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Implications of Ammonia Emissions from Post-Combustion Carbon Capture for Airborne Particulate Matter

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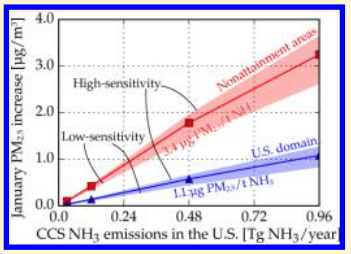
Supporting Information

ABSTRACT: Amine scrubbing, a mature post-combustion carbon capture and storage (CCS) technology, could increase ambient concentrations of fine particulate matter ($PM_{2.5}$) due to its ammonia emissions. To capture 2.0 Gt CO_2/year, for example, it could emit 32 Gg NH_3/year in the United States given current design targets or 15 times higher (480 Gg NH_3/year) at rates typical of current pilot plants. Employing a chemical transport model, we found that the latter emission rate would cause an increase of 2.0 μg $PM_{2.5}/m^3$ in nonattainment areas during wintertime, which would be troublesome for $PM_{2.5}$-burdened areas, and much lower increases during other seasons. Wintertime $PM_{2.5}$ increases in nonattainment areas were fairly linear at a rate of 3.4 μg $PM_{2.5}/m^3$ per 1 Tg NH_3, allowing these results to be applied to other CCS emissions scenarios. The $PM_{2.5}$ impacts are modestly uncertain (±20%) depending on future emissions of SO_2, NO_x, and NH_3. The public health costs of CCS NH_3 emissions were valued at $31−68 per tonne CO_2 captured, comparable to the social cost of carbon itself. Because the costs of solvent loss to CCS operators are lower than the social costs of CCS ammonia, there is a regulatory interest to limit ammonia emissions from CCS.



INTRODUCTION

Carbon capture and storage (CCS) technology is considered an important potential climate change mitigation option.[1−3] Amine scrubbing is currently the most mature post-combustion capture technology.[4] Ammonia-based CO_2 capture, which uses aqueous ammonia as a solvent for CO_2 instead of amines, is another promising post-combustion option because it may have energy and cost advantages over the amine-based system.[5]

There have been various environmental concerns associated with using amines for CCS.[6] One that is the focus of this study is that amine scrubbing could create an air quality problem associated with its ammonia emissions. Ammonia is a significant precursor of $PM_{2.5}$,[7,8] which refers to particulate matter having a diameter of 2.5 μm and smaller. Exposures to $PM_{2.5}$ pollution are strongly associated with increases in mortality and morbidity.[9]

Another concern is that amine systems produce a hazardous waste. Amines react with acid gas impurities such as SO_2, SO_3, NO_x, and HCl to form corrosive heat-stable salts (HSS).[6,10] While some amines can be released from HSS for reuse by adding a strong alkali, the remaining HSS must be treated as a hazardous waste. In addition, amines emitted to the atmosphere may react with NO_x to form nitrosamines, which are known carcinogens. However, nitrosamines are broken down rapidly by photolysis under sunlight,[11] and nitrosamines were not detected in an experimental study on amines emitted by amine-based CO_2 capture technology.[12,13] Lastly, ammonia emissions may also increase nitrogen deposition. Ecosystems with excess nitrogen could suffer from eutrophication and soil acidification.[14,15]

The role of ammonia in $PM_{2.5}$ formation is largely determined by nonlinear interactions between SO_2, NO_x, NH_3, and their products.[7,8,16] A unit ammonia emission from CCS may result in highly variable impacts on $PM_{2.5}$ concentrations depending on the ambient concentrations of these species as shown in Figure S1 of the Supporting Information. Once emitted to the air, ammonia may remain in the gas phase if sulfuric acid and nitric acid are not available, which therefore causes no change in $PM_{2.5}$ concentrations. If unneutralized sulfuric acid exists, ammonia first reacts with it to form PM sulfate ($(NH_4)_2SO_4$). Because unneutralized sulfuric acid already exists overwhelmingly in the particle phase, this reaction increases $PM_{2.5}$ concentrations only marginally by replacing hydrogen with ammonium. If sulfate is neutralized, however, any remaining ammonia may form PM nitrate (NH_4NO_3) by reacting with nitric acid. The formation of ammonium nitrate may be limited either by ammonia or by nitric acid. When ammonia is the limiting reagent, a unit of ammonia emitted creates much more $PM_{2.5}$ mass by PM nitrate formation than by neutralizing sulfate. Because PM nitrate formation is favored at cold temperatures, ammonia emissions may create a significant amount of $PM_{2.5}$ especially in winter or at night. Therefore, changes in ammonia emissions will tend to have stronger impacts on $PM_{2.5}$ in regions where ammonia is limiting PM nitrate formation, which corresponds to cold temperatures, lower SO_2 emissions, higher NO_x emissions, and

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intermediate ammonia levels (sufficient to neutralize sulfate but limiting for ammonium nitrate formation). Such conditions occur regularly in the eastern United States in winter.[7,8,16]

Because the role of ammonia in $PM_{2.5}$ formation in the atmosphere is sensitive to ambient copollutants and atmospheric conditions, it is necessary to employ a chemical transport model to understand the impacts of CCS ammonia on ambient $PM_{2.5}$ concentrations. Although there have been studies looking into the environmental impacts of amine capture systems,[17−26] no study has been done yet to explore the actual physical and chemical interactions of the emitted ammonia in the atmosphere, which determine their consequences to society.

