# Hidden Cost of U.S. Agricultural Exports: Particulate Matter from Ammonia Emissions

Fabien Paulot* and Daniel J. Jacob

School of Engineering and Applied Sciences, Harvard University, Cambridge, Massachusetts 02138, United States

ABSTRACT: We use a model of agricultural sources of ammonia (NH_3) coupled to a chemical transport model to estimate the impact of U.S. food export on particulate matter concentrations ($PM_{2.5}$). We find that food export accounts for 11% of total U.S. NH_3 emissions (13% of agricultural emissions) and that it increases the population-weighted exposure of the U.S. population to $PM_{2.5}$ by 0.36 μg m^{-3} on average. Our estimate is sensitive to the proper representation of the impact of NH_3 on ammonium nitrate, which reflects the interplay between agricultural (NH_3) and combustion emissions (NO, SO_2). Eliminating NH_3 emissions from food

export would achieve greater health benefits than the reduction of the National Ambient Air Quality Standards for $PM_{2.5}$ from 15 to 12 μg m^{-3}. Valuation of the increased premature mortality associated with $PM_{2.5}$ from food export (36 billion US$ (2006) per year) amounts to 50% of the gross food export value. Livestock operations in densely populated areas have particularly large health costs. Decreasing SO_2 and NO_x emissions will indirectly reduce health impact of food export as an ancillary benefit.
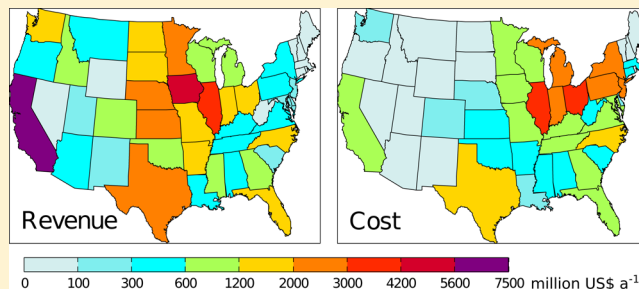
■ INTRODUCTION

Increasing nitrogen inputs from fertilizer application have contributed to greater agricultural outputs in the last 50 years.[1] They have also resulted in the release of nitrogen to the environment, in particular through emissions of ammonia (NH_3) to the atmosphere. In the U.S., Houlton et al.[2] estimated that ~25% of the nitrogen used as fertilizer is lost to the atmosphere as ammonia (NH_3), costing farmers ~6 billion US$ a^{-1}. Beyond the direct economic liability, this makes agriculture the largest source of NH_3 to the atmosphere with important consequences for human health, ecosystems, and climate.[3−5] The most costly impact, human health,[6] is due to the production of fine inorganic particulate matter ($PM_{2.5}$) as ammonium−sulfate−nitrate salts, a major contributor to $PM_{2.5}$ mass.[7] $PM_{2.5}$ is a well-documented factor for premature mortality.[8,9]

Estimates of the health cost of NH_3 emissions through $PM_{2.5}$ require accurate representation of the sensitivity of $PM_{2.5}$ to changes in NH_3 emissions, of the relationship between $PM_{2.5}$ and the health outcome (e.g., premature mortality), and of the valuation of the health impact. Previous work suggests that the average U.S. annual health cost (morbity + mortality) of 1 kg of NH_3 emitted to the atmosphere ranges from 3 to 13 US$ (2006) depending on the valuation method, 2 and 9 times greater than the cost of 1 kg of SO_2 and NO_x, respectively.[10] The cost can also vary depending on the source type. For instance, Muller and Mendelsohn[11] found that the cost of 1 kg of NH_3 can vary from 0.1 to 73 US $(2006). This variability reflects in part the spatial distributions associated with different NH_3 sources, with sources located closer to population centers having a greater impact.

The above estimates rely on simplified source-receptor (S-R) models that do not capture the complex nonlinear relationship between NH_3 emissions and $PM_{2.5}$.[12,13] This relationship is controlled by the thermodynamic equilibrium between $NH_x \equiv NH_{3(g)} + NH_4^+$, $NO_{3T} \equiv HNO_{3(g)} + NO_3^-$, and $SO_{4T} \equiv SO_4^{2-} + HSO_4^- + H_2SO_4$, where (g) denotes the gas-phase and other species are in the aerosol phase.[14] The impact of NH_3 emissions on $PM_{2.5}$ thus depends on meteorological parameters (e.g., temperature, relative humidity), the magnitude of the perturbation to NH_3 emissions, and the abundance of NO_{3T} and SO_{4T}, which are the products of the oxidation of SO_2 and NO_x, two byproducts of combustion.[15−17]

Here, we focus on quantifying the cost of NH_3 emission associated with food export. Unlike previous valuation studies, we use a chemical transport model with detailed representation of aerosol thermodynamics and NH_x losses to calculate the impact of a change in NH_3 emissions on $PM_{2.5}$. We also account for the temporal and spatial heterogeneity of the various NH_3 sources using the model for the magnitude and seasonality of agricultural emissions (MASAGE[18]).

