## **PERSPECTIVE**

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# A shift in sulfur-cycle manipulation from atmospheric emissions to agricultural additions

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Burning fossil fuels has resulted in a prominent yet unintended manipulation of the global sulfur cycle. Emissions of sulfur dioxide and reactive sulfur to the atmosphere have caused widespread health and environmental impacts and have led, ultimately, to calls to decrease sulfur emissions. However, anthropogenic modification of the sulfur cycle is far from over. Using four contrasting case studies from across the United States, we show how high levels of sulfur are added to croplands as fertilizers and pesticides and constitute a major yet under-studied environmental perturbation. Long-term sulfur additions to crops probably cause similar consequences for the health of soil and downstream aquatic ecosystems as those observed in regions historically impacted by acid rain, yet the cascade of effects has not been broadly explored. A new wave of research on the sulfur cycle will require studies that examine the integrated roles of climate, hydrology and other element cycles in modifying sulfur processes and flows within and downgradient of agricultural source areas. Such research must include not only scientists, but also farmers, regulating authorities and land managers who are engaged in developing approaches to monitor and mitigate environmental and human health impacts.

ince the Industrial Revolution, the exploitation of geologic reservoirs for energy production has been the most important human manipulation of the global sulfur (S) cycle. Through mining and subsequent fossil fuel combustion, humans have mobilized forms of biologically available and chemically reactive S in air, land and water systems, more than doubling background concentrations<sup>1</sup>. Emissions of sulfur dioxide, the formation of sulfate aerosols, and the transport and distribution of these constituents in the environment have occurred at rates that far outpace a return of anthropogenic S to more stable geologic forms<sup>2</sup>.

Beginning in the 1950s, researchers demonstrated the unintended consequences of chronic atmospheric S deposition<sup>3,4</sup>. Large releases of reactive S to the atmosphere from fossil fuel combustion have dramatically degraded air, soil and water quality, impacting ecosystems hundreds of kilometres from pollution sources<sup>5</sup>, and resulting in substantial public health effects due to impairment of respiratory function<sup>6</sup>. Acid rain lowered pH, increased nutrient cation losses, and mobilized trace metals in soils and aquatic ecosystems, most notably in the eastern United States and Europe<sup>7</sup>. Over time, the structure and function of forested and aquatic ecosystems changed in response to inadvertent S deposition<sup>8,9</sup>, resulting in decline or deaths of forest species and toxicity of surface waters from aluminium<sup>10</sup>.

In the United States, these research findings inspired the 1970 Clean Air Act (CAA) and the Title IV Amendments of the 1990 CAA, which have resulted in more than threefold reductions of sulfur dioxide emissions since the mid-1970s<sup>11</sup>. Today, atmospheric S deposition has decreased to near pre-industrial levels in the United States, including the Ohio River Valley and the Appalachian Mountains, which experienced some of the highest historical rates of atmospheric S deposition (Fig. 1). We estimate that in 1989, the areal atmospheric S load over the contiguous United States was 6.6 kg S ha<sup>-1</sup> yr<sup>-1</sup>; the total load of 5.3 Tg S yr<sup>-1</sup> was considerably less than US emissions at that time, 10.6 Tg S yr<sup>-1</sup> (ref. <sup>12</sup>) (see Supplementary

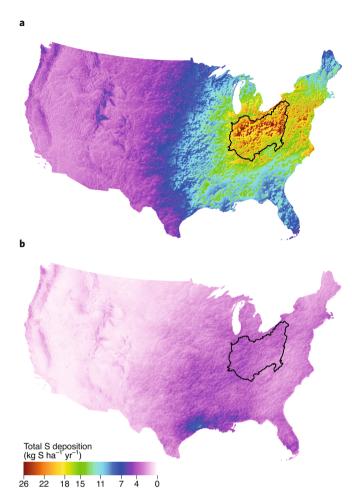
Information). In 2017, areal atmospheric S deposition was 1.6 kg S ha<sup>-1</sup> yr<sup>-1</sup>, and the total load was similar to US emissions of  $\sim$ 1.3 Tg S yr<sup>-1</sup>. The historical projections probably underestimate the contribution of dry deposition and reflect S export to Canada and/or the ocean. While India and parts of China, for example, may not yet have reached their peak in S emissions<sup>1,13</sup>, the magnitude of decline in the United States signals a transition in the S story.

