

1 Summertime nitrate aerosol in the upper troposphere and lower stratosphere
2 over the Tibetan Plateau and the South Asian summer monsoon region

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26 **Abstract**

27 We use the global three-dimensional Goddard Earth Observing System
28 chemical transport model (GEOS-Chem) to examine the contribution of nitrate
29 aerosol to aerosol concentrations in the upper troposphere and lower
30 stratosphere (UTLS) over the Tibetan Plateau and the South Asian summer
31 monsoon (TP/SASM) region during summertime of year 2005. Simulated
32 surface-layer aerosol concentrations are compared with ground-based
33 observations, and simulated aerosols in the UTLS are evaluated by using the
34 Stratospheric Aerosol and Gas Experiment II satellite data. Simulations show
35 elevated aerosol concentrations of sulfate, nitrate, ammonium, black carbon,
36 organic carbon, and PM_{2.5} (particles with diameter equal or less than 2.5 μm,
37 defined as the sum of sulfate, nitrate, ammonium, black carbon, and organic
38 carbon aerosols in this study) in the UTLS over the TP/SASM region
39 throughout the summer. Nitrate aerosol is simulated to be of secondary
40 importance near the surface but the most dominant aerosol species in the
41 UTLS over the studied region. Averaged over summertime and over the
42 TP/SASM region, C_{NIT} (the ratio of nitrate concentration to PM_{2.5} concentration)
43 values are 5–35% at the surface, 25–50% at 200 hPa, and could exceed 60%
44 at 100 hPa. The mechanisms for the accumulation of nitrate in the UTLS over
45 the TP/SASM region include vertical transport and the gas-to-aerosol
46 conversion of HNO₃ to form nitrate. The high relative humidity and low
47 temperature associated with the deep convection over the TP/SASM region
48 are favorable for the gas-to-aerosol conversion of HNO₃.

49 **1 Introduction**

50 Aerosols in the upper troposphere and lower stratosphere (UTLS) have much
51 longer residence times than those in the lower troposphere, influencing
52 atmospheric chemistry and the Earth's climate with large spatial and temporal
53 coverage (Rasch et al., 2008). Aerosols in the UTLS influence the
54 concentrations of chemical species via changes in photolysis rates and
55 heterogeneous reactions (Pitari et al., 2014). For example, heterogeneous
56 reactions on sulfate aerosol can perturb the chemical partitioning in the lower
57 stratosphere, leading to significant O₃ depletion through enhanced chlorine,
58 bromine, and odd-hydrogen catalytic cycle (Zhao et al., 1997; Considine et al.,
59 2001; Talukdar et al., 2012; Tang et al., 2014; Pitari et al., 2014). Aerosols in
60 the UTLS also influence climate by altering properties of cirrus clouds via
61 homogeneous or heterogeneous ice nucleation (Li et al., 2005; Liu et al., 2009;
62 Yin et al., 2012; Fadnavis et al., 2013). Injection of aerosols into the UTLS has
63 been reported to induce complex responses in circulation, temperature, and
64 water vapor (Liu et al., 2009; Wu et al., 2011; Su et al., 2011; Fadnavis et al.,
65 2013).

66 Aerosols over the Tibetan Plateau (TP) and the Asian summer monsoon
67 region are especially important. The TP is surrounded by countries with large
68 anthropogenic emissions (Li et al., 2005; Lau et al., 2006). Aerosols from India,
69 Southeast Asia, and southern China can be transported to the TP by prevailing
70 winds in the premonsoon and monsoon seasons (Lawrence and Lelieveld,
71 2010; Xia et al., 2011). Observational and modeling studies have shown that
72 persistent maxima of atmospheric constituents, such as water vapor
73 (Gettelman et al., 2004; Randel and Park, 2006; Park et al., 2007), CO (Kar et

74 al., 2004; Li et al., 2005; Park et al., 2007, 2008, 2009), CH₄ (M. Park et al.,
75 2004; Xiong et al., 2009), NO_x (M. Park et al., 2004), HCN (Park et al., 2008;
76 Randel et al., 2010), C₂H₆ and C₂H₂ (Park et al., 2008), exist in the UTLS
77 above the TP and the South Asian summer monsoon (SASM) region because
78 of the deep convection during boreal summer. Satellite observations
79 suggested that the convection associated with the SASM is a vital pathway to
80 transport air mass from the lower troposphere into the stratosphere (Chen et
81 al., 2006; Randel and Park, 2006; Randel et al., 2010; Bian et al., 2011a). The
82 heating associated with the persistent deep convection during summertime
83 leads to the formation of the Tibetan anticyclone in the UTLS, which acts to
84 isolate air within the anticyclone and traps the uplifted pollutants at that altitude
85 (Park et al., 2007; Vernier et al., 2011; Bourgeois et al., 2012; Fadnavis et al.,
86 2013; He et al., 2014). The stratosphere-troposphere exchange (STE) over the
87 TP contributes largely to the global STE (Chen et al., 2006).

88 Previous studies have reported that aerosols exist in the UTLS over the
89 TP/SASM region. Kim et al. (2003) carried out optical measurements with a
90 ground-based lidar in Lhasa from August to October of 1999, and found an
91 enhancement in aerosol concentration near the local tropopause with
92 scattering ratio (SR, the ratio of aerosol plus molecular backscatter to
93 molecular backscatter alone) of 1.1–1.2. Tobo et al. (2007) reported an
94 enhancement of sub-micron aerosols (effective radius $r = 0.15\text{--}0.6\ \mu\text{m}$) near
95 the summertime tropopause (about 130 to 70 hPa), on the basis of in situ
96 balloon measurements from an Optical Particle Counter at the same location in
97 August of 1999. Vernier et al. (2009) examined satellite measurements from
98 the Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) onboard

99 Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation (CALIPSO)
100 and reported the presence of small depolarizing particles with high SR values
101 (about 1.20 at 532 nm) at 16–17 km altitude over South Asia in July and
102 August of 2007 and 2008. Bourgeois et al. (2012) found that an aerosol layer
103 existed at 16–18 km altitude over the Asian continent and Indian Ocean
104 (20°S–30°N, 5–105°E) on the basis of the CALIOP observations. Recently, He
105 et al. (2014) examined the vertical profiles of aerosol extinction coefficients
106 measured with a Micro Pulse Lidar at Naqu, a meteorological station located in
107 the central part of the TP, and also showed a maximum in aerosol extinction
108 coefficient ($\sim 2.10^{-3} \text{ km}^{-1}$) in the UTLS (18–19 km) during the summer of 2011.

109 A number of previous studies have attempted to understand the chemical
110 composition of aerosols in the UTLS. Froyd et al. (2009) measured aerosol
111 composition with the National Oceanic and Atmospheric Administration (NOAA)
112 single-particle mass spectrometer aboard the National Aeronautics and Space
113 Administration (NASA) WB-57 high altitude aircraft platform, and reported that
114 particles in the tropical tropopause layer were rich in nitrogen. Vernier et al.
115 (2011) suggested that aerosol layer at the tropopause of Asia could be sulfur
116 and/or organics, considering that Asian pollutants consisted of black carbon,
117 organic carbon, SO₂, and NO_x (Park et al., 2009; Randel et al., 2010). Weigel
118 et al. (2011) analyzed the volatility of aerosols obtained from in situ airborne
119 measurements and reported that about 75–90 % of the particles in the tropical
120 tropopause layer were volatile, but this study did not give any detailed
121 analyses of chemical composition of aerosols. Bourgeois et al. (2012) showed,
122 by using the ECHAM5.5-HAM2 model, that sulfate, water, and OC contributed,
123 respectively, 53%, 29%, and 11% to aerosol extinction in the vicinity of the

124 tropical tropopause layer. The ECHAM5.5-HAM2 model used by Bourgeois et
125 al. (2012) simulated all major aerosol species in the atmosphere except for
126 nitrate.

127 Few previous studies have examined nitrate aerosol in the UTLS, although
128 nitrate is expected to be important for the following reasons. First, emissions of
129 precursors of nitrate, such as NO_x and NH_3 , are high over India, Southeast
130 Asia, and China (Streets et al., 2003; Datta et al., 2012; Huang et al., 2012).
131 Second, simulated nitrate concentrations are high over those regions (Liao and
132 Seinfeld, 2005; Mu and Liao, 2014; Lou et al., 2014). Third, measured
133 concentrations of nitrate are comparable to or larger than those of sulfate at
134 rural and urban sites in the SASM region. Shrestha et al. (2000) carried out
135 measurements of aerosols at Phortse, Nepal, during September
136 1996–November 1997, and showed that the average concentration of nitrate
137 during the monsoon season (June–September) was $0.34 \mu\text{g m}^{-3}$, higher than
138 that of sulfate ($0.17 \mu\text{g m}^{-3}$). Decesari et al. (2010) reported, on the basis of
139 measurements at the Nepal Climate Observatory–Pyramid from 2006 to 2008,
140 that the concentrations of nitrate and sulfate were $0.37 \mu\text{g m}^{-3}$ and $0.50 \mu\text{g m}^{-3}$,
141 respectively, during the monsoon season. Chatterjee et al. (2010) measured
142 aerosols at a high altitude station in northeastern Himalayas during
143 January–December 2005. They found that the average concentrations of
144 fine-mode nitrate and sulfate were $3.31 \pm 2.25 \mu\text{g m}^{-3}$ and $3.80 \pm 2.9 \mu\text{g m}^{-3}$,
145 respectively. At Lahore, an urban site in Pakistan, the observed daytime nitrate
146 concentration of $21.8 \mu\text{g m}^{-3}$ was also higher than sulfate concentration of 12.6
147 $\mu\text{g m}^{-3}$ (Lodhi et al., 2009), as the observations were averaged over November
148 2005 to March 2006. Fourth, the low temperatures in the UTLS would favor

149 nitrate formation (Seinfeld and Pandis, 2006). Therefore, it is of interest to take
150 nitrate aerosol into consideration when we examine aerosols in the UTLS.

151 In this work we simulate nitrate aerosol and its contribution to aerosol
152 concentrations in the UTLS over the TP (70–105°E, 25–40°N) and the SASM
153 region (70–105°E, 10–25°N) by using the global chemical transport model
154 GEOS-Chem driven by the assimilated meteorological fields. These regions of
155 interest are shown in Fig. 1. Simulated surface-layer aerosol concentrations
156 are compared with ground-based observations, and simulated aerosols in the
157 UTLS are evaluated by using the Stratospheric Aerosol and Gas Experiment
158 II (SAGE II) satellite data. Section 2 is a brief description of the GEOS-Chem
159 model and numerical experiment. Section 3 presents the simulation and
160 evaluation of distributions and concentrations of HNO₃ and O₃ to show model's
161 capability in simulating the NO_x-O₃-HNO₃ cycle over the studied regions.
162 Section 4 shows simulated aerosols and Section 5 presents the simulated
163 contribution of nitrate to aerosol concentrations in the UTLS over the TP and
164 the SASM region. Section 6 discusses the mechanisms for high concentrations
165 of nitrate in the UTLS. Section 7 discusses the impacts of uncertainties in
166 surface-layer aerosol concentrations on simulated nitrate in the UTLS.

167

168 **2 Model description and numerical experiment**

169 **2.1 GEOS-Chem model**

170 We simulate gas-phase species and aerosols using the global chemical
171 transport model GEOS-Chem (version 9-01-03, <http://geos-chem.org>) driven
172 by the GEOS-5 assimilated meteorological fields from the Goddard Earth
173 Observing System of the NASA Global Modeling and Assimilation Office. The

174 version of the model used here has a horizontal resolution of 2° latitude by 2.5°
175 longitude and 47 vertical layers extending from the surface to 0.01 hPa. Over
176 the TP and the SASM region, the model has about 34 layers in the troposphere
177 and 12 layers in the stratosphere.

178 The GEOS-Chem model has a fully coupled treatment of tropospheric
179 NO_x -CO-hydrocarbon-aerosol chemistry and aerosols including sulfate (SO_4^{2-}),
180 nitrate (NO_3^-), ammonium (NH_4^+), organic carbon (OC), black carbon (BC) (R. J.
181 Park et al., 2003; 2004; Pye et al., 2009), mineral dust (Fairlie et al., 2007), and
182 sea salt (Alexander et al., 2005; Jaeglé et al., 2011). Anthropogenic aerosols
183 are treated as bulk mass concentrations (particles of SO_4^{2-} , NO_3^- , NH_4^+ , BC,
184 and OC are not size-resolved). Sea Salt mass is simulated for two size bins
185 (0.1–0.5 and 0.5–8 μm) and mineral dust is simulated for four size bins (0.1–
186 1.0, 1.0–1.8, 1.8–3.0, and 3.0–6.0 μm). Both BC and OC consist of hydrophilic
187 and hydrophobic fractions in the model. It is assumed that 80% of BC and 50%
188 of OC emitted from all primary sources are hydrophobic (Cooke et al., 1999;
189 Chin et al., 2002; Chung and Seinfeld, 2002), which become hydrophilic with
190 an e-folding time of 1.2 days following Cooke et al. (1999) and Chin et al.
191 (2002). All secondary OC is assumed to be hydrophilic. Hydrophilic fractions of
192 both BC and OC aerosols are assumed to be fully soluble.

193 The gas-aerosol partitioning of nitric acid and ammonium is calculated
194 using the ISORROPIA II thermodynamic equilibrium module (Fountoukis and
195 Nenes, 2007). In the version of the GEOS-Chem model used in this work, ions
196 considered in ISORROPIA II include $\text{H}^+/\text{Na}^+/\text{NH}_4^+/\text{Cl}^-/\text{SO}_4^{2-}/\text{HSO}_4^-/\text{NO}_3^-/\text{OH}^-$. The
197 two-way coupling between aerosols and gas phase chemistry provides
198 consistent chemical fields for aerosol simulation and aerosol mass for

199 heterogeneous processes and calculations of gas-phase photolysis rates.
200 Heterogeneous reactions include hydrolysis of N_2O_5 (Evans and Jacob, 2005),
201 irreversible absorption of NO_3 and NO_2 on wet aerosols (Jacob, 2000), and the
202 uptake of HO_2 by aerosols (Liao and Seinfeld, 2005; Thornton et al., 2008).
203 Aerosol species are treated as an external mixture in the calculation of aerosol
204 optical properties.

205 With respect to chemistry in the stratosphere, stratospheric O_3
206 concentrations are calculated using the linearized parameterization scheme
207 (McLinden et al., 2000). The monthly mean production rates and loss
208 frequencies of other stratospheric species (including long-lived species such
209 as CFCs and N_2O) use those from NASA Global Modeling Initiative (GMI)
210 Combo simulations (Duncan et al., 2007; Considine et al., 2008; Murray et al.,
211 2012).