We choose to focus on food exports to reflect their growing importance for the U.S. trade balance. The U.S. is presently the largest world exporter of wheat, corn, soybeans, cotton, pork, and poultry,[19] which makes it a central component of global food security.[20] From 2000 to 2009, 20% of U.S. agricultural production was exported, which amounted to 74 billion US$ (2006) per year and accounted for 9% of the total value of U.S.

exports.[21] The value of U.S. agricultural export is increasing faster than other exports (doubling from 2000 to 2010), reflecting in part growing demand from China, which has become the leading export destination for U.S. food.[22]

MATERIALS AND METHODS

We use the GEOS-Chem global CTM (v9.1.3) to calculate the sensitivity of $PM_{2.5}$ to NH_3 emissions from agricultural exports. GEOS-Chem includes a detailed representation of the photochemical production of SO_{4T} and NO_{3T}.[23,24] Thermodynamic equilibria between SO_{4T}, NH_x, and NO_{3T} are simulated using ISORROPIA II.[25] Wet scavenging is simulated as described by Liu et al.[26] and Wang et al.[27] for aerosols and by Amos et al.[28] for gases. Comparisons with observations[24,28,29] have shown these parametrizations provide an unbiased representation of wet scavenging. Dry deposition is calculated using a standard resistance-in-series model[30,31] applied to a surface-type database from.[32] GEOS-Chem is driven by assimilated meteorological data from the NASA Goddard Earth Observing System (GEOS-5) with horizontal resolution of $0.5° \times 0.67°$ and 72 vertical levels. We degrade the horizontal resolution to $2° \times 2.5°$ for computational efficiency.

US anthropogenic emissions of NO_x and SO_2 are taken from the U.S. Environmental Protection Agency (EPA) National Emission Inventory for 2005 (NEI05). The parametrization of soil NO_x emissions takes into account the effect of N deposition and fertilizer application as described by Hudman et al.[33] Our simulation also includes NO_x emissions from biomass burning (GFED3 with monthly temporal resolution[34]) and lightning.[35]

Agricultural emissions of NH_3 are calculated using the MASAGE model. MASAGE estimates the magnitude, seasonality, and spatial distribution of NH_3 emissions associated with 19 crops and 7 livestock types.[18] This detailed representation is necessary to capture the emission profiles associated with different commodities. For instance, soybean requires little N input, whereas beef cattle, the largest source of NH_3 in the U.S., generates NH_3 emissions not only from manure but also from fertilizer used to grow feed crops (40% of corn grown in the U.S. is used as livestock feed). We also include in the model other anthropogenic sources of NH_3 from transportation and biofuel,[36] natural sources from soil, ocean, and wild animals,[37] and open fire emissions from GFED3.[34] U.S. NH_3 emissions in MASAGE are 2.7 Tg N a^{-1}, to which agriculture contributes more than 80%. MASAGE U.S. NH_3 emissions are 15% lower than the emissions from the U.S. EPA National Emission Inventory for 2006. Paulot et al.[18] found good agreement between NH_3 emissions estimated by MASAGE and those inferred from inversion of NH_4^+ wet deposition fluxes.

We estimate the U.S. NH_3 emissions associated with food export by scaling crop acreages and livestock head numbers based on the commodity–specific export fraction by weight reported by the United States Department of Agriculture Economic Research Service (USDA ERS[38]) and the Food and Agriculture Organization (FAO[39]) averaged from 2000 to 2009. The export fraction for each commodity is assumed uniform across the U.S. We use the USDA feed index to account for the indirect export of feed crops through consumption by exported livestock.[38] We also account for the contribution of food export to NO emissions. Food exports account for 20% of soil NO_x emissions from fertilizer but this represents less than 1% of

total U.S. NO_x emissions. The effect on surface ozone is small (<0.6 ppbv anywhere) and will not be discussed further.

