

# Modelling the deep convective transport of trace gases (CO, NH<sub>3</sub> and SO<sub>2</sub>) from the planetary boundary layer to the Asian summer monsoon anticyclone

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**Abstract.** Deep convection plays a vital role in transporting Asian pollutants from the planetary boundary layer (PBL) into the Asian summer monsoon anticyclone (ASMA). However, the efficiency and effectiveness of transporting pollutants with various chemical and physical properties to the ASMA remain unclear. In this study, we use the global atmospheric chemistry and climate model EMAC to investigate the deep convective transport of trace gases such as CO, NH<sub>3</sub> and SO<sub>2</sub> from the PBL to the ASMA over the years 2010-2020. We quantify the deep convective transport efficiency of different trace gases into the ASMA. We show that the strongest convective transport tendency occurs over northern India and the southern edge of the Tibetan Plateau for CO (0.2-0.5 ppbv hr<sup>-1</sup>), over the south and eastern parts of the Tibetan Plateau for NH<sub>3</sub> (0.02-0.05 ppbv hr<sup>-1</sup>), and over central India and eastern China for SO<sub>2</sub> (0.002-0.005 ppbv hr<sup>-1</sup>). We find that, in contrast to CO and NH<sub>3</sub>, the SO<sub>2</sub> enhancements within the ASMA are very weak, and there ~~is~~ can even be a decrease in SO<sub>2</sub> over the southern Tibetan Plateau relative to the surroundings. Our analysis indicates that gas-liquid partitioning in clouds and subsequent wet deposition over South Asia are more effective at reducing SO<sub>2</sub> than NH<sub>3</sub> reaching the Tibetan Plateau and the ASMA. In view of ongoing changes in regional emissions, the effects of deep convective transport of various pollutants and associated gas-aerosol-cloud interactions on the chemical features of the ASMA require continued investigation.

## 1 Introduction

Deep convection plays an important role in the vertical redistribution of trace gases and the pollution transport from the planetary boundary layer (PBL) to the upper troposphere and lower stratosphere (UTLS) (Chatfield and Crutzen, 1984; Dickerson et al., 1987; Lelieveld and Crutzen, 1994; Pan et al., 2024). The rapid transport of reactive pollutants from the PBL to the free troposphere and the tropopause region can significantly impact the UTLS chemistry globally (Pickering et al., 1990; Thompson et al., 1994; Thornton et al., 1997; Barth et al., 2007; Bertram et al., 2007; Barth et al., 2012). The deep and widespread convection associated with the Asian summer monsoon is ~~a convective manifestation~~ observed in June, July, and August that is absent in the Northern Hemisphere wintertime (Houze Jr. et al., 2015). Deep convection can transport Asian pollutants, especially those from South Asia and East Asia, to the Asian summer monsoon anticyclone (ASMA) (Hoskins and Rodwell, 1995), where they are confined by the ASMA flow, forming a distinct chemical regime in the UTLS of the Northern Hemisphere during summertime (Randel and Park, 2006; Park et al., 2007; Park et al., 2008; Randel et al., 2010; Smith et al., 2025a). The strength of the barrier separating air masses inside and outside the anticyclone depends on altitude, and there are also daily and yearly variations of the ASMA in relation to its boundary, area, and dynamic behaviour (Ploeger et al., 2015; Kachula et al., 2025). Satellite observations have revealed increasing levels of tropospheric gases (e.g., CO, HCN and NH<sub>3</sub>) within the ASMA relative to ~~outside its surroundings~~ (Kar et al., 2004; Randel et al., 2010; Höpfner et al., 2016; Santee et al., 2017). A widespread strong enhancement of aerosols within the ASMA, namely the Asian Tropopause Aerosol Layer (ATAL), has also been detected by satellites (Vernier et al., 2011; Thomason and Vernier, 2013; Vernier et al., 2015).

The ASMA can serve as “~~an efficient smokestack vertical conduit~~” venting Asian surface pollutants to the global stratosphere (Yu et al., 2017; Bian et al., 2020). While the central role of the ASMA is recognized, there is still inconsistency about the most efficient pathway for the transport of moisture and air pollutants to the UTLS (Chen et al., 2012; Chen et al., 2024). The Tibetan Plateau plays a critical role in transporting air pollutants from the lower troposphere to the stratosphere during the Asian summer monsoon (Zhou et al., 1995). Deep convection over the Tibetan Plateau and its southern slope is considered “a short circuit” for the transport of water vapor and polluted air to the global stratosphere (Li et al., 2005; Fu et al., 2006). The middle troposphere centered over the southern Tibetan Plateau can act as “a well-defined conduit”, where strong convection lofts air parcels in the boundary layer into the ASMA (Bergman et al., 2013; Bian et al., 2020). In addition, the northern Bay of Bengal and adjacent land area, where air pollution from the Indian subcontinent converges, has been identified as the central aerosol-convection source area for the ATAL (e.g., Park et al., 2009; Fadnavis et al., 2013; He et al., 2021; Nützel et al., 2022). The emissions from South Asia have been shown to make a dominant contribution to gaseous and aerosol pollutants trapped in the ASMA (Bergman et al., 2013; Lelieveld et al., 2018; Ma et al., 2022). While SO<sub>2</sub> and NO<sub>2</sub> emissions in India have been observed to increase rapidly (Krotkov et al., 2016), how these South Asian pollutants are transported to the ASMA and how they affect its chemical features remains an area in need of in-depth investigation.

The ASMA constitutes a seasonally persistent, zonally restricted circulation pattern transporting climate-relevant emissions rapidly from surface sources to higher altitudes (Vogel et al., 2015; Vogel et al., 2016; Ploeger et al., 2017; Vogel et al., 2023).

Transport by deep convection generally occurs alongside scavenging and multiphase chemical reactions involving reactive trace gases and aerosols, and it is also accompanied by various gas-aerosol-cloud interactions (Iribarne and Pyshnov, 1990; Pruppacher and Klett, 1997; Zondlo et al., 1997; Seinfeld and Pandis, 2006; Bertram et al., 2007). In addition to the convective ~~dynamic~~-activity, the deep convective transport efficiency of air pollutants, which reflects the amount of gases or aerosols reaching the outflow at the cloud top relative to that of the inflow into the convective mass near the cloud base, can change significantly with the difference in their solubility, hydrophilicity and reactivity (Barth et al., 2007; Barth et al., 2015; Yang et al., 2015; Bela et al., 2016). Therefore, understanding the various physical and chemical processes involved in the deep convective transport is essential for revealing the chemical characteristics of air masses within the ASMA and the formation mechanism of the ATAL. Several measurement campaigns have been conducted successfully to explore the chemical composition and spatiotemporal distributions of air pollutants in the UTLS over the Asian summer monsoon region, including the High Altitude and Long Range Research Aircraft (HALO) measurement (e.g., Gottschaldt et al., 2017), the Oxidation Mechanism Observations (OMO) campaign (Lelieveld et al., 2018; Hottmann et al., 2020), the Balloon Measurements of the Asian Tropopause Aerosol Layer (BATAL) (Vernier et al., 2018; Vernier et al., 2022), the StratoClim field campaign (Höpfner et al., 2019; Johansson et al., 2020; Lee et al., 2021; von Hobe et al., 2021; Appel et al., 2022; Singer et al., 2022; Ebert et al., 2024; Johansson et al., 2024), the Asian Summer Monsoon Chemical and CLimate Impact Project (ACCLIP) (e.g., Pan et al., 2025; Smith et al., 2025a), and the Probing High Latitude Export of Air from the Asian Summer Monsoon (PHILEAS) campaign (Riese et al., 2025; Vogel et al., 2025). However, these measurements in Asia have still not fully characterized and quantified the deep convective transport of air pollutants from the PBL to the ASMA. The chemical composition of the convective inflow and outflow was measured during ~~compared to~~ the Deep Convective Clouds and Chemistry (DC3) field experiment taking place in North America (e.g., Barth et al., 2015); these airborne measurements alone, while valuable, are not sufficiently to fully characterize and quantify deep convective transport due especially to limited sampling in space and time, and numerical modeling considering detailed chemical, physical and dynamical processes are still needed to provide such estimates (Barth et al., 2007; Barth et al., 2012; Barth et al., 2026).

In the present study, we use the ECHAM/MESSy Atmospheric Chemistry (EMAC) model (Jöckel et al., 2006; Jöckel et al., 2010) to investigate the deep convective transport of trace gases, including carbon monoxide (CO), ammonia (NH<sub>3</sub>) and sulfur dioxide (SO<sub>2</sub>), from the PBL to the ASMA. CO is a typical pollution tracer that is transported into and trapped within the ASMA (Fu et al., 2006; Park et al., 2007). NH<sub>3</sub> and SO<sub>2</sub> have a much higher solubility and reactivity in water clouds than CO, and can act as gaseous precursors of ammonium sulfate aerosols (Seinfeld and Pandis, 2006). Therefore, deep convective transport of NH<sub>3</sub> and SO<sub>2</sub> from the PBL to the ASMA may significantly influence the formation processes and chemical composition of the ATAL (Höpfner et al., 2019). There is substantial interannual variability in the ASMA. For example, in summer 2015, the monsoon, in particular upward transport in the ASMA, was strongly influenced by El Niño, which has a global impact lasting over many months (Kunze et al., 2016; Santoso et al., 2017; Yan et al., 2018; Fadnavis et al., 2019; Ravindra Babu et al., 2021; Becker et al., 2025). Further, the composition of the ASMA tends to have strong day-to-day variability, influenced by various meteorological conditions such as typhoons (Vogel et al., 2014; Hanumanthu et al., 2020; Li et al.,

2020;Li et al., 2021;Li et al., 2023). For this study, we performed model simulations using EMAC for a relatively long period, from January 2010 through December 2020. This study aims to investigate the deep convective transport of selected trace gases to the ASMA in June-August (JJA) for a climatology of 2010-2020.

