



Sensitivity of nitrate optical depth to ammonia emissions and nitrate chemistry

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Sensitivity of nitrate aerosols to ammonia emissions and to nitrate chemistry: implications for present and future nitrate optical depth

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Abstract

We update and evaluate the treatment of nitrate aerosols in the Geophysical Fluid Dynamics Laboratory (GFDL) atmospheric model (AM3). Accounting for the radiative effects of nitrate aerosols generally improves the simulated aerosol optical depth, although nitrate concentrations at the surface are biased high. This bias can be reduced by increasing the deposition of nitrate to account for the near-surface volatilization of ammonium nitrate or by neglecting the heterogeneous production of nitric acid to account for the inhibition of N_2O_5 reactive uptake at high nitrate concentrations. Globally, uncertainties in these processes can impact the simulated nitrate optical depth by up to 25 %, much more than the impact of uncertainties in the seasonality of ammonia emissions (6 %) or in the uptake of nitric acid on dust (13 %). Our best estimate for present-day fine nitrate optical depth at 550 nm is 0.006 (0.005–0.008). We only find a modest increase of nitrate optical depth (< 30 %) in response to the projected changes in the emissions of SO_2 (–40 %) and ammonia (+38 %) from 2010 to 2050. Nitrate burden is projected to increase in the tropics and in the free troposphere, but to decrease at the surface in the midlatitudes because of lower nitric acid concentrations. Our results suggest that better constraints on the heterogeneous chemistry of nitric acid on dust, on tropical ammonia emissions, and on the transport of ammonia to the free troposphere are needed to improve projections of aerosol optical depth.

1 Introduction

Ammonium nitrate (NH_4NO_3) aerosols are produced by the reaction of nitric acid (HNO_3), a photochemical product of NO oxidation, and ammonia (NH_3). Emissions of NH_3 and NO are primarily from anthropogenic origin: fossil fuel combustion for NO and agriculture for NH_3 (Bouwman et al., 1997; Paulot et al., 2014). The formation of NH_4NO_3 is favored by cold temperatures and high relative humidity (Stelson and Seinfeld, 1982). NH_4NO_3 production competes with that of ammonium sulfate, which

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Ming et al. (2006). For radiative calculations, aerosols are assumed to be externally mixed except for sulfate and hydrophilic black carbon, which are assumed internally mixed (Donner et al., 2011). Nitrate is not considered for radiative calculations in AM3.

A new configuration of AM3 is introduced (referred to as AM3N hereafter) with the following changes aimed at improving the simulation of nitrate aerosols (see Sect. 3):

Aerosol chemistry – We use ISORROPIA to simulate the sulfate-nitrate-ammonia thermodynamic equilibrium (Fountoukis and Nenes, 2007). Equilibrium between gas and aerosol is assumed to be reached at each model time step (30 min), which is generally justified for $PM_{2.5}$ (Meng and Seinfeld, 1996). In-cloud oxidation of SO_2 is restricted to liquid clouds and we revise the calculation of cloud pH to account for the partitioning of HNO_3/NO_3^- and NH_3/NH_4^+ between the gas phase and cloud water.

Heterogeneous chemistry on dust – We include the heterogeneous uptake of HNO_3 , NO_3^- , N_2O_5 , SO_2 , and H_2SO_4 on dust particles (Table S1 in the Supplement). The uptake of HNO_3 , NO_3^- , and N_2O_5 is assumed to be limited by alkalinity (Song and Carmichael, 2001). Following Fairlie et al. (2010), dust alkalinity is comprised of calcium and magnesium carbonates, with calcium and magnesium constituting 3 and 0.6 % (by mass) of coarse dust emissions (radius > 1 μm), respectively. Observations suggest alkalinity is primarily found in the coarse mode (Claquin et al., 1999); we assume that fine dust carries half as much alkalinity per kg as coarse dust.

Nitrate optical depth – The optical properties and the mixing with black carbon of ammonium nitrate are assumed to be identical to those of ammonium sulfate. This approximation introduces an error in mass extinction at 550 nm of less than 20 % for $RH < 95\%$. We neglect the optical properties of NO_3^- on dust.

Dry deposition – Similar to AM3, the dry deposition of gases and fine aerosols are calculated based on a monthly climatology of deposition velocities. We update this climatology to account for recent observations of rapid deposition of H_2O_2 and some oxygenated volatile organic compounds (OVOCs), using the deposition velocities calculated in the GEOS-Chem chemical transport model as described by Nguyen et al. (2015).

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Wet deposition – In AM3, aerosol removal by snow is treated like that by rain. In AM3N, water-soluble aerosols are not removed by snow, when the snow is formed via the Wegener–Bergeron–Findeisen mechanism (referred to as Bergeron mechanism hereafter), i.e., when water evaporates from liquid cloud droplets and condenses onto growing ice crystals. This treatment is consistent with observations (Henning et al., 2004) and similar to that used in other global models (Liu et al., 2011; Wang et al., 2011; Fan et al., 2012). Scavenging by snow formed via riming and homogeneous freezing is treated like that by rain. Gases are not scavenged by snow except HNO₃ (Neu and Prather, 2012). Convective plumes are discretized on a vertical grid that has finer vertical resolution than AM3 (Donner, 1993). The improved discretization of the convective plume increases the convective wet removal of tracers as we will show in Sect. 3.

2.2 Emissions

We use anthropogenic emissions from the Hemispheric Transport of Air Pollution v2 (HTAP_v2) task force regridded to 0.5° × 0.5° for years 2008 and 2010 (Janssens-Maenhout et al., 2015). HTAP_v2 aircraft emissions are distributed vertically following Lamarque et al. (2010). Daily biomass burning emissions are based on the NCAR Fire INventory (FINNv1, Wiedinmyer et al., 2011) and emitted in the model surface layer. Average dust emissions are parameterized following Ginoux et al. (2001), as:

$$F_p = CSs_p u_{10m}^2 (u_{10m} - u_t) \text{ if } u_{10m} > u_t \quad (1)$$

where C is a dimensional factor ($\mu\text{gs}^2\text{m}^{-5}$), S is the source function based on topography, u_{10m} is the horizontal wind at 10 m (ms^{-1}), u_t is the threshold velocity (ms^{-1}), and s_p is the fraction of total dust emitted in the size class p as defined by Li et al. (2008). Over the 2008–2010 period, dust emission is 1640Tga^{-1} . This includes 1230Tga^{-1} from natural sources (S from Ginoux et al. (2001), $C = 0.125\mu\text{gs}^2\text{m}^{-5}$, $u_t = 1\text{ms}^{-1}$), similar to the AEROCOM multi-model mean (Huneus et al., 2011), and 410Tga^{-1}