Today, an important perturbation of the S cycle is occurring throughout the world, yet it has gone largely under-appreciated: the addition of reactive S in fertilizers, pesticides and soil conditioners to large-scale croplands. Compared with nitrogen (N) and phosphorus (P), agricultural S use has received scant attention, yet has increased S flows through a host of environmental pathways. With continued changes in climate, crop intensification and decreased atmospheric S deposition, agricultural S inputs will likely increase, carrying with them a cascade of biogeochemical and ecological consequences. In this Perspective, we explore the shift in human manipulation of the S cycle—from atmospheric emissions to agricultural additions—and propose the next wave of research efforts to address its effects.

#### Sulfur additions to croplands

Globally, agricultural S applications are over 50% of the annual S produced, ~70 Tg yr $^{-1}$  (ref.  $^{14}$ ). In the United States, S additions to croplands are substantial: average areal S applications are well in excess of the 1973 peak in acid rain deposition to the northeastern United States, ~20 kg S ha $^{-1}$  (ref.  $^{10}$ ) (Fig. 2), and, in some cases, comparable to additions of N and P. We estimate that S inputs for major crops are ~4 kg S ha $^{-1}$  yr $^{-1}$  averaged over the contiguous United States (total S load, 3.3 Tg S yr $^{-1}$ ; Fig. 2, Supplementary Table 1). However, if averaged only over croplands where S amendments are regularly used, the inputs are ~40 kg S ha $^{-1}$  yr $^{-1}$  (see Supplementary Information). In China, where sulfur dioxide emissions have declined from 25 Tg S yr $^{-1}$  in 2006 to <10 Tg S yr $^{-1}$  in 2017, average

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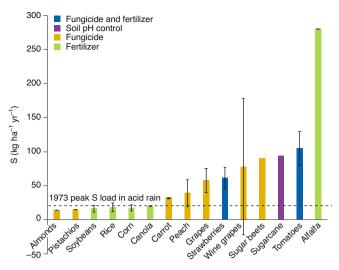


**Fig. 1 | Atmospheric S deposition to the United States. a**, 1989. **b**, 2017. The Ohio River Valley, a location of historically high atmospheric S deposition, is outlined in each map for reference. See Supplementary Information for a description of how S loads were modelled.

S fertilizer inputs are currently ~16 kg ha<sup>-1</sup> yr<sup>-1</sup> and projected to increase<sup>15</sup>. These estimates illustrate that agricultural S use is on a par with elevated atmospheric S deposition.

Sulfur is a fundamental nutritional requirement for all organisms, and critical for plant uptake of N<sup>16–18</sup>. Crops have a range of S requirements. For example, canola, a major crop in the United States and Canada, requires ~20 kg S ha<sup>-1</sup> yr<sup>-1</sup> (refs. <sup>19,20</sup>), whereas average applications to alfalfa, a common forage crop, are 220–340 kg S ha<sup>-1</sup> every few years<sup>21</sup>. Reductions in atmospheric S deposition due to regulation in the United States and Europe are increasing the need for S applications to croplands<sup>22–24</sup> (Fig. 2). Indeed, farmers are now increasing fertilizer S applications to canola in Germany<sup>25</sup>, soybean<sup>26</sup> and corn<sup>27–30</sup> in the Midwestern United States, and alfalfa in New York, United States<sup>31</sup>, and Ontario, Canada<sup>32</sup>, for example, to compensate for reductions in 'free' S deposition.

Sulfur is also a potent pesticide and fungicide used in large quantities for crops like grapes and sugar beets, both of which are plagued by powdery mildew disease<sup>33</sup> (Fig. 2). In California, a major agricultural producer, elemental S is the most widely applied pesticide, exceeding 22 Gg yr<sup>-1</sup> or ~25% of the state's total annual pesticide use<sup>34</sup>. Broadly beyond nutritional requirements, agricultural S uses include as a carrier anion (sulfate) for other nutrient amendments such as N, as a regulator of soil pH for P availability<sup>35,36</sup>, and as gypsum to provide a number of



**Fig. 2** | Current S use in US crops relative to the peak of atmospheric deposition (1973). Data are mean  $\pm 1$  s.d., where a range or multiple values are reported in the literature. Data are from the US Department of Agriculture<sup>33</sup> for all crops except canola<sup>19,20</sup>, sugarcane<sup>36</sup>, wine grapes<sup>34</sup> and alfalfa<sup>21</sup>. In all cases, data are for California, except soybeans, corn, canola and peaches (United States); sugarcane (Florida); and wine grapes (Napa, California). For alfalfa, we report the application rate that is used in years when S is applied, but S is not applied to this crop every year.

benefits (for example, to improve water retention, decrease erosion and reduce P losses)<sup>37–39</sup>.

