



# Transport of aerosols into the UTLS and their impact on the Asian monsoon region as seen in a global model simulation

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Received: 17 October 2012 – Published in Atmos. Chem. Phys. Discuss.: 21 November 2012

Revised: 12 June 2013 – Accepted: 27 July 2013 – Published: 3 September 2013

**Abstract.** An eight-member ensemble of ECHAM5-HAMMOZ simulations for a boreal summer season is analysed to study the transport of aerosols in the upper troposphere and lower stratosphere (UTLS) during the Asian summer monsoon (ASM). The simulations show persistent maxima in black carbon, organic carbon, sulfate, and mineral dust aerosols within the anticyclone in the UTLS throughout the ASM (period from July to September), when convective activity over the Indian subcontinent is highest, indicating that boundary layer aerosol pollution is the source of this UTLS aerosol layer. The simulations identify deep convection and the associated heat-driven circulation over the southern flanks of the Himalayas as the dominant transport pathway of aerosols and water vapour into the tropical tropopause layer (TTL). Comparison of model simulations with and without aerosols indicates that anthropogenic aerosols are central to the formation of this transport pathway. Aerosols act to increase cloud ice, water vapour, and temperature in the model UTLS. Evidence of ASM transport of aerosols into the stratosphere is also found, in agreement with aerosol extinction measurements from the Halogen Occultation Experiment (HALOE) and Stratospheric Aerosol and Gas Experiment (SAGE) II. As suggested by the observations, aerosols are transported into the Southern Hemisphere around the tropical tropopause by large-scale mixing processes. Aerosol-induced circulation changes also include a weakening of the main branch of the Hadley circulation and a reduction of monsoon precipitation over India.

## 1 Introduction

East Asia is one of the largest source regions of gaseous and aerosol pollutants, having both high anthropogenic emissions and important natural sources such as wildfires and dust storms (Park et al., 2010; Jeong et al., 2011). Developing nations of East Asia are experiencing a dramatic increase in the levels of aerosol pollution because of the rapid industrialization (Liu and Diamond, 2005; Venkataraman et al., 2005). The increase in atmospheric concentrations of anthropogenic aerosols (including black carbon (BC), organic carbon (OC), and sulfate ( $\text{SO}_4^{2-}$ )) from such sources as transportation, industry, agriculture, and urban land development affect weather and climate (IPCC, 2007). Absorbing aerosols such as dust and BC heat the atmosphere due to short-wave absorption. Non-absorbing aerosols such as  $\text{SO}_4^{2-}$  cause surface cooling by scattering solar radiation, and have a relatively small heating effect on the atmosphere itself. Recent studies suggest that increased aerosol loading may change the energy balance in the atmosphere and at the Earth's surface, as well as alter the global water cycle (IPCC, 2007). Ramanathan et al. (2005) reported that the polluted layer of BC, OC, and dust over the Asian monsoon region leads to a weakening of the summer monsoon rainfall based on ensemble simulations. However, Lau et al. (2006) argued that absorbing aerosols over the elevated Tibetan Plateau, with its high surface albedo, should intensify the Indian summer monsoon through the increased heat pump effect.

Numerical experiments have suggested that atmospheric circulation anomalies induced by BC may be a cause of

drought over northern China and of excessive rainfall over southern China and India (Menon et al., 2002). These studies indicate that aerosol effects can induce large changes in precipitation patterns, which in turn may change climate on a regional or even global scale.

Aerosol-induced circulation changes might furthermore affect the vertical transport of pollution from the surface into the upper troposphere and lower stratosphere (UTLS). Recent satellite observations provide evidence that the Asian summer monsoon (ASM) circulation provides a pathway for pollution transport into the stratosphere (Randel et al., 2010) with potential implication for stratosphere dynamics. A persistent maximum of tropospheric chemical constituents ( $\text{H}_2\text{O}$ ,  $\text{CO}$ ,  $\text{C}_2\text{H}_6$ ,  $\text{CH}_4$ ,  $\text{N}_2\text{O}$ ,  $\text{HCN}$ , and aerosols) has been observed inside the ASM anticyclone in the UTLS during boreal summer (Park et al., 2004; Li et al., 2005; Randel and Park, 2006; Fu et al., 2006; Xiong et al., 2009; Randel et al., 2010). These results have been confirmed by modelling studies (Li et al., 2005; Park et al., 2007; Randel et al., 2010; Chen et al., 2012). Recently, a layer of aerosols at the tropopause during the Asian monsoon has been observed in CALIPSO lidar measurements (Vernier et al., 2011). The UTLS aerosol layer extends from the eastern Mediterranean to western China and vertically from 13 to 18 km, indicating the role of deep convection associated with the monsoon in transporting aerosols in the upper troposphere.

Observations and modelling studies suggest that the occurrence of aerosol layers near the tropopause could strongly affect the formation of cirrus clouds and precipitation (Li et al., 2005; Yin et al., 2005; Su et al., 2011). Liu et al. (2009) reported that the homogeneous freezing of  $\text{SO}_4^{2-}$  particles dominates cirrus cloud formation in the upper troposphere. Anthropogenic  $\text{SO}_4^{2-}$  results in a global annual mean change of long-wave cloud forcing (LWCF) of  $0.20 \pm 0.09 \text{ W m}^{-2}$  and short-wave cloud forcing (SWCF) of  $0.30 \pm 0.17 \text{ W m}^{-2}$  and an increase of UTLS water vapour by  $\sim 10\%$ . When both homogeneous and heterogeneous ice nucleation are taken into account, anthropogenic soot may increase global cirrus cloud cover by  $\sim 2\%$  and UTLS water vapour by 40% with a change in LWCF of  $1.5 \text{ W m}^{-2}$ . Su et al. (2011) hypothesized that aerosol semi-direct radiative heating and changes in cirrus radiative heating may contribute to the observed increase in tropical tropopause layer temperatures and to elevated water vapour concentrations in polluted clouds. Several other studies (e.g. Randall et al., 1989; Ramaswamy and Ramanathan, 1989; Liu et al., 2003a, b; Dodion et al., 2008) pointed out that cirrus clouds are likely to affect the intensity of the large-scale circulation in the tropics.

In the stratosphere, aerosols have a much longer residence time than in the troposphere, and they get dispersed over a larger area. Through their interaction with ultraviolet, visible, and infrared radiation, stratospheric aerosols are likely to play a significant role in the Earth's radiation budget and climate (Dodion et al., 2008). Simulations of stratospheric

geo-engineering with  $\text{SO}_4^{2-}$  (Heckendorn et al., 2009) or BC aerosols (Kravitz et al., 2012) show that continuous injection of aerosol into the lower stratosphere would cause large changes in UTLS temperatures.

As can be seen from the above discussions the ASM plays a dominant role in the transport of boundary layer pollution into the UTLS. Fu et al. (2006) provided observational evidence that the region poleward of  $25^\circ \text{N}$  encompassing the southern slopes of the Himalayas and the Tibetan Plateau provides the main pathway for cross-tropopause transport. In contrast, Park et al. (2009) concluded based on CTM simulations that deep monsoon convection south of  $25^\circ \text{N}$  over the Bay of Bengal and extending to the South China Sea is the primary entry point into the UTLS. According to their analysis, convection in this region transports boundary layer pollutants up to  $\sim 200 \text{ hPa}$ , and a fraction of the air is lifted to 16 km by large-scale upward motion on the eastern side of the anticyclone. These differing pictures of the transport pathways indicate that there is need for additional research to increase our understanding. The purpose of the study presented here is to use a GCM with fully coupled aerosol effects on convection and radiative transfer to investigate the impact of aerosols on transport and associated changes in the energy balance and the hydrological cycle in the ASM region.

We employ the state-of-the-art ECHAM5-HAMMOZ aerosol–chemistry–climate model (Pozzoli et al., 2008a, 2011) to simulate BC, OC (OC), mineral dust, and  $\text{SO}_4^{2-}$  aerosols during an Asian summer monsoon season. The model results are evaluated through comparison with HALOE and SAGE II data. To understand the effect of aerosols, the difference between model simulations with fully interactive aerosols (CTRL) and with only passively transported aerosol (NOAER) are analysed. The paper is organized as follows. The ECHAM5-HAMMOZ model and satellite data (SAGE II and HALOE) are described in Sect. 2. The influence of ASM convection on the distribution of aerosols in the UTLS is discussed in Sect. 3, and impact of aerosols on cloud ice, temperature, water vapour, and changes in circulation and precipitation are presented in Sect. 4. Section 5 concludes this study.

## 2 Model simulations and data analysis

### 2.1 ECHAM5-HAMMOZ model simulation and experimental setup

The ECHAM5-HAMMOZ aerosol–chemistry–climate model used in the present study comprises the general circulation model ECHAM5 (Roeckner et al., 2003), the tropospheric chemistry module, MOZ (Horowitz et al., 2003), and the aerosol module, the Hamburg Aerosol Model (HAM) (Stier et al., 2005). The tropospheric chemistry module MOZ and the aerosol module HAM are fully

interactive and implemented together in ECHAM5 (Pozzoli et al., 2008a). The HAM module takes into account the major aerosol compounds, namely  $\text{SO}_4^{2-}$ , BC, OC, sea salt, and mineral dust. It represents aerosols as internal and external mixtures with four soluble and three insoluble modes (Vignati et al., 2004). Details of the aerosol categorization and their parameterization schemes are documented by Stier et al. (2005). The chemical scheme used in the tropospheric chemistry module, MOZ, is identical to the MOZART-2 model with small modifications as described by Pozzoli et al. (2008a). It includes 63 tracers and 168 reactions to represent  $\text{O}_x$ – $\text{NO}_x$ –hydrocarbon chemistry.

We used the RETRO project data set of the year 2000 (<http://www.retro.enes.org/>) for the surface  $\text{CO}$ ,  $\text{NO}_x$ , and hydrocarbon emissions from anthropogenic sources and biomass burning (Schultz et al., 2008). The anthropogenic and fire emissions of  $\text{SO}_2$ , BC, and OC are based on the AEROCOM emission inventory (Dentener et al., 2006) representative of the year 2000. DMS, predominantly emitted from the oceans, is a  $\text{SO}_4^{2-}$  aerosol precursor. Its emissions depend on seawater DMS concentrations associated with phytoplankton blooms (Kettle and Andreae, 2000) and model surface wind speed, which determines the DMS sea–air exchange (Nightingale et al., 2000). Terrestrial biogenic DMS emissions follow Pham et al. (1995). The emissions of sea salt are based on Schulz et al. (2004). Mineral dust emissions are calculated online using the ECHAM5 wind speed, hydrological parameters (e.g. soil moisture), and soil properties, following the work of Tegen et al. (2002) and Cheng et al. (2008). The anthropogenic and natural emissions are also described in detail by Pozzoli et al. (2008a, b).

The ECHAM5, HAM, and MOZ model performance against observations have been presented in earlier studies (e.g. Stier et al., 2005; Auvray et al., 2007; Pozzoli et al., 2008a, b, 2011). All of these studies found good agreement between simulated and observed  $\text{SO}_4^{2-}$ , BC, and OC surface concentrations in several world regions.

The simulations described in this study were performed using the coupled model ECHAM5–HAMMOZ with a spectral resolution of T42 corresponding to about  $2.8^\circ \times 2.8^\circ$  in the horizontal dimension. The vertical resolution of the model is described by 31 hybrid  $\sigma$ – $p$  levels from the surface up to 10 hPa, which corresponds to layer heights of  $\sim 50$  m near the surface and  $\sim 1$  km near the tropopause. Time step length was 20 min. AMIP sea surface temperature (SST) and sea ice cover (SIC) from 2003 were used as lower boundary conditions. Each run covered the time period from June to September with two months of spin-up time. In order to achieve more robust results, we performed eight-member ensemble runs by varying the initial conditions (both SST and SIC) between 24 and 31 March 2003.