This study focuses on an amine system using monoethanolamine (MEA, C_2H_7NO), the most common solvent found in the literature, but the results are readily applicable to other post-combustion capture systems such as an ammonia-based process. We focus on the $PM_{2.5}$ impacts of the ammonia emissions themselves even though CCS may also reduce SO_2 emissions and, therefore, $PM_{2.5}$. We feel that this framing is cleaner and more decision-relevant for two reasons. First, independent of any decision to deploy CCS, the normal processes of air quality regulation[27−29] will continue to reduce SO_2 emissions. Therefore, attribution of these SO_2 reductions in the future involves considerable guesswork about the course of air quality regulation in future decades. Furthermore, once the decision to deploy CCS is made, the SO_2 reductions come either from CCS or air quality regulations, whereas regulators and operators are left with a separate decision about how much to control the associated ammonia emissions, which we seek to inform in our analysis. This study does not consider the potential contribution of amines themselves to $PM_{2.5}$ creation[12,30] due to the lack of data on emissions and atmospheric chemistry of amines. It has also been suggested that ammonia and/or amines contribute to the number concentration of ultrafine particles by enhancing the rates and frequencies of new particle formation events,[11,31−33] but this chemistry is still highly uncertain and is not considered here. Our calculations neglect potential effects of pH changes on organic $PM_{2.5}$ via acid-catalyzed oligomerization. However, the importance of this process is debated and uncertain[34] and has not been generally adopted in chemical transport models.

This study aims to evaluate the potential changes in $PM_{2.5}$ concentrations and resulting health impacts from amine scrubbing CCS in the United States. We estimated the ammonia emissions under an aggressive amine scrubbing deployment scenario in 2050. To demonstrate the potential for $PM_{2.5}$ impacts, we chose a CCS ammonia emission rate typical of current pilot plants, although these are substantially higher than design targets. Then, we simulated $PM_{2.5}$ concentrations with and without CCS ammonia for 2050. Several additional simulation analyses were carried out to test the sensitivity of our results to major uncertainties and to make our results applicable to a wide range of CCS ammonia emissions. Finally, the health impacts and associated social costs of the $PM_{2.5}$ changes were evaluated.

All monetary values in this study were converted to year 2010 U.S. dollars unless otherwise noted. GraphSketcher[35] was used to create Figures 1 and 2, and Matplotlib[36] was used to create all other figures.

AMMONIA EMISSIONS FROM AMINE SCRUBBING

Ammonia is created from the oxidative degradation of amines in the scrubbing process.[6,37] It has been reported that 30−50% of the amine lost in the process oxidizes to ammonia.[6,38] Differences in ammonia emissions between coal and natural gas plants have not been found in the literature. Because ammonia emissions are controllable by after-treatment, ammonia emissions probably will not depend on fuel type.

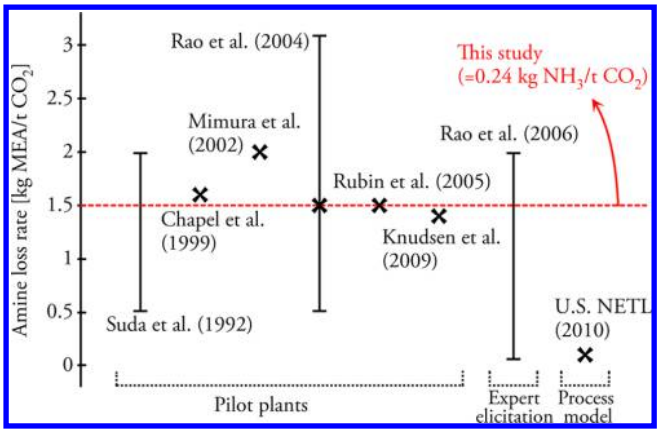

Figure 1. Amine loss rates reported or estimated in the literature. An ammonia emissions of 0.24 kg NH_3/t CO_2 was chosen for this study, which was reported in Rubin et al.[45] based on a coal power plant model assuming an amine loss rate of 1.5 kg MEA/t CO_2.[6] Note that the chosen rate is 15 times higher than the current U.S. NETL design target.

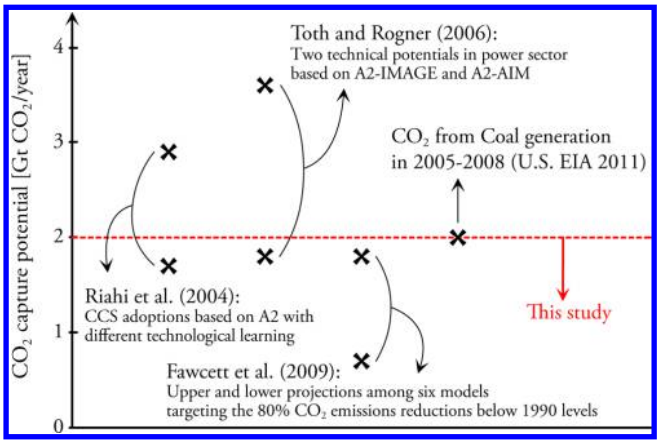

Figure 2. CCS potential in the United States. This study assumes that amine scrubbing CCS will capture 2.0 Gt CO_2/year in 2050.

