Thanks to co-Editor for your review. Here is our point-to-point reply.

Concerning responses to R1:

R1 comment #1: “They attribute these model differences to differences in 1) pH-dependent wet deposition of NH4+, 2) nitrate formation on the surface of sea salt and dust aerosol, and 3) the nitrate coarse mode fraction. They find that nitrate production on sea salt and dust is important to include in models as it tends to dominate nitrate production and controls its partitioning between the fine and coarse mode. In that sense, it seems to me that 2 and 3 above are referring to the same process.”

Author’s response: “We intend to separate discussion of section 5.2 and 5.3 because the nitrate formation on the surface of sea salt and dust aerosol (section 5.3) is important, but not the only factor, to determining nitrate size distribution (section 5.2). Also, the former focuses more on chemical process and the later on physical process and climate implication.” I find that the authors did not address the reviewer’s concern. The formation of nitrate on coarse mode dust and sea salt particles is the major factor controlling the size distribution and the ratio of coarse/total particulate nitrate in models. This is not clearly explained in the manuscript, and should be clarified in section 5.

Authors’ response: we added on lines 691-695 “Coarse mode aerosol nitrate is formed due to presence of dust and/or sea salt. In other words, the formation of nitrate on coarse mode dust and sea salt particles is the major factor controlling the size distribution. Other factors, such as NH3/NH4+/NO3- chemistry and atmospheric transport and removal processes, also affect nitrate size distribution.”

R1 comment #2: Authors did not address the reviewer’s concern “What the authors are referring to by the use of “heterogeneous chemistry” is what I would call thermodynamic partitioning between the gas and aerosol phase.” I agree with the reviewer that the use of the heterogeneous chemistry should not be applied to refer to the gas-particle thermodynamic partitioning. It is unclear in the revised manuscript whether the models are treating the formation of coarse mode nitrate by a heterogeneous uptake of HNO3 onto dust and sea-salt particles or by an equilibrium approach. This must be clarified.

Authors’ response: On lines 225-227, we state “Please note that the heterogeneous chemical production of particulate nitrate mentioned in this paper refers only to the first order loss reaction of HNO3 on the surface of dust and sea salt particles.” On lines 279-285, we indicate that there are two ways to account for the contribution of dust and sea salt to nitrate formation. Some models (EMAC, Oslo-CTM3, and Oslo-CTM2) include dust and/or sea salt components in their TEQM models directly (marked as TEQM in table 1 under column “How do CHEMDUSS”), while some models (EMEP, GISS-OMA, GMI, and INCA) use an approach of first order loss rate outside their TEQMs to account for the heterogeneous reactions of HNO3 on the surface of dust and sea salt (marked as HETCHEM in table 1). We have gone through the paper to clarify “heterogeneous chemistry”, see lines 34-35, 222, 226-227, and 826.

Concerning responses to R2:

In the added text please change the 1st sentence to: “Our work presents an initial effort to assess
nitrate simulation from chemical and physical processes (deposition).” You should be more specific by what chemical and physical processes you have looked at. And please have the rest of the paragraph corrected for English.

Author’s response: Changed the sentence to “Our work presents a first effort to assess nitrate simulation from chemical (e.g. chemistry among NH₃, NH₄⁺, NO₃⁻, SO₄²⁻, dust and sea salt) and physical processes (e.g. emission, dry deposition, and wet deposition).” The whole paragraph has been revised for English.

Why would you want to compromise the accuracy and efficiency, this sentence is misleading (line 619: “Several approximations, therefore, have been developed to compromise accuracy and efficiency.”) Is this what you meant: “Several approximations have been developed to allow computational efficiency although they might compromise the model accuracy.” Please reword.

Author’s response: Done as suggested.

Additional comments:

1) The confusion about the use of the “nitrate” term:

It should be clearly stated in the title that the paper is evaluating the particulate nitrate: “Investigation of global particulate nitrate from the AeroCom Phase III experiment.”

Instead of using nitrate aerosol it would be preferable to use particulate nitrate when referring to the particle phase as aerosol term refers to both gas and particulate fraction that are in equilibrium.

Author’s response: Done as suggested.

Line 122: Is this particulate or gas-phase nitrate: “If fixed Nr is deposited as nitrate in forests...”

Authors’ response: Nr (Reactive nitrogen) is a term used for a variety of nitrogen compounds that support growth directly or indirectly. Nr includes the gases nitrogen oxides (NOₓ), ammonia (NH₃), nitrous oxide (N₂O), as well as gas and particulate nitrate (NO₃⁻). I added an explanation “including gas and particulate NO₃⁻ and other nitrogen compounds” in lines 124-125.

2) As mentioned by R1, N₂O₅ hydrolysis is an important heterogeneous reaction when investigating the nitrate budgets that is typically included in global models. It should be clearly stated in Table 1 or 2 and in the paper how this reaction is treated and if it is included. And some discussion on the uncertainty due to this reaction and references should be added in the manuscript.

Authors’ response: We added a column “N₂O₅ hydrolysis” in table 1. We also added these sentences in lines 209-215.

“Meanwhile, all models consider N₂O₅ hydrolysis, the conversion of N₂O₅ to HNO₃. The first order loss reaction occurs on the surface of tropospheric aerosols and assumes irreversible instant reaction. However, the models differ in N₂O₅ hydrolysis by considering the reaction on the surface of different aerosol types. Uptake coefficients (aka gamma factors) also differ in their relationship to temperature and RH. CHASER model is special as it allows N₂O₅ conversion to HNO₃ on liquid cloud particles. Please refer to Table 1 and the listed references for details.”

3) Add “relative” Line 78: More importantly, the relative importance of aerosol nitrate

Authors’ response: Done.
4) Add “particulate” Line 102: First, the formation of particulate nitrate, 
Authors’ response: Done.

5) Given that coarse mode nitrate measurements are sparse, please include measurements that have been done in Paris during the ESQUIF campaign that found that the coarse nitrate fraction represents up to 60% of total particulate nitrate mass during the night and 80% during the day. See either Figure 13c of “Hodzic et al., ACP 2006 Aerosol chemical and optical properties over the Paris area within ESQUIF project”, or Figure 6 of Hodzic et al, AE 2006: A model evaluation of coarse-mode nitrate heterogeneous formation on dust particles.” 
Authors’ response: A sentence has been added on lines 746-748: “Measurements taken in Paris during the ESQUIF campaign found that the coarse nitrate fraction represents up to 60% of the total particulate nitrate mass at night and 80% at day (Hodzic et al., 2006a, 2006b).”

6) Clarify what you mean by feedback in this sentence: Line 202: “All models use full gas phase O3-NOx-HOx chemistry to produce HNO3 and consider the feedback of nitrate aerosol formation on HNO3 calculation.” Do you mean radiative feedbacks on photolysis or changes in the HNO3 concentrations due to the gas/particle equilibrium?
Authors’ response: We added an explanation on lines 208-209: “changes in HNO3 concentrations due to the gas/particle equilibrium).”
Investigation of global particulate nitrate from the AeroCom Phase III experiment

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Abstract

An assessment of global particulate nitrate and ammonium aerosol based on simulations from nine models participating in the AeroCom Phase III study is presented. A budget analyses was conducted to understand the typical magnitude, distribution, and diversity of the aerosols and their precursors among the models. To gain confidence on model performance, the model results were evaluated with various observations globally, including ground station measurements over North America, Europe, and East Asia for tracer concentrations and dry and wet depositions, as well as with aircraft measurements in the Northern Hemisphere mid-high latitudes for tracer vertical distributions. Given the unique chemical and physical features of the nitrate occurrence, we further investigated the similarity and differentiation among the models by examining: 1) the pH-dependent NH3 wet deposition; 2) the nitrate formation via heterogeneous chemistry on the surface of dust and sea-salt particles or thermodynamic equilibrium calculation including dust and sea salt ions; and 3) the nitrate coarse mode fraction (i.e., coarse/total). It is found that HNO3, which is simulated explicitly based on full O3-HOx-NOx-aerosol chemistry by all models, differs by up to a factor of 9 among the models in its global tropospheric burden. This partially contributes to a large difference in NO3−, whose atmospheric burden differs by up to a factor of 13. The atmospheric burdens of NH3 and NH4+ differ by 17 and 4, respectively. Analyses at the process level show that the large diversity in atmospheric burdens of NO3−, NH3, and NH4+ is also related to deposition processes. Wet deposition seems to be the dominant process in determining the diversity in NH3 and NH4+ lifetimes. It is critical to correctly account for contributions of heterogeneous chemical production of nitrate on dust and sea-salt, because this process overwhelmingly controls atmospheric nitrate production (typically >80%) and determines the coarse and fine mode distribution of nitrate aerosol.

1. Introduction
Atmospheric aerosols adversely affect human health and play an important role in changing the Earth’s climate. A series of multimodel studies have been coordinated by the international activity of Aerosol Comparisons between Observations and Models (AeroCom) in its Phase I and II model experiments that have systematically assessed the presence and influence of almost all major atmospheric anthropogenic and natural aerosols (such as sulfate, dust, and carbonaceous aerosols) (e.g., Kinne et al., 2006; Schulz et al., 2006; Textor et al., 2006; Koch et al., 2009; Huneu et al., 2011; Tsirigidis et al., 2014; Kim et al., 2015). Very little attention has been drawn to nitrate aerosol (hereafter “nitrate” referring to particulate nitrate unless otherwise specified) other than its contribution to radiative forcing (Myhre et al., 2013). One obvious reason is that not many models used to include nitrate owing to the chemical complexity of nitrate formation. However, atmospheric nitrate aerosol not only exerts direct effects on air quality and climate, but also uniquely impacts the Earth system by being directly involved in tropospheric chemistry and constraining net primary productivity, hence altering carbon sequestration and ecological effects, via its deposition (Prentice et al., 2001).

Atmospheric nitrate contributes notably to total aerosol mass in the present-day, especially in urban areas and agriculture regions. Nitrate is about a quarter of sulfate in terms of overall global burden, AOD, and direct forcing at the present-day according to the study of AeroCom II direct forcing experiment (Myhre et al., 2013). This conclusion is confirmed by recent publications using various individual models and emission inventories (Bellouin et al.; 2011; Bauer et al., 2007; Hauglustaine 2014; Karydis et al., 2016; Mezuman et al., 2016; Paulot et al., 2016). Regionally, considerable evidences from in-situ measurements (Bessagnet et al., 2014; Haywood et al., 2008; Jimenez et al., 2009; Malm et al., 1994; Vieno et al., 2016) and model results (Karydis et al., 2011; Ensberg et al., 2013; Trump et al., 2015) indicate that nitrate becomes one of the major aerosol species in urban and agriculture environments. For example, nitrate concentration is about half of sulfate during the summer season in Beijing (Zhou et al., 2016) and represents a large portion of wintertime aerosol mass in the San Joaquin Valley in California (Pusede et al., 2016).

More importantly, the relative importance of aerosol nitrate is likely to increase over the century with a projected decline in SO2 and NOx emissions and increase in NH3 emissions (IPCC, 2013). With the reduction of SO2 emissions, less atmospheric NH3 is required to neutralize the strong acid H2SO4. The excess of NH3 results in gaseous HNO3 and NH3 entering the condensed phase, and their subsequent dissociation yields nitrate and ammonium ions. The trend of future nitrate depends on which is the limited species, NOx or NH3, for nitrate formation (Tsimpidi et al., 2007; 2008). Generally, our atmosphere, at its current and foreseeable near future, is still in an NH3-limited condition according to sensitivity studies by Heald et al. (2012) and Walker et al. (2012). Almost all global models predicted an overall increase of atmospheric nitrate burden during this century based on current available emission inventories (Bauer et al 2007; 2016; Bellouin et al., 2011; Hauglustaine et al., 2014; Li et al., 2014). For example, using CMIP5 future emission projections, Bellouin et al. (2011) concluded that, by 2090, nitrate would become an important aerosol species in Europe and Asia, contributing up to two thirds of
the globally averaged anthropogenic optical depth. However, the predicted trend of surface nitrate is mixed. Some studies estimated a consistent increase of surface nitrate (Bellouin et al., 2011), while others pointed out that this increase might vanish or even reverse over some regional urban areas due to the decline of NOx emissions (Bauer et al., 2016; Pusede et al., 2016; Trail et al., 2014). Nevertheless, the potentially increasing importance of nitrate in climate and its large uncertainty in future surface nitrate predictions urge us to characterize model performance and understand the physicochemical mechanisms behind the diversity of nitrate simulations.

