

***Interactive comment on “Modelling the aerosol chemical composition of the tropopause over the Tibetan Plateau during the Asian summer monsoon” by Jianzhong Ma et al.: Author responses***

We would like to thank the two referees for their insightful and constructive comments.

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**Reply to Anonymous Referee #1**

Referee comments are in black. Author responses are in blue.

10 **General comments**

Ma et al. study the composition of the Asian Tropopause Aerosol Layer (ATAL), a feature associated with the upper tropospheric Asian Summer Monsoon (ASM) anticyclone. The ASM, and particularly ATAL, has received quite some attention recently. Timely publication of this paper might e.g. inform upcoming studies from the StratoClim project (<http://www.stratoclim.org/>), which addresses similar research questions.

The paper is based on a high resolution global simulation with a detailed representation of gas phase chemistry, aerosols and particularly mineral dust. Previous studies disagree on the ATAL composition. Ma et al. highlight the importance of the latter, also discussing the source areas. The methodology is sound, the paper nicely complements previous studies, it is very well written and well suited for ACP(D).

Before publication in ACP the following three aspects need to be addressed, which might or might not need major revisions:

25 We thank the anonymous referee for his/her insightful comments. Below are our point-by-point responses to referee's comments in detail.

(1) From the references provided it is not clear to me, if the used or a similar setup has ever been evaluated for the EMAC model. If there is such an evaluation, please specifically refer to it. If there is no such evaluation of the T106L90 resolution with chemistry yet, a separate evaluation paper (e.g. in GMD) might be considered – or at least a dedicated supplement. The solitary comparison to a measured profile of aerosol extinction coefficients is just a start. Furthermore, there are a lot of comparative statements about the simulation in the text. It is often not clear, whether those refer to observations, other simulations, or something else. This needs to be clarified, e.g. by an

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explicit evaluation against observations or other simulations.

EMAC is a very complicated model system coupled with different modules to simulate various physical and chemical processes simultaneously in both the troposphere and stratosphere. The model is updated and improved, by introducing new and advanced modules continuously. The references we provided in the first paragraph of Sect. 2 describe these updates and improvements as well as evaluations against observations. In subsequent paragraphs, we describe the model setup, i.e., modules adopted in this study, such as MECCA, JVAL, GMXe, ISORROPIA-II, MECCA\_KHET, AEROPT, SCAV, DRYDEP and so on, with their references provided. The standard versions of EMAC introduced by Jöckel et al. (2006, 2010, 2016) and its modules, e.g. GMXe (Pringle et al., 2010), were evaluated for a typical horizontal resolution of T42. Indeed, there were several studies using the T106 resolution and, as suggested by the reviewer, we have added some sentences to the end of Sect. 2. to describe them in detail:

*“Pozzer et al. (2012) used the same modules (e.g., GMXe and ISORROPIA-II for aerosols) as in this study but different emission data (e.g., dust emissions from AEROCOM) in the T106L31 simulations of tropospheric aerosols for the years 2005-2008. By comparing the model output with observations from different measurement networks and satellite remote sensing instruments, they found that the main spatial and temporal atmospheric distribution of sulfate, ammonium and nitrate aerosols were well reproduced in general, but there was an underestimation of aeolian dust emissions in the dust outflow regions (Pozzer et al., 2012). Klingmüller et al. (2018) develop an advanced dust emission scheme, with the updated land cover classification, the inclusion of the topography factor and the modification of the sandblasting efficiency function. They performed the T106L31 simulations for the year 2011; by comparing with the aerosol optical depth (AOD), dust concentrations and deposition results from various observational platforms, they concluded that the update significantly improves agreement with the observations (Klingmüller et al., 2018). With respect to aerosol and stratospheric chemistry, the model setup in this study is similar to those used in Brühl et al. (2015, 2018), the latter of which (Brühl et al., 2018) used the T106L90 resolution for a 1-year sensitivity test”.*

Theoretically, chemical processes simulated with state-of-science chemical reactions in the models like EMAC should be independent of model resolution. However, due to large gradients in emission rates of its precursors, e.g., around megacities, and non-linear ozone chemistry, simulated ozone in some regions might be affected by model resolution (e.g., Kentarchos et al., 2001; Stock et al., 2014). Wild and Prather (2006) showed that compared to the observations, the simulated ozone at T21, T42, T63 and T106 is increasingly realistic with resolution. Even so, non-linear ozone chemistry associated with pollution emissions and its effect on aerosols should not be so significant in remote regions, in particular at the tropopause. We think that dynamical processes that are strongly affected by the topography of the Tibetan Plateau and simulated by state-of-art parameterizations in the model, such as convection and

lightning as pointed out by the reviewer in Question 2, and associated chemical and physical processes may have a larger resolution effect on the simulation of the ATAL. A full evaluation of the T106L90 with chemistry, as suggested by the reviewer, can be considered in other paper(s).

With regards to unclear comparative statements in the text, we have revised those statements accordingly, as described in our response to specific questions below.

(2) Convection and the associated emission of lightning NO<sub>x</sub> plays a central role in the ASM anticyclone, affecting also nitrate aerosols. Convection and lightning NO<sub>x</sub> emissions are known to depend on model resolution. How does the simulation perform with respect to observed convection? Are the lightning NO<sub>x</sub> emissions in a reasonable range, globally and within the anticyclone? Those are notorious uncertainties in modelling, but the discussion of the relative importance of nitrate aerosols depends on the availability of NO<sub>x</sub>.

We agree that convection and the associated lightning NO<sub>x</sub> emissions play a central role in the NO<sub>x</sub> and OH budget in the ASM anticyclone (Gottschaldt et al., 2018; Lelieveld et al., 2018), which may have large impacts on nitrate aerosols (Tost, 2017). Both parameterizations are a notorious source of uncertainty in global models (Gottschaldt et al., 2018). As suggested by the reviewer, we have added the following paragraph in the middle part of Sect. 2 to describe the parameterization schemes for convection and lightning NO<sub>x</sub> emissions used in our simulations:

*“Convection is parameterized by the submodel CONVECT, in which the Tiedtke (1989) scheme with modifications by Nordeng (1994) is set as default (Tost et al., 2006b) and used in this study. This scheme was developed for resolution T63. As discussed in Brihl et al. (2018), it overestimates vertical transport in the UTLS for T42 and underestimates it for T106. The NO<sub>x</sub> emissions from lightning activity are calculated online using the submodel LNOX (Tost et al., 2007b). In this study we apply the parameterization developed by Grewe et al. (2001), which links the flash frequency to the updraft velocity. Here flash frequency obtained by this parameterization is scaled by a factor of 0.0695 for T106L90 simulations. In this study global emissions of NO<sub>x</sub> from lightning are simulated to be 7.9, 6.7 and 6.3 Tg (N) a<sup>-1</sup> for the years 2010, 2011 and 2012, respectively, falling into the range of 2–8 Tg (N) a<sup>-1</sup> suggested by Schumann and Huntrieser (2007). The parameterisation schemes for convection (Tiedtke, 1989; Nordeng, 1994; Tost et al., 2006b) and lightning (Grewe et al., 2001) used in this study have been evaluated with simulations at coarser resolution (Tost et al., 2007b; Lopez, 2016; Gottschaldt et al., 2018) and the agreement is particularly noticeable for the ASM anticyclone region (Gottschaldt et al., 2018)”.*

We have also added the following sentences at the end of Sect. 3.3 to highlight the important role of lightning over the Tibetan Plateau and its potential effects on nitrate

aerosols in the UTLS.