This paper is organized as follows. In Sect. 2, we describe the EMAC model and settings used for this study. In Sect. 3, we present the model simulation results, including the tendency and efficiency of the deep convective transport of CO, NH<sub>3</sub> and SO<sub>2</sub> into the ASMA. ~~In Sect. 4, we provide a discussion, and~~ exploring the effects of partitioning into the clouds and wet scavenging processes on the deep convective transport of NH<sub>3</sub> and SO<sub>2</sub> into the ASMA. A summary of the main findings and conclusions is given in Sect. ~~5~~4.

## 2 Model description and setup

10 The EMAC model is a global atmospheric chemistry and climate model that combines the 5th generation European Centre – Hamburg general circulation model (ECHAM5) (Roeckner et al., 2006) with the Modular Earth Submodel System (MESSy) Atmospheric Chemistry system (Jöckel et al., 2006;Jöckel et al., 2010) to simulate atmospheric processes from the troposphere to the middle atmosphere and their interactions with oceans, land, and anthropogenic influences. For this study, we used the EMAC (ECHAM5 version 5.3.02, MESSy version 2.55.0), which includes the MESSy submodels describing various chemical, 15 physical and dynamical processes in detail (Jöckel et al., 2016). The model resolution used in this study is T63L90, corresponding to a horizontal grid resolution of about  $1.875^\circ \times 1.875^\circ$  and 90 vertical layers extending from the Earth's surface to an altitude of 0.01 hPa (~80 km).

In EMAC, heterogeneous and gas-phase chemistry are simulated online using the MECCA submodel (Sander et al., 2011;Sander et al., 2019). MECCA calculates the concentration of a range of gases and radicals, including reactive and aerosol 20 precursor species, such as CO, SO<sub>2</sub>, NH<sub>3</sub>, nitrogen oxides (NO<sub>x</sub>  $\equiv$  NO +NO<sub>2</sub>), and volatile organic compounds (VOCs), and major oxidant species like OH, O<sub>3</sub>, H<sub>2</sub>O<sub>2</sub>, and NO<sub>3</sub>. In this study, the Mainz isoprene mechanism (Taraborrelli et al., 2009), and halogen and sulfur stratospheric chemistry (Brühl et al., 2015), were included in the MECCA calculation, following our previous work (Ma et al., 2019). Photolysis rates for the troposphere up to the mesosphere are calculated by the JVAL submodel (Jöckel et al., 2006), which considers absorption and scattering by gases, aerosols and clouds using a delta-two-stream method. 25 The uptake of SO<sub>2</sub> and NH<sub>3</sub> and the aqueous-phase oxidation of SO<sub>2</sub> by H<sub>2</sub>O<sub>2</sub> and O<sub>3</sub> in cloud droplets are calculated by the SCAV submodel (Tost et al., 2006a;Tost et al., 2007). The removal of gases and aerosols through wet deposition is calculated by the SCAV submodel (Tost et al., 2006a), and dry deposition is calculated by the DRYDEP submodel (Kerkweg et al., 2006).

Aerosol microphysics and gas/aerosol partitioning are treated by the GMXe submodel (Pringle et al., 2010), which uses seven interacting lognormal modes (M7) to describe the typical size range of aerosol species, including the nucleation mode 30 and hydrophilic and hydrophobic Aitken, accumulation and coarse modes. The properties of aerosols in each mode are represented by the number concentration, total mass (internal mixture of contributing species), density, median radius, and

width of the lognormal distribution. The inorganic aerosol composition is simulated by the ISORROPIA-II thermodynamic equilibrium model (Fountoukis and Nenes, 2007), with updates as discussed in the work of Capps et al. (2012).

Convective cloud processes and convective tracer transport are calculated using the CONVECT and CVTRANS submodels (Tost et al., 2006b; Tost et al., 2010), respectively. CONVECT consists of an interface for selecting different convection schemes (Tost et al., 2006b), and in this study we used the Tiedtke convection scheme with Nordeng closure (Tiedtke, 1989; Nordeng, 1994), which has proven to be the best-performing, including in the recent work of Xenofontos et al. (2025). Convective cloud microphysics is based on temperature and moisture profiles, though without interactively accounting for the influence of aerosols on liquid droplet or ice formation processes. Large-scale cloud processes are described by the CLOUD submodel (Roeckner et al., 2006), and the original ECHAM5 cloud scheme without aerosol–cloud interactions was used for this study. CVTRANS calculates the tracer transport following the bulk approach (“leaky pipe”) based on Lawrence and Rasch (2005). It is implemented with an interface to collect the required updraft and downdraft air mass fluxes and the respective entrainment and detrainment rates from CONVECT for a selected convection scheme (e.g. the Tiedtke-Nordeng scheme in this study) (Tost et al., 2010) (Jeske and Tost, 2025).

The CAMS-GLOB-ANT emission inventory (v4.2, <https://eccad.sedoo.fr>, last access: 1 November 2025), with a horizontal grid resolution of  $0.5^\circ \times 0.5^\circ$  at monthly intervals, was utilized for surface anthropogenic emissions of the trace gases (including CO, NH<sub>3</sub> and SO<sub>2</sub>) and aerosols in this study. For aircraft emissions, the CAMS-GLOB-AIR emission inventory (v1.1, <https://eccad.sedoo.fr>, last access: 1 November 2025) was used. The land and water biological emissions of NH<sub>3</sub> and non-methane hydrocarbons (NMHCs) are based on the Global Emissions Inventory Activity (GEIA) database (Bouwman et al., 1997). NO<sub>x</sub> produced by lightning is calculated online and distributed vertically based on the parameterization of Price and Rind (1992). The NO soil emissions are calculated online according to the algorithm of Yienger and Levy II (1995). Biomass burning emissions are calculated by the BIOBURN submodel (Kaiser et al., 2012), which determines the fluxes based on biomass burning emission factors and dry matter combustion rates from the Global Fire Assimilation System (GFAS).

The SO<sub>2</sub> emissions from explosive volcanic eruptions occurring at different locations and latitudes were taken into account using the volcanic SO<sub>2</sub> plumes detected by various (occultation and limb-based) satellite instruments (Schallock et al., 2023). There were about 146 explosive volcanic eruption events accounted for the years 2010–2020, and a three-dimensional volcanic SO<sub>2</sub> plume (in unit of volume mixing ratio) for each event was added on-line to the background values of SO<sub>2</sub> in the UTLS at the corresponding time during the model simulation, as done in our previous work (Brühl et al., 2018; Ma et al., 2019).

The EMAC model has been evaluated against various observations of trace gases and aerosols in both the troposphere and stratosphere, including aircraft measurements conducted within the ASMA in some specific years (Gottschaldt et al., 2017; Lelieveld et al., 2018; Tomsche et al., 2019; Johansson et al., 2020; Xenofontos et al., 2024). These comparisons indicate that EMAC can simulate complex dynamic, physical and chemical processes from the PBL to the UTLS over the Asian summer monsoon region (e.g., Gottschaldt et al., 2018; Ma et al., 2019; Rosanka et al., 2021; Becker et al., 2025). In this study, the model simulation was performed for the years 2010–2020 at an integration time step of 10 minutes. The simulation was nudged by the ECMWF's ERA5 meteorological re-analysis data at time intervals of 6 hours (Hersbach et al., 2020). The nudging was

exerted for temperature, vorticity, divergence, and surface pressure, with maximum weights at the model levels from about 10 hPa to 706 hPa (except for surface pressure). The chemical initial conditions of trace gases and aerosols were provided by the results of a previous simulation using EMAC T106L90 (Ma et al., 2019). The simulation results were output at 5-hour intervals for analysis. Since each hour of the day is accounted once for a period of about 5-days, daily variation characteristic will not be lost when a seasonally or longer-term averaged value is calculated.

### 3 Results and discussion

~~The objective of this study is to investigate the deep convective transport of the primary gaseous pollutants CO, NH<sub>3</sub> and SO<sub>2</sub> from the planetary boundary layer (PBL) to the Asian summer monsoon anticyclone (ASMA). During the summer season, i.e., June-August (JJA), deep convection associated with the Asian summer monsoon is observed each year (Houze Jr. et al., 2015) and is selected for the analysis. We focus on the climatology of deep convective transport of selected trace gases into the ASMA in JJA during 2010-2020.~~

#### 3.1 Spatial distributions of CO, NH<sub>3</sub> and SO<sub>2</sub> in the PBL during the Asian summer monsoon

The Asian summer monsoon region is characterized by enhanced precipitation, strong and deep convection, and a distinct vertical structure in the circulation, with a cyclonic flow and convergence in the lower troposphere and a strong anticyclone and divergence in the UTLS (Krishnamurti and Bhalme, 1976; Hoskins and Rodwell, 1995; Wang and LinHo, 2002). These climatic features associated with the Asian summer monsoon are well captured by our EMAC model with 11 years of simulated meteorological data (see Fig. S1). Figure 1 shows the averaged PBL's wind field and volume mixing ratios of CO, NH<sub>3</sub> and SO<sub>2</sub> in JJA over the years 2010-2020. The influence and footprints of the large anthropogenic emission sources near the Earth's surface, especially those in northern India and eastern China (see Fig. S2), are clearly evident in the geographic distributions of these primary gaseous pollutants within the PBL. As shown in Fig. 1, in the PBL, the pollutants from South Asia (northern India in particular) can be transported by the cyclonic flow (dominated by southwesterlies over the Indian subcontinent) to the southern flank of the Tibetan Plateau. Further transport of these pollutants to the large Tibetan Plateau platform appears to be limited by the topographical block at the steep southern slopes of the plateau with less thermal impact (Boos and Kuang, 2010).