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(Lamsal et al., 2010). Nitrate formation is expected to be sensitive to this reaction as both NH_4NO_3 and N_2O_5 are more stable under cold conditions. The impact of heterogeneous chemistry on NO_y depends on the choice of reaction probabilities (γ ; Paulot et al., 2013; Macintyre and Evans, 2010), which remain uncertain (Chang et al., 2011). Field and laboratory observations show that N_2O_5 heterogeneous uptake is suppressed by aerosol nitrate and organics (Brown et al., 2009; Brown and Stutz, 2012; Wagner et al., 2013; Gaston et al., 2014), but favored by cold temperatures (Griffiths and Anthony Cox, 2009; Wagner et al., 2013). In AM3N, we use $\gamma_{\text{N}_2\text{O}_5} = 0.01$, one order of magnitude less than in AM3 (Mao et al., 2013c) and similar to the global mean value $\gamma_{\text{N}_2\text{O}_5} = 0.02$ derived by Evans and Jacob (2005). In AM3N_nhet, we neglect the heterogeneous production of HNO_3 via NO_3 , NO_2 , and N_2O_5 aerosol uptake, similar to Bellouin et al. (2011). We also evaluate the impact of heterogeneous chemistry on dust. In AM3N_ndust, we neglect the uptake of HNO_3 , N_2O_5 , NO_3 , H_2SO_4 , and SO_2 on dust, similar to Pye et al. (2009) and Bellouin et al. (2011).

2.3.3 Surface removal of fine NO_3^-

In AM3N, the dry deposition of NH_4NO_3 is slow similar to other fine aerosols. Several field observations have reported steeper vertical gradients and faster deposition velocities (v_d) for NO_3^- than for SO_4^{2-} (Huebert et al., 1988; Wyers and Duyzer, 1997; Van Oss et al., 1998; Rattray and Sievering, 2001; Nemitz et al., 2004; Fowler et al., 2009; Wolff et al., 2010; Barbaro et al., 2015). This difference stems from gradients in temperature, RH, and HNO_3 within the boundary layer, which reduce the stability of NH_4NO_3 near the surface. The volatilization NH_4NO_3 may result in an underestimate of the surface deposition of $\text{TNO}_3 \equiv \text{HNO}_3 + \text{NO}_3^-$ since $v_d(\text{NH}_4\text{NO}_3) \ll v_d(\text{HNO}_3)$. As an upper bound, we assume that the surface removal of fine NO_3^- is limited by turbulent transport by setting $v_d(\text{NO}_3^-) = v_d(\text{HNO}_3)$ in AM3N_fdep.

in AM3N than in AM3 reflects the change in the speciation of NH_x and the faster dry deposition of NH_3 relative to NH_4^+ . The lifetime of NH_x in AM3N (2.5 days) is similar to that derived by Xu and Penner (2012) and Hauglustaine et al. (2014) (2.3 days).

AM3N and AM3 differ most strikingly in their simulations of NO_y . The contribution of HNO_3 to the removal of NO_y decreases from 81 % (AM3) to 56 % (AM3N). In contrast, the contribution of aerosols to NO_y removal increases from 2 to 22 %. Recent studies (Hauglustaine et al., 2014; Xu and Penner, 2012) find an even greater contribution of aerosols to the removal of NO_y (> 30 %); this different may reflect the lack of HNO_3 uptake by seasalt in AM3N. Organic nitrogen contributes 10 % of NO_y removal in both AM3 and AM3N. The much lower fraction of NO_y deposited as HNO_3 in AM3N relative to AM3 reflects both the increased production of NH_4NO_3 and the uptake of HNO_3 on dust. The total heterogeneous production of HNO_3 by N_2O_5 (9.7 Tg Na^{-1}), NO_2 (0.6 Tg Na^{-1}), and NO_3^- (0.4 Tg Na^{-1}) uptake on fine aerosols is reduced by 50 % in AM3N relative to AM3. This decrease is primarily driven by reduced reaction probabilities for NO_2 and NO_3^- uptake. In contrast, the change of $\gamma_{\text{N}_2\text{O}_5}$ from 0.1 (AM3) to 0.01 (AM3N) reduces the heterogeneous uptake of N_2O_5 by only 20 % because of the large increase in the sulfate surface area in winter (see Sect. 3). The magnitude of the N_2O_5 source of HNO_3 in AM3N is three times as large as reported by Hauglustaine et al. (2014). This may reflect greater reactive aerosol surface area in AM3N, as N_2O_5 hydrolysis can take place on SO_4^{2-} , BC, OC, and NO_3^- aerosols, while only SO_4^{2-} is considered by Hauglustaine et al. (2014). Reduction in the simulated HNO_3 burden – driven by faster NO_3^- deposition (AM3N_fdep), heterogeneous uptake of HNO_3 on dust (AM3N_ndust), or reduced heterogeneous production of HNO_3 (AM3N_nhet) – increase cloud pH, which favors the oxidation of SO_2 by O_3 (Table S2).

Figure 2 shows the burden of fine NO_3^- , NO_3^- on dust, NH_4^+ , and NH_3 in AM3N. The simulated global burdens fall within the range of previous estimates (Bauer et al., 2007; Feng and Penner, 2007; Pye et al., 2009; Pringle et al., 2010; Bellouin et al., 2011; Xu and Penner, 2012; Hauglustaine et al., 2014) for fine NO_3^- (0.04–0.11 TgN), NO_3^- on dust (0.07–0.41 TgN), NH_4^+ (0.21–0.27 TgN), and NH_3 (0.07–0.29 TgN). The burden

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defined as:

$$GR = \frac{[NH_3] + [NH_4^+] - 2[SO_4^{2-}]}{[HNO_3] + [NO_3^-]} \quad (2)$$

GR defines three different regimes: (a) $GR > 1$, in which NH_4NO_3 formation is limited by the availability of HNO_3 , (b) $0 < GR < 1$, in which NH_4NO_3 is limited by the availability of NH_3 , and (c) $GR < 0$, in which NH_4NO_3 is inhibited by SO_4^{2-} . We define the degree of limitation of NH_4NO_3 by HNO_3 ($\mathcal{L}(HNO_3)$) as the fraction of the time when $GR > 1$. In winter, NH_4NO_3 is most frequently limited by HNO_3 ($\mathcal{L}(HNO_3) = 78\%$ in AM3N). Figure 4 (bottom panel) shows $\mathcal{L}(HNO_3)$ binned by NO_3^- concentrations. NH_4NO_3 is most limited by HNO_3 availability at low $[NO_3^-]$, while NH_3 becomes more limiting at high $[NO_3^-]$. This suggests that even in an environment that is generally NH_3 -rich with respect to NH_4NO_3 formation, NH_3 emissions modulates NO_3^- production during high- NO_3^- episodes (AM3N_ns).