212 Convective transport in GEOS-Chem mimics that in the parent GEOS
213 general circulation model (GCM) (Hack, 1994; Zhang and McFarlane, 1995),
214 which accounts for updraft, downdraft, and entrainment mass fluxes for deep
215 and shallow convection (Wu et al., 2007). The aerosol wet deposition scheme
216 in the GEOS-Chem follows that of Liu et al. (2001). For the scavenging of
217 aerosols, SO_4^{2-} , NO_3^- , NH_4^+ , and hydrophilic OC and hydrophilic BC aerosols
218 are assumed to be fully soluble. Dry deposition follows the standard
219 resistance-in-series model of Wesely (1989).

220 Global emissions of aerosols and their precursors in the GEOS-Chem
221 follow R. J. Park et al. (2003, 2004), with anthropogenic emissions of NO_x , CO,
222 SO_2 , and non-methane volatile organic compounds (NMVOC) in Asia
223 overwritten by David Streets' 2006 emission inventory

224 (<http://mic.greenresource.cn/intex-b2006>). Emissions of NH₃ in Asia are taken
225 from Streets et al. (2003). Since NH₃ emissions in China showed large
226 uncertainties in previous studies (Streets et al., 2003; Kim et al., 2006; Y.
227 Zhang et al., 2010; Huang et al., 2011, 2012), we use the most recent estimate
228 of NH₃ emissions in China by Huang et al. (2012), which is 9.8 Tg yr⁻¹, instead
229 of 13.5 Tg yr⁻¹ from Streets et al. (2003). Table 1 summarizes the annual
230 emissions of NO_x, SO₂, NH₃, OC, and BC in Asia domain (60–155°E,
231 10–55°N).

232 Natural NO_x emissions from lightning are calculated using the scheme
233 described by Sauvage et al. (2007) and Murray et al. (2012), and those from
234 soil are simulated following Wang et al. (1998). Natural NH₃ emissions from
235 soil, vegetation, and the oceans are taken from the Global Emissions Inventory
236 Activity inventory (Bouwman et al., 1997). Biomass burning emissions are from
237 the monthly Global Fire Emissions Database (GFED v3) driven by satellite
238 observations of fire activity (van der Werf et al., 2010). Biogenic VOC (volatile
239 organic compounds) emissions are calculated from the Model of Emissions of
240 Gases and Aerosols from Nature (Guenther et al., 2006).

241 The monthly variations of emissions of SO₂ and NO_x follow Wang et al.
242 (2013) and those of BC and OC follow Lou et al. (2014). The monthly scaling
243 factors for NH₃ emissions follow the global inventory described in Fisher et al.
244 (2011). Monthly variations of emissions (anthropogenic plus natural emissions)
245 of NO_x SO₂, NH₃, OC, and BC over Asia are displayed in Fig. 2. The emissions
246 of NH₃ are the highest in June as a result of the agriculture practice and high
247 temperatures (Wang et al., 2013).

248 **2.2 Numerical experiment**

249 To examine the contribution of nitrate to aerosol concentrations in the UTLS
250 over the TP/SASM region, we simulate aerosol concentrations by using the
251 emissions of and meteorological fields of year 2005. Year 2005 is chosen so
252 that we can use the observational datasets for this year from SAGE II and MLS,
253 as described in Sects. 3 and 4. Following Rasch et al. (2008), we perform a
254 10-year spin-up run to generate the initial conditions (to allow the stratospheric
255 species to reach quasi-steady state conditions). We would consider that the
256 tropospheric simulation can be representative of year 2005 but stratosphere
257 simulation should represent a multi-year average, because the production
258 rates and loss frequencies in the stratosphere are the averages over years of
259 2004–2010 ([http://wiki.seas.harvard.edu/geos-chem/index.php/Stratospheric_](http://wiki.seas.harvard.edu/geos-chem/index.php/Stratospheric_chemistry)
260 [chemistry](http://wiki.seas.harvard.edu/geos-chem/index.php/Stratospheric_chemistry)).

261

262 **3 Simulated concentrations of HNO₃ and O₃ and model evaluation**

263 Nitrate aerosol forms when nitric acid (HNO₃) reacts with alkaline gases (for
264 example, ammonia) in the atmosphere (Seinfeld and Pandis, 2006). HNO₃, as
265 the important precursor of nitrate, is the major oxidation product of nitrogen
266 oxides (NO_x = NO+NO₂) (Seinfeld and Pandis, 2006). To show the model's
267 capability in simulating the NO_x-O₃-HNO₃ cycle over the studied regions, we
268 present and evaluate the simulated HNO₃ and O₃ in this section.

269 Simulated mixing ratios of HNO₃ and O₃ in the UTLS are evaluated by
270 using datasets from the limb viewing satellite instrument of Microwave Limb
271 Sounder (MLS, version 3.3, level 2,
272 ftp://acdisc.gsfc.nasa.gov/data/s4pa///Aura_MLS_Level2/). The MLS datasets
273 provide valuable information on atmospheric compositions in the UTLS

274 (Waters et al., 2006). For HNO₃, the MLS provides scientifically useful datasets
275 for 215 to 1.5 hPa, with a vertical resolution of 3–4 km and a horizontal
276 resolution of 400–500 km. Since further evaluations are needed for datasets at
277 altitudes with pressures higher than 215 hPa (Livesey et al., 2011), we use
278 only datasets for pressures lower than that. For O₃, the MLS provides
279 scientifically useful datasets for 261 to 0.02 hPa, with a vertical resolution of
280 2.5–3 km and a horizontal resolution of 300–400 km in the UTLS (Santee et al.,
281 2007; Livesey et al., 2011). The uncertainties of the MLS HNO₃ and O₃
282 datasets in the UTLS are discussed in Livesey et al. (2011). The MLS data
283 HNO₃ product at 100 hPa represents an average of a 3–4 km layer reaching
284 from below the tropopause layer into the lower stratosphere (Duncan et al.,
285 2007). The data screening is conducted strictly according to the instructions of
286 Livesey et al. (2011). For comparison with the MLS observations, the
287 simulated HNO₃ and O₃ mixing ratios are converted to \hat{x} , following the method
288 of Livesey et al. (2011):

$$\hat{x} = x_a + A(x - x_a)$$

289 where A is the averaging kernel matrix of the MLS, x is the modeled vertical
290 profile of HNO₃ or O₃ interpolated at the vertical grid of MLS, and x_a is the
291 HNO₃ or O₃ prior profile of MLS.

292 **3.1 HNO₃**

293 Figure 3(a) shows the simulated global distribution of HNO₃ concentrations
294 averaged over June-August of 2005. Concentrations of HNO₃ exceed 1 ppbv
295 over the industrialized areas such as Europe, North America, central and
296 eastern Asia, and over biomass burning regions in the tropics, in agreement
297 with the distributions and magnitudes reported in Liao et al. (2003). Over South

298 Asia, simulated HNO₃ concentrations are high (0.3–1 ppbv) in the northern
299 Indian subcontinent, because the emissions of NO_x and NH₃ are high in this
300 region (Streets et al., 2003; Zhang et al., 2009; Datta et al., 2012).

301 Figures 4(a)-4(b) show the simulated HNO₃ concentrations in the UTLS
302 averaged over June-August of 2005. Since the tropopause is located at
303 70–150 hPa (12–15 km) over the TP/SASM region (Li et al., 2005; Bian et al.,
304 2011b; Fadnavis et al., 2014), we choose the vertical layers of 200 hPa and
305 100 hPa to represent the UTLS. At both 200 hPa and 100 hPa, the highest
306 HNO₃ concentrations are simulated to occur in the high latitude regions in the
307 Northern Hemisphere (NH) (Fig. 4(a) and Fig. 4(b)). Simulated HNO₃
308 concentrations at 100 hPa are low over the region of 40–100°E and 10–30°N,
309 which is part of the anticyclone region defined in Fig. 1. Figure 4(c) shows the
310 latitude-altitude cross section of simulated seasonal mean HNO₃ mixing ratios
311 averaged over 70–105°E. In boreal summer, the highest HNO₃ mixing ratios
312 are simulated to occur at 30 hPa over the Polar Regions in both hemispheres.
313 Over high latitudes, HNO₃ concentrations in the Southern Hemisphere (SH)
314 are simulated to be higher than those in the NH.

315 To evaluate the simulated HNO₃, Figures 4(d)-4(f) show HNO₃
316 concentrations in the UTLS from MLS that are averaged over June-August of
317 2005. At 200 and 100 hPa altitudes, the observed HNO₃ mixing ratios are high
318 in the high latitudes in the NH, which are captured by the GEOS-Chem model.
319 The observed HNO₃ at 100 hPa exhibits low values of less than 400 pptv over
320 30–100°E and 10–30°N in the Asian monsoon anticyclone region (Fig. 4(e)). At
321 100 hPa, the observed HNO₃ mixing ratio averaged over the TP/SASM region
322 (70–105°E, 10–40°N) is 335.4 pptv, which is lower than the simulated value of

323 372.6 pptv. Considering all the grid cells with MLS HNO₃ data available, the
324 simulated seasonal mean HNO₃ concentrations show normalized mean bias
325 (NMB) of +11.1 % at 100 hPa over the TP/SASM region in summer of year
326 2005. The observed pattern of the HNO₃ vertical distribution (Fig. 4(f)) is also
327 captured by the GEOS-Chem model (Fig. 4(c)). The distributions of HNO₃ in
328 the UTLS are associated with the Brewer-Dobson circulation proposed by
329 Brewer (1949) and Dobson (1956), traveling upwards across the tropopause to
330 the stratosphere at the equator and downwards to the troposphere near the
331 Polar region.

332 **3.2 O₃**

333 Figure 3(b) shows the global distribution of simulated summertime
334 surface-layer O₃ concentrations. Simulated O₃ concentrations are in a range of
335 40–70 ppbv over Europe, North America, China, and the biomass burning
336 region of South Africa. Our model results agree closely with the simulated
337 distributions and magnitudes reported in Mickley et al. (1999), Collins et al.,
338 (2000), Liao et al. (2003), Wu et al., (2008), Zeng et al. (2008), and Fadnavis et
339 al. (2014). Fadnavis et al. (2014) also presented aircraft measurements over
340 India in September of 2010 during the Cloud Aerosol Interaction and
341 Precipitation Enhancement Experiment (CAIPEEX). Our simulated O₃
342 concentrations of 30–40 ppbv over India agree with the CAIPEEX
343 measurements.

344 Figures 5(a)-5(b) show the simulated O₃ concentrations in the UTLS
345 averaged over June-August of 2005. The distributions of O₃ concentrations in
346 the UTLS are similar to those of HNO₃, with elevated values in the high
347 latitudes of the NH. Relatively low O₃ mixing ratios of less than 200 ppbv are

348 simulated at 100 hPa over 10–30°N, 20–110°E, within the anticyclone region
349 defined in Fig. 1. Our simulated distributions and magnitudes of O₃ agree with
350 those reported in Bian et al. (2011b), which examined the summertime
351 distributions of O₃ in the UTLS during 2005–2009 by using the MLS version 2.2
352 level 2 products (Livesey et al., 2008). Because the background O₃
353 concentrations are generally high in the UTLS and the stratosphere, the low O₃
354 concentrations in the UTLS over the TP/SASM region are caused by the deep
355 convection that transports O₃-poor air upward (Fu et al., 2006; Randel and
356 Park, 2006; Park et al., 2007; Bian et al., 2011b). Figure 5(c) displays the
357 latitude-altitude cross section of seasonal mean O₃ mixing ratios averaged
358 over 70–105°E. As a result of the Brewer-Dobson circulation, O₃
359 concentrations in the UTLS are lower over the tropics than in the Polar
360 Regions, even though the maximum O₃ concentrations are located around 10
361 hPa over the tropics (Brewer, 1949). Our simulated O₃ concentrations in the
362 UTLS agree well with the measurements from MLS (Fig. 5(d)-5(f)). At 100 hPa,
363 simulated and MLS observed O₃ mixing ratios averaged over the TP/SASM
364 region (70–105°E, 10–40°N) are 151.7 and 146.6 ppbv, respectively.
365 Compared to MLS observations, simulated O₃ concentrations at 100 hPa have
366 a NMB of +3.5 % over the TP/SASM region in summer of 2005. Our simulated
367 global STE of O₃ is 420 Tg yr⁻¹, which is within the range reported in previous
368 studies (475±120 Tg yr⁻¹ in McLinden et al. (2000), 420 Tg yr⁻¹ in Škerlak et al.
369 (2014), and 556±154 Tg yr⁻¹ in Stevenson et al. (2006)).

370 In addition to the comparisons against MLS products, the simulated O₃
371 profiles are compared with balloon-borne sonde measurements in Fig. 6. The
372 measurements were carried out at Kunming (KM, 102.7°E, 25.0°N) in August

373 of 2009 and 2012, and at Lhasa (LH, 91.1°E, 29.7°N) in August of 2010 and
374 2013. The uncertainties of the observed O₃ mixing ratios were estimated to be
375 within 5–10% (Bian et al. 2012). The comparisons with multi-year observations
376 show that the model can reproduce the vertical distributions of O₃ above 12 km
377 in Kunming and Lhasa. At 100 hPa, the simulated monthly mean O₃ mixing
378 ratio in KM is 112.6 ppbv, and the observed value is 124.2 ppbv in 2009 and
379 113.5 ppbv in 2012. In LH, the simulated monthly O₃ mixing ratio at 100 hPa is
380 152.6 ppbv, and the observed O₃ mixing ratio at that altitude is 142.4 ppbv in
381 2010 and 167.9 ppbv in 2013. The magnitudes of O₃ mixing ratios from these
382 balloon-borne sonde measurements support those from MLS; O₃ mixing ratios
383 in the UTLS are less than 200 ppbv over the TP/SASM region.

384

385 **4 Simulated aerosols and model evaluation**

386 **4.1 Simulated aerosols**

387 Figure 7 (a) shows the simulated surface-layer concentrations of SO₄²⁻, NO₃⁻,
388 NH₄⁺, OC, BC, and PM_{2.5} (the sum of the mass of SO₄²⁻, NO₃⁻, NH₄⁺, BC, and
389 OC aerosols) averaged over June-August of year 2005. As expected,
390 simulated aerosol concentrations are high over polluted regions such as India
391 and eastern China as a result of the high anthropogenic emissions of aerosol
392 precursors and aerosols (Streets et al., 2003; Huang et al., 2012). Over the
393 TP/SASM region (70–105°E, 10–40°N), the average concentrations of SO₄²⁻,
394 NO₃⁻, NH₄⁺, BC, and OC are 1.70, 0.94, 0.85, 0.30, and 0.94 μg m⁻³,
395 respectively. NO₃⁻ is simulated to be of secondary importance at the surface
396 over the region of our interest. The simulated distributions and magnitudes of
397 these aerosol species are similar to those reported in Wang et al. (2013) and

398 Mu and Liao (2014).

399 Figures 7(b) and 7(c) also show the simulated concentrations of SO_4^{2-} ,
400 NO_3^- , NH_4^+ , OC, BC, and $\text{PM}_{2.5}$ in the UTLS. Elevated concentrations of SO_4^{2-} ,
401 NO_3^- , NH_4^+ , OC, BC and $\text{PM}_{2.5}$ are simulated over the TP and Plateau south
402 slope at 200 hPa altitude, and extend from eastern Mediterranean to western
403 China at 100 hPa. The simulated enhanced concentrations of SO_4^{2-} , OC, and
404 BC at 100 hPa over the anticyclone region (20–120°E, 10–40°N) agree with
405 previous observational and modeling studies (Lelieveld et al., 2001; Li et al.,
406 2005; Fadnavis et al., 2013). Li et al. (2005) reported elevated CO
407 concentrations in the upper troposphere over the TP, on the basis of both MLS
408 measurements and the GEOS-Chem simulation for September 2004.
409 Fadnavis et al. (2013) also simulated maximum concentrations of SO_4^{2-} , OC,
410 BC, and mineral dust aerosols in the UTLS during the Asian summer monsoon
411 season owing to convective uplifting of the boundary layer pollutants. With
412 NO_3^- aerosol accounted for in our simulation, NO_3^- is simulated to be the most
413 dominant aerosol species in the UTLS over the TP/SASM region, followed
414 by SO_4^{2-} , NH_4^+ , OC, and BC. At 100 hPa, the averaged concentrations of SO_4^{2-} ,
415 NO_3^- , NH_4^+ , OC, and BC over the TP/SASM region (70–105°E, 10–40°N)
416 region are 0.026, 0.069, 0.014, 0.011, and 0.002 $\mu\text{g m}^{-3}$, respectively.