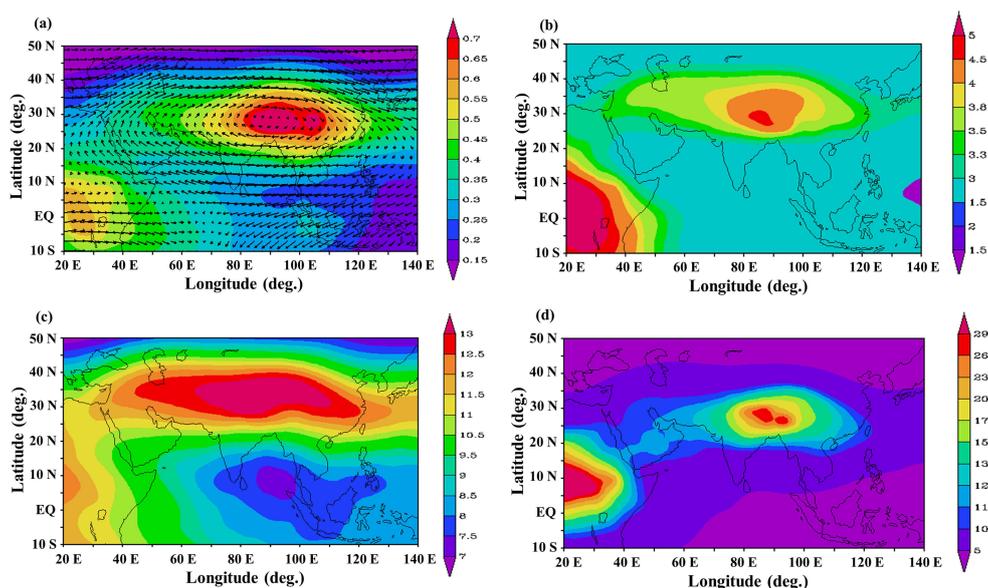
We compare two sets of results: (1) the baseline simulations (CTRL), where the simulated aerosol concentrations are used in the calculation of the radiative budget and of the cloud droplet (CDNC, Lohmann et al., 1999;

Lin and Leaitch, 1997) and the ice crystal (ICNC Kärcher and Lohmann, 2002) number concentrations; and (2) the NOAER simulation, where the standard ECHAM5 cloud scheme is used (Lohmann and Roeckner, 1996; Roeckner et al., 2003) and aerosols are turned off. The parameterizations for precipitation formation in warm clouds, cold clouds, and mixed-phase clouds are discussed in detail in Sect. 10.3.4 of the MPI-Report No. 349 “The atmospheric general circulation model ECHAM5, Part I” ([http://www.mpimet.mpg.de/fileadmin/publikationen/Reports/max\\_scirep\\_349.pdf](http://www.mpimet.mpg.de/fileadmin/publikationen/Reports/max_scirep_349.pdf)).

## 2.2 Satellite observations: SAGE II and HALOE

To evaluate the model results we have analysed the distribution of aerosol extinction from two satellite-based instruments: Stratospheric Aerosol and Gas Experiment II (SAGE II) on board the Earth Radiation Budget Satellite (ERBS), and Halogen Occultation Experiment (HALOE) on board the Upper Atmospheric Research Satellite (UARS). HALOE is a limb-viewing solar occultation instrument that obtains transmittance profiles in the infrared region of the spectrum giving 15 sunrise and 15 sunset measurements each day (Russell et al., 1993). HALOE was operational from September 1991 to November 2005. The vertical resolution is  $\sim 2$  km or less. Temperature and aerosols are retrieved at four wavelengths (2.45, 3.40, 3.46, 5.26  $\mu\text{m}$ ). The HALOE aerosol retrievals are described in detail by Hervig et al. (1995). The validation of these measurements suggests uncertainties of  $\sim 15$ – $20\%$  (Hervig et al., 1995). Aerosol extinction at 5.26  $\mu\text{m}$  is analysed to study transport of aerosols in the UTLS region. HALOE data are available at <http://haloe.gats-inc.com/download/index.php>.

SAGE II is also a solar occultation instrument where measurements are only made during limb-viewing conditions, providing 15 sunrise and 15 sunset measurements per day. During each sunrise and sunset encountered by the orbiting spacecraft, the instrument uses the solar occultation technique, measuring the attenuated solar radiation through the Earth’s limb in seven channels with central wavelengths ranging from 0.385 to 0.1020  $\mu\text{m}$ . The transmittance measurements are inverted using the “onion-peeling” approach (Antuña et al., 2002) to retrieve the aerosol extinction coefficient ( $\text{km}^{-1}$ ). SAGE II has a horizontal resolution of the order of 200 km and a vertical resolution of 1 km (Kent et al., 1998). The 0.525  $\mu\text{m}$  extinction data used here are based on the v6.2 retrieval algorithm. SAGE II aerosol data have been validated by Oberbeck et al. (1989) indicating an extinction uncertainty of  $\sim 20$ – $30\%$ . Temporal and spatial coverage of HALOE are similar to those of SAGE II (Terao and Logan, 2007). SAGE II data are available at [http://badc/sage2/data/version\\_6.2](http://badc/sage2/data/version_6.2).



**Fig. 1.** Horizontal structure of June–September 2003 averages at 110 hPa of aerosol concentrations ( $\text{ng m}^{-3}$ ): (a) BC and horizontal wind fields (shown as vectors), (b) OC, (c)  $\text{SO}_4^{2-}$ , and (d) mineral dust.

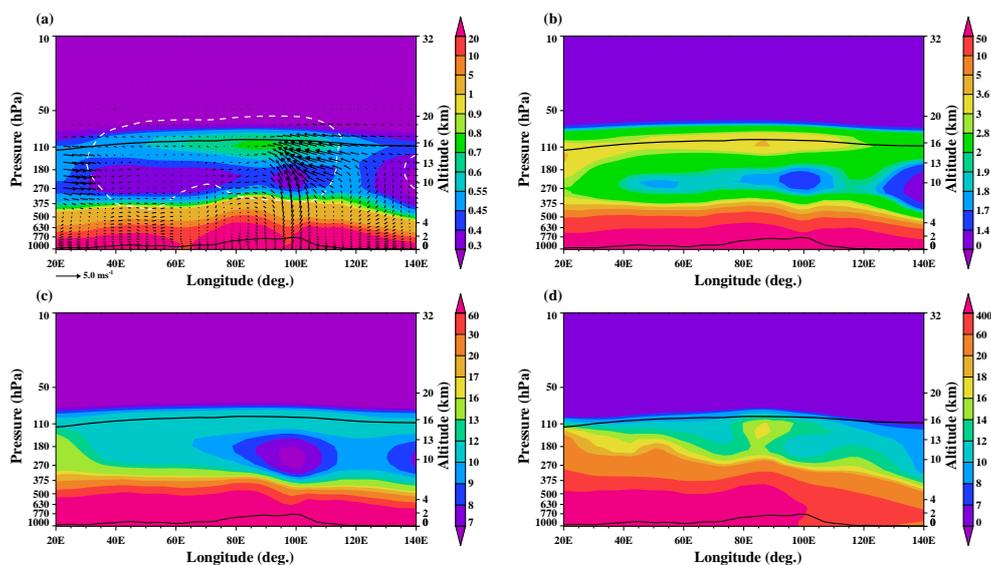
### 3 Transport aerosols into the UTLS region due to ASM convection

In order to study the influence of ASM convection on the distribution of aerosols in the UTLS, ensemble-average monthly mean concentrations of black BC,  $\text{SO}_4^{2-}$ , OC, and mineral dust aerosols from the ECHAM5-HAMMOZ ensemble are analysed during the summer season (June–September) of the year 2003. Figure 1 shows the seasonal mean distributions of BC,  $\text{SO}_4^{2-}$ , OC, and mineral dust aerosol, respectively, for the CTRL simulation at 110 hPa. Figure 1a also displays the average wind field at the same pressure level. The winds show a strong anticyclonic circulation between 20 and 120° E, 12 and 40° N collocated with a maximum in all aerosol fields. Confinement of aerosols within the anticyclonic circulation, with a pronounced maximum on its eastern part covering South East Asia and India is quite obvious.

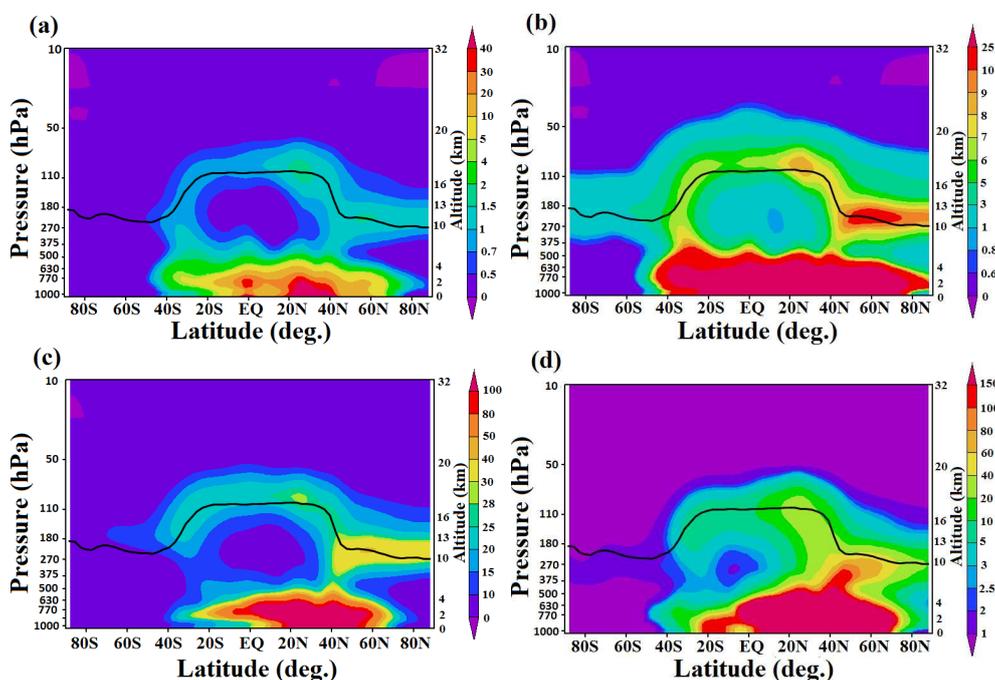
To illustrate vertical transport, Fig. 2 shows a seasonal mean longitude–pressure cross section averaged over 15–35° N and for June–September as obtained from CTRL simulation. In agreement with satellite observations (Dodion et al., 2008; Vernier et al., 2011), ECHAM5-HAMMOZ shows a layer of aerosols in the UTLS region ( $\sim$  between 416 and 70 hPa). Transport of boundary layer aerosols from 60 to 120° E to the UTLS region due to large-scale monsoon convection is evident. There are two regions of vertical transport eastward of 60° E: first at the eastern end of the anticyclone (around 85° E) and second around 120° E. This second region over the South China Sea is another pathway for aerosols into the UTLS, and it is likely that some of this aerosol is transported westward by the equatorial easterly

winds and is trapped in the anticyclone as identified by the aerosol maximum around 85° E and 110 hPa. The latitude–pressure cross sections (averaged over 60–120° E) of aerosol concentrations (BC, OC,  $\text{SO}_4^{2-}$ , and mineral dust) averaged during the Asian monsoon season are plotted in Fig. 3. Cross-tropopause transport (extending up to 70 hPa) is evident in all the aerosol distributions, but the details vary. The vertical distribution of aerosol concentrations does not vary much between individual months of the summer monsoon season.