Figure 4 also shows that AM3N_nhet and AM3N_fdep produce different distributions of daily $[NO_3^-]$ although they have similar mean monthly $[NO_3^-]$ (top panel). AM3N_fdep reproduces observations at low NO_3^- concentrations well but underestimates the frequency of high NO_3^- events, when NH_4NO_3 exhibits significant sensitivity to NH_3 . Under these conditions, less volatilization of NH_4NO_3 near the surface is expected as NH_3 is not depleted near the surface like HNO_3 . AM3_nhet $[NO_3^-]$ is most consistent with observations at high $[NO_3^-]$, conditions under which N_2O_5 heterogeneous uptake has been observed to be inhibited both in laboratory and field settings (Bertram and Thornton, 2009; Wagner et al., 2013). The ability of AM3N_fdep and AM3N_nhet to capture NO_3^- under different conditions emphasizes the need to represent the dynamic nature of $\gamma(N_2O_5)$ and TNO₃ surface removal.

Figure 5 shows the observed and simulated monthly AOD at Bondville. Observed AOD peaks in summer and reaches a minimum in winter. This seasonality is well captured by AM3 (top panel), while AOD in AM3N_fdep_diu (bottom panel) peaks in spring

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AM3N_fdep_diu better captures surface $[\text{NO}_3^-]$ and we will focus on this configuration in the following. Note that the diurnal cycle of NH_3 emissions has a small effect on the simulated $[\text{NO}_3^-]$ concentration on a continental scale, but reduces surface $[\text{NH}_3]$ and increases the export of NH_3 to the free troposphere.

Figure 8 shows the average monthly variation of AOD from 2008 to 2010 over different regions as observed by MODIS (Remer et al., 2008) and MISR (Kahn et al., 2009) and simulated by AM3 and AM3N_fdep_diu. Although AM3 does not exhibit a large bias on a global scale, it fails to capture the seasonality of AOD over most continental regions. Over North America, AOD is biased low in winter and high in summer in AM3, consistent with the biases in surface $[\text{SO}_4^{2-}]$. The spring bias may be exacerbated by insufficient transport of aerosols from Asia. AM3 is biased high over tropical land masses, consistent with insufficient convective removal of aerosols. AM3N_fdep_diu AOD shows improved correlations with observations over most continental regions (see also Fig. S18). The increased AOD in winter and spring can be partly attributed to nitrate optical depth, which accounts for up to $\sim 35\%$ of AOD over North America.

Following Lee and Adams (2010) and Shindell et al. (2013), we further evaluate the performances of AM3 and AM3N in locations within the top decile of simulated NO_3^- and SO_4^{2-} burden against observations from MODIS, MISR, and AERONET. AM3 AOD is biased high over high SO_4^{2-} regions and low over high NO_3^- regions consistent with the analysis of Shindell et al. (2013). The bias over high SO_4^{2-} regions is greatly reduced in AM3N, while the model exhibits a high bias against satellite AOD observations but little bias against AERONET observations in high NO_3^- regions. More detailed comparisons with AERONET show that AM3N better captures AOD at high latitude in spring (Fig. S18), which lends support to the changes made to the representation of in-cloud sulfate production and wet deposition.

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nario as well as an increase in the conversion rate from NH_3 to NO_3^- from 0.36 day^{-1} to 0.57 day^{-1} .

Figure 11 shows that the simulated NO_3^- optical depth decreases in all AM3N configurations over Europe and China, increases over India, and exhibits little change over the US. In all regions SO_2 emissions are projected to decrease. This results in greater sensitivity of NO_3^- optical depth to HNO_3 , which is reflected in the increase of the sensitivity of NO_3^- optical depth to the uptake of HNO_3 by dust and lower sensitivity to temporal variations of NH_3 emissions (seasonality, diurnal cycle). The sensitivity of NO_3^- optical depth to the heterogeneous production of HNO_3 is reduced despite the increased sensitivity of NO_3^- to HNO_3 . This follows the decrease in aerosol surface area associated with the reduction of the SO_4^{2-} burden.

The simulated changes in NO_3^- optical depth from present-day to 2050 over the US, China, and Europe are consistent with surface NH_4NO_3 limitations. For instance, surface NH_4NO_3 is primarily limited by HNO_3 in Europe and China and the decrease of NO_3^- optical depth is driven by the reduction of NO emissions. In these regions, AM3N simulates similar NO_3^- optical depth using different anthropogenic emissions of NH_3 for 2050, which is also consistent with the reduced sensitivity to NH_3 emissions. However, surface NH_4NO_3 limitation patterns cannot explain the increase of NO_3^- optical depth over India.

Figure 12 shows that the NO_3^- burden is projected to shift equatorward in the Northern Hemisphere in response to changes in anthropogenic emission from present-day to 2050. This shift is less pronounced when using anthropogenic NH_3 emissions from RCP8.5 for 2050 because of lower NH_3 emissions over India. The lower NH_3 emissions in RCP8.5 over India also result in opposite trends for NO_3^- optical depth in this region (Fig. 11). Figure 12 also shows that the meridional redistribution of NO_3^- is associated with an increase of the free tropospheric burden of NO_3^- , while NO_3^- near the surface decreases. A similar vertical redistribution of NO_3^- was also noted by Hauglustaine et al. (2014). In the free troposphere, NH_4NO_3 formation is limited by NH_3 (Fig. 7),

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which suggests that the increase of the NO_3^- burden in AM3N from present-day to 2050 is primarily driven by an increase of the NH_3 flux to the free troposphere associated with greater tropical emissions. The lower tropical NO_3^- burden in AM3N compared to the estimate of Hauglustaine et al. (2014) likely results from a lower flux of NH_3 to the free troposphere in AM3N (e.g., from different biomass burning injection heights, convective schemes). Our study suggests that changes in this flux due to changes in emission and climate will play a large role in determining future NO_3^- optical depth.