While the PBL's CO levels are comparable between northern India and eastern China, the PBL's NH<sub>3</sub> levels are 5-10 ppbv higher in northern India than in eastern China and, in contrast, the PBL's SO<sub>2</sub> levels are 4-8 ppbv lower in northern India than in eastern China (Fig. 1). For NH<sub>3</sub>, IASI satellite observations also showed a similar spatial distribution pattern, with higher abundance over northern India than eastern China, due to stronger emission strength from the former (Liu et al., 2022; Luo et al., 2022). As can be estimated from Fig. 1, the ratios of the PBL's NH<sub>3</sub> volume mixing ratio to the PBL's SO<sub>2</sub> volume mixing ratio are about 4-6 in northern India, and these ratios drop to 1.5-2 in eastern China. Since NH<sub>3</sub> is an alkaline gas and SO<sub>2</sub> is an acidic gas, changes in their relative abundance will affect aqueous-phase chemistry and the scavenging process when they are dissolved in clouds (Seinfeld and Pandis, 2006). It should be noted that CAMS emissions of SO<sub>2</sub> over China might

underestimate recent reductions due to environmental policies over 2010-2020, which can result in an overestimation of its PBL abundance and convective transport to the upper troposphere over eastern China (Smith et al., 2025b).

### **3.2 Deep convection frequency and spatial distributions of CO, NH<sub>3</sub> and SO<sub>2</sub> within the ASMA over the Tibetan Plateau and the surrounding areas**

5 Deep convection usually occurs as part of a mesoscale convective system with a sufficient moisture supply at cloud base (Houze Jr., 2004). Strong ascent with deep convection can reach the upper troposphere above 10 km above sea level (a.s.l., hereafter, all altitudes are referred to a.s.l. unless specified otherwise), and even higher into the lower stratosphere (Meenu et al., 2010; Emmanuel et al., 2021). The ASMA is located at an altitude range of about 10 km to 18 km in the vertical, and horizontally it covers a larger area ranging 30-120°E and 20-40°E, which can be highlighted using the geopotential height  
10 distribution at 100 hPa (Fig. S1) as done by Basha et al. (2020). For this study, we used the convective cloud top heights output at intervals of 5 hours over the simulation period to count the events of deep convection reaching a selected altitude (e.g., 10-14 km) or higher which corresponds to a middle level in the vertical range of the ASMA. Then, we calculated the frequency of deep convection at the defined altitudes by dividing the number of deep convection events by the total number across all datasets (including both cloudy and cloud-free cases) used for counting. In this study, we employed the all-day Global Cloud Product derived from a single-layer cloud retrieval model within the DaYu cloud analysis system (DaYu-GCP), based on the merged thermal infrared brightness temperature of the global geostationary satellite from the Gridded Satellite project (Knapp et al., 2011), to evaluate the EMAC model. DaYu-GCP has a temporal resolution of 3 hours and a spatial resolution of 0.07°, covering regions from 70°S to 70°N with a total temporal span of 23 years (2000–2022), including cloud phase, cloud top height, cloud effective radius, and cloud optical thickness (Zhao et al., 2026). For the analysis in this research, data from 2010 to 2020 were used.

Figure 2 presents the deep convection frequency for JJA during 2010-2020 for the convective cloud top heights above 10 km, 12 km, 14 km, and 16 km, respectively, corresponding to the main vertical range of the ASMA) from both the satellite observations and EMAC simulations. Except for the oceanic regions, the frequency of deep convection reaching altitudes above 10 km is the highest (up to ~20%) over the Tibetan Plateau among the continental Asian summer monsoon regions. Both satellite observations and model simulations show that at for altitudes above 14 km, the frequency of deep convection is prominently high (up to reaches 10%) over the southern Tibetan Plateau, higher than that over compared to the surrounding regions within the ASMA. However, the satellite observations show a maximum in the deep convection frequency over southern India (around 20°N), which is not well predicted by EMAC, probably due to inappropriate convective parameterization in the model, e.g., using the same parameters for the ocean and land. It should also be noted that there are still some uncertainties in the deep convection frequency estimated from different satellite observations (Fu et al. 2006; Smith et al., 2025a). It is also shown that deep convection can reach to altitudes above 16 km over the Tibetan Plateau, although the corresponding frequencies are as small as ~1% or less, lower than those over the surrounding areas to its south within the ASMA. Our model results of enhanced deep convection frequency over the Tibetan Plateau are also supported by satellite

observations presented in previous work (see Figure 2 of Fu et al. (2006), which has shown that number counts of convective clouds over the Tibetan are larger than those over the Tibetan southern slope at an altitude range of 11-16 km and those over the Indian monsoon region throughout all the altitude range. These results are also in agreement with previous studies in that convective clouds form frequently over the Tibetan Plateau due to the abundant water vapor convergence, elevated land surface and strong radiative heating (e.g., Sugimoto and Ueno, 2010; Xu et al., 2014). They are also consistent with the work of Legras and Bucci (2020), which shows that the Tibetan Plateau, with the largest number of high clouds, favours the deep convective transport of compounds released from ground level to much higher altitudes, even into the lower stratosphere.

### **3.3 Spatial distributions of CO, NH<sub>3</sub> and SO<sub>2</sub> within the ASMA**

We evaluate the model's performance in simulating CO, NH<sub>3</sub> and SO<sub>2</sub> within the ASMA, using satellite measurements of CO from the Aura Microwave Limb Sounder (MLS) (Livesey et al., 2018; Yan et al., 2019 and reference therein), NH<sub>3</sub> and SO<sub>2</sub> from the Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) (Höpfner et al., 2015b; Höpfner et al., 2016). The model output data for each model grid cell at 5-hr intervals under cloud-free conditions were selected by using a cloud fraction of 0.3 as threshold, and derived data were analysed statistically and compared with satellite observations (see Fig. 3). Comparing between MLS observed CO (Fig. 3a1) and EMAC simulated CO (Fig. 3a2) at 178 hPa for JJA during 2010-2020, one can see an enhancement of CO on the southern parts of the ASMA from both the satellite observations and model simulations. EMAC well reproduced the concentration level and distribution pattern of CO within the ASMA observed by MLS.

There are only two years (2010 and 2011) of data that overlap between MIPAS observations and model simulations for this study. Fig. 3 b1-b2 (and c1-c2) show comparisons between MIPAS observed NH<sub>3</sub> (and SO<sub>2</sub>) and EMAC simulated NH<sub>3</sub> (and SO<sub>2</sub>) at 15 km height averaged for JJA 2010-2011. Apparently, MIPAS observations do not support the EMAC model simulations, the latter showing increased NH<sub>3</sub> over northern India and the southern Tibetan Plateau and decreased SO<sub>2</sub> over large areas within the ASMA. In contrast to the model, MIPAS detected an enhancement of NH<sub>3</sub> over the northwest Tibetan Plateau (70-90 °E, 30-40 °N) and a reduction of NH<sub>3</sub> over the southern Tibetan Plateau and the area to its south (80-100 °E, 20-30 °N). Such satellite-observed NH<sub>3</sub> distribution pattern in the ASMA is typical during 2002-2011 when MIPAS data are available (not shown in the paper). An explanation for these differences might be that EMAC ignored some large NH<sub>3</sub> emission sources near the northwest Tibetan Plateau and overestimated NH<sub>3</sub> transported to the ASMA over the southern Tibetan Plateau and larger surrounding areas due to the use of an inappropriate emission inventory or convective/scavenging parametrization. MIPAS observations might have been perturbed by optically absorptive dust aerosols (which can be lofted to the UTLS (Ma et al., 2019)) over the northern Tibetan Plateau and by optically scattered cloud particles over the southern Tibetan Plateau and Asian monsoon region. However, such perturbations are generally removed from the analysed MIPAS- spectra by the applied cloud-filter. For the remaining spectra, as aerosols and clouds exhibit a relatively smooth radiance signal compared to the

spectral lines of gases, such perturbations are accounted for by the baseline-fit during retrieval. Thus, we do not estimate that such effects can explain the considerable differences between observation and model simulation in the case of NH<sub>3</sub>.

The volcanic eruption's impacts on the regional distributions of SO<sub>2</sub> are visible from both MIPAS observations and EMAC simulations shown in Fig. 3 c1-c2 (the latter considered the SO<sub>2</sub> emissions from explosive volcanic eruptions as described in Sect. 2). The impacts of the Nabro volcano erupted on 12–13 June 2011 can be seen more clearly by comparing the regional distributions of SO<sub>2</sub> between 2010 and 2010 (see Figs. S3 and S4). It should be noted that MIPAS samples might not align well with those from EMAC due to limited overpass time for satellite observations, resulting in sparser SO<sub>2</sub> hot spots at the ASMA's edge compared to the model results. As shown in Fig. S5, there are much fewer cloud-free samples in the southern parts of the ASMA than in the northwest parts for both the satellite and the model. These might induce large uncertainties in the comparison between satellite observations and model simulations. The comparisons presented here have provided the regional distribution characteristics of trace gases in the ASMA and its surroundings under cloud-free conditions. Since convective transport is generally associated with cloud processes, below we analyse the model results using the data for both the clear-sky and cloudy conditions.