Figure 1 summarizes the amine loss rates reported in the literature. Current pilot-scale applications show amine loss rates of 0.5−2 kg MEA/t CO_2. Pilot-scale natural gas power plants equipped with the Fluor Daniel Econamine system reported 1.5,[6,39] 1.6,[40] and 0.5−2 kg MEA/t CO_2.[41] A pilot-scale coal-fired power plant with an amine system reported losses of 1.4 kg MEA/t CO_2.[38]

However, other studies suggest that the amine loss could be smaller in the future. An expert elicitation study[42] reported that experts on amine-based CCS expected losses to be 0.05−2 kg MEA/t CO_2 by 2015 assuming modest R&D. A commercial power plant was able to reduce the solvent loss to 0.35 kg/t CO_2 using the amine solvent, KS-1, and further down to 0.1−0.2 kg/t CO_2 by modifying operational conditions.[43] On the basis of engineering modeling, U.S. NETL[44] projected a loss rate of 0.1 kg MEA/t CO_2.

This study selected an ammonia emission rate of 0.24 kg NH_3/t CO_2 from an amine loss rate of 1.5 kg MEA/t CO_2,

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which is based on a supercritical pulverized coal power plant model with amine scrubbing and including a typical water wash.[45] The performance of the plant model was reported in the IPCC Special Report on Carbon Capture and Storage.[2] However, it should be noted that our chosen value is substantially higher than the current U.S. NETL design target listed above. We have deliberately selected this value because it is supported by current operations, and we wish to evaluate whether CCS has the potential to create air quality problems. Because amines and ammonia are highly soluble in water, their emissions are technically controllable, and control strategies can be designed depending on the economics of and/or regulations on amine scrubbing.

The other important variable is the level of CCS deployment in 2050, which is difficult to estimate because amine scrubbing systems are only now being demonstrated at the commercial scale and carbon mitigation plans are not yet clear in the United States nor in most other nations. Figure 2 shows the context for the CCS deployment assumed in this study. On the basis of the IPCC SRES A2 scenario,[46] Toth and Rogner[47] estimated that the technical potential of CCS in the United States would be 3.6 Gt CO_2/year in the power sector in 2050 under the A2-IMAGE scenario and 1.8 Gt CO_2 under the A2-AIM scenario. Riahi et al.[48] reported that OECD90, defined as all members of OECD in 1990, would capture 3.5−5.9 Gt CO_2 in 2050. About 50% of this potential, or 1.7−2.9 Gt CO_2, would come from the United States, reflecting coal primary energy consumption in 2000.[49] The Energy Modeling Forum 22 study[50] reported that coal electricity production with CCS ranges from 2.8 to 6.7 EJ/year among six models for United States transition scenarios targeting 80% emissions reductions below 1990 levels. This would be equivalent to 0.7−1.8 Gt CO_2/year if they are captured from a coal plant similar to the plant model cited above. All these deployment levels are not limited to post-combustion technology or amine scrubbing systems.

To estimate the potential air quality problem from CCS ammonia, we assumed that amine scrubbing in the United States would capture 2.0 Gt CO_2/year from coal-fired power plants and large industrial facilities in 2050. This assumption represents a future with aggressive amine scrubbing deployment because the amount is similar to the CCS deployment levels comprised of all CCS technologies in the scenario studies mentioned above but is realized with only amine scrubbing. The amount of captured CO_2 we assumed is similar to the amount of CO_2 emitted by coal power plants alone annually from 2005 to 2008.[51] The CO_2 emissions from natural gas power plants were 320−360 Mt CO_2/year during the same period. Recent shale gas development and new air quality regulations may force old power plants to retire and result in more intensive use of natural gas in electricity generation. Although a natural gas combined cycle (NGCC) emits about half the carbon dioxide to generate a unit of electricity compared to conventional coal plants,[44,52] it would be necessary to equip a portion of the NGCC fleet with CCS to achieve large (~80%) GHG reductions.[50]

From the two factors assumed above, the NH_3 emissions per CO_2 captured of 0.24 kg NH_3/t CO_2 and the amount of CO_2 captured with amine scrubbing of 2.0 Gt CO_2/year, the amount of ammonia emitted from amine scrubbing CCS was estimated to be 480 Gg NH_3/year. This amount of CCS ammonia is ~10% of the current anthropogenic ammonia emissions in the United States, which are 3.5−4.0 Tg NH_3/year.[53]

Non-CCS NH_3 emissions are larger in spring and summer than in other seasons because animal husbandry and synthetic fertilizer application are dominant sources of NH_3.[53] Thus, the CCS NH_3 emissions would result in a relatively larger increase of NH_3 in winter than in summer, precisely when $PM_{2.5}$ concentrations are most sensitive to ammonia emissions.