417 **4.2 Comparisons of simulated aerosol concentrations with in-situ** 418 **observations**

419 The simulated aerosol concentrations in East Asia in the GEOS-Chem model
420 have been evaluated in previous studies (L. Zhang et al., 2010; Fu et al., 2012;
421 Jeong and Park, 2013; Jiang et al., 2013; Wang et al., 2013; Lou et al., 2014).
422 Here we are focused on the evaluation of aerosols in the South Asian

423 monsoon region. For lack of publicly accessible in situ measurements of
424 summertime aerosols in South Asia monsoon area, we compiled monthly or
425 seasonal mean measured concentrations of each aerosol species based on
426 measurements reported in the literature (see Table S1 in the Supplementary
427 Material). These measurements were carried out over years of 1992–2010.
428 The locations of sites with measurements available are shown in Fig. 8(a).
429 Most sites are located in the upwind directions of the TP, with pollutants that
430 can be transported to the UTLS during the South Asian summer monsoon
431 season. The observed PM_{10} concentrations listed in Table S1 are multiplied by
432 0.6 to convert to $PM_{2.5}$ for model evaluation, following the suggestions in
433 Zhang et al. (2002) and Chatterjee et al. (2010).

434 Figures 8(b)–8(f) show the scatterplots of simulated versus observed
435 seasonal mean aerosol concentrations. Compared with measurements,
436 simulated SO_4^{2-} , NO_3^- , NH_4^+ , OC and BC have NMBs of -17.0% , $+38.8\%$,
437 $+42.0\%$, -69.7% and -41.0% , respectively, as the concentrations of all
438 seasons are considered. The correlations between model results and
439 observations have R values of 0.49–0.85 for all aerosol species, indicating that
440 the model is capable of capturing the spatial distributions and seasonal
441 variations of each aerosol species in the South Asian monsoon region despite
442 the biases in concentrations. If we consider simulated and measured
443 concentrations for JJA alone, the simulated concentrations of SO_4^{2-} , NO_3^- ,
444 NH_4^+ , OC and BC exhibit seasonal NMBs of -14.7% , $+51.5\%$, $+74.9\%$, -57.2%
445 and -32.2% , respectively, and the values of R are in the range of 0.24–0.85.
446 Note that the measurements of NO_3^- and NH_4^+ are quite limited in terms of the
447 number of samples, and the discrepancies between model results and

448 measurements may also arise from the mismatch of the model year 2005 with
449 the years of 1992–2010 with observations available.

450 **4.3 Comparisons of simulated aerosol extinction coefficients with SAGE**

451 **II datasets**

452 Satellite datasets from the Stratospheric Aerosol and Gas Experiment II
453 (SAGE II, https://eosweb.larc.nasa.gov/project/sage2/sage2_v620_table) are
454 used to evaluate the simulated aerosol extinction in the UTLS. The SAGE II
455 instrument was launched in October 1984 aboard the Earth Radiation Budget
456 Satellite (ERBS) and terminated on 8 September 2005 (McCormick et al. 1987;
457 Chu et al. 1989). The datasets used here are aerosol extinction coefficients at
458 525 nm from the version 6.20 SAGE retrievals, covering from 0.5 to 40 km with
459 a vertical resolution of 0.5 km. Many validation studies have been conducted
460 on the SAGE II aerosol data (Russell and McCormick, 1989; Oberbeck et al.,
461 1989; Wang et al., 1989), which indicated that extinction coefficients have
462 uncertainties of 20–30%. The extinction coefficients of aerosols in the
463 GEOS-Chem model are calculated using aerosol mass concentration,
464 extinction efficiency, effective radius, particle mass density, and the assumed
465 aerosol size distribution (Drury et al., 2010). The hygroscopic growth of each
466 aerosol species with relative humidity is accounted for, using the hygroscopic
467 growth factors listed in Martin et al. (2003).

468 Figure 9(a) presents the simulated monthly mean distribution of aerosol
469 extinction coefficients at 100 hPa for July of 2005. At 100 hPa, the simulated
470 aerosol extinction coefficients are relatively high over the anticyclone region,
471 where anthropogenic aerosol species (Fig. 7) and natural aerosols such as
472 mineral dust and sea salt contribute to aerosol extinction coefficients in

473 summer. Note that the contributions of sulfate, nitrate, ammonium, OC, sea
474 salt, and mineral dust are all considered when we calculate aerosol extinction
475 coefficients. Aerosol extinction coefficients are simulated to be $1.2\text{--}2\times 10^{-3}$
476 km^{-1} at 100 hPa over the Asian continent and Indian Ocean ($20^{\circ}\text{S}\text{--}30^{\circ}\text{N}$,
477 $30^{\circ}\text{--}105^{\circ}\text{E}$). These values agree closely with aerosol extinction coefficients
478 measured at Naqu during August of 2011 for the same altitude, the maximum
479 of which was $2.4\times 10^{-3} \text{ km}^{-1}$ (He et al., 2014). Vernier et al. (2011) also
480 identified this Asian aerosol layer with high SR at 100 hPa by observations of
481 CALIPSO for JJA of 2006–2008.

482 Figure 9(b) displays the monthly mean vertical profiles of aerosol extinction
483 coefficients averaged over the Asian monsoon anticyclone region ($20\text{--}120^{\circ}\text{E}$
484 $10\text{--}40^{\circ}\text{N}$) (Fig. 1) for July of 2005. The SAGE II datasets are available for July
485 only in 2005. The profiles from SAGE II and the GEOS-Chem simulation are all
486 shown. The vertical distributions of aerosol extinction coefficients “with nitrate”
487 and “without nitrate” are both from the baseline run with full chemistry. The
488 vertical distribution of aerosol extinction coefficient “with nitrate” (or “without
489 nitrate”) indicates that the contribution of nitrate aerosol to aerosol extinction is
490 (or is not) accounted for. Accounting for all aerosol species, the GEOS-Chem
491 model reproduces well the aerosol extinction coefficients above 10 km, but the
492 discrepancies are rather large in altitudes less than 10 km. Note that the
493 uncertainties in satellite datasets increase as the altitude decreases
494 (Vanhellemont et al., 2008; Kulkarni and Ramachandran, 2015), and the
495 missing data in the lower troposphere along the satellite trajectories over the
496 region of our interest also contribute to the discrepancies

497 Comparisons of profiles of aerosol extinction coefficients with and without

498 nitrate aerosol indicate that the profiles show small differences in altitudes less
499 than 6 km but large discrepancies from 6 km to the tropopause. With nitrate
500 aerosol accounted for, the simulated aerosol extinction coefficients agree
501 closely with SAGE II datasets in the UTLS (averaged over 14–16 km, the
502 simulated value is $8.6 \times 10^{-4} \text{ km}^{-1}$ while the observed value is $8.0 \times 10^{-4} \text{ km}^{-1}$).
503 Without nitrate aerosol, the simulated aerosol extinction coefficient at 14–16
504 km altitude is $1.5 \times 10^{-4} \text{ km}^{-1}$, which underestimates the aerosol extinction
505 coefficient by 82.6% compared to that calculated with all the aerosol species.
506 These comparisons of extinction coefficients with and without nitrate aerosol
507 suggest that nitrate aerosol plays an important role in aerosol extinction in the
508 UTLS over the region of our interest.

509

510 **5 Contribution of nitrate to aerosol concentrations in the UTLS**

511 Since nitrate aerosol is simulated to be the most abundant aerosol species in
512 the UTLS over the TP/SASM region, we analyze the contribution of nitrate to
513 $\text{PM}_{2.5}$ concentration ($C_{\text{NIT}} = \text{nitrate concentration} / \text{PM}_{2.5} \text{ concentration}$) in this
514 section. Figure 10 shows the simulated seasonal mean distributions of C_{NIT} for
515 June-August of year 2005. At the surface layer (Fig. 10(a)), simulated high C_{NIT}
516 values are located over the areas with high nitrate concentrations (India and
517 eastern China) as well as the oceans where NO_3^- also forms on sea salt and
518 mineral dust particles (Arimoto et al., 1996; Nakamura et al., 2005; George and
519 Nair, 2008). Over the TP/SASM region, the C_{NIT} values in JJA are 5–35% at the
520 surface, 25–50% at 200 hPa (Fig. 10(b)), and could exceed 60% at 100 hPa
521 (Fig. 10(c)). The latitude-altitude cross section of C_{NIT} (Fig. 10(d)) shows that
522 C_{NIT} over 20–40°N increases with altitude and reaches maximum values

523 around the extratropical tropopause.

524 Table 2 lists the mean concentrations of SO_4^{2-} , NO_3^- , NH_4^+ , BC and OC, and
525 their contributions to $\text{PM}_{2.5}$ during summertime of 2005 over the TP/SASM, TP,
526 and SASM regions. Over the TP/SASM region, SO_4^{2-} , NO_3^- , NH_4^+ , BC and OC
527 are simulated to contribute 35.9%, 19.8%, 18.1%, 6.4%, and 19.8%,
528 respectively, to $\text{PM}_{2.5}$ mass concentration at the surface layer. The
529 contributions increase significantly in the UTLS. The largest C_{NIT} is simulated
530 in the SASM region at 100 hPa, where NO_3^- accounts for 60.5% of $\text{PM}_{2.5}$ mass
531 concentration. The high C_{NIT} values indicate that NO_3^- plays an important role
532 in the aerosol layer in the UTLS over the TP/SASM region.

533 Considering the large uncertainties in simulated sea salt (Jaeglé et al.,
534 2011) and mineral dust (Fairlie et al., 2007) aerosols, we tend to be focused on
535 anthropogenic aerosol species (SO_4^{2-} , NO_3^- , NH_4^+ , BC, and OC) in this work. In
536 our model, concentrations of sea salt (or mineral dust) are simulated to be
537 $1.0\text{--}1.7 \text{ ng m}^{-3}$ (or $5.0\text{--}7.0 \text{ ng m}^{-3}$) over the studied region in the summer of
538 2005, which contribute less than 1.2% (or 5.0%) to total aerosol mass at 100
539 hPa. Therefore the consideration of sea salt and mineral dust can slightly
540 reduce C_{NIT} values, but C_{NIT} values at 100 hPa are still as high as 45-65% over
541 the TP/SASM region in summer.

542

543 **6 Mechanisms for high nitrate concentrations in the UTLS**

544 **6.1 Upward transport of nitrate from the lower troposphere**

545 The intense convective transport of chemical species into the UTLS over the
546 TP/SASM region during summertime has been widely discussed in previous
547 studies (Randel et al., 2010; Bian et al., 2011a; Fadnavis et al., 2013, 2014;

548 Qie et al., 2014; He et al., 2014), evidenced by both the satellite observations
549 (Fu et al., 2006; Luo et al., 2011) and the calculation of the outgoing long-wave
550 radiation which is a convective proxy over the region (Randel and Park, 2006;
551 Park, et al., 2007; Fadnavis et al., 2013). Since nitrate aerosol is simulated to
552 be of secondary abundant aerosol species in the surface layer over the
553 TP/SASM region (Fig. 7), the vertical mass transport through the deep
554 convection in this region contributes to the accumulation of NO_3^- in the UTLS.
555 Figure 11 shows the latitude-altitude cross sections of simulated
556 concentrations of SO_4^{2-} and NO_3^- averaged over 70–105°E in June-August of
557 2005, together with the wind vectors obtained from the European Centre for
558 Medium-Range Weather Forecasts (ECMWF) ERA-Interim Reanalysis data.
559 Note that the assimilated GEOS-5 meteorological fields do not have vertical
560 winds
561 (http://wiki.seas.harvard.edu/geos-chem/index.php/List_of_GEOS-5_met_fields
562 s), so we use the ECMWF reanalysis wind fields to do the analysis here. High
563 values of aerosol concentrations are found on the south slope of the
564 Himalayas, where the deep convection exists. Although both SO_4^{2-} and NO_3^-
565 are transported upward to the extratropical tropopause, the details of the
566 vertical distributions are different. At altitudes higher than 8 km, the
567 concentrations of NO_3^- do not decrease with altitude as quickly as those of
568 SO_4^{2-} , and the concentrations of NO_3^- over 10–40°N are higher than those of
569 SO_4^{2-} .

570 The chemical mechanisms for the formation of SO_4^{2-} , NO_3^- , and NH_4^+
571 aerosols in the GEOS-Chem model were described in R. J. Park et al. (2004),
572 which are comprehensive and have been used extensively in previous studies

573 to simulate these three aerosol species (R. J. Park et al., 2004; Pye et al.,
574 2009; L. Zhang et al., 2010; Zhu et al., 2012; Jiang et al., 2013; Lou et al.,
575 2014). Sulfate aerosol forms from gas-phase oxidation of SO₂ by OH and from
576 in-cloud oxidation of SO₂ by O₃ and H₂O₂. Nitrate forms from the partitioning of
577 HNO₃ between gas and aerosol phases, which is calculated by the
578 ISORROPIA II thermodynamic equilibrium module (Fountoukis and Nenes,
579 2007) in the GEOS-Chem model. HNO₃ is produced by the reaction of NO₂
580 with OH during daytime and by hydrolysis of N₂O₅ on aerosol surfaces at night
581 (Table 3). The chemical mechanisms for SO₄²⁻ and NO₃⁻ have different
582 sensitivity to meteorological conditions. During the vertical transport,
583 temperature decreases, which reduces the gas-phase oxidation of SO₂ (Yao et
584 al., 2002; Seinfeld and Pandis 2006) but promotes the formation of NO₃⁻ by
585 shifting gas-particle equilibria (Dawson et al., 2007; Liao et al., 2009). Dawson
586 et al. (2007) examined the sensitivities of sulfate and nitrate concentrations to
587 temperature by using the Particulate Matter Comprehensive Air Quality Model
588 with extensions (PMCAMx). The sensitivity test was performed by fixing all
589 meteorological parameters but perturbing temperature. Their sensitivity
590 simulations showed that the increases in temperature led to increases in
591 sulfate concentrations and decreases in nitrate concentrations. Compared to
592 nitrate, sulfate concentrations showed smaller sensitivity to temperature
593 changes (Dawson et al., 2007); as temperature increased, nitrate
594 concentrations decreased by 19% K⁻¹ and 17% K⁻¹ in January and July
595 respectively, while sulfate concentration increased by 0.12% K⁻¹ and 1.3% K⁻¹
596 in January and July, respectively. Therefore the different chemical mechanisms
597 for SO₄²⁻ and NO₃⁻ formation contribute to the differences in their vertical

598 distributions.