Nitrate is also important in that its formation directly affects tropospheric chemistry. First, the formation of particulate nitrate, through either aqueous phase chemical reaction between HNO3 and NH3 (Metzger et al., 2002; Kim et al., 1993) or heterogeneous reaction of nitrogen species such as HNO3, NO3, and N2O5 on the surface of dust and sea salt aerosol particles (Bauer et al., 2004; 2005; Bian et al., 2003; Dentener 1996; Liao et al., 2003), converts gas phase nitrogen species into aerosols. Consequently, the global tropospheric NOx concentration and the rate of conversion of N2O5 to HNO3 will be reduced (Riemer et al., 2003), which in turn leads to the reduction of atmospheric oxidants. For example, global tropospheric O3 can be reduced by 5% (Bauer et al., 2007) and tropical Atlantic OH by 10% (Bian et al., 2003) just through the heterogeneous reactions of nitrogen radicals on dust. Second, the most important removal path for nitrogen from the atmosphere is the formation of HNO3, which is subsequently deposited (Riemer et al., 2003). Since HNO3 is subject to partitioning between the gas and aerosol phases, the lifetimes of nitrogen species can be shortened by the formation of tropospheric nitrate aerosol because the loss of total HNO3 will be accelerated by a much higher dry deposition in the aerosol phase.

Large nitrogen deposition occurs over both land and ocean (Dentener et al., 2006; Kanakidou et al., 2012; 2016). Nitrogen deposition can either benefit or impair ecosystem productivity depending on the initial balance of nutrients since different ecosystems have different Nr (reactive nitrogen including gas and particulate NO3− and other nitrogen compounds) availability and retention (Galloway et al., 2004; Prentice et al., 2001). If fixed Nr is deposited as nitrate in forests, it may act as a "fertilizer," stimulating growth and thus enhancing carbon sequestration (Fowler et al., 2015). But when the accumulated deposition exceeds the nutritional needs of the ecosystem, nitrogen saturation may result (Fenn et al., 1996). Soil fertility declines due to the leaching of cations (Milegroet and Cole, 1984) and, thus, carbon uptake diminishes. The balance between fertilization and saturation depends on the spatial and temporal extent of nitrogen deposition. In order to determine the extent to which the emissions of air pollutants will have to be reduced and whether the environment needs to be protected from damage, it is essential to know where and by how much N deposition exceeds nature's tolerance (Dentener et al. 2006; Lamarque et al., 2005; Phoenix et al., 2006).

Here we present a nitrate-focused study that has been organized as a part of the series of AeroCom phase III experiments (https://wiki.met.no/aerocom/phase3-experiments). The goals of this activity are to (1) address the diversity of the nitrate simulation by the AeroCom multi-models and diagnose the driving processes for the diversity, (2) explore...
the uncertainty of the model nitrate simulations constrained against various
measurements from ground station networks and aircraft campaigns, and (3) investigate
how the formation of nitrate changes in different models in response to perturbation on
key precursors and factors that determine nitrate formation. We focus on the first two
objectives in this paper. Such a study directs us on how to improve the representation of
nitrate aerosol formation and size distribution in climate chemistry models and reveals
nitrate effects on global air quality and climate.

Building upon the analysis of the multi-model diversity, three additional sensitivity
experiments are designed using the GMI model to further explore the potential sources
for the diversity on physical and chemical process-level. First, we explore the impact of
pH-dependent NH₃ wet deposition on atmospheric NH₃ and associated nitrogen species.
We then reveal the importance of mineral dust and sea salt in the nitrate formation and
check the resultant nitrate aerosol size distribution that is particularly important in nitrate
forcing estimation.

The paper is organized as follows. Section 2 introduces the experiment setup including
the emission inventories used and the participating AeroCom models. Observations of
surface tracer concentrations and dry and wet depositions over U.S., Europe, and East
Asia, as well as aircraft measurements in the ARCTAS campaigns are described in
section 3. We present AeroCom model inter-comparison and the model evaluation using
aforementioned observations in section 4. Based on the knowledge from previous
sections, we further discuss nitrate formation in response to physiochemical
methodologies in section 5 and summarize our major findings in section 6.

2. Experiment setup and AeroCom model description

2.1 Experiment setup

The AeroCom III nitrate experiment comprises one baseline and six perturbation
simulations, with the latter designed for assessing the possible future changes of emission
and meteorological fields relevant to nitrate formation. Models are advised to use the
same prescribed emission datasets for gases and aerosols. Emissions from anthropogenic,
aircraft, and ship for aerosol and ozone simulations are obtained from the recently
developed HTAP v2 database (Janssens-Maenhout et al., 2015) that provides high spatial
resolution monthly emission. For the tracers that are included in ozone chemistry but are
not provided by HTAP v2 (i.e. some volatile organic compounds), they should be
obtained from CMIP5 RCP85 with a linear interpolation between 2005 and 2010.
Biomass burning emissions are the emissions of GFED3 (Werf et al., 2010) in 2008
[http://www.globalfiredata.org/data.html]. The NH₃ emission from ocean is adopted
based on the compilation of GEIA emission inventory [Bouwman et al., 1997].
Participating modeling groups use their own emissions of dimethyl sulfide (DMS), dust,
sea salt, and NO from lightning, since they are calculated based on models’
meteorological fields.

A full year simulation for 2008 is required for the nitrate model experiment. There are
several in-situ observation datasets available in 2008 for model evaluation, including the
surface concentration and deposition measurements over the US (CastNet, AMoN, NDAP/NTN), Europe (EMEP), and Asia (EANET), and the aircraft measurements of vertical profiles (e.g. ARCTAS-A, ARCTAS-CARB, and ARCTAS-B). All participating models are required to use the reanalysis or nudged meteorological data for 2008 and allow one-year spin up for the baseline simulation.

2.2 AeroCom models

Nine models participate in the AeroCom III nitrate experiment. Their general nitrate-related physiochemical mechanisms are summarized in Table 1. Further detailed information on their thermodynamic equilibrium model (TEQM) is given in Table 2.

The models participating in this study are divided into two groups. Group one (CHASER, EMAC, INCA, GISS-MATRIX, and GISS-OMA) run chemical fields together with meteorological fields, while group two (EMEP, GMI, OsloCTM2, and OsloCTM3) simulate chemical fields using archived meteorological fields. Most models in this study have a horizontal resolution around 2-3 degrees except EMEP with 0.5 degree. Vertically, most models cover both the troposphere and the stratosphere with a peak altitude up to 0.01 hPa except EMEP that extends vertically up to 100 hPa into the troposphere only.

All models use full gas phase O$_3$-NO$_x$-HO$_x$ chemistry to produce HNO$_3$ and consider the feedback of nitrate aerosol formation on HNO$_3$ calculation (i.e. changes in HNO$_3$ concentrations due to the gas/particle equilibrium). Meanwhile, all models consider N$_2$O$_5$ hydrolysis, the conversion of N$_2$O$_5$ to HNO$_3$. The first order loss reaction occurs on the surface of tropospheric aerosols and assumes irreversible instant reaction. However, the models differ in N$_2$O$_5$ hydrolysis by considering the reaction on the surface of different aerosol types. Uptake coefficients (aka gamma factors) also differ in their relationship to temperature and RH. CHASER model is special as it allows N$_2$O$_5$ conversion to HNO$_3$ on liquid cloud particles. Please refer to Table 1 and the listed references for details.

However, due to the complexity of chemical mechanisms for organic nitrate compounds and different recommendations for reaction rates, HNO$_3$ fields produced by the models differ greatly. This difference propagates into the subsequent gas-aerosol reactions for nitrate formation.

These models are very different in their approaches on gas-aerosol reactions in nitrate formation. All models consider reactions between NH$_3$ and HNO$_3$. However, models differ dramatically in whether to include contributions of heterogeneous reactions on dust and sea salt (Table 1). Some account for both, some for only dust or sea salt, and some do not account for any of them at all. The methods used by the models in accounting for NH$_3$ and dust/sea salt contributions are also different. Please also note that the heterogeneous chemical production of particulate nitrate mentioned in this paper refers only to the first order loss reaction of HNO$_3$ on the surface of dust and sea salt particles. A series of reactions, such as N$_2$O$_5$ hydrolysis and BrONO$_2$ hydrolysis, affect HNO$_3$ simulation. These reactions are typically considered in O$_3$-NO$_x$-HO$_x$ chemistry and their discussion is beyond the scope of this paper.
All participating models adopt TEQM to deal with aqueous and solid phase reactions and gas-aerosol partitioning (Tables 1 and 2). This is based on the assumption that volatile species in the gas and aerosol phases are generally in chemical equilibrium. However, the assumption is not always warranted in some cases, as we will discuss in section 5.2. Even with the TEQM approach, nitrate calculation could differ due to treatments of equilibrium constants or chemical potentials, solute activity coefficients, water activity, and relative humidity of deliquescence (RHD). The parameterizations adopted by the models to deal with multicomponent activity coefficient, binary activity coefficient, and water activity are given in table 2. GISS-OMA, Oslo-CTM2 and Oslo-CTM3 are special in that they assume aerosols to be metastable so that the model does not take into account formation of solids in this study. All other models do consider the effect of the hysteresis of particle phase transitions. All models also assume that the overall particles are large enough to neglect the Kelvin effect.

The participating models call the TEQMs in different ways to account for aerosol size effect. All the TEQMs (ISORROPIA-I, ISORROPIA-II, MARS, RPMIRES, INCA, and EQSAM3) assume particles to be internally mixed, i.e. all particles of the same size have the same composition. However, some parent models (CHASER, EMEP, GMI, INCA, GISS-MATRIX and GISS-OMA) call their TEQMs only once for fine mode aerosol particles, while the others (EMAC, OsloCTM2 and OsloCTM3) call their TEQMs from different aerosol size bins. For example, Oslo-CTM2 and Oslo-CTM3 consider a bimodal aerosol size-spectrum with two major aerosol modes, fine and coarse, and calculate gas-aerosol equilibrium partitioning with EQSAM3 first for fine mode and then for coarse mode. Additionally, to account for kinetic limitations, EMAC calculates the phase partitioning in two stages. In the first stage, the amount of the gas-phase species that is able to kinetically condense onto the aerosol phase within the model time step is calculated, while in the second stage, the TEQM redistributes the mass between the two phases assuming instant equilibrium (Pringle et al., 2010).

The TEQMs also differ in the chemical components considered. Specifically, the TEQMs in CHASE, EMEP, GISS-MATRIX, GISS-OMA, GMI and INCA include only species of sulfate, nitrate, ammonium and their gas, liquid, and solid components. The models Oslo-CTM2 and Oslo-CTM3 add NaCl and HCl, while the model EMAC further expands the species by including dust-related crustal material such as Ca\(^{2+}\), K\(^+\), and Mg\(^{2+}\).

These TEQMs differ in their computational approaches as well. Computational efficiency is a prime consideration for a TEQM that is designed for incorporation into a global air quality and climate study. To speed up the calculation, TEQMs typically divide the system into sub-domains based on RH and concentrations of ammonium, sodium, crustal cations, and sulfate. Corresponding approximation could be adopted for each sub-domain with the minimum numbers of equilibriums and unknown components. As listed in table 2, the numbers of sub-domains are 4, 5, 4, 2, 3, and 3 for the TEQM ISORROPIA-I, ISORROPIA-II, MARS, RPMIRES, INCA, and EQSAM3, respectively.

The ways to account for the contribution of dust and sea salt to nitrate formation are also different (see Table 1 column “How do CHEMDUSS”). Some models (EMAC, Oslo-
CTM3, and Oslo-CTM2) include dust and/or sea salt components in their TEQM models directly (marked as TEQM in Table 1 under column “How do CHEMDUSS”), while some models (EMEP, GISS-OMA, GMI, and INCA) use an approach of first order loss rate outside their TEQMs to account for the heterogeneous reactions of HNO3 on the surface of dust and sea salt (marked as HETCHEM in Table 1). For the latter approach, the gamma rates and their RH dependence adopted by the models differ as well.

Dry and wet deposition of NH3, ammonium nitrate, and ammonium sulfate are treated similarly to other gas and aerosol tracers in the models. It is worth pointing out that there is a different consideration for Henry’s law constant of NH3 used by the models. Some models modify it based on the pH value of cloud water while others do not. We will discuss the impact of these two treatments on nitrate simulation in section 5.1.