EMISSIONS SCENARIOS AND SENSITIVITY SIMULATIONS

Main Scenarios. We have designed three main scenarios to explore the role of CCS ammonia based on reasonable current and future levels of ambient SO_2, NO_x, and non-CCS NH_3 as shown in Figure S2 of the Supporting Information. We focused on these three species because the effect of CCS ammonia on ambient $PM_{2.5}$ depends on their relative availability as discussed above.

The first one is *Current*, which corresponds to the current air quality resulting from the emissions database of year 2005,[54] which was built for a U.S. EPA regulatory impact assessment.[27] The database includes emissions from Canada and Mexico and from marine vessels over the oceans. However, in the following scenarios, we did not change these emissions but only those emitted on land over the contiguous U.S. domain.

Next, *No-CCS-NH_3 2050* represents a future with significant CCS deployment but without any CCS NH_3 emissions. Because the future emissions of SO_2, NO_x, and NH_3 would be reduced by CCS or normal air quality regulation,[27−29] we assumed that the net impact of these factors is a reduction of 85% of SO_2 point emissions relative to 2005, 50% of SO_2 area emissions, 50% of NO_x emissions, and 30% of NH_3 emissions. Amine-based CCS removes almost all SO_2 because SO_2 reacts with amines to form heat stable salts. Therefore, a future with high CCS adoption would easily achieve an 85% reduction of SO_2 point emissions by 2050. Although more difficult than SO_2 point sources, SO_2 area emissions and NO_x emissions may also be substantially reduced. Although NH_3 emissions are not currently regulated, a 30% reduction in NH_3 emissions is assumed because NH_3 reduction is a cost-effective $PM_{2.5}$ control measure and regulatory interest in it has increased.[7,55,56]

Lastly, *CCS-NH_3 2050* is the same as the *No-CCS-NH_3 2050* scenario just described but with the additional 480 Gg NH_3/year of CCS ammonia as estimated above. Assuming large SO_2 sources represent the likely locations of future CCS plants, either coal plants or other large industrial sources, we added CCS NH_3 to the largest SO_2 point sources, which in total emit the same amount of SO_2 emissions by electricity generation in our 2005 emissions inventory.[54] We distributed CCS ammonia to the SO_2 point sources proportionally to their SO_2 emissions on an hourly basis throughout the year.

Sensitivity to Future Emissions, to CCS Ammonia Emissions and Locations, and to Climate Change. We did sensitivity analyses to address four major uncertainties associated with our main scenarios developed above. One is the future emissions of SO_2, NO_x, and non-CCS NH_3. In order to explore this uncertainty, two more sets of scenarios are developed—High-sensitivity and Low-sensitivity—as shown in Table S1 of the Supportiung Information. Because it is computationally too expensive to run the Comprehensive Air Quality Model with Extensions (CAMx) with many possible emissions combinations, the two scenarios are developed based on the understanding of inorganic $PM_{2.5}$ thermodynamics discussed above. High-sensitivity represents a future combination of SO_2, NO_x, and non-CCS NH_3 emissions that would

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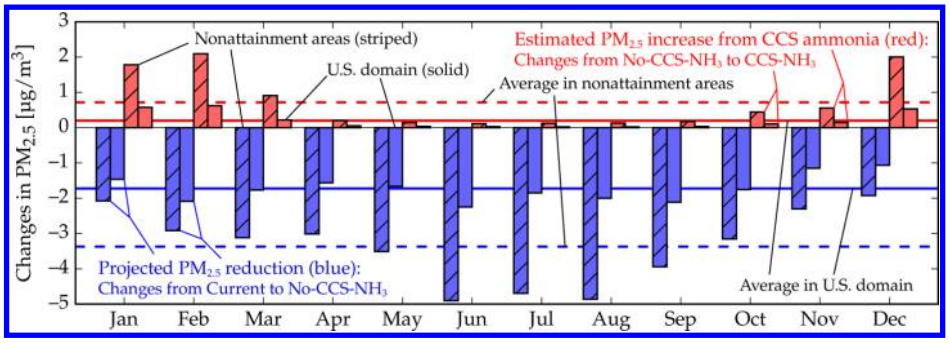

Figure 3. Monthly changes in $PM_{2.5}$ concentrations. United States domain is the contiguous United States in the simulation grid. Estimated $PM_{2.5}$ increases from CCS ammonia (red) represent a future scenario that captures 2.0 Gt CO_2/year at 0.24 kg NH_3/t CO_2, an ammonia emission rate typical of current pilot plants.

result in more $PM_{2.5}$ formation per unit CCS ammonia emissions, and Low-sensitivity represents one that would result in less $PM_{2.5}$ formation. SO_2 is assumed to decrease by 95% for High-sensitivity, considering a thorough reduction of SO_2 by amine scrubbing and other measures, and by 70% for Low-sensitivity, considering a future that would capture a substantial amount of CO_2 from natural-gas burning facilities while keeping a part of coal generation without CCS. NO_x is assumed to decrease by 70% for Low-sensitivity considering aggressive reduction efforts and by 20% for High-sensitivity considering modest control efforts. Lastly, non-CCS NH_3 is assumed to be reduced by 50% for High-sensitivity considering the cost effectiveness of NH_3 control[7,55,56] and by 0% for Low-sensitivity considering no action for NH_3 control.

