Environmental Bulletin*, *16*(1), 91–98. https://doi.org/10.1007/978-94-007-1363-5_16.

Miller, N. H. J. (1905). The amounts of nitrogen as ammonia and as nitric acid, and of chlorine in the rainwater collected at Rothamsted. *The Journal of Agricultural Science*, *1*(3), 280–303. https://doi.org/10.1017/S0021859600000320

Moore, J. W., Schindler, D. E., Scheuerell, M. D., & Smith, D. (2003). Lake eutrophication at the urban fringe, Seattle region, USA. *Ambio*, *32*(1), 13–18. https://doi.org/10.1579/0044-7447-32.1.13

Nadim, F., Trahiotis, M. M., Stapcinskaite, S., Perkins, C., Carley, R. J., Hoa, G. E., & Yan, X. (2001). Estimation of wet, dry and bulk deposition of atmospheric nitrogen in Connecticut. *Journal of Environmental Monitoring*, *3*(6), 671–680. https://doi.org/10.1039/b107008h

National Climatic Data Center. (2016). Climate data online: data tools. https://www.ncdc.noaa.gov/cdo-web/datatools/normals.

Nixon, S. W., Buckley, B. A., Granger, S. L., Harris, L. A., Oczkowski, A. J., Fulweiler, R. W., & Cole, L. W. (2008). Nitrogen and phosphorus inputs to Narragansett Bay: Past, present, and future. In A. Desbonnet, B. A. Costa-Pierce (Eds.), *Science for Ecosystem Based Management* (pp. 101–175). New York, NY: Springer-Verlag. https://doi.org/10.1007/978-0-387-35299-2_5

Nixon, S. W., Granger, S. L., & Nowicki, B. L. (1995). An assessment of the annual mass balance of carbon, nitrogen, and phosphorus in Narragansett Bay. *Biogeochemistry*, *31*(1), 15–61. https://doi.org/10.1007/BF00000805.

Nowak, D. J., & Crane, D. E. (2002). Carbon storage and sequestration by urban trees in the USA. *Environmental Pollution*, *116*(3), 381–389. https://doi.org/10.1016/S0269-7491(01)00214-7

Nowak, D. J., Hirabayashi, S., Bodine, A., & Hoehn, R. (2013). Modeled PM 2.5 removal by trees in ten U.S. cities and associated health effects. *Environmental Pollution*, *178*, 395–402. https://doi.org/10.1016/j.envpol.2013.03.050

Ormerod, S. J., Donald, A. P., & Brown, S. J. (1989). The influence of plantation forestry on the pH and aluminum concentration of upland welsh streams: A re-examination. *Environmental Pollution*, *62*, 47–62. https://doi.org/10.1016/0269-7491(89)90095-X

Osono, T., Hobara, S., Koba, K., & Kameda, K. (2006). Reduction of fungal growth and lignin decomposition in needle litter by avian excreta. *Soil Biology and Biochemistry*, *38*, 1623–1630. https://doi.org/10.1016/j.soilbio.2005.12.001

Paerl, H. W. (1997). Coastal eutrophication and harmful algal blooms: Importance of atmospheric deposition and groundwater as new nitrogen and other nutrient sources. *Limnology and Oceanography*, *42*(5, part 2), 1154–1165. https://doi.org/10.4319/lo.1997.42.5_part_2.1154

Pandey, J., Pandey, U., Singh, A. V., Tripathi, S., & Mishra, V. (2016). Atmospheric N and P deposition in the Ganges Basin. *Current Science India*, *110*(6), 974–976.

Pardo, L. H., Fenn, M. E., Goodale, C. L., Geiser, L. H., Driscoll, C. T., Allen, E. B., … Dennis, R. L. (2011). Effects of nitrogen deposition and empirical nitrogen critical loads for ecoregions of the United States. *Ecological Applications*, *21*(8), 3049–3082. https://doi.org/10.1890/10-2341.1

Parron, L. M., Bustamante, M. M. C., & Markewitz, D. (2011). Fluxes of nitrogen and phosphorus in a gallery forest in the Cerrado of central Brazil. *Biogeochemistry*, *105*, 89–104. https://doi.org/10.1007/s10533-010-9537-z

Pedersen, L. B., & Bille-Hansen, J. (1995). Effects of nitrogen load to the forest floor in Sitka spruce stands (*Picea sitchensis*) as affected by difference in deposition and spruce aphid infestations. *Water, Air, and Soil Pollution*, *85*(3), 1173–1178. https://doi.org/10.1007/BF00477140

Piirainen, S., Finer, L., & Starr, M. (1998). Canopy and soil retention of nitrogen deposition in a mixed boreal forest in eastern Finland. *Water, Air, and Soil Pollution*, *105*, 165–174. https://doi.org/10.1007/978-94-017-0906-4_16.

