frontiers
in Environmental Science

ORIGINAL RESEARCH
published: 29 March 2022
doi: 10.3389/fenvs.2022.867908

# Modeling Reactive Ammonia Uptake by Secondary Organic Aerosol in a Changing Climate: A WRF-CMAQ Evaluation

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In addition to the well-constrained inorganic acid-base chemistry of ammonia resulting in fine particulate matter ($PM_{2.5}$) formation, ammonia also reacts with certain organic compounds in secondary organic aerosol (SOA) to produce less basic nitrogen-containing organic compounds. In this study, the potential meteorology and air quality impacts of the heterogeneous uptake of $NH_3$ by SOA are investigated using the WRF-CMAQ two-way coupled model, which calculates the two-way radiative forcing feedback caused by aerosol between meteorology and chemistry in a single simulation. Simulations with and without the $NH_3$-SOA uptake are performed over the contiguous US for July 2014 and July 2050 under the RCP 8.5 IPCC scenario to study the potential impact of climate change. A comparison with multiple observation network data shows that the $NH_3$-SOA uptake improves the model performance for $PM_{2.5}$ prediction (bias reduced from −22% to −17%), especially the underestimation of organic carbon over the Southeastern US (bias reduced from −17% to −7%). Secondly, the addition of the $NH_3$-SOA chemistry significantly impacts the concentration of $NH_3$ and $NH_4^+$, thus affecting the modeled particle acidity. Including the $NH_3$-SOA uptake also impacts the meteorological conditions through the WRF-CMAQ two-way feedback. Moreover, the impact on meteorological conditions results in different windspeed or dispersion conditions, thus affecting air quality predictions. Finally, simulations including the $NH_3$-SOA uptake under the warmer climate conditions of 2050 show a smaller impact on air quality predictions than it does under current climate conditions. This study confirms the importance and necessity of including $NH_3$-SOA chemistry in air quality predictions.

**Keywords: ammonia, secondary organic aerosol (SOA), particulate matter, ammonium nitrate (AN), climate change, coupled meteorological and air quality modeling**

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***Specialty section:***
*This article was submitted to Atmosphere and Climate, a section of the journal Frontiers in Environmental Science*

***Received:*** *01 February 2022*
***Accepted:*** *11 March 2022*
***Published:*** *29 March 2022*

***Citation:***
*Zhu S, Wu K, Nizkorodov SA and Dabdub D (2022) Modeling Reactive Ammonia Uptake by Secondary Organic Aerosol in a Changing Climate: A WRF-CMAQ Evaluation. Front. Environ. Sci. 10:867908. doi: 10.3389/fenvs.2022.867908*

## 1 INTRODUCTION

As an important atmospheric trace gas, ammonia ($NH_3$) is found in various sources of natural and anthropogenic emissions (Bouwman et al., 1997). Globally, roughly half of the $NH_3$ emissions originates from agricultural activities, for example, satellite observations show ammonia hot spots in major agricultural areas (Clarisse et al., 2009). As a highly water-soluble molecule, the majority of atmospheric ammonia is lost by deposition on wet particles and aquatic surfaces (Dentener et al.,

2006) as part of the global nitrogen cycle. Ammonia has long been considered an important precursor to the inorganic component of fine particulate matter ($PM_{2.5}$, particles with sizes below 2.5 µm that penetrate easily the respiratory tract). Ammonia is converted into inorganic ammonium salts with low volatility by neutralizing inorganic acids commonly found in polluted air, such as sulfuric acid and nitric acid. Such salts accumulate as solids in $PM_{2.5}$ (Seinfeld and Pandis 2016). Ammonium nitrate and ammonium sulfate constitute an important fraction of the total $PM_{2.5}$ in many geographical areas (Behera and Sharma 2010).

In addition to the well-constrained inorganic acid-base chemistry of ammonia leading to $PM_{2.5}$ formation, there is experimental evidence that ammonia reacts with certain organic compounds in secondary organic aerosol (SOA) leading to the formation of nitrogen-containing organic compounds (NOC) (Bones et al., 2010). SOA produced by photooxidation of biogenic and anthropogenic volatile organic compounds (VOCs) represents a large fraction of $PM_{2.5}$. Although considerable uncertainty still exists over chemical reactions between $NH_3$ and SOA, the general mechanism involves the reaction of ammonia with carbonyls within the organic particles to produce relatively unstable imines and amines, which may be stabilized by intramolecular cyclization into heteroaromatic NOC based on imidazole, pyrrole, indole, etc., (Montoya-Aguilera et al., 2018). These reactions change the chemical composition of organic particles and their physical properties, such as color, viscosity, and condensed-phase diffusivity (Laskin, Laskin, and Nizkorodov 2015; Li et al., 2015; Bell et al., 2017; Liu et al., 2018).

Until recently, interactions between $NH_3$ and SOA (SOA-$NH_3$ uptake) were not considered in air quality models. In our previous work to incorporate such interactions into air quality models, a significant decrease was found in predicted $NH_3$ and inorganic $PM_{2.5}$ concentration in a regional study (Horne et al., 2018) on the South Coast Air Basin of California (SoCAB) using the UCI-CIT model (Nguyen and Dabdub 2002). Furthermore, we found a significant SOA concentration increase in the southeast of the US during the summertime in a national-scale study (Zhu et al., 2018) resulting from enhanced acid-catalyzed reactions (Pye et al., 2013) when the SOA-$NH_3$ uptake was incorporated into the Community Multiscale Air Quality (CMAQ v5.2) model (Byun and Schere 2006). In general, the ammonia loss to the SOA uptake is smaller than the traditional inorganic acidic uptake pathway. However, there are situations in which the SOA-$NH_3$ uptake becomes competitive and has a modest impact on organic aerosol and $PM_{2.5}$ concentrations.