Figure 34 shows the averaged volume mixing ratios of CO, NH<sub>3</sub> and SO<sub>2</sub> at the selected altitudes of 10-12 km and 14-16 km in JJA over the years 2010-2020, respectively. It can be seen that the spatial distributions of CO, NH<sub>3</sub>, and SO<sub>2</sub> within the ASMA differ. The enhancements of CO and NH<sub>3</sub> within the ASMA are clearly visible, with the maxima occurring over northern India and the southern edge of the Tibetan Plateau for CO and over the southern and eastern parts of the Tibetan Plateau for NH<sub>3</sub>. The CO spatial distribution character within the ASMA, as simulated in this study, has been well recognized by a large number of previous studies (e.g., Park et al., 2007; Pan et al., 2016). The NH<sub>3</sub> enhancements within the ASMA characterized by this study are also in agreement with the satellite observations and past model study before (Ge et al., 2018; Ma et al., 2019).

An analysis of the MIPAS satellite data showed the enhanced mixing ratios of SO<sub>2</sub> at altitudes of 16-18 km within the ASMA (Höpfner et al., 2015a), but these enhancements of SO<sub>2</sub> are not as significant as those of CO and NH<sub>3</sub>. Figure 3-4 indicates that SO<sub>2</sub> enhancements occur over central India (a small area around 20 °N latitude) and eastern China, but these enhancements are very weak. In contrast, our simulation shows a decrease of SO<sub>2</sub> over the southern Tibetan Plateau relative to the surroundings within the ASMA, which was not detected by satellite measurements. According to our simulation, SO<sub>2</sub> levels within the ASMA are very low (below 0.05 ppbv), similar to those reported by Höpfner et al. (2015a) for cloud-free conditions. For SO<sub>2</sub>, the averaged mixing ratios in the upper troposphere within the ASMA are about two orders of magnitude lower than those in the polluted area within the PBL, whereas they are about one order of magnitude lower for NH<sub>3</sub> and only a few times lower for CO.

### **3.3.4 Tendency and efficiency of the deep convective transport of CO, NH<sub>3</sub> and SO<sub>2</sub> into the ASMA**

As mentioned in Sect. 2, convective transport of the tracers was simulated using the CVTRANS submodel, with the updraft and downdraft mass fluxes, entrainment and detrainment calculated by the CONVECT submodel. In the CVTRANS scheme,

convective transport is calculated separately from scavenging, and the tracers are redistributed vertically without a net gain or loss in the whole convective column (Jeske and Tost, 2025). The tendency of a tracer due to convective transport alone can be obtained by extracting a corresponding variable (named 'xtte\_cvtrans'), which ~~from~~ is the difference in the tracer's mixing ratio before and after the implementation of CVTRANS. Simulated results for xtte\_cvtrans are saved at 5-hr intervals as done for other variables, and these instantaneous values of xtte\_cvtrans can be considered as averages over each time interval, with its unit changed from mol mol<sup>-1</sup> s<sup>-1</sup> to nmol mol<sup>-1</sup> hr<sup>-1</sup> (ppbv hr<sup>-1</sup>) or pmol mol<sup>-1</sup> hr<sup>-1</sup> (pptv hr<sup>-1</sup>). For the cases without convection, the tendency is set as zero and is accounted for when doing seasonal and climatic averaging. Here, in this study, the mean tendency is calculated by averaging the data across all time intervals, rather than only for deep convection events.

Figure 45 shows the averaged deep convective transport tendency of CO, NH<sub>3</sub> and SO<sub>2</sub> at the selected altitudes of 10-12 km and 14-16 km in JJA over the years 2010-2020, respectively. Strong deep convective transport tendency within the ASMA can be found over northern India and the southern edge of the Tibetan Plateau for CO (0.2-0.5 ppbv hr<sup>-1</sup>), over the southern and eastern parts of the Tibetan Plateau for NH<sub>3</sub> (0.02-0.05 ppbv hr<sup>-1</sup>), and over central India and eastern China for SO<sub>2</sub> (0.002-0.005 ppbv hr<sup>-1</sup>). These spatial distribution patterns in the deep convective transport tendency are similar to those in the volume mixing ratios of CO, NH<sub>3</sub> and SO<sub>2</sub> shown in Fig. 34, indicating a ~~dominant~~ role of deep convection in the enhancements of CO, NH<sub>3</sub> and SO<sub>2</sub> within the ASMA. It can be seen that the maximum tendency for SO<sub>2</sub> (e.g., over central India) is much lower (by an order of magnitude) than that for NH<sub>3</sub> (e.g., over the southern Tibetan Plateau). Moreover, in contrast to CO and NH<sub>3</sub>, an enhancement of the strong deep convective transport tendency cannot be found for SO<sub>2</sub> over the southern Tibetan Plateau. It is indicated that, in addition to the deep convective transport itself, other factors, e.g., the scavenging process associated with it, influence the amount of pollutants reaching the ASMA.

In this study, the deep convective transport efficiency of a trace gas into the ASMA is defined by the ratio of the updraft mass flux of this trace gas at a selected height (i.e.g., 10 km) to its maximum near the cloud base in ~~the a~~ convective column (see Fig. S3). The updraft mass flux of a trace gas is calculated as the updraft mass flux of the air times the mass mixing ratio of this trace gas at the same altitude. It can be expected that, in addition to convective activity, cloud scavenging may influence the deep convective transport efficiency of the trace gas by reducing the amount that reaches the ASMA.

Figure 56 shows the ~~mean~~averaged deep convective transport efficiency of CO, NH<sub>3</sub> and SO<sub>2</sub> into the ASMA (defined with a lower boundary of 10 km height) in JJA over the years 2010-2020, respectively. The deep convective transport efficiency is ~~very high~~ (above 50%) over the central and southern Tibetan Plateau for all three trace gases considered, indicating that the middle troposphere of the Tibetan Plateau ~~is~~ can be a potentially an effective pathway for transporting pollutants from the PBL to the ASMA. With a low solubility and thus not affected by scavenging, CO has a higher deep convective transport efficiency than NH<sub>3</sub> and SO<sub>2</sub> over all the continental regions where deep convection occurs. For instance, over the southern Tibetan Plateau, the maximum in the deep convective transport efficiency reaches about 80%-90% for CO, 60%-70% for NH<sub>3</sub>, and 60%-80% for SO<sub>2</sub>. Over northern India (excluding the southern flank of the Tibetan Plateau), the deep convective transport efficiency is 20%-30% for CO, 2%-4% for NH<sub>3</sub>, and less than 1% for SO<sub>2</sub>. For the deep convective transport efficiency of these trace gases at 14 km (a middle altitude of the ASMA), maximum values are also found over the southern Tibetan Plateau

(Fig. S6). The deep convective transport efficiency of SO<sub>2</sub> calculated here appears to be comparable to that of NH<sub>3</sub> over the southern Tibetan Plateau. It should be noted that there could be an overestimation of the deep convective transport efficiency of SO<sub>2</sub> here since a considerable fraction of SO<sub>2</sub> in the upper troposphere might come from other sources than Asian surface emissions, e.g., volcanic SO<sub>2</sub> from explosive eruptions (Neely et al., 2014; Ma et al., 2019).

## 5 **4 Discussion**

~~In this section, we aim to explain the results presented in the above section (Sect. 3) by investigating the cloud and wet scavenging processes over the Asian summer monsoon region during the same period (i.e., JJA during 2010-2020). Among the three trace gases, only the cloud and wet scavenging processes of NH<sub>3</sub> and SO<sub>2</sub> are investigated since CO has a low solubility and is not considered in the EMAC submodel SCAV. The focus is on the causes of the difference between NH<sub>3</sub> and SO<sub>2</sub> in the deep convective transport from the PBL to the ASMA.~~

### **3.4.1.5 Partitioning of NH<sub>3</sub> and SO<sub>2</sub> between the gas and liquid phases in clouds**

In the above section, we show that the middle troposphere over the Tibetan Plateau is an effective pathway of transporting the pollutants from the PBL to the ASMA by deep convection, with some differences in the deep convective transport efficiency for different trace gases, e.g., CO vs. NH<sub>3</sub>, and NH<sub>3</sub> vs. SO<sub>2</sub> (Fig. S6). Among the three trace gases, NH<sub>3</sub> and SO<sub>2</sub> are only involved in the cloud and wet scavenging processes of NH<sub>3</sub> and SO<sub>2</sub> are investigated since, and CO has a low solubility and is not considered in the EMAC submodel SCAV. To investigate the effect of cloud process on the differences in the deep convective transport efficiency, we looked at the vertical column densities of gaseous NH<sub>3</sub> and SO<sub>2</sub> in the air (denoted as NH<sub>3</sub>(air) and SO<sub>2</sub>(air), simply as NH<sub>3</sub> and SO<sub>2</sub>), their dissolving and reaction products in the clouds (denoted as NH<sub>x</sub>(liq) and SO<sub>x</sub>(liq)), and the total (i.e., NH<sub>3</sub>(air)+NH<sub>x</sub>(liq) and SO<sub>2</sub>(air)+SO<sub>x</sub>(liq)) within a height range of 6-10 km for the simulation period (see Fig. S47). The NH<sub>x</sub> in the clouds is the sum of liquid ammonia and ammonium, i.e., NH<sub>x</sub>(liq) ≡ NH<sub>3</sub>(liq) + NH<sub>4</sub><sup>+</sup>(liq). The SO<sub>x</sub> in the clouds is the sum of liquid SO<sub>2</sub> and its dissociation and oxidation products (denoted by S(IV) and S(VI)), i.e., SO<sub>x</sub>(liq) ≡ S(IV, liq) + S(VI, liq), where S(IV, liq) ≡ SO<sub>2</sub>(liq) + HSO<sub>3</sub><sup>-</sup>(liq) + SO<sub>3</sub><sup>2-</sup>(liq) and S(VI, liq) ≡ H<sub>2</sub>SO<sub>4</sub>(liq) + HSO<sub>4</sub><sup>-</sup>(liq) + SO<sub>4</sub><sup>2-</sup>(liq). It is noted that for the investigated region and period of this study, NH<sub>x</sub>(liq) and SO<sub>x</sub>(liq) are dominated by NH<sub>4</sub><sup>+</sup>(liq) (>99.9%) and SO<sub>4</sub><sup>2-</sup>(liq) plus HSO<sub>4</sub><sup>-</sup>(liq) (>99%), respectively.