The other major uncertainty is the amount of ammonia emitted from CCS. Despite nonlinearities in the thermodynamics of inorganic $PM_{2.5}$, we assume that the impacts will be approximately proportional to emissions. To test the linearity of impacts over the range of possible CCS ammonia emissions, CAMx was run for CCS-NH_3 2050, Low-sensitivity, and High-sensitivity scenarios that have 6.25%, 25%, 100%, and 200% of the CCS ammonia emissions assumed in CCS-NH_3 2050 scenario.

Also, in order to test the sensitivity of our results to the spatial distribution of CCS NH_3, we performed an additional sensitivity simulation in which we added the CCS ammonia to large NO_x point sources. This also allows us to look at the case of deploying CCS to natural gas power plants and other large natural gas burning facilities as well as coal plants.

Lastly, future temperature increase may affect our results. Under a strong warming climate scenario (Representative Concentration Pathways 8.5), climate models estimate the mean United States temperature may increase by 2 °C by 2050 on average.[57] We analyzed a case in which we imposed a 2 °C increase uniformly in space and time on the 2050 meteorology as a sensitivity scenario for this potential effect.

METHODS

Air Quality Simulations. We used the Comprehensive Air Quality Model with Extensions (CAMx) version 5.41[58] to simulate the air quality of the scenarios. CAMx is a state-of-the-art CTM that simulates horizontal and vertical advection, dispersion, wet and dry deposition, gas and liquid phase chemistry, and aerosol formation and growth. We used the CAMx air quality modeling platform, which was evaluated as a part of a U.S. EPA regulatory impact analysis.[27] The platform covers the continental United States with 36 km × 36 km horizontal grid resolution and 14 vertical layers reaching up to 16 km, which is fine enough for $PM_{2.5}$ human health impact analysis.[59] The initial and boundary conditions were provided by a global chemical transport model.[60] For inorganic $PM_{2.5}$ species, the modeling system showed a good performance (a 10−30% bias compared to observations).[60] Additional evaluations are summarized in the Supporting Information. Figure S3 of the Supporting Information presents CAMx results, showing simulated $PM_{2.5}$ concentrations with our 2005 database.

We ran CAMx for an entire year for each of the three main scenarios. However, due to high computational costs, we limited our sensitivity cases to four months (January, April, July, and October). We ran 7 days before each simulation period as ramp-up to minimize the effect from initial conditions. Special attention is paid to the $PM_{2.5}$ nonattainment areas designated for 1997 and 2006 standards[61] (Figure S4, Supporting Information), which are referred to here as $PM_{2.5}$-burdened areas.

Public Health Impacts. The health impacts from CCS-related $PM_{2.5}$ increases were quantified using standard methods adopted by the U.S. EPA.[62,63] First, for each model grid cell, we estimated the changes in mortality rate given the changes in annual-average $PM_{2.5}$ concentrations associated with air quality improvements in 2050 (Current to No-CCS-NH_3 2050) and with CCS ammonia impacts (No-CCS-NH_3 2050 to CCS-NH_3 2050). We used the concentration−response relations from two landmark cohort-based PM mortality studies; for each $PM_{2.5}$ concentration increase of 10 μg $PM_{2.5}/m^3$. Lepeule et al.[64] reported that all-cause mortality increases by 14% (95% confidence interval: 7−22%), and Krewski et al.[65] reported 6% (95% confidence interval: 4−8%). We quantified only the $PM_{2.5}$ impact on mortality as this accounts for more than 90% of monetized costs.[62,63,66] We also assumed that all $PM_{2.5}$ species have the same health effect on a mass basis because there is not sufficient epidemiological evidence that supports a metric better than $PM_{2.5}$ mass.[62,67] Second, for each grid cell, we estimated the number of premature deaths by multiplying population by the changed mortality rates. We used the year 2040 population forecast provided in the environmental Benefits Mapping and Analysis Program (BenMAP)[68] based on Woods & Poole Economics, Inc.,[69] which is 37% larger than the population in 2010. Although our scenarios were developed for 2050, we used the BenMAP population forecast for 2040, as no later forecast is available. Finally, we multiplied the number of premature deaths by the value of a statistical life (VSL), which is people's willingness-to-pay to avoid the risk of