#### Where does S manipulation occur?

Here we present four case studies from regions within the United States that have contrasting histories of atmospheric S deposition, intensity of S-treated cropland, form of and reason for S applications, and potential future trajectories (Fig. 3). They highlight many of the same environmental issues and unknowns that persist in intensive crop systems globally.

A reference: Wild River, northeastern United States. The Wild River is 27.7 km in length and drains the White Mountains of New Hampshire and Maine. The watershed is 180 km² of predominantly montane forest. Agriculture is a relatively minor component of the landscape and the dominant S input is atmospheric deposition. As with other areas of the northeastern United States, trends in atmospheric S deposition have closely followed the decline in national emissions of sulfur dioxide<sup>40</sup>.

In the late 1980s, atmospheric S deposition to the watershed was elevated, estimated to be 10.5 kg S ha<sup>-1</sup> yr<sup>-1</sup>. The Wild River is relatively close to the Hubbard Brook Experimental Forest, New Hampshire, the site of the longest continuous record of precipitation chemistry in the United States. Observations there suggest that bulk S deposition peaked in 1973 at 17.5 kg S ha<sup>-1</sup> yr<sup>-1</sup> and has declined since. Thus, the pattern of elevated atmospheric S deposition to the Wild River watershed probably predates our analysis; we show a decline from the early elevated values to 2 kg ha<sup>-1</sup> yr<sup>-1</sup> in 2017 (Fig. 4a).

Sulfate-S export in the Wild River generally follows the reductions in atmospheric S deposition. Over the period of record, stream sulfate-S flux has significantly declined by over 50% from 13.0 kg S ha<sup>-1</sup> yr<sup>-1</sup> in 1989 to 6.1 kg S ha<sup>-1</sup> yr<sup>-1</sup> in 2017; a block bootstrap test reported a likelihood estimate of 0.984 for the declining trend. The trends in S fluxes are characterized by year-to-year variations

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due to changes in discharge (see Supplementary Information). The cumulative S mass balance over the monitoring interval suggests that early in the record the watershed was a net sink for atmospheric S deposition. Cumulative S retention peaked in the early 2000s and since that time the watershed has been a net S source, with around two- to threefold higher S export than atmospheric S deposition.

Sulfate-S concentration-discharge relationships for the Wild River support the shift in sink-source behaviour (Supplementary Fig. 2a). Early in the record there was a strong increase in sulfate-S concentrations with discharge, particularly following the summer growing season, suggesting soil flushing with increases in autumn discharge. Over time, the concentration-discharge relationships have changed to ones of dilution with increases in discharge. This pattern is consistent with montane watersheds along the eastern United States<sup>41</sup>, which have either shifted from net sink to source with continued declines in atmospheric S deposition or are projected to do so over the next decade. Watersheds in the northeastern United States, at latitudes comparable to the Wild River, exhibited this transition from a net S sink to source in the early 2000s. The date of transition is delayed with decreases in latitude due to soil characteristics that favour sulfate retention and decreases in discharge due to increases in evapotranspiration<sup>41</sup>.

Corn and soy in the Midwestern United States. The Ohio River crosses seven states and covers the portion of the United States receiving the highest historical atmospheric S deposition (Fig. 1). At its peak, atmospheric S deposition to the region was 21.2 kg ha $^{-1}$  yr $^{-1}$ . Today, values have decreased to  $\sim 3$  kg ha $^{-1}$  yr $^{-1}$ , near pre-industrial levels (Fig. 4b). The 1,579 km Ohio River originates in Pennsylvania and empties into the Mississippi River in Illinois, draining 490,600 km $^2$  that predominately includes corn and soy. Historically, the supply of atmospheric S deposition met crop requirements, but this pattern is changing; increasingly farmers report using S fertilizers Corn requires 6–28 kg S ha $^{-1}$  yr $^{-1}$ , dependent on existing soil S $^{30,42,43}$ , while recommendations for soybean are mixed  $^{44}$ .