We introduce only the major characteristics of thermodynamic equilibrium models since this study aims for the evaluation and explanation of overall nitrate diversity among the GCM/CTM models from all potential aspects. The detailed discussion of the models chemical mechanism of gas phase reactions and the aerosol optical properties adopted by the models is also beyond this work. Readers could refer to the references listed in Tables 1 and 2 for any further details.

3. Observations

We use surface measurements from ground station networks and aircraft campaigns to evaluate modeled surface concentrations, dry and wet depositions, and vertical distributions of nitrate and related species (Table 3).

3.1 Surface measurements of concentrations and deposition rates

Ambient concentrations of sulfur and nitrogen species throughout the US and Canada have been measured by the ground station network CASTNET (Clean Air Status and Trends Network) (Figure 1). The measurements use a 3-stage filter pack with a controlled flow rate. The measurements of CASTNET do not include NH3. AMoN (Ammonia Monitoring Network), measuring concentrations of ambient NH3, has been deployed at CASTNET sites starting from October 2007 using passive samplers. The corresponding tracers’ surface concentration measurements over Europe have been conducted by EMEP (The European Monitoring and Evaluation Programme). The measured sites of all these networks are located in rural areas or sensitive ecosystems, representing a larger region by avoiding influences and contamination from local sources. Surface concentrations over East Asia are inferred from the measurement of dry deposition by EANET (Acid Deposition Monitoring Network in East Asia). This network provides acid deposition from a regional monitoring network including 13 countries in East Asia using standardized monitoring methods and analytical techniques.

CASTNET also provides dry deposition of sulfate and nitrogen species. Direct measurements of dry deposition fluxes (D) are expensive so D is calculated as the measured pollutant concentration (C) multiplied by the modeled dry deposition velocity (V_d). V_d is either estimated by the Multi-Layer Model fed with measured hourly
meteorological data or derived from historical average $V_4$ for sites with discontinued meteorological parameters.

Direct measurements of wet deposition fluxes of sulfate, nitrate, and other ions have also been performed by NADP/NTN (the National Atmospheric Deposition Program / National Trends Network) across the contiguous US, Canada, Alaska, and the US Virgin Islands and EANET over East Asia. Sites are predominantly located away from urban areas and point sources of pollution. Each site has a precipitation chemistry collector and gauge. Both networks can measure wet deposition for a continuous period (weekly for NADP/NTN and daily for EANET), or every precipitation event if using an automated collector (wet-only sampling).

Data is quality assured for all measurements. Measurements over North America use automated screening techniques, semi-annual calibration results, site operator comments, and manual data review. Quality assurance of EMEP is carried out on both the national level and by the Chemical Co-ordinating Centre (CCC). The quality of EMEP measurements is not equal at the national level (Schaap et al., 2002; 2004). Sites in North, Western and Central Europe were generally well equipped and performing, while sites in the rest of Europe suffered from inadequate sampling and calibrating methods due to political and/or economical reasons. The quality of ammonia measurement is relatively low since some laboratories experienced contamination problems (Williams et al., 1992). Although EANET adopts standardized monitoring methods and analytical techniques, quality assurance is carried out on the national level.

3.2 Aircraft measurements of vertical profiles

Aircraft campaign measurements during the 2008 Arctic Research of the Composition of the Troposphere from Aircraft and Satellites (ARCTAS) are used to evaluate tracer vertical distribution simulated by the models (Bian et al., 2013; Jacob et al., 2010). Three phases of the campaign, ranging from Northern Hemisphere mid-latitude industrial region (ARCTAS-CARB, June 2008) to high latitude Arctic regions influenced by long-range pollution transport (ARCTAS-A, April 2008) and by local boreal biomass burning (ARCTAS-B, July 2008), provide well encompassing environment observations. All flights were conducted by the NASA DC-8 aircraft and the flight tracks of these three phases are presented in Figure 2. An onboard HR-ToF-AMS instrument (Cubison et al., 2011; DeCarlo et al., 2006) measured fine mode aerosol concentrations (PM1) along the flight track including NO$_3^-$, NH$_4^+$, and SO$_4^{2-}$ at STP conditions (1013mb and 273.15K) at a sampling time interval of $\sim$12 seconds. Accuracy estimate of 2-standard deviations, likely conservative, is 34% for inorganics, dominated by the uncertainty in particle collection efficiency due to particle bouncing (Huffman et al., 2005).

4. Model intercomparison and evaluation

4.1 AeroCom model inter-comparisons of global distributions and budgets

4.1.1 NH$_3$ and NH$_4^+$

Six models use HTAP2 anthropogenic emissions, two (GISS-MATRIX and GISS-OMA) use CMIP5 emissions, and one (INCA) uses ECLIPSE emissions. Table 4b shows that
eight models have the annual NH₃ emission values within 5% of the value from the 
AeroCom experiment recommended emission inventories, but INCA is 11% higher. The 
similar emission distributions ensure that the examined inter-model diversities are truly 
caused by the differences in physicochemical processes among the models. The 
normalized root-mean-square deviation (NRMSD) of NH₃ global burden among models 
is 1.17 and 0.33 with and without EMAC included. This drastic change in global burden 
by EMAC is caused by its special treatment of wet deposition. In fact, the 
removal of trace gases and aerosol particles by clouds and precipitation in EMAC is not 
calculated based on empirically determined, fixed scavenging coefficients, but rather by 
solving a system of coupled ordinary differential equations, explicitly describing the 
processes involved (Tost et al., 2006). This method resolves feedback mechanisms 
between the multi-phase chemistry and transport processes involved. The liquid phase 
reaction set used converts all the scavenged NH₃ (or HNO₃) into NH₄⁺ (or NO₃⁻) in the 
liquid phase so that at the end everything that is deposited is the total NH₄⁺ and NH₃. 

Atmospheric NH₄⁺ is produced entirely by NH₃ chemical transformation. The models 
simulate NH₂⁺ much closer in chemical production (difference less than a factor of 2) than 
in lifetime (difference up to a factor of 5.2), indicating removing rates are a key factor in 
controlling the global burden of NH₄⁺. For example, CHASER has a much longer lifetime 
of NH₂⁺ (i.e. 9.8 days versus 4.3 days in average), which indicates a slow deposition 
removal of NH₂⁺ from the atmosphere. Consequently, CHASER simulates a much higher 
atmospheric NH₂⁺ burden than other models.

4.1.2 HNO₃ and NO₃⁻
HNO₃, an important nitrate precursor, differs by up to a factor of 9 in its global 
tropospheric burden among the models (Table 4c). All models simulated HNO3 based on 
a full gas phase O₃-HOx-NOx chemistry and coupled it with aerosol chemistry. This 
HNO₃ diversity will naturally be propagated into the NO₃⁻ simulation. However, further 
discussion of the detailed consideration of full gas-aerosol chemistry for HNO3 diversity 
among the models is beyond the scope of this study.

The resultant aerosol product (i.e., NO₃⁻) does not entirely follow its precursor (i.e., 
HNO₃) in terms of global burden: EMEP has very low HNO₃ but high NO₃⁻, two GISS 
models (MATRIX and OMA) simulate high HNO₃ but low NO₃⁻, while OsloCTM3 has 
an average HNO₃ but more than triple high NO₃⁻ than average (Tables 4a and 4c). 
Furthermore, the difference in NO₃⁻ global burden (up to a factor of 13) is larger than that 
of HNO₃. Differences in chemical mechanisms of NO₃⁻ production could be a potential 
explanation along with the difference in HNO₃ precursor. Unfortunately, only GMI and 
INCA provide a detailed NO₃⁻ chemistry budget analysis. Nevertheless, we can infer that 
the total chemical production of NO₃⁻ must be very low (~ 10Tg) in the two GISS models 
while very high (> 100 Tg) in OsloCTM2 and OsloCTM3 based on the reported total 
NO₃⁻ loss. Combining this information with the HNO₃ global tropospheric burden (Table 
4c), we can further infer that the chemical conversion from HNO₃ to NO₃⁻ must be lowest 
in the two GISS models while highest in the two Oslo models. Several factors could 
influence this conversion, such as the availability of alkaline species of mineral dust and 
sea-salt particles and the physicochemical mechanism of nitrate formation on dust and
sea-salt, availability of NH₃ after combining with SO₄²⁻, and the atmospheric meteorological fields of temperature and relative humidity. More discussions are given in sections 5.2 and 5.3.

Atmospheric lifetime of NO₃⁻ differs up to a factor of 4, from about 2 days in GMI and OsloCTM2 to larger than 7 days in GISS-OMA and GISS-MATRIX. The slower removal processes in the two GISS models compensate the low chemical production and help to maintain their NO₃⁻ atmospheric burden (Figure 3 and Table 4a).

4.2 Model-observation comparisons

4.2.1 Comparisons of surface concentrations over North America, Europe, and East Asia

Understanding diversity among model simulations and potential physiochemical processes behind the difference is important but not sufficient. The information has to be combined with the knowledge of model performance obtained directly from comparisons, particularly down to processes level, against various measurements to gain a direction of any improvement. Figures 4a-c show a model-observation comparison for surface mass/volume mixing ratios of NO₃⁻, NH₄⁺, NH₃, HNO₃, and SO₄²⁻ over North America (CastNET), Europe (EMEP), and East Asia (EANET). Each point represents a monthly mean concentration at one observational site. Generally, the agreement between model and observation is better for aerosol components than for gas tracers (i.e. the precursor species NH₃ and HNO₃) over all three regions. All models underestimate NH₃ surface volume mixing ratio with a ratio of model to observation down to 0.14, while most models overestimate surface HNO₃ volume mixing ratio with a ratio up to 3.9 over North America. The worse performances of NH₃ against observations may be also associated to their relatively lower measurement accuracy, i.e. easier to be contaminated during measurement (Williams et al., 1992). Among aerosol simulations, model performance is very similar for NH₄⁺ and SO₄²⁻, while slightly worse for NO₃⁻ that is dispersed further away from the 1:1 line, particularly at low NO₃⁻ values. The NO₃⁻ simulation over East Asia is worst with the average normalized root mean square to be 1.3 and 1.8 higher than that over North America and Europe, respectively.

4.2.2 Comparisons of vertical profiles with aircraft measurements during the ARCTAS field campaign

Evaluation of model performance presented in 4.2.1 for the surface concentrations in the source regions is highly dependent on the accuracy of the emission inventory. On the other hand, evaluation using aircraft measurements, particularly over remote regions, provides further examination of models’ physicochemical evolution during transport. Here we use data from three phases of the ARCTAS aircraft campaign (section 3), and the results are shown in Figure 5. All model results of NO₃⁻, NH₄⁺, and SO₄²⁻ are sampled along flight track and averaged regionally within 1km vertically for each campaign phase before comparing with the corresponding aircraft measurements. Note that only EMAC, EMEP and GMI report daily 3D global tracer concentrations, while the others report monthly only. Note also that only EMEP and GMI adopt daily biomass burning emission while the others use monthly emission. To verify the representativeness of monthly mean
concentration in capturing the main features exhibited in model-observation comparisons, daily and monthly concentrations of the three models are used in the same spatial sampling to compare with the measurements (see the green lines for daily and red for monthly in the figure). The comparison keeps its main features as shown when using both daily and monthly model data.

During ARCTAS-A, which was conducted in April 2008 and was based in Fairbanks, Alaska, none of the models captures the long-range transport of aerosols primarily from Asia, which enter Polar Regions at altitudes between 2-7 km (Fig. 3 in Bian et al., 2013). Except CHASER and EMAC, all models also report a significant underestimation of NH$_4^+$ and SO$_4^{2-}$ in boundary layer. A previous assessment of pollution transport to the Arctic indicated that aerosol wet removal plays an important role in the uncertainty of Arctic aerosols (Shindell et al., 2008). Another potential reason is that some large fire activities in Siberia during April 2008 (Jacob et al., 2010) may be missed in the GFED3 emission inventory. The underestimation of SO$_4^{2-}$ may help bring up NO$_3^-$ production, particularly at high altitudes. During ARCTAS-CARB, which was conducted in June 2008 based in Palmdale, California, agreement between model and measurements is much improved. Almost all models show a rapid vertical decease from surface to free troposphere, which is consistent with the measurements of SO$_4^{2-}$ and NH$_4^+$, but not NO$_3^-$. The observation shows a maximum of NO$_3^-$ at about 1.5 km, which is not represented by any of the models. During ARCTAS-B, which was conducted in July 2008 and was based in Cold Lake, Canada, when there were frequent local wild fires, model performances are mixed. In general, most models underestimate concentrations of NO$_3^-$, NH$_4^+$ and SO$_4^{2-}$ below 4 km. CHASER model is special in that it overestimates SO$_4^{2-}$ significantly. This may be contributed to high (near surface) to comparable (free troposphere) model simulation of NH$_4^+$ but an underestimation of NO$_3^-$. Different from other models, the INCA model shows an enhancement of pollutants in the upper troposphere with concentrations much higher (more than 5 times) than observations. This behavior may be derived from a much vigorous vertical uplifting to the upper troposphere as revealed from Fig. 3a-3b combined with a low NH$_3$ Henry’s law constant used by INCA, see discussion in section 5.2.