An air quality model can either be an offline model where the meteorology is specified from a foreign source or an online model where the meteorology is adjusted based on the chemical constituents within the model. Our previous SOA-$NH_3$ uptake sensitivity studies (Horne et al., 2018; Zhu et al., 2018) were based on offline air quality models, which means no feedback was considered between pollutant concentration and meteorological conditions (Grell and Baklanov 2011). However, field experiments and satellite observation have shown that chemistry-atmosphere feedbacks play an essential role in the Earth system (Kaufman and Fraser 1997; Rosenfeld et al., 2007, 2008; Pausata et al., 2015). On the one hand, meteorological conditions dictate both the formation and transport of air pollutants. On the other hand, chemical species can also influence the meteorological parameters by changing the atmospheric radiation budget and through cloud formation. In this study, a more comprehensive air quality impact study of the effects of SOA-$NH_3$ uptake is conducted by including the chemistry-atmosphere feedbacks using the two-way coupled Weather Research and Forecasting—Community Multi-scale Air Quality (WRF-CMAQ) model (Wong et al., 2012). Developed by the US Environmental Protection Agency (EPA), the CMAQ model is an offline model historically and is widely used for air quality simulations worldwide (X. H. Liu et al., 2010; Canty et al., 2015; Sharma et al., 2016; Matthias 2008; de Almeida Albuquerque et al., 2018). In recent years, it was further developed and coupled with the Weather Research and Forecasting (WRF) model to simulate the two-way radiative forcing feedbacks caused by the aerosol direct effect between chemistry and meteorology (Wong et al., 2012). This new WRF-CMAQ coupled model has been evaluated in several applications and used to investigate aerosol direct effects on meteorology (Wang et al., 2014; Gan et al., 2015; Hogrefe et al., 2015; Xing et al., 2016; Hong et al., 2017).

Many studies (Brasseur et al., 2006; Fiore et al., 2012; Lin, Penner, and Zhou 2016; Campbell et al., 2018; Zhu et al., 2019) have demonstrated that climate change could impact significantly the physical and chemical processes that govern the formation of atmospheric pollutants, including ammonia and SOA. Furthermore, both emissions of ammonia (Skjøth and Geels 2013) and VOC precursors (Constable et al., 1999) are expected to increase in the future due to the rising temperature as a result of climate change. Combining these two factors is likely to lead to significant changes in the mass concentration and chemical composition of SOA. As a preliminary study, the potential impact of the physical and chemical processes alone due to the implementation of the SOA-$NH_3$ uptake mechanism is investigated under the changing climate using the WRF-CMAQ model. The Representative Concentration Pathway (RCP) 8.5 (business-as-usual) dataset for the year 2050 (Bruyère et al., 2013; Monaghan et al., 2014) is used here to provide a midterm outlook and the worst scenario estimation. Our results show that the inclusion of the $NH_3$-SOA uptake under the warmer climate conditions of 2050 produces a smaller impact on air quality predictions than under current climate conditions. But overall, this study highlights the importance and necessity of including $NH_3$-SOA chemistry in air quality predictions.

## 2 METHODOLOGY

### 2.1 Model Description

This study uses the two-way coupled WRF-CMAQ (WRF v3.8 and CMAQ v5.2) as the base platform to integrate the SOA-$NH_3$ uptake process to conduct regional meteorology and air quality simulations. WRF is the meteorology driver of the model, while

CMAQ handles the air quality dynamics. A detailed description of the two-way coupled WRF-CMAQ model is presented in Wong et al. (2012). The publicly available version used in this study supports the Rapid and accurate Radiative Transfer Model for General Circulation Models (RRTMG) radiation scheme (Clough et al., 2005) for shortwave aerosol direct effects. It uses a core-shell model to perform the aerosol optics calculation. The aerosol indirect effects that result from interactions between aerosols and cloud formation are not considered in this version.

For WRF configuration, the Asymmetric Convective model version 2 (ACM2) (Pleim 2007) is used as the planetary boundary layer scheme with the Pleim-Xiu land surface model (Xiu and Pleim 2001). The Morrison double-moment scheme (Morrison, Thompson, and Tatarskii 2009) is used for the microphysics option of WRF, and version 2 of Kain-Fritsch convective parametrization (Kain 2004) is used for cumulus physics. Both longwave and shortwave radiations are solved by the RRTMG scheme (Iacono et al., 2008). For the current scenarios, WRF input is derived from the National Centers for Environmental Prediction (NCEP) North American Mesoscale Forecast System (NAM) 12 km analysis data (NCEP 2015). For the future (year 2050) climate scenarios, the RCP 8.5 National Center for Atmospheric Research (NCAR) Community Earth System Model (CESM) global bias-corrected CMIP5 dataset (Monaghan et al., 2014) is used to provide a midterm outlook and the worst scenario estimation, where CAM-Chem (Lamarque et al., 2012) is used for chemistry-climate modeling and MOZART as the chemical mechanism (Emmons et al., 2010).

The CMAQ model is configured using the Carbon Bond 2006 (CB06) chemical mechanism for gas-phase chemistry (Yarwood et al., 2010), including 127 species as detailed on the model's website (Adams 2017). The aerosol dynamics are solved by the sixth-generation CMAQ aerosol module (AERO6) (Appel et al., 2013), which includes 21 inorganic species and 34 organic species (28 SOA and 6 primary organic species) as detailed on the CMASWIKI website (Pye 2016). The modeling domain is the same as Zhu et al. (2018), which covers the contiguous US (CONUS) with a 12 km × 12 km horizontal-grid resolution and a 29-layers logarithmic vertical structure. The initial and boundary conditions are downscaled from the Model for Ozone and Related Chemical Tracers (MOZART v4.0) (Emmons et al., 2010). Emissions are generated based on the 2014 National Emissions Inventory (NEI) version 2 (US EPA 2018) and spatially/temporally resolved using the Sparse Matrix Operator Kernel Emission (SMOKE, version 4.5) processor (US EPA 2017). Biogenic emissions are calculated using the Biogenic Emission Inventory System (BEIS) (Schwede et al., 2005). As a preliminary study and only accounting for the change in climate alone, the same emissions are used for both 2014 and 2050 cases.