Figure 67 shows the relative contributions of the liquid NH<sub>x</sub> and SO<sub>x</sub> in the clouds to the total (gas-phase plus liquid-phase) within a vertical column of 6-10 km in JJA during 2010-2020, respectively. One can see considerable amounts of gaseous NH<sub>3</sub> and SO<sub>2</sub> at heights above 6 km over the Tibetan Plateau partitioning into the clouds before reaching the ASMA. Similar to the spatial distributions of NH<sub>3</sub> and SO<sub>2</sub>, the enhancements in the 6-10 km vertical columns of cloudy NH<sub>x</sub>(liq) and SO<sub>x</sub>(liq) occur over the Tibetan Plateau as well (Fig. 4S7). The relative contributions of the 6-10 km column in the clouds to the total (gas-phase plus liquid-phase) over the Tibetan Plateau are about 10-30% for NH<sub>x</sub>(liq) and 50-80% for SO<sub>x</sub>(liq), respectively. ~~In this study, t~~he primary partitioning of the soluble gases between the gas and liquid phases ~~was~~ can be treated

with Henry's law, with a Henry's law constant of  $203 \text{ M atm}^{-1}$  for  $\text{NH}_3$  and  $3.13 \text{ M atm}^{-1}$  for  $\text{SO}_2$  at a temperature of  $0 \text{ }^\circ\text{C}$ , respectively. While the environmental conditions, including the cloud liquid water content, are the same, the lower Henry's law constant for  $\text{SO}_2$  than for  $\text{NH}_3$  cannot explain the higher fraction of  $\text{SO}_2$  than  $\text{NH}_3$  partitioning into the liquid phase simulated by the model. The gases dissolved in water can dissociate into ions, e.g.,  $\text{NH}_4^+(\text{liq})$ ,  $\text{HSO}_3^-(\text{liq})$ , and  $\text{SO}_3^{2-}(\text{liq})$ , following chemical equilibria included in the SCAV submodel. Therefore, the effective Henry's law coefficients, which consider trace gas dissociating into ions (as done in our model simulations), are better to represent the partitioning of the trace gases like  $\text{NH}_3$  between the gas and liquid phases (Seinfeld and Pandis, 2006). Under atmospheric conditions, practically all dissolved ammonia in clouds exists as the ammonium ion. Moreover, in the clouds the concentration level of  $\text{S(VI, liq)}$  is two orders of magnitude higher than that of  $\text{S(IV, liq)}$  according to our simulation (not shown). This indicates that the oxidation of  $\text{S(IV, liq)}$  to  $\text{S(VI, liq)}$  (which is included in our model simulations) is an important factor influencing the partitioning of  $\text{SO}_2$  between the gas and liquid phases, and it can result in higher fractions of  $\text{SO}_2$  than  $\text{NH}_3$  partitioning into the liquid phase, as shown in Fig. 67.

### **4.3.2.6 Wet removal of $\text{NH}_3$ and $\text{SO}_2$ through aqueous $\text{NH}_4^+$ and $\text{S(VI)}$ in the ASM region**

Being very soluble and reactive in water,  $\text{NH}_3$  and  $\text{SO}_2$  partitioned into the cloud droplets can be efficiently removed from the atmosphere by wet deposition of their aqueous reaction products  $\text{NH}_x(\text{liq})$  and  $\text{SO}_x(\text{liq})$ . As for  $\text{NH}_x(\text{liq})$  and  $\text{SO}_x(\text{liq})$  in the clouds mentioned above (Sect. 4.1.3.5),  $\text{NH}_x(\text{liq})$  and  $\text{SO}_x(\text{liq})$  in ~~rain~~ the precipitation downdrafts are also mainly (>99%) in the form of  $\text{NH}_4^+(\text{liq})$  and  $\text{SO}_4^{2-}(\text{liq})$  plus  $\text{HSO}_4^-(\text{liq})$ , respectively. Figure 78 shows the wet deposition fluxes of  $\text{NH}_x(\text{liq})$  and  $\text{SO}_x(\text{liq})$  averaged for JJA over the years 2010-2020, respectively. Strong wet deposition fluxes of  $\text{NH}_x(\text{liq})$  and  $\text{SO}_x(\text{liq})$  occur in the polluted regions, such as northern India and eastern China, corresponding to the large emission rates of  $\text{NH}_3$  and  $\text{SO}_2$  there (Fig. S2). They are also affected by the precipitation rate and spatial distribution (Fig. S1c). For example, the simulated  $\text{SO}_x(\text{liq})$  wet deposition fluxes are negligible in Saudi Arabia due to extremely low precipitation, although the  $\text{SO}_2$  emissions are large in some areas there. The maximum wet deposition fluxes of both  $\text{NH}_x(\text{liq})$  and  $\text{SO}_x(\text{liq})$  are found on the southern and eastern slopes of the Tibetan Plateau, corresponding to the highest precipitation rates there.

Numerical studies have highlighted the very strong precipitation occurring along the southern slopes of the Tibetan Plateau and its role in the monsoon circulation (e.g., Bao and Li, 2020). This precipitation removes aerosols over the Tibetan Plateau by wet scavenging (Liu et al., 2023). Our model results indicate that in addition to the topographical block, the removal by wet scavenging along the southern slopes is very effective at reducing the amounts of  $\text{NH}_3$  and  $\text{SO}_2$  reaching the Tibetan Plateau (Fig. 1 and Fig. 78). Such scavenging is more effective for  $\text{SO}_2$  than  $\text{NH}_3$ , resulting much lower amounts of  $\text{SO}_2$  available for the deep convective transport from the lower troposphere of the Tibetan Plateau to the ASMA. While the deep convection frequency (shown in Fig. 2) is a dominant factor, the concentration levels of the tracers in the PBL (shown in Fig. 1) and scavenging efficiency are also important for determining the amount of the tracers transported into the ASMA (Fig. 45). This can explain the causes of the minima in both the deep convective transport tendency and volume mixing ratio of  $\text{SO}_2$  within the ASMA occurring over the southern Tibetan Plateau.

We quantify the effectiveness of precipitation in removing NH<sub>3</sub> and SO<sub>2</sub> by comparing their atmospheric lifetimes ~~against~~ ~~with~~ wet deposition, calculated as the tropospheric vertical column density (below 10 km, ~~see Fig. S8~~) divided by the wet deposition flux for each trace gas (~~Fig. 8~~). Figure ~~89~~ shows the mean atmospheric lifetime of tropospheric NH<sub>3</sub> and SO<sub>2</sub> against wet deposition in JJA during 2010-2020. It can be seen that over the southern parts and slopes of the Tibetan Plateau, the lifetime against wet deposition is around 1-2 days for NH<sub>3</sub> and less than 1 day for SO<sub>2</sub>, ~~respectively~~. The lifetime of SO<sub>2</sub> ~~against wet deposition~~ is shorter than that of NH<sub>3</sub> ~~against wet deposition~~ over northern India, the Tibetan Plateau and its southern slopes, and vice versa over eastern China. Such a turnaround could be due to differences in the relative amounts of tropospheric SO<sub>2</sub> and NH<sub>3</sub> over these regions. The ratio of the tropospheric vertical column density of NH<sub>3</sub> to that of SO<sub>2</sub> appears to be much larger over northern India than over eastern China (see Fig. ~~S58~~). The NH<sub>3</sub> column over northern India is the largest over the globe as reported in a previous study (Wang et al., 2020). It should be noted that NH<sub>3</sub> and SO<sub>2</sub> are alkaline and acidic gases, tending to neutralize each other to form NH<sub>4</sub><sup>+</sup>(liq) and SO<sub>4</sub><sup>2-</sup>(liq) or HSO<sub>4</sub><sup>-</sup>(liq) in the clouds, respectively. The large and excess amounts of NH<sub>3</sub> relative to SO<sub>2</sub> over northern India not only favour the deep convective transport of more NH<sub>3</sub> from the PBL to the ASMA, but also limit the amounts of SO<sub>2</sub> transported to the Tibetan Plateau by enhancing its partitioning into the clouds and removal by wet deposition.

#### 5-4 Conclusions

We have investigated the deep convective transport of trace gases CO, NH<sub>3</sub> and SO<sub>2</sub> from the planetary boundary layer (PBL) to the Asian summer monsoon anticyclone (ASMA) using the global atmospheric chemistry and climate model EMAC at T63L90 resolution. The model simulation was performed for the period from January 2010 to December 2020, and for this study 11 years of the seasonally averaged results for June–August (JJA) have been analyzed. The focus is on the similarities and differences in the transport efficiency of CO, NH<sub>3</sub>, and SO<sub>2</sub> from the PBL, in particular from the South Asian PBL, to the ASMA.