“As demonstrated in previous studies, lightning  $\text{NO}_x$  clearly dominates the  $\text{NO}_x$  budget from the tropopause to 100 hPa below it, and its emissions are much stronger in the Tibetan part (Gottschaldt et al., 2018). Lightning  $\text{NO}_x$  also plays a central role in sustaining upper tropospheric OH concentrations over the monsoon (Lelieveld et al., 2018). In our simulations, lightning  $\text{NO}_x$  emissions within the ASM anticyclone (20-140 °E and 10-45 °N) are very intensive, with estimated values of 1, 1, 0.5 and 0.9 Tg (N)  $\text{a}^{-1}$  in the years 2010, 2011 and 2012, respectively. High  $\text{NO}_x$  and OH concentrations are in favour of  $\text{HNO}_3$  formation via the reaction  $\text{NO}_2 + \text{OH}$ , which may provide an important source of nitrate within the anticyclone. Tost (2017) found 60% of aerosol nitrate between 500 hPa and the tropopause being produced from lightning in the ASM and its outflow under the present climatic condition. In addition to the available nitric acid, other factors such as neutralising ions (e.g.,  $\text{Ca}^{2+}$ ,  $\text{NH}_4^+$  and  $\text{SO}_4^{2-}$ ) and temperature can also influence the amount of nitrate aerosols in the ATAL. How these factors and associated processes affect nitrate aerosols in the UTLS over the Tibetan Plateau will be investigated thoroughly in a future study”.

(3) Please consider to add a statement about data availability (model code, setup).

We have added the statements about Code and data availability, Competing interests and Special issue in the revised manuscript.

### Specific comments

In the following URLs or full citations are only given for references not provided in the draft. Otherwise please refer to the references' list in Ma et al.

So do for our responses.

P1L23: emissions, chemistry and transport

Done.

P2L3: Isn't the UTLS rather defined by the exchange processes between stratosphere and troposphere than by some exact distance to the (which?) tropopause? Please discuss or specifically provide a citation for that definition.

We agree that it seems better to define the UTLS by exchange processes rather than by some exact distance. Here we use a rough distance value from the work (2nd sentence in Introduction) of Gettelman et al. (2011) merely to give a concept about the possible range of the ULS. The phrase has been changed to “ranging about  $\pm 5$  km around the tropopause (Gettelman et al., 2011)” in the revised manuscript.

P2L15: The ASM anticyclone is not only driven by deep convection, but also by the interaction of orography with a sea breeze and heating of the Tibetan plateau. No need to discuss in detail. Just appreciate the complexity, particularly since your simulation represents topography better than coarser simulations (see P4L18).

We agree. In the beginning of Sect. 3.1, we state “*The ASM circulation is characterized by cyclonic flow and convergence in the lower troposphere, and a strong anticyclone and divergence in the UTLS (Krishnamurti and Bhalme, 1976); its structure is primarily a response to diabatic heating associated with deep convection (Hoskins and Rodwell, 1995). Known as the “sensible heat pump”, the Tibetan Plateau modifies monsoon circulation (Wu and Zhang, 1998) and even modulates large-scale atmospheric circulations over the Northern Hemisphere (Zhao and Chen, 2001; Zhou et al., 2009)”* .

P4L18: The Jöckel et al. papers often refer to T42L90, which seems to be some kind of standard resolution for EMAC and should also be mentioned here.

Done.

P5L1: If there is an evaluation for T106L90, please specifically refer to that.

As stated in our response to General comments (Q1) above, we have specified the evaluations for T106L90 in the end of Sect. 2. of the revised manuscript: “*Pozzer et al. (2012) used the same modules .....the T106L90 resolution for a 1-year sensitivity test*”.

P5L5: Consider adding Jöckel et al. (2016) to the listing of EMAC model evaluation studies.

Done.

P5L9: Consider to move the statement about spin-up from the end of the section to here.

Done.

P5L11: Please provide more details about the chemical mechanism or provide a citation to make it reproducible.

We have added the following sentence in the revised manuscript: “*The chemical mechanism used in this study is primarily based on stratospheric chemistry used by Brihl et al. (2015) plus VOC chemistry reported by Taraborrelli et al. (2012)*”.

P6L7: Isn't the aerosol just facilitating gas phase reactions in this context, i.e. educts and products are both gaseous?

In some cases, the aerosol is just facilitating gas-phase reactions. In other cases, educts and products are not released as gases, and instead they are kept in the aerosol, either in solid or liquid state. These depend on what reaction it is and what region (the stratosphere or the troposphere) the reaction takes place. For example, for the reaction  $\text{N}_2\text{O}_5 + \text{H}_2\text{O} \rightarrow \text{HNO}_3$  on aerosol surfaces,  $\text{N}_2\text{O}_5$  is converted into aqueous-phase nitrate in the troposphere, whereas in the dry stratosphere, gaseous  $\text{HNO}_3$  is produced. For detailed information, please see Sect. 7.1.2 MECCA KHET in the work of Jöckel et al. (2010).

P6L23: Is there something like a "DLR-MACCity" inventory? Is this MACCity with species redistributed to the chemical mechanism used by Jöckel et al. (2016)?

It is actually the same inventory as reported by Jöckel et al. (2016). To avoid confusion, we have removed “*DLR-*” in the revised manuscript.

P6L25: Please elaborate to avoid quotation marks. Is the worst case scenario the best match for real emissions in that period?

The quotation mask has been removed in the revised manuscript. It is an estimate of historical emissions, instead of projected emissions based on different scenarios. Maybe we misunderstand something. For the period 2005 to 2015 the worst case scenario matches observations best for most species.