A pronounced maximum of BC, OC,  $\text{SO}_4^{2-}$ , and mineral dust in the UTLS near 30° N is associated with the ASM anticyclone. High concentrations extend to the equator and are then transported poleward and downward in the Southern Hemisphere to  $\sim$  30° S by the Brewer–Dobson circulation, thereby forming an arch. Most of the transport appears to occur around the tropical tropopause, suggesting mixing by breaking Rossby waves in the lower tropical stratosphere and in the upper tropical tropopause layer (TTL). This is consistent with the results of Homeyer and Bowman (2013), who analysed ECMWF ERA-Interim Reanalysis data. Similar transport is also observed in ACE-FTS observations of HCN (Randel et al., 2010). From Figs. 2 and 3 one can also infer that large-scale vertical transport within the anticyclone is related to deep monsoon convection over the region 10–35° N, 60–120° E. Figure 2a shows vectors of the resolved circulation, which exhibits a strong upwelling around 100° E. This is a region of deep convection on the southern flanks of the Himalayas as can be seen from Fig. 4a and b, which shows the combined distribution of CDNC and ICNC. The upwelling shown in Fig. 2a is induced by convective heating, but does not represent the full transport experienced by



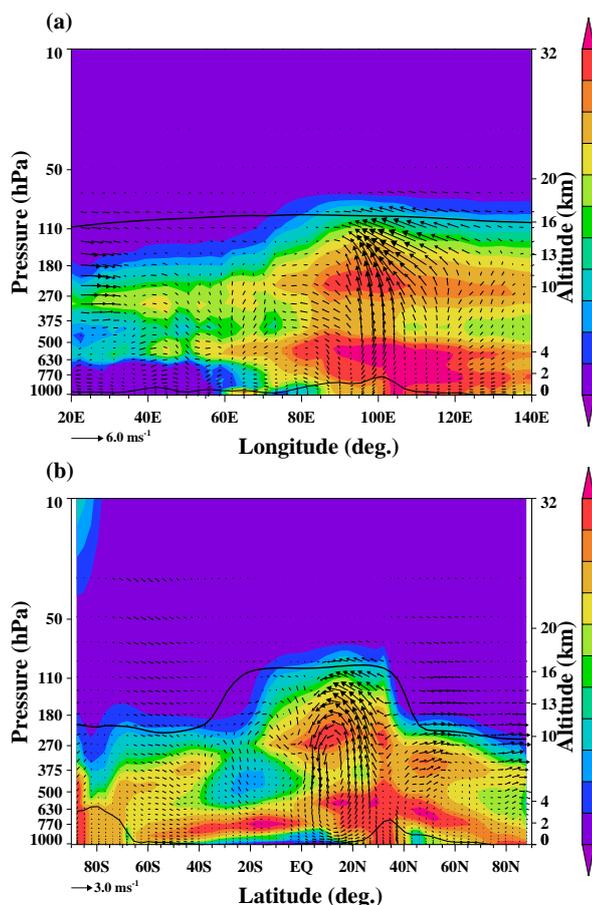
**Fig. 2.** “cross section” (averaged for June–September and 15–35° N) of concentrations ( $\text{ng m}^{-3}$ ) of (a) BC aerosols, location of the monsoon anticyclone indicated with dotted line, (b) OC aerosols, (c)  $\text{SO}_4^{2-}$ , and (d) mineral dust aerosols. Vectors of the circulation are shown in (a) with the vertical wind scaled by 300. The upper black line marks the latitudinally averaged tropopause showing increased height in the ASM region. The lower black line marks the topography. The dashed white line is the negative  $10^{-5}$  vorticity contour showing the approximate region of the anticyclone.



**Fig. 3.** “cross section” (averaged for June–September and 60–120° E) of concentrations ( $\text{ng m}^{-3}$ ) of (a) BC aerosols, (b) OC aerosols, (c)  $\text{SO}_4^{2-}$  aerosols, and (d) mineral dust aerosols. The black line marks the longitudinally averaged tropopause showing increased height in the ASM region.

aerosols and other tracers, which are subject to vertical redistribution by the sub-grid-scale deep convection parameterization. The distributions of the four aerosol species shown in Fig. 2 all have a minimum between 180 and 270 hPa around

100° E with a distinct trench extending to below 500 hPa. The aerosol values increase above 180 hPa, which suggests deep convective transport extending above 180 hPa acting to remove aerosols from lower altitudes. The resolved circulation



**Fig. 4.** Distribution of cloud droplet number concentration (CDNC) and ice crystal number concentration (ICNC) ( $1 \text{ mg}^{-1}$ ) averaged for June–September 2003. (a) Latitude average between  $20$  and  $30^\circ \text{ N}$  and (b) zonal average between  $60$  and  $120^\circ \text{ E}$ .

transports aerosols into the anticyclone core, where they accumulate (Fig. 2a) so that the UT maxima occur between  $80$  and  $90^\circ \text{ E}$ . There is also a convective transport pathway over the Tibetan Plateau poleward of  $40^\circ \text{ N}$  (see Fig. 4b), which is distinct from the one over the southern slopes.

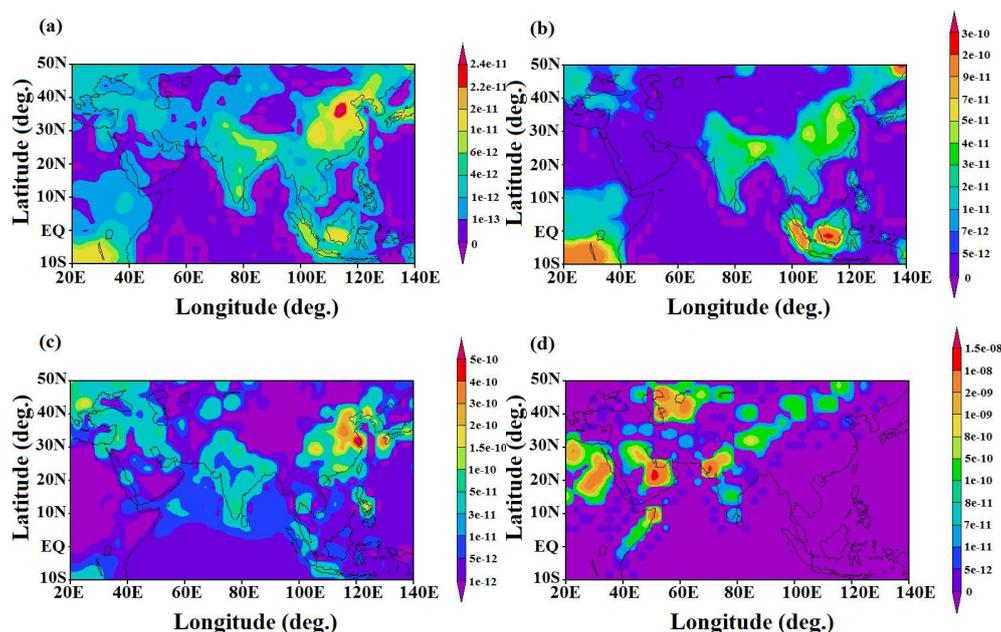
The latitude–altitude cross sections in Fig. 3 show that aerosols accumulate around the extratropical tropopause. It is likely that large-scale Rossby wave mixing processes are acting to transport some of these aerosols towards the equator (e.g. Scott and Cammas, 2002; Homeyer and Bowman, 2013).

The aerosol arch feature in the tropical tropopause region seen in Fig. 3 cannot be explained by direct convective transport. Overshooting convection accounts for only about 5% of the total deep convective activity in the tropics, including the ASM region (Alcala and Dessler, 2002). Most convection does not penetrate above the zero diabatic heating surface at about  $15 \text{ km}$  (e.g. Folkins and Martin, 2005). The transport of water and hence aerosols to  $100 \text{ hPa}$  and above between  $20$  and  $40^\circ \text{ N}$  is achieved via large-scale transport in

the ASM anticyclone (Gettelman et al., 2004; James et al., 2008), and this is the primary channel for injection into the lower stratosphere. As noted above, transport by synoptic-scale Rossby wave breaking towards the equator from the subtropics occurs around the  $380 \text{ K}$  isentrope in summer and fall. Mixing near the equator can occur through Kelvin waves (Fujiwara et al., 1998) and gravity waves (Moustaoui et al., 2004). Transport by synoptic-scale Rossby wave breaking near  $380 \text{ K}$  in the Southern Hemisphere during June–July–August–September (JJAS) occurs both poleward and equatorward in equal measure, as found by Homeyer and Bowman (2013). This will result in mixing of aerosols crossing the equator towards the Southern Hemisphere subtropics.

Figure 5 shows emissions of BC, OC, mineral dust, and sulfur over the Asian region and surrounding area. Sulfur emissions are primarily from coal-fired power plants. Asia is one of the highest sulfur-producing regions in the world (Vernier et al., 2011). The industrial and residential sectors emit BC and OC which are produced through incomplete combustion of coal and bio-fuel and also in wildland fires. These emissions are high over south west China and the Indo Gangetic Plain (IGP). Source regions of high mineral dust are in North Africa, the Taklimakan Desert of north-west China, part of Arabia, Iran, and the shores of the Caspian Sea. The low-level convergence during ASM may collect these pollutants from the aforementioned regions, and deep convective activity lifts them upward. As can be seen in Figs. 2 and 3, transport of aerosols into the UTLS primarily occurs over the convective region ( $15$ – $30^\circ \text{ N}$ ,  $60$ – $120^\circ \text{ E}$ ).

Altitude–latitude cross sections of aerosol extinction obtained from HALOE ( $5.26 \mu\text{m}$ ) and SAGE II ( $0.525 \mu\text{m}$ ) and ECHAM5-HAMMOZ CTRL ( $0.550 \mu\text{m}$ ) simulations, averaged over longitudes  $60$ – $120^\circ \text{ E}$  and for the ASM season (of the year 2003) are shown in Fig. 6a–c, respectively. Since the number of profiles obtained from HALOE decreases rapidly at altitudes below  $16 \text{ km}$ , aerosol extinction is plotted over the altitudes  $16$ – $32 \text{ km}$ . Figure 6a and b show evidence of enhanced aerosol concentrations in the lower stratosphere transported into the Southern Hemisphere across the equator with descent over the southern subtropics. There is an arch feature in the aerosol distribution between  $40^\circ \text{ S}$  and  $15$ – $20^\circ \text{ N}$  above  $18 \text{ km}$  which is clearly separated from the aerosols in the lower TTL. This feature is similar to the one found in the ECHAM5-HAMMOZ simulation (see Fig. 6c). The extra aerosol features seen in the observations and not in the model in the lower TTL are likely due to a combination of sampling bias and under-representation of overshooting convection in the model due to the coarse resolution. In addition, we are comparing an ensemble average of free runs with observations. The free runs do not reproduce the observed state of the atmosphere, and averaging obscures much of the fine structure of individual ensemble realizations. The purpose of the comparison done here is to highlight similarities with the large-scale aerosol distribution, and it should be treated as qualitative.



**Fig. 5.** Distribution of emission mass flux ( $\text{kg m}^{-2} \text{s}^{-1}$ ) averaged for June–September 2003 for (a) BC, (b) OC, (c)  $\text{SO}_4^{2-}$ , and (d) mineral dust.