The SOA-$NH_3$ uptake mechanism is incorporated into the AERO6 module using the same method as our previous study (Zhu et al., 2018). In AERO6, all particles are considered spherical and internally mixed, which means a homogeneous distribution of all chemical substances within the particle. However, as the uptake coefficient used in this study is measured from pure SOA particles, how the uptake coefficient could be changed with SOA mass ratios within the particle is unknown. In the absence of better information, we assume that the uptake coefficient is proportional to the SOA mass fraction within the particle. In general, the uptake of $NH_3$ by SOA is calculated based on the representative wet surface area concentration of SOA ($S_{SOA}$) and the reactive uptake coefficient $\gamma$. The calculation of $S_{SOA}$ is based on the SOA mass ratios within the particle. Details of the calculation are presented in our previous work (Zhu et al., 2018). In general, the amount of $NH_3$ uptake by SOA is reduced from the bulk $NH_3$ concentration at each timestep before the all-other chemical mass-transfer calculation, and the mass of organic aerosol is kept constant during the process. The effective first-order rate constant for of the $NH_3$ uptake by SOA is calculated as follows:

**TABLE 1** | Simulation case definition.

| Conditions | 2014 | 2050 |
|---|---|---|
| No SOA-$NH_3$ uptake | Base_14 | Base_50 |
| With SOA-$NH_3$ uptake | UpTk_14 | UpTk_50 |

$$k = \gamma \times \frac{v_{NH3} \times S_{SOA}}{4}$$

where $v_{NH3}$ is the average speed of $NH_3$ molecules (609 $ms^{-1}$ at 298 K). This first-order rate constant is then multiplied by the gas-phase $NH_3$ concentration to determine the loss rate of $NH_3$ in each grid cell at each time step. As explained in Zhu et al. (2018) in this method, the direct change of SOA mass due to the $NH_3$ uptake is neglected to simplify the calculation. This assumption is supported by the experimental observation that SOA particles exposed to ammonia in a smog chamber did not change their size distribution but showed clear evidence of incorporation of organic nitrogen into the particles in online and offline mass spectra, as described in Horne et al. (2018). As the existing laboratory data are still insufficient to determine the exact uptake coefficient for individual SOA species, a range of uptake coefficient $\gamma$ is selected for sensitivity studies in our previous work (Zhu et al., 2018) between $\gamma = 10^{-5}$ to $\gamma = 10^{-3}$ based on the values reported (Y. J. Li et al., 2015). To avoid redundant sensitivity analysis, we only select the highest uptake coefficient $\gamma = 10^{-3}$ in this study to provide the largest possible impact estimation of the SOA-$NH_3$ uptake process.

In total, four simulations with two-way feedback are performed in this study, two for 2014 and two for 2050. For each year, there is one case considering the SOA-$NH_3$ uptake and one case without the uptake. **Table 1** summarizes the naming and definition of these six simulation cases. July is selected as the simulation period, with the first week discarded as a spin-up. Our previous study (Zhu et al., 2018) showed that SOA-$NH_3$ uptake produced an additional impact on acid-catalyzed SOA formation during the summer (Pye et al., 2013), suggesting that July is a good choice for this simulation.

**TABLE 2 |** Model performance statistics for $PM_{2.5}$, $NH_4^+$, and Organic Carbon (OC) under different scenarios. $PM_{2.5}$ and OC observation (Obs.) data are from three networks (IMPROVE, CSN and AQS); $NH_4^+$ observation data are from four networks (IMPROVE, CSN, CASTNET, and AQS). (Average—Avg.; Simulation—Sim.; Normalized Mean Bias—NMB; Normalized Mean Error—NME).

| Case | Avg. Obs. (μg/m$^3$) | Avg. Sim. (μg/m$^3$) | NMB (%) | NME (%) | Correlation (%) |
|---|---|---|---|---|---|
| — | **$PM_{2.5}$ (1,391 sites)** | | | | |
| Base_14 | 9.35 | 7.28 | −22.14 | 22.44 | 60.69 |
| UpTk_14 | 9.35 | 7.75 | −17.08 | 21.06 | 51.41 |
| — | **$NH_4^+$ (578 sites)** | | | | |
| Base_14 | 0.54 | 0.34 | −37.01 | 44.43 | 47.74 |
| UpTk_14 | 0.54 | 0.39 | −27.39 | 36.86 | 56.91 |
| — | **OC (484 sites)** | | | | |
| Base_14 | 1.57 | 1.29 | −17.51 | 24.88 | 44.08 |
| UpTk_14 | 1.57 | 1.46 | −6.96 | 24.83 | 49.09 |

## 2.2 Observational Data and Model Validation

Model validation and performance analysis are quantified using the Atmospheric Model Evaluation Tool (AMET) (Appel et al., 2011) developed by the US EPA. AMET organizes, provides consistency, and speeds up the evaluation process for operational meteorological and air quality model simulations. It can match the model output for particular locations to the corresponding observed values from one or more monitor networks. For meteorological model evaluation, observations are obtained from NCEP Meteorological Assimilation Data Ingest System (MADIS) (NCEP 2019) and Baseline Surface Radiation Network (BSRN) (AWI 2019). For air quality model evaluation, measurement network data provided by AMET includes AErosol RObotic NETwork (AERONET) (NASA 2019), Ammonia Monitoring Network (AMoN) (NADP 2019), Air Quality System (AQS) (US EPA 2019a), Clean Air Status and Trends Network (CASTNET) (US EPA 2019c), Chemical Speciation Network (CSN) (US EPA 2019b), Interagency Monitoring of Protected Visual Environments (IMPROVE) (USGS 2019a), National Atmospheric Deposition Program (NADP), National Air Pollution Surveillance (NAPS) (Canada 2019), South-Eastern Aerosol Research and Characterization (SEARCH) (USGS 2019b), Tropospheric Ozone Assessment Report (TOAR) (Schultz et al., 2017). The model performance is evaluated and compared for the two 2014 cases in **section 3.1**.

# 3 RESULTS AND DISCUSSION

In this section, the impact on model performance by including the $NH_3$-SOA chemistry is first presented. Then, the chemical and meteorological impacts of the new chemistry are further investigated for the two 2014 cases. Finally, the sensitivity variation resulting from climate change is discussed by comparing the 2014 and 2050 cases.

