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Temporal trajectories of wet deposition across hydro-climatic regimes: Role of urbanization and regulations at U.S. and East Asia sites

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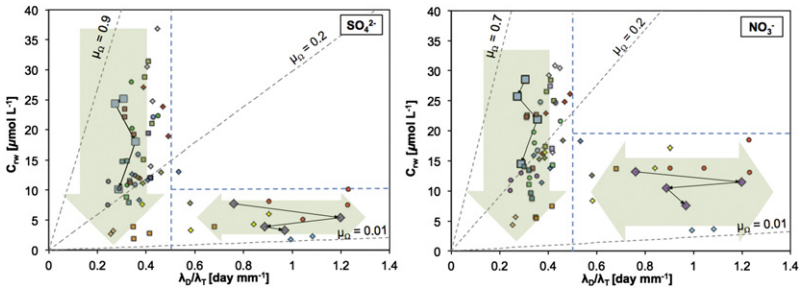
HIGHLIGHTS

- Hydro-climatology drives inter- and intra-annual variability of wet deposition flux.
- Anthropogenic controls can homogenize wet deposition trajectories of NO_3 and SO_4.
- Stochastic simulations enabled evaluation of the relative importance of drivers.
- Results are useful in identifying locations that can benefit from strong regulations.

GRAPHICAL ABSTRACT



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ABSTRACT

Dominant global patterns of urbanization and industrialization contribute to large-scale modification of the drivers for hydrologic and biogeochemical processes, as evident in Asia, Africa, and South America which are experiencing rapid population and economic growth. One manifestation of urbanization and economic development is decreases in air quality, increases in dry/wet deposition fluxes, and growing adverse impacts on public health and ecosystem integrity. We examined available long-term (1980–2010) observational data, gathered at weekly intervals, for wet deposition at 19 urban sites in the U.S., and monitoring data (2000–2009) available for 17 urban sites at a monthly scale in East Asia. Our analyses are based on data for four constituents (SO_4^{2-}, NO_3^-, Ca^{2+}, and Mg^{2+}); differences in atmospheric chemistry and terrestrial sources of these constituents enabled a robust comparative analysis. We examined intra-annual variability and the long-term temporal trajectories of wet deposition fluxes to discern the relative role of anthropogenic and stochastic hydro-climatic forcing. Here, we show that: (1) temporal variability in wet deposition fluxes follows an exponential probability density function at all sites, evidence that stochasticity of rainfall is the dominant control of wet deposition variability; (2) the mean wet deposition flux, μ_Ω ($ML^{-2}T^{-1}$), has decreased in the U.S. over time since enactment of the Clean Air Act, with μ_Ω having become homogenized across varying hydro-climatic regimes; and (3) in contrast, μ_Ω values for East Asian cities are 3–10 times higher than U.S. cities, attributed to lax regulatory enforcement. Based on the observed patterns, we suggest a stochastic model that generates ellipses within which the μ_Ω temporal trajectories are inscribed. In the U.S., anthropogenic forcing (regulations) is dominant in the humid regions, while variability in hydro-climatic forcing explains

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inter-annual variability in arid regions. Our stochastic analysis facilitates projections of the temporal trajectory shifts in wet deposition fluxes as a result of urbanization and other land-use changes, climate change, and regulatory enforcement.

1. Introduction

The broad goal of understanding how the coupling of natural and anthropogenic drivers affect environmental response patterns at regional and global scales regarding atmospheric systems is evident in three parallel developments: (1) the development of air quality and wet deposition models at various spatial scales (e.g., Dentener et al., 2006; Ge et al., 2011), (2) networks for long-term monitoring at various scales (e.g., Baumgardner et al., 2002; Fagerli and Aas, 2008) and (3) remote sensing (e.g., Gupta et al., 2006; Martin, 2008). Studies on large-scale deposition of atmospheric pollutants have focused on characterizing long-term deposition trends to identify the efficacy of policies and emission controls (Lin et al., 2012), correlating high wet deposition with emission "hotspots" (Butler et al., 2005) or with impacts on social-ecological systems (Baron et al., 2011). The present study focuses on temporal and spatial trends ("trajectories") of atmospheric deposition of NO_3^-, SO_4^{2-}, Ca^{2+}, and Mg^{2+} because relatively long-term observational records are available in many countries. Such records may allow examination of the temporal trajectories of wet deposition over urban regions, and enable prediction of patterns for other urban regions in the earlier stages of economic development with similar hydro-climatic conditions. Also, analysis of long-term data at diverse sites allows an examination of large-scale emergent phenomena resulting from interplay between anthropogenic activities and diverse hydro-climatic conditions.