Note that all measurements and model data we discussed above are for fine mode aerosols. Total NO$_3^-$ (orange line using monthly model output) is also shown in the figure to reveal whether a changing of partitioning of fine and coarse mode NO$_3^-$ could improve the model-observation comparison. It seems that the new version of OsloCTM3 may put too much of NO$_3^-$ in coarse mode.

4.3 Model-observation comparison for dry and wet deposition

4.3.1 Dry deposition

The budget analyses in section 4.1 concluded that dry and/or wet depositions are most likely the main processes driving the diversity in the model simulations. Thus, further evaluation of deposition processes is needed to identify any potential problematic model.
The dry depositions of NO$_3^-$, NH$_4^+$, HNO$_3$, and SO$_4^{2-}$ simulated by the models are compared against CASTNET measurements over North America (Figure 6). Generally, the overestimation of surface HNO$_3$ concentrations (Figure 3a) results in the higher dry depositions of HNO$_3$, but this is not the case for NO$_3^-$. Meanwhile, most of the models give a better dry deposition simulation for aerosol SO$_4^{2-}$ and NH$_4^+$ than for aerosol NO$_3^-$. Specifically, GISS-OMA and GISS-MATRIX have wide spread dry NO$_3^-$ deposition at any given measurement value. In other words, the two models underestimate NO$_3^-$ dry deposition significantly at many observational stations, which does not occur in the other models. This low dry deposition simulation may occur outside North America as well because the global dry depositions of the two models are lower than others (Table 4a). OsloCTM2 overestimates NO$_3^-$ dry deposition significantly, which is probably linked to its larger coarse fraction of the nitrate aerosol (see discussion in section 5.3). OsloCTM3 improved its dry deposition scheme although the model still overestimates the dry deposition. We will discuss the OsloCTM2 NO$_3^-$ simulation over North America by combining the model’s wet deposition in the next section. NH$_4^+$ dry deposition is low in GMI but very high in CHASER. This performance is also extended globally as summarized in Table 4b.

4.3.2 Wet deposition

The wet deposition simulations from the nine models are compared with surface measurement over North America (Figure 7a) and East Asia (Figure 7b) for oxidized NO$_3^-$ (i.e. total NO$_3^-$ and HNO$_3$), total NH$_4^+$ and NH$_3$ (tNH$_4^+$), and SO$_4^{2-}$. All models tend to underestimate the wet deposition of tNH$_4^+$ and SO$_4^{2-}$ over the two regions. Models EMAC, GMI, OsloCTM2 and OsloCTM3 have relatively high wet removal for oxidized NO$_3^-$, while EMEP removes much less than others over North America. All models’ wet deposition of oxidized NO$_3^-$ is biased low over East Asia. As we discussed above, OsloCTM2 and OsloCTM3 have very high dry NO$_3^-$ depositions (Figure 6) compared with CASTNET observations. The overall high dry and wet NO$_3^-$ depositions along with high atmospheric concentrations (Figure 4a) indicate that the chemical formation of NO$_3^-$ in the two models must be also high. This performance might be also true on global scale since the inferred chemical productions of NO$_3^-$ in the two models are the highest (Table 4a). CHASER has the lowest tNH$_4^+$ wet deposition. This may result in a very high NH$_4^+$ dry deposition (Figure 6) and concentration (Figures 4a-c, 5) compared with observations and other models. Overall, wet deposition seems to be the dominant process in determining the diversity in NH$_3$ and NH$_4^+$ lifetime (Table 4b).

Note that we use the traditional approach of comparing models’ grid box mean values with observations, which does not take into account the impact of the models’ horizontal resolutions in their representation of observations (Schutgens et al., 2016). Since majority models (except EMEP) have horizontal resolutions around 2-3 degrees, the models grid box means tend to smooth out extreme (i.e. very low or high) observations. Consequently, the slopes of the fitting lines are generally less than 1 on the scattering plots with model as y-axis and observation as x-axis (e.g. Figures 4a-d, 6, 7a-b).

5. Discussion of major uncertainties in nitrate formation

Large uncertainties of nitrate studies result from the complexity of the simulations which
must consider a comprehensive NOx-NMHC-O3-NH3 chemistry and a thermodynamic equilibrium model (TEQM) to partition semi-volatile ammonium nitrate between the gas and aerosol phases. Nitrate aerosol concentrations depend on temperature, relative humidity (RH), and concentrations of HNO3, NH3, NH4+, SO4²−, Cl−, Na+, Ca²+, K+, Mg²+, organic acids, among others. A further complicating factor is that the equilibrium for the coarse mode is somewhat questionable (Feng and Penner, 2007). In addition, wet removal of NH3 is very sensitive to the pH in cloud water. We will discuss some of these uncertainties below.

5.1 pH-dependent NH3 wet deposition

Gas tracer NH3, a precursor of ammonium aerosol, experiences atmospheric wet deposition and its deposition rate is typically calculated using Henry’s Law. Henry’s law constant (H) of gases in water is usually given at 298 K (indicated by Θ in superscript) and can be adjusted by temperature (T).

\[
H(T) = H^Θ \times \exp \left( -\frac{\Delta H_{sol}}{R} \left( \frac{1}{T} - \frac{1}{T^Θ} \right) \right)
\]

(1)

Here ΔHsol is the enthalpy of dissolution and R is the gas constant.

For some acidic/basic gases, including NH3, Henry’s law constant is also a function of pH in cloud water (a.k.a effective Henry’s law constant H^Θ*). As explained in the Appendix, the H^Θ* is inferred from H^Θ with a correction of pH (pH = -log10[H⁺]) as

\[
H^Θ* = H^Θ \times \frac{K_{al}[H⁺]}{K_w}
\]

(5)

Here, K_al ≈ 1.8x10⁻⁵ and K_w = 1.0x10⁻¹⁴ at 298 K in pure water (see Appendix). However, not every model accounts for pH adjustment (i.e. the reaction of equation 2 in Appendix) for NH3 dissolution. More accurately, the EMAC model implicitly calculates the effective Henry’s law constant by solving a set of partial differential equations, which includes not only the gas-liquid phase equilibria, but also the reactions in the liquid phase (i.e. dissociation or acid-base equilibria, Redox reactions and photolysis reactions in the liquid phase, see Tost et al. (2006)). Therefore, the gas-liquid phase equilibrium is explicitly calculated based on the chemical mechanism used in the liquid phase. As listed in Table 5, the rest of the models are generally divided into two groups based on their effective Henry’s law constant: (1) INCA, GISS-OMA and GISS-MATRIX has H^Θ* ≤ 100 (L-theta without pH correction) and (2) CHASER, GMI, OsloCTM2 and Oslo-CTM3 has H^Θ* > 10⁵ (H-theta with pH correction). The NH3’s H^Θ* adopted by the models varies dramatically, up to an order of 6 in magnitude among all the models and a factor of 10 just for the models in H-theta group (Table 5). The latter corresponds to a range of pH from 4.5 (Oslo-CTM2) to 5.5 (CHASER).

To examine how sensitive of NH3, NH4+ and NO3− simulations in response to the magnitude of NH3’s H^Θ*, we performed a sensitivity experiment, named TWET, in the GMI model in which there was no pH adjustment for NH3 Henry’s law constant (i.e. H^Θ* = 61 instead of 1.05e+6, see table 6). The resultant annual budgets of dry/wet deposition, chemistry production and loss, and atmospheric loading of NH3, NH4+ and NO3− are summarized in Table 7, the tracers’ vertical zonal mean distributions are shown in Figure 8, and the comparisons with the ARCTAS measurements for
NH$_4^+$ and NO$_3^-$ are shown in Figure 9. For convenient comparison, the GMI baseline results are given in the table and figures as well. There is a dramatic decrease (from 17.5 to 1.1 Tg) in NH$_3$ wet deposition when using pure water NH$_3$ Henry’s law constant. Consequently, NH$_3$ will remain in the atmosphere (i.e. ~ 8 times more atmospheric NH$_3$) to produce ~1.6 times more NH$_4^+$ chemically. This, in turn, greatly increases atmospheric NO$_3^-$ to 0.97 Tg from 0.26 Tg reported in baseline simulation. A large portion of the increased NH$_3$, NH$_4^+$ and NO$_3^-$ resides in the upper troposphere and close to the tropopause region, while the changes of the tracers in the lower troposphere are relatively small, as shown in Figure 8. These accumulations at high altitudes are far above (i.e. ~ 50 times for NH$_4^+$ and NO$_3^-$) the ARCTAS observed tracer amounts as shown in Figure 9. The TWET experiment might be an explanation of NH$_4^+$ and NO$_3^-$ accumulations near the tropopause region (Figure 3a-b) in the INCA model whose NH$_3$ Henry’s law constant H$^\Theta$ is 74 without pH correction (i.e. a L-theta model, table 5). However, it is puzzling that the NH$_3$ simulations by GISS-MATRIX and GISS-OMA, those are the models with L-theta, are closer to the simulations of the models with H-theta, i.e. no NH$_4^+$ and NO$_3^-$ accumulation near the tropopause and comparable removal of NH$_4^+$ (Figure 3a-b and Table 4b).

### 5.2 Contribution of dust and sea salt on nitrate formation

In the presence of acidic accumulation-mode sulfuric acid containing aerosols, HNO$_3$, NO$_3$ radicals, and N$_2$O$_5$ will deposit on larger alkaline mineral or salt particles (Dentener et al., 1996; Gard et al., 1998; Hauglustaine 2014; Karydis et al., 2016; Murphy and Thomson 1997; Paulot et al., 2016). Considerable evidence shows that the majority of atmospheric nitrate is formed via reactions associated with dust and sea salt (Allen et al., 2015; Itahashi et al., 2016; Karydis et al., 2016). Coarse mode nitrate overwhelmingly dominates over remote oceanic regions (Itahashi et al., 2016). Over wide land regions, nitrate also quite often exists in the form of supermicron NO$_3^-$ balanced by the presence of mineral cations arising from transport of crustal dust and sea spray aerosol (Allen et al., 2015; Lefer and Talbot; 2001).

Investigation of nitrate interactions with mineral dust and sea salt depends on the simulation approach adopted in a model. The traditional equilibrium approach to partition semi-volatile HNO$_3$ between the gas and aerosol phases is no longer possible since the time to reach equilibrium on coarse mode particles (several hours to days) is typically much longer than the chemical time step used in a global model (less than 1 hour) (John et al., 1989; Myhre et al., 2006). Meng and Seinfeld (1996) found that on longer time scales, when NH$_3$/HNO$_3$ started to condense on larger aerosols, their gas phase concentrations decreased so that some of the condensed matter can be driven back to the gas phase from the small semi-volatile aerosols. A fix to a non-equilibrium state would be to implement a kinetic formulation for the particles that have a long equilibrium time scale (Feng and Penner, 2007; Karydis et al., 2010). However, implementing explicit kinetics in a global model would be computationally expensive and, hence, is not feasible for long-term climate simulations. Several approximations have been developed to allow computational efficiency although they might compromise model accuracy. Several approximations, therefore, have been developed to compromise accuracy and efficiency.
Four such approximations are adopted by the nine models participating in this study: 1) using equilibrium calculations for fine mode particles only while neglecting nitrate formation on coarse mode particles (CHASER and GISS-MATRIX); 2) combining equilibrium calculation for a solution of $SO_4^{2-}$-$NO_3^-$-$NH_4^+$-$H_2O$ and heterogeneous reaction calculation for nitrogen uptake on dust and sea-salt using a first-order loss rate (EMEP, GMI, GISS-OMA and INCA); 3) running equilibrium model including NH$_3$ dust and sea salt repeatedly for aerosol sizes from fine mode to coarse mode (Oslo-CTM2 and Oslo-CTM3); and 4) using only the fraction of the gas that can kinetically condense within the time step of the model in the equilibrium calculations for each aerosol size mode (EMAC).

