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Atmospheric deposition of carbon and nutrients across an arid metropolitan area

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ABSTRACT

Urbanization is increasing rapidly in semi-arid environments and is predicted to alter atmospheric deposition of nutrients and pollutants to cities as well as to ecosystems downwind. We examined patterns of wet and coarse dry deposition chemistry over a five-year period at 7 sites across the Central Arizona-Phoenix (CAP) study area, one of two urban sites within the National Science Foundation's Long-Term Ecological Research (LTER) program. Wet and dry deposition of organic carbon (oC) were significantly elevated in the urban core; in contrast, mean annual wet and dry fluxes of nitrogen (N) were low (<6 kg ha^{-1} yr^{-1}) compared to previous estimates and did not differ significantly among sites. Wet deposition of sulfate (SO_4^{2-}) was high across CAP (mean 1.39 kg ha^{-1} yr^{-1} as S) and represented the dominant anion in rainfall. Dry deposition rates did not show strong seasonal trends with the exception of oC, which was 3-fold higher in winter than in summer; ammonium (NH_4^+) deposition was high but more variable. Dry deposition of NO_3^- and oC was strongly correlated with particulate base cations and dust-derived soluble reactive phosphorus (SRP), suggesting that urban-derived dust is scrubbing the atmosphere of acidic gases and entrained particles and increasing local deposition. Differences between measured and predicted rates of dry N deposition to the urban core may be explained by incomplete collection of gas phase N on surrogate deposition surfaces in this hot and arid environment. The extent of urban enhancement of cations and oC inputs to desert ecosystems appears to be restricted to the urbanized metropolitan area rather than extending far downwind, although a low number of sites make it difficult to resolve this spatial pattern. Nevertheless, wet and dry inputs may be important for biogeochemical cycles in nutrient and carbon-poor desert ecosystems within and near arid cities.

1. Introduction

Atmospheric deposition is recognized as an important source of nutrients to many ecosystems, particularly those in arid environments (West and Skunjins, 1977). Urbanization contributes to an increase in the sources of airborne pollutants such as nitrogen (N), sulfur (S), and organic carbon (oC), and can lead to significant deposition of both nutrients and pollutants in cities as well as to

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ecosystems downwind, with potentially detrimental consequences for human health and ecological functioning (Greenfelt and Hultberg, 1986; Russell et al., 1993; Bytnerowicz and Fenn, 1996; Howarth et al., 1996; Lovett et al., 2000; Smith et al., 2000; Burian et al., 2002; Fenn et al., 2003a; Grimm et al., 2008). In particular, studies have shown urbanization can increase emission and deposition of particulate oC compounds that have a variety of adverse health effects (e.g., Pope et al., 2002; Krewski and Rainham, 2007). Other studies have shown that elevated N deposition can result in changes in plant and microbial community composition and declines in sensitive organisms in both aquatic and terrestrial ecosystems (see Fenn et al., 2003b for review). Despite a global trend towards increased urbanization, with biogeochemical cycles progressively more influenced by human activities (Grimm et al., 2008), there are few spatially explicit deposition data for most urban areas (Lovett et al., 2000).

Cities are not well represented in national monitoring networks (e.g., National Atmospheric Deposition Program (NADP) for wet deposition and the Clean Air Status and Trends Network for dry deposition), since such programs are aimed at monitoring regional and national patterns. Meanwhile, air-quality monitoring networks operated by state agencies are concentrated on cities but they typically focus on health-related pollutants (e.g., ozone, particulate matter (PM), carbon monoxide (CO), and fine-particulate materials) rather than nutrients of ecological interest. This situation is particularly true for rapidly urbanizing, inland, semi-arid to arid regions of the western USA (Fenn et al., 2003a). Given that arid and semi-arid lands represent a third of global terrestrial lands (Graf, 2002) and are expected to experience disproportionate increases in human populations and land transformation worldwide (United Nations, 2005), they are increasingly subject to altered atmospheric deposition, the effects of which have not been fully investigated. Quantifying the spatial and temporal patterns of inputs of oC and nutrients to urban areas and surrounding semi-arid systems is a critical first step in understanding their impact on these ecosystems (Kaye et al., 2006).