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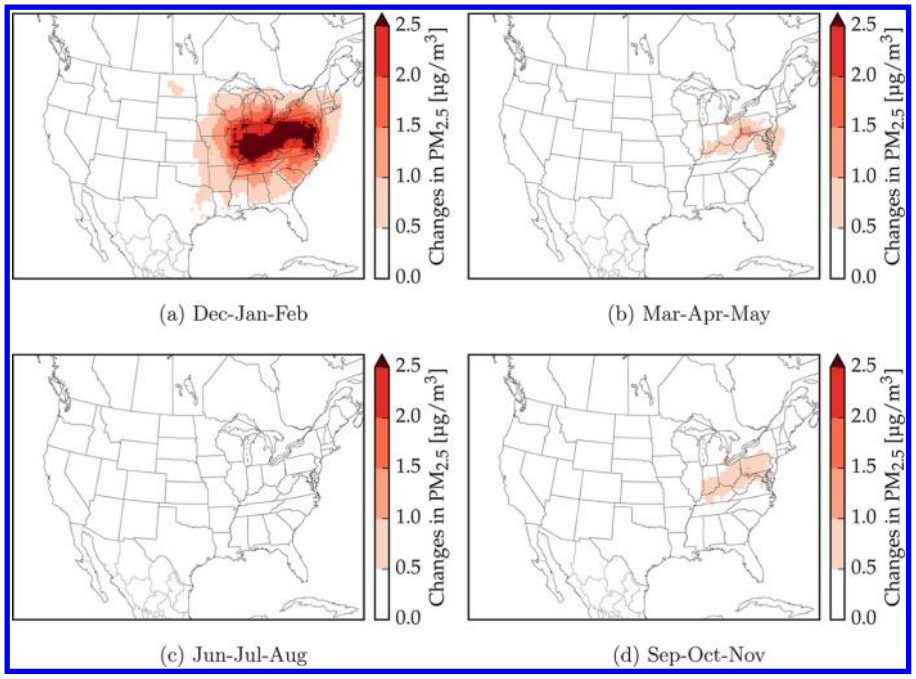

Figure 4. Estimated increase in $PM_{2.5}$ concentrations due to CCS ammonia in 2050. $PM_{2.5}$ increase is most sensitive to ammonia emissions during wintertime and relatively insensitive during summertime.

premature death. We used a Weibull distribution having a mean VSL of $8 million, which is recommended by the U.S. EPA.[70] This value is derived primarily based on "revealed preference" studies that use marketplace behaviors to infer the willingness-to-pay of individuals to avoid mortality risks and "stated preference" studies that surveys people how they would choose in various hypothetical situations of different mortality risks. We carried out Monte Carlo simulations, each with 5000 iterations, to quantify uncertainties surrounding the concentration−response relation and VSL.

RESULTS

$PM_{2.5}$ Impacts. The monthly changes in $PM_{2.5}$ concentrations are presented in Figure 3. Nonattainment regions show larger changes than the entire United States domain because changes in emissions occur relatively nearby to nonattainment regions. The assumed air quality controls between now and 2050 result in a significant reduction of 3.4 $\mu g/m^3$ in $PM_{2.5}$ (Current to No-CCS-NH_3 2050) for the annual average over nonattainment areas and 1.7 $\mu g/m^3$ over the contiguous United States domain. The annual $PM_{2.5}$ concentration increases due to CCS (between No-CCS-NH_3 2050 and CCS-NH_3 2050) are smaller but significant: 0.72 $\mu g/m^3$ over nonattainment areas and 0.20 $\mu g/m^3$ over the United States domain. To better visualize the CTM results, difference maps of $PM_{2.5}$ concentrations are presented in Figure 4 and Figure S5 of the Supporting Information. A summary of the PM concentrations of all scenarios is presented in Table S2 of the Supporting Information.

Whereas the projected $PM_{2.5}$ reduction is the least in January and the largest in July (Figure S5, Supporting Information), the $PM_{2.5}$ increase from CCS ammonia is the largest in January and the lowest in July (Figure 4). This result agrees with the known $PM_{2.5}$ thermodynamics discussed above. Wintertime $PM_{2.5}$ is sensitive to additional ammonia emissions, and summertime $PM_{2.5}$ is generally sensitive to reductions in SO_2 emissions.[7,8,16] In winter, the impacts of CCS ammonia offset 86% of the projected future air quality improvements for the nonattainment areas and 38% for the United States domain. In summer, by contrast, CCS ammonia impacts on $PM_{2.5}$ concentrations are negligible. The $PM_{2.5}$ increases in nonattainment areas in spring and fall by CCS ammonia are about 20% of the increase in winter.

$PM_{2.5}$ concentrations increase linearly over a wide range of CCS ammonia for all four months as shown in Figure 5 and Figure S6 of the Supporting Information. The slope in January is 3.4 μg $PM_{2.5}/m^3$ per Tg NH_3/year for nonattainment areas and 1.1 μg $PM_{2.5}/m^3$ per Tg NH_3/year for the United States domain. The sensitivity of the $PM_{2.5}$ increase to CCS ammonia is also linear in other months, although the slopes are shallower. Figure S6a of the Supporting Information shows that the impact of CCS ammonia on $PM_{2.5}$ has a modest sensitivity to the mix of other pollutants: SO_2, NO_x, and non-CCS NH_3. In addition, our results are not sensitive to the location of CCS ammonia and the temperature increase as shown in Figures S7 and S8 of the Supporting Information.