By analysing the spatial distributions of convective cloud top height, ~~we our model simulations~~ show that the frequency of deep convection reaching into the ASMA (~~defined as having a lower boundary at 10 km~~) is the highest (up to ~~2010%~~ ~~at 14 km~~) over the Tibetan Plateau among the continental Asian summer monsoon regions. Such a high deep convection frequency favours the efficient transport of trace gases from the lower troposphere of the Tibetan Plateau to the ASMA. ~~Here for the first time, w~~We quantify the deep convective transport efficiency of the trace gases CO, NH<sub>3</sub> and SO<sub>2</sub> into the ASMA by the ratio of the updraft mass flux of trace gas at the lower boundary of the ASMA (i.e., 10 km height) to its maximum near the convective cloud base. The results show that the deep convective transport efficiency is very high (above 50%) over the Tibetan Plateau for all three trace gases considered, with maximum values of about 80%-90% for CO, 60%-70% for NH<sub>3</sub>, and 60%-80% for SO<sub>2</sub>. Over the Indo-Gangetic plane, the deep convective transport efficiency is 20%-30% for CO, 2%-4% for NH<sub>3</sub>, and less than 1% for SO<sub>2</sub>.

Our model simulations show that the strongest deep convective transport tendency within the ASMA tends to occur over northern India and the southern edge of the Tibetan Plateau for CO (0.2-0.5 ppbv hr<sup>-1</sup>), over the southern and eastern parts of

the Tibetan Plateau for  $\text{NH}_3$  (0.02-0.05 ppbv  $\text{hr}^{-1}$ ), and over central India and eastern China for  $\text{SO}_2$  (0.002-0.005 ppbv  $\text{hr}^{-1}$ ). These spatial distribution patterns of deep convective transport tendency match well with those of the volume mixing ratios of CO,  $\text{NH}_3$  and  $\text{SO}_2$  within the ASMA, respectively. While the enhancements of CO and  $\text{NH}_3$  within the ASMA are evident, the enhancements of  $\text{SO}_2$  are very weak. In contrast to CO and  $\text{NH}_3$ , we find a decrease in  $\text{SO}_2$  over the southern Tibetan Plateau relative to the surroundings within the ASMA.

We find considerable amounts of gaseous  $\text{NH}_3$  and  $\text{SO}_2$  at heights above 6 km over the Tibetan Plateau partitioning into the clouds before reaching the ASMA, with the relative contributions of the 6-10 km column in the clouds (liquid-phase) to the total (gas-phase plus liquid-phase) of about 10-30% for  $\text{NH}_3$  (in the form of  $\text{NH}_4^+(\text{liq})$ ) and 50-80% for  $\text{SO}_2$  (mainly in the form of  $\text{SO}_4^{2-}(\text{liq})$  and  $\text{HSO}_4^-(\text{liq})$ ), respectively. Our model simulations show that the removal by wet deposition on the southern slopes plays an important role in reducing the amounts of  $\text{NH}_3$  and  $\text{SO}_2$  reaching the Tibetan Plateau platform. Such scavenging is more effective for  $\text{SO}_2$  than for  $\text{NH}_3$ , with lifetimes of 1-2 days for  $\text{NH}_3$  and less than 1 day for  $\text{SO}_2$ . We argue that the large and excess amounts of  $\text{NH}_3$  relative to  $\text{SO}_2$  over northern India not only favour the deep convective transport of more  $\text{NH}_3$  from the PBL to the ASMA, but also limit the amounts of  $\text{SO}_2$  transported to the Tibetan Plateau and then the ASMA by enhancing its partitioning into the clouds and removal by wet deposition.

The ASMA has unique physical-chemical features, influenced by deep convective transport of pollutants from the Asian PBL, with the Tibetan Plateau acting as a well-defined conduit. In this study, our model simulations, together with satellite-based observational data analysis, further demonstrate the role of deep convection over the Tibetan Plateau in effectively transporting CO, an insoluble reactive gas, from the Southern Asian PBL to the ASMA. Our model simulations show that deep convective transport to the ASMA over the Tibetan Plateau is also effective for  $\text{NH}_3$ , but ineffective for  $\text{SO}_2$ . However, these model results for  $\text{NH}_3$  and  $\text{SO}_2$ , the two soluble reactive gases, are not entirely supported by the satellite observations. The causes of the discrepancy between model simulations and satellite observations of  $\text{NH}_3$  and  $\text{SO}_2$  in the ASMA cannot be determined in this study, posing a challenge for modeling and remote sensing of these two gases in future research. Moreover, deep convection is generally accompanied by lightning activity, which can release  $\text{NO}_x$  and further produce  $\text{HNO}_3$  through oxidation reactions. While  $\text{NH}_3$  is alkaline,  $\text{SO}_2$  and  $\text{HNO}_3$  are acidic-gases, and they are all gaseous precursors of aerosols in the ATAL<sub>2</sub>. Ongoing changes in emissions of  $\text{NH}_3$ ,  $\text{SO}_2$  and  $\text{NO}_x$  and their interactions through multi-phase chemistry during deep convective transport may have significant impacts on the chemical characteristics of ASMA, which require continued investigation.

**Data availability.** The DaYu-GCP dataset, which is stored in the Network Common Data Format (NetCDF), is freely available on the Science Data Bank at <https://doi.org/10.57760/sciencedb.26292> (Zhao et al., 2026). The MLS CO data are available at <http://mirador.gsfc.nasa.gov>. The MIPAS  $\text{NH}_3$  and  $\text{SO}_2$  dataset is available upon request from Michael Höpfner or at <http://www.imk-asf.kit.edu/english/308.php>. The usage of MESSy (Modular Earth Submodel System) and access to the source code is licensed to all affiliates of institutions which are members of the MESSy Consortium. Institutions can become members

of the MESSy Consortium by signing the “MESSy Memorandum of Understanding”. More information can be found on the MESSy Consortium website: <http://www.messy-interface.org> (last access: 3 November 2025). The code used in this study has been based on MESSy version 2.55 and is archived with a restricted-access DOI (<https://doi.org/10.5281/zenodo.8379120>, The MESSy Consortium, 2023). The data produced in the study are available from the authors upon request.

5 **Supplement.** The supplement related to this article is available online.

**Author contributions.** JM, QH and JL initiated the project. JM performed the model simulations and analyzed the data. BC, QH, XY, GW, ~~and SC~~, and MH contributed to the data analysis and model evaluation. BS, CB, HT, AP, and JL contributed to the model simulation. JM prepared the manuscript with contributions from all co-authors.

**Competing interests.** At least one of the (co-)authors is a member of the editorial board of Atmospheric Chemistry and Physics.  
10 The authors have no other competing interests to declare.