Pitman, R. M., Vanguelova, E. I., & Benham, S. E. (2010). The effects of phytophagous insects on water and soil nutrient concentrations and fluxes through forest stands of the level II monitoring network in the UK. *Science of the Total Environment*, *409*(1), 169–181. https://doi.org/10.1016/j.scitotenv.2010.09.029

Pollman, C. D., Landing, W. M., Perry Jr., J. J., & Fitzpatrick, T. (2002). Wet deposition of phosphorus in Florida. *Atmospheric Environment*, *36*, 2309–2318. https://doi.org/10.1016/S1352-2310(02)00199-1.

Ponette-González, A., Perroni, Y., Weathers, K. C., DeSouza, P., Garcia-Oliva, F., & DeMello, W. Z. (2017). Nitrogen cycling in tropical Atlantic Forest differing in exposure to urban atmospheric nitrogen deposition. *Plant and Soil*, *420*, 451–465. https://doi.org/10.1007/s11104-017-3421-8

Ponette-González, A. A. G., Weathers, K. C., & Curran, L. M. (2015). Tropical land-cover change alters biogeochemical inputs to ecosystems in a Mexican montane landscape. *Ecological Applications*, *20*(7), 1820–1837. https://doi.org/10.1890/09-1125.1

Potter, C. (1991). Nutrient leaching from *Acer rubrum* leaves by experimental acid rainfall. *Canadian Journal of Forest Research*, *21*, 222–229. https://doi.org/10.1139/x91-027

Qualls, R. G., Haines, B. L., & Swank, W. T. (1991). Fluxes of dissolved organic nutrients and humic substances in a deciduous forest. *Ecology*, *72*(1), 254–266. https://doi.org/10.2307/1938919

Raciti, S. M., Hutyra, L. R., & Newell, J. D. (2014). Mapping carbon storage in urban trees with multi-source remote sensing data: Relationships between biomass, land use, and demographics in Boston neighborhoods. *Science of the Total Environment*, *500-501C*, 72–83. https://doi.org/10.1016/j.scitotenv.2014.08.070

Rao, P., Hutyra, L. R., Raciti, S. M., & Templer, P. H. (2014). Atmospheric nitrogen inputs and losses along an urbanization gradient from Boston to Harvard Forest, MA. *Biogeochemistry*, *121*, 229–245. https://doi.org/10.1007/s10533-013-9861-1

Reiners, W. A., & Olson, R. K. (1984). Effects of canopy components on throughfall chemistry: An experimental analysis. *Oecologia*, *63*, 320–330. https://doi.org/10.1007/BF00390660

Reynolds, B., Neal, C., Hornung, M., & Stevens, P. A. (1986). Baseflow buffering of streamwater acidity in five mid-Wales catchments. *Journal of Hydrology*, *87*, 167–185. https://doi.org/10.1016/0022-1694(86)90121-6

Runyan, C. W., Odorico, P. D., Vandecar, K. L., Das, R., Schmook, B., & Lawrence, D. (2013). Positive feedbacks between phosphorus deposition and forest canopy trapping, evidence from southern Mexico. *Journal of Geophysical Research – Biogeosciences*, *118*, 1521–1531. https://doi.org/10.1002/2013JG002384

Sakugawa, H., & Kaplan, I. R. (1993). Measurements of H2O2, aldehydes, and organic acids in Los Angeles rainwater: Their sources and deposition rates. *Atmospheric Environment*, *27B*(2), 203–219. https://doi.org/10.1016/0957-1272(93)90006-R