599 **6.2 Net chemical production of HNO₃ during the vertical transport**

600 As mentioned above, the formation of gas-phase HNO₃ and the
601 partitioning of HNO₃ between gas and aerosol phases are the two major
602 chemical processes that influence NO₃⁻ concentrations. The ability of the
603 GEOS-Chem model to simulate gas-phase HNO₃ has been evaluated in
604 Section 3.1 (by comparisons of our model results with MLS observations and
605 concentrations from previous modeling studies). Major reactions for the
606 production and loss of HNO₃ are listed in Table 3. Figure 12 shows the net
607 chemical production of HNO₃ by gas-phase reactions and heterogeneous
608 reactions (chemical production by reactions R1-R23 minus chemical loss by
609 reactions R24-R25 in Table 3) summed over the TP/SASM region. The net
610 chemical production has an overall trend of decreasing with altitude. Since
611 NO_x emissions from aircraft and lightning are located between 200–300 hPa
612 over the TP/SASM region during summer (Martin et al., 2007; Murray et al.,
613 2012; Pitari et al., 2015), net chemical production of HNO₃ shows a small peak
614 at those altitudes. The mixing ratio of HNO₃ decreases with altitude between
615 500 and 200 hPa and increases with altitude above 200 hPa. At 100 hPa, the
616 average HNO₃ mixing ratio over the TP/SASM region agrees with the values of
617 300–400 pptv over the same region shown in Fig. 4.

618 Figure 12 also shows the transport of HNO₃ over the TP/SASM region,
619 defined as convergence of horizontal flux of HNO₃ (inflow minus outflow) plus
620 convergence of vertical flux of HNO₃ (inflow minus outflow) for a specific
621 vertical model layer. At altitudes below 200 hPa, concentrations of HNO₃ over
622 the TP/SASM region are dependent on the net chemical production by

623 gas-phase reactions and heterogeneous reactions as well as the transport of
624 HNO_3 . The vertical variation of the horizontal transport (the north-south and the
625 east-west transport) follows the development of the anticyclone in the upper
626 troposphere during summertime. As the altitude increases, the westerlies
627 enhance over the TP and the easterlies develop over the SASM region (see
628 Fig. 1 for our definitions of TP and SASM regions). At latitudes between 100–
629 500 hPa, vertical transport of HNO_3 occurs over the TP due to the deep
630 convection activities over the region which can arrive at 100–200 hPa (Fig. 11;
631 Fadnavis et al., 2013; Qie et al., 2014).

632 **6.3 The gas-to-aerosol conversion of HNO_3 to form nitrate during the** 633 **vertical transport**

634 NO_3^- formation from gas-to-aerosol conversion of HNO_3 is calculated by using
635 the ISORROPIA II thermodynamic equilibrium module (Fountoukis and Nenes,
636 2007). As shown in Fig. 12, the nitrate formation from gas to aerosol
637 conversion of HNO_3 peaks between 100–300 hPa, indicating that the
638 gas-aerosol partitioning plays an important role in the enhancement of nitrate
639 in the UTLS.

640 The gas-to-aerosol conversion of HNO_3 to form NO_3^- is very sensitive to
641 relative humidity (RH) and temperature (Fountoukis and Nenes 2007; Dawson
642 et al., 2007). Low temperature and high RH are favorable for NO_3^- formation.
643 Figure 13 shows the seasonal mean horizontal distributions of RH and
644 temperature at 100 hPa and the latitude-altitude cross sections of these two
645 parameters averaged over 70–105°E. RH exhibits high values in the TP/SASM
646 region, which are consistent with the high H_2O mixing ratios in this area
647 reported in Gettelman et al. (2004), M. Park et al. (2004), and Fu et al. (2006).

648 At 100 hPa, the locations with high RH of exceeding 45% correspond well with
649 those with high C_{NIT} values (Fig. 10(c)). The latitude-altitude cross section of
650 RH (Fig. 13(c)) shows that RH has high values over the places with intense
651 upward transport (Fig. 11). For temperature, as Fig. 13(b) and 13(d) show,
652 summertime temperatures are cold (190–200 K) at 100 hPa in the TP/SASM
653 region, consistent with the distribution and magnitude reported for August,
654 2011, in He et al. (2014) on the basis of the NCEP Reanalysis data. The low
655 temperatures over the TP/SASM region are associated with the adiabatic
656 expansion of ascending air mass of the deep convections (Yanai et al., 1992;
657 Park et al., 2007; He et al., 2014).

658 Because of the favorable conditions of RH and decreasing temperature,
659 the gas-to-aerosol conversion of HNO_3 to form nitrate can occur during the
660 upward transport and in the UTLS. Figure 14 shows the mass budget for
661 nitrate aerosol within the selected box of (70–105°E, 10–40°N, 8–16 km) to
662 see the role of nitrate formation over the TP/SASM region. The horizontal
663 mass fluxes have a net negative value of $0.10 \text{ Tg season}^{-1}$, reducing nitrate
664 aerosol in the selected box. The vertical transport and the gas-to-aerosol
665 conversion of HNO_3 increase nitrate mass in the selected box, with values of
666 $0.09 \text{ Tg season}^{-1}$ and $0.11 \text{ Tg season}^{-1}$, respectively, indicating that the
667 gas-to-aerosol conversion plays an important role in the enhancement of
668 nitrate in the UTLS over the TP/SASM region. Although relatively high RH
669 exists near the tropopause of the TP/SASM region, the air near the tropopause
670 is still dryer compared to that in the lower altitudes. Model results show that the
671 gas-to-aerosol partition of HNO_3 decreases with altitude between 100–300
672 hPa, indicating that the gas to aerosol conversion contributes to nitrate

673 accumulation in the UTLS mainly during the process of upward transport.

674 Previous studies have also reported that nitric acid trihydrates (NAT,
675 $\text{HNO}_3 \cdot (\text{H}_2\text{O})_3$) could form in the polar and tropical stratosphere at low
676 temperatures through two mechanisms: (1) the homogeneous nucleation out
677 of supercooled ternary solutions, and (2) the heterogeneous formation on ice
678 particles (Hofmann et al., 1989; Carslaw et al., 1998; Voigt et al., 2000; Popp et
679 al., 2006; Kirner et al., 2011). A typical NAT condensation temperature is
680 approximate 193 K (Kirner et al., 2011). As shown in Fig. 13, the temperatures
681 around 100 hPa over the TP/SASM region are in the range of 190–200 K,
682 which are low enough to produce some NAT particles. However, balloon-borne
683 measurements of depolarization ratio and backscattering ratio of aerosols at
684 Lhasa during August-October of 1999 by Kim et al. (2003) and Tobo et al.
685 (2007) suggested that coarse and aspherical particles such as NAT are scarce
686 in the UTLS of the TP/SASM.

687

688 **7 Sensitivities of simulated nitrate in the UTLS to anthropogenic NO_x , 689 NH_3 , and SO_2 emissions in Asia**

690 Since simulated SO_4^{2-} , NO_3^- and NH_4^+ concentrations have, respectively,
691 NMBs of -17.0% , $+38.8\%$, and $+42.0\%$ on an annual mean basis and of
692 -14.7% , $+51.5\%$, and $+74.9\%$ in summer (Section 4.2), we perform four
693 sensitivity simulations to examine the impacts of uncertainties in surface-layer
694 aerosol concentrations on simulated nitrate in the UTLS. In the first three
695 cases, anthropogenic emissions of NO_x , NH_3 , and SO_2 in Asia are changed by
696 -50% , -50% , and $+20\%$, respectively, relative to those in our standard
697 simulation. In the last case, anthropogenic emissions of all these three species

698 are changed simultaneously, with NO_x reduced by 50%, NH_3 reduced by 50%,
699 and SO_2 increased by 20% in Asia relative to the standard case. The purpose
700 of these sensitivity studies is to reduce NMBs of simulated surface-layer
701 concentrations of SO_4^{2-} , NO_3^- and NH_4^+ and see whether NO_3^- is still the
702 most dominant aerosol species in the UTLS. Model results from these
703 sensitivity studies for summer of 2005 are presented in Table 4.

704 As anthropogenic emissions of SO_2 in Asia are increased by 20%, the
705 NMB of simulated surface-layer SO_4^{2-} concentrations is -4.4% , which is an
706 improvement compared to the NMB of -14.7% in the standard simulation.
707 However, the increases in SO_2 emissions lead to larger NMBs of surface-layer
708 NO_3^- and NH_4^+ because of the increased formation of ammonium sulfate or
709 ammonium bisulfate. The percentage contributions of SO_4^{2-} to total aerosol
710 mass in the UTLS increase slightly by 2.7% at 200 hPa and by 1.6% at 100
711 hPa, and nitrate in the UTLS also shows small sensitivity to the change in SO_2
712 emissions.

713 With anthropogenic emissions of NO_x in Asia reduced by 50%, the NMB of
714 simulated surface-layer NO_3^- concentrations changes from $+51.5\%$ in the
715 standard simulation to -11.7% in this sensitivity run. The contribution of each
716 of SO_4^{2-} , NO_3^- and NH_4^+ aerosols to total aerosol mass in the UTLS is not
717 sensitive to this reduction in NO_x emissions at the surface; the percentage
718 contribution obtained from this sensitivity run is very close to the value
719 obtained in the standard simulation (Table 4). Similarly, in the sensitivity study
720 with NH_3 emissions reduced by 50% in Asia, simulated surface-layer
721 concentrations of NO_3^- and NH_4^+ are improved in terms of the values of NMBs,
722 but the improvement in simulated aerosol concentrations at the surface-layer

723 does not influence our conclusion of high nitrate aerosol concentration in the
724 UTLS.

725 As shown in Table 4, for the surface layer, simulated nitrate concentration
726 over the TP/SASM region decreases by 46.8% (from $0.94 \mu\text{g m}^{-3}$ to $0.50 \mu\text{g m}^{-3}$)
727 with a 50% reduction in anthropogenic NO_x emissions in Asia, and it decreases
728 by 22.3% (from $0.94 \mu\text{g m}^{-3}$ to $0.73 \mu\text{g m}^{-3}$) when anthropogenic NH_3 emissions
729 are reduced by the same percentage, indicating that surface-layer nitrate
730 aerosol is more sensitive to anthropogenic emissions of NO_x than to those of
731 NH_3 . Relative to the baseline simulation, simulated nitrate concentrations at
732 200 hPa and 100 hPa decrease, respectively, by 49.0% (from $7.57 \times 10^{-2} \mu\text{g m}^{-3}$
733 to $3.86 \times 10^{-2} \mu\text{g m}^{-3}$) and 17.7% (from $6.90 \times 10^{-2} \mu\text{g m}^{-3}$ to $5.68 \times 10^{-2} \mu\text{g m}^{-3}$)
734 with a 50% reduction in NH_3 emissions, whereas only by 2.1% (from 7.57×10^{-2}
735 $\mu\text{g m}^{-3}$ to $7.41 \times 10^{-2} \mu\text{g m}^{-3}$) and 1.3% (from $6.90 \times 10^{-2} \mu\text{g m}^{-3}$ to $6.81 \times 10^{-2} \mu\text{g}$
736 m^{-3}) with a 50% reduction in NO_x emissions. Over the studied region, the role
737 of NH_3 in the sulfate-nitrate-ammonium aerosol system can be quantified by
738 the gas ratio of $\text{GR} = \frac{\text{free ammonia}}{\text{total nitrate}} = \frac{\text{TA} - 2 \times \text{TS}}{\text{TN}}$ (Ansari and Pandis, 1998), where
739 $\text{TA} = \text{NH}_3 + \text{NH}_4^+$, $\text{TS} = \text{SO}_4^{2-}$, and $\text{TN} = \text{HNO}_3 + \text{NO}_3^-$. Over the TP/SASM
740 region, GR is generally positive both at the surface and in the UTLS, especially
741 over $20\text{--}40^\circ\text{N}$ where deep convection exits (Fig. 11), indicating that free
742 ammonia is available to react with nitrate (Seinfeld and Pandis 2006). However,
743 GR is generally less than 1.0 above 400 hPa in summer over the TP/SASM
744 region, which indicates nitrate concentrations are most sensitive to changes in
745 NH_3 and explains the small sensitivity of nitrate aerosol to NO_x emissions in
746 the UTLS.

747 In the sensitivity study with emissions of NO_x , NH_3 , and SO_2 in Asia

748 changed simultaneously, simulated surface-layer concentrations of SO_4^{2-} , NO_3^-
749 and NH_4^+ have NMBs of -8.3% , -27.0% and $+55.4\%$, respectively, which are
750 all improved compared to those in the standard simulation. Even though nitrate
751 aerosol is now underestimated at the surface, it still accounts for 53.3% of the
752 $\text{PM}_{2.5}$ concentration at 100 hPa over the TP/SASM region in summer.

753 It should be noted that the concentrations of OC and BC are also
754 underestimated, with NMBs of -57.2% and -32.2% , respectively, in summer
755 (Section 4.2). We have done a simple calculation with the concentrations of
756 OC and BC in the UTLS multiplied by 2.3 and 1.5, respectively, and nitrate is
757 still the most dominant aerosol species in summertime in the UTLS over the
758 TP/SASM region (not shown in Table 4). Therefore the uncertainties in surface
759 aerosol concentrations do not compromise the conclusion of this study.

760

761 **8 Conclusions**

762 In this work we simulate nitrate aerosol and its contribution to aerosol
763 concentrations in the UTLS over the TP/SASM region ($70\text{--}105^\circ\text{E}$, $10\text{--}40^\circ\text{N}$)
764 for summertime of year 2005, using the global chemical transport model
765 GEOS-Chem driven by the assimilated meteorological fields.

766 Simulated HNO_3 and O_3 are evaluated to show the model's ability to
767 simulate the $\text{NO}_x\text{-O}_3\text{-HNO}_3$ cycle over the studied region. In the UTLS, both
768 the horizontal and vertical distributions of simulated HNO_3 and O_3 agree well
769 with the MLS observations. At 100 hPa, simulated seasonal mean HNO_3 and
770 O_3 mixing ratios show NMBs of $+11.1\%$ and $+3.5\%$, respectively, over the
771 TP/SASM region ($70\text{--}105^\circ\text{E}$, $10\text{--}40^\circ\text{N}$) in summer of year 2005, and the model
772 biases lie within the confidence range of the MLS instruments. Both simulated

773 and observed O₃ concentrations show relatively low values of less than 200
774 ppbv at 100 hPa over the TP/SASM region.