5 Conclusions

We have developed a new configuration of AM3 (AM3N) with revised treatment of nitrate and sulfate chemistry and deposition. We showed that AM3N better captures observed AOD than a configuration of AM3 similar to that used for ACCMIP and CMIP5. AM3N overestimates surface NO_3^- concentration especially in the US. This bias may reflect neglect in AM3N of the dynamic nature of N_2O_5 uptake and near-surface volatilization of NH_4NO_3 .

We have evaluated the sensitivity of NO_3^- optical depth to poorly constrained aspects of NO_3^- chemistry (heterogeneous production of HNO_3 , uptake of HNO_3 by natural and anthropogenic dust, surface removal of NH_4NO_3) and NH_3 emissions (diurnal cycle, seasonality). Globally, the formation of NH_4NO_3 is more limited by HNO_3 than NH_3 , such that NO_3^- optical depth is more sensitive to the representation of the heterogeneous chemistry of HNO_3 than to uncertainties in NH_3 emissions. Simulated present-day NO_3^- optical depth ranges from 0.0054 to 0.0082, depending on the treatment of reactive nitrogen. Differences in the treatment of reactive nitrogen alone are unlikely to account for the large spread in estimates of present-day NO_3^- optical depth (0.0023–0.025).

We have examined the response of simulated NO_3^- optical depth to projected changes in anthropogenic emissions from 2010 to 2050 in RCP8.5. AM3N simulates only a small increase in NO_3^- optical depth (< 30 %) under present-day climate con-

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**Table 1.** Simulated budget of SO_4 , NH_x , and NO_y in 2010.

	AM3	AM3N
SO_4		
Production (TgSa^{-1})	37.3	33.1
OH	10.4	7.7
H_2O_2	26.7	16.2
O_3	0.1	4.5
dust	0.0	1.9
Loss (TgSa^{-1})	37.4	33.3
Dry deposition	4.7	4.6
SO_4	4.7	3.8
SO_4 on dust	0.0	0.8
Wet deposition	32.7	28.7
SO_4	32.7	27.5
SO_4 on dust	0.0	1.1
Lifetime (days)	4.9	3.8
NH_x		
NH_3 emission (TgNa^{-1})	54.5	54.5
Loss (TgNa^{-1})	54.8	55.0
Dry deposition	14.4	23.5
Wet deposition	40.4	30.7
Gas oxidation	0.0	0.8
Lifetime (days)	5.5	2.5
NO_y		
NO emission (TgNa^{-1})	51.4	51.8
Loss (TgNa^{-1})	51.3	51.0
Dry deposition	25.4	23.1
HNO_3	18.3	10.7
NO_3 on dust	0.0	3.4
NH_4NO_3	0.7	0.8
Organic nitrogen	3.9	4.0
Wet deposition	25.6	27.6
HNO_3	23.4	17.8
NO_3 on dust	0.0	3.7
NH_4NO_3	0.5	3.5
Organic nitrogen	1.7	2.6
Lifetime (days)	22.7	13.4

* SO_2 emissions are 74.0 TgSa^{-1} including 16.0 TgSa^{-1} from DMS oxidation.

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Table 2. Configurations of AM3N used in this study.

	Temporal variation of NH ₃ emissions	Heterogeneous chemistry on dust	Heterogeneous production of HNO ₃	Dry deposition of NH ₄ NO ₃
AM3N	Monthly	Yes	Yes	SO ₄ ²⁻
AM3N_fdep	Monthly	Yes	Yes	HNO ₃
AM3N_diu	Monthly+Diurnal	Yes	Yes	SO ₄ ²⁻
AM3N_ns	No	Yes	Yes	SO ₄ ²⁻
AM3N_nhet	Monthly	Yes	No	SO ₄ ²⁻
AM3N_ndust	Monthly	No	Yes	SO ₄ ²⁻
AM3N_fdep_diu	Monthly+Diurnal	Yes	Yes	HNO ₃

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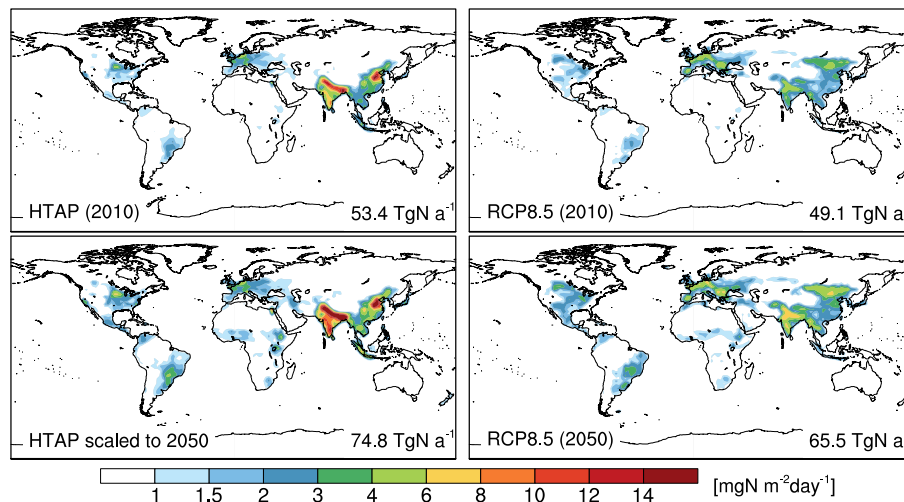


Figure 1. Average annual emissions of NH_3 for 2010 (top row) and 2050 (bottom row) based on anthropogenic NH_3 emissions from HTAP_v2 (left column) and from RCP8.5 (right column). Non anthropogenic emissions (including biomass burning) are the same in all scenarios. Total annual emissions are indicated inset.