## 3.1 Impact on Model Performance

The model performance of the 2014 simulations with and without ammonia uptake is evaluated for $PM_{2.5}$, $NH_4^+$, and Organic Carbon (OC) against multiple observation networks detailed in **section 2.2** (**Table 2**). The $PM_{2.5}$ model performance of both Base_14 and UpTk_14 satisfies the recommended performance criteria proposed by Emery et al. (2017), with normalized mean bias (NMB) < ± 30%, normalized mean error (NME) <40% and correlation >40%. The $PM_{2.5}$ is underestimated by 22.14% in the base case, while the $NH_3$-SOA uptake process reduced such underestimation bias to 17.08% across the U.S. The overall model error is also reduced, as most model improvements occur over the southeastern US, as presented in **Figure 1A**. The model performance for $NH_4^+$ is also largely improved after including the $NH_3$-SOA uptake process, with model bias reduced from −37.01% to −27.39%, model error reduced from 44.43 to 36.86% and correlation increased from 47.74 to 56.91%. With the $NH_3$-SOA uptake process included in the model, the underestimation of OC is also significantly improved, and the model bias is reduced from −17.51% to −6.96%. With more than 60% of the observation sites show decreased model error, **Figure 1B** shows the OC prediction is largely improved over the Southern and Eastern US, especially for the northeastern metropolitan regions like New York and Washington D.C. As underestimating OC is a common problem for air quality models (J. Li et al., 2017) such an improvement on OC prediction is promising. It confirms the importance of including the $NH_3$-SOA chemistry in air quality models. However, the $NH_3$-SOA uptake process only has little to no impact on the meteorological side of model performance [**Supplementary Figure S1** in the Supporting Information (SI)], such as the temperature and relative humidity (RH), only slight improvement is found for the stational pressure estimation. Although the overall averaged impact is minimum, impacts on some specific locations could still be visible and could influence air pollutant distribution through feedback. Those feedback impacts are driven by the changes in the short-wave radiation balance due to the changes in aerosol concentrations (Clough et al., 2005). In the following section, the impact on meteorological parameters will be more carefully examined.

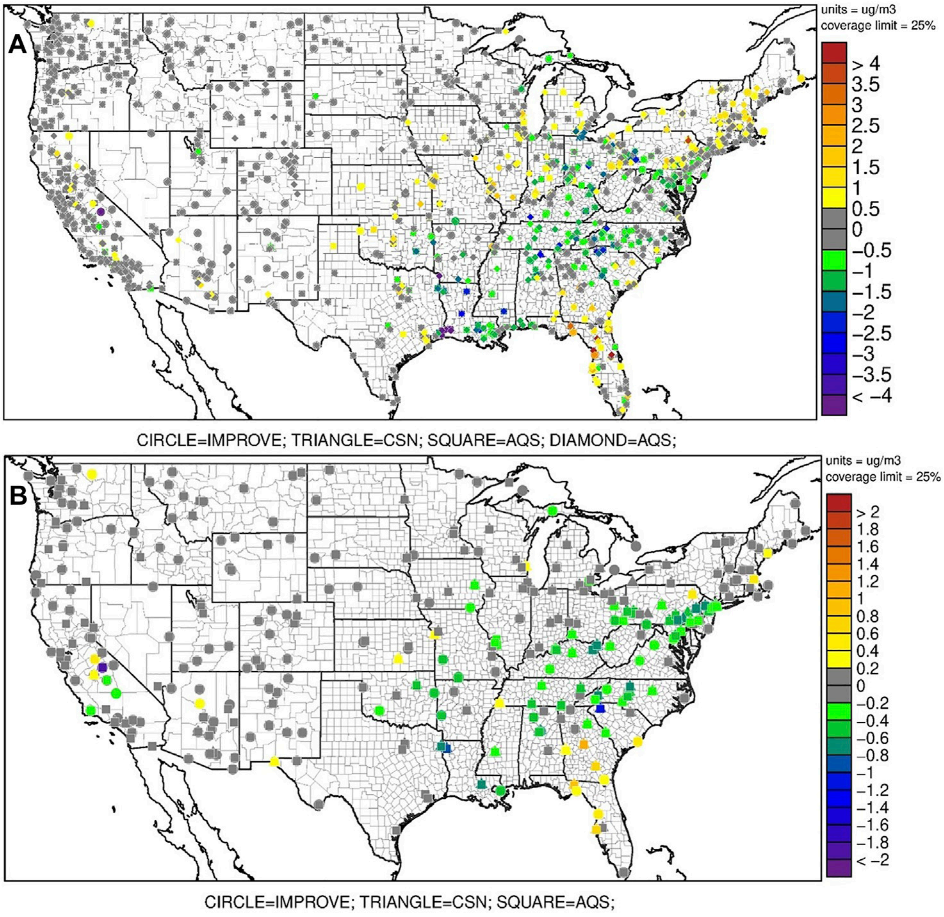

**FIGURE 1 |** Changes in model errors between Base_14 and UpTk_14 for **(A)** $PM_{2.5}$ and **(B)** OC at each observation site. Negative values indicate reduced model errors in UpTk_14 compares to Base-14. For **(A)**, the SQUARE (AQS sites) only records daily mean value, the DIAMOND (AQS sites) records hourly data.