775 Averaged over the TP/SASM region, the surface-layer concentrations of
776 SO₄²⁻, NO₃⁻, NH₄⁺, BC, and OC are simulated to be 1.70, 0.94, 0.85, 0.30, and
777 0.94 μg m⁻³, respectively. Nitrate aerosol is simulated to be of secondary
778 importance near the surface over the region of our interest. Comparisons of
779 simulated aerosol concentrations with ground-based observations show that
780 simulated summertime concentrations of SO₄²⁻, NO₃⁻, NH₄⁺, OC and BC have
781 NMB of -14.7%, +51.5%, +74.9%, -57.2% and -32.2%, respectively. Note that
782 the measurements of NO₃⁻ and NH₄⁺ are quite limited in terms of the number
783 of samples.

784 Model results show elevated concentrations of SO₄²⁻, NO₃⁻, NH₄⁺, OC, BC
785 and PM_{2.5} in the UTLS over the TP/SASM region throughout the summer. NO₃⁻
786 is simulated to be the most dominant aerosol species in the UTLS of the
787 TP/SASM region. Accounting for NO₃⁻ aerosol, the GEOS-Chem model
788 reproduces well the magnitude of aerosol extinctions above 10 km, as model
789 results are compared with the SAGE II measurements. The discrepancies
790 between the simulated and observed aerosol extinction coefficient are within 8%
791 in the UTLS (averaged over 14–16 km). Simulated vertical profiles of aerosol
792 extinction coefficients with and without nitrate aerosol show large
793 discrepancies from 6 km to tropopause, indicating the important role of nitrate
794 in aerosol layer in the UTLS over the TP/SASM region.

795 The contribution of NO₃⁻ to aerosols in the TP/SASM region is quantified
796 by C_{NIT} (the ratio of nitrate concentration to PM_{2.5} concentration). Over the
797 TP/SASM region, the C_{NIT} values in summer are 5–35% at the surface, 25–

798 50% at 200 hPa, and could exceed 60% at 100 hPa. The mechanisms for the
799 accumulation of nitrate in the UTLS over the TP/SASM region include vertical
800 transport and the gas-to-aerosol conversion of HNO_3 to form nitrate. Such
801 gas-to-aerosol conversion occurs during the upward transport and in the UTLS.
802 The high relative humidity and low temperature associated with the deep
803 convection over the TP/SASM region are favorable for nitrate formation.

804 Results from the present study indicate that nitrate is an important aerosol
805 species in the UTLS over the TP/SASM region. Considering the scarce
806 measurements of nitrate in the UTLS and the model uncertainties, more
807 observational and modeling studies are needed to further explore the aerosol
808 composition in the Asian tropopause aerosol layer. Further simulations of
809 nitrate aerosol in the UTLS also need to account for NAT formation at low
810 temperatures (Kirner et al., 2011) and the roles of natural aerosols, including
811 the transport of mineral dust and sea salt to the UTLS as well as nitrate
812 formation on these natural particles (Ma et al., 2003).

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821

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Table 1. Summary of Annual Emissions of Aerosols and Aerosol Precursors in Asia (60°E-155°E, 10°N-55°N)

| Species | Global | Asia |
|--|--------|-------|
| NO_x (Tg N yr⁻¹) | | |
| Aircraft | 0.5 | 0.08 |
| Anthropogenic | 28.6 | 9.96 |
| Biomass burning | 4.7 | 0.27 |
| Fertilizer | 0.7 | 0.31 |
| Lightning | 5.9 | 0.87 |
| Soil | 5.9 | 0.96 |
| Total | 46.3 | 12.45 |
| SO₂ (Tg S yr⁻¹) | | |
| Aircraft | 0.1 | 0.01 |
| Anthropogenic | 52.6 | 23.46 |
| Biomass burning | 1.2 | 0.07 |
| Volcanoes | 4.4 | 1.04 |
| No_eruption | 8.9 | 1.78 |
| Ship | 7.4 | 0.94 |
| Total | 74.6 | 27.30 |
| NH₃ (Tg N yr⁻¹) | | |
| Anthropogenic | 34.9 | 17.83 |
| Natural | 14.2 | 2.01 |
| Biomass burning | 3.5 | 0.21 |
| Biofuel | 1.6 | 0.71 |
| Total | 54.2 | 20.76 |
| OC (Tg C yr⁻¹) | | |
| Anthropogenic | 3.1 | 1.42 |
| Biomass burning | 18.7 | 1.10 |
| Biofuel | 6.3 | 3.28 |
| Biogenic | 9.7 | 1.22 |
| Total | 37.8 | 7.02 |
| BC (Tg C yr⁻¹) | | |
| Anthropogenic | 3.0 | 1.43 |
| Biomass burning | 2.2 | 0.12 |
| Biofuel | 1.6 | 0.86 |
| Total | 6.8 | 2.41 |

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Table 2. Simulated seasonal mean concentrations of aerosols and their contributions to PM_{2.5} (in percentages in parentheses) during summertime (June-August) of 2005 for the TP/SASM, TP, and SASM regions. The unit is $\mu\text{g m}^{-3}$ for concentrations at the surface, and $10^{-2} \mu\text{g m}^{-3}$ for concentrations at 200 hPa and 100 hPa.

| | PM _{2.5} | SO ₄ ²⁻ | NO ₃ ⁻ | NH ₄ ⁺ | OC | BC |
|---------|-------------------|-------------------------------|------------------------------|------------------------------|-------------|------------|
| TP/SASM | | | | | | |
| Surface | 4.73 | 1.70(35.9%) | 0.94(19.8%) | 0.85(18.1%) | 0.94(19.8%) | 0.30(6.4%) |
| 200 hPa | 16.19 | 3.27(20.2%) | 7.57(46.8%) | 2.67(16.5%) | 2.22(13.7%) | 0.44(2.7%) |
| 100 hPa | 12.14 | 2.60(21.4%) | 6.90(56.8%) | 1.43(11.8%) | 1.05(8.6%) | 0.16(1.3%) |
| TP | | | | | | |
| Surface | 5.44 | 2.12(39.0%) | 1.05(19.3%) | 1.08(19.9%) | 0.88(16.1%) | 0.31(5.7%) |
| 200 hPa | 19.80 | 4.16(21.0%) | 9.43(47.6%) | 3.25(16.4%) | 2.49(12.6%) | 0.47(2.4%) |
| 100 hPa | 10.58 | 2.60(24.6%) | 5.51(52.0%) | 1.35(12.7%) | 0.99(9.4%) | 0.14(1.3%) |
| SASM | | | | | | |
| Surface | 4.02 | 1.28(31.8%) | 0.83(20.5%) | 0.63(15.6%) | 1.00(24.8%) | 0.29(7.2%) |
| 200 hPa | 12.57 | 2.38(18.9%) | 5.72(45.5%) | 2.10(16.7%) | 1.95(15.5%) | 0.41(3.3%) |
| 100 hPa | 13.71 | 2.60(19.0%) | 8.30(60.5%) | 1.52(11.1%) | 1.11(8.1%) | 0.18(1.3%) |

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Table 3. List of gas-phase reactions and heterogeneous reactions involve HNO₃ in the GEOS-Chem model (Version 9-01-03)

| | Reactants | | Products |
|----------------------|---|---|------------------------------------|
| Chemical productions | | | |
| R1 | NO ₂ + OH + M | → | HNO ₃ + M |
| R2 | NO ₃ + CH ₂ O | → | HNO ₃ + prod. |
| R3 | ALD ₂ + NO ₃ | → | HNO ₃ + prod. |
| R4 | RIO ₁ + NO | → | HNO ₃ + prod. |
| R5 | IAO ₂ + NO | → | 0.08HNO ₃ + prod. |
| R6 | ISN ₁ + NO | → | 0.05HNO ₃ + prod. |
| R7 | VRO ₂ + NO | → | HNO ₃ + prod. |
| R8 | MRO ₂ + NO | → | HNO ₃ + prod. |
| R9 | INO ₂ +NO | → | 0.85HNO ₃ + prod. |
| R10 | ALK ₄ + NO ₃ | → | HNO ₃ + prod. |
| R11 | RCHO + NO ₃ | → | HNO ₃ + prod. |
| R12 | MEK + NO ₃ | → | HNO ₃ + prod. |
| R13 | INO ₂ + MO ₂ | → | 0.425HNO ₃ + prod. |
| R14 | GLYX + NO ₃ | → | HNO ₃ + prod. |
| R15 | MGLY + NO ₃ | → | HNO ₃ + prod. |
| R16 | MACR + NO ₃ | → | HNO ₃ + prod. |
| R17 | C ₂ H ₆ + NO ₃ | → | HNO ₃ + prod |
| R18 | INO ₂ + MCO ₃ | → | 0.85HNO ₃ + prod. |
| R19 | NO ₂ + (aerosol) | → | 0.5HNO ₃ + prod. |
| R20 | NO ₃ + (aerosol) | → | HNO ₃ + prod. |
| R21 | N ₂ O ₅ + (aerosol) | → | 2HNO ₃ |
| R22 | DMS + NO ₃ | → | HNO ₃ + prod. |
| R23 | BrNO ₃ + H ₂ O(l,s) | → | HNO ₃ + prod. |
| Chemical loss | | | |
| R24 | HNO ₃ + OH | → | H ₂ O + NO ₃ |
| R25 | HNO ₃ + hv | → | OH + NO ₂ |

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1435 Table 4. Sensitivity simulations to examine the impacts of uncertainties in surface-layer aerosol concentrations on simulated NO_3^-
 1436 in the UTLS. “Conc” and “Ctr” denote, respectively, simulated seasonal mean concentrations of SO_4^{2-} , NO_3^- , NH_4^+ , OC, BC and
 1437 their contributions to $\text{PM}_{2.5}$ (in percentages) during summertime (June-August) of 2005. The mass concentrations are averaged
 1438 over the TP/SASM region, with unit of $\mu\text{g m}^{-3}$ at the surface layer and of $10^{-2} \mu\text{g m}^{-3}$ at 200 hPa and 100 hPa. Also shown are the
 1439 NMBs, as the simulated surface-layer concentrations are compared with measurements described in Section 4.2.

| Species | Baseline Case | | | SO_2 (+20%) | | | NO_x (-50%) | | | NH_3 (-50%) | | | All Change | | |
|--------------------|---------------|-------|--------|----------------------|-------|--------|----------------------|-------|--------|----------------------|-------|--------|------------|-------|--------|
| | Conc. | Ctri. | NMB | Conc. | Ctri. | NMB | Conc. | Ctri. | NMB | Conc. | Ctri. | NMB | Conc. | Ctri. | NMB |
| Surface | | | | | | | | | | | | | | | |
| SO_4^{2-} | 1.70 | 35.9% | -14.7% | 1.92 | 38.1% | -4.4% | 1.58 | 39.5% | -18.1% | 1.70 | 38.1% | -14.7% | 1.78 | 43.2% | -8.3% |
| NO_3^- | 0.94 | 19.8% | +51.5% | 0.94 | 18.7% | +53.5% | 0.50 | 12.5% | -11.7% | 0.73 | 16.4% | +24.1% | 0.39 | 9.5% | -27.0% |
| NH_4^+ | 0.85 | 18.1% | +74.9% | 0.94 | 18.6% | +93.8% | 0.68 | 17.1% | +44.1% | 0.78 | 17.6% | +64.6% | 0.71 | 17.3% | +55.4% |
| OC | 0.94 | 19.8% | -57.2% | 0.94 | 18.6% | -57.2% | 0.94 | 23.4% | -57.2% | 0.94 | 21.0% | -57.2% | 0.94 | 22.7% | -57.2% |
| BC | 0.30 | 6.4% | -32.2% | 0.30 | 6.0% | -32.2% | 0.30 | 7.5% | -32.2% | 0.30 | 6.8% | -32.2% | 0.30 | 7.3% | -32.2% |
| 200 hPa | | | | | | | | | | | | | | | |
| SO_4^{2-} | 3.27 | 20.2% | | 3.67 | 22.9% | | 3.31 | 20.6% | | 3.29 | 29.1% | | 3.74 | 33.7% | |
| NO_3^- | 7.57 | 46.8% | | 7.05 | 43.9% | | 7.41 | 46.0% | | 3.86 | 34.2% | | 3.19 | 28.7% | |
| NH_4^+ | 2.67 | 16.5% | | 2.67 | 16.6% | | 2.71 | 16.8% | | 1.49 | 13.2% | | 1.50 | 13.5% | |
| OC | 2.22 | 13.7% | | 2.22 | 13.8% | | 2.22 | 13.8% | | 2.22 | 19.7% | | 2.22 | 20.0% | |
| BC | 0.44 | 2.7% | | 0.44 | 2.7% | | 0.44 | 2.7% | | 0.44 | 3.9% | | 0.44 | 4.0% | |
| 100 hPa | | | | | | | | | | | | | | | |
| SO_4^{2-} | 2.60 | 21.4% | | 2.80 | 23.0% | | 2.66 | 21.9% | | 2.60 | 25.2% | | 2.87 | 27.2% | |
| NO_3^- | 6.90 | 56.8% | | 6.72 | 55.3% | | 6.81 | 56.1% | | 5.68 | 55.0% | | 5.62 | 53.3% | |
| NH_4^+ | 1.43 | 11.8% | | 1.43 | 11.7% | | 1.45 | 12.0% | | 0.84 | 8.1% | | 0.84 | 8.0% | |
| OC | 1.05 | 8.6% | | 1.05 | 8.6% | | 1.05 | 8.7% | | 1.05 | 10.2% | | 1.05 | 10.0% | |
| BC | 0.16 | 1.3% | | 0.16 | 1.3% | | 0.16 | 1.3% | | 0.16 | 1.6% | | 0.16 | 1.5% | |

1440 **Figure Captions**

1441

1442 **Figure. 1.** Regions examined in this study: the Tibetan Plateau region (TP, 70–
1443 105°E, 25–40°N), the SASM region (SASM, 70–105°E, 10–25°N), and the
1444 anticyclone region of (20–120°E, 10–40°N).

1445

1446 **Figure. 2.** Monthly variations in emissions of NO_x (Tg N month^{-1}), SO_2 (Tg S
1447 month^{-1}), NH_3 (Tg N month^{-1}), OC (Tg C month^{-1}), and BC (Tg C month^{-1})
1448 over Asia. Values shown are the total emissions (anthropogenic plus natural
1449 emissions listed in Table 1).

1450

1451 **Figure. 3.** Simulated global distributions of surface-layer HNO_3 (pptv) and O_3
1452 (ppbv) averaged over June-August, 2005.