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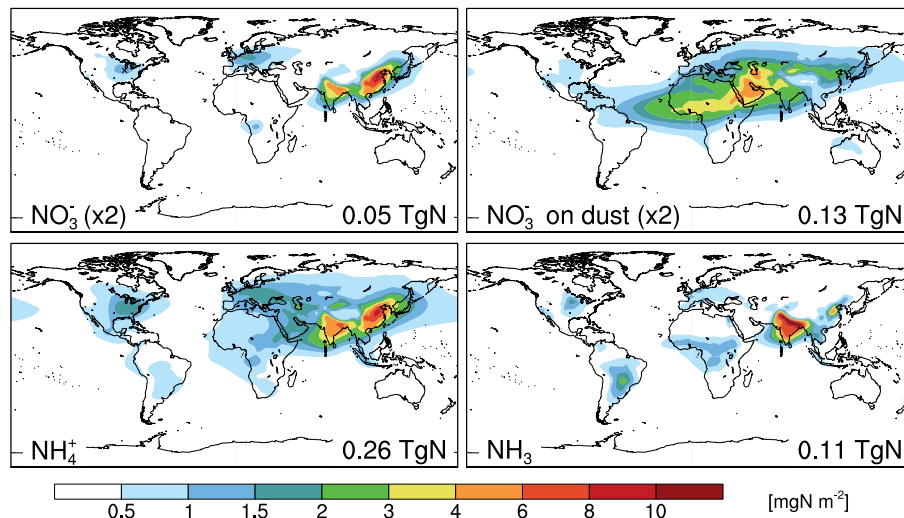


Figure 2. Annual mean burden of NO_3^- , NO_3^- on dust, NH_4^+ , and NH_3 in mg N m^{-2} in AM3N from 2008 to 2010. Global burdens are indicated inset. The location of the Bondville site is indicated by a black cross.

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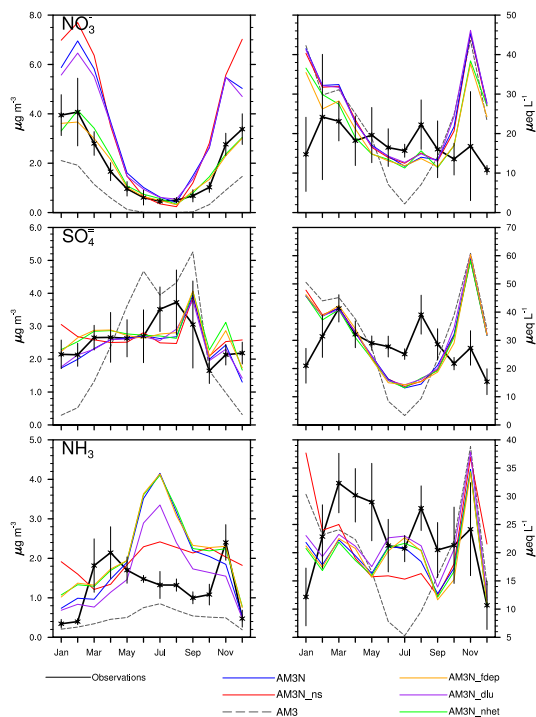


Figure 3. Observed (black) and simulated monthly concentrations of NO_3^- , SO_4^{2-} , and NH_3 at Bondville (40.1°N , 88.4°W) in surface air (left panel) and precipitated water (right panel). Observations are averaged from the 2006 to 2012, while model output is from 2008 to 2010. The vertical bars denote one standard deviation of the mean monthly observations. The different model sensitivity experiments are described in Table 2.

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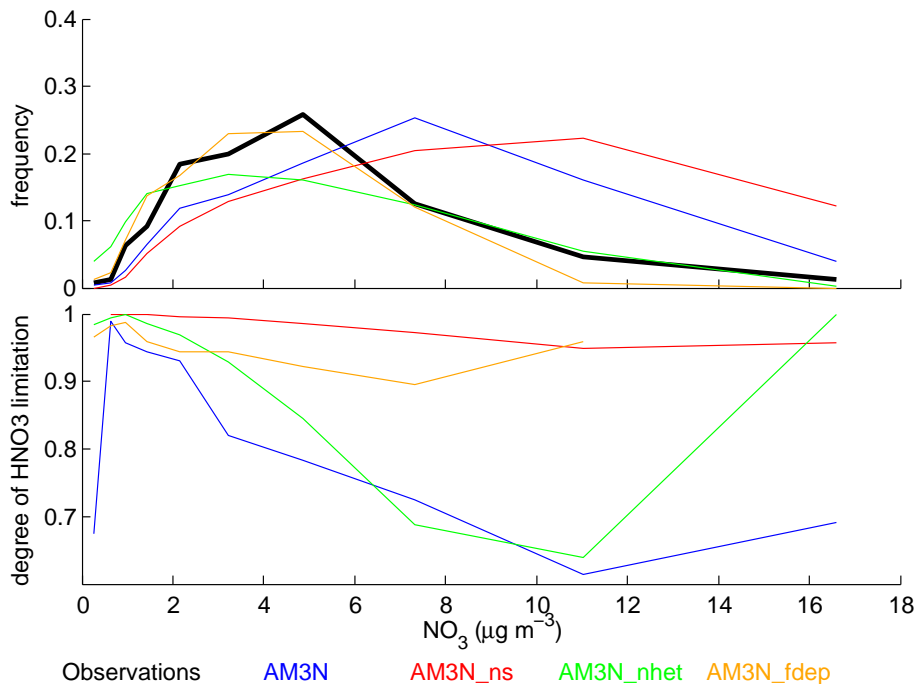


Figure 4. Observed and simulated distribution of daily NO_3^- concentration at Bondville (40.1°N , 88.4°W) in winter (top panel) from 2006 to 2012 (observations) and 2008 to 2010 (model). The degree of HNO_3 limitation for NH_4NO_3 formation ($\text{GR} > 1$) is shown in the bottom panel. The different model sensitivity experiments are described in Table 2.