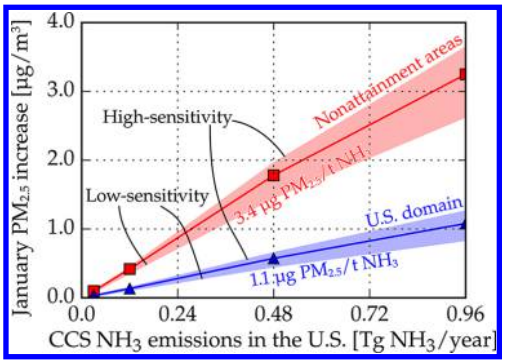

Figure 5. Sensitivity analysis of January $PM_{2.5}$ to future emissions of copollutants (SO_2, NO_x, and non-CCS NH_3) and the amount of ammonia emitted by amine scrubbing.

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Estimation and Valuation of Premature Deaths. The projected changes in annual premature deaths and their valuations are presented in Figure S9 of the Supporting Information. Two mean estimates calculated based on the two epidemiological studies are presented as an estimated range here. Comparing improved air quality in 2050 without CCS ammonia to the present, the number of annual premature deaths is expected to decrease by 51,000−120,000, which is evaluated at $410 billion to $930 billion. Under the increased $PM_{2.5}$ from CCS ammonia, the number of annual premature deaths attributed to CCS ammonia is estimated to be 7600−17,000, a social cost of $61 billion to $140 billion. Given the seasonality of the $PM_{2.5}$ response discussed previously, 68% of the annual-average $PM_{2.5}$ increase resulted from wintertime $PM_{2.5}$ changes with a negligible contribution from summertime changes.

On the basis of these results, the per unit social health costs of CCS ammonia is calculated to be $130,000−280,000/t NH_3. Wintertime CCS NH_3 costs are higher at $340,000−770,000/t NH_3. On the basis of CO_2 captured, the costs of CCS ammonia are calculated to be $31−68/t CO_2 per year and $82−186/t CO_2 during the winter.

DISCUSSION

This paper has explored the air quality and human health impacts that could be imposed by ammonia emissions from amine-based post-combustion CO_2 capture processes. First, we estimated potential ammonia emissions based on current emission factors and analyzed the possible changes in concentrations of fine particulate matter ($PM_{2.5}$), of which ammonia is a major precursor, with a state-of-science chemical transport model, CAMx. Then, we estimated the premature mortality associated with the $PM_{2.5}$ formation and monetized the impacts. We also explored major uncertainties surrounding our results.

We found that ammonia emissions from amine-based carbon capture systems at a rate typical of current pilot plants would create a significant increase in $PM_{2.5}$ concentrations, resulting in worrisome public health impacts, although these could be lessened greatly if the current U.S. NETL design target[44] is achieved. With an emission factor of 0.24 kg NH_3/t CO_2, a substantial deployment of amine scrubbing to capture 2Gt CO_2/year would emit 480 Gg NH_3/year in the United States. This amounts to 14% of annual ammonia emissions or 34% of winter emissions of the United States in 2005. This scenario is intentionally chosen to demonstrate the potential for significant $PM_{2.5}$ impacts, but sensitivity to differing emissions rates was analyzed. Such emissions would increase the winter $PM_{2.5}$ concentrations in nonattainment areas by 2.0 $\mu g/m^3$ on average and up to 4.3 $\mu g/m^3$ in some locations.

This work has examined the key uncertainties governing the impacts of CCS NH_3, which are summarized in Table S3 of the Supporting Information. Because CCS ammonia emissions are uncertain and because ammonia impacts depend on the levels of copollutants available from other sources, we performed a sensitivity analysis over a wide range of CCS ammonia emissions and potential emissions of copollutants (SO_2, NO_x, and non-CCS NH_3) as shown in Figure S6 of the Supporting Information. We showed that $PM_{2.5}$ impacts are fairly linear with CCS ammonia emissions, and concentrations increase with CCS ammonia at a rate of 3.4 $\mu g/m^3$ per Tg NH_3 in nonattainment areas in January. The $PM_{2.5}$ increase in nonattainment areas in January could vary by about 20% depending upon the future emissions of the copollutants. The approximately linear response is useful. Because ammonia emissions from future systems may be lower than current pilot plants, the $PM_{2.5}$ impacts considered here may be scaled accordingly, noting that Figure S6 of the Supporting Information shows somewhat higher unit impacts for smaller CCS emissions. In addition, our results are not sensitive to the details of how CCS ammonia emissions are distributed around the nation nor to potentially warmer future temperatures (Figures S7 and S8, Supporting Information). As is always the case with $PM_{2.5}$ health valuations, uncertainties in concentration−response relations and VSL are significant (−90% to +160%).