Our current understanding of spatial patterns of atmospheric deposition in urban settings is limited mostly to short-term studies concentrated in temperate or coastal cities (Lovett et al., 2000; Sun et al., 2006). For example, Lovett et al. (2000) quantified throughfall and bulk deposition fluxes along an urban to rural gradient, within and to the north of New York City (NYC) over two one-month study periods, and showed that concentrations and fluxes of nutrients decreased significantly with distance from the city. Lovett et al. (2000) suggested that the urban atmosphere acts as a scrubber, removing nitric acid vapor with dust particles containing calcium and magnesium oxides and depositing them near the city as coarse particles. Most other studies have focused on deposition of nitrate, sulfate, and organic carbon compounds such as polychlorinated biphenyls (PCBs) and polycyclic aromatic hydrocarbons (PAHs) in particular urban centers (Noll et al., 1990; Holsen et al., 1991; Lovett et al., 2000; Hughes et al., 2002; Lestari et al., 2003). Thus, questions remain about the extent to which total fluxes of nutrients and oC can be predicted by proximity to the urban core and how they vary with intra- and inter-annual climate conditions.

Compared to mesic metropolitan areas, atmospheric deposition of nutrients to inland urban areas of the western USA is likely to vary substantially both in terms of timing and relative importance of dry versus wet inputs. A large extent of the western USA is characterized by an arid to semi-arid climate where dryfall as particulate and gaseous material can be the dominant pathway of atmospheric deposition (Katrinak et al., 1995; Malm et al., 2004). The few studies of cities in this region have focused on the Los Angeles metropolitan area and surrounding coastal mountains in California; these studies documented high N deposition rates (20–45 kg N ha^{-1} yr^{-1}, maximum 90 ha^{-1} yr^{-1}; Fenn et al., 1996; Fenn and Poth, 1999; Hughes et al., 2002). Loads for other locations are primarily derived from atmospheric deposition models (Fenn et al., 2003a) or site-specific measures (Katrinak et al., 1995). For example, Fenn et al. (2003a) reported modeled N deposition loads of 7–18 kg N ha^{-1} yr^{-1} for the rapidly urbanizing region of central Arizona, with the highest predicted deposition rates in downwind desert ecosystems to the east of Phoenix. To date, however, no empirical studies have evaluated these modeled deposition estimates, or the spatial extent of the urban influence in the surrounding regional landscape.

The goal of this study was to characterize atmospheric deposition across the Central Arizona-Phoenix (CAP) region, including the developed urban core and outlying desert. Specifically, we asked, 'To what extent are concentrations and fluxes of materials enhanced at sites within the urban core relative to undeveloped desert sites upwind and downwind of the city'? We expected higher rates of wet and dry deposition within the urban core and at downwind locations compared to an upwind desert site. We also expected patterns of deposition to vary as a function of proximity to the urban core (e.g., Lovett et al., 2000). We present event-based concentrations and annual wet deposition fluxes of carbon and nutrients across the CAP metropolitan area over a 5-year period (2000–2005). We also examine patterns of coarse dry deposition and compare these estimates with published atmospheric fine-particle concentrations for Phoenix and surrounding locations within Arizona collected as part of the Interagency Monitoring of Protected Visual Environments (IMPROVE) network (Malm et al., 1994).

2. Materials and methods

2.1. *Study region*

The study was conducted at the Central Arizona-Phoenix (CAP) Long-Term Ecological Research (LTER) site in Phoenix, Arizona, USA, one of two urban ecosystems within the National Science Foundation's LTER program. The CAP LTER study site is located in a large alluvial basin in central Arizona surrounded by eroded mountain remnants that together provide the physical template upon which the city has developed. Since 1990, the population of this region has increased by 47% to over 3.5 million people (U.S. Census Bureau, 2002). The CAP LTER encompasses the rapidly expanding Phoenix metropolitan area (13% of the total LTER site) and includes 23 municipalities, surrounding agricultural lands (10%), and undeveloped or sparsely populated Sonoran desert lots and parks distributed throughout the urban matrix (77%; Fig. 1).