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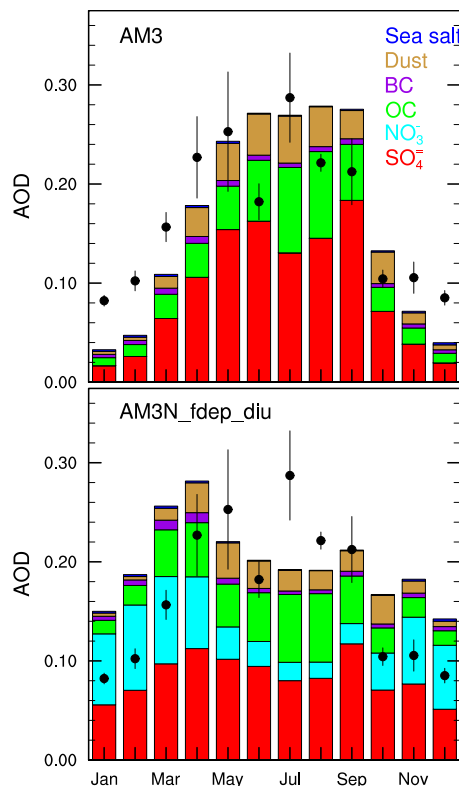


Figure 5. Observed and simulated aerosol optical depth at 550 nm at Bondville (40.1° N, 88.4° W) in AM3 and AM3N_fdep_diu. Observations (black crosses) are averaged from 2006 to 2012 and the thin vertical black bars denote one standard deviation of the mean. Thick color bars show the simulated optical depth of SO_4^{2-} (red), NO_3^- (cyan), OC (green), BC (purple), dust (brown), and sea salt (blue) for AM3N_fdep_diu (2008–2010 average).

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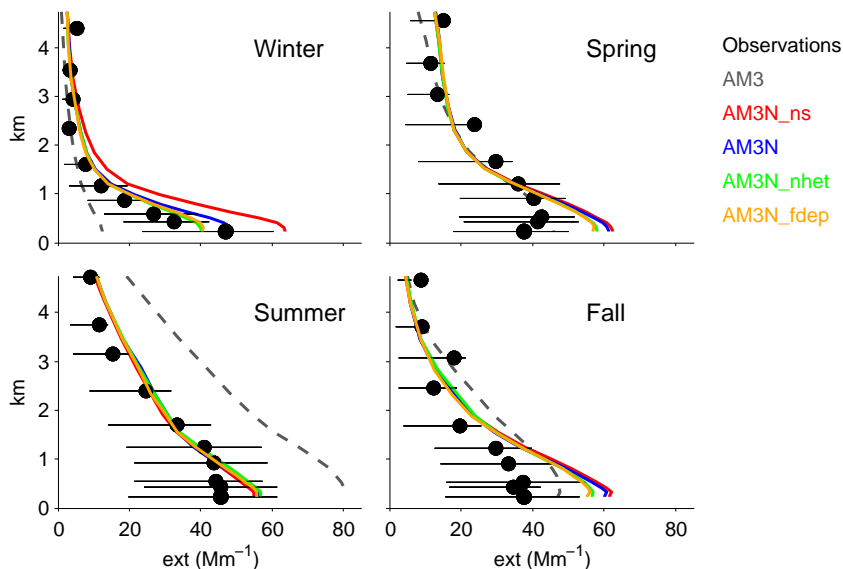


Figure 6. Mean seasonal observed (black dots) and simulated surface and vertical profiles of aerosol dry extinction at Bondville (40.1° N, 88.4° W). The vertical profile show the average of all observations by the Airborne Aerosol Observatory from 2006 to 2009 collected during daytime (10 a.m.–4 p.m. local time). Surface observations reflect the average of all daytime observations at the ESRL BND station from 2006 to 2012 with no local pollution. Model is averaged for daytime from 2008 to 2010. Horizontal lines show the 25th to 75th percentiles of observed dry aerosol extinctions. Dry extinctions are reported at standard temperature and pressure (273.15 K, 1 atm). We multiply the modeled nitrate extinction by 0.8 to account for the evaporation of ammonium nitrate in the nephelometer (Bergin et al., 1997). The different model sensitivity experiments are described in Table 2.

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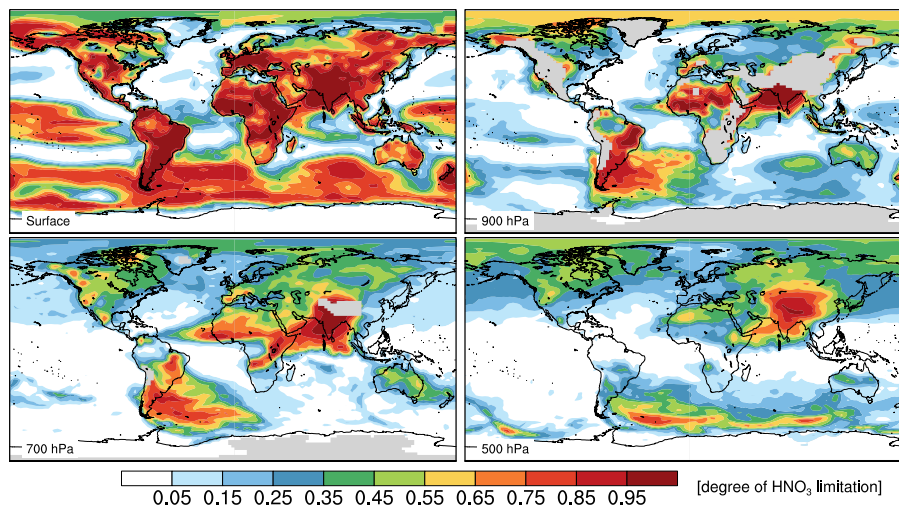


Figure 7. Simulated degree of limitation of NH_4NO_3 formation by HNO_3 ($\text{GR} > 1$) weighted by NH_4NO_3 concentration at different pressure levels in AM3N for 2010.