If ammonia emissions were allowed at a level typical of current CCS pilot plants, the $PM_{2.5}$ increase would significantly compromise air quality. Especially, the wintertime $PM_{2.5}$ increase can offset in nonattainment areas 86% of all future air quality improvements including the contribution of CCS to large SO_2 reductions. An increased $PM_{2.5}$ concentration of 2.0 $\mu g/m^3$ is significant when one considers that current nonattainment areas often seek to cut 1−2 $\mu g/m^3$ to meet the $PM_{2.5}$ National Ambient Air Quality Standards (NAAQS). It may also cause other areas to slip into nonattainment, especially if more stringent NAAQS standards are adopted in the future. If future amine scrubbing plants are operated with lower ammonia emissions, the impact will be lower accordingly. For example, the current U.S. NETL design target,[44] which emits 15 times less ammonia than current pilot plots, would result in an average increase of 0.1 μg $PM_{2.5}/m^3$ for nonattainment areas in January.

Our per-tonne costs, $130,000−280,000/t NH_3, are somewhat larger than those in the literature. This is likely because $PM_{2.5}$ formation is more sensitive to ammonia emissions in the atmosphere in 2050 than we assumed, and we used the 2040 population forecast, which is 37% larger than the 2010 population. For comparison, we converted the following literature-reported ammonia social costs to 2010 U.S. dollars and metric ton from their reported units. With the Response Surface Model,[71] an air quality model, Fann et al.[72] reported social costs per ton of NH_3 emitted from mobile sources were $120,000/t NH_3 at the national level and $52,000−170,000/t NH_3 over nine urban areas based on a concentration response relation[73] similar to Lepeule et al.[64] For area source NH_3, they estimated a social health cost of $46,000/ton NH_3 at the national level. With a reduced-form air quality model, Muller et al.[74] reported the costs of NH_3 for all US counties using a VSL similar to this study and a concentration−response relation[75] similar to Krewski et al.[65] They vary from $2200/t NH_3 (fifth percentile) to $130,000/t NH_3 (95th percentile) with a mean of $38,000/t NH_3.

In the absence of controls on ammonia emissions, the $PM_{2.5}$ problem resulting from CCS ammonia emissions could be compared to the climate benefits of the avoided CO_2 emissions. Using a standard method of valuing $PM_{2.5}$ mortality, we estimated the social cost of CCS ammonia at $31−68 per tonne CO_2 captured. Estimates of the social cost of carbon, which includes CO_2 damages on human health, property, and ecosystem services, are uncertain and vary widely, but a United States government interagency working group estimated the social cost of carbon in 2050 to be $28−102/t CO_2.[76] When compared to these estimates, the public health impacts from CCS ammonia emissions are significant in comparison to the climate benefits from CO_2 emissions reductions from CCS and

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deserve close attention in the future. CCS ammonia impacts could be minimized compared to CO_2 benefits by reducing CCS NH_3 emission factors below those used here.

Operators of CCS facilities have a natural incentive to reduce amine losses. For a solvent loss rate of 1.5 kg MEA/t CO_2 and an assumed amine solvent cost of \$2250/t MEA,[44] the amine consumption costs about \$3.4/t CO_2. However, our analysis shows that the $PM_{2.5}$ social costs are still much higher than the private costs borne by the operators in the form of solvent makeup. Therefore, it makes sense for regulators to impose limits on ammonia and amine emissions from CCS in order to protect the public interest. Because 68% of the burden occurs in winter and virtually none during the summer, it could be considered to enforce more stringent ammonia controls on a seasonal basis.

The concerns noted here suggest a need to proceed cautiously, but the air quality impacts of CCS ammonia are not necessarily prohibitive of the technology. Because ammonia is highly soluble in water, it is not technically difficult to control by installing more or better water wash units. Water wash units are already included in plant design mainly to reduce solvent loss from mechanical entrainment and evaporation.[44] Water wash systems could be better designed to minimize ammonia and amine emissions to the atmosphere in addition to the current purpose of minimizing solvent losses. If CCS ammonia is managed, for example, at the current ammonia control level of the selective catalytic reduction system (2−10 ppm), our estimate for the $PM_{2.5}$ impact from CCS ammonia is reduced by a factor of 10.

We based our analysis on MEA systems because this capture technology is reasonably well understood, but the results are readily applicable to other post-combustion capture systems such as an ammonia-based process. Although little information is currently available about ammonia leakage from such systems, the impacts per unit ammonia emitted could be applied to these systems given such data. Because the material cost of ammonia is much lower than MEA in terms of solvent costs per tonne of CO_2 captured,[77] an ammonia-based CCS power plant may afford to lose more solvent to the atmosphere than an amine-based one. Therefore, there would be an even stronger need for regulatory intervention to protect the public health.

In summary, widespread deployment of CCS technology could result in significant unwanted increases in $PM_{2.5}$ levels and potentially other impacts on air quality as well. There is a need for regulators to be pro-active in considering appropriate emissions-based standards to avoid such an outcome. Currently, there is no federal regulation on ammonia emissions from power plants. Emissions-based standards low enough to prevent significant air quality degradation will incur some cost but should be technically feasible, and the impact assessment performed here provides quantitative guidance for what level of control is appropriate.

■ ASSOCIATED CONTENT

Supporting Information

Supplemental figures and tables and additional evaluation of the CAMx air quality modeling platform. This material is available free of charge via the Internet at http://pubs.acs.org.

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Notes

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