1453

1454 **Figure. 4.** Comparisons of simulated HNO_3 concentrations (pptv) with
1455 observations (pptv) from MLS. (a) and (b) are simulated concentrations at 200
1456 hPa and 100 hPa, respectively. (c) is the latitude-altitude cross section of
1457 simulated HNO_3 concentrations averaged over 70–105°E. (d)-(f) are the same
1458 as (a)-(c), except that (d)-(f) are observations from MLS. The GEOS-Chem
1459 HNO_3 simulations are smoothed by the corresponding averaging kernels (AK).
1460 White areas indicate lack of data meeting the retrieval quality criteria. All the
1461 data are averaged over June-August of 2005.

1462

1463 **Figure. 5.** Comparisons of simulated O_3 concentrations (ppbv) with
1464 observations (ppbv) from MLS. (a) and (b) are simulated concentrations at 200
1465 hPa and 100 hPa, respectively. (c) is the latitude-altitude cross section of
1466 simulated O_3 concentrations averaged over 70–105°E. (d)-(f) are the same
1467 as (a)-(c), except that (d)-(f) are observations from MLS. The GEOS-Chem O_3
1468 simulations are smoothed by the corresponding averaging kernels. White
1469 areas indicate lack of data meeting the retrieval quality criteria. All the data are
1470 averaged over June-August of 2005.

1471

1472 **Figure. 6.** The simulated and observed vertical profiles of monthly mean O_3
1473 mixing ratios at (a) Kunming and (b) Lhasa in August. The model results are
1474 from the simulation of year 2005. The observations in Kunming were
1475 conducted during August 7–13 (11 profiles of O_3 collected) in 2009 and during
1476 August 12–31 in 2012 (daily observations). The observations in Lhasa were
1477 conducted during August 22–28 in 2010 (12 profiles of O_3 collected) and
1478 during August 4–26 in 2013 (daily observations).

1479

1480 **Figure. 7.** Simulated seasonal mean concentrations ($\mu\text{g m}^{-3}$) of sulfate, nitrate,
1481 ammonium, organic carbon, black carbon, and $\text{PM}_{2.5}$ (defined as the sum of
1482 sulfate, nitrate, ammonium, BC, and OC) at (a) the surface layer, (b) 200 hPa,
1483 and (c) 100 hPa, during summer (June-August) of year 2005. Note that color
1484 bars are different for concentrations at the surface, 200 hPa, and 100 hPa.

1485

1486 **Figure. 8.** (a) Locations with measured aerosol concentrations from previous
1487 studies. Also shown are surface winds during summertime. (b)–(f) show the
1488 comparisons of simulated seasonal mean concentrations of sulfate, nitrate,

1489 ammonium, OC, and BC with measured values, respectively. Also shown in
1490 (b)–(f) are the 1:1 line (dashed), linear fit (solid line and equation), correlation
1491 coefficient between simulated and measured concentrations (R), and
1492 normalized mean bias (NMB) (defined as $NMB = \frac{\sum_{i=1}^n (P_i - O_i)}{\sum_{i=1}^n O_i} \times 100\%$, where P_i
1493 and O_i are predicted and observed concentrations at station i for each aerosol
1494 species).

1495
1496 **Figure. 9.** (a) Monthly mean distribution of aerosol extinction coefficients (km^{-1})
1497 at 100 hPa for July of 2005. (b) Monthly mean vertical distributions of aerosol
1498 extinction coefficients (at 525 nm for SAGE II and 550 nm for GEOS-Chem,
1499 with the contributions of both anthropogenic and natural aerosols considered)
1500 (km^{-1}) averaged over the Asian monsoon anticyclone region ($20\text{--}120^\circ\text{E}$,
1501 $10\text{--}40^\circ\text{N}$) for July of 2005. The horizontal dashed line represents the
1502 tropopause averaged over the Asian monsoon anticyclone region simulated by
1503 the GEOS-Chem model.

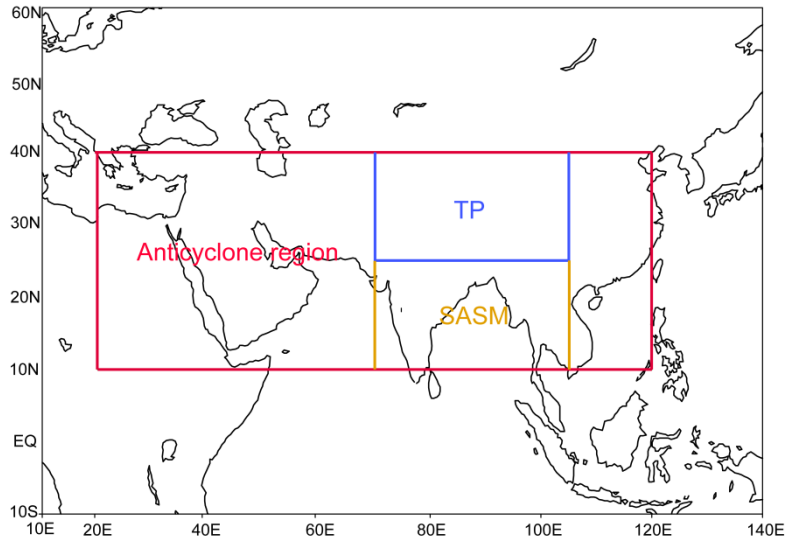
1504
1505 **Figure. 10.** Simulated contributions of nitrate to $\text{PM}_{2.5}$ ($C_{\text{NIT}} = [\text{NIT}] / [\text{PM}_{2.5}]$
1506 $\times 100\%$) averaged over summer (June–August) of year 2005 at (a)
1507 surface-layer, (b) 200 hPa, and (c) 100 hPa. (d) The latitude-altitude cross
1508 section of simulated C_{NIT} (%) averaged over $70\text{--}105^\circ\text{E}$.

1509
1510 **Figure. 11.** Latitude-altitude cross sections of simulated concentrations (color
1511 shades, $\mu\text{g m}^{-3}$) of SO_4^{2-} and NO_3^- averaged over $70\text{--}105^\circ\text{E}$ in June–August
1512 of 2005, together with the wind vectors obtained from the European Centre for
1513 Medium-Range Weather Forecasts (ECMWF) ERA-Interim Reanalysis data.
1514 The black line is the tropopause simulated by the GEOS-Chem model.

1515
1516 **Figure. 12.** Profiles of the net chemical production of HNO_3 by gas-phase
1517 reactions and heterogeneous reactions (red dotted line), the production of
1518 nitrate from gas to aerosol conversion of HNO_3 (blue dotted line) and the
1519 transport contributions of HNO_3 over the TP/SASM ($70\text{--}105^\circ\text{E}$, $10\text{--}40^\circ\text{N}$). Also
1520 shown is the average HNO_3 mixing ratios (black dotted line) over the TP/SASM
1521 region during summertime of year 2005.

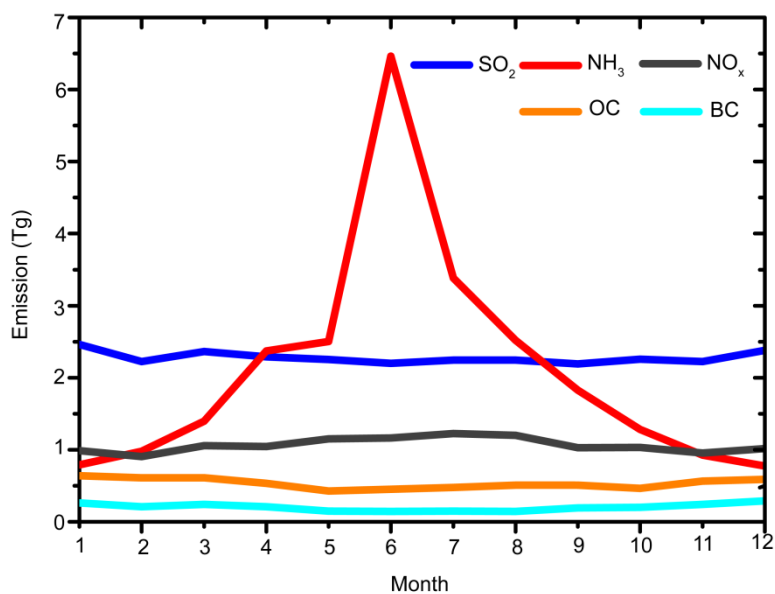
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1523 **Figure. 13.** (a)–(b) Distributions of RH (%) and temperature (K) at 100 hPa.
1524 (c)–(d) The latitude-altitude cross sections of RH (%) and temperature (K)
1525 averaged over $70\text{--}105^\circ\text{E}$. RH and temperature are from the GEOS5
1526 assimilated meteorological fields, and all the values are the averages over
1527 June–August of year 2005.

1528
1529 **Figure. 14.** Mass budget for nitrate aerosol within the selected box of (70--
1530 105°E , $10\text{--}40^\circ\text{N}$, $8\text{--}16$ km). E/W transport indicates net mass flux through the
1531 east and west lateral boundaries, N/S transport indicates net mass flux through
1532 the north and south lateral boundaries, and upward transport is the net mass
1533 flux through the top and bottom sides of the box. The mass flux is positive if it
1534 increases nitrate mass within the box. Unit of fluxes is Tg season^{-1} . All the
1535 values are the averages over June–August of 2005.



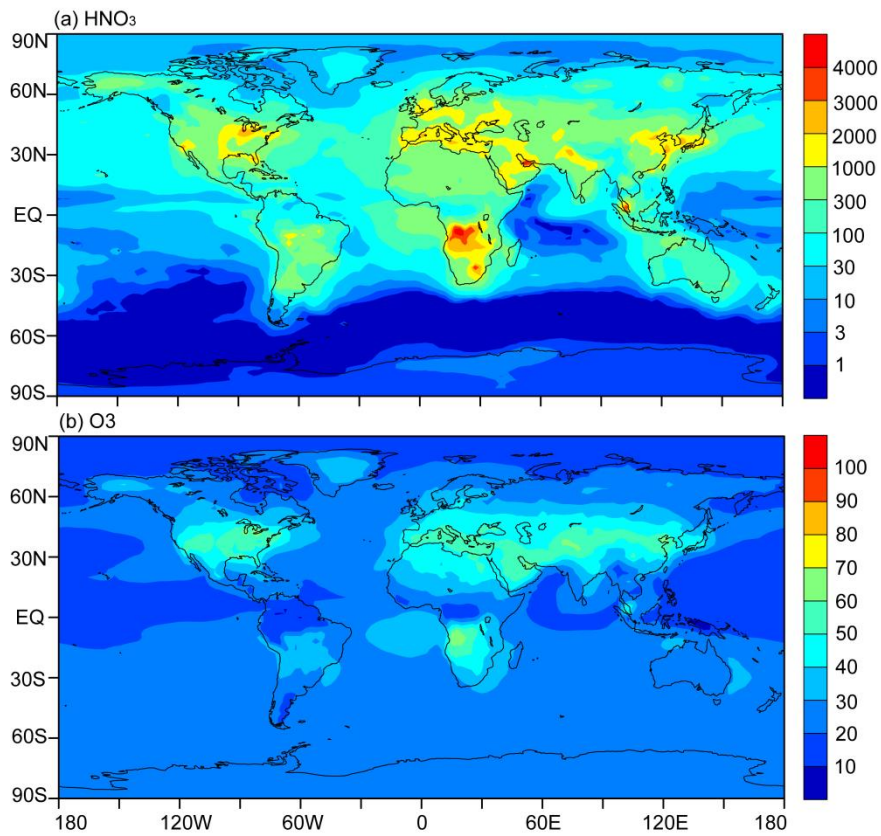
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Figure. 1. Regions examined in this study: the Tibetan Plateau region (TP, 70–105°E, 25–40°N), the SASM region (SASM, 70–105°E, 10–25°N), and the anticyclone region of (20–120°E, 10–40°N).



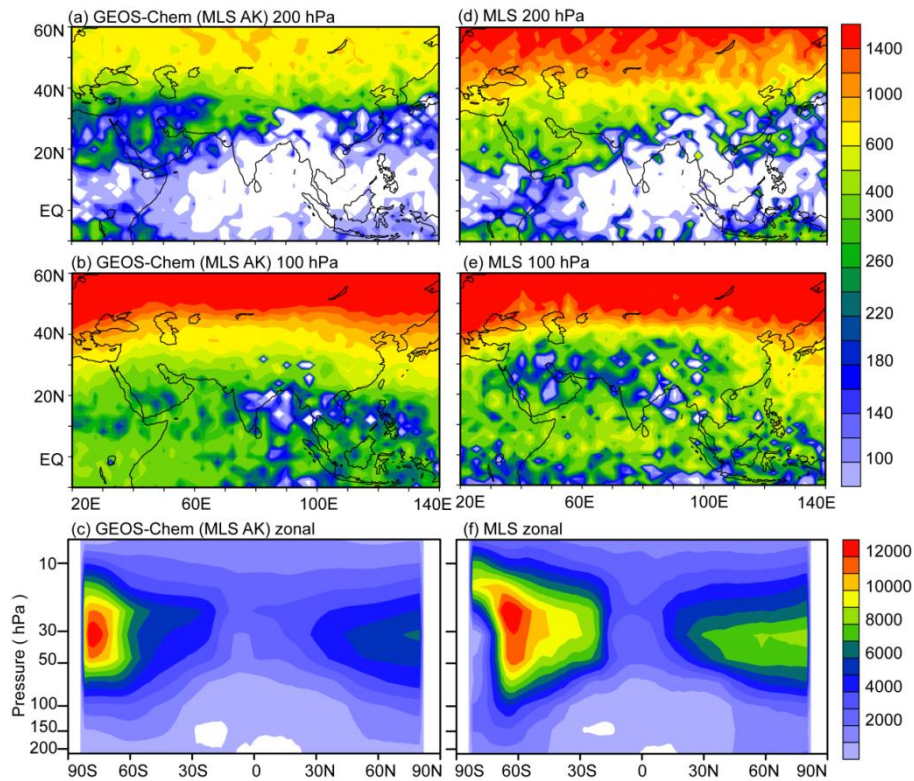
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Figure. 2. Monthly variations in emissions of NO_x (Tg N month⁻¹), SO₂ (Tg S month⁻¹), NH₃ (Tg N month⁻¹), OC (Tg C month⁻¹), and BC (Tg C month⁻¹) over Asia. Values shown are the total emissions (anthropogenic plus natural emissions listed in Table 1).