P6L28: Lightning emissions depend on model resolution and parameterizations contain tuning factors. How big is your annual global lightning NO<sub>x</sub> emissions or towards which value did you tune? Please consider adding a more thorough analysis and discussion of lightning NO<sub>x</sub>, because this is a crucial aspect of atmospheric composition in the ASM anticyclone (Barret et al., 2016; Gottschaldt et al., 2018) and affects nitrate aerosols.

As stated in our response to General comments (Q2) above, the global NO<sub>x</sub> emissions from lightning are simulated to be 7.9, 6.7 and 6.3 Tg (N) a<sup>-1</sup> for the years 2010, 2011 and 2012, respectively, falling into the range of 2–8 Tg (N) a<sup>-1</sup> suggested by Schumann and Huntrieser (2007). We have added some sentences at the end of Sect. 3.3 to highlight the important role of lightning over the Tibetan Plateau and its potential effects on nitrate aerosols in the UTLs. It should be noted that the formation of nitrate aerosols and partitioning of nitrate between gaseous and aerosol phases are complicated, depending on other more factors (e.g., gaseous NH<sub>3</sub>, pH of aerosol liquid solution, and temperature) than gaseous NO<sub>x</sub> (specifically HNO<sub>3</sub>), and will be investigated thoroughly in another paper in the near future.

P7L16: Is “crustal species” = minerals of the Earth’s crust? Why do the given species not sum up to 100 percent?

Yes, here we mean species in selected ionic form. They sum up not to 100% because the rest exist in other form, which are dominant and still treated as chemical inert dust in the model.

P7L21: Do you use the same inventory for volcanic SO<sub>2</sub> as Brühl et al (2018)?

Yes, we do. To avoid confusion, we have changed the phrase to “*As in the work of Brühl et al. (2018),*” in the revised manuscript.

P7L34: Were there any major eruptions that should have been included?

We have added the following state in the revised manuscript: “*There were two large major volcanic eruptions occurring in 2012, namely Soputan (124.73 °E, 1.11 °N) on 18 September 2012 and Copahue (288.8 °E, 37.86 °S) on 22 December 2012, with the amounts of ejected SO<sub>2</sub> being 1-2 orders of magnitude smaller than that of Nabro, as reported by Mills et al. (2016). These two volcanic eruptions are not included for the simulations of this study*”. Since they occurred after the ASM period we are investigating, neglecting them in the simulations should have not any effects on the results for this study.

P8L9: Is this one of the nudging profiles used by Jöckel et al. (2016)?

Yes. The nudging profile should be the same as demonstrated in Fig. S5 of Jöckel et al. (2016). We have added the phrase “*as used by Jöckel et al. (2016)*” in the revised manuscript.

P8L24: “regionally averaged” ... Which region? The one covered by fig. 2?

It is the region covered by Fig. 2. We have added the phrase “*over 20–140 °E and 10–45 °N*” in the revised manuscript.

P8L27: There is eddy shedding to the east and to the west of the ASM anticyclone.

We have added this sentence to the revised manuscript as suggested by the reviewer.

P8L31: “well represented” ... Such a statement needs comparisons to observations.

Here we mean the general or climatic horizontal distributions of the wind field, O<sub>3</sub> and CO associated with the ASM anticyclone. In the end of this paragraph, we cited other model studies (i.e., Gettelman et al., 2004; Park et al., 2007; Park et al., 2009; Barret et al., 2016; Santee et al., 2017) for such O<sub>3</sub> and CO distributions. We also have added the following sentence in the revised manuscript: “*It was shown recently that EMAC realistically simulates reactive gases and radicals within the ASM anticyclone by comparing the simulation results with aircraft measurements (Lelieveld et al., 2018)*”.

P9L5-6: Those studies did not use EMAC, did they? Consider rephrasing.

No, they didn't. The phrase has been changed to “*This feature has been well reproduced by EMAC using O<sub>3</sub> as a stratospheric tracer and CO as a tropospheric tracer as also done in other model studies*”.

P9L8: Why not showing the average of multiple years? Is 2011 somehow representative for the climatological mean state?

By comparing Fig. 3 with Fig. S1 and Fig. S2, one can see nearly the same global distributions of these aerosol columns (e.g., dust) among 2010, 2011 and 2012. So the year 2011 is representative for the climatological mean state in the early 2010s. We show the three years of aerosol columns separately considering that one may want to look at a specific year for comparison with aerosols in the UTLS, which are investigated year on year.

P9L18: Consider scanning the literature for results of the StratoClim campaign(s). For instance, Brunamonti et al. 2018 (<https://www.atmos-chem-phys.net/18/15937/2018/>) show aerosol measurements within the ASM anticyclone.

We have added the following statement in the revised manuscript “*By balloon-borne measurements, Brunamonti et al. (2018) found the maximum aerosol backscatter occurring at the cold-point tropopause, revealing the thermodynamically significant levels of the ATAL*”.

P9L33: Consider putting this into perspective by discussing differences between 2011 and 2015.

Do you mean P10L33 and between 2011 and 2012? We have added this into the second paragraph of the Conclusions in the revised manuscript: “*In contrast to the absolute K<sub>e</sub>*



from mineral dust, which is at the same level in the three years, the absolute  $K_e$  contributed by water-soluble species (WASO) and aerosol liquid water (ALW) is much higher in 2011 than in 2010 and 2012, especially in the lower stratosphere (e.g., at 17 km)".

5 P10L5: "reproduced" ... To which observations do you refer here?

We have added the phrase "*as shown by Vernier et al. (2011) and Thomason and Vernier (2013)*" in the revised manuscript.

P10L9: Are there observations for 2011, showing a similar anti-ATAL effect?

We did not find any literature reporting such observations for 2011.

10 P10L32: Why do you focus on 2011, given that it is an unusual year for ATAL?

Here we try to explain this phenomenon caused by the volcanic eruption, which is unusual for ATAL but does happen once every few years.

P11L22: This is only true without dynamic instabilities (see e.g. Gottschaldt et al., 2018). Please consider a more differentiated formulation.

15 We have changed the statement to "*The ASM anticyclone not only traps tropospheric pollutants inside but also to some extent blocks intrusions of stratospheric ozone and aerosols from outside. It should be noted that here we only investigate a seasonal mean aspect of the ASM anticyclone. Due to the dynamical instabilities of the ASM anticyclone, entrainment of stratospheric tracers does occur frequently, approximately*  
20 *twice a month (Gottschaldt et al., 2018)*".

P11L31: This statement seems to contradict the previous statement about blocking (P11L22).

Please see our reply above (P11L22).