## 3.2 Impact on Modeled NH3 and PM2.5

By comparing simulations UpTk_14 and Base_14, more details can be revealed on the impact on $NH_3$ and $PM_{2.5}$. **Figure 2A** shows the time-averaged $NH_3$ concentration over the simulation domain for Base_14. Hot spots can be identified in regions with high agricultural $NH_3$ emissions (e.g., California, North Carolina, Iowa, and Idaho, **Supplementary Figure S2** in the SI) or regions with intense wildfire activities (e.g., Washington, Oregon, and Texas, **Supplementary Figure S3** in the SI). **Figure 2B** shows the $NH_3$ concentration difference between UpTk_14 and Base_14. The inclusion of the $NH_3$-SOA uptake process results in an overall decrease in $NH_3$ concentration throughout the domain, with the most significant decrease of almost 30% in California. The enhanced $NH_3$ reduction for California is most likely due to the unique spatial overlap between domains of high $NH_3$ and SOA concentrations (**Supplementary Figure S4** in the SI). Among the states with top $NH_3$ emissions, Iowa is the only one increasing $NH_3$ concentration after including the $NH_3$-SOA uptake. A more detailed investigation shows this is most likely due to the meteorological feedback resulting from the changing air quality conditions. **Figure 2D** shows the changes of 10-m wind speed for Iowa after the addition of $NH_3$-SOA uptake. The overall reduction in wind speed indicates a loss in dispersion capability, especially over the northwest part of the state, where most of the $NH_3$ emissions are located (**Figure 2C**). As a result, more $NH_3$ is accumulated over those regions, resulting in the unusual increase of $NH_3$ concentrations (**Figure 2E**). This phenomenon also implies that the most effective meteorological factor that could impact the atmospheric chemical concentrations from the chemistry-atmosphere feedbacks would be the change in atmospheric dispersion capability.

The inclusion of $NH_3$-SOA uptake also resulted in significant changes on modeled $PM_{2.5}$. **Figure 3A** shows the changes of $PM_{2.5}$ and its components (e.g., $NH_4^+$ and SOA) for the entire CONUS and four states with the highest $NH_3$ emissions. In general, $PM_{2.5}$ and SOA increased after the inclusion of $NH_3$-SOA uptake, while $NH_4^+$ decreased. Among them, North Carolina shows the most significant decrease in $NH_4^+$ concentration and the largest increase in SOA. However, western states like California and Idaho exhibit both $NH_4^+$ and SOA decreases after implementing the $NH_3$-SOA uptake. **Figure 4A** shows the mean $PM_{2.5}$ concentration for Base_14. The high concentration along the southeast boundary of the domain is

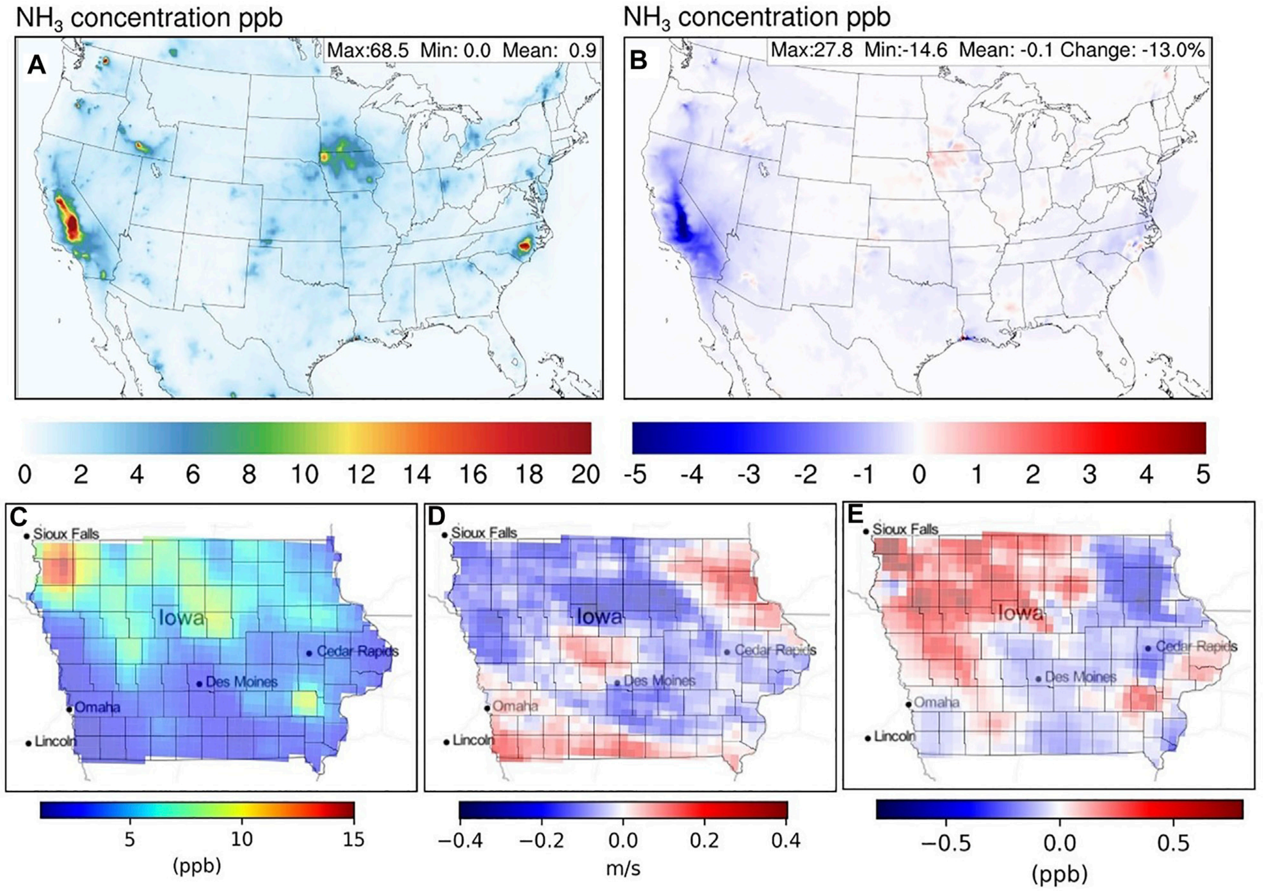

**FIGURE 2 |** Spatial distribution of time-averaged $NH_3$ concentration for Base_14 **(A)** CONUS and **(C)** State of Iowa. Spatial distribution of differences between UpTk_14 and Base_14 uptake case: **(B)** $NH_3$ concentration for CONUS, **(D)** 10-m wind speed for Iowa and **(E)** $NH_3$ concentration for Iowa.

caused by foreign $PM_{2.5}$ that originated from the western Sahara Desert in Africa.