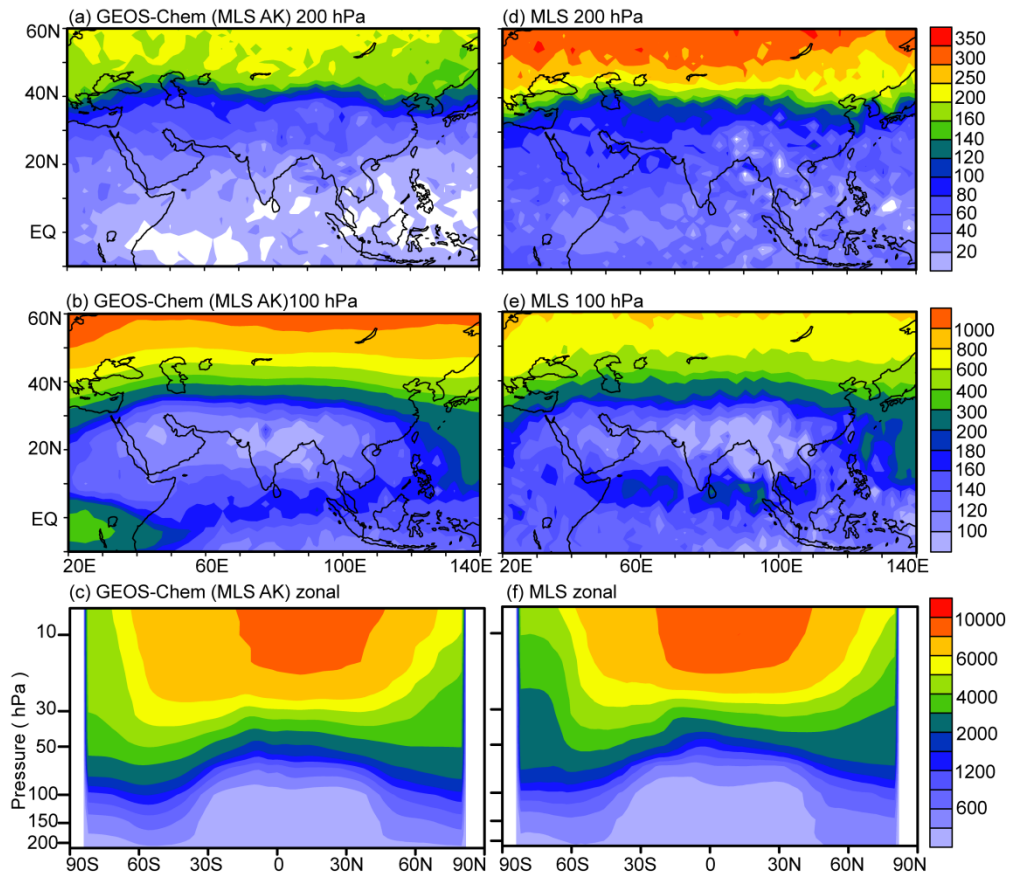
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Figure. 3. Simulated global distributions of surface-layer HNO_3 (pptv) and O_3 (ppbv) averaged over June-August, 2005.



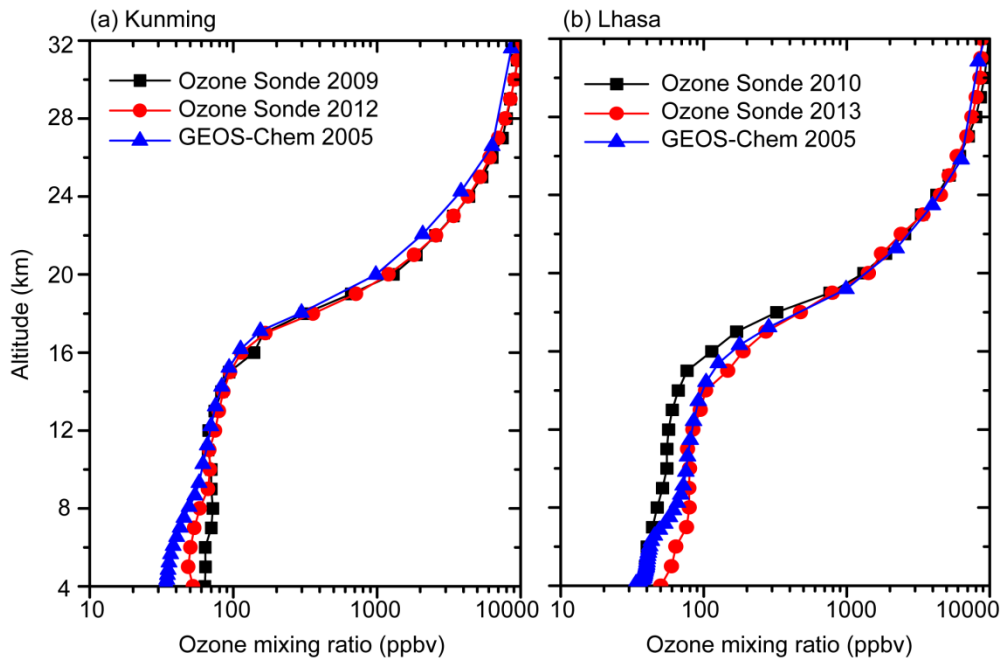
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1553 **Figure 4.** Comparisons of simulated HNO_3 concentrations (pptv) with
 1554 observations (pptv) from MLS. (a) and (b) are simulated concentrations at 200
 1555 hPa and 100 hPa, respectively. (c) is the latitude-altitude cross section of
 1556 simulated HNO_3 concentrations averaged over 70–105°E. (d)-(f) are the same
 1557 as (a)-(c), except that (d)-(f) are observations from MLS. The GEOS-Chem
 1558 HNO_3 simulations are smoothed by the corresponding averaging kernels (AK).
 1559 White areas indicate lack of data meeting the retrieval quality criteria. All the
 1560 data are averaged over June-August of 2005.



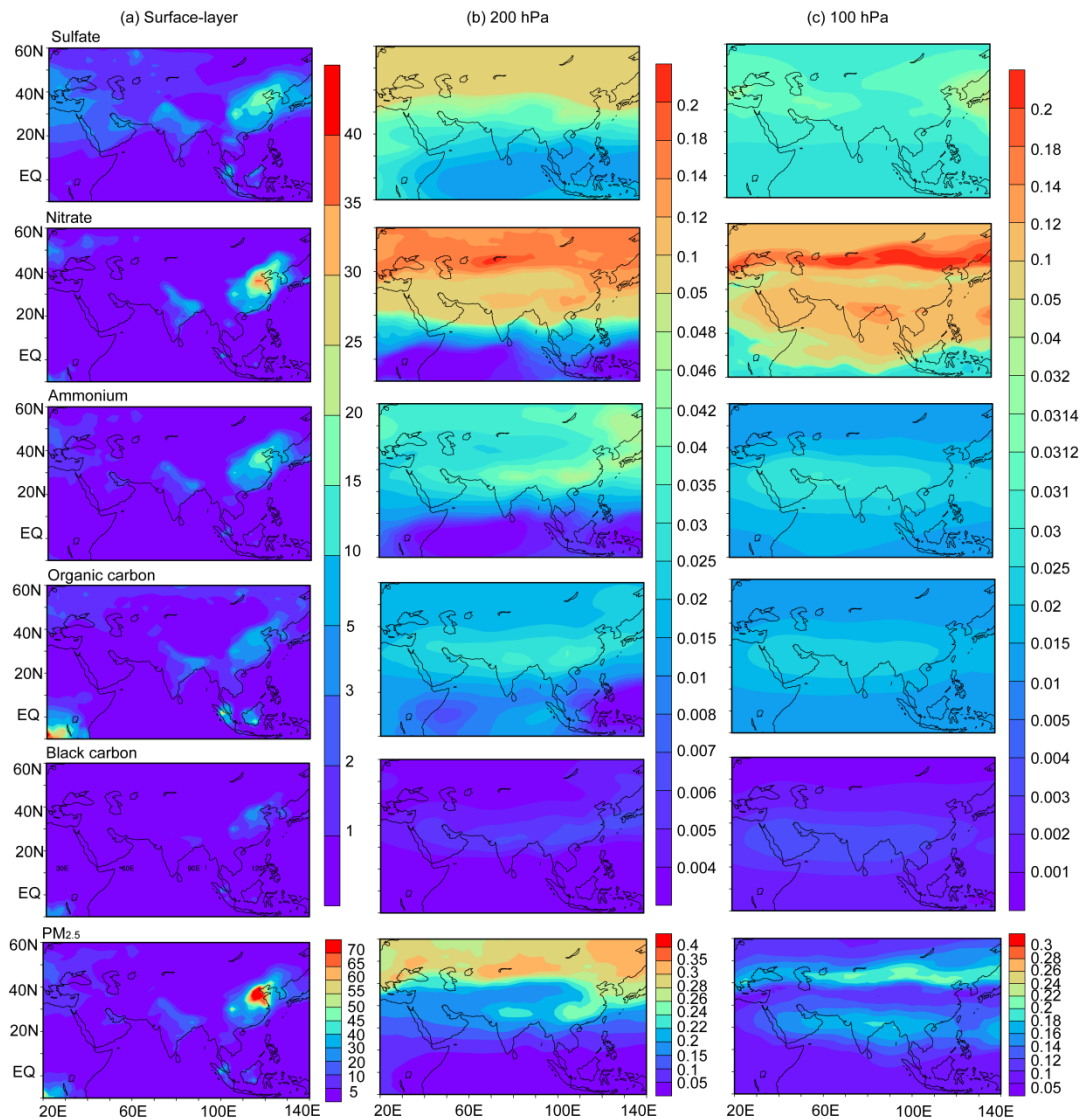
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Figure. 5. Comparisons of simulated O_3 concentrations (ppbv) with observations (ppbv) from MLS. (a) and (b) are simulated concentrations at 200 hPa and 100 hPa, respectively. (c) is the latitude-altitude cross section of simulated O_3 concentrations averaged over 70–105°E. (d)-(f) are the same as (a)-(c), except that (d)-(f) are observations from MLS. The GEOS-Chem O_3 simulations are smoothed by the corresponding averaging kernels. White areas indicate lack of data meeting the retrieval quality criteria. All the data are averaged over June-August of 2005.



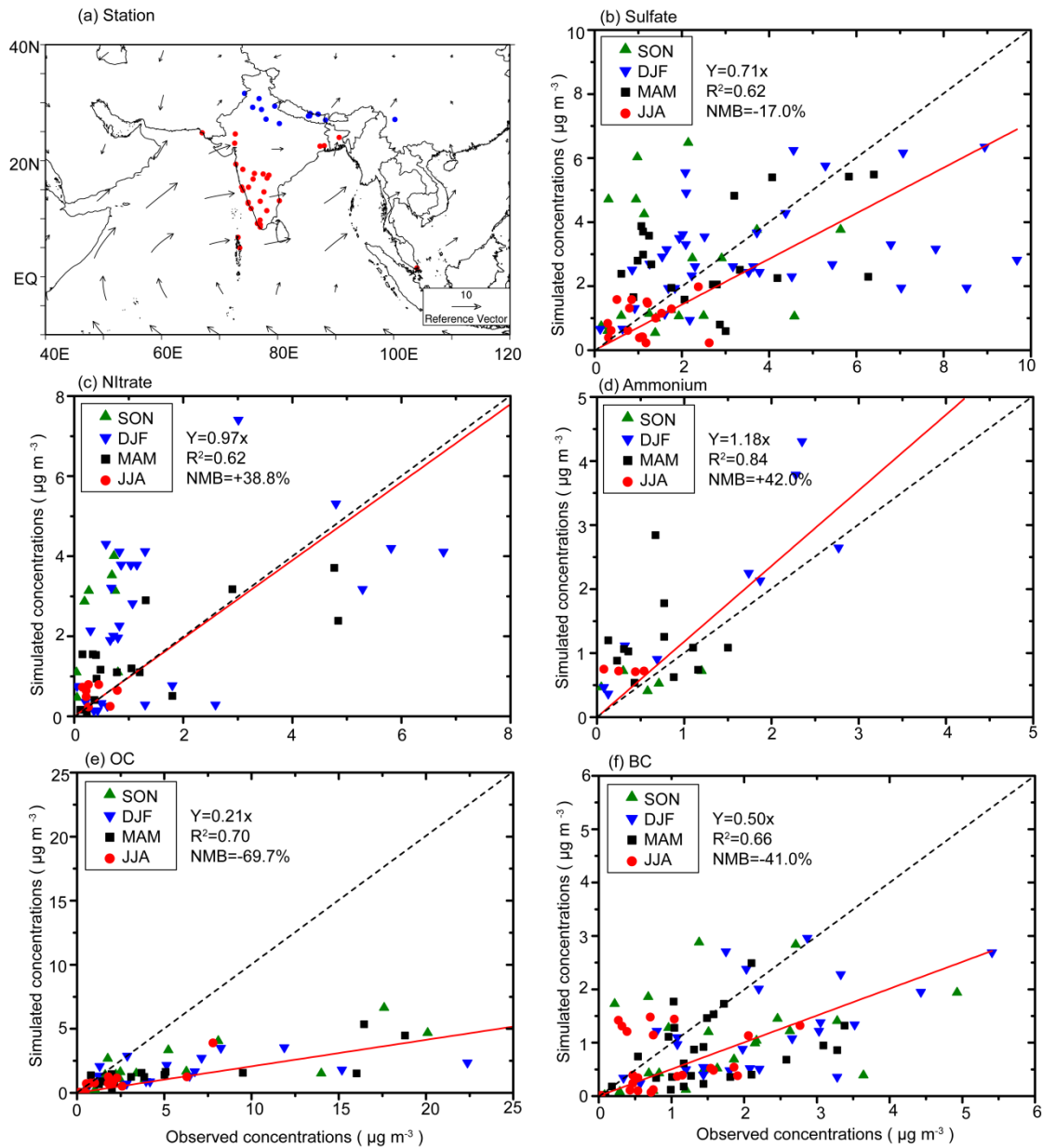
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Figure 6. The simulated and observed vertical profiles of monthly mean O_3 mixing ratios at (a) Kunming and (b) Lhasa in August. The model results are from the simulation of year 2005. The observations in Kunming were conducted during August 7–13 (11 profiles of O_3 collected) in 2009 and during August 12–31 in 2012 (daily observations). The observations in Lhasa were conducted during August 22–28 in 2010 (12 profiles of O_3 collected) and during August 4–26 in 2013 (daily observations).



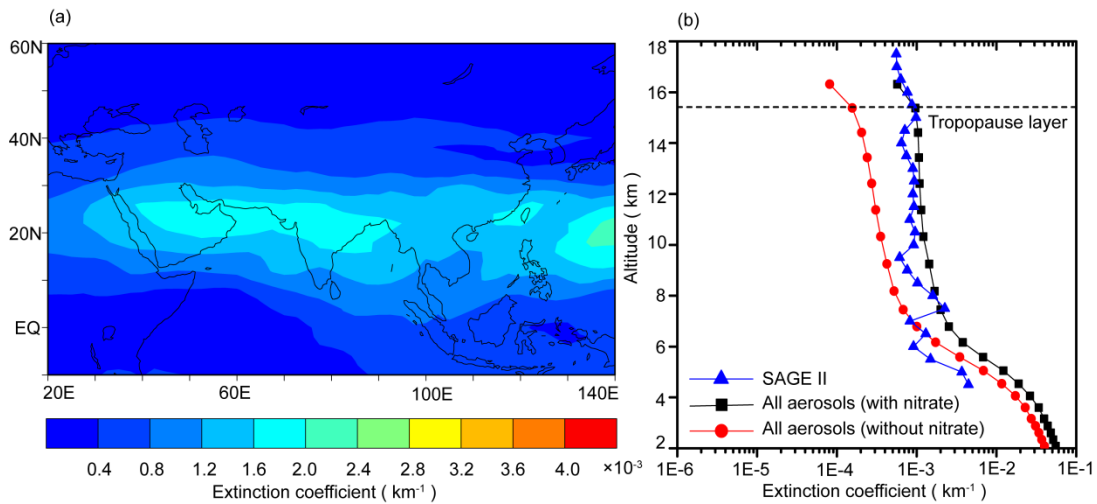
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1582 **Figure. 7.** Simulated seasonal mean concentrations ($\mu\text{g m}^{-3}$) of sulfate, nitrate,
 1583 ammonium, organic carbon, black carbon, and $\text{PM}_{2.5}$ (defined as the sum of
 1584 sulfate, nitrate, ammonium, BC, and OC) at (a) the surface layer, (b) 200 hPa,
 1585 and (c) 100 hPa, during summer (June-August) of year 2005. Note that color
 1586 bars are different for concentrations at the surface, 200 hPa, and 100 hPa.