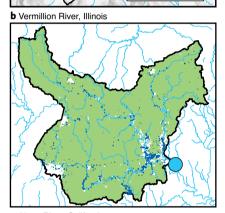
Currently, S fertilizer inputs to corn in Illinois, for example, are minimal relative to other crops, at  $\sim\!2$  kg ha $^{-1}$  yr $^{-1}$ , and comparable to atmospheric S deposition (Fig. 4b). These areal estimates may be low because S fertilizer is typically added within fields where S deficiency is observed. In addition, mineralization and/or desorption of stored soil S may supplement S additions. Given the intensity of corn and soy farming in the United States—359,000 km² of corn and over 360,000 km² of soy in 2018 $^{33}$ —we and others $^{25,45}$  anticipate that fertilizer S use will increase over time, particularly if atmospheric S deposition remains low and below-ground S pools decrease.

While agricultural S application appears to be on the rise, stream sulfate-S fluxes in the Ohio River watershed demonstrate a strongly decreasing trend since the late 1980s. The temporal patterns exhibited by the Vermillion River (Fig. 4b) and other tributaries (see Supplementary Information) represent those of the broader watershed. Sulfate-S fluxes have declined 51% between 1989 and 2017. The magnitude of S export, from >100 kg S ha<sup>-1</sup> yr<sup>-1</sup> in 1994 to <40 kg S ha<sup>-1</sup> yr<sup>-1</sup> in 2017 is more than tenfold greater than estimates of atmospheric S deposition during this period. The long-term

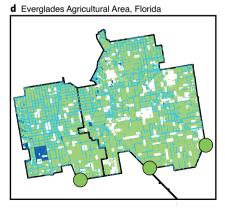
Fig. 3 | Case study areas used to evaluate S mass balances. a-d, Wild River (a), Vermillion River (b), Napa River (c) and Everglades Agricultural Area (d). Cropland is shown in green; circles mark United States Geological Survey stream gauge locations (a-c) and South Florida Water Management District gauges (d) (see Supplementary Table 2 for gauge locations). The inset shows the locations of case study areas within the United States. Maps were prepared by J. Pierson and created using ArcGIS software by Esri (www.esri.com). Credit: Airbus, USGS, NGA, NASA, CGIAR, NCEAS, NLS, OS, NMA, Geodatastyrelsen, GSA, GSI and the GIS User Community.

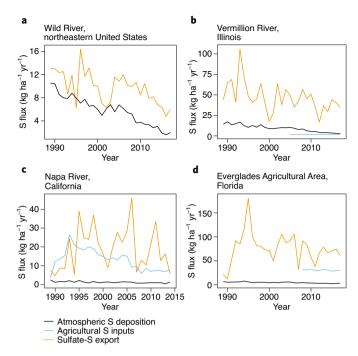
decrease suggests that the surface water record is dominated by continued decline of atmospheric S deposition in response to air quality regulation. In fact, the sulfate-S concentration-discharge relationships reflect a very similar pattern to those of the Wild River after that region became a net S source (Fig. 4a). The high sulfate-S fluxes early in the record are noteworthy and suggest that we











**Fig. 4 | Sulfur mass balances for regional case study areas. a-d**, Wild River (a), Vermillion River (b), Napa River (c) and Everglades Agricultural Area (d). The watershed areas used for the mass-balance calculations are shown in Fig. 3. Data and the modelling approach used to develop the mass balances, as well as sources of uncertainty, are described in the Supplementary Information.

underestimate highly uncertain, historical S sources, including dry deposition and geologic contributions.

Wine-growing in California. The United States is the fourth largest producer of wine in the world, with California as the majority contributor<sup>46</sup>. Powdery mildew is a major threat to wine grapes, so much so that grape growers use elemental S preventatively. Regular, low dose (6–14 kg S ha<sup>-1</sup>) applications of elemental S coat the crop and soil with a reactive skin<sup>47</sup>. During the dry growing season (April–September) cumulative loads are variable (Fig. 2), with a mean of 80 kg S ha<sup>-1</sup> yr<sup>-1</sup> in Napa Valley<sup>34</sup>. Although some grape growers use elemental S in combination with alternative fungicides<sup>48</sup>, S is extremely effective and has not been replaced. While wine grapes are grown across the state, Napa Valley is considered one of the premier wine-growing regions, and wine grapes are grown nearly exclusively (Fig. 4c).