As the Asian summer monsoon transport of pollution occurs every year (Park et al., 2009; Kunze et al., 2010; Vernier et al., 2011), a similar distribution of aerosols is also observed in Atmospheric Chemistry Experiment (ACE) measurements for the year 2005 (see Fig. 6 in Dodion et al., 2008). Dodion et al. (2008) reported that the observed aerosol layer indicates the presence of high subvisible cirrus clouds. They also observed a correlation of latitudinal variation of cirrus clouds with the seasonal variation of the Intertropical Convergence Zone (ITCZ), which suggests a dependence on aerosol redistribution.

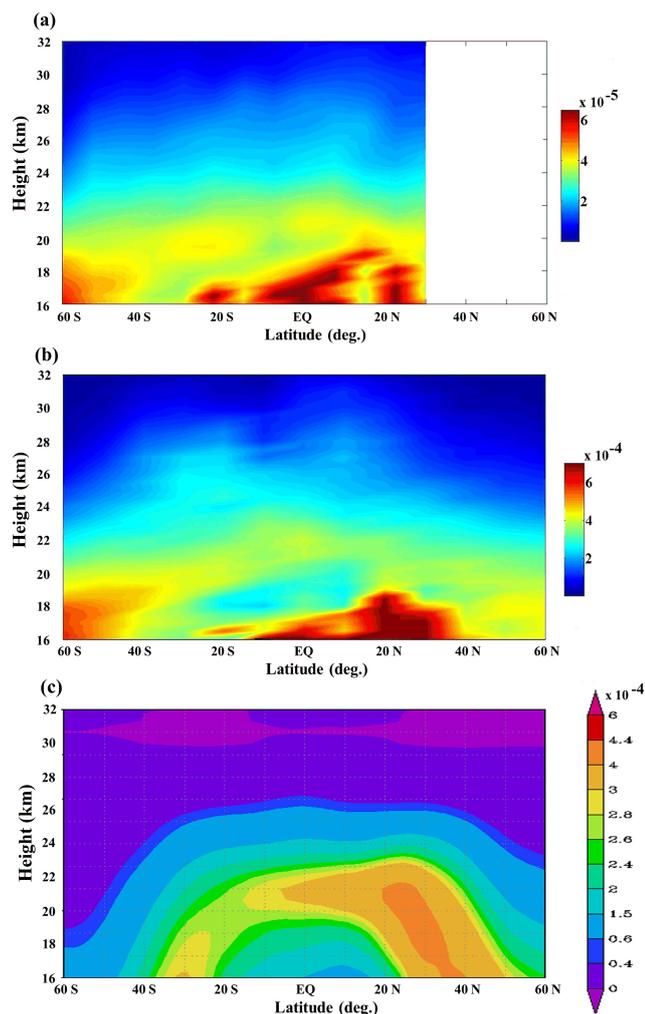
Deep convection occurs frequently over the region spanned by  $15\text{--}30^\circ \text{N}$  and  $60\text{--}120^\circ \text{E}$ . Outgoing long-wave radiation (OLR) averaged over this region is an indicator of convective forcing; hence the OLR time series can be used as a proxy of monsoon convection (Randel and Park, 2006). Association of deep convection with aerosols in the UTLS is examined by observing simultaneously time variations of OLR and BC, OC, and  $\text{SO}_4^{2-}$  aerosols at 110 hPa averaged over the central monsoon region ( $20\text{--}30^\circ \text{N}$ ,  $60\text{--}120^\circ \text{E}$ ). Figure 7 illustrates the coupling of deep convection with aerosol transport within the anticyclone. A 10–20 day periodicity in convection is evident in the aerosols. Time series of BC, OC, and  $\text{SO}_4^{2-}$  aerosols mixing ratio show statistically significant (at 95 % confidence level) anti-correlation greater than 0.5 with OLR, and thus all the aerosols vary coherently with OLR. During the strongest deep convective event in late June aerosol concentrations abruptly increase and they remain relatively high during the monsoon season. A similar coherent variation with convection was observed in AIRS data for wa-

ter vapour (Randel and Park, 2006) and in Aura MLS retrievals for CO (Park et al., 2007). This indicates that the model is able to qualitatively reproduce the influence of ASM convection on UTLS aerosols.

## 4 Impact of aerosols on the UTLS region

### 4.1 Impact of aerosols on clouds

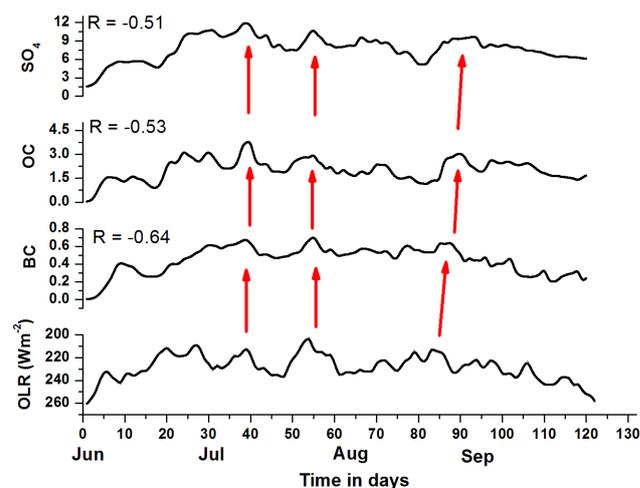
Li et al. (2005) proposed that uplifted boundary layer aerosols trapped by the Tibetan anticyclone could enhance high-altitude cloud formation and would have consequences for precipitation. From Microwave Limb Sounder (MLS) observations they showed evidence of elevated ice water content collocated with a pollution-generated CO maximum during the ASM. Global Ozone Monitoring by Occultation of Stars (GOMOS) and Atmospheric Chemistry Experiment-Fourier Transform Spectrometer (ACE-FTS) measurements also showed the coexistence of an aerosol layer and subvisual tropical cirrus clouds during 2004 and 2005 at an altitude of  $15\text{--}17 \text{ km}$  (Dodion et al., 2008). Spatial distributions of ECHAM5-HAMMOZ (CTRL runs) simulated ice cloud water (ICW) and ICNC at 110 hPa are shown in Fig. 8a and b. It shows high amounts of ICW ( $2\text{--}4 \text{ decigram m}^{-2}$ ) and ICNC ( $0.8\text{--}4 \text{ mg}^{-1}$ ) at the eastern flank of the anticyclone. Figure 8c and d exhibit time-average and latitude–pressure cross sections (averaged over  $60\text{--}120^\circ \text{E}$ ) of ICW and ICNC. Maxima in ICW ( $50\text{--}70 \text{ decigram m}^{-2}$ ) and ICNC ( $16\text{--}24 \text{ mg}^{-1}$ ) collocated with the aerosol maximum (Fig. 1) indicate that transport of aerosol- and water-



**Fig. 6.** “cross section” (averaged for June–September and 60–120° E) of aerosol extinction ( $\text{km}^{-1}$ ) as obtained from (a) HALOE 5.26  $\mu\text{m}$  for the year 2003 (b) SAGE II 0.525  $\mu\text{m}$  (c) ECHAM5-HAMMOZ simulations 0.550  $\mu\text{m}$  for the year 2003 at the altitude 16–32 km.

vapour-rich air by ITCZ may enhance cloud ice formation in the Northern Hemisphere subtropics. The ECHAM5-HAMMOZ simulated ICNC over this region is  $\sim 0.4\text{--}8\text{ cm}^{-3}$  (Fig. 8d), which is larger than the values obtained by Liu et al. (2009) ( $\sim 0.03\text{--}0.5\text{ cm}^{-3}$ ). It is unclear if these differences result from differences in the model parameterizations, different region definitions, or different time periods considered. In the present study ICNC are averaged over the anticyclone area and for the ASM season, while distribution of ICNC reported by Liu et al. (2009) is an annual average and shown as a zonal cross section across the globe.

High amounts of ICW and ICNC near the tropical tropopause layer indicate that uplifted boundary layer aerosols trapped by the Tibetan anticyclone may elevate cloud ice formation. The distribution of ICW and ICNC in this region is consistent with the aerosol distributions shown

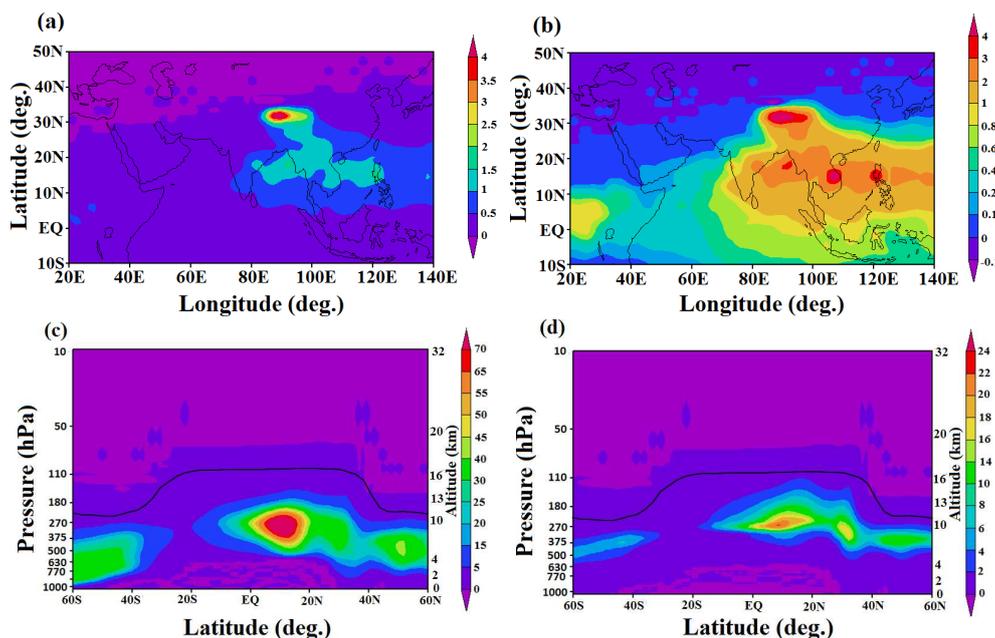


**Fig. 7.** Time series of outgoing long-wave radiation (OLR) ( $\text{W m}^{-2}$ ) averaged over 15–30° N, 60–120° E and ECHAM5-HAMMOZ-simulated concentrations ( $\text{ng m}^{-3}$ ) of BC, OC,  $\text{SO}_4^{2-}$ , averaged over 20–30° N and 60–120° E. Note that the scale for OLR is inverted. The anti-correlation coefficient of BC, OC, and  $\text{SO}_4^{2-}$  with OLR is indicated at the top of the each panel.

in Figs. 3 and 6. High amounts of ICW and ICNC at lower latitudes are related to deep convection partly over the Indian Ocean and partly over South East Asia and the Indonesian archipelago. So the ICW and ICNC concentrations between 10 and 20° N are related to the ASM.

The difference between CTRL and NOAER simulations (aerosol-induced anomalies) is shown in Figs. 9–11. Figure 9a shows the horizontal distribution of the aerosol-induced cloud ice ( $\mu\text{g m}^{-3}$ ) anomalies at 181 hPa averaged during the ASM season. It shows a prominent feature at the eastern end of the anticyclone region, where the positive cloud ice anomaly has a maximum ( $\sim 15\text{ mg m}^{-3}$ ). Figure 9b shows the seasonal and zonal average (60–120° E) latitude versus pressure cross section of aerosol-induced cloud ice anomalies ( $\mu\text{g m}^{-3}$ ). It can be seen that cloud ice shows increases of up to  $10\text{ mg m}^{-3}$  near the tropical tropopause due to aerosol loading. This value is again substantially larger than the result from Liu et al. (2009), which may again be due to differences in averaging over different areas and for different times of the year.