25 P12L13: Is there a reason for not using isentropic coordinates, which should better reflect lateral transport?

Yes, we agree that isentropic coordinates should be better to reflect lateral transport. Here we use altitudes considering that they are popular for satellite products and sounding measurements. The model uses hybrid pressure coordinates.

30 P12L15: Consider rephrasing the rationale for not showing those: Ok for comparisons to corresponding observations, but distinction might make sense for analysing model results only.

35 The statement has been rephrased as "*For simplicity, the crustal species  $\text{Ca}^{2+}$ ,  $\text{Na}^+$ ,  $\text{K}^+$  and  $\text{Mg}^{2+}$  are not shown individually, considering that these ionic species within the ASM anticyclone originate predominantly from mineral dust, having the same constant relative fractions as their emission sources do (see Sect. 2 above)*".

P12L26: Do you refer to the interior or the surroundings of the ASM anticyclone here?

We refer to the interior of the ASM anticyclone, in which the Tibetan Plateau is situated.

P13L7: It's probably not only due to the dust scheme, but also to the high resolution.



We have added a statement about the resolution effect in the revised manuscript.

P13L10: Did Fadnavis et al. not consider volcanic sulfate, e.g. via some inventory?

This was not clearly described in the work of Fadnavis et al. (2013). We have rephrased the statement as *“since volcanic sulfate was most likely not considered in their simulation”* in the revised manuscript.

P13L18: Such a striking discrepancy deserves elaboration. A thorough discussion of the differences might be difficult, but are there any ideas to explain this?

We have added the following discussion in the revised manuscript: *“Similar to the comparison with Fadnavis et al. (2013) mentioned above, the different dust emission scheme and model resolution used by Yu et al. (2015) might lead to significant discrepancy of dust concentration in the UTLs within the ASM anticyclone compared to this study. Moreover, Yu et al. (2015) did not take into account stratospheric volcanic emissions and nitrate aerosols in their simulation, which might underestimate the contribution of inorganic aerosols to the ATAL with respect to this study”*.

P13L23: This is much less than simulated by EMAC (fig. 10, 11) -> Reasons for this discrepancy?

Gu et al. (2016) referred Fairlie et al. (2007) for the mineral dust treatment in GEOS-Chem. Fairlie et al. (2007) reported that global dust emissions were 178 Tg a<sup>-1</sup> for accumulation mode particles (radius of 0.1–1.0 μm) and 1453 Tg a<sup>-1</sup> in total (radius at 0.1–6.0 μm) for the year 2001. The dust scheme of EMAC used in this study is based on the work of Klingmüller et al. (2018). According to Klingmüller et al. (2018), global dust emissions were 148 Tg a<sup>-1</sup> for accumulation mode particles and 1310 Tg a<sup>-1</sup> in total for the year 2011. It seems that the estimates in global dust emissions between the two models (GEOS-Chem and EMAC) are comparable. For the reasons of much less dust (mainly for the accumulation mode) in the UTLs within the ASM anticyclone, we assume that both soil-related database and topography-associated modeling strategy for the Tibetan Plateau can influence the results. These should be investigated thoroughly in future studies.

P14L4: Consider a more cautious formulation here, because there is no consensus with other studies.

We have changed the phrase *“As shown above”* to *“According to our EMAC simulations”* in the revised manuscript.

P14L19: Tagging the emission regions would help here. However, the connection of the plume over the Tibetan plateau to the surface is convincing. Consider to substitute “spatial distribution” by a more explicit formulation.

We have changed the expression to *“The plume transport pattern that connects the maximums of mineral dust aerosols over the Tibetan Plateau to the surface indicates”* in the revised manuscript.

P14L23: This is strange, because near surface concentrations in Fig. 12 are not

increased north of the conduit. However, it's hard to judge from fig. 3. The apparent shift to the north could just be an artefact of the colour scale and the white line over the maximum in Fig. 3. Please clarify.

Fig. 3 shows the tropospheric columns ( $\text{mg m}^{-2}$ ), which are derived by integrating the aerosol loads in all model levels from the surface to the tropopause. Fig. 12 presents aerosol mass mixing ratios ( $\mu\text{g kg}^{-1}$ ) at the surface and selected altitudes above sea level (a.s.l.), with the latter starting from 6 km a.s.l. If a lower altitude, e.g. 4 or 5 km a.s.l., is selected, one can see large blank areas on the plateau, simply because they are below the ground levels (see Fig. A1 below). This terrain effect can result in the shift of the column maximum to the north shown in Fig. 3.

P15L15-17: Didn't Brühl et al. just consider T106L31? Please try to disentangle the effects of vertical and horizontal resolution more clearly.

Brühl et al. (2018) considered T106L31 and T106L90.

P15L25-28: Do you recommend T106L90 for this type of study in general OR only if there was some improvements to the convection scheme?

We recommend T106L90 with improved convection parameterization. We have changed the statements to: *"Such difference might partly be attributed to the difference in simulated mineral dust, but also it is likely due to less efficiently convective transport of anthropogenic aerosols and their gaseous precursors in the T106L90 simulations compared to T63L90 simulations. It should be noted that deep convection events occur much less frequently over the northern part of the Tibetan Plateau than the southern part of it, to the latter pollution from South Asia tends to accumulate (Lelieveld et al., 2018). Our EMAC simulation at a relatively high resolution (i.e., T106L90) reveals clearly the important role of emissions and the orographic forcing in mineral dust transport over the Tibetan Plateau. T106L90 with improved convection parameterization is suggested to investigate the transport of aerosols and their gaseous precursors associated with complex topography and finer structure of the anticyclone"*.

P15L25-28: T63L90 fits best to observations, because the convection scheme was developed for this resolution. Is this correct?

As described in the manuscript, the convection scheme we use for this study was developed by Tiedtke (1989) based on T63 resolution.

P15L25-28: You claim that T106L90 is better for transport over complex terrain. That is intuitively clear, but strictly would need to be shown. However, what is dominating transport to the UTLS: convection or orographic forcing from the terrain?

This should depend on the regions. On the northern part of the plateau, orographic forcing may be more important, due to less moisture and deep convection. See our reply to the first question about P15L25-28 above.

P15L25-28: Another point to consider is whether or not the occurrence of convective events is represented more realistically by T63 or T106. Please discuss those aspects to better justify your recommendation for T106L90. Consider comparing convection or

lightning activity to observations to get a better idea of how realistically your simulation is (see also General comments).