Nitrate is formed primarily on dust and sea salt by GMI (88%) and INCA (82%) (see Table 4a). INCA further separates the formation as 45% on dust and 37% on sea-salt. The above-mentioned approach 1 is problematic due to absence of coarse mode nitrate, an important portion of nitrate, which results in relatively low nitrate burdens for CHASER and GISS-MATRIX. Unfortunately, the other models are missing a detailed nitrate chemistry budget report. A potential impact of dust and sea-salt on nitrate formation, nevertheless, can be inferred from the approach adopted by a model. For example, OsloCTM2 and OsloCTM3 adopt approach 3. Although the model allows fine mode particles to reach equilibrium first, the subsequent equilibrium calculation for coarse mode particles may still produce coarse mode nitrate too quickly, see discussion of the ratio of coarse model nitrate in the next subsection. To avoid such overestimations on the production of coarse mode nitrate, EMAC allows only a fraction of HNO$_3$ to partition in the aerosol phase by assuming diffusion limited condensation (Pringle et al., 2010).

To further understand the role of homogeneous and heterogeneous chemical reaction processes in nitrate formation, we conducted two more sensitivity experiments, TnoCNH$_3$ and TnoCHET, with the GMI model (Table 6). Experiment TnoCNH$_3$ turned off chemical conversion of NH$_3$ to NH$_4^+$ in the GMI thermodynamic equilibrium model, while experiment TnoCHET excluded the nitrate formation via heterogeneous reaction of gas HNO$_3$ on the particles of dust and sea salt. The budget report, vertical zonal mean distribution and model-observation comparison of NH$_3$, NH$_4^+$ and NO$_3^-$ are given in Table 7 and Figures 8-9, respectively. It is not surprising that experiment TnoCNH$_3$ gives a higher atmospheric NH$_3$ burden (0.32 Tg) compared with baseline (0.11 Tg) with little NH$_4^+$ left (from its initial field). The interesting thing is that the formed NO$_3^-$ has only slightly decreased compared with baseline (from 0.26 to 0.20 Tg), confirming the importance of NO$_3^-$ formation via dust and sea salt. For experiment TnoCHET, the simulations of NH$_3$ and NH$_4^+$ stay the same but the formed NO$_3^-$ is decreased dramatically (from 0.26 to 0.10), indicating that NO$_3^-$ formation via NH$_3$ chemistry alone in the GMI model is relatively small. The chemical production of NO$_3^-$ is about 6 times larger in TnoCNH$_3$ (via dust and sea salt) than in TnoCHET (via NH$_3$). However, the NO$_3^-$ produced via NH$_3$ chemistry (TnoCHET) is non-negligible over remote regions impacted by long-range transport, as shown in the analysis of April Alaska observations in Figure 9.

5.3 Nitrate size distribution
Unlike sulfate aerosol, a noticeable fraction of nitrate aerosol is in coarse mode. Coarse mode aerosol nitrate is formed due to presence of dust and/or sea salt. In other words, the formation of nitrate on coarse mode dust and sea salt particles is the major factor controlling size distribution. Other factors, such as $\text{NH}_3/\text{NH}_4^+\sim\text{NO}_3^-$ chemistry and atmospheric transport and removal processes, also affect nitrate size distribution. Having an accurate aerosol size distribution is critical in climate forcing estimations, since large size particles have a relatively small optical cross section at a given aerosol mass loading and the nitrate material coating on dust particles has almost no direct impact on the dust optics, although the greatly impact dust lifetime (Bauer et al., 2007). Given that the deposition velocity of a coarse particle is greater than that of a fine particle, an accurate size distribution is also necessary to estimate deposition of particulate nitrates (Yeatman et al., 2001; Sadanaga et al., 2008). This estimation is particularly important over oceans where coarse mode nitrate dominates (Itahashi et al., 2016) and nitrogen supply is often in deficit (Hansell and Follows, 2008).

As we have discussed in section 5.2, nitrate size distribution varies with the approaches adopted for nitrate formation on coarse mode aerosols (i.e. dust and sea salt). Figure 10 gives the burdens of nitrate in fine mode and coarse mode portions and the ratio between coarse mode and total ($f_c$) for the eight discussed models. The ratio is ranging from 0 (CHASER and GISS-OMA), ~50% (EMAC, GMI and INCA), ~80% (EMEP and OsloCTM2), and 97% (OsloCTM3). The two OsloCTMs give the highest $f_c$ partially because they run TEQM model for coarse model particles.

A wide range of $f_c$, from 0 to > 90%, has been reported previously by model simulations (Adams et al., 2001; Bauer et al., 2007; Jacobson 2001), while the range is narrowed down to 40-60% for the model studies using the approach that solves dynamic mass transfer equation for coarse mode particles (Feng and Penner, 2007; Xu and Penner, 2012).

It is worth pointing out that aerosol microphysics modify aerosol size as well. For example, a process like coagulation would also allow $\text{NO}_3^-$ to mix with other particles and enter coarse mode aerosol. New particle formation/nucleation would add $\text{NH}_3/\text{NH}_4^+\sim\text{NO}_3^-$ into the ultra fine mode. Except EMAC and GISS-MATRIX, majority models involved in this study are bulk aerosol models that do not account for aerosol microphysics.

It is challenging to verify the nitrate size distribution globally due to the limited measurements on time and space. Measurements over regional and station sites indicated that the ratio of $f_c$ could be very high and vary seasonally over oceanic sites. For example, annual mean $f_c$ during 2002-2004 from the Fukue supersite observatory is about 72% with a seasonal variation of 60–80% in winter and of around 80% in summer (Itahashi et al., 2016).

However, the ratio could be varied dramatically over land or the areas affected by land pollution. For example, observations of fine and coarse particulate nitrate at several rural locations in the United States indicated that nitrate was predominantly in submicron ammonium nitrate particles during the Bondville and San Gorgonio (April) campaigns, in
coarse mode nitrate particles at Grand Canyon (May) and Great Smoky Mountains (July/August), and both fine and coarse mode nitrate during the studies at Brigantine and San Gorgonio (July) (Lee et al., 2008). Allen et al. (2015) examined aerosol composition data collected during the summer 2013 SOAS and concluded that inorganic nitrate in the southeastern United States likely exists in the form of supermicron \( \text{NO}_3^- \), balanced by the presence of mineral cations arising from the transport of crustal dust and sea spray aerosol. The measurements over Harvard Forest, a rural site in central Massachusetts, supported that the majority of nitrate mass was associated with water-soluble supermicron soil-derived \( \text{Ca}^{2+} \) in an acidic environment (Lefer and Talbot, 2001). Measurements taken in Paris during the ESQUIF campaign found that the coarse nitrate fraction represents up to 60% of total particulate nitrate mass at night and 80% at day (Hodzic et al., 2006a, 2006b). Measurements of coarse-mode aerosol nitrate and ammonium at two polluted coastal sites, Weybourne, England and Mace Head, Ireland, during polluted flow when the air had passed over strong source regions of the UK and northern Europe, showed 40–60% of the nitrate was found in particles with diameter \( >1 \mu \text{m} \), but under clean marine conditions almost 100% conversion was seen (Yeatman et al., 2001).

6. Conclusions

We present the AeroCom phase III nitrate study by assessing aerosol simulations of nitrate and ammonium and their precursors with nine global models. Five of the models couple the chemical calculation online with meteorological simulation, and four use archived meteorological fields driving chemistry. To focus on chemical-physical processes behind the diversity of nitrate simulation, all participating models are encouraged to use HTAP2 emission inventory for aerosol and gas emissions from anthropogenic, aircraft, and ship sources. The simulated aerosols of nitrate and ammonium and their precursors are compared among the models and evaluated against various measurements including surface concentrations and dry/wet depositions from surface measurements, and vertical distributions from aircraft measurements.

All models capture the main features of the distribution of nitrate and ammonium: large surface and column amounts over China, South Asia, Europe, and U.S. These regions are typically densely populated with large \( \text{NH}_3 \) and \( \text{NO}_x \) emissions. Many models also show enhanced nitrate and ammonium over the Middle East and continents over the Southern Hemisphere. The former undergoes huge dust pollution and the latter experiences fires that emit both \( \text{NH}_3 \) and \( \text{NO}_x \).

The diversity of nitrate and ammonium simulations among the models is large: the ratio of the maximum to minimum quantities among the nine models is 13.4 and 4.4 for model simulated global mass burdens of nitrate and ammonium, respectively, and 3.9 and 5.2 for the corresponding lifetimes. These values are also larger than those of sulfate: 4.0 for global burden and 3.0 for lifetime. The agreement between models and observations is better for aerosol components than for gas tracers. All models underestimate \( \text{NH}_3 \) surface mass concentrations but most models overestimate surface \( \text{HNO}_3 \) concentrations over North America and East Asia. Performance of \( \text{NH}_3 \) is the worst: this could partially be
associated to its relatively lower measurement accuracy, i.e. a loss of ammonia possibly on the filters designed to collect NH$_3$ (Williams et al., 1992). Among aerosol simulations, model performance based on evaluation of surface mixing ratio and dry/wet depositions is very similar for NH$_4^+$ and SO$_4^{2-}$, while slightly worse for NO$_3^-$. Models severely underestimate the aerosol concentrations with only a few exceptions when compared with aircraft measurements and this problem is worse over regions impacted by long-range transport than those closer to sources.

There are many intrinsic reasons for a larger diversity in nitrate simulations among models. Nitrate is involved in much more complicated chemistry: the chemical mechanism needs to handle a multiphase multicomponent solution system. The system sometimes cannot even be solved using the thermodynamic equilibrium approach when coarse mode dust and sea salt particles present. A reasonable nitrate simulation also depends on good simulations of various precursors, such as NH$_3$, HNO$_3$, dust and sea salt, although models account for impact of dust and sea salt very differently. Even an accurate simulation of SO$_4^{2-}$ is a prerequisite because SO$_4^{2-}$ surpasses NO$_3^-$ at reacting with NH$_4^+$.

The models’ intercomparison and model-observation comparison revealed at least two critical issues in nitrate simulation that demand further exploration: NH$_3$ wet deposition and relative contribution to NO$_3^-$ formation via NH$_3$ and dust/sea salt. The nine participating models adopt very different effective Henry’s law constants for NH$_3$, with one group having a value equal or less than 100 (in pure water) and the other larger than 1.e+05 (with pH correction). Sensitivity studies using the GMI model indicated that without pH correction, NH$_3$ wet deposition decreases massively (from 17.5 to 1.1 Tg), which prolongs atmospheric NH$_3$ lifetime (from 0.67 to 5.2 days) and enhances its atmospheric burden (from 0.11 to 0.85 Tg), and thus the atmospheric burden of NH$_4^+$ (from 0.17 to 0.48 Tg) and NO$_3^-$ (from 0.26 to 0.97 Tg) as well. These enhanced tracers tend to accumulate in the upper troposphere and close to the tropopause, and are too high when compared with aircraft measurements. Since liquid-phase reaction 2 in Appendix can reach equilibrium quickly within a chemical time step, we recommend including it in accounting for NH$_3$ solution. Theoretically, a more accurate approach is to combine wet removal with liquid-phase chemistry calculation. In other words, instead of using an implicit calculation of effective Henry’s law constant, the gas-liquid phase equilibrium is explicitly calculated based on the chemical mechanism used in the liquid phase. The solution of NH$_3$ is calculated by solving a set of partial differential equations, which includes not only the gas-liquid phase equilibrium, but also all the important reactions in the liquid phase, as adopted in EMAC model.

All the models use thermodynamic equilibrium to solve the chemical process of NH$_3$/NH$_4^+$ to NO$_3^-$ formation in fine mode aerosols. However, the models adopt very different ways in accounting for the contribution of these reactions on the surface of dust and sea salt particles: some account for both dust and sea salt, some account for only dust or only sea salt, and two models even do not account for any heterogeneous reactions on dust and sea salt. The methodologies that take dust and sea salt into account are also very different, i.e. together with NH$_4^+$ using thermodynamic equilibrium model or simply...
adopting a first order loss rate on dust and sea salt surfaces. The chemical budget reported
by GMI and INCA indicates that the majority (>80%) of global $\text{NO}_3^-$ formation is via
reaction on dust and sea salt. Two sensitivity experiments using the GMI model by
tagging the $\text{NO}_3^-$ formation from either NH$_3$/NH$_4^+$ chemistry or heterogeneous reactions
on dust and sea salt confirm the critical importance of the latter process, and indicate that
the former process is relatively important in remote regions. The importance of $\text{NO}_3^-$
formation on dust and sea salt lies also in its determination on nitrate particle size
distribution, so that has an implication in air quality and climate studies as well.