Figure 9b shows cloud ice increase around 20° N which connects the lower troposphere to the upper troposphere and is associated with a region of enhanced vertical transport (see Fig. 4). Increases of cloud ice near the tropical tropopause are associated with warming due to increased reflection of upwelling infrared radiation by cirrus clouds (not shown).



**Fig. 8.** (a) Horizontal structure as obtained from ECHAM5-HAMMOZ CTRL simulations (averaged for ASM) of (a) cloud ice water ( $\text{decigram m}^{-2}$ ) at 110 hPa, (b) ice crystal number concentration ( $1\text{ mg}^{-1}$ ) at 110 hPa, (c) latitude–pressure cross section (averaged for June–September and  $60\text{--}120^\circ\text{ E}$ ) of cloud ice water ( $\text{decigram m}^{-2}$ ), and (d) latitude–pressure cross section (averaged for June–September and  $60\text{--}120^\circ\text{ E}$ ) of ice crystal number concentration (ICNC) ( $1\text{ mg}^{-1}$ ).

#### 4.2 Impact of aerosols on temperature, water vapour, and circulation

Figure 10a–c show the aerosol-induced change in the atmospheric temperature, water vapour, and meridional circulation (vertical upwelling). Temperature increases by  $\sim 1\text{--}5\text{ K}$  near the tropical tropopause. A similar increase in temperature is reported by Lau et al. (2006), Lau and Kim (2006), Liu et al. (2009), and Su et al. (2011). The interior of the anticyclone over the Tibetan Plateau experiences a significant warming as well. Here, the warming in ECHAM5-HAMMOZ is greater than that found by Lau et al. (2006).

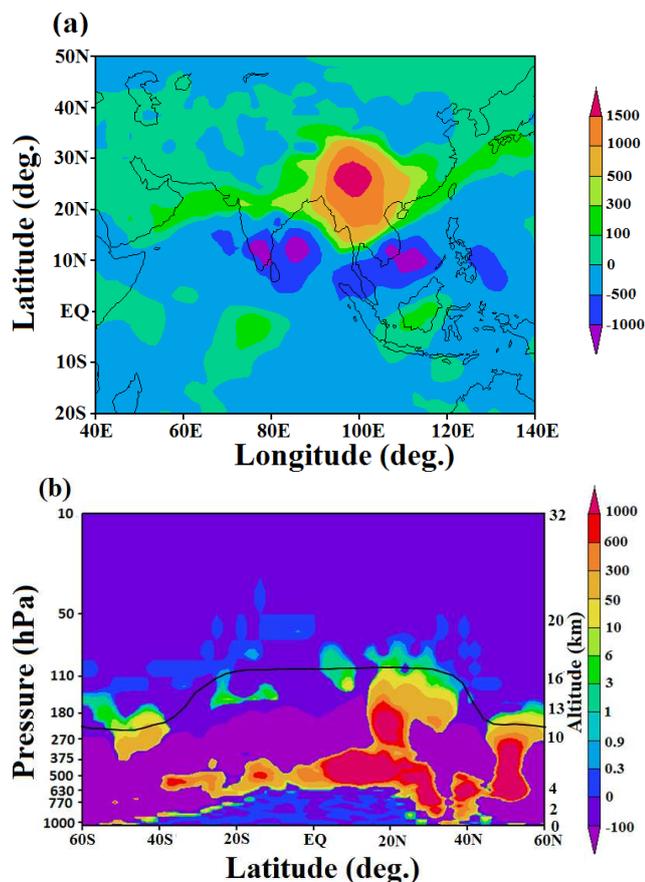
The aerosol-induced meridional circulation anomaly (Fig. 10c) shows descent (or decreased ascent) at about  $0\text{--}10^\circ\text{ N}$  and  $35\text{--}40^\circ\text{ S}$ . This pattern indicates a slight contraction and weakening of the dominant Hadley circulation cell which straddles the equator. A weakening of the Hadley circulation due to aerosol forcing is consistent with the findings of Ramanathan et al. (2005) and Bollasina et al. (2011), although there are significant differences in the model scenarios being considered. Weaker upwelling in the ITCZ around  $5^\circ\text{ N}$  is associated with reduced water vapour transport into the middle troposphere and warmer temperatures at the cold point due to lower cloud top heights. It also produces a cooling around 7 km and  $5^\circ\text{ N}$  from reduced release of latent heat by cloud condensate.

The weak reduction in upwelling through the tropical tropopause (Fig. 10c) is partly associated with the temperature increase seen in Fig. 10a. This feature is likely due

to aerosol-induced changes in the Brewer–Dobson circulation in the low latitude UTLS. The region of warming in the stratosphere above the tropical tropopause can influence wave propagation and dissipation. There are also significant dynamical changes in the Northern Hemisphere subtropics associated with the ASM that are likely modifying the synoptic-scale Rossby wave flux (see discussion below).

The results presented here broadly agree with the transport picture presented by Fu et al. (2006) in that the primary route for tracers into the TTL in the ASM region is via the convective zone on the southern flanks of the Himalayas. A prominent feature seen in Fig. 10 is the increase in vertical transport over this region. Air masses from the boundary layer reach the TTL and penetrate into the stratosphere. A large fraction of this circulation increase results from aerosol-induced enhancement of convection. However, the circulation increase above 200 hPa seen in Fig. 10 cannot be simply attributed to a change in the anticyclone. The zonal average over  $90\text{--}110^\circ\text{ E}$  (not shown) shows that the tropopause is elevated noticeably over the region of increased convective activity on the southern flank of the Himalayas. This indicates that convection is extending above 200 hPa in this region.

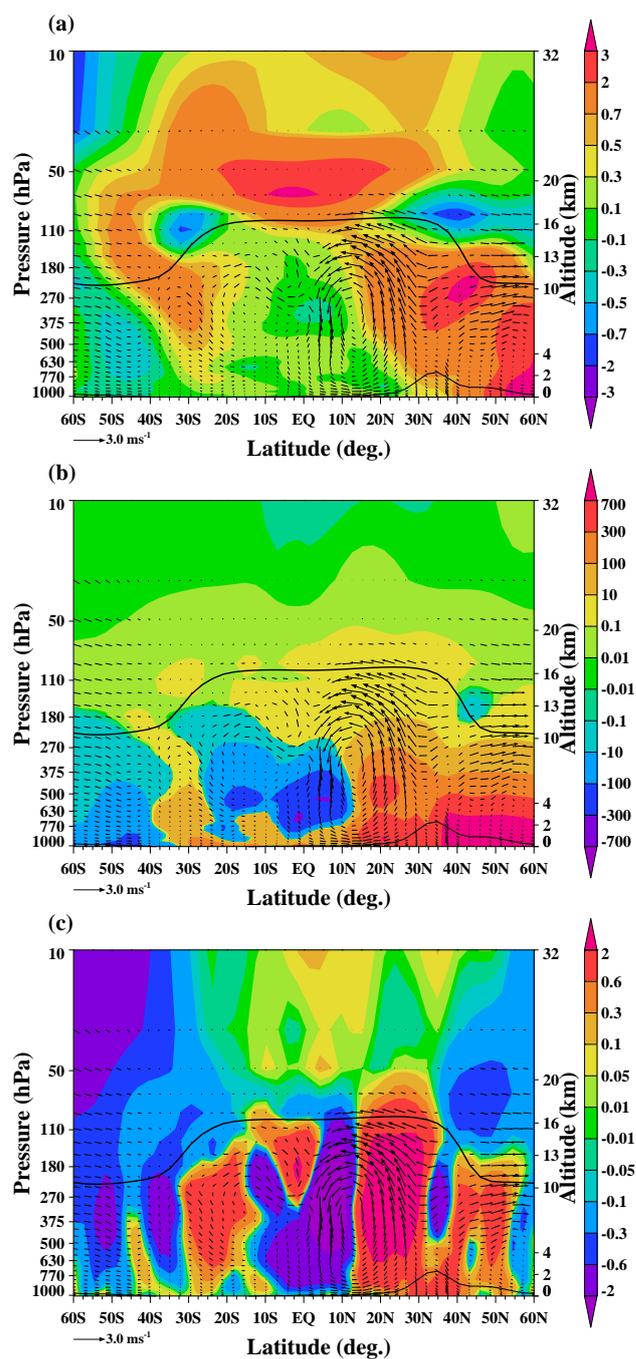
The tropopause height increase can also be attributed in part to an intensification of the Brewer–Dobson circulation evident in Fig. 10, which exhibits increased poleward and downward transport in the subtropics that has to be balanced by more tropical upwelling (Fig. 10c). This regional Brewer–



**Fig. 9.** Spatial structure of mean cloud ice ( $\mu\text{g m}^{-3}$ ) anomalies as obtained from ECHAM5-HAMMOZ CTRL–NOAER runs during ASM. (a) Pressure level slice at 181 hPa. (b) Latitude–pressure distribution averaged over 60–120° E.

Dobson circulation anomaly produces peak upwelling and downwelling over  $0.3 \text{ mm s}^{-1}$  (compare this with the mean upwelling of  $0.2\text{--}0.4 \text{ mm s}^{-1}$  between 16 and 20 km inferred from observations by Mote et al., 1998), which facilitates the entry of tropospheric constituents into the lower tropical stratosphere. The increased Brewer–Dobson circulation implies an increase in wave drag. From the change in the zonal wind (not shown) it appears that both orographic gravity wave drag and synoptic-scale Rossby wave drag have been modified due to aerosols. The former is due to an increase in the surface westerlies over India, and the latter is due to an intensification of the subtropical jet and the associated change in vertical shear (e.g. Garcia and Randel, 2008). Previous studies indicate that parameterized non-orographic gravity wave drag plays a role in simulated changes in tropical upwelling in response to subtropical jet changes (McLan-dress and Shepherd, 2009; Calvo et al., 2010).

The meridional circulation anomaly between the equator and 30° N in the troposphere seen in Fig. 10c can be interpreted as the result of increased convective heating over the southern flanks of the Himalayas around 100° E and between



**Fig. 10.** Latitude–pressure structure (filled contours, averaged over 60–120° E) of difference between ECHAM5-HAMMOZ CTRL and NOAER runs during ASM for (a) temperature (K), (b) water vapour (ppmv), and (c) vertical wind ( $\text{mm s}^{-1}$ ; not scaled). The meridional circulation is shown as a vector field (the vertical velocity field has been scaled by 300 and the units are  $\text{m s}^{-1}$ ).

20 and 25° N. It drives a thermal circulation (like the Hadley circulation) and is subject to the same asymmetry that is discussed by Lindzen and Hou (1988), namely that a localized heating anomaly centred off the equator but still in the tropics

has a two-cell circulation pattern with a dominant circulation cell on the equatorward side. The intensification of this secondary circulation contributes to the decreased ascent around  $5^{\circ}$  N. It should be noted that the change in the Hadley circulation induced by aerosols is small and there is no significant change in its spatial structure.

Although the convective transport pathway in the CTRL simulations is distinct from that identified by Park et al. (2009), being centred on the southern flanks of the Himalayas, there is some agreement with their results. From the circulation pattern shown in Fig. 4a it is clear that there is large-scale, resolved upward and westward flow from the region eastward of  $100^{\circ}$  E which is penetrating inside the ASM anticyclone in the upper troposphere (delineated by the white contour in Fig. 2a). The region of the convective upwelling anomaly also extends to  $15^{\circ}$  N, which overlaps the region spanning the Bay of Bengal and South East Asia. Therefore it is possible for aerosol and trace gas species to enter the anticyclone from this region in agreement with the results of Park et al. (2009).