Table 1 summarizes the export fractions and the gross and net values for the main agricultural commodities exported by

Table 1. Food Export and Associated NH_3 Emissions

commodity	fraction exported[a] %	export value[a] billion US$	net export value[b] billion US $	NH_3 emissions[c] GgN a^{-1}
livestock		10.1	3.4	165
poultry	13%	2.8	2.1[d]	40
pork	12%	2.6	0.6	40
beef	7%	2.9	0.1	75
dairy	2%	1.8	0.6	10
crops		45	20.2	130
cotton	69%	3.8	0.8	15
soybeans	56%	10.5[e]	7	5
wheat	49%	5.5	2.7	45
rice	48%	1.3	0.5	5
feed, other grain	23%	5.3	1.3	20
corn	22%	7.3	3.4	35
fruits, vegetables, nuts	19%	11.2	4.5	<5
total		55.1[f]	23.5	295

[a]Averaged from 2000 to 2009 from USDA ERS[38] and FAOSTAT.[39] [b]Calculated by multiplying the export value by the ratio of production value minus operating cost to production value (USDA ERS[38]). [c]Calculated using the MASAGE model (see text). [d]http://extension.umd.edu/publications/pdfs/eb373.pdf [e]http://www.soystats.com [f]Total U.S. export value over the 2000−2009 period was 74 billion US $(2006). The remainder includes a number of minor products (such as tobacco and hides) that are not considered here.

the U.S. Together they account for 75% of the gross value of U.S. export. Fruits, vegetables and nuts, soybeans, corn, wheat, and cotton are the most important exported commodities by value. Other commodities such as tobacco and hides account for the remaining 25% and are not considered here.

RESULTS AND DISCUSSION

We estimate that U.S. NH_3 emissions from food export are 0.3 Tg N a^{-1} or 13% of U.S. agricultural emissions. This is less than the exported fraction of U.S. agricultural production (20%), reflecting the large contribution of crop production to U.S. exports. Livestock production, which is more N intensive than crops,[40] accounts for 50% of export emissions but only 20% of the export value. Beef alone accounts for 25% of U.S. export emissions. Figure 1 shows the spatial distribution of NH_3 emissions associated with food export. Beef export is the largest contributor in the South, while pork, corn, and wheat exports are the largest contributors in the upper Midwest.

Figure 2 shows the increase in annual mean $PM_{2.5}$ from NH_3 emissions associated with food export, as estimated by difference between a GEOS-Chem simulation including all emissions and a simulation excluding the NH_3 and NO_x emissions associated with agricultural exports. The increase of $PM_{2.5}$ exceeds 1 $\mu g \ m^{-3}$ in Indiana and Ohio and is dominated by ammonium nitrate (NH_4NO_3). The maximum increase is not collocated with the maximum in export emissions (Figure 1), reflecting the different sensitivity of $PM_{2.5}$ to NH_x under
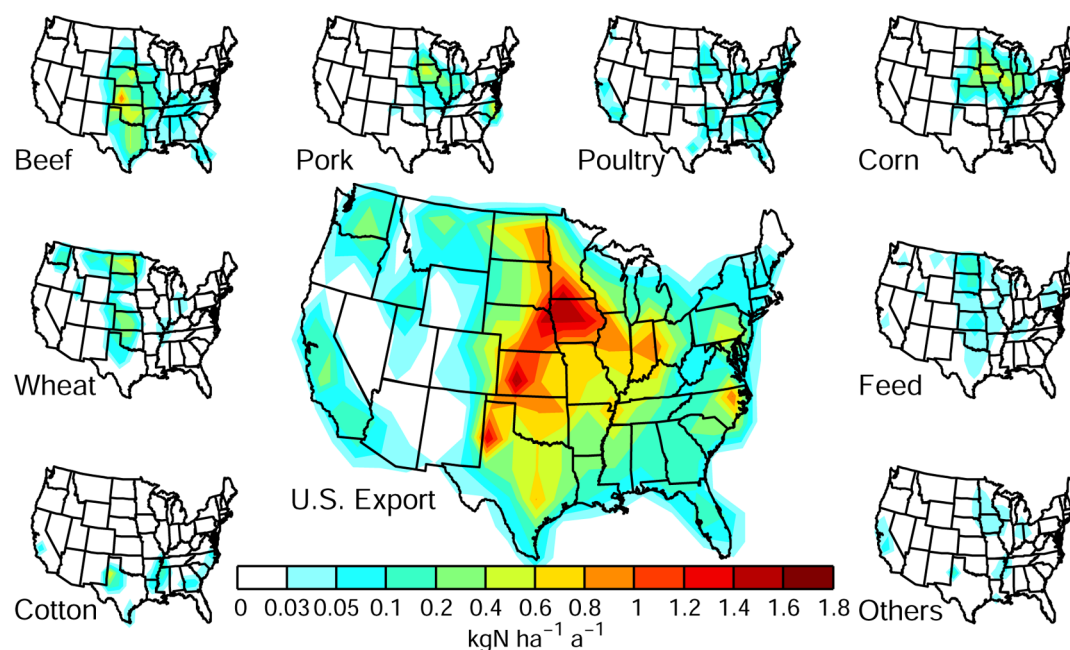
Figure 1. NH_3 emissions associated with the production of exported food.