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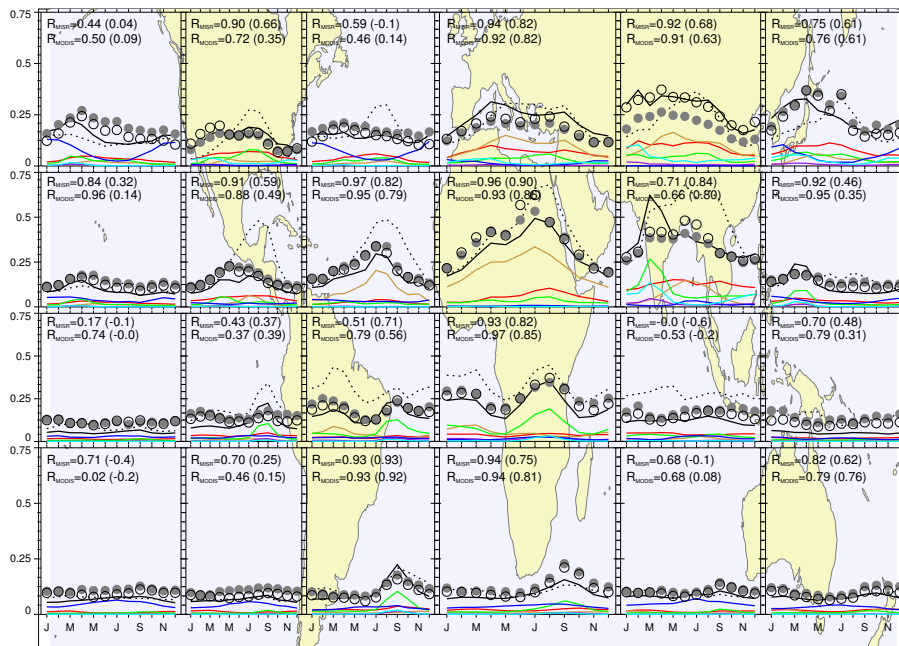


Figure 8. Observed and simulated monthly AOD at 550 nm in different regions averaged over the 2008–2010 period. Circles show observations from MODIS (open circles) and MISR (filled circles). The solid and dash black line show the AOD simulated by AM3N_fdep_diu and AM3 respectively. We also show the simulated optical depths of sulfate (red), nitrate (cyan), dust (brown), organic carbon (green), black carbon (purple), and sea salt (blue) in AM3N_fdep_diu. The model is sampled to match the the location and time of valid measurements by both MODIS and MISR in each region. Correlations between simulated and observed AOD are shown inset for AM3N_fdep_diu and AM3 (in parenthesis).

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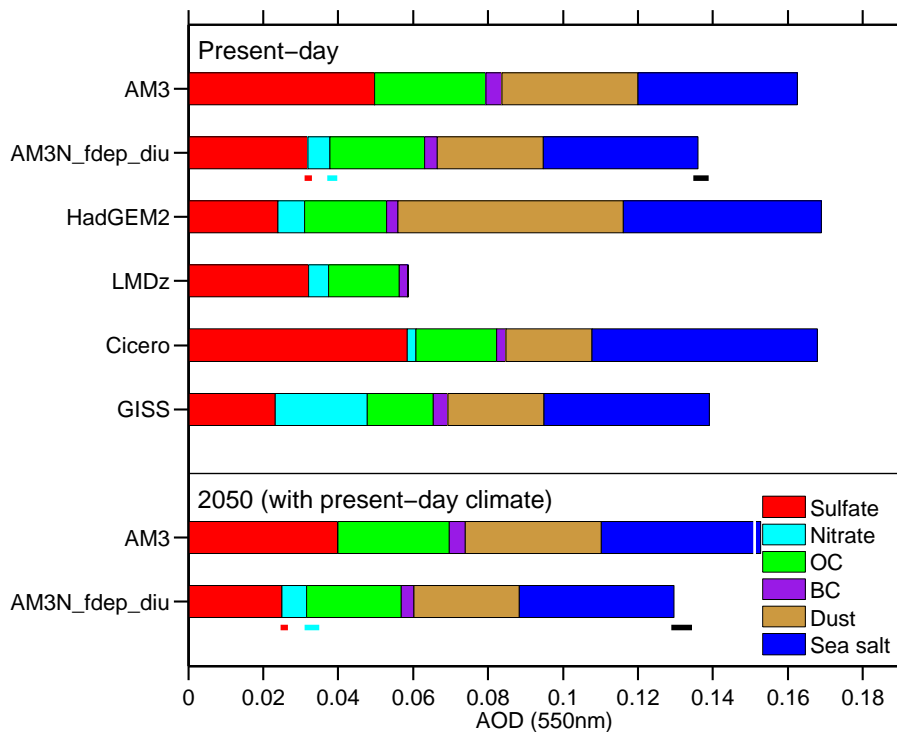


Figure 9. Contribution of different aerosol types to the global mean annual aerosol optical depth at 550 nm in AM3, AM3N, and other climate models considering NO_3^- aerosol. Note that clear-sky AOD is used for the GISS model. AM3 and AM3N AOD are representative of 2010 conditions, while other models reflect 2000 conditions. The range of SO_4^{2-} , NO_3^- , and total AOD across AM3N are shown by red, light blue, and black horizontal bars respectively.

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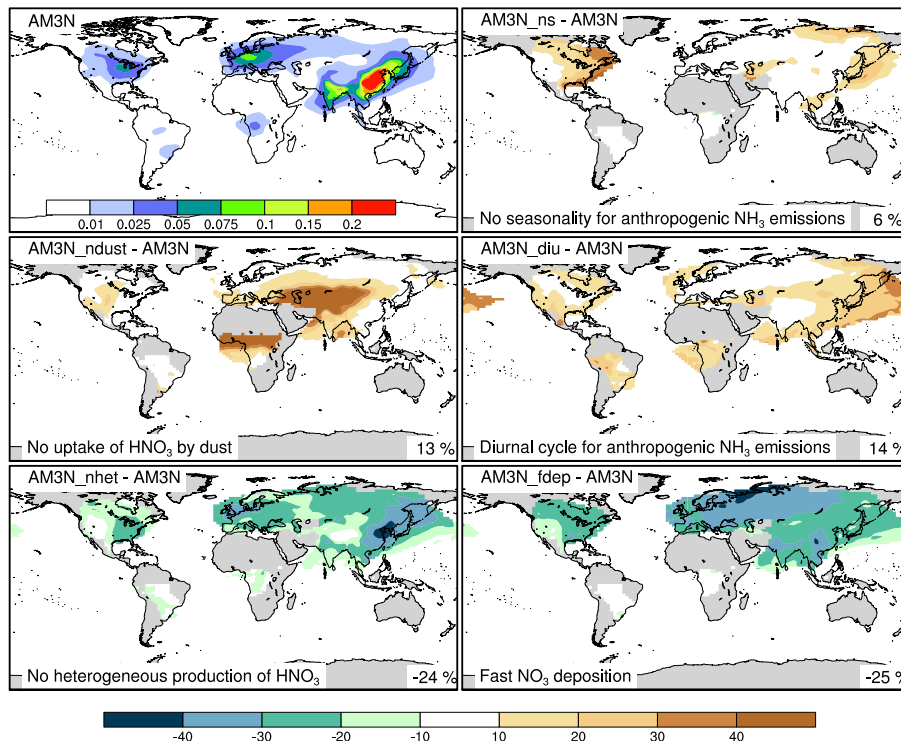


Figure 10. Annual mean NO_3^- optical depth at 550 nm in AM3N (top left panel) and its relative sensitivity to the treatment of NH_3 emissions, NO_3^- production, and loss in % for 2008–2010 conditions. The change in NO_3^- optical depth relative to AM3N is indicated in the bottom left for each configuration. The sensitivity is only shown in regions, where NO_3^- optical depth is greater than 0.005.