Figure 10a also shows a cold stratospheric temperature anomaly above a warm tropospheric temperature anomaly around  $30$ – $40^{\circ}$  N and  $30$ – $40^{\circ}$  S. This structure is a manifestation of a “Gill-type solution” identified by Park et al. (2007), where two anticyclonic vortices on both sides of the equator are induced by a near-equatorial heat source, from deep convection, lying to the east. Analysis of the CTRL–NOAER geopotential and horizontal circulation difference at 100 hPa for June–September in our simulations (not shown) identifies two positive anomalies corresponding to a weaker anticyclone in the SH which mirrors a stronger anticyclone in the NH. Aerosols amplify the anticyclonic circulation in both hemispheres by intensifying deep convective heating in the regions of the ASM and north of Indonesia.

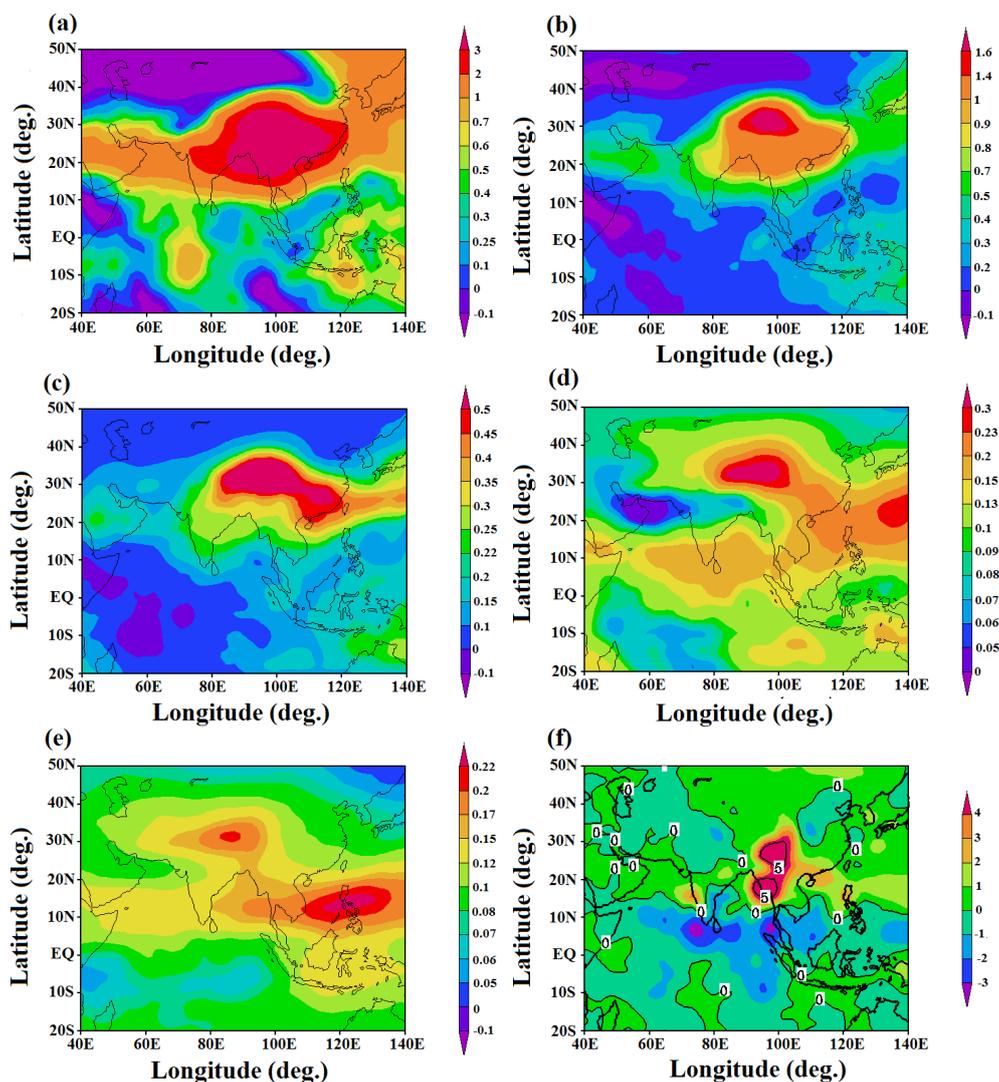
Warming of the tropical cold point tropopause seen in the present study agrees with a geo-engineering study by Heckendorn et al. (2009) which demonstrated that continuous emission of sulfur into the tropical lower stratosphere would lead to increased cold point temperatures. Warming of this sensitive region would then induce moistening of the stratosphere which is apparent in Fig. 10b.

To further diagnose the impact of aerosols on UTLS vapour, we analysed water vapour anomalies as obtained from difference between CTRL and NOAER simulations at different pressure levels. Figure 11a–e show water vapour anomalies at 155 hPa, 132 hPa, 110 hPa, 90 hPa, and 70 hPa, respectively. There are positive water vapour anomalies (0.2–3 ppmv) in the ASM anticyclone throughout the UTLS region induced by aerosols. Using the CAM3 climate model, Liu et al. (2009) demonstrated that increasing  $\text{SO}_4^{2-}$  and soot pollution lead to increases in lower stratospheric water vapour  $\sim 0.3$  ppmv (10 %) and  $\sim 0.6$  ppmv (20 %), respectively. These results are in agreement with the present study. Above the tropopause there is a noticeable increase in water vapour between the equator and  $15^{\circ}$  N (Fig. 11d). This struc-

ture appears to be a horizontal transport feature, the source of which is moisture injected into the lower tropical stratosphere over the South China Sea as well as northern India and the Himalayas. Figure 11c shows that this feature is not transported from below. From Fig. 11a–e it is clear that aerosol enhances transport of water vapour into the lower stratosphere through the northern edge of the tropical tropopause inside the ASM anticyclone.

### 4.3 Impact of aerosols on precipitation

Figure 11f shows the distribution of aerosol-induced changes in precipitation (averaged over the ASM). There are negative precipitation anomalies ( $-1$  to  $-3$  mm day $^{-1}$ ) to the south of India and extending northward along its eastern half. There is a small decrease over northern India and the Tibetan Plateau west of  $90^{\circ}$  E (about  $-1$  mm day $^{-1}$ ). This feature is most likely due to weakening of Hadley circulation discussed above. Positive precipitation anomalies (0–3 mm day $^{-1}$ ) are found over western India, which extends across to the northeast around  $20^{\circ}$  N. At the eastern end of the anticyclone region ( $90$ – $100^{\circ}$  E and  $12$ – $30^{\circ}$  N) there is significant increase in precipitation anomalies in the range of  $5$ – $7$  mm day $^{-1}$ . The positive vertical upwelling anomaly seen in Fig. 10c around  $20^{\circ}$  N is most intense between  $90$  and  $100^{\circ}$  E and is associated with the increased convection indicated by this precipitation anomaly. The weakening of the Hadley circulation applies to both the primary branch and the weak northern branch. This helps to intensify the convective system in question since the upwelling increases. However, aerosols are modifying the convection as well, so the increase in the secondary thermally direct circulation at the base of the Himalayas is not due just to changes in the Hadley circulation. As with the Hadley circulation, there is a feedback from the non-local thermally direct circulation onto the convection, intensifying it through increased vertical upwelling. Simultaneous analysis of aerosol-induced circulation changes (in Fig. 10a, b) and precipitation changes (in Fig. 10f) shows increased precipitation anomalies over the region of strong convection,  $15$ – $30^{\circ}$  N. Negative precipitation anomalies are observed in the region of reduced upwelling ( $5$ – $10^{\circ}$  N). The region of increased precipitation also extends over the South China Sea ( $1$ – $3$  mm day $^{-1}$ ). Lau et al. (2006) reported that rainfall response is not just a direct response to local aerosol forcing, but rather the result of a large-scale dynamical adjustment to aerosol initiated horizontal and vertical heating gradients to the atmosphere and land surface, modulating the climatological seasonal heating in spring and summer. So the impact of aerosols during ASM on precipitation requires a more comprehensive analysis, which is beyond the scope of this study.



**Fig. 11.** (a) Spatial structure of mean water vapour (ppmv) anomalies obtained from ECHAM5-HAMMOZ CTRL–NOAER runs during ASM at (a) 155 hPa, (b) 132 hPa, (c) 110 hPa, (d) 90 hPa, and (e) 70 hPa, and (f) spatial distribution of mean (JJAS) precipitation ( $\text{mm day}^{-1}$ ) anomalies.

## 5 Conclusions

An eight-member ensemble of ECHAM5-HAMMOZ simulations of a boreal summer season is analysed to study the transport of aerosols during Asian summer monsoon in the UTLS. Simulations show persistent maxima in BC, OC,  $\text{SO}_4^{2-}$ , and mineral dust aerosols within the anticyclone in the UTLS throughout summer. Model simulations indicate the transport of boundary layer aerosol pollutant by ASM convection to the UTLS. The ASM transport of aerosols into the TTL and the lower tropical stratosphere is observed in HALOE and SAGE II aerosol extinction. Model simulations and satellite data show that high concentrations of BC, OC,  $\text{SO}_4^{2-}$ , and mineral dust around the tropical tropopause extend to low latitudes and are transported across the equator poleward and downward in the Southern Hemisphere to

$\sim 30^\circ$  S. Variations in all four types of aerosols in the anticyclone are closely related to deep convection.

The impact of aerosols in the UTLS region is analysed by computing differences between the CTRL and NOAER simulations. Transport of anthropogenic aerosols into the UTLS produces a complex response in temperature, water vapour, and cloud ice in this region. There is warming of the tropical tropopause and the lower tropical stratosphere which is associated with direct and indirect effects of aerosols on transport and dynamics. A notable feature is the increased vertical transport between 15 and  $30^\circ$  N and around  $100^\circ$  E, which reaches into the tropical lower stratosphere. This is the primary transport pathway into the TTL and tropical lower stratosphere, and agrees with the observational findings of Fu et al. (2006) that the southern slopes of the Himalayas are

the region with the deepest-penetrating convection. However, the convective region from the Bay of Bengal to the South China Sea is also a source of aerosols given the structure of the large-scale circulation in agreement with the pathway identified by Park et al. (2009). In addition to convection and the associated diabatic circulation change in the troposphere, the transport change in this region involves the stratospheric Brewer–Dobson circulation near the tropopause. There is a significant increase in cross-tropopause transport between 15 and 35° N.

Aerosols induce a weakening of the Hadley circulation, as noted in previous studies (e.g. Ramanathan et al., 2005; Bollasina et al., 2011), and an intensification of a secondary thermally direct circulation associated with the region of strong convection on the southern flanks of the Himalayas (15–30° N and around 100° E). The effect is to reduce vertical upwelling around 5° N, which reduces transport of water vapour into the mid-troposphere near the equator. At the same time, there is increased transport of water vapour around 15–30° N. Significant positive precipitation anomalies (5–7 mm day<sup>-1</sup>) are observed over the region of intensified convection, at the eastern end of the anticyclone (90–100° E). There are negative precipitation anomalies (–1 to –3 mm day<sup>-1</sup>) to the south of India and extending northward along its eastern half. There is a small decrease over northern India and the Tibetan Plateau west of 90° E (about –1 mm day<sup>-1</sup>). Negative precipitation anomalies are collocated with reduced upwelling around 5–10° N. This is most likely due to weakening of Hadley circulation upwelling at these latitudes. The impact of aerosols on precipitation requires a more detailed study to be done later.

