LETTER

Rise and fall of nitrogen deposition in the United States

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Understanding the changes in rates and composition of nitrogen (N) deposition has important implications for the regulation of anthropogenic N emissions and is a prerequisite for the assessment of the consequent ecological impacts. By comparing observed data on wet deposition between the periods 1990–1992 and 2010–2012, Li et al. (1) show that total wet inorganic N deposition in the United States shifted from being nitrate-dominated to ammonium-dominated, in line with the conclusions of an earlier report by Du et al. (2). However, Li et al.'s (1) two-period comparison fails to uncover the detailed temporal changes in the rates and composition of N deposition. Based on an analysis of wet inorganic N deposition (nadp.sws.uiuc.edu/) in the United States during the period 1985–2012, I herein show that the national mean wet inorganic N deposition first showed a rise (0.069 kg N ha^{-1} yr^{-2}) during the period 1985–1995 and thereafter a significant fall (–0.036 kg N ha^{-1} yr^{-2}) (Fig. 1*A*).

The nonlinear change in the total wet inorganic N deposition was mainly driven by the change of wet nitrate deposition rather than wet ammonium deposition. Because of an absent regulation of ammonia (NH_3) emissions, national mean wet ammonium deposition increased continuously (0.019 kg N ha^{-1} yr^{-2}) over the period 1985–2012 (Fig. 1*B*). In line with the temporal trend of total wet inorganic N deposition, national mean wet nitrate deposition showed a rise (0.019 kg N ha^{-1} yr^{-2}) during the period 1985–1996 and thereafter a significant fall (–0.055 kg N ha^{-1} yr^{-2}) (Fig. 1*C*). The rise and fall of wet nitrate deposition is consistent with the estimated change in N oxides (NO_x) emissions under the Clean Air Act of 1970 and the Clean Air Act Amendments of 1990 (3, 4).

The composition of wet inorganic N deposition has been remarkably changed over the period 1985–2012, leading to a transition of the dominant N species from nitrate to ammonium. The contribution of ammonium to total wet inorganic N deposition showed a significant increase (0.38% per year) over the period 1985–1999 (Fig. 1*D*) because of a more rapid increase of wet ammonium deposition than wet nitrate deposition (0.05 vs. 0.019 kg N ha^{-1} yr^{-2}). Thereafter, contribution of ammonium has increased more rapidly (0.93% per year) because of an increase in wet ammonium deposition and a decrease in wet nitrate deposition.

Because NO_x emissions has been well curbed by the National Ambient Air Quality Standard (https://www3.epa.gov/airtrends/nitrogen.html), the ongoing increase of ammonium deposition as a result of the absence of control policy for NH_3 emissions should be mainly responsible for the threat of N deposition to many ecosystems in the United States. The changes in rates and composition of N deposition generally reflect the trend in N emissions, which deliver an important message that the regulation of NH_3 emissions is crucial for the future reduction of N deposition. Moreover, the practice of the N emission regulation in the United States also has useful implications for N regulation in developing countries (e.g., China and India), which are currently growing as anthropogenic N emitters.

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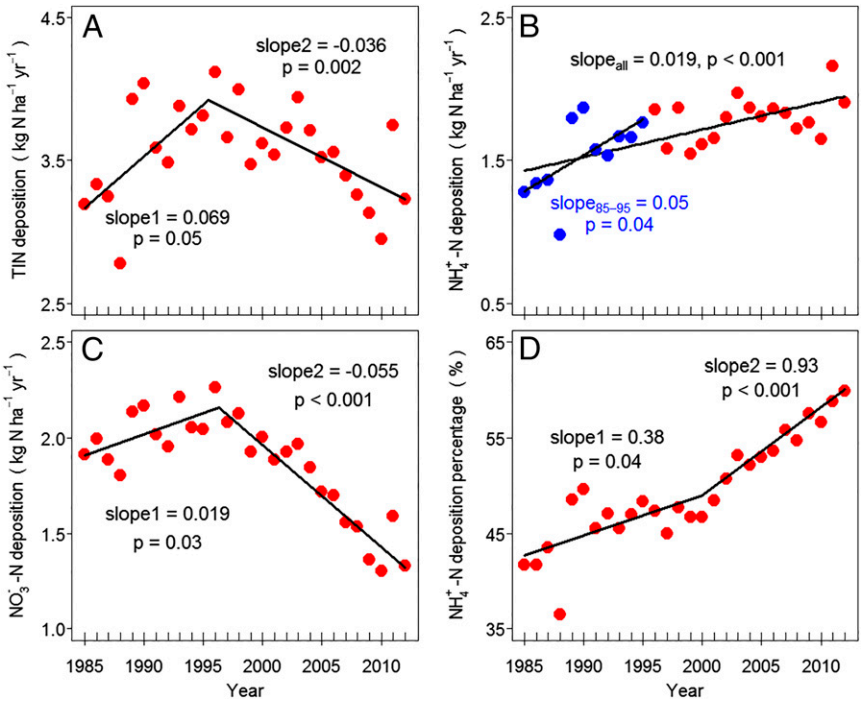

Fig. 1. Changes in national mean wet deposition (unit: kg N ha^{-1} yr^{-1}) of (*A*) total inorganic N (TIN), (*B*) ammonium (NH_4^+), (*C*) nitrate (NO_3^-), and (*D*) ammonium percentage of wet inorganic N deposition in the United States between 1985 and 2012. Observed data of wet deposition were available from National Atmospheric Deposition Program (nadp.sws.uiuc.edu/). Sites were excluded if data were missing for 1 or more years and totally 151 sites were used in this analysis. The piecewise linear regression analysis was conducted by using the "segmented" package (5) for R software.

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