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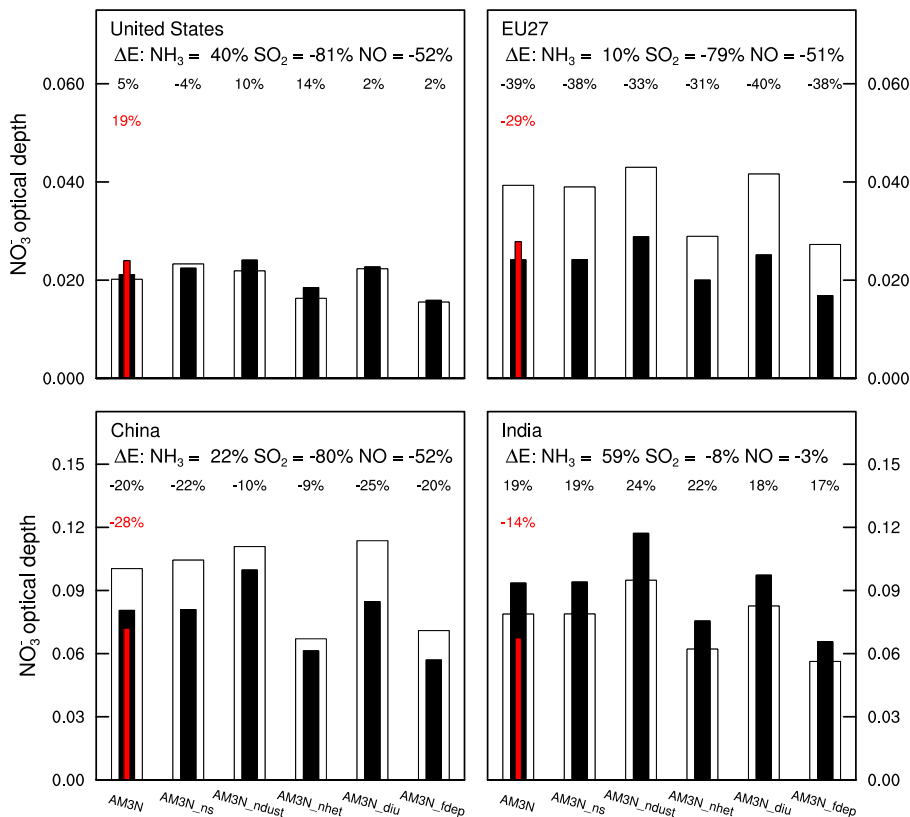


Figure 11. Nitrate optical depth at 550 nm over the United States, Europe, China, and India for 2008–2010 (white bars) and 2050 (black bars) anthropogenic emissions for different configurations of AM3N. The thin red bar indicates the nitrate optical depth calculated using RCP8.5 2050 NH₃ emissions in AM3N. The relative changes between 2008–2010 and 2050 in NO₃ optical depth and surface emissions of NH₃, SO₂, and NO are indicated for each region.

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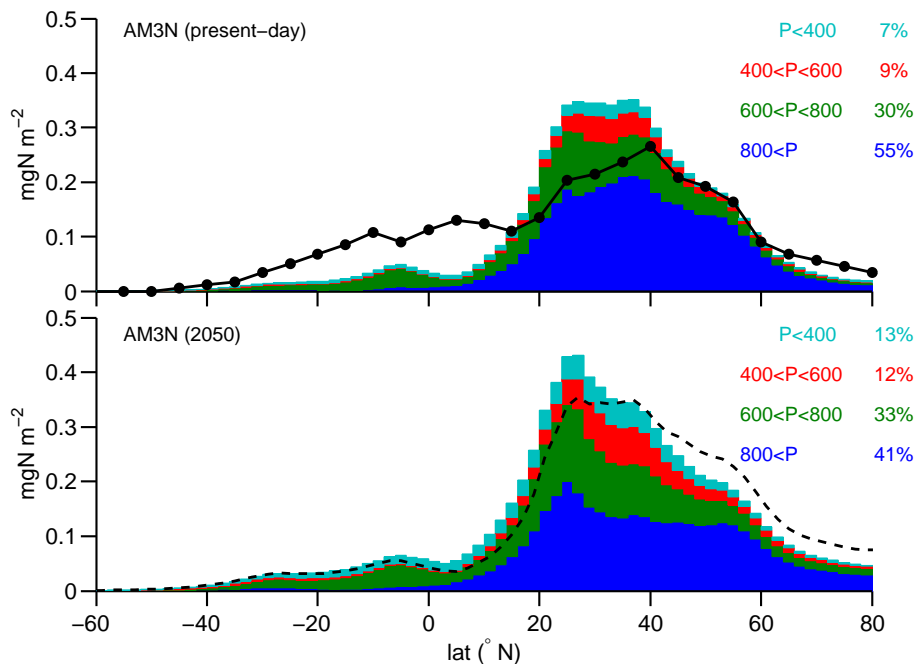


Figure 12. Annual zonal mean distribution of NO_3^- in AM3N with present-day anthropogenic emissions (top) and 2050 anthropogenic emissions (from RCP8.5 except for NH_3 , see text). The blue, green, red, and cyan regions denote the NO_3^- burden located above 800 hPa, between 600 and 800 hPa, between 400 and 600 hPa, and below 400 hPa, with the global fraction in each pressure range indicated inset. The black solid line in the top panel indicates the 2000 annual zonal mean NO_3^- burden simulated by Hauglustaine et al. (2014). The dash black line in the bottom panel indicates the annual zonal mean NO_3^- burden simulated by AM3N using 2050 anthropogenic emissions from RCP8.5 for all compounds.