Maxima in ICW and ICNC collocated with aerosol maxima indicate that transport of aerosol- and water-vapour-rich air by deep convection may enhance high-level cloud ice formation in the Northern Hemisphere subtropics. Changes in cloud properties have an impact on the hydrological cycle and climate, and cirrus clouds in particular play an important role (Liou, 1986). However models do not capture cirrus clouds effectively with existing cloud schemes (Kärcher et al., 2006). Thin cirrus clouds were an uncertain component which has been recently found to be a significant fraction of the total cirrus distribution (e.g. Lee et al., 2009). Several studies (e.g. Randall et al., 1989; Ramaswamy and Ramanathan, 1989; Liu et al., 2003a, b) point out that cirrus clouds are likely to have great impact on the radiation and hence the intensity of the large-scale circulation in the tropics. Hence, anthropogenic aerosols transported by the ASM convection to the UTLS region may impact the hydrological cycle and climate. Given the limitations of cirrus cloud processes in our model, our results are in agreement with Su et al. (2011) and other studies indicating that pollution warms the TTL (by 0.3–2.5 K) and leads to a moistening of the stratosphere. Cloud processes require more detailed analysis and improved microphysical parameterizations, which are beyond the scope of this study.

*Acknowledgements.* S. Fadnavis acknowledges with gratitude B. N. Goswami, Director of IITM, for his encouragement during the course of this study. The authors thank the SAGE II and HALOE teams for providing data and the High Power Computing Centre (HPC) in IITM, Pune, India, for providing computer resources. We thank anonymous reviewers for their useful comments that helped to improve the manuscript.

Edited by: F. Fierli

## References

- Alcala, C. M. and Dessler, A. E.: Observations of deep convection in the tropics using the TRMM precipitation radar, *J. Geophys. Res.*, 107, 4792, doi:10.1029/2002JD002457, 2002.
- Antuña, J. C., Robock, A., Stenchikov, G. L., Thomason, L. W., and Barnes, J. E.: Lidar validation of SAGE II aerosol measurements after the 1991 Mount Pinatubo eruption, *J. Geophys. Res.*, 107, 4194, doi:10.1029/2001JD001441, 2002.
- Auvray, M., Bey, I., Lllull, E., Schultz, M. G., and Rast, S.: A model investigation of tropospheric ozone chemical tendencies in long-range transported pollution plumes, *J. Geophys. Res.*, 112, D05304, doi:10.1029/2006JD007137, 2007.
- Bollasina, M. A., Ming, Y., and Ramaswamy, V.: Anthropogenic Aerosols and the Weakening of the South Asian Summer Monsoon, *Science*, 334, 6055, 502–505, doi:10.1126/science.1204994, 2011.
- Calvo, N., Garcia, R. R., Randel, W. J., and Marsh, D.: Dynamical mechanism for the increase in tropical upwelling in the lowermost tropical stratosphere during warm ENSO events, *J. Atmos. Sci.*, 67, 2331–2340, doi:10.1175/2010JAS3433.1, 2010.
- Chen, B., Xu, X. D., Yang, S., and Zhao, T. L.: Climatological perspectives of air transport from atmospheric boundary layer to tropopause layer over Asian monsoon regions during boreal summer inferred from Lagrangian approach, *Atmos. Chem. Phys. Discuss.*, 12, 4185–4219, doi:10.5194/acpd-12-4185-2012, 2012.
- Cheng, T., Peng, Y., Feichter, J., and Tegen, I.: An improvement on the dust emission scheme in the global aerosol-climate model ECHAM5-HAM, *Atmos. Chem. Phys.*, 8, 1105–1117, doi:10.5194/acp-8-1105-2008, 2008.
- Dentener, F., Kinne, S., Bond, T., Boucher, O., Cofala, J., Generoso, S., Ginoux, P., Gong, S., Hoelzemann, J. J., Ito, A., Marelli, L., Penner, J. E., Putaud, J.-P., Textor, C., Schulz, M., van der Werf, G. R., and Wilson, J.: Emissions of primary aerosol and precursor gases in the years 2000 and 1750 prescribed data-sets for AeroCom, *Atmos. Chem. Phys.*, 6, 4321–4344, doi:10.5194/acp-6-4321-2006, 2006.
- Dodion, J., Fussen, D., Vanhellemont, F., Bingen, C., Matshvili, N., Gilbert, K., Skelton, R., Turnball, D., McLeod, S. D., Boone, C. D., Walker, K. A., and Bernath P. F.: Aerosols and clouds in the upper troposphere-lower stratosphere region detected by GOMOS and ACE: Intercomparison and analysis of the years 2004 and 2005, *Adv. Space Res.*, 42, 1730–1742, doi:10.1016/j.asr.2007.09.027, 2008.
- Folkens, I. and Martin, R. V.: The Vertical Structure of Tropical Convection and Its Impact on the Budgets of Water Vapour and Ozone, *J. Atmos. Sci.*, 62, 1560–1573, doi:10.1175/JAS3407.1, 2005.

- Fu, R., Hu, Y., Wright, J. S., Jiang, J. H., Dickinson, R. E., Chen, M., Filipiak, M., Read, W. G., Waters, J. W., and Wu, D. L.: Short circuit of water vapour and polluted air to the global stratosphere by convective transport over the Tibetan Plateau, *P. Natl. Acad. Sci. USA*, 103, 5664–5669, 2006.
- Fujiwara, M., Kita, K., and Ogawa, T.: Stratosphere-troposphere exchange of ozone associated with the equatorial Kelvin wave as observed with ozonesondes and rawinsondes, *J. Geophys. Res.*, 103, 19173–19182, doi:10.1029/98JD01419, 1998.
- Garcia, R. R. and Randel, W. J.: Acceleration of the Brewer–Dobson circulation due to increases in greenhouse gases, *J. Atmos. Sci.*, 65, 2731–2739, doi:10.1175/2008JAS2712.1, 2008.
- Gottelman, A., Kinnison, D. E., Dunkerton, T. J., and Brasseur, G. P.: The impact of monsoon circulations on the upper troposphere and lower stratosphere, *J. Geophys. Res.*, 109, D22101, doi:10.1029/2004JD004878, 2004.
- Heckendorn, P., Weisenstein, D., Fueglistaler, S., Luo, B. P., Rozanov, E., Schraner, M., Thomason, L. W., and Peter, T.: The impact of geoengineering aerosols on stratospheric temperature and ozone, *Environ. Res. Lett.*, 4, 045108, doi:10.1088/1748-9326/4/4/045108, 2009.
- Hervig, M. E., Russell III, J. M., Gordley, L. L., Daniels, J., Drayson, S. R., and Park, J. H.: Aerosol effects and corrections in the Halogen Occultation Experiment, *J. Geophys. Res.*, 100, 1067–1079, 1995.
- Homeyer, C. R. and Bowman, K. P.: Rossby wave breaking and transport between the tropics and extratropics above the subtropical jet, *J. Atmos. Sci.*, 70, 607–626, doi:10.1175/JAS-D-12-0198.1, 2013.
- Horowitz, L. W., Walters, S., Mauzerall, D. L., Emmons, L. K., Rasch, P. J., Granier, C., Tie, X., Lamarque, J., Schultz, M. G., Tyndall, G. S., Orlando, J. J., and Brasseur, G. P.: A global simulation of tropospheric ozone and related tracers: Description and evaluation of MOZART, version 2, *J. Geophys. Res.*, 108, 4784, doi:10.1029/2002JD002853, 2003.
- IPCC (Intergovernmental Panel on Climate Change): *Climate Change 2007 – The Physical Science Basis*, Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change, Cambridge University Press, Cambridge, UK and New York, 2007.
- James, R., Bonazzola, M., Legras, B., Surbled, K., and Fueglistaler, S.: Water vapour transport and dehydration above the convective outflow during Asian monsoon, *Geophys. Res. Lett.*, 35, L20810, doi:10.1029/2008GL035441, 2008.
- Jeong, J. I., Park, R. J., Woo, J. H., Han, Y. J., and Yi, S. M.: Source contributions to carbonaceous aerosols in Korea, *Atmos. Environ.*, 45, 1116–1125, 2011.
- Kärcher, B. and Lohmann, U.: A parameterization of cirrus cloud formation: Homogeneous freezing of supercooled aerosols, *J. Geophys. Res.*, 107, 4010, doi:10.1029/2001JD000470, 2002.
- Kärcher, B., Hendricks, J., and Lohmann U.: Physically based parameterization of cirrus cloud formation for use in global atmospheric models, *J. Geophys. Res.*, 111, D01205, doi:10.1029/2005JD006219, 2006.
- Kent, G. S., Trepte, C. R., Skeens, K. M., and Winker, D. M.: LITE and SAGE II measurements of aerosols in the southern hemisphere upper troposphere, *J. Geophys. Res.*, 103, 19111–19127, doi:10.1029/98JD00364, 1998.
- Kettle, A. and Andreae, M.: Flux of dimethylsulfide from the oceans: A comparison of updated data seas and flux models, *J. Geophys. Res.-Atmos.*, 105, 26793–26808, 2000.
- Kravitz, B., Robock, A., Shindell, D. T., and Miller, M. A.: Sensitivity of stratospheric geoengineering with black carbon to aerosol size and altitude of injection, *J. Geophys. Res.*, 117, D09203, doi:10.1029/2011JD017341, 2012.
- Kunze, M., Braesicke, P., Langematz, U., Stiller, G., Bekki, S., Brühl, C., Chipperfield, M., Dameris, M., Garcia, R., and Giorgetta, M.: Influences of the Indian Summer Monsoon on Water Vapour and Ozone Concentrations in the UTLS as Simulated by Chemistry-Climate Models, *J. Climate*, 23, 3525–3544, 2010.
- Lau, K. M. and Kim, K. M.: Observational relationships between aerosol and Asian monsoon rainfall, and circulation, *Geophys. Res. Lett.*, 33, L21810, doi:10.1029/2006GL027546, 2006.
- Lau, K. M., Kim, M. K., and Kim, K. M.: Asian summer monsoon anomalies induced by aerosol direct forcing: the role of the Tibetan Plateau, *Clim. Dynam.*, 26, 855–864, 2006.
- Lee, J., Yang, P., Dessler, A. E., Gao, B.-C., and Platnick, S.: Distribution and radiative forcing of tropical thin cirrus clouds, *J. Atmos. Sci.*, 66, 3721–3731, doi:10.1175/2009JAS3183.1, 2009.
- Li, Q., Jiang, J. H., Wu, D. L., Read, W. G., Livesey, N. J., Waters, J. W., Zhang, Y., Wang, B., Filipiak, M. J., Davis, C. P., Turquety, S., Wu, S., Park R. J., Yantosca R. M., and Jacob D. J.: Convective outflow of South Asian pollution: A global CTM simulation compared with EOS MLS observations, *Geophys. Res. Lett.*, 32, L14826, doi:10.1029/2005GL022762, 2005.
- Liou, K.-N.: Influence of cirrus clouds on weather and climate processes: A global perspective, *Mon. Weather Rev.*, 114, 1167–1199, 1986.
- Lin, H. and Leaitch, R.: Development of an in-cloud aerosol activation parameterization for climate modelling, in: *Proc. WMO Workshop on Measurements of Cloud Properties for Forecasts of Weather, Air Quality and Climate*, Mexico City, 13562, 13563, 13602, 1997.
- Lindzen, R. S. and Hou, A. Y.: Hadley circulations for zonally averaged heating centered off the equator, *J. Atmos. Sci.*, 45, 2416–2427, 1988.
- Liu, J. and Diamond, J.: China's Environment in a Globalizing World, *Nature*, 435, 1179–1186, 2005.
- Liu, H. L., Wang, P. K., and Schlesinger, R. E.: A numerical study of cirrus clouds. Part I: Model description, *J. Atmos. Sci.*, 60, 1075–1084, 2003a.
- Liu, H. L., Wang, P. K., and Schlesinger, R. E.: A numerical study of cirrus clouds. Part II: Effects of ambient temperature, stability, radiation, ice microphysics, and microdynamics on cirrus evolution, *J. Atmos. Sci.*, 60, 1097–1119, 2003b.
- Liu, X., Penner, J. E., and Wang M.: Influence of anthropogenic sulfate and black carbon on upper tropospheric clouds in the NCAR CAM3 model coupled to the IMPACT global aerosol model, *J. Geophys. Res.*, 114, D03204, doi:10.1029/2008JD010492, 2009.
- Lohmann, U. and Roeckner, E.: Design and performance of a new cloud microphysics scheme developed for the ECHAM general circulation model, *Clim. Dynam.*, 12, 557–572, 1996.
- Lohmann, U., Feichter, J., Chuang, C. C., and Penner, J. E.: Prediction of the number of cloud droplets in the ECHAM GCM, *J. Geophys. Res.*, 104, 9169–9198, doi:10.1029/1999JD900046, 1999.