The Napa River watershed is 1,103 km², with the Napa River draining multiple tributaries along its 89 km length from St. Helena to San Pablo Bay. Atmospheric S deposition is low relative to the eastern United States (1.2  $\pm$  0.5 kg ha<sup>-1</sup> yr<sup>-1</sup>, averaged over 1989–2017; Fig. 4c). Sulfur inputs to wine-growing regions dwarf atmospheric S loading, and, from 1989 to 2016, total S inputs have nearly doubled over Napa vineyards³4. The net increase in S applications coincides with a 30% increase in grape acreage from 1989 to 2016⁴9.

Currently, the Napa River watershed appears to be a source of S in most years; sulfate-S export increased 41% over 1989–2017 and has exceeded agricultural S inputs on an areal-basis for the past seven years of the record (Fig. 4c, Supplementary Fig. 2). However, like the Wild River, dramatic discharge-driven changes in sulfate-S export across years have resulted in intervals of net S storage, including prior to 1994, the period of dramatic increases in S applications. Recent drought in the region (beginning in 2011)<sup>50</sup> appears to have

caused a decline in S export—moving the system again towards net storage—while S inputs have remained relatively constant (Fig. 4c).

**Sugarcane in Florida.** The Everglades Agricultural Area (EAA) is almost 3,000 km² and extends south from Lake Okeechobee to the northern extent of the Everglades wetlands. Approximately 2,000 km² of the EAA is dedicated to sugar production. Elemental S is added to increase acidity and improve P availability within the calcium-rich soils of the EAA $^{51-53}$ . We estimate agricultural S usage in the EAA to be  $94 \text{ kg S ha}^{-1} \text{ yr}^{-1}$  (Fig. 4d). However, this application rate is highly uncertain across sources. Schueneman $^{54}$  estimates S applications of  $37 \text{ kg S ha}^{-1} \text{ yr}^{-1}$  based on interviews with commercial fertilizer vendors in the region, while the University of Florida extension recommends  $280-560 \text{ kg S ha}^{-1} \text{ yr}^{-1}$ , particularly for soils with pH > 7.5 (refs.  $^{51,52}$ ); farmers interviewed as part of this survey report using only 20% of the recommended S applications.

Like northern California, atmospheric S deposition to the EAA is relatively low,  $4.8 \pm 1.3 \text{ kg S ha}^{-1} \text{ yr}^{-1}$ , averaged over the 1989–2017 period; in 2017, atmospheric S deposition was ~3 kg S ha $^{-1} \text{ yr}^{-1}$ . In contrast, annual S outflow from canals draining the EAA shows elevated losses with a highly variable temporal pattern largely driven by precipitation to this highly managed hydrologic regime (Fig. 4d, Supplementary Table 2). The difference between atmospheric S deposition and canal S export in the EAA watershed, or net S release, is ~72 kg S ha $^{-1} \text{ yr}^{-1}$ . This discrepancy is within the range of estimated S applications to the EAA. Additional contributions may come from net S mineralization in soils; indeed, chronic peat loss from drainage and agriculture is a management concern for the region  $^{53}$ .

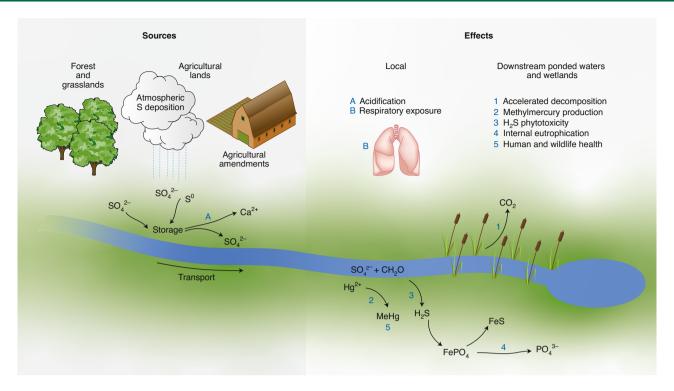
#### Potential consequences of high S applications

The four case studies demonstrate that both in areas historically impacted by acid rain and in croplands receiving S additions, there is a large amount of reactive S that is initially retained in the source area and subsequently transported into adjacent, S-limited ecosystems. This manipulation creates the potential for both local and downstream effects on biogeochemical cycling, ecosystem function and human health (Fig. 5). Within ecosystems, S is subject to multiple transformations, yielding forms that span covalent states from -2 to +6. The large body of research on the impacts of inadvertent S deposition to forests provides insight into those that may occur in croplands and regions recovering from historic atmospheric S deposition.