Fig. A2 shows the geographical distributions of averaged daily deep convection events for each month in 2011, from our EMAC T106L90 simulation. It seems that the distribution patterns are realistic. We also think that these comparisons are interesting, and we will do that in the near future when we have found a optimistic method to improve the convection scheme for T106L90 resolution.

P15L32: “improve” ... Compared to what?

We have skipped this sentence in the revised manuscript considering that this advanced dust flux scheme has been used and tested before.

P16L1: “enhance” ... Compared to what?

We have skipped this sentence in the revised manuscript considering that MIPAS SO<sub>2</sub> data was not firstly used in this study.

P28L4: Consider using SI units

Done.

P30L3: Confusing description of the quantity: Do you mean just the tropospheric burden?

Yes. The confusing phrase “*of the column concentrations*” has been deleted in the revised manuscript.

## Technical corrections

P31, Fig. 4: Figure blurry -> consider higher resolution or a vector graphics format  
The .ps files will be provided for figure production.

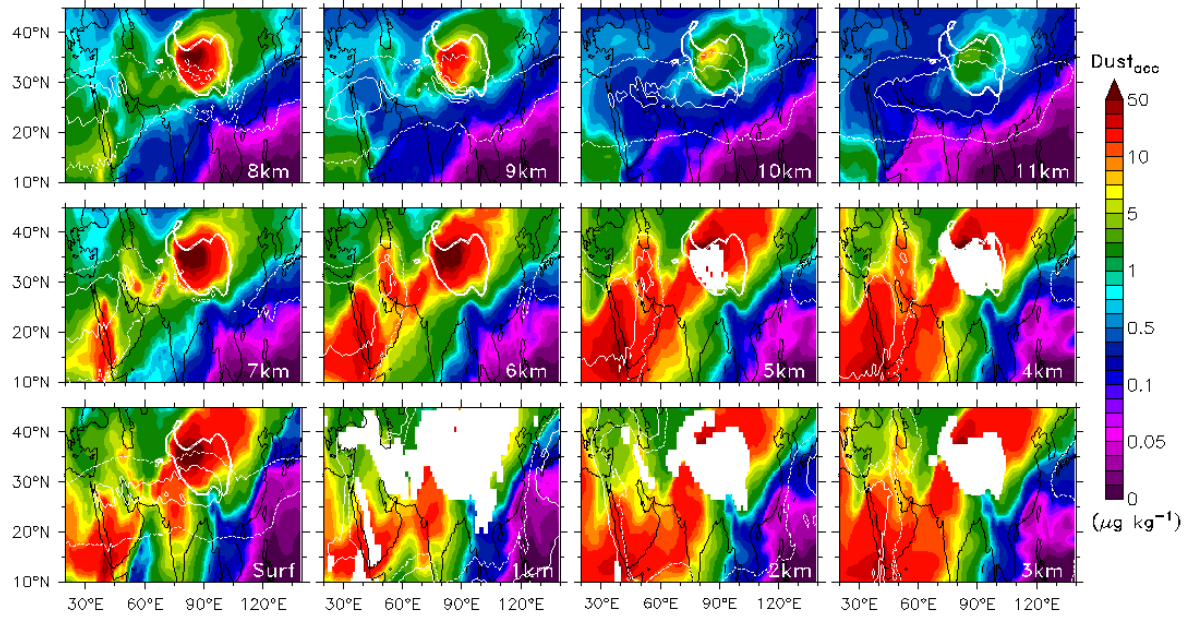
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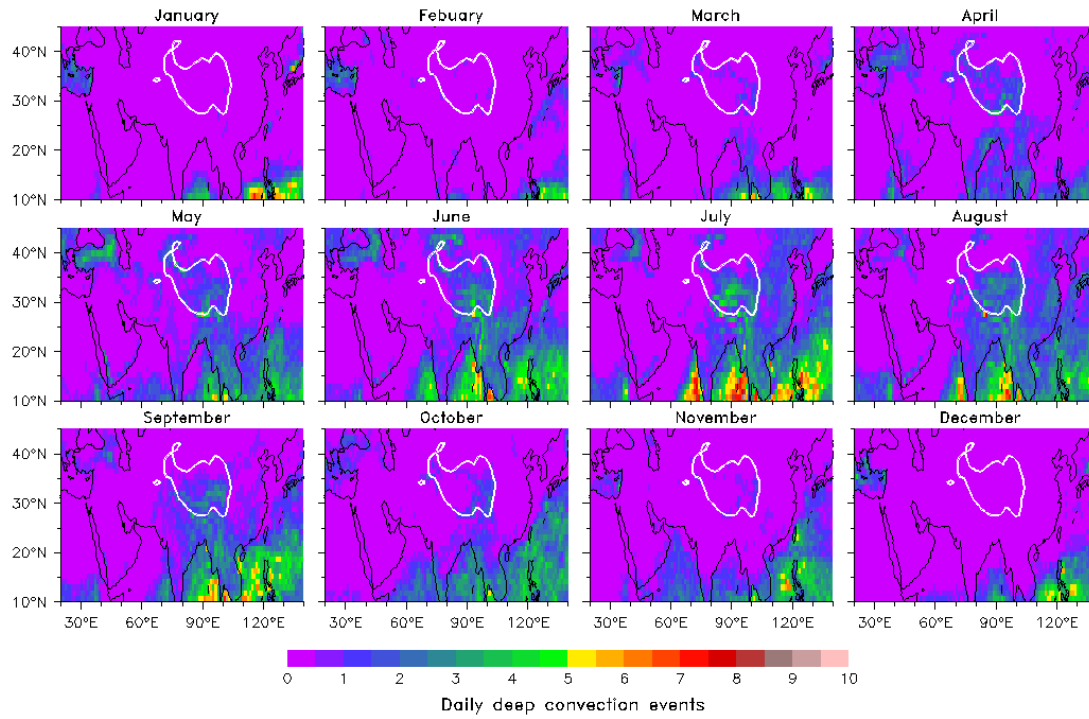
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**Figure A1.** EMAC simulated dust mass concentrations in the accumulation mode,  $Dust_{acc}$ , in units of microgram of dust per kilogram of air ( $\mu g\ kg^{-1}$ ) (note that a molecular weight of 40.08 is used for dust), at selected altitudes, i.e. at the surface, 1 km, 2 km, 3 km, 4 km, 5 km, 6 km, 7 km, 8km, 9 km, 10 km, and 11 km, averaged for July–August 2011. Thin white lines indicate refers to the contour of pressure deviation, as defined in Fig. 2, and thick white lines highlight the Tibetan Plateau area.



**Figure A2.** EMAC simulated averaged daily deep convection events for each month in 2011. White lines highlight the Tibetan Plateau area.