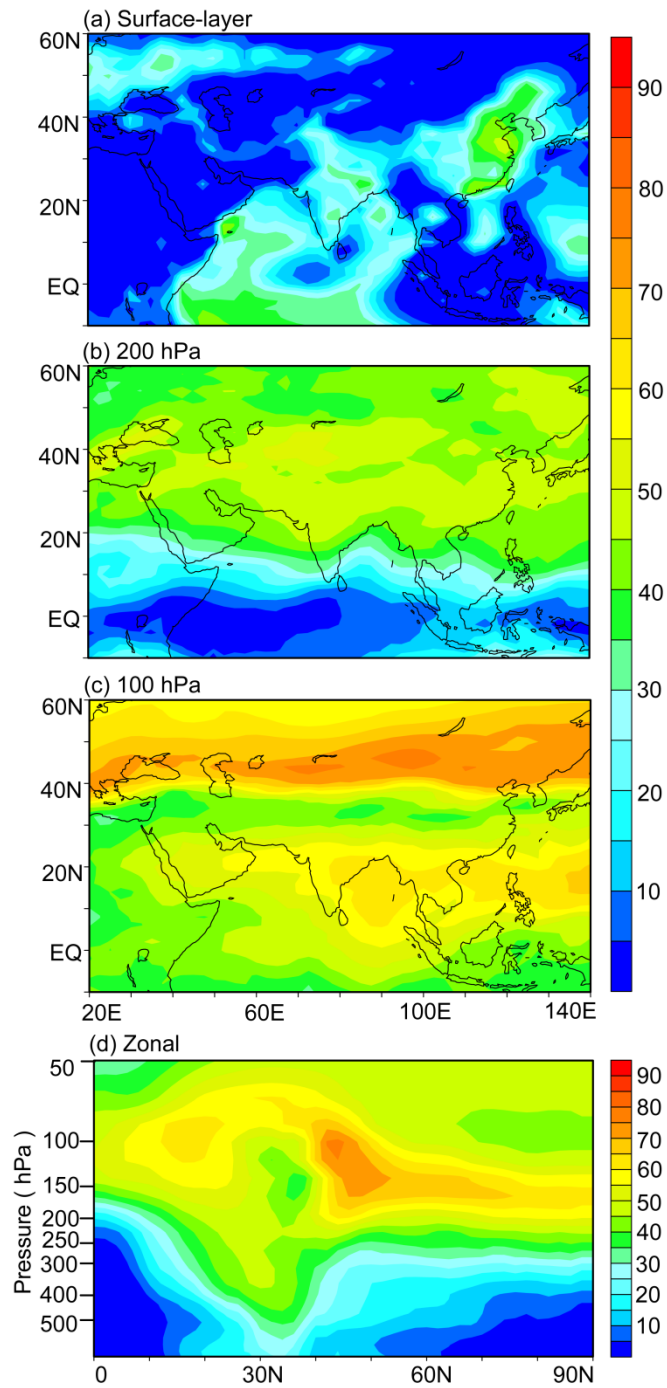
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1589 **Figure 8.** (a) Locations with measured aerosol concentrations from previous
1590 studies over the TP (blue) and SASM (red) region. Also shown are surface
1591 winds during summertime. (b)–(f) show the comparisons of simulated seasonal
1592 mean concentrations of sulfate, nitrate, ammonium, OC, and BC with
1593 measured values, respectively. Also shown in (b)–(f) are the 1:1 line (dashed),
1594 linear fit (solid line and equation), correlation coefficient between simulated
1595 and measured concentrations (R), and normalized mean bias (NMB) (defined
1596 as $NMB = \frac{\sum_{i=1}^n (P_i - O_i)}{\sum_{i=1}^n O_i} \times 100\%$, where P_i and O_i are predicted and observed
1597 concentrations at station i for each aerosol species).



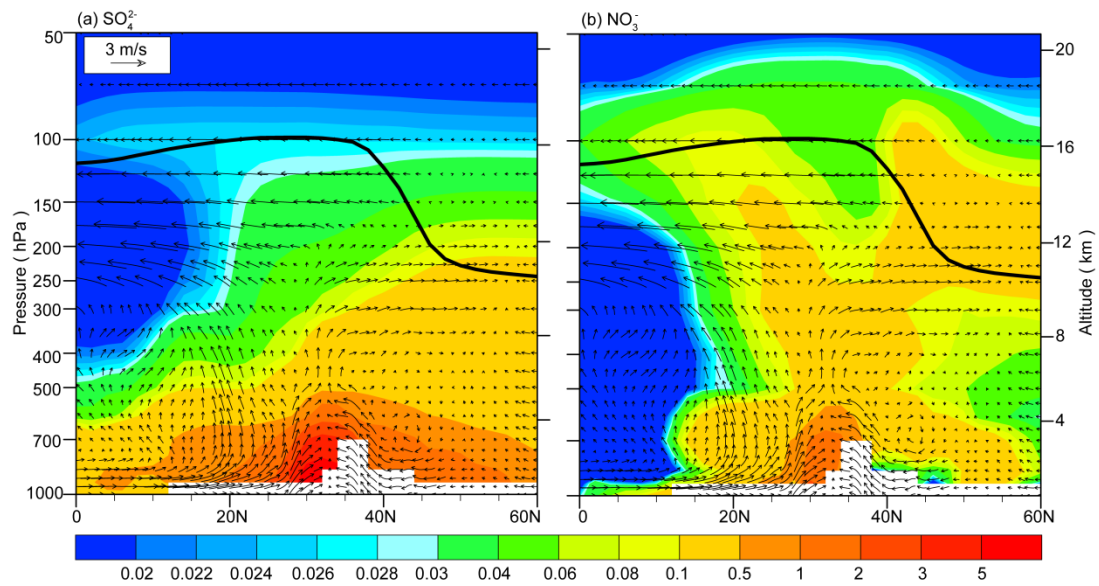
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1600 **Figure. 9.** (a) Monthly mean distribution of aerosol extinction coefficients (km^{-1})
1601 at 100 hPa for July of 2005. (b) Monthly mean vertical distributions of aerosol
1602 extinction coefficients (at 525 nm for SAGE II and 550 nm for GEOS-Chem,
1603 with the contributions of both anthropogenic and natural aerosols considered)
1604 (km^{-1}) averaged over the Asian monsoon anticyclone region (20–120°E,
1605 10–40°N) for July of 2005. The horizontal dashed line represents the
1606 tropopause averaged over the Asian monsoon anticyclone region simulated by
1607 the GEOS-Chem model.



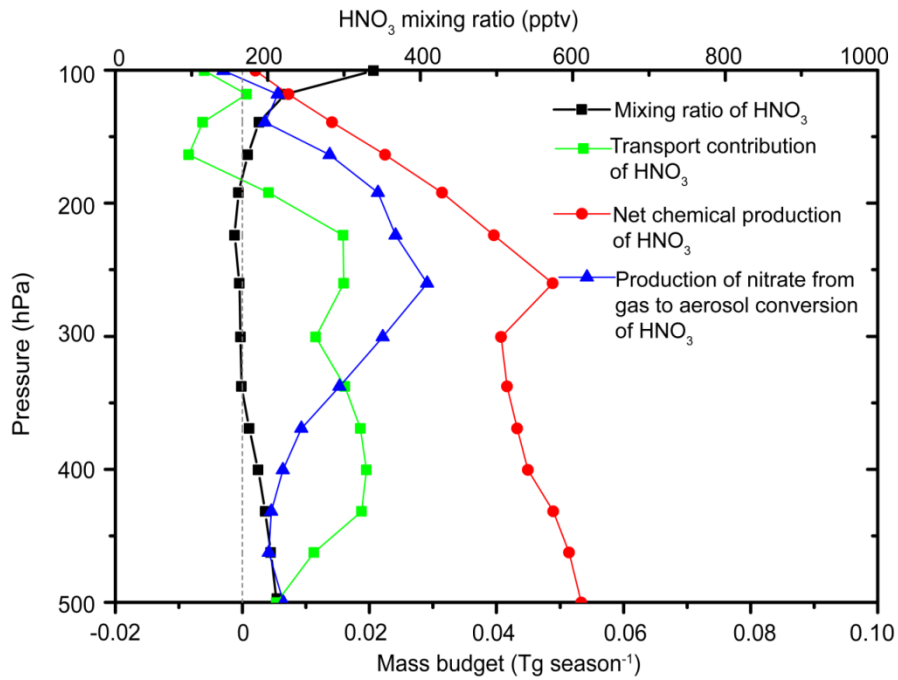
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Figure. 10. Simulated contributions of nitrate to PM_{2.5} ($C_{\text{NIT}} = [\text{NIT}] / [\text{PM}_{2.5}] \times 100\%$) averaged over summer (June-August) of year 2005 at (a) surface-layer, (b) 200 hPa, and (c) 100 hPa. (d) The latitude-altitude cross section of simulated C_{NIT} (%) averaged over 70–105°E.



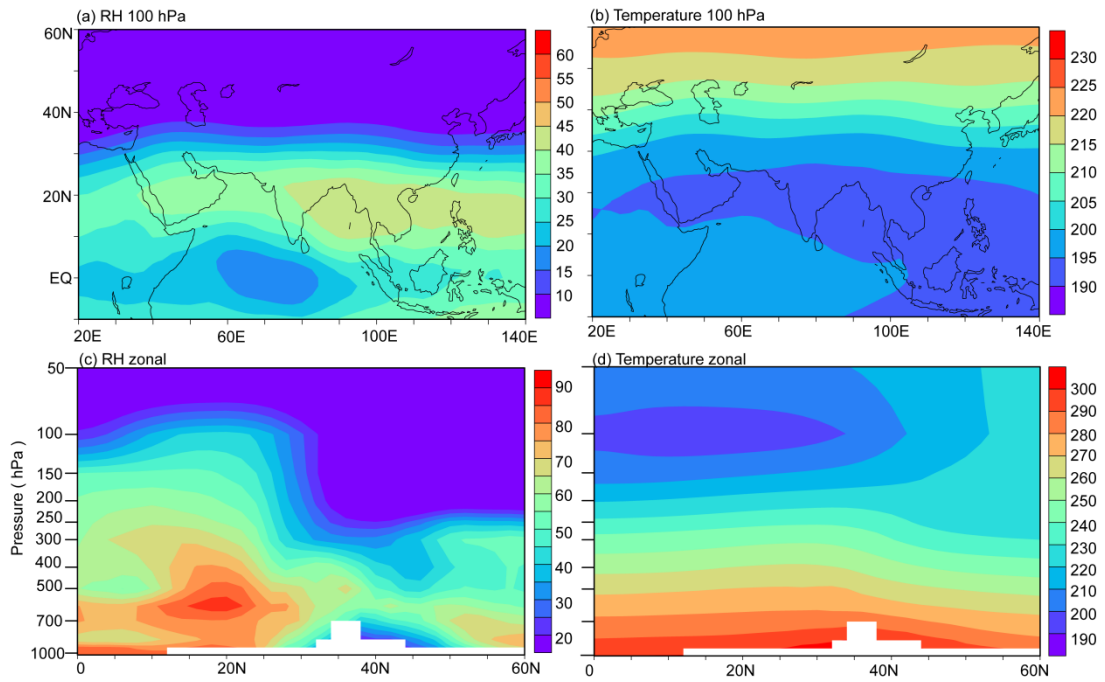
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Figure. 11. Latitude-altitude cross sections of simulated concentrations (color shades, $\mu\text{g m}^{-3}$) of SO_4^{2-} and NO_3^- averaged over $70\text{--}105^\circ\text{E}$ in June-August of 2005, together with the wind vectors obtained from the European Centre for Medium-Range Weather Forecasts (ECMWF) ERA-Interim Reanalysis data. The black line is the tropopause simulated by the GEOS-Chem model.



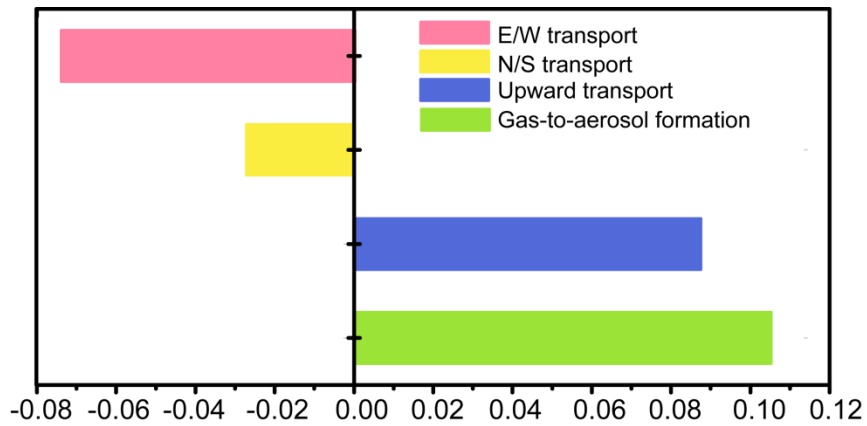
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Figure 12. Profiles of the net chemical production of HNO₃ by gas-phase reactions and heterogeneous reactions (red dotted line), the production of nitrate from gas to aerosol conversion of HNO₃ (blue dotted line) and the transport contributions of HNO₃ over the TP/SASM (70–105°E, 10–40°N). Also shown is the average HNO₃ mixing ratios (black dotted line) over the TP/SASM region during summertime of year 2005.



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Figure. 13. (a)-(b) Distributions of RH (%) and temperature (K) at 100 hPa. (c)-(d) The latitude-altitude cross sections of RH (%) and temperature (K) averaged over 70–105°E. RH and temperature are from the GEOS5 assimilated meteorological fields, and all the values are the averages over June-August of year 2005.



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Figure. 14. Mass budget for nitrate aerosol within the selected box of (70–105°E, 10–40°N, 8–16 km). E/W transport indicates net mass flux through the east and west lateral boundaries, N/S transport indicates net mass flux through the north and south lateral boundaries, and upward transport is the net mass flux through the top and bottom sides of the box. The mass flux is positive if it increases nitrate mass within the box. Unit of fluxes is Tg season⁻¹. All the values are the averages over June-August of 2005.