Our work presents a first effort to assess nitrate simulation from chemical (e.g. chemistry
among NH$_3$, NH$_4^+$, $\text{NO}_3^-$, $\text{SO}_4^{2-}$, dust and sea salt) and physical processes (e.g. emission,
dry deposition, and wet deposition). A companion study is proposed by AeroCom III
nitrate activity to investigate how sensitive is nitrate formation is in response to the
possible future changes of in emission and meteorological fields. These perturbation
fields include increasing NH$_3$ emission, decreasing NOx, SOx, and dust emissions, and
increasing atmospheric temperature and relative humidity. It would be particularly
interesting to examine how aerosol pH changes and its influence on atmospheric
acid/base gas-particle system during the experiment. Future aerosol pH does not
necessarily increase with SO$_2$ emission reduction. Indeed, studies over US southeast
indicated that its aerosol has been getting more acidic over the past decade
although SO$_2$ emission decreased and NH$_3$ emission stayed constant [Silvern et al., 2017;
Weber et al., 2016]. This environment of high aerosol acidity hinders the formation of
nitrate aerosol, which only occurs when pH is over ~2 to 3 [Weber et al., 2016]. In
addition, understanding why and how the system is insensitive to changing SO$_2$ level due
to buffering of the partitioning of semivolatile NH$_3$ over regions such as US southeast
helps us to gain some insight into how errors in sulfate (and ammonium) may propagate
to errors in aerosol nitrate. In particular, the correlation between model predictions and
observations for SO$_4^{2-}$ and NH$_4^+$ is quite poor for some models (Figure 4). If would also
be interesting to include organic gas/aerosol into the system since they are not only
important atmospheric components, but also reduce the uptake of NH$_3$. Competition for
uptake between NH$_3$ and organic gases considerably slows down the approach to
thermodynamic equilibrium [Silvern et al., 2017]. Based on the findings of this work,
modelers should pay particular attention to incorporating dust and sea salt and treating
NH$_3$ wet deposition to improve nitrate simulation. Further evaluation using satellite
measurements, such as NH$_3$ products from IASI and TES, is desired and will be
conducted. Such evaluation requires global 3-dimensional high frequency model data.
Potential future study also includes estimation of nitrate forcing for climate change.

Appendix

For some acidic/basic gases, including NH$_3$, Henry’s law constant is also a function of
pH in water (a.k.a effective Henry’s law constant). This is because not only does the
aqueous chemistry reaction NH$_3$ + H$_2$O (equation 1) reach equilibrium within a chemical
time step but its product NH$_3$H$_2$O (equation 2) does as well.

\[ \text{NH}_3 + \text{H}_2\text{O} \Leftrightarrow \text{NH}_3 \cdot \text{H}_2\text{O} \]  
\[ \text{NH}_3 \cdot \text{H}_2\text{O} \Leftrightarrow \text{NH}_4^+ + \text{OH}^- \]
Here, NH$_4^+$ is the ammonium ion and OH$^-$ is the hydroxide ion. The total dissolved ammonia [NH$_3^{T}$] is given by

$$[NH_3^{T}] = [NH_3 \cdot H_2O] + [NH_4^+]$$

$$= p_{NH_3} T \left( 1 + \frac{K_{al}[H^+]}{K_w} \right)$$

$$\approx p_{NH_3} T \left( \frac{K_{al}[H^+]}{K_w} \right)$$  \hspace{1cm} (3)$$

Here, $p_{NH_3}$ is the partial pressure of NH$_3$, $K_{al} = [NH_4^+][OH^-]/[NH_3 \cdot H_2O] \approx 1.8 \times 10^{-5}$, and $K_w = 1.0 \times 10^{-14}$ at 298 K in pure water. So the effective Henry’s law constant $H^\theta*$ is inferred from $H^\theta$ with a correction of pH ($pH = -\log_{10}[H^+]$) as

$$H^{\theta*} = H^\theta \frac{K_{al}[H^+]}{K_w}$$  \hspace{1cm} (4)$$

References:


Myhre, G., B. H., Samset, M. Schulz, Y. Balkanski, S. Bauer, T. K. Berntsen, H. Bian, N. Bellouin, M.


Phoenix, G., W. K. Hicks, S. Cinderby, J. C. I. Kuylenstierna, W. D. Stock, et al. (2006), Atmospheric nitrogen deposition in world biodiversity hotspots: the need for a greater global perspective in


Schaap, M., Müller, K., & Ten Brink, H. M. (2002). Constructing the European aerosol nitrate concentration field from quality analysed data. Atmospheric Environment, 36(8), 1323-1335.


Table 1. Nitrate chemical mechanism and physical properties of AeroCom models

<table>
<thead>
<tr>
<th>Model</th>
<th>CHEM-EQM</th>
<th>HNO₃ mechanism</th>
<th>N₂O₅ Hydrolysis</th>
<th>CHEM DUST</th>
<th>CHEM SEASALT</th>
<th>How do CHEMDUSS?</th>
<th>Bins for nitrate</th>
<th>Model Name &amp; resolution</th>
<th>References</th>
</tr>
</thead>
<tbody>
<tr>
<td>CHASER</td>
<td>ISORROPIA-I</td>
<td>CHASER (Sudo et al., 2002)</td>
<td>y⁺ (0.1 for SO₄²⁻, NO₃⁻, OC, DU, and SS, and 0.05 for liquid cloud particles) (Dentener and Crutzen, 1993)</td>
<td>No</td>
<td>No</td>
<td>---</td>
<td>Fine mode</td>
<td>MIROC, GCM, 2.8°x2.8°x64</td>
<td>Watanabe et al., 2011</td>
</tr>
<tr>
<td>EMAC</td>
<td>ISORROPIA-II</td>
<td>MESSy2 (Jöckel et al., 2010)</td>
<td>γ (STA), STA+: climatological aerosol in Aitken, accumulation, and coarse soluble modes (Jöckel et al 2010)</td>
<td>Yes</td>
<td>Yes</td>
<td>ISORROPIA-II (TEQM)</td>
<td>4 bins: Nucleation, Aitken, Accumulation, Coarse</td>
<td>ECHAM5, GCM, 2.8°x2.8°x31</td>
<td>Karydis et al., 2016</td>
</tr>
<tr>
<td>EMEP</td>
<td>MARS</td>
<td>EMEP EmChem09 (Simpson et al., 2012)</td>
<td>γ (STA, T, RH), STA: NH₄⁺, SO₄²⁻, NO₃⁻ (Evans and Jacob, 2005; Davis et al.,2008)</td>
<td>Yes</td>
<td>Yes</td>
<td>First order loss (HETCHEM)</td>
<td>Fine and coarse</td>
<td></td>
<td>Simpson et al., 2012</td>
</tr>
<tr>
<td>GMI</td>
<td>RPMARES</td>
<td>GMi (Straham et al., 2007)</td>
<td>γ(STA, T, RH), STA: BC, OC, SO₄²⁻, DU, SS (Evans and Jacob, 2005).</td>
<td>Yes</td>
<td>Yes</td>
<td>first order loss (HETCHEM)</td>
<td>3 bins: (D&lt;0.1, 0.1 – 2.5, &gt; 2.5 um)</td>
<td>MERRA2, CTM, 2.5°x2°x72</td>
<td>Bian et al., 2009</td>
</tr>
<tr>
<td>INCA</td>
<td>INCA</td>
<td>INCA tropospheric chemistry (Hauglustaine et al., 2004)</td>
<td>γ(STA, T, RH), STA: BC, SO₄²⁻, DU, SS (Evans and Jacob, 2005).</td>
<td>Yes</td>
<td>Yes</td>
<td>first order loss (HETCHEM)</td>
<td>2 bins : (D&lt; 1µm and 1 - 10µm)</td>
<td>LMD-v4, GCM, 1.9°x3.75°x39</td>
<td>Hauglustaine et al., 2014</td>
</tr>
<tr>
<td>GISS MATRIX</td>
<td>ISORROPIA-II</td>
<td>MATRIX Bauer (2008) and tropospheric chemistry (Shindell et al., 2003)</td>
<td>γ (STA), STA: SO₄²⁻¹⁻ (Dentener and Crutzen, 1993)</td>
<td>No</td>
<td>No</td>
<td>No</td>
<td>Distributed over all mixing states e.g. size distributions.</td>
<td>NASA GISS-E2, GCM, 2°x2.5°x60</td>
<td>Schmidt et al 2014</td>
</tr>
<tr>
<td>GISS OMA</td>
<td>EQSAM_v03d</td>
<td>OMA (Bauer 2007) and tropospheric chemistry (Shindell et al., 2003)</td>
<td>γ (STA), STA: SO₄²⁻¹⁻ (Dentener and Crutzen, 1993)</td>
<td>Yes</td>
<td>No</td>
<td>No</td>
<td>Fine mode</td>
<td>NASA GISS-E2, GCM, 2°x2.5°x60</td>
<td>Schmidt et al 2014</td>
</tr>
<tr>
<td>Oslo CTM2</td>
<td>EQSAM_v03d</td>
<td>Oslo CTM2 (Berntsen and Isaksen, 1997)</td>
<td>γ (STA), STA: climatology aerosol (Dentener and Crutzen, 1993; Søvde et al., 2012).</td>
<td>No</td>
<td>Yes</td>
<td>EQSAM_v03d (TEQM)</td>
<td>2 bins: fine and coarse mode</td>
<td>ECMWF, CTM, 2.8°x2.8°x60</td>
<td>Myhre et al., 2006</td>
</tr>
<tr>
<td>Oslo CTM3</td>
<td>EQSAM_v03d</td>
<td>Oslo CTM2 (Berntsen and Isaksen, 1997)</td>
<td>γ (STA), STA: climatology aerosol (Dentener and Crutzen, 1993; Søvde et al., 2012).</td>
<td>No</td>
<td>Yes</td>
<td>EQSAM_v03d (TEQM)</td>
<td>2 bins: fine and coarse mode</td>
<td>ECMWF, CTM, 2.25°x2.25°x60</td>
<td>Myhre et al., 2006</td>
</tr>
</tbody>
</table>

*CHEMDUSS: Chemistry reaction on dust and sea salt particles

γ: the dimensionless uptake coefficient

 STA: Surface of Tropospheric Aerosols

Metastable: where the aerosol is composed only of a supersaturated aqueous phase
<table>
<thead>
<tr>
<th>Model</th>
<th>CHEM-GQM</th>
<th>NO2-chem mechanism</th>
<th>CHEM DUST</th>
<th>How-do CHEMDUST</th>
<th>Bin-for nitrate</th>
<th>Model-name Accumulation</th>
<th>References</th>
</tr>
</thead>
<tbody>
<tr>
<td>CHASER</td>
<td>ISORROPIA-I</td>
<td>CHASER (Sudo et al., 2002)</td>
<td>No</td>
<td>No</td>
<td>—</td>
<td>PM-mode</td>
<td>MIROC-GCM, ECHAM4-MO主意</td>
</tr>
<tr>
<td>EMAC</td>
<td>ISORROPIA-II (Stable state)</td>
<td>MESSy2 (Jöckel et al., 2010)</td>
<td>Yes</td>
<td>Yes</td>
<td>ISORROPIA-II, Adaption, Reduction, Accumulation, Coarse</td>
<td>ECMWF-IFS, CTM, ROMS (mass)</td>
<td>Karydis et al., 2016</td>
</tr>
<tr>
<td>EMIC</td>
<td>MARS</td>
<td>EMIC for Chem09 (Simpson et al., 2009)</td>
<td>Yes</td>
<td>Yes</td>
<td>First-order loss</td>
<td>Fine and course</td>
<td>ISORROPIA-II, ISORROPIA, ECHAM5, GCM, 2.8°x2.8°x31</td>
</tr>
<tr>
<td>CMA</td>
<td>SPMAARIES (Stable state)</td>
<td>CMA (Buchardt et al., 2009)</td>
<td>Yes</td>
<td>Yes</td>
<td>First-order loss</td>
<td>Fine and course</td>
<td>ISORROPIA-II, ISORROPIA, ECHAM5, GCM, 2.8°x2.8°x31</td>
</tr>
<tr>
<td>CALA</td>
<td>INCA</td>
<td>INCA tropospheric chemistry (Hauglustaine et al., 2004)</td>
<td>Yes</td>
<td>Yes</td>
<td>First-order loss</td>
<td>Simultaneous (in 5µm and 1-10µm)</td>
<td>LMD-IAM, CTM, 1.7°Cx2.5°</td>
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<tr>
<td>GISS</td>
<td>MATRIX</td>
<td>MATRIX-Bauer (2008) and tropospheric chemistry (Shindell et al., 2003)</td>
<td>No</td>
<td>No</td>
<td>NA</td>
<td>Distributed aerosol mixing states e.g. size distributions</td>
<td>GISS-E2, CTM, 2.5°Cx2.5°</td>
</tr>
<tr>
<td>GISS</td>
<td>OMA</td>
<td>OMA (Bauer 2007) and tropospheric chemistry (Shindell et al., 2003)</td>
<td>Yes</td>
<td>No</td>
<td>Bauer and Koch, 2005</td>
<td>Fine-mode</td>
<td>GISS-E2, CTM, 2.5°Cx2.5°</td>
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<tr>
<td>Oslo</td>
<td>CTM2</td>
<td>Oslo-CTM2 (Hannigan and Koch, 2005)</td>
<td>Yes</td>
<td>Yes</td>
<td>OZh3-CO2d</td>
<td>Zero-first and coarse mode</td>
<td>ECMWF-CTM, 2.5°Cx2.5°</td>
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</tbody>
</table>
Table 2. Characteristics of thermodynamic equilibrium models