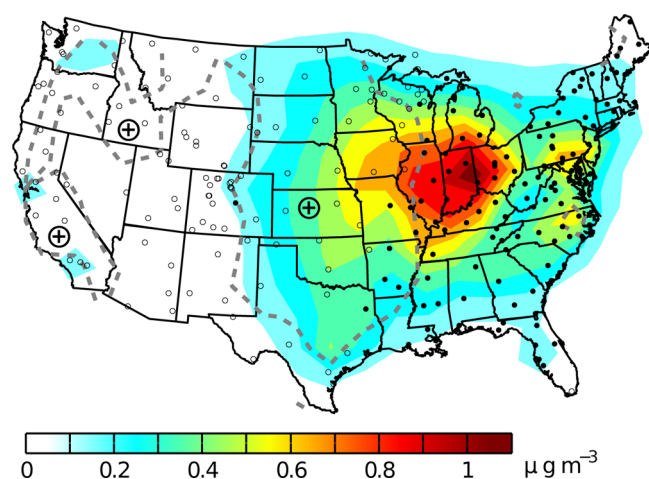**Figure 2.** Impact of NH_3 emissions from food export on annual mean surface $PM_{2.5}$ concentration. The GR = 1 contour line is shown as dashed and delimitates the region, where NH_x is sufficient to neutralize both NO_{3T} and SO_4 (shown by a $\oplus$ symbol). Black (white) dots designate stations from the National Acid Deposition Program with annual average pH below 5 (above 5) in 2006.

different chemical regimes. This variable sensitivity can be diagnosed using the gas-ratio (GR):[15]

$$GR = \frac{[NH_x] - 2[SO_4]_T}{[NO_{3T}]} \qquad (1)$$

When GR is greater than 1, there is enough NH_x to neutralize both NO_{3T} and SO_{4T} and NH_4NO_3 is only weakly sensitive to changes in NH_3 emissions. When $0 < GR < 1$, all SO_{4T} is neutralized and NH_4NO_3 formation is limited by NH_x, such that an increase in NH_3 emission correspondingly increases NH_4NO_3 mass. When GR is lower than 0, $(NH_4)_2SO_4$ and NH_4HSO_4 formation are limited by NH_x, and increasing NH_3 emissions has a small effect on $PM_{2.5}$ by adding mass to pre-existing sulfate aerosol. This latter regime is not found in our simulations anywhere in the U.S., consistent with observations

that $PM_{2.5}$ is in general fully neutralized.[23] The dashed gray line in Figure 2 shows the simulated transition between the NH_x saturated and NO_{3T} saturated regimes (GR = 1). The transition region is consistent with measurements of precipitation pH, where it corresponds roughly to pH 5. An atmosphere with GR > 1 would have a rainwater pH > 5,[41] although correspondence is complicated by the variation of GR with altitude. The maximum increase in $PM_{2.5}$ is in agricultural regions located near large SO_2 and NO sources (coal-fired power plants, urban centers), which promote low GR. This highlights the detrimental impact on air quality of the interaction between agricultural and combustion (transport and energy) emissions.

The average annual exposure of the contiguous US population[42] to $PM_{2.5}$ from food export is 0.36 μg m^{-3}. Based on commodity−specific export fluxes over the 2000−2009 period,[39] intercontinental food exports account for 80% of the NH_3 emissions associated with agricultural export, with Asia contributing 46%. Japan is the largest single contributor as a leading export destination of pork (37.5% of all export pork) and beef (14%). The contribution of intercontinental food export to the $PM_{2.5}$ exposure of the US population far exceeds that from intercontinental atmospheric transport of sulfate, organic, and black carbon (0.085 μg m^{-3} [43]).