We explore here long-term patterns in wet deposition in urban areas as influenced by three groups of *stochastic* and *non-stationary* controls: (1) anthropogenic drivers, which determine net emissions from multiple, mobile and fixed sources, with increasing urbanization and economic growth and the adoption of environmental regulations; (2) hydro-climatic drivers, especially with inter- and intra-annual changes in precipitation patterns; and (3) transport and transformation processes in the atmospheric boundary layer (ABL) that control regional-scale export/import of atmospheric constituents. Changes in mobile-source emissions depend on increases in the number and type of automobiles, fuel sources, and changes in traffic patterns, while emissions from fixed sources (e.g., power plants and industrial centers; domestic heating or cooking) increase with growing energy demands (Moomaw, 2002). However, emissions from mobile and fixed sources and atmospheric deposition fluxes can be reduced if mitigation measures (e.g., installation of control technologies) are adopted either voluntarily or as a result of strict environmental regulations, thus improving air quality and reducing associated ecological and health impacts (Warby et al., 2005). Additionally, expansion of urban infrastructure and metropolitan areas – and landscape changes in surrounding rural areas – produces local and regional shifts in hydro-climatic forcing (Niyogi et al., 2011; NRC, 2012).

Both hydro-climatic forcing and anthropogenic forcing control the spatiotemporal patterns in wet and dry deposition fluxes. We examine the relative role of these two types of stochastic forcing on wet deposition patterns in the U.S. and East Asia. Wet deposition temporal trajectories for U.S. cities are re-examined as a case study for illuminating the success of regulations in reducing the environmental impact of anthropogenic activities. Similar success has been achieved in Western European nations (Hjellbrekke and Fjaeraa, 2011). For example, Endo et al. (2011) reported that average total (wet + dry) sulfur depositions observed by CASTNET (U.S.) and EMEP (Europe) during 2003–2007 were both 6.2 kg S ha^{-1}, while it was 17.6 kg S ha^{-1} for EANET (East Asia). Using the U.S. as the reference case, we then analyze wet deposition patterns for 17 cities in East Asia to reveal highly variable patterns that depend on rate of urbanization, economic development, and enforcement of regulatory controls. We also present a simple stochastic model that reproduces the temporal trajectories of wet deposition with non-stationary source emissions and natural hydro-climatic drivers.

2. Material and methods

We used long-term (1980–2010) weekly data for wet deposition at 19 locations (Fig. 1a, Table A.1) in the conterminous U.S., maintained by the U.S. National Atmospheric Deposition Program (NADP) (U.S. EPA, 2011). The U.S. sites were selected to represent the range of hydro-climatic regimes and areas likely impacted by urbanization. U.S. Metropolitan Statistical Area (MSA) maps (U.S. Census Bureau, 2009) were used to assess the location of the NADP sites with respect to urban boundaries. Different from administrative boundaries, MSAs map contiguous regions with high population densities that have strong economic and social ties to the central counties. Although, most NADP sites were intended to be installed in non-urban areas, using MSAs allowed the examination of wet deposition impacted by urban regions. Of the 19 sites, 8 sites are within MSAs with population greater than 1 million, 7 sites are within MSAs with population less than 1 million, and 4 sites are in micropolitan areas (Table A.1). We also used wet deposition data (2000–2009), available at a monthly scale at 17 urban sites in East Asia (Fig. 1b, Table A.2) from the Acid Deposition Monitoring Network in East Asia (EANET) (Network Center for EANET, 2011).

Monitoring data were available as rainwater concentration (in ionic weights) of constituents at weekly (CASTNET) or monthly (EANET) intervals. Using these data, mean annual precipitation-weighted concentrations, C_{rw} (ML^{-3}), were calculated as:

$$C_{\mathrm{rw}} = \frac{\sum C_i P_i}{\sum P_i} \tag{1}$$

where C_i is the weekly (or monthly) concentration (ML^{-3}) in rainwater and P_i is rainfall amount (L^3/L^2) collected at weekly (or monthly) intervals.

Our analysis is based on data for SO_4^{2-}, NO_3^-, Ca^{2+}, and Mg^{2+}; differences in atmospheric chemistry and transport, terrestrial emission sources, and regulatory controls of these constituents enabled a robust comparative analysis. We recognize that NH_4^+ contributes to the total N deposition, and has ecological consequences. Holland et al. (2005) reported that, average annual NH_4^+ wet deposition flux in U.S. was 1.38 kg N ha^{-1} while NO_3^- was 1.64 kg N ha^{-1} during 1978–1994. Total N ($NH_4^+ + NO_3^-$) wet deposition flux data for two contrasting sites (NY99 and CA88) conformed to exponential *pdf*s (data not shown). We excluded NH_4^+ from the present analysis because our focus was to examine and compare the role of anthropogenic controls (regulations) to natural forcing over urban regions. Agricultural activities are the dominant sources of NH_4^+ (Fenn et al., 2003), thus its temporal trajectory