<table>
<thead>
<tr>
<th>Species</th>
<th>ISORROPIA-I</th>
<th>ISORROPIA-II</th>
<th>MARS</th>
<th>RPMARES</th>
<th>INCA</th>
<th>EQSAM v03d</th>
</tr>
</thead>
<tbody>
<tr>
<td># of components</td>
<td>23</td>
<td>34</td>
<td>16</td>
<td>11</td>
<td>9</td>
<td>18</td>
</tr>
<tr>
<td># of reactions</td>
<td>15</td>
<td>27</td>
<td>7</td>
<td>6</td>
<td>4</td>
<td>25</td>
</tr>
<tr>
<td>Multicomponent activity coefficient</td>
<td>Bromley</td>
<td>Bromley</td>
<td>Bromley</td>
<td>Bromley</td>
<td>Seinfeld and Pandis</td>
<td>Metzger</td>
</tr>
<tr>
<td>Binary activity coefficient</td>
<td>Kusik and Meissner</td>
<td>Kusik and Meissner</td>
<td>Pitzer</td>
<td>Pitzer</td>
<td>Seinfeld and Pandis</td>
<td>Metzger</td>
</tr>
<tr>
<td>Water activity</td>
<td>ZSR</td>
<td>ZSR</td>
<td>ZSR</td>
<td>ZSR</td>
<td>ZSR</td>
<td></td>
</tr>
<tr>
<td>Kelvin effect</td>
<td>No</td>
<td>No</td>
<td>No</td>
<td>No</td>
<td>No</td>
<td>No</td>
</tr>
<tr>
<td>Quantities that determine subdomains</td>
<td>[Na⁺, [NH₄]⁺, [SO₄²⁻] ]</td>
<td>[Ca²⁺, [K⁺], [Mg²⁺], [Na⁺], [NH₄]⁺, [SO₄²⁻] ]</td>
<td>RH, [NH₄]⁺, [SO₄²⁻]</td>
<td>[NH₄]⁺, [SO₄²⁻]</td>
<td>[NH₄]⁺, [SO₄²⁻]</td>
<td></td>
</tr>
<tr>
<td># of subdomains</td>
<td>4</td>
<td>5</td>
<td>4</td>
<td>2</td>
<td>3</td>
<td>3</td>
</tr>
</tbody>
</table>

*ZSR: Zdanovskii-Stokes-Robinson

Table 3. Summary of the observational data used in this study

<table>
<thead>
<tr>
<th>SURFACE NETWORK</th>
<th>QUANTITY</th>
<th>COVER AREA</th>
<th># of sites in 2008</th>
<th>SAMPLE FREQUENCY</th>
<th>SOURCE</th>
</tr>
</thead>
<tbody>
<tr>
<td>CASTNET</td>
<td>Concentration of HNO₃, NO₃⁻, NH₄⁺, SO₄²⁻</td>
<td>North America</td>
<td>83</td>
<td>weekly</td>
<td><a href="http://www.epa.gov/castnet/clearsession.do">www.epa.gov/castnet/clearsession.do</a></td>
</tr>
<tr>
<td>AMoN</td>
<td>Concentration of NH₃</td>
<td>U.S.</td>
<td>19</td>
<td>2-weekly</td>
<td><a href="http://nadp.isws.illinois.edu/">http://nadp.isws.illinois.edu/</a></td>
</tr>
<tr>
<td>NADP/NTN</td>
<td>Wet deposition of HNO₃, NO₂⁻, NH₄⁺, SO₄²⁻</td>
<td>U.S.</td>
<td>253</td>
<td>weekly</td>
<td>nadp.isws.illinois.edu</td>
</tr>
<tr>
<td>EMEP</td>
<td>Concentration of HNO₃, NH₃, NO₃⁻, NH₄⁺, SO₄²⁻</td>
<td>Europe</td>
<td>35</td>
<td>daily</td>
<td><a href="http://www.nilu.no/pr">http://www.nilu.no/pr</a> ojects/ccc/index.html</td>
</tr>
<tr>
<td>EANET</td>
<td>Concentration of HNO₃, NH₃, NO₂⁻, NH₄⁺, SO₄²⁻</td>
<td>East Asia</td>
<td>56</td>
<td>Daily to 2-weekly</td>
<td><a href="http://www.eanet.asia">http://www.eanet.asia</a> /eanet/brief.html</td>
</tr>
<tr>
<td>AIRCRAFT CAMPAIGNS</td>
<td>QUANTITY</td>
<td>COVER AREA</td>
<td># of Flights</td>
<td>CAMPAIGN PERIOD</td>
<td>SOURCE</td>
</tr>
<tr>
<td>ARCTAS-A</td>
<td>Concentration of NO₃⁻, NH₄⁺, SO₄²⁻</td>
<td>Alaska, U.S.</td>
<td>11</td>
<td>March-April</td>
<td><a href="http://www-air.larc.nasa.gov/cgi-bin/arctas-c">http://www-air.larc.nasa.gov/cgi-bin/arctas-c</a></td>
</tr>
<tr>
<td>ARCTAS-CARB</td>
<td>Concentration of NO₃⁻, NH₄⁺, SO₄²⁻</td>
<td>California Bay area U.S.</td>
<td>6</td>
<td>June</td>
<td></td>
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<tr>
<td>ARCTAS-B</td>
<td>Concentration of NO₃⁻, NH₄⁺, SO₄²⁻</td>
<td>Central Canada</td>
<td>7</td>
<td>July</td>
<td></td>
</tr>
</tbody>
</table>

Table 4a. NO₃⁻ global budget for each model

<table>
<thead>
<tr>
<th>Tracer</th>
<th>Model</th>
<th>Burden</th>
<th>SConc</th>
<th>DDep</th>
<th>WDep</th>
<th>ChemDUSS</th>
<th>ChemPS</th>
<th>Lifetime</th>
<th>AODp</th>
</tr>
</thead>
</table>

*Stable state: where salts precipitate once the aqueous phase becomes saturated
*Metastable: where the aerosol is composed only of a supersaturated aqueous phase
*CHEMDUSS: Chemistry reaction on dust and sea salt particles
Table 4b NH3 and NH$_4^+$ global budget for each model

<table>
<thead>
<tr>
<th>Tracer</th>
<th>Model</th>
<th>Emi (Tg a$^{-1}$)</th>
<th>Burden (Tg)</th>
<th>SConc (μg kg$^{-1}$)</th>
<th>DDep (Tg a$^{-1}$)</th>
<th>WDep (Tg a$^{-1}$)</th>
<th>ChemP/L$^a$ (Tg a$^{-1}$)</th>
<th>Lifetime (days)</th>
<th>AOD</th>
</tr>
</thead>
<tbody>
<tr>
<td>NH$_4^+$</td>
<td>CHASER</td>
<td>0.16</td>
<td>0.18</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>0.0076</td>
<td></td>
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<tr>
<td></td>
<td>EMAC</td>
<td>0.67</td>
<td>0.47</td>
<td>46.3</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>0.0073</td>
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</tr>
<tr>
<td></td>
<td>EMEP</td>
<td>0.96</td>
<td>0.30</td>
<td>15.0</td>
<td>62.7</td>
<td>(71.7)$^c$</td>
<td>4.5</td>
<td>0.0073</td>
<td></td>
</tr>
<tr>
<td></td>
<td>GISS-MATRIX</td>
<td>0.22</td>
<td>0.06</td>
<td>1.3</td>
<td>9.6</td>
<td>(10.9)</td>
<td>7.4</td>
<td>-</td>
<td></td>
</tr>
<tr>
<td></td>
<td>GISS-OMA</td>
<td>0.14</td>
<td>0.05</td>
<td>1.1</td>
<td>5.5</td>
<td>(6.6)</td>
<td>7.8</td>
<td>0.0153</td>
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<tr>
<td></td>
<td>GMI</td>
<td>0.26</td>
<td>0.22</td>
<td>14.9</td>
<td>31.5</td>
<td>41.9</td>
<td>4.8</td>
<td>2.1</td>
<td>0.0047</td>
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<td></td>
<td>INCA</td>
<td>0.79</td>
<td>0.17</td>
<td>4.5</td>
<td>44.6</td>
<td>44.1</td>
<td>9.8</td>
<td>5.9</td>
<td>0.0064</td>
</tr>
<tr>
<td></td>
<td>Oslo-CTM2</td>
<td>0.60</td>
<td>0.25</td>
<td>47.0</td>
<td>61.5</td>
<td>(109.3)</td>
<td>2.0</td>
<td>0.0018</td>
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<tr>
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<td>Oslo-CTM3</td>
<td>1.88</td>
<td>0.36</td>
<td>34.6</td>
<td>90.6</td>
<td>(125.2)</td>
<td>5.5</td>
<td>-</td>
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</tr>
<tr>
<td></td>
<td>Avg</td>
<td>0.63</td>
<td>0.23</td>
<td>20.7</td>
<td>45.9</td>
<td>60.6</td>
<td>6.0</td>
<td>5.0</td>
<td>0.0072</td>
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<tr>
<td></td>
<td>Med</td>
<td>0.60</td>
<td>0.22</td>
<td>15.0</td>
<td>44.6</td>
<td>46.7</td>
<td>5.5</td>
<td>3.9</td>
<td>8.5</td>
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<tr>
<td>Ratio$^d$</td>
<td>13.4</td>
<td>9.4</td>
<td>43.5</td>
<td>16.5</td>
<td>19.0</td>
<td>3.9</td>
<td>8.5</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

$^a$: ChemP refers to NO$_2$ chemical production associated with NH$_3$/NH$_4^+$

$^b$: AOD here includes NH$_4^+$ that is associated to NO$_3^-$ for all models except EMEP

$^c$: value inside parenthesis is estimated total NO$_3^-$ chemical production based on its total loss, while budget without parenthesis is reported directly by model.