At the same time, some hot spots with high $PM_{2.5}$ concentrations are caused by wildfires, like those in Washington, Oregon, and Texas. The result of the $PM_{2.5}$ distribution shows a clear pattern with the majority of the pollution happening over the eastern part of the CONUS, except for California. **Figure 4D** presents the change in $PM_{2.5}$ concentration after introducing the $NH_3$-SOA uptake. The overall impact on $PM_{2.5}$ is smaller than the impact on $NH_3$. The 4% increase of domain-wide $PM_{2.5}$ concentrations is still significant, with peak changes as large as 160 $\mu g/m^3$. Most of the increases occur over the eastern part of the CONUS, overlapped mainly with the region with high $PM_{2.5}$ concentration, except California. Such a distribution pattern is consistent with previous studies (Horne et al., 2018; Zhu et al., 2018). More detailed investigation shows that almost half of the $PM_{2.5}$ changes result from the increase of SOA concentrations (**Figure 4E**). The most impacted region states close to the lower Mississippi River (e.g., Louisiana, Alabama, Mississippi, and Georgia), which also have elevated SOA background concentrations (**Figure 4B**). The drive behind the increase of SOA concentration is a change in particle acidity as more $NH_3$ is consumed by the $NH_3$-SOA uptake process (**Figure 4C**). The acid-catalyzed SOA formation pathway of isoprene epoxydiols is enhanced by increasing particle acidity (Pye et al., 2013), which is also observed in our previous works (Zhu et al., 2018; Wu et al., 2021).

Interestingly, the situation is different in Florida, where a decrease in $PM_{2.5}$ and SOA concentration is predicted. Like the anomaly of increasing $NH_3$ in Iowa, this particular case also results from the meteorological feedback effects. **Figure 4F** shows the changes of 10-m wind speed after adding the $NH_3$-SOA uptake process, where a substantial increase in atmospheric dispersion is observed over Florida. Such an increase in wind speed clears out $PM_{2.5}$ and decreases its overall concentration. The exact magnitude correlations between the change in wind and air pollutant concentrations are likely to be affected by multiple factors (e.g., boundary lay high, topographic, etc.) and would be an interesting subject for future investigations.

## 3.3 Impact due to Climate Change

Changes in background meteorological conditions under RCP 8.5 climate scenarios largely altered the atmospheric chemistry conditions in the model. From the SI, **Supplementary Figure S5**

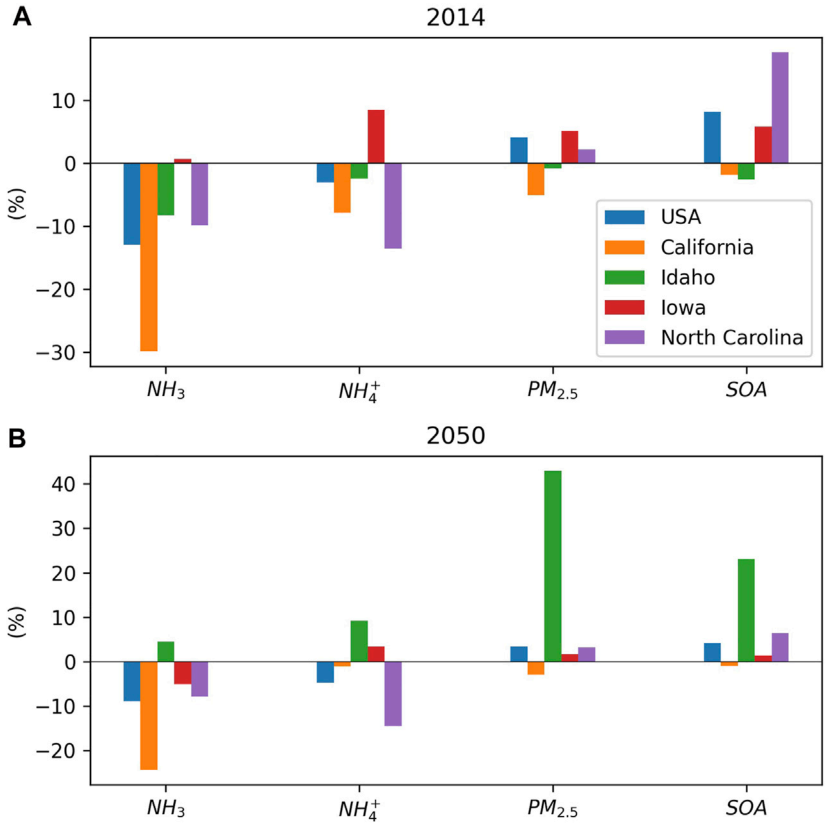

**FIGURE 3 |** Normalized averaged concentration changes due to the $NH_3$- SOA uptake for the United States and four states with top anthropogenic $NH_3$ emissions for the **(A)** 2014 case and **(B)** 2050 case.

shows the meteorological and air quality changes between Base_50 and Base_14. First, there is a general increase in the surface temperature and a decrease in 10-m wind speed over the continent. The particle acidity also experiences dramatic changes. Furthermore, there is some substantial increase in pH values over the southeastern of the US, largely overlapping with the high SOA region in **Figure 4B**. For air pollutants, the overall $NH_3$ concentration is decreased by 2.1%, with significant decreases in Washington, Oregon, and Texas. Decreases in those states have a direct connection with wildfire emissions. The plume rise or vertical distribution of wildfire emissions are calculated inline with the CMAQ model based on a Briggs plume rise formulation (Briggs 1982), which means the change in meteorological conditions (e.g., temperature, wind speed, and mixing height) could broadly impact the distribution of related emissions. In addition, the change in dispersion directions alters the distribution of $NH_3$ concentrations in California and North Carolina. Also, some significant increase in $PM_{2.5}$ concentration are observed in the southeast corner of the simulation domain, most likely due to the spread of the west Sahara original sandstorm from the boundary conditions under the new meteorology. Interestingly, SOA concentration decreases significantly over the southeastern CONUS, leading to a 15% decrease domain wide.