## Reply to Anonymous Referee #2

Referee comments are in black. Author responses are in blue.

### General comments:

This interesting work studies the aerosol chemical composition in the Asian Tropopause Aerosol Layer (ATAL), and uses EMAC model running at a high resolution to investigate the links between ATAL and Asian Summer Monsoon (ASM). The aerosol properties in the ATAL have been the subject of discussion over the past decade and have received quite some attention recently. I agree with the Reviewer#1 that the results of this work nicely complement previous studies. The manuscript is well written, the methodology is detailed described and sound, and the results are well presented and discussed. The authors have addressed most of my concerns in the response to Reviewer#1. I feel this work is suitable for publication in ACP after address the following minor concerns.

We thank the anonymous referee for his/her insightful comments. Below are our point-by-point responses to referee's comments in detail.

### Specific comments:

1) It would be best to rephrase some expressions in the manuscript, considering the similarity index of 23% (see Similarity Report). No doubt regarding the originality of this work, which will be mentioned in my later comments, but I feel some re-wording to credit the previous works in a better way would help improve this manuscript.

The similarity report for our manuscript showed that the matching words resulted mostly from common terms, such as “*the upper troposphere and lower stratosphere*”, “*over the Tibetan Plateau*” and “*the Asian summer monsoon (ASM)*”, as well as from some phrases used to describe the model and methods. They were distributed over different paragraphs and pages. As suggested by the reviewer, we have strived as much as possible to rephrase these expressions so that the percentage of similarity can be dramatically reduced. Specific revisions can be found in the marked-up version of revised manuscript.

2) About the altitudes of 15-18 km. In order to avoid confusion, please find a suitable place to clearly state that whether it is above the sea level or over ground.

In the first paragraph of our manuscript (P2L9-10 of the ACPD version), we stated “*between 13-18 km above sea level (a.s.l., hereafter all altitudes are referred to a.s.l. except when specified differently)*”. This statement appears to be not entirely sufficient for clarification. As suggested by the reviewer, we have added such note to the legends of related figures in the revised manuscript.

3) P5L24. I am wondering that how is organic aerosol formation simulated in the ORACLE sub-model, if partitioning of secondary organic aerosol between gas and particle phases is not considered? Some elaboration may be needed here.

Thanks to the referee, we have noticed that this text does not correspond to the simulation performed, as ORACLE was not used in our numerical experiment. We have rewritten the sentences in the revised manuscript as “*The primary OC aerosol is assumed to be emitted by 65% as hydrophilic and 35% as hydrophobic (Pringle et al., 2010). Following Pringle et al. (2010) and Pozzer et al. (2012), secondary OC particles are directly emitted as primary OC. The organic aerosol formation and chemical aging can be calculated by the ORACLE submodel in EMAC (Tsimpidi et al., 2014). However, in this work the partitioning of secondary organic compound between the gaseous and aerosol phases is not considered and the ORACLE module was not used in the simulation. The aging that leads the conversion of hydrophobic BC and OC to hydrophilic ones is realized by the collisions with hydrophilic particles and the condensation of inorganic gases such as sulfuric acid (Pringle et al., 2010 and references therein)*”.

4) P10L19. The acronym ‘SS’ for sea spray. Please place it at the first time when ‘sea spray’ was used.

We have given up using the acronym ‘SS’ in the revised manuscript considering that sea spray (or sea salt) is not frequently discussed in the paper.

5) P10L32. The contribution of WASO and ALW to aerosol extinction is much higher in 2011 than in 2010/2012. Is it due to the pronounced Nabro eruption in 2011? This eruption may enhance the highly hygroscopic components in UTLS such as sulfate originated from SO<sub>2</sub>, and enlarge the contributions from WASO and ALW? Some discussion about the difference between 2011 and 2010/2012 would be interesting to see. The distributions of ionic (hygroscopic) species, including SO<sub>4</sub><sup>2-</sup>, HSO<sub>4</sub><sup>-</sup> and H<sup>+</sup>, and the effect of the 2011 Nabro eruption are shown and discussed in Sect. 3.3. We have added the statements “*The pronounced Nabro eruption of SO<sub>2</sub> in 2011 can enhance sulfuric acid and sulfate aerosols in the UTLS, and increase the contributions of WASO and ALW to the aerosol in the UTLS, especially in the lower stratosphere and outside the ASM cyclone, as shown and discussed in the following section*” to the last paragraph of Sect. 3.2, and “*Due to the effect of the Nabro eruption, higher SO<sub>4</sub><sup>2-</sup>, HSO<sub>4</sub><sup>-</sup> and H<sup>+</sup> concentrations are found in the year 2011 than in 2010 and 2012 over the ASM region and the whole northern hemisphere, most profoundly in the lower stratosphere*” to the third paragraph of Sect. 3.3 in the revised manuscript.

6) P11L11. The definition of ‘Ca<sup>2+\*</sup>’ needs to be clarified. Cations of Na, K, Mg and Ca are accounted for as ‘Ca<sup>2+\*</sup>’. Here, you mean by mass, mol, or charge balancing? Furthermore, Na<sup>+</sup> is also a typical tracer for sea salt. Drop sodium and just sum up calcium, magnesium and calcium. Would this be a better tracer for mineral dust?

As shown in Figs. S7, 8, and 9, all these ionic species are accounted for by charge balancing. We have rewritten the sentences as “(4) *for simplicity, Na<sup>+</sup>, K<sup>+</sup> and Mg<sup>2+</sup>*”



together with  $\text{Ca}^{2+}$  in the mineral dust are accounted for as  $\text{Ca}^{2+,*}$  in the equivalent amounts since  $\text{Ca}^{2+}$  is a typical ionic tracer for mineral dust (Ma et al., 2013). Note that these four ionic species are also contained in sea salt and generally  $\text{Na}^+$  is taken as a tracer for sea salt. However, mineral dust appears to be a dominant source of all these species in the UTLS over the Tibetan Plateau since they are well correlated with the dust plume and the concentrations of  $\text{Ca}^{2+}$  are much higher than those of  $\text{Na}^+$  there". The distribution patterns of  $\text{Ca}^{2+,*}$  with  $\text{Na}^+$  being dropped (Fig. A3) are similar to those with  $\text{Na}^+$  being included (Fig. 9). So we still take  $\text{Na}^+$  into account for  $\text{Ca}^{2+,*}$  as the maximum of  $\text{Na}^+$  concentrations in the UTLS over the Tibetan Plateau is also dominated by the contribution from mineral dust.

7) P13L27. Typo? ‘ACM’ change to ASM ?

Done.

