Supplementary Information

Sulfur fertiliser use in the Midwestern US increases as atmospheric sulfur deposition declines with improved air quality

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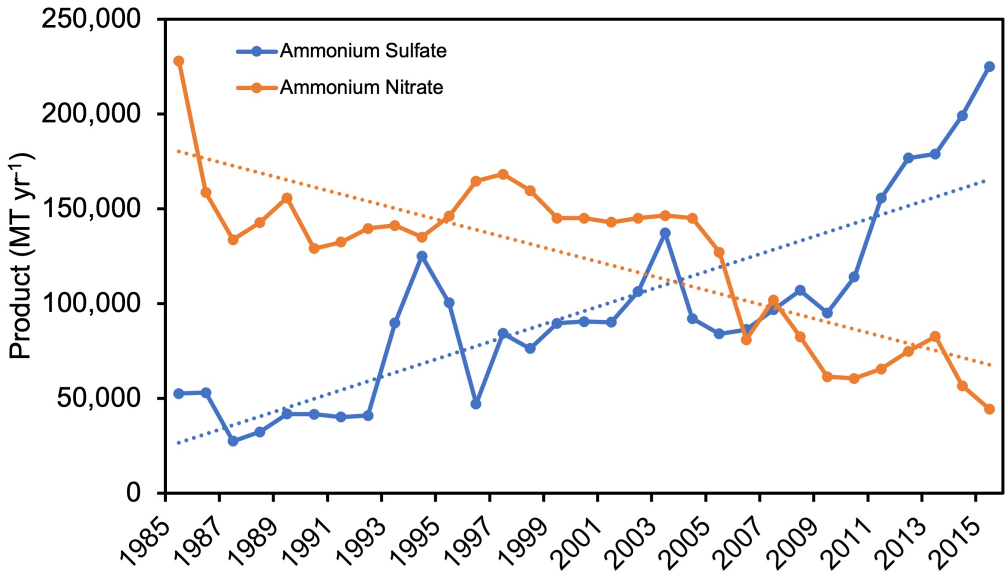
Supplementary Note 1. *Additional explanations for the increase in agricultural S applications*

While declines in atmospheric sulfur (S) deposition may be part of the explanation for increased S fertiliser inputs and S loading to Midwestern United States croplands, other factors should also be considered. In the main text, we describe how yield dynamics driven by climate and soil type, as well as the decision-making of individual farmers and management companies, likely determine fertiliser S inputs, while soil S cycling processes are likely a control on the release and bioavailability of legacy stored S. Yet another factor that may explain increases in S loading over time is increase in application of ammonium sulfate products (providing both nitrogen and S) in lieu of applying ammonium nitrate. Over our period of interest (1985-2015), the latter has been increasingly regulated in the U.S. and elsewhere following its use in several bombings and terrorist attacks. The trends in ammonium sulfate and ammonium nitrate over the 30-year period are shown in Supplementary Figure 1. Indeed, ammonium sulfate increases while ammonium nitrate declines. Even if the increased use of ammonium sulfate is primarily to provide a source of N, it also provides an S source, contributes to overall increases in S loading over the Midwestern U.S. agricultural region, and likely contributes to increased soil acidity (i.e., through nitrification of ammonium). Data are from the Association of American Plant Food Control Officials (AAPFCO).



Supplementary Figure 1. Ammonium nitrate and ammonium sulfate fertiliser trends from 1985-2015. Best fit lines are slope = 4623 MT yr^{-1}, $R^2 = 0.71$ (ammonium sulfate) and slope = -3749 MT yr^{-1}, $R^2 = 0.66$ (ammonium nitrate). The linear models are statistically significant ($p < 0.001$). Both trend lines are significantly different from zero ($p < 0.001$).

Supplementary Note 2. *Estimation of soil S pools across the Midwestern U.S.*

Estimation of soil S pools followed the methodologies outlined in Olson and colleagues (*1*). Briefly, soil S concentrations were obtained from a USGS soil survey which measured S in three samples (0-5 cm, A horizon, C horizon) at approximately 4,857 sites across the Conterminous United States (CONUS) (*2*). Equal Area Quadratic Smoothing Splines (EAQSS) were used to convert the three discrete S concentrations into continuous, site-specific S profiles, from which average S concentrations were calculated for 10 depths (i.e., 0-5 cm, 5-10 cm, 10-20 cm, 20-30 cm, etc.) corresponding to the top 0.9 m. For each of these layers, S concentrations were modeled using a generalized additive model (GAM):

$$\mathrm{Log}_{10}S = \alpha + (Lat, Lon) + s_1(\mathrm{SOC}) + s_2(\mathrm{Elev}) + s_3(\mathrm{Prec}) + \varepsilon \qquad (1)$$

where, S is soil sulfur concentration (%), α is the model intercept, *Lat* is latitude (degrees) *Lon* is longitude (degrees), *Elev* is elevation (m), and *Prec* is precipitation (kg m^{-2} s^{-1}) and ε is the error term. The above model was applied to CONUS SOC, elevation, and precipitation coverages to produce a continuous soil S coverage for the three soil depths. Soil S pools were calculated using:

$$P = S \times BD \times (1 - RC) \times D \qquad (2)$$

where P is the sulfur pool (cg cm^{-2}), S is the modeled concentration of sulfur (%), BD is bulk density (g cm^{-3}), RC is the fraction of soil that consists of rock > 2 mm in size, and D is the depth of the layer (cm). Once generated for CONUS, soil pools were calculated for the WNCEN and ENCEN regions.

The SOC and bulk density data used for pool calculation were obtained from the United States Department of Agriculture (USDA) Gridded National Soil Survey Geographic Database (gNATSGO) database. The GEOS-5 FP meteorology product from the NASA Global Modeling and Assimilation Office and processed by the GEOS-Chem Support Team was used to obtain precipitation data (0.25° x 0.3125°) (*3,4*). Elevation data (30 m x 30 m) were obtained through ESRI ARCGIS Image service. Rock fragment volume data were based on USDA State Soil Geographic Database (STATSGO) data and were obtained from the Soil Information for Environmental Modeling and Ecosystem Management (Pennsylvania State University) (*5*). For discussion of the strengths and limitations of this approach, see (*1*).

Supplementary References

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