Soil degradation. Historic atmospheric S deposition, as well as more recent agricultural S inputs, increase local concentrations of sulfate, a strong acid anion, in soil solutions and surface waters. High soil sulfate may help to meet crop S requirements but can also facilitate cation leaching as solutions maintain their charge balance. This disturbance has been chronicled in inherently base-poor forest soils, particularly within formerly glaciated areas of the northeastern United States. Recent research has shown that long-term acidification of forest soils has impacted the water balance—intensifying vegetation water use, increasing evapotranspiration and depleting soil water—in both temperate<sup>55</sup> and tropical<sup>56</sup> systems.

We anticipate that in crop soils, the degree to which S additions generate acidity and leach cations will depend on climate forcing, soil types, intensity of S use, and soil management. The effects of long-term S inputs may be mitigated through liming, tillage practices, or other fertilizer additions. These possible interventions differentiate the potential response of croplands from that of upland forests. However, there is still a need for studies that determine the effects of long-term S inputs on crop soils and agronomic trials that explore management solutions. For example, the extent to which long-term elemental S applications have degraded California vineyard soils is unknown. Past research shows that elemental S causes

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**Fig. 5 | Sources and effects of S in non-agricultural and agricultural areas.** There are multiple forms of S inputs used in agricultural systems, including elemental S and sulfate, among others. Transformations within soils (for example, oxidation of elemental S or reduction of sulfate) result in different fates of S inputs. Potential consequences include effects on human respiratory health (for example, efflux of gaseous S following applications to croplands), human and wildlife health (for example, via stimulation of methylmercury production, a neurotoxin), changes to biogeochemical processes (for example, cycling of carbon, phosphorus and heavy metals), and shifts in the ecology of downstream aquatic systems (for example, conditions that influence plant community composition). Illustration by K. M. Driscoll.

episodic acidification of soils $^{47}$  and suggests that, over time, its use could diminish soil health.

Methylmercury production. Methylmercury (MeHg) formation is a potentially notable consequence of S pollution in aquatic ecosystems downgradient of agricultural source areas. Generated in sediments and reducing environments where sulfate-reducing bacteria and archaea are active<sup>57</sup>, MeHg readily bioaccumulates and biomagnifies through food webs. Concentrations of MeHg drive the degree of assimilation into living organisms and MeHg is the dominant form of mercury in top predators<sup>58</sup>. All 50 US states have fish-consumption advisories and more exist for mercury than all other contaminants combined 59,60. The supply of sulfate can be an important determinant of fish mercury contamination, and this link has been explored extensively in the Florida Everglades<sup>61</sup> and in limited non-agricultural areas 62-65. Sulfate transported from the Florida EAA causes a peak in sediment MeHg production and fish mercury concentrations near the centre of the Everglades<sup>61,65</sup>. Observations of elevated MeHg production have been made in the wetlands downstream of California vineyards<sup>66,67</sup>, and it has long been a concern in the Midwestern United States<sup>68</sup>. However, outside of the Florida EAA, the link between MeHg production and agricultural S use is unexplored. Broadening the scope of this research is an important next step.

**Too much sulfide.** In ponded waters, interaction between S and iron (Fe) cycling can influence P supply from aquatic sediments, an important mechanism contributing to eutrophication. In a well-oxygenated water column, Fe(III) in surface sediments strongly retains P. Under reducing conditions, Fe(II) is ineffective at retaining P, mobilizing it to overlying water. With elevated sulfate loading, the reduction to sulfide and strong binding of sulfide with Fe limits

P retention<sup>69</sup>. As a result, sulfate pollution can exacerbate eutrophication of lakes and wetlands, as demonstrated in non-agricultural environments<sup>70–72</sup>.

A second effect of high sulfate loading to low S freshwater and saline ecosystems is sulfide toxicity<sup>73,74</sup>. Plant tolerances for hydrogen sulfide vary, yet the signs of toxicity are common: inhibition of anoxic energy production and decreases in nutrient uptake and overall plant growth<sup>75</sup>. The factors that detoxify hydrogen sulfide include sequestration by Fe, radial pumping of oxygen by roots, and oxidation by microorganisms living in association with roots<sup>74</sup>. Hydrogen sulfide toxicity is primarily a concern for adjacent, non-agricultural ecosystems; however, it has been observed in rice paddies receiving S<sup>76</sup>. The connection between atmospheric S deposition and hydrogen sulfide toxicity has been established in non-agricultural contexts<sup>77</sup>, but exploration in areas downgradient of high-S croplands has been limited<sup>74,78</sup> and the full extent unquantified.