We estimate the excess mortality (ΔM) associated with agricultural exports as

$$\Delta M = \mathcal{P} \mathcal{M}_0 (1 - \exp(-\beta \Delta C)) \qquad (2)$$

where $\mathcal{P}$ is the population over 30 years old, $\mathcal{M}_0$ is the annual mortality rate (a^{-1}), ΔC is the change in annual mean $PM_{2.5}$ (in μg m^{-3}). β, the impact parameter, is taken from Krewski et al.[44] ($\beta = 5.8 \times 10^{-3}$ $m^3 \mu g^{-1}$). The Krewski et al.[44] study is used by the U.S. EPA[12,45,46] to estimate the health impact of SO_2 and NO_x emissions. It shows a significantly lower health impact for $PM_{2.5}$ than the Harvard Six-Cities Study ($\beta \sim 14 \times 10^{-3} m^3 \mu g^{-1}$[47,48]) and thus provides a more conservative estimate of the excess mortality from food export. ΔM is calculated using the US EPA BenMAP version 4.0 software.[42] We estimate that $PM_{2.5}$ from food export is responsible for
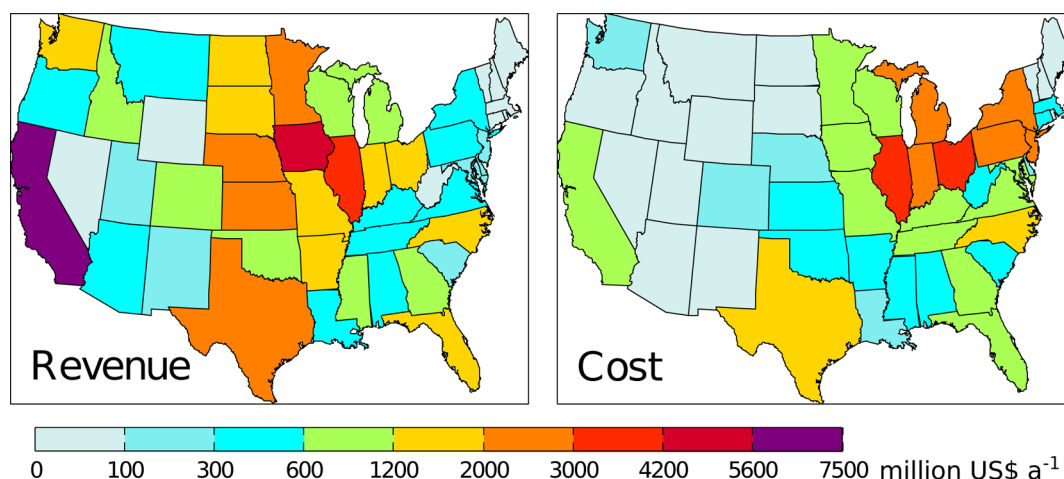
Figure 3. Comparison between annual gross revenue and health cost of agricultural export for individual states. The health cost as computed here is solely driven by increased exposure to $PM_{2.5}$ due to NH_3 emissions from agricultural export.

5100 premature deaths (3400−6700; 95% confidence interval associated with the uncertainty in β) per year. This corresponds to ~4% of the health impact of all anthropogenic $PM_{2.5}$[45] and is much larger than the reduction in premature mortality (460 premature deaths) that would be achieved by a reduction of the National Ambient Air Quality Standards for $PM_{2.5}$ from 15 to 12 μg m^{-3}.[46]

We estimate that the resulting annual health cost of $PM_{2.5}$ from food export is 36 (4−100) billion US$ (2006) or 100 US$ (2006) per kg of NH_3. Our estimate reflects the "willingness to pay" of individuals in the U.S. for a small reduction in the risk of premature death, which is summarized through the value of a statistical life (VSL). Here we use a VSL of 4.8 million US$ (1990) (7.9 million US$ (2006)) based on 26 wage-risk and contingent valuation studies.[42] Premature mortality is expected to lag $PM_{2.5}$ exposure therefore we need to discount the cost.[49] Our estimate is based on a discount rate of 3%, which reflects the rate of return of long-term government debt,[49] and a 20 year lag-stage structure as recommended by the U.S. EPA Science Advisory Board.[46]

Our estimate of the health cost of 1 kg of NH_3 is greater than previous estimates: 12 € in the European Union,[6] 0.1 to 73 US $(2006) in the U.S.[10,11,50] Part of this discrepancy can be attributed to differences in valuation methods. A discount rate of 7%, which reflects the rate of return of private capital in the U.S.,[49] would reduce our estimate by 10%, while a 15 year homogeneous lag would reduce our estimate by 27% and 57% for a discount rate of 3% and 7% respectively.