- McLandress, C. and Shepherd, T. G.: Simulated Anthropogenic Changes in the Brewer-Dobson Circulation, Including Its Extension to High Latitudes, *J. Climate*, 22, 1516–1540, doi:10.1175/2008JCLI2679.1, 2009.
- Menon, S., Hansen, J., Nazarenko, L., and Luo, Y.: Climate effects of black carbon aerosols in China and India, *Science*, 297, 2250–2253, 2002.
- Mote, P. W., Dunkerton, T. J., McIntyre, M. E., Ray, E. A., Haynes, P. H., and Russell III, J. M.: Vertical velocity, vertical diffusion, and dilution by midlatitude air in the tropical lower stratosphere, *J. Geophys. Res.*, 103, 8651–8666, doi:10.1029/98JD00203, 1998.
- Moustauoui, M., Joseph, B., and Teitelbaum, H.: Mixing layer formation near the tropopause due to gravity wave-critical level interactions in a cloud-resolving model, *J. Atmos. Sci.*, 61, 3112–3124, doi:10.1175/JAS-3289.1, 2004.
- Nightingale, P., Malin, G., Law, C., Watson, A., Liss, P., Liddicoat, M., Boutin, J., and Upstill-Goddard, R.: In situ evaluation of air-sea gas exchange parameterizations using novel conservative and volatile tracers, *Global Biogeochem. Cy.*, 14, 373–387, 2000.
- Oberbeck, V. R., Livingston, J. M., Russell, P. B., Pueschel, R. F., Rosen, J. N., Osborn, M. T., Kritiz, M. A., Snetsinger, K. G., and Ferry, G. V.: SAGE II aerosol validation: Selected altitude measurements, including particle micrometeorology, *J. Geophys. Res.*, 94, 8367–8380, doi:10.1029/JD094iD06p08367, 1989.
- Park, M., Randel, W. J., Kinnison, D. E., Garcia, R. R., and Choi, W.: Seasonal variation of methane, water vapour, and nitrogen oxides near the tropopause: Satellite observations and model simulations, *J. Geophys. Res.*, 109, D03302, doi:10.1029/2003JD003706, 2004.
- Park M., Randel W. J., Gettleman, A., Massie, S. T., and Jiang, J. H.: Transport above the Asian summer monsoon anticyclone inferred from Aura Microwave Limb Sounder tracers, *J. Geophys. Res.*, 112, D16309, doi:10.1029/2006JD008294, 2007.
- Park, M., Randel, W. J., Emmons, L. K., and Livesey, N. J.: Transport pathways of carbon monoxide in the Asian summer monsoon diagnosed from Model of Ozone and Related Tracers (MOZART), *J. Geophys. Res.*, 114, D08303, doi:10.1029/2008JD010621, 2009.
- Park, R. J., Kim, M. J., Jeong, J. I., Youn, D., and Kim, S.: A contribution of brown carbon aerosol to the aerosol light absorption and its radiative forcing in East Asia, *Atmos. Environ.*, 44, 1414–1421, 2010.
- Pham, M., Muller, J., Brasseur, G., Granier, C., and Megie, G.: A three-dimensional study of the tropospheric sulfur cycle, *J. Geophys. Res.-Atmos.*, 100, 26061–26092, 1995.
- Pozzoli, L., Bey, I., Rast, J. S., Schultz, M. G., Stier, P., and Feichter, J.: Trace gas and aerosol interactions in the fully coupled model of aerosol-chemistry-climate ECHAM5-HAMMOZ: 1. Model description and insights from the spring 2001 TRACE-P experiment, *J. Geophys. Res.*, 113, D07308, doi:10.1029/2007JD009007, 2008a.
- Pozzoli, L., Bey, I., Rast, J. S., Schultz, M. G., Stier, P., and Feichter, J.: Trace gas and aerosol interactions in the fully coupled model of aerosol-chemistry-climate ECHAM5-HAMMOZ: 2. Impact of heterogeneous chemistry on the global aerosol distributions, *J. Geophys. Res.*, 113, D07309, doi:10.1029/2007JD009008, 2008b.
- Pozzoli, L., Janssens-Maenhout, G., Diehl, T., Bey, I., Schultz, M. G., Feichter, J., Vignati, E., and Dentener, F.: Re-analysis of tropospheric sulfate aerosol and ozone for the period 1980–2005 using the aerosol-chemistry-climate model ECHAM5-HAMMOZ, *Atmos. Chem. Phys.*, 11, 9563–9594, doi:10.5194/acp-11-9563-2011, 2011.
- Ramanathan, V., Chung, C., Kim, D., Bettge, T., Buja, L., Kiehl, J. T., Washington, W. M., Fu, Q., Sikka, D. R., and Wild M.: Atmospheric Brown Clouds: Impacts on South Asian Climate and Hydrological Cycle. *P. Natl. Acad. Sci.*, 102, 5326–5333, doi:10.1073/pnas.0500656102, 2005.
- Ramaswamy, V. and Ramanathan V.: Solar absorption of cirrus clouds and the maintenance of the tropical upper troposphere thermal structure, *J. Atmos. Sci.*, 46, 2293–2310, 1989.
- Randall, D. A., Harshvardan, D. A. Dazlich, and Corsetti, T. G.: Interactions among radiation, convection, and large-scale dynamics in a general circulation model, *J. Atmos. Sci.*, 46, 1943–1970, 1989.
- Randel, W. J. and Park, M.: Deep convective influence on the Asian summer monsoon anticyclone and associated tracer variability observed with Atmospheric Infrared Sounder (AIRS), *J. Geophys. Res.*, 111, D12314, doi:10.1029/2005JD006490, 2006.
- Randel, W. J., Park, M., Emmons, L., Kinnison, D., Bernath, P., Walker, K. A., Boone, C., and Pumphrey H.: Asian monsoon transport of pollution to the stratosphere, *Science*, 328, 611–613, 2010.
- Roeckner, E., Bauml, G., Bonaventura, L., Brokopf, R., Esch, M., Giorgetta, M., Hagemann, S., Kirchner, I., Kornbluh, L., Manzini, E., Rhodin, A., Schlese, U., Schulzweida, U., and Tompkins, A.: The atmospheric general circulation model ECHAM5: Part 1, Tech. Rep. 349, Max Planck Institute for Meteorology, Hamburg, 2003.
- Russell, J. M., Gordley, L. L., Park, J. H., Drayson, S. R., Hesketh, W. D., Cicerone, R. J., Tuck, A. F., Frederick, J. E., Harries, J. E., and Crutzen P. J.: The Halogen occultation Experiment, *J. Geophys. Res.*, 98, 10777–10797, 1993.
- Schulz, M., de Leeuw, G., and Balkanski, Y.: Emission of Atmospheric Trace Compounds, chap. Sea-salt aerosol source functions and emissions, 333–359, Kluwer, 2004.
- Schultz, M., Heil, A., Hoelzemann, J., Spessa, A., Thonicke, K., Goldammer, J., Held, A., Pereira, J., and van het Bolscher, M.: Global wildland fire emissions from 1960 to 2000, *Global Biogeochem. Cy.*, 22, GB2002, doi:10.1029/2007GB003031, 2008.
- Scott, R. K. and Cammas, J.-P.: Wave breaking and mixing at the subtropical tropopause, *J. Atmos. Sci.*, 59, 2347–2361, doi:10.1175/1520-0469(2002)059<2347:WBAMAT>2.0.CO;2, 2002.
- Stier, P., Feichter, J., Kinne, S., Kloster, S., Vignati, E., Wilson, J., Ganzeveld, L., Tegen, I., Werner, M., Balkanski, Y., Schulz, M., Boucher, O., Minikin, A., and Petzold, A.: The aerosol-climate model ECHAM5-HAM, *Atmos. Chem. Phys.*, 5, 1125–1156, doi:10.5194/acp-5-1125-2005, 2005.
- Su, H., Jiang, J. H., Liu, X., Penner, J. E., Read, W. G., Massie, S., Schoeberl, M. R., Colarco, P., Livesey, N. J., and Santee, M. L.: Observed increase of TTL temperature and water vapour in polluted clouds over Asia, *J. Climate*, 24, 2728–2736, doi:10.1175/2010JCLI3749.1, 2011.
- Tegen, I., Harrison, S., Kohfeld, K., Prentice, I., Coe, M., and Heimann, M.: Impact of vegetation and preferential source areas

- on global dust aerosol: Results from a model study, *J. Geophys. Res.-Atmos.*, 107, 4576, doi:10.1029/2001JD000963, 2002.
- Terao, Y. and Logan, J. A.: Consistency of time series and trends of stratospheric ozone as seen by ozonesonde, SAGE II, HALOE, and SBUV(/2), *J. Geophys. Res.*, 112, D06310, doi:10.1029/2006JD007667, 2007.
- Venkataraman C., Habib, G., Eiguren-Fernandez, A., Miguel, A. H., and Friedlander, S. K.: Residential Biofuels in South Asia: Carbonaceous Aerosol Emissions and Climate Impacts, *Science*, 307, 1454, doi:10.1126/science.1104359, 2005.
- Vernier, J. P., Thomason, L. W., and Kar, J.: CALIPSO detection of an Asian tropopause aerosol layer, *Geophys. Res. Lett.*, 38, L07804, doi:10.1029/2010GL046614, 2011.
- Vignati, E., Wilson, J., and Stier, P.: M7: An efficient size-resolved aerosol microphysics module for large-scale aerosol transport models, *J. Geophys. Res.*, 109, D22202, doi:10.1029/2003JD004485, 2004.
- Xiong, X., Houweling, S., Wei, J., Maddy, E., Sun, F., and Barnett, C.: Methane plume over south Asia during the monsoon season: satellite observation and model simulation, *Atmos. Chem. Phys.*, 9, 783–794, doi:10.5194/acp-9-783-2009, 2009.
- Yin, Y., Carslaw, K. S., and Feingold, G.: Vertical transport and processing of aerosols in a mixed-phase convective cloud and the feedback on cloud development, *Q. J. Roy. Meteorol. Soc.*, 131, 221–246, 2005.