$^d$: a ratio between maximum to minimum model simulations

(109.3)

(71.7)

(10.9)

(6.6)

(125.2)

(31.9)

(32.0)

(109.3)

(125.2)

(31.9)

(32.0)

(109.3)

(125.2)

(31.9)

(32.0)

(109.3)

(125.2)

(31.9)

(32.0)

(109.3)

(125.2)

(31.9)

(32.0)

(109.3)

(125.2)

(31.9)

(32.0)

(109.3)

(125.2)

(31.9)

(32.0)

(109.3)

(125.2)

(31.9)

(32.0)

(109.3)

(125.2)

(31.9)

(32.0)

(109.3)

(125.2)

(31.9)

(32.0)

(109.3)

(125.2)

(31.9)

(32.0)

(109.3)

(125.2)

(31.9)

(32.0)

(109.3)

(125.2)

(31.9)

(32.0)

(109.3)

(125.2)

(31.9)

(32.0)

(109.3)

(125.2)

(31.9)

(32.0)

(109.3)

(125.2)

(31.9)

(32.0)

(109.3)

(125.2)

(31.9)

(32.0)

(109.3)

(125.2)

(31.9)

(32.0)

(109.3)

(125.2)

(31.9)

(32.0)

(109.3)

(125.2)

(31.9)

(32.0)

(109.3)

(125.2)

(31.9)

(32.0)

(109.3)

(125.2)

(31.9)

(32.0)

(109.3)

(125.2)

(31.9)
b chemical budgets inside parenthesis are inferred based on the reported emission and total deposition
c EMAC gives total wet deposition of NH$_4^+$ and NH$_3$
d Statistic values of NH$_4^+$ wet deposition do not include EMAC
e INCA uses ECLIPSE anthropogenic emissions, two GISS models use CMIP5 anthropogenic emission, and all other models use HTAPv2 anthropogenic emissions

### Table 4c. HNO$_3$ global budget for each model

<table>
<thead>
<tr>
<th>Tracer</th>
<th>Model</th>
<th>Burden ($Tg$)</th>
<th>SConc (μg kg$^{-1}$)</th>
<th>DDep ($Tg$ a$^{-1}$)</th>
<th>WDep ($Tg$ a$^{-1}$)</th>
<th>CheAP ($Tg$ a$^{-1}$)</th>
<th>CheGP ($Tg$ a$^{-1}$)</th>
<th>CheAL ($Tg$ a$^{-1}$)</th>
<th>CheGL ($Tg$ a$^{-1}$)</th>
<th>Lifetime (days)</th>
</tr>
</thead>
<tbody>
<tr>
<td>HNO$_3$</td>
<td>CHASER</td>
<td>1.1</td>
<td>0.29</td>
<td>74.0</td>
<td>120.9</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>EMAC</td>
<td>3.1</td>
<td>0.32</td>
<td>56.1</td>
<td>136.0</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>EMEP</td>
<td>0.66</td>
<td>0.04</td>
<td>39.2</td>
<td>123.9</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>GISS-MATRIX</td>
<td>5.7</td>
<td>0.12</td>
<td>61.7</td>
<td>167.5</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>GISS-OMA</td>
<td>5.3</td>
<td>0.10</td>
<td>49.8</td>
<td>148.2</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>GMI</td>
<td>1.8</td>
<td>0.18</td>
<td>39.7</td>
<td>128.1</td>
<td>413</td>
<td>42.6</td>
<td>299</td>
<td>3.5</td>
<td></td>
</tr>
<tr>
<td></td>
<td>INCA</td>
<td>1.5</td>
<td>0.09</td>
<td>47.7</td>
<td>77.5</td>
<td>569</td>
<td>10.0</td>
<td>210</td>
<td>5.7</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Oslo-CTM2</td>
<td>1.3</td>
<td>0.05</td>
<td>36.1</td>
<td>66.0</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>Oslo-CTM3</td>
<td>2.3</td>
<td>0.04</td>
<td>36.0</td>
<td>49.3</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>Avg</td>
<td>2.5</td>
<td>0.14</td>
<td>45.8</td>
<td>108.7</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>Med</td>
<td>1.8</td>
<td>0.10</td>
<td>43.7</td>
<td>123.9</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>Ratio</td>
<td>8.6</td>
<td>8.0</td>
<td>1.6</td>
<td>3.4</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
</tbody>
</table>

**SO$_4^{2-}$ burden for the atmosphere up to 100 hPa**

**CheAP:** chemistry production from aerosol phase

**CheGP:** chemistry production from gas phase

**CheAL:** chemistry loss from aerosol phase

**CheGL:** chemistry loss from gas phase

**for both HNO$_3$ and NO$_3^-$**

**statistical values do not include CHASER and EMAC that report total dry or wet deposition of HNO$_3$ and NO$_3^-$**

### Table 4d. SO$_4^{2-}$ global budget for each model

<table>
<thead>
<tr>
<th>Tracer</th>
<th>Model</th>
<th>Emi SO2 ($Tg$ a$^{-1}$)</th>
<th>Emi SO4 ($Tg$ a$^{-1}$)</th>
<th>Burden ($Tg$)</th>
<th>SConc (μg kg$^{-1}$)</th>
<th>DDep ($Tg$ a$^{-1}$)</th>
<th>WDep ($Tg$ a$^{-1}$)</th>
<th>Chem GP ($Tg$ a$^{-1}$)</th>
<th>Chem AqPb ($Tg$ a$^{-1}$)</th>
<th>Lifetime (days)</th>
<th>AOD</th>
</tr>
</thead>
<tbody>
<tr>
<td>SO$_4^{2-}$</td>
<td>CHASER</td>
<td>111</td>
<td>0</td>
<td>3.3</td>
<td>1.44</td>
<td>22.1</td>
<td>137</td>
<td>(159)</td>
<td>7.6</td>
<td>0.0826</td>
<td></td>
</tr>
<tr>
<td></td>
<td>EMAC</td>
<td>138</td>
<td>6.19</td>
<td>1.9</td>
<td>1.72</td>
<td>504</td>
<td>302</td>
<td>(187)</td>
<td>0.86</td>
<td>-</td>
<td></td>
</tr>
<tr>
<td></td>
<td>EMEP</td>
<td>109</td>
<td>0.83</td>
<td>0.45</td>
<td>10.2</td>
<td>109</td>
<td>(119)</td>
<td>2.5</td>
<td>0.0232</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>GISS-MATRIX</td>
<td>133</td>
<td>5.1</td>
<td>1.3</td>
<td>0.63</td>
<td>16.6</td>
<td>97</td>
<td>(109)</td>
<td>4.2</td>
<td>-</td>
<td></td>
</tr>
<tr>
<td></td>
<td>GISS-OMA</td>
<td>133</td>
<td>5.1</td>
<td>1.1</td>
<td>0.53</td>
<td>11.8</td>
<td>112</td>
<td>52.7</td>
<td>66.2</td>
<td>3.3</td>
<td>0.0714</td>
</tr>
<tr>
<td></td>
<td>GMI</td>
<td>111</td>
<td>0</td>
<td>1.1</td>
<td>0.58</td>
<td>7.5</td>
<td>205</td>
<td>126.5</td>
<td>86.1</td>
<td>3.6</td>
<td>0.0257</td>
</tr>
<tr>
<td></td>
<td>INCA</td>
<td>116</td>
<td>8.0</td>
<td>1.8</td>
<td>0.34</td>
<td>8.4</td>
<td>116</td>
<td>42.2</td>
<td>75.1</td>
<td>5.3</td>
<td>0.0417</td>
</tr>
<tr>
<td></td>
<td>Oslo-CTM2</td>
<td>133</td>
<td>4.1</td>
<td>2.0</td>
<td>0.49</td>
<td>17.6</td>
<td>184</td>
<td>(198)</td>
<td>3.6</td>
<td>0.0366</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Oslo-CTM3</td>
<td>133</td>
<td>4.1</td>
<td>2.7</td>
<td>0.55</td>
<td>20.2</td>
<td>160</td>
<td>(176)</td>
<td>5.5</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Avg</td>
<td>122</td>
<td>1.8</td>
<td>0.63</td>
<td>14.3</td>
<td>140</td>
<td>151</td>
<td>4.5</td>
<td>0.0469</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
Table 5: Effective Henry Law constant used in the models

<table>
<thead>
<tr>
<th>Aerocom Model</th>
<th>$H^*$ (M/atm)</th>
<th>$-\Delta H_{m}/R$ (K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>CHASER</td>
<td>3.0e+5</td>
<td>3400</td>
</tr>
<tr>
<td>EMAC</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>EMEP</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>GIS MATRIX</td>
<td>1.0e+2</td>
<td>3415</td>
</tr>
<tr>
<td>GISS OMA</td>
<td>1.0e+2</td>
<td>3415</td>
</tr>
<tr>
<td>GMI</td>
<td>1.05e6</td>
<td>4200</td>
</tr>
<tr>
<td>INCA</td>
<td>7.4e+1</td>
<td>3400</td>
</tr>
<tr>
<td>Oslo-CTM2</td>
<td>3.3e+6</td>
<td>0</td>
</tr>
<tr>
<td>Oslo-CTM3</td>
<td>3.3e+6</td>
<td>0</td>
</tr>
</tbody>
</table>

a EMAC: See its wet deposition description in section 4.1.1.
b EMEP: The model does not use Henry law but applies simple empirical scavenging ratio, which for NH$_3$ is 1.4e6 for in-cloud and 0.5e6 for below-cloud scavenging. The scavenging ratio by definition is the ratio the concentration of a certain pollutant in precipitation divided by the concentration of the pollutant in air.

c Effective Henry Law constant from 1.05e+6 (pH=5.0) to 62 (pure water)
d EMAC dry deposition includes sedimentation and SO$_4^{2-}$ sedimentation is very high since it has assumed that 7.7% of sea salt is SO$_4^{2-}$
e Statistical values do not include EMAC

Table 6. Baseline and three sensitivity experiments in the GMI model

<table>
<thead>
<tr>
<th>Experiment</th>
<th>Setup</th>
<th>Purpose</th>
</tr>
</thead>
<tbody>
<tr>
<td>BASE</td>
<td>Standard simulation as described in section 2.1</td>
<td>Baseline simulation</td>
</tr>
<tr>
<td>TWET</td>
<td>Set NH$_3$ effective Henry law constant from 1.05e+6 (pH=5.0) to 62 (pure water)</td>
<td>Review impact of NH$_3$ wet deposition</td>
</tr>
<tr>
<td>TnoNH3</td>
<td>Turn off NO$_3^-$ production from NH$_3$/NH$_4^+$</td>
<td>Identify how large/where the NO$_3^-$ formation from NH$_3$/NH$_4^+$</td>
</tr>
<tr>
<td>TnoHET</td>
<td>Turn off NO$_3^-$ production from dust and sea salt</td>
<td>Identify how large/where the NO$_3^-$ formation from dust and sea salt</td>
</tr>
</tbody>
</table>

Table 7: NO$_3^-$, NH$_4^+$, NH$_3$ and HNO$_3$ budgets from the base simulation and three sensitivity experiments

<table>
<thead>
<tr>
<th>Tracer</th>
<th>Exp</th>
<th>Burden (Tg)</th>
<th>SConc (μg kg$^{-1}$)</th>
<th>DDep (Tg a$^{-1}$)</th>
<th>WDep (Tg a$^{-1}$)</th>
<th>ChemDUSS (Tg a$^{-1}$)</th>
<th>ChemP (Tg a$^{-1}$)</th>
<th>Lifetime (days)</th>
</tr>
</thead>
<tbody>
<tr>
<td>NO$_3^-$</td>
<td>BASE</td>
<td>0.26</td>
<td>0.22</td>
<td>14.9</td>
<td>31.5</td>
<td>41.9</td>
<td>4.8</td>
<td>2.1</td>
</tr>
<tr>
<td></td>
<td>Twet</td>
<td>0.97</td>
<td>0.23</td>
<td>14.8</td>
<td>43.3</td>
<td>41.0</td>
<td>18.3</td>
<td>6.0</td>
</tr>
<tr>
<td></td>
<td>TnoNH3</td>
<td>0.20</td>
<td>0.17</td>
<td>14.7</td>
<td>27.5</td>
<td>42.3</td>
<td>0</td>
<td>1.7</td>
</tr>
<tr>
<td></td>
<td>TnoHET</td>
<td>0.099</td>
<td>0.065</td>
<td>0.61</td>
<td>6.70</td>
<td>0</td>
<td>7.1</td>
<td>5.0</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Tracer</th>
<th>Model</th>
<th>Emi</th>
<th>Burden (Tg)</th>
<th>SConc</th>
<th>DDep</th>
<th>WDep</th>
<th>ChemP/L</th>
<th>Lifetime (days)</th>
</tr>
</thead>
</table>

33
NH₄⁺ BASE 0.17 0.14 1.7 30.6 32.2 1.9
Twet 0.48 0.16 1.9 50.7 53.0 3.4
TnoNH3 - - - - - -
TnoHET 0.17 0.14 1.6 30.6 32.2 1.9

NH₃ BASE 60.4 0.11 0.40 12.6 17.5 30.4 0.67
Twet 0.85 0.81 8.70 1.1 50.1 5.2
TnoNH3 0.32 0.58 20.9 39.3 0 1.9
TnoHET 0.10 0.40 12.6 17.4 30.4 1.2

Figure 1. The observational station locations for CASTNET surface concentrations (CASTNET Conc), Ammonia surface monitor network over U.S. (AMON), CASTNET dry deposition (CASTNET DDEP); National Acid Deposition Network for wet deposition over U.S. (NADP NTN), surface concentrations over Europe (EMEP), and surface dry and wet deposition over Asia (EANET).
Figure 2. Flight-tracks of ARTCTA-A (left), ARCTAS-CARB (middle), and ARCTAS-B (right). The colors represent observations during different days.