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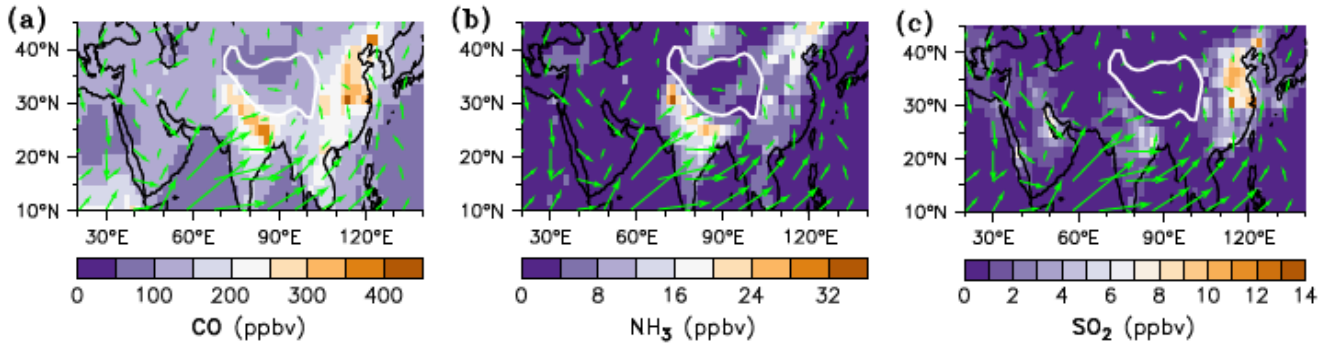
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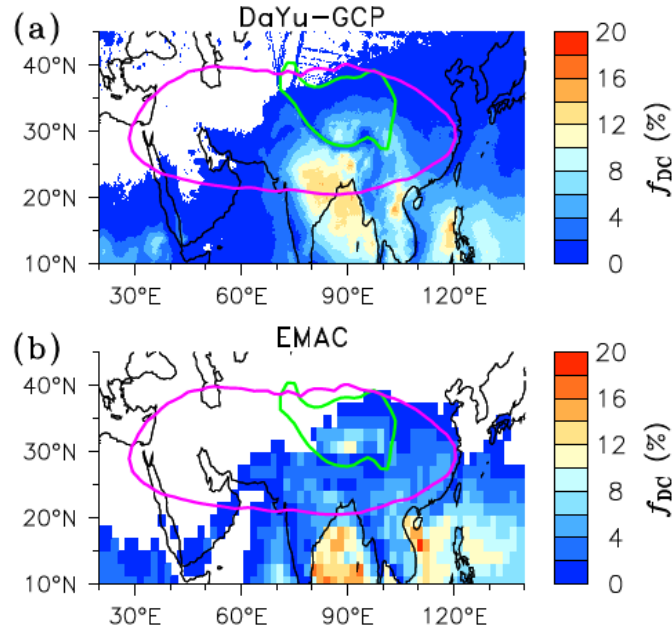
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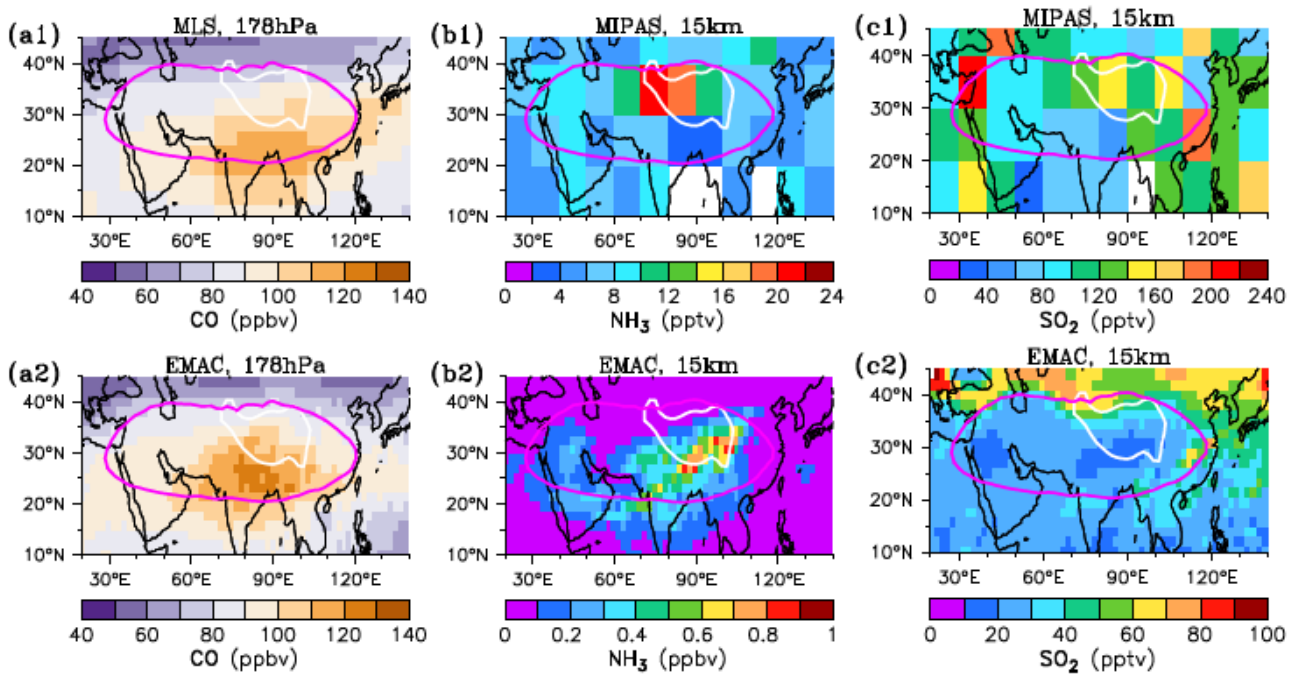
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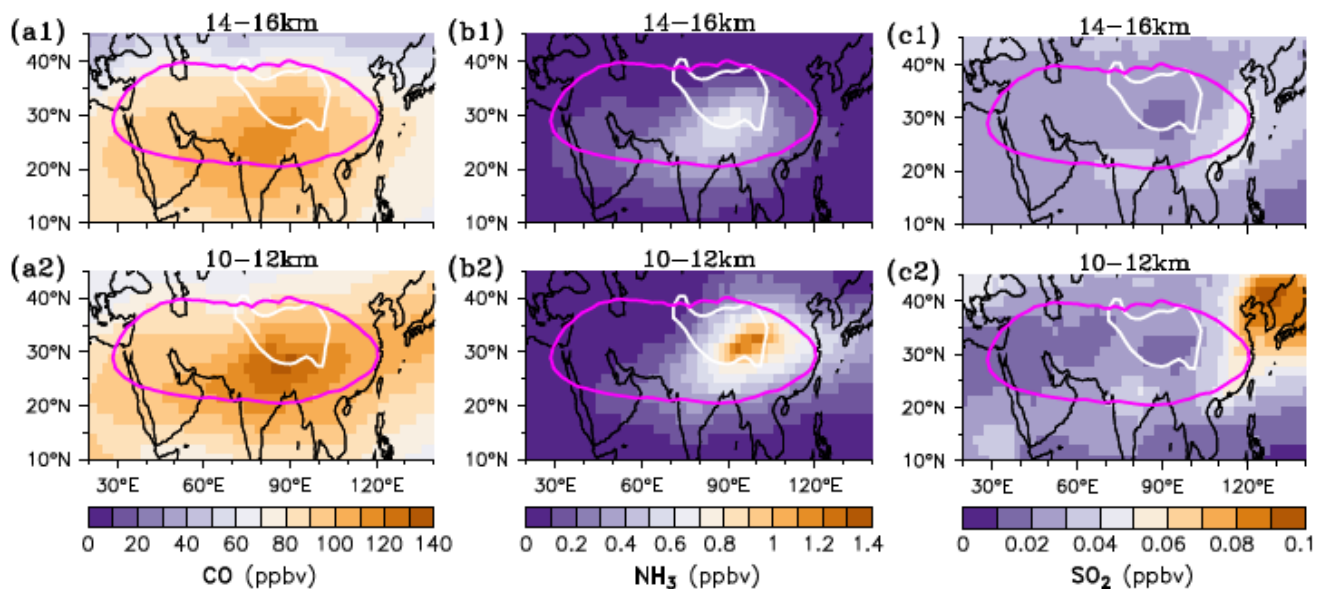
**Figure 1.** EMAC simulated volume mixing ratios of CO (a), NH<sub>3</sub> (b) and SO<sub>2</sub> (c) overlaid with the wind vectors in the PBL, averaged for JJA over the years 2010-2020. White lines represent the 3 km terrain height contour, highlighting the Tibetan Plateau.



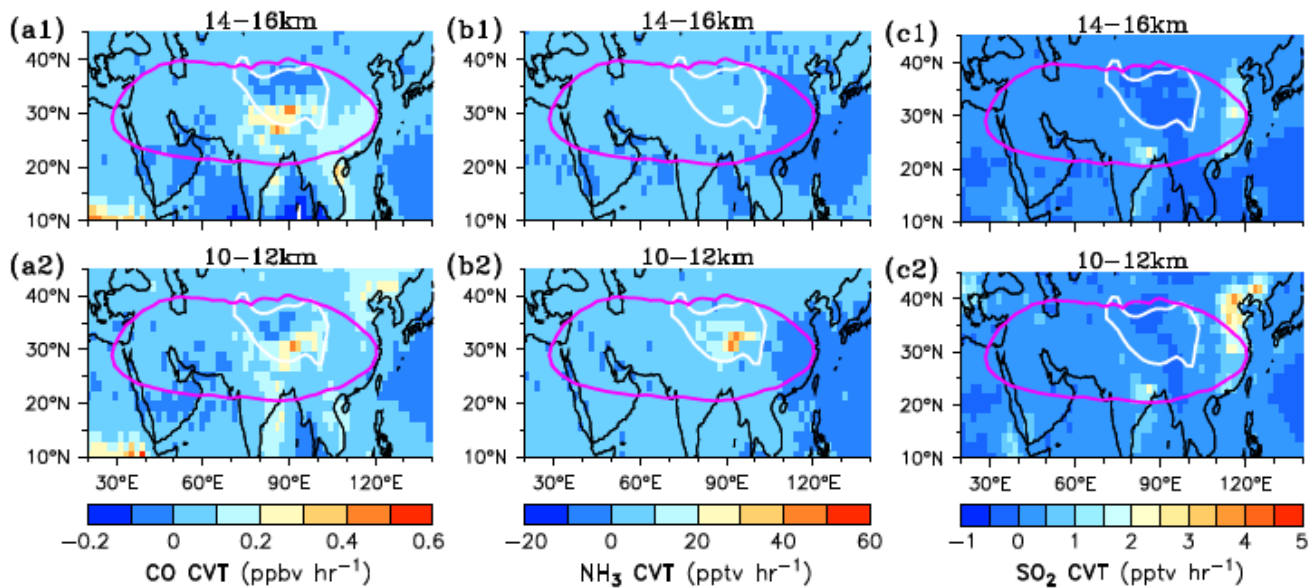
**Figure 2.** Comparison of EMAC simulated relative deep convection frequency ( $f_{DC}$  in percent) for ~~selected~~ convective cloud top heights, i.e., reaching above ~~16 km (a)~~, 14 km above sea level with DaYu-GPC satellite data (a)(b) ~~12 km (c)~~, and 10 km (d) above sea level, in JJA during the years 2010-2020. Purple lines are the 16.64 km geopotential height contour at 100 hPa, highlighting the main ASMA area (see Figure S1). Green lines represent the 3 km terrain height contour, highlighting the Tibetan Plateau.