**Human health.** In grapes and other crops where S is broadcast sprayed as a pesticide, there is the potential for impacts on human health. In particular, chronic exposure may lead to respiratory issues, such as asthma<sup>79,80</sup>. While in the United States farm safety regulations include personal protective equipment and post-spray exclusion periods, they do not cover worker exposure that may occur after exclusion periods as S products continue to react (as, for example, with gaseous forms) or exposure of local residents to pesticide drift. The long-term effects of S exposure across demographics in major agricultural areas—including children and people with pre-existing health conditions—are not well documented.

#### **Future research directions**

There is a strong need to quantify agricultural S inputs broadly and determine where S deficiencies are emerging with continued

declines in atmospheric S deposition and intensification of crop management. Determining which S products are best for meeting S demands and how to use them prudently<sup>81</sup> remain open questions. In addition, the degree to which crop S demands are satisfied by plants mining legacy atmospheric S deposition is unclear. Efforts to understand storage of legacy S in the soil profile, the timescales over which it is released and becomes available to crops, and the degree to which it must be supplemented with S fertilizers to satisfy plant demands are important next steps to inform sustainable management plans.

It is also important to evaluate the effects of long-term S applications on soil health and fertility. High applications of N fertilizers in the United States and China, for example, are already a major problem, causing acidification and loss of base cations from crop soils<sup>82,83</sup>. Long-term use of elemental S, which oxidizes to sulfate and increases acidity, may pose an additional risk if not countered with lime applications. In order to address the long-term impacts of high S applications on crop soils using a mass-balance approach or to conduct agronomic trials to explore management solutions, there must be better accounting and public reporting of agricultural S inputs. We suggest periodic surveys and mandatory reporting, such as that implemented by the US Department of Agriculture and the State of California, respectively.

Similar to widespread efforts to investigate human manipulation of the N cycle<sup>84</sup>, it is crucial to determine the fate and consequences of agricultural S broadly. In recent decades, critical research tools have advanced, including stable isotope ratio mass spectrometry, metagenomics and spectroscopy, which can help researchers identify unique agricultural S signatures, investigate microbial communities in high S soils, and quantify S species. In addition, it will be necessary to integrate the study of hydrology and other interacting biogeochemical cycles. Irrigation management, altered rainfall patterns, and changes to wet–dry cycles will have profound effects on S reactivity and transport. Similarly, the effects of agricultural S on the quality and reactivity of dissolved organic matter and cycling of other elements (for example, Fe, P and Hg) in downstream areas must be quantified.

#### **Conclusions**

It will require concerted effort to understand the consequences of shifting from unintended atmospheric emissions to deliberate agricultural applications of S. Countries like China and India, which are still working to lower air pollution while simultaneously intensifying agricultural production, will probably experience compounded effects of these S sources. With continued changes in climate and hydrology, the behaviour of S and the extent of consequences will vary over space and time. This calls for integrated studies across environmental science, agronomy, public health and social science that explicitly include farmers and other stakeholders in codeveloping research and solutions—uniquely possible in agricultural systems. We believe that by drawing on the decades of research on the consequences of acid rain and excess use of N and P fertilizers, scientists and stakeholders can swiftly and proactively guide actions to understand and address today's changes to the S cycle.

#### Data availability

Data for the atmospheric S deposition estimates are available through the National Atmospheric Deposition Program (https://nadp.slh.wisc.edu/) for wet deposition, the US Environmental Protection Agency Clean Air Status and Trends Network (https://www.epa.gov/castnet) for dry deposition, and the PRISM spatial climate database (https://www.prism.oregonstate.edu/) for precipitation quantity. Data for sulfate export and stream discharge are available through the United States Geological Survey (https://waterdata.usgs.gov/nwis).

#### Code availability

Code for the kriging analysis and modelling of atmospheric S deposition is available on GitHub (https://github.com/h-fakhraei/s\_deposition.git). Code and information about Weighted Regressions on Time, Discharge and Season modelling of sulfate export are available at https://github.com/USGS-R/EGRET.

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#### **Competing interests**

The authors declare no competing interests.

#### **Additional information**

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