**Figure 3B** shows the impacts on air pollutants after including the $NH_3$-SOA uptake in the 2050 climate scenario. Compared to the impacts for the 2014 case (**Figure 3A**), the domain-wide $NH_3$ reduction is 30% smaller under the future climate scenario. One reason is that the lower background $NH_3$ concentration produced less $NH_3$ available for uptake. Another more important reason is the meteorological feedback due to the $NH_3$-SOA uptake changed the wildfire emission, and the gains in wildfire emissions counterbalanced the reduction caused by the $NH_3$-SOA uptake (**Figure 5A**). The significant increase in pollutants concentrations in Idaho is also caused by wildfire emissions (**Figures 5A–D**). For Iowa, the $NH_3$ anomaly is largely reduced as the overall wind speed increased by 1.7% (**Figure 5F**). Due to the decrease in background SOA concentration, the impact of SOA (**Figure 5C**) and particle acidity (**Figure 5E**) over the southeastern CONUS due to $NH_3$-SOA uptake is largely reduced compares to the 2014 case (**Figure 4**). Even with the additional gain from altered wildfire emissions, the increase of SOA is only about half of the 2014 case, and the decrease in pH is only 47%. Even for California, the impact caused by the $NH_3$-SOA uptake is also largely reduced. Only North Carolina shows similar impacts to the 2014 cases. Although, climate change could also impact the upstream emissions that have not been considered in this study.

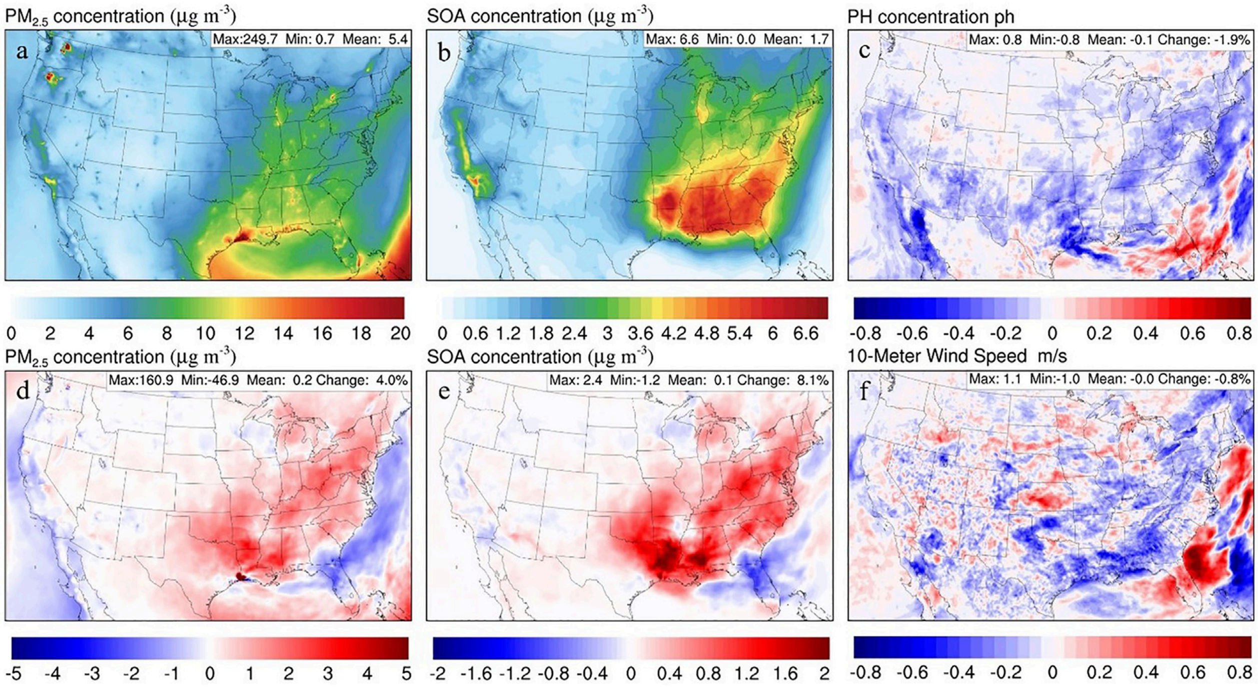

**FIGURE 4 |** Spatial distribution of time-averaged concentration for Base_14 **(A)** $PM_{2.5}$ and **(B)** SOA. Spatial distribution of differences between UpTk_14 and Base_14 uptake case: **(D)** $PM_{2.5}$, **(E)** SOA, **(C)** pH, and **(F)** 10-metter wind speed.