Salehi, M., Amiri, G. Z., Attarod, P., Salehi, A., Brunner, I., Schleppi, P., & Thimonier, A. (2016). Seasonal variations of throughfall chemistry in pure and mixed stands of oriental beech (*Fagus orientalis* Lipsky) in Hyrcanian forests (Iran). *Annals of Forest Science*, *73*, 371–380. https://doi.org/10.1007/s13595-015-0525-2

Santos, S. M., Santos, E. B. H., & Duarte, A. C. (2014). Dissolved organic and inorganic matter in bulk deposition of a coastal urban area: An integrated approach. *Journal of Environmental Management*, *145*, 71–78. https://doi.org/10.1016/j.jenvman.2014.04.022

Schmidt, B. H. M., Wang, C. P., Chang, S. C., & Matzner, E. (2010). High precipitation causes large fluxes of dissolved organic carbon and nitrogen in a subtropical montane *Chamaecyparis* forest in Taiwan. *Biogeochemistry*, *101*, 243–256. https://doi.org/10.1007/s10533-010-9470-1

Sims, G. K., Ellsworth, T. R., & Mulvaney, R. L. (1995). Microscale determination of inorganic nitrogen in water and soil extracts. *Communications in Soil Science and Plant Analysis*, *26*(1&2), 303–316. https://doi.org/10.1080/00103629509369298

Siudek, P., Frankowski, M., & Siepak, J. (2015). Seasonal variations of dissolved organic carbon in precipitation over urban and forest sites in central Poland. *Environmental Science and Pollution Research*, *22*, 11087–11096. https://doi.org/10.1007/s11356-015-4356-3

Smil, V. (2000). Phosphorus in the environment: Natural flows and human interferences. *Annual Review of Energy and the Environment*, *25*, 53–88. https://doi.org/10.1146/annurev.energy.25.1.53

Sparks, J. P. (2009). Ecological ramifications of the direct foliar uptake of nitrogen. *Oecologia*, *159*, 1–13. https://doi.org/10.1007/s00442-008-1188-6

Stadler, B., & Michalzik, B. (1998). Linking aphid honeydew, throughfall, and forest floor solution chemistry of Norway spruce. *Ecology Letters*, *1*, 13–16. https://doi.org/10.1046/j.1461-0248.1998.00006.x

Sun, L., Liu, Y., Wang, J., Khalil, M. A. K., Zou, J., & Zhengqin, X. (2014). Atmospheric nitrogen and phosphorus deposition at three sites in Nanjing, China, and possible links to nitrogen deposition sources. *Clean: Soil, Air, Water*, *42*(11), 1650–1659. https://doi.org/10.1002/clen.201300692

Tallis, M., Taylor, G., Sinnett, D., & Freer-Smith, P. (2011). Estimating the removal of atmospheric particulate pollution by the urban tree canopy of London, under current and future environments. *Landscape and Urban Planning*, *103*(2), 129–138. https://doi.org/10.1016/j.landurbplan.2011.07.003

Templer, P. H., & McCann, T. M. (2010). Effects of the hemlock woolly adelgid on nitrogen losses from urban and rural northern forest ecosystems. *Ecosystems*, *13*, 1215–1226. https://doi.org/10.1007/s10021-010-9382-x

Templer, P. H., Weathers, K. C., Ewing, H. A., Dawson, T. E., Mambelli, S., Lindsey, A. M., … Firestone, M. K. (2015b). Fog as a source of nitrogen for redwood trees: Evidence from fluxes and stable isotopes. *Journal of Ecology*, *103*, 1397–1407. https://doi.org/10.1111/1365-2745.12462

Templer, P. H., Weathers, K. C., Lindsey, A., Lenoir, K., & Scott, L. (2015a). Atmospheric inputs and nitrogen saturation status in and adjacent to class I wilderness areas of the northeastern US. *Oecologia*, *177*(1), 5–15. https://doi.org/10.1007/s00442-014-3121-5

Tipping, E., Benham, S., Boyle, J. F., Crow, P., Davies, J., Fischer, U., … Toberman, H. (2014). Atmospheric deposition of phosphorus to land and freshwater. *Environmental Science: Processes & Impacts*, *16*, 1608–1617. https://doi.org/10.1039/c3em00641g

Tukey Jr., H. B. (1970). The leaching of substances from plants. *Annual Review of Plant Physiology*, *21*, 305–324. https://doi.org/10.1146/annurev.pp.21.060170.001513

Tulloss, E. M., & Cadenasso, M. L. (2015). Nitrogen deposition across scales: Hotspots and gradients in a California savanna landscape. *Ecosphere*, *6*(9), 1–12. https://doi.org/10.1890/ES14-00440.1.