8) Section 3.5. A nice discussion to figure out the source of dust in the UTLS by holographic scanning the transport pathway from surface to 18 km. My first suggestion is: would it be more generally representative to show the results of 2010/2012, but place the results of 2011 in supplement? Anyway, it would not change the results significantly, because ‘transport patterns are the same in 2010 and 2012 as in 2011’; however, 2011 is a special year tagged by the pronounced Nabro eruption with potential impacts on climate the general circulation, which may lead to unnecessary doubt from audience about its representative. The second suggestion is: would it be possible to have some discussion about the source or transport pathway of WASO and ALW (or water vapor), which also considerably contribute to aerosol extinction coefficient in the ATAL?

According to the reviewer comments, we have used the results of 2012 for both Fig. 3 and Fig. 12, and have moved the results of 2011 to the Supplement of the revised manuscript. For the second suggestion of the reviewer, we have added Figures S11, S12 and 13 (and Figures S13, S14 and 14) and related discussions about the distributions and transport of water vapor (and ammonia) in the revised manuscript, as can be seen in the last two paragraphs of Sect. 3.5: “Figures S11, S12 and 13 present the geographical distributions of specific humidity,  $q_v$ , at various altitudes from the surface to 18 km over the ASM region from our EMAC simulations, averaged for July–August in the years 2010, 2011 and 2012, respectively. It is shown that deep convection of moisture occurs over the southern edge of the Tibetan Plateau, with the maximum  $q_v$  attained from 10–11 km altitude upward. This is similar to the reanalysis results from previous studies, e.g., Xu et al. (2014), who found the most significantly relative enhanced  $q_v$  at 250–300 hPa over the same area. The geographical position of the mineral dust maximum in the upper troposphere over the Tibetan Plateau is apparently different from that of the moisture maximum, indicating that different pathways for the transport of aerosols and their gaseous precursors from the lower troposphere to the UTLS over the Tibetan Plateau.

Figures S13, S14 and 14 present the geographical distributions of gaseous ammonia ( $\text{NH}_3$ ) at various altitudes from the surface to 18 km over the ASM region from our

EMAC simulations, averaged for July–August in the years 2010, 2011 and 2013, respectively. In addition to northern India, which is located to the southwest of the Tibetan Plateau, an enhancement of surface  $\text{NH}_3$  is also found in the central main body of the Tibetan Plateau, although the maximum  $\text{NH}_3$  mixing ratio in the latter area ( $\sim 10$  ppbv) is about half of that in the former area ( $\sim 20$  ppbv). It is shown that surface ammonia can be more efficiently convected and transported to the UTLS from the Tibetan Plateau than from the Indian region, leading to high levels of  $\text{NH}_3$ , e.g., about 0.1 ppbv at 17 km altitude. Such large amounts of lofted ammonia accumulate within the ASM anticyclone, providing the basic gases favourable for the formation of nitrate aerosols (especially in the years 2010 and 2012 when the volcanic eruption effect is relatively small with respect to 2011), as discussed in Sect. 3.3.”.

The sources, formation mechanisms and transport pathways of aerosols in the ATAL are very complicated and relevant to various physiochemical, cloud and dynamical processes, which will be investigated thoroughly in the next study.

9) P17L9. I am curious that why finer resolution (T106L90) underestimate the convective vertical transport, but not the coarser resolution. Should the finer resolution represent convective processes better, although none of them explicitly describe the convective processes? Would you help me understand it.

As pointed out by the reviewer, the vertical velocity manifests on finer horizontal resolution, approaching to be more realistically representing the convective processes. On the other hand, cumulus convection is a sub-grid scale phenomenon in all GCMs and has to be parameterized using model’s resolved variables. For the extended versions of the convective parameterization scheme at ECMWF developed by Tiedtke (1989) and Nordeng (1994), the large scale convective activity is related to the convective available potential energy (CAPE), and the cloud base mass flux ( $M_B$ ) can be expressed as:

$$M_B = \frac{\text{CAPE}}{\tau} \left\{ \int_{\text{base}}^{\text{top}} \left[ \frac{(1+\delta\bar{q})}{C_p \bar{T}_v} \frac{\partial \bar{s}}{\partial z} + \delta \frac{\partial \bar{q}}{\partial z} \right] \eta \frac{g}{\bar{p}} dz \right\}^{-1}$$

(see the paper of Nordeng (1994, equation 33) for detailed information about the variables and formula).  $M_B$  is inversely proportional to the adjustment time scale  $\tau$ , giving too little convective mass flux when a too long adjustment time is chosen.

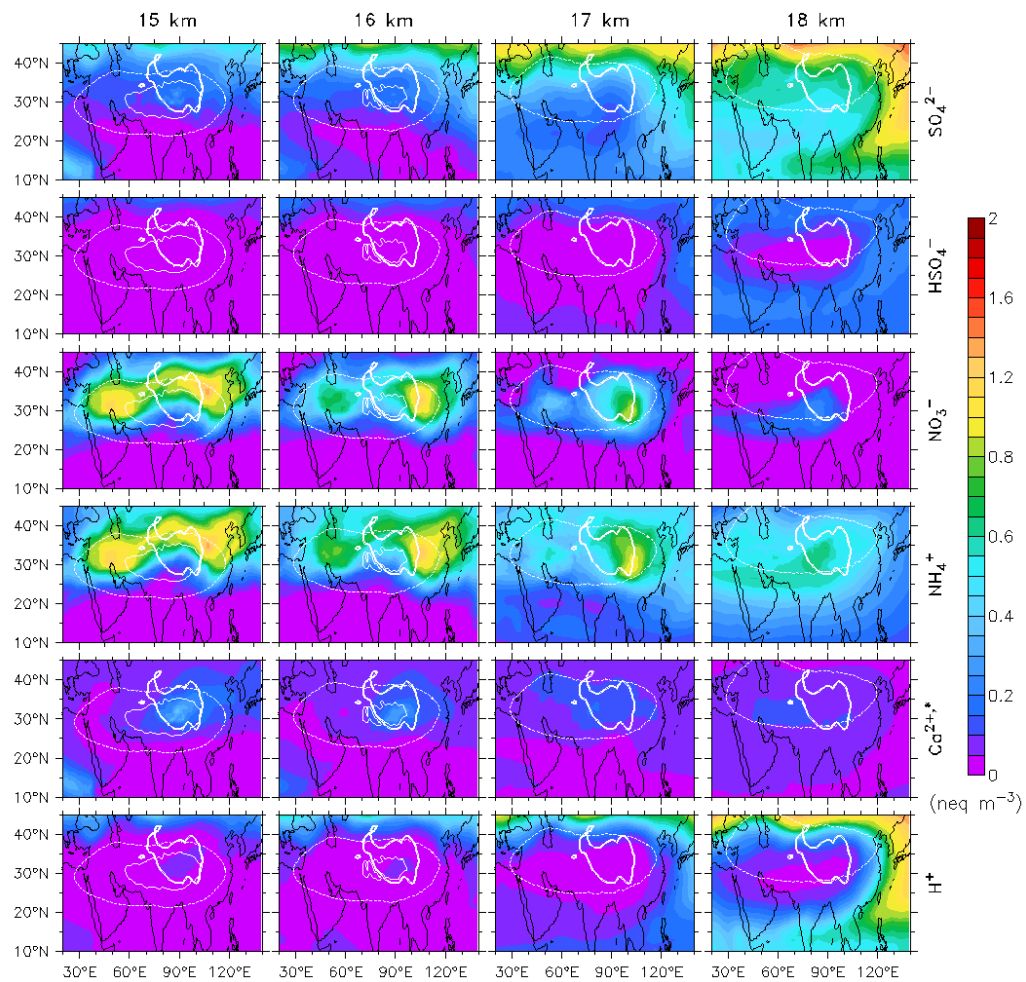
To estimate the cloud ensemble properties properly, among other parameters, the convection relaxation time  $\tau$ , which should be smaller (larger) with increasing (decreasing) horizontal resolution, also needs to be adjusted. Following Nordeng (1994), who suggested that as a rule of thumb the change in  $\tau$  should be inverse proportional to the change in resolution, an algorithm similar to that used in the ECMWF model, i.e.,  $\tau[\text{s}] = \min(3 \cdot 3600, 2 \cdot 3600 \cdot 63/N)$ , where  $N$  denotes the spectral resolution, is applied in EMAC. In this study,  $N$  was set to be 106 since we performed T106 simulations. However, such algorithm was tuned to T63 simulations and might lead to