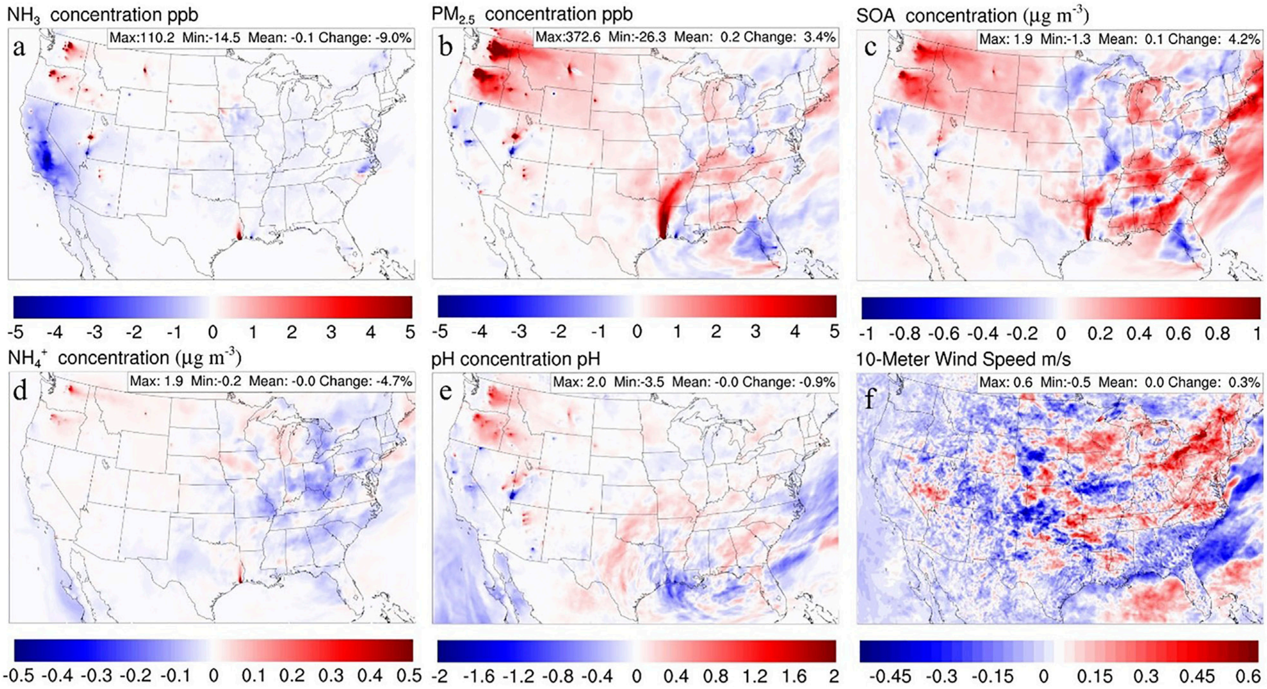

**FIGURE 5 |** Spatial distribution of differences between UpTk_50 and Base_50 uptake case: **(A)** $NH_3$, **(B)** $PM_{2.5}$, **(C)** SOA, **(D)** $NH_4^+$, **(E)** pH, and **(F)** 10-metter wind speed.

These preliminary results imply that under a warmer climate, the impact of the $NH_3$-SOA uptake might be reduced compared to current climate conditions. Also, the impact on smoke plume rise characters could be a significant factor during the meteorology and air quality two-way feedback, as observed in this study.

## 4 CONCLUSION

In this study, the potential meteorology and air quality impacts of the chemical uptake of $NH_3$ by SOA, resulting in nitrogen-containing organics, are investigated with the WRF-CMAQ two-way coupled model. Simulations over the continental US are performed for July 2014 and July 2050 under the RCP 8.5 IPCC scenario to study the potential joint impact due to climate change. First, results show that the $NH_3$-SOA uptake process could reduce the model underestimation of $PM_{2.5}$ (23% bias reduction), especially the underestimation of SOA over the southeastern CONUS (61% error reduction). Such an improvement in model performance indicates the importance of including $NH_3$-SOA chemistry in the air quality model and its potential benefits. Secondly, a significant reduction in the concentration of $NH_3$ was observed with the $NH_3$-SOA uptake turned on, which also increased the model predicted particle acidity. In general, the inclusion of the $NH_3$-SOA uptake would lead to lower ammonium concentrations but higher SOA concentrations, with some exceptions. Western U.S. states usually have decreased $PM_{2.5}$ concentrations with the uptake included, whereas Eastern U.S. states usually exhibit increased $PM_{2.5}$ concentrations. Most of the impact on pollution concentrations caused by the air quality and meteorology two-way feedback results from changes in wind speed and resulting dispersion conditions. As illustrated by the 2050 scenario, the impact on wildfire plume calculation due to the atmospheric chemistry and meteorology interaction through the two-way feedback could also significantly impact the resulting pollution distribution. Finally, as a preliminary study, we observe that under a warmer climate, including the $NH_3$-SOA uptake produces a lower impact on air quality predictions than it does in the current climate.

Results show that the inclusion of the two-way feedback process has significant impacts on model behaviors when the new mechanism is implemented. It is noted that results from this study only consider the shortwave feedback. There are other feedback processes between the air pollutant and meteorology, including longwave feedbacks, the oceanic response and other indirect effects (Seinfeld and Pandis 2016) that are not included in this study and could be explored in future research. Indirect effects like changes in cloud formation and cloud albedo due to changes in aerosol concentration could enhance the meteorological feedback and further affect aerosol predations (Menon et al., 2002). It is likely the indirect effects could enhance the feedback impact over the southeast of the U.S. The oceanic response due to changes in aerosol estimation could also affect large-scale circulations (Cai et al., 2006), which should be considered for long-term simulations but most likely to be insignificant in our short-term study. Moreover, it is also noted that the $NH_3$ uptake coefficient used in this study is on the higher end of current laboratory measurement ($\gamma = 10^{-3}$). Namely, the actual impact due to the $NH_3$ uptake process could be even smaller. Our previous study shows the impact on $NH_3$concentration is likely to be 7–12 times lower at the lower end of the uptake coefficient ($\gamma = 10^{-5}$), and the impact on $PM_{2.5}$ could be about 48–88 times lower (Zhu et al., 2018; Wu et al., 2021). Developing a more accurate and comprehensive SOA-$NH_3$ parameterization with improved observation data should be of significant priority for future work. Finally, results of this study highlight the potential impacts that can be caused due to climate change in model development.

## DATA AVAILABILITY STATEMENT

The raw data supporting the conclusion of this article will be made available by the authors, without undue reservation.

## AUTHOR CONTRIBUTIONS

SZ designed and conducted WRF-CMAQ simulations and SZ wrote the manuscript. SZ, KW, SN and DD reviewed and modified the manuscript. KW contributes analysis on meteorological results. SN provides the chemical mechanism of ammonia uptake by SOA. DD administered the project. All authors contributed to manuscript revision and read and approved the submitted version.

## ACKNOWLEDGMENTS

This publication was developed under Assistance Agreement No. EPA 83588101 was awarded by the US Environmental Protection Agency to the Regents of the University of California. It has not been formally reviewed by the EPA. The views expressed in this document are solely those of the authors and do not necessarily reflect those of the agency. The EPA does not endorse any products or commercial services mentioned in this publication.

## SUPPLEMENTARY MATERIAL

The Supplementary Material for this article can be found online at: https://www.frontiersin.org/articles/10.3389/fenvs.2022.867908/full#supplementary-material

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**Conflict of Interest:** The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.