United Nations, Department of Economic and Social Affairs, Population Division (2015). *World Urbanization Prospects: The 2014 Revision, (ST/ESA/SER.A/366)*. New York: United Nations, Department of Economic and Social Affairs, Population Division.

United States Census Bureau. (2013). Population change for metropolitan and micropolitan statistical areas in the United States and Puerto Rico (February 2013 Delineations): 2000 to 2010. http://www.census.gov/population/www/cen2010/cph-t/cph-t-5.html.

United States Environmental Protection Agency (2014). *EPA's Report on the Environment (ROE) 2014 Draft*. Washington DC: U.S. Environmental Protection Agency.

Valiela, I. G., Collins, G. Kremer, J., Lajtha, K., Geist, M., Seeley, B., … Sham, C. H. (1997). Nitrogen loading from coastal watersheds to receiving estuaries: New method and application. *Ecological Applications*, *7*(2), 358–380. https://doi.org/10.1023/A:1006345024374

Varenik, A., Konovalov, S., & Stanichny, S. (2015). Quantifying importance and scaling effects of atmospheric deposition of inorganic fixed nitrogen for the eutrophic Black Sea. *Biogeosciences*, *12*, 6479–6491. https://doi.org/10.5194/bg-12-6479-2015

Velinsky, D. J., Wade, T. L., & Wong, G. T. F. (1986). Atmospheric deposition of organic carbon to Chesapeake Bay. *Atmospheric Environment*, *20*(5), 941–947. https://doi.org/10.1016/0004-6981(86)90278-7

Vos, P. E. J., Maiheu, B., Vankerkom, J., & Janssen, S. (2013). Improving local air quality in cities: To tree or not to tree? *Environmental Pollution*, *183*, 113–122. https://doi.org/10.1016/j.envpol.2012.10.021

Wang, X., Ge, T., & Xu, C. (2016). Carbon isotopic (^{14}C and ^{13}C) characterization of fossil-fuel derived dissolved organic carbon in wet precipitation in Shandong Province, China. *Journal of Atmospheric Chemistry*, *73*, 207–221. https://doi.org/10.1007/s10874-015-9323-3

Willey, D., Kieber, R. J., Eyman, M. S., & Avery Jr., G. B. (2000). Rainwater dissolved organic carbon: Concentrations and global flux. *Global Biogeochemical Cycles*, *14*(1), 139–148. https://doi.org/10.1029/1999GB900036

Wollheim, W. M., Pellerin, B. A., Vörösmarty, C. J., & Hopkinson, C. S. (2005). N retention in urbanizing headwater catchments. *Ecosystems*, *8*, 871–884. https://doi.org/10.1007/s10021-005-0178-3

Yan, G., & Kim, G. (2012). Dissolved organic carbon in the precipitation of Seoul, Korea: Implications for global wet depositional flux of fossil-fuel derived organic carbon. *Atmospheric Environment*, *59*, 117–124. https://doi.org/10.1016/j.atmosenv.2012.05.044

Young, R. F. (2011). Planting the living city. *Journal of the American Planning Association*, *77*(4), 368–381. https://doi.org/10.1080/01944363.2011.616996

Zamora, L. M., Prospero, J. M., & Hansell, D. A. (2011). Organic nitrogen in aerosols and precipitation at Barbados and Miami : Implications regarding sources, transport and deposition to the western subtropical North Atlantic. *Journal of Geophysical Research*, *116*, 1–17. https://doi.org/10.1029/2011JD015660

Zimmermann, F., Plessow, K., Queck, R., Bernhofer, C., & Matschullat, J. (2006). Atmospheric N- and S-fluxes to a spruce forest — Comparison of inferential modelling and the throughfall method. *Atmospheric Environment*, *40*, 4782–4796. https://doi.org/10.1016/j.atmosenv.2006.03.056.