The toxicity of $PM_{2.5}$ is also uncertain. Here, we assume that the toxicity of $PM_{2.5}$ is independent of its speciation. In contrast, the ExternE model, which is used to evaluate the costs and benefits of European policies (http://www.externe.info) assumes that nitrate are 50% less toxic than sulfate. Unlike metals,[51] neither the health impact of nitrate or sulfate has been conclusively established by toxicological studies.[52] Based on epidemiological studies[53] nitrate is associated with more cardiovascular hospital admissions than $PM_{2.5}$ but fewer respiratory hospital admissions.

The largest difference with previous valuations of the cost of NH_3 emissions lies in the characterization of the relationship between NH_3 and $PM_{2.5}$. Previous studies relied on S−R relationships derived from reduced-form techniques, which allow to explore many different scenarios through a simplified treatment of transport, wet and dry deposition, emissions, and photochemistry.[12,13] For instance, NH_3 emissions are assumed to have no seasonality[10] and ammonium nitrate production is reduced by 75% to account for the effect of temperature on the NO_{3T}−NH_x−SO_{4T} equilibrium.[54] Because ammonium nitrate is very sensitive to wintertime NH_3 concentrations,[24,55] these simplifications suggest that S−R models may not be suitable to characterize the sensitivity of ammonium nitrate to NH_3 emissions.[13] Indeed the CRDM model, a S−R model used for many valuation studies, suggests a near linear relationship between NH_3 emissions and cost in the U.S. over a large range of emissions,[50,56] but this is incorrect given the heterogeneity of the gas-ratio. Recent studies in Europe using detailed chemical transport models and time-resolved NH_3 emissions clearly illustrate the strong nonlinearity between $PM_{2.5}$ and NH_3 emissions.[57] found that $PM_{2.5}$ was more sensitive to a 50% change in NH_3 emissions than to a 50% change in NO_x or SO_2 emissions. In contrast, Pay et al.[58] found that $PM_{2.5}$ was only weakly sensitive to small changes in NH_3 emissions.

Comparison between the cost of the increased health risk (36 (4−100) billion US$ (2006) for NH_3 emissions alone) associated with agricultural exports and the gross (55 billion US $ (2006)) and net value (23.5 billion US$ (2006)) of these exports (Table 1) indicates extensive negative externalities. Taking into account other impacts of agriculture (e.g., eutrophication,[59,60] loss of biodiversity,[4,61−63] and greenhouse gases emissions from production and transportation[64]) would further diminish the net value of agricultural exports.

Figure 3 highlights the different geographical distribution of the $PM_{2.5}$ health cost and direct gross revenue associated with agricultural exports. Regions with large agricultural activities (high NH_3 emissions) and low population densities (low SO_2 and NO emissions) clearly benefit from food exports. Most of the cost is born by populated states in the Northeast and Great Lakes region, where $PM_{2.5}$ formation is promoted by upwind NH_3 sources. In the Northeast, where agricultural production is small, the cost is driven by interstate transport of NH_x.[17]

Export data for 2010 onward suggest that the exported fraction of the US agricultural production is stable or increasing. For instance, 21% of the U.S. pork production was exported in 2011,[38] a near-doubling over the 2000−2009 average fraction used in this study (Table 1). This trend may foretell the continuing growth of the U.S. NH_3 emissions

attributable to food export as world food demand increases.[65] Greater focus on N-efficient crops (e.g., soybeans) would reduce the health impact of food exports. Previous studies have also shown that NH_3 emissions could be reduced through changes in fertilizer types and applications as well as manure management.[6,50,55] Such measures have proven effective in Europe. where NH_3 emissions have decreased by nearly 30% from 1990 to 2010.[66] Reduction of the health impact of NH_3 emissions could also be achieved indirectly though reduction of NO_x and SO_2 emissions, which would lower the sensitivity of $PM_{2.5}$ to NH_3. Our work further suggests that the health impact of food production could be diminished through greater spatial segregation between food production and densely populated regions. Such an approach would result in greater food transport ("food miles") but the impact on the carbon footprint of the food chain would likely be small.[67]

■ AUTHOR INFORMATION

Corresponding Author
*E-mail: paulot@seas.harvard.edu.

Notes
The authors declare no competing financial interest.

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