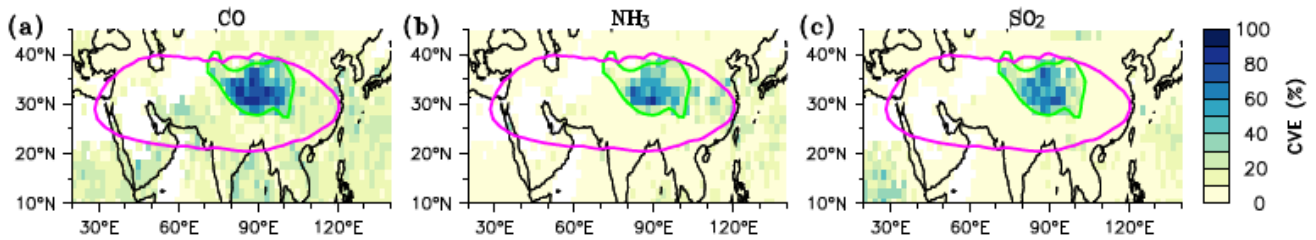
**Figure 3.** Comparisons of EMAC simulated cloud-free CO (**a2**) with MLS observed CO (**a1**) at 178 hPa for JJA over the years 2010-2020 and EMAC simulated cloud-free NH<sub>3</sub> (**b2**) and SO<sub>2</sub> (**c2**) with MIPAS observed NH<sub>3</sub> (**b1**) and SO<sub>2</sub> (**c1**) at 15 km above sea level for JJA 2010-2011. Purple lines show the 16.64 km geopotential height contour at 100 hPa, highlighting the main ASMA area. White lines represent the 3 km terrain height contour, highlighting the Tibetan Plateau. White grid cells in (**b1**) and (**c1**) indicate missing data in the MIPAS dataset due to cloud interference.



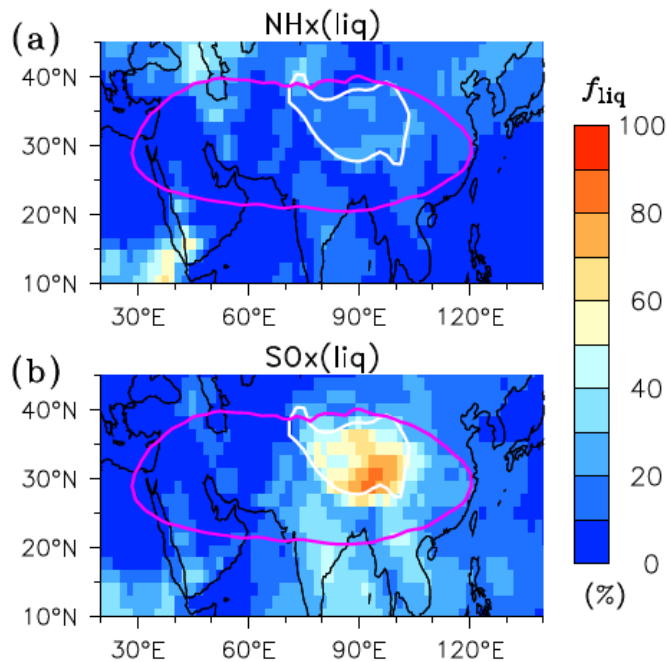
**Figure 34.** EMAC simulated volume mixing ratios of CO (**a1** and **a2**),  $\text{NH}_3$  (**b1** and **b2**) and  $\text{SO}_2$  (**c1** and **c2**) at selected altitudes, i.e., 14-16 km (**a1**, **b1** and **c1**) and 10-12 km (**a2**, **b2** and **c2**) above sea level, averaged for JJA over the years 2010-2020. Purple lines show the 16.64 km geopotential height contour at 100 hPa, highlighting the main ASMA area (see Figure S1). White lines represent the 3 km terrain height contour, highlighting the Tibetan Plateau.



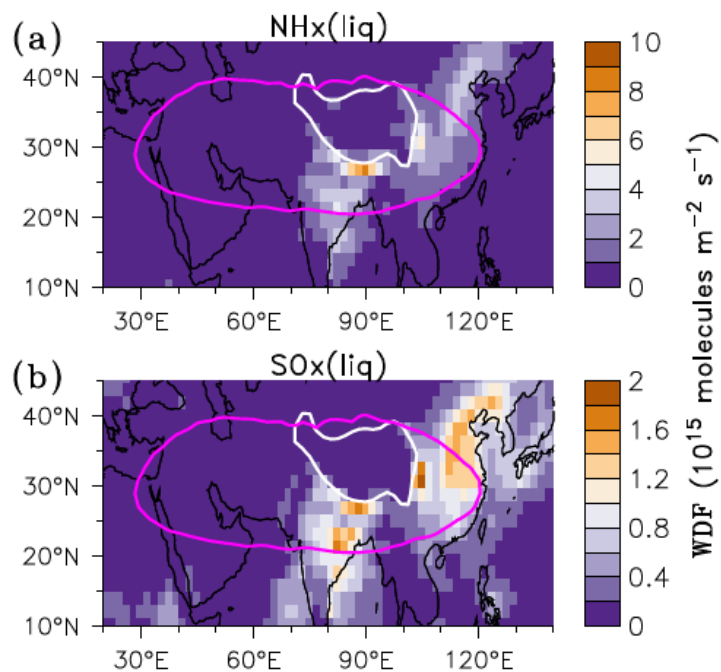
**Figure 45.** EMAC simulated mean deep convective transport tendency (CVT) of CO (**a1** and **a2**), NH<sub>3</sub> (**b1** and **b2**) and SO<sub>2</sub> (**c1** and **c2**) at selected altitudes, i.e., 14-16 km (**a1**, **b1** and **c1**) and 10-12 km (**a2**, **b2** and **c2**) above sea level, averaged for JJA over the years 2010-2020. Purple lines show the 16.64 km geopotential height contour at 100 hPa, highlighting the main ASMA area (see Figure S1). White lines represent the 3 km terrain height contour, highlighting the Tibetan Plateau.



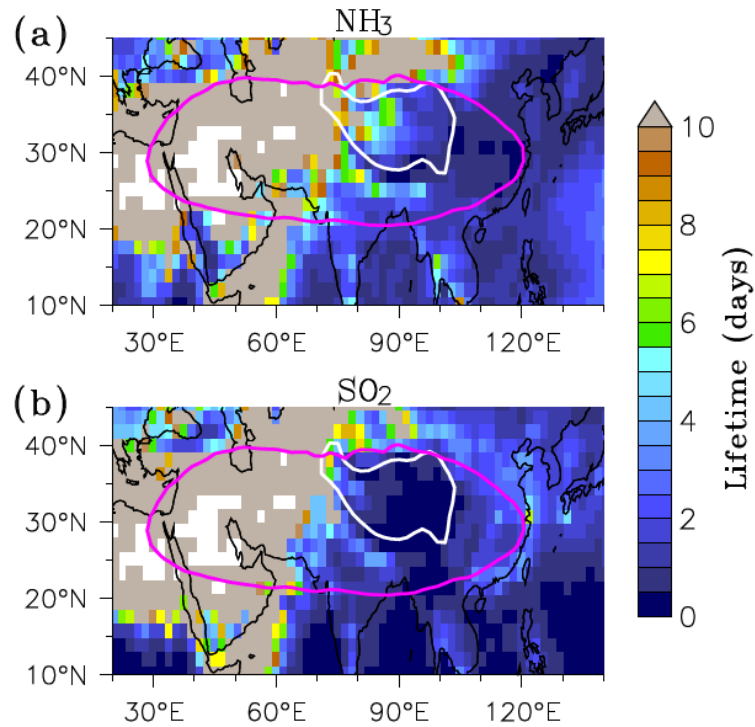
**Figure 56.** EMAC simulated mean averages of deep convective transport efficiency (CVE), i.e., the ratio of the updraft mass flux (UMF) at 10 km height above sea level to its maximum in the deep each convection column (expressed in percent), for CO (a), NH<sub>3</sub> (b) and SO<sub>2</sub> (c), in JJA during the years 2010-2020. Purple lines show the 16.64 km geopotential height contour at 100 hPa, highlighting the main ASMA area (see Figure S1). Green lines represent the 3 km terrain height contour, highlighting the Tibetan Plateau.



**Figure 67.** EMAC simulated mean relative contributions of the liquid-phase amount to the total (gas-phase plus liquid-phase) within a vertical column of 6-10 km above sea level ( $f_{liq}$ ), for NH<sub>3</sub> and its reaction products in the clouds (denoted by NH<sub>x</sub>) (a) and SO<sub>2</sub> and its reaction products in the clouds (denoted by SO<sub>x</sub>) (b), in JJA during the years 2010-2020. Purple lines show the 16.64 km geopotential height contour at 100 hPa, highlighting the main ASMA area (see Figure S1). White lines represent the 3 km terrain height contour, highlighting the Tibetan Plateau.



**Figure 78.** EMAC simulated wet deposition fluxes (WDF) of  $\text{NH}_3$  and its reaction products in the rains-precipitating downdrafts (denoted by  $\text{NH}_x$ ) (a) and  $\text{SO}_2$  and its reaction products in the rains-precipitating downdrafts (denoted by  $\text{SO}_x$ ) (b), averaged for JJA over the years 2010-2020. Purple lines show the 16.64 km geopotential height contour at 100 hPa, highlighting the main ASMA area (see Figure S1). White lines represent the 3 km terrain height contour, highlighting the Tibetan Plateau.



**Figure 89.** EMAC simulated mean atmospheric lifetimes of tropospheric  $\text{NH}_3$  (a) and  $\text{SO}_2$  (b) (below 10 km height above sea level) against wet deposition in JJA during the years 2010-2020. Purple lines show the 16.64 km geopotential height contour at 100 hPa, highlighting the main ASMA area (see Figure S1). White lines represent the 3 km terrain height contour, highlighting the Tibetan Plateau.