biases (either positive or negative) when applying to other resolutions. Actually, Nordeng (1994, chapter 6.1) reports deviations from his rule of thumb. In going from T106 to T213, which is a doubling in resolution, he proposed according to his estimate a halving in  $\tau$ . But the change from T63 to T106, which increases resolution just by a factor of 1.67, also required a halving in  $\tau$  in his model. We will try to improve the convection parameterizations for T106L90 simulations in the next study.

10) The conclusion. I feel add some clear statement to highlight the originality and novelty of this work would help audience get the full picture of this nice study. In the introduction and results discussion, authors made extensive comparisons with previous studies to evaluate the results of this study. Add some sentences to clearly state the new findings or improvements of this study will be helpful. For example, including nitrate which is missing in lots of previous studies, find more dust contribution by updating dust emission scheme and etc..

Thanks to the reviewer, we have added the following sentences to the end of the second last paragraph and the last paragraph of the conclusions in the revised manuscript, respectively: *“We also find that the emissions and convective transport of ammonia in the central main body of the Tibetan Plateau contribute significantly to the enhancement of gaseous  $\text{NH}_3$  in the UTLS over the Tibetan Plateau and entire ASM anticyclone area.*

*..... We find a larger dust contribution to the ATAL after updating the dust emission scheme. Our results are also consistent with the recently reported observation of enhanced nitrate (Höpfner et al, 2019), which appears to be missing in previous studies. Both the ATAL and Anti-ATAL characteristics are well simulated by our model in general”.*



**Figure A3.** Same as Fig. 9, but with  $\text{Na}^+$  being excluded from  $\text{Ca}^{2+,*}$ .

# Modelling the aerosol chemical composition of the tropopause over the Tibetan Plateau during the Asian summer monsoon

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**Abstract.** Enhanced aerosol abundance in the upper troposphere and lower stratosphere (UTLS) associated with the Asian summer monsoon (ASM), is referred to as the Asian Tropopause Aerosol Layer (ATAL). The chemical composition, microphysical properties and climate effects of aerosols in the ATAL have been the subject of discussion over the past decade. In this work, we use the ECHAM/MESSy Atmospheric Chemistry (EMAC) general circulation model at a relatively fine grid resolution (about 1.1×1.1 degrees) to numerically simulate the emissions~~and~~, chemistry and transport of aerosols and their precursors in the UTLS within the ASM anticyclone during the years 2010–2012. We find a pronounced maximum ~~in~~of aerosol extinction in the UTLS over the Tibetan Plateau, which to a large extent is caused by mineral dust emitted from the northern Tibetan Plateau and slope areas, lofted to an altitude of at least 10 km, and accumulating within the anticyclonic circulation. We also find that the emissions and convection of ammonia in the central main body of the Tibetan Plateau make a great contribution to the enhancement of gasphase NH<sub>3</sub> in the UTLS over the Tibetan Plateau and ASM anticyclone region. Our simulations show that mineral dust, water soluble compounds, such as nitrate and sulfate, and associated liquid water dominate aerosol extinction in the UTLS within the ASM anticyclone. Due to shielding of high background sulfate concentrations outside the anticyclone from volcanoes, a relative minimum of aerosol extinction within the anticyclone in the lower stratosphere is simulated, being most pronounced in 2011 when the Nabro eruption occurred. In contrast to mineral dust and nitrate concentrations, sulfate increases with increasing altitude due to the larger volcano effects in the lower stratosphere compared to the upper troposphere. Our study indicates that the UTLS over the Tibetan Plateau can act as a well-defined conduit for natural and anthropogenic gases and aerosols into the stratosphere.

## 1 Introduction

The upper troposphere and lower stratosphere (UTLS) is a transition region between the troposphere and stratosphere, ranging [about](#)  $\pm 5$  km around the tropopause (Gottelman et al., 2011), and the region plays an important role in the stratosphere-troposphere exchange (STE) and influences the chemistry of both the troposphere and stratosphere (Holton et al., 1995; Fueglistaler et al., 2009; Gottelman et al., 2011). Aerosols in the UTLS have large impacts on stratospheric ozone and [Earth's global climate system](#) through heterogeneous chemistry and radiative forcing (Solomon, 1999; Solomon et al., 2007; Solomon et al., 2011). By analyzing the Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation (CALIPSO) scattering ratio data, Vernier et al. (2011) found an Asian Tropopause Aerosol Layer (ATAL) associated with the Asian summer monsoon (ASM), i.e. an enhancement of background aerosol levels in the UTLS, between 13-18 km above sea level (a.s.l., hereafter all altitudes are referred to a.s.l. except when specified differently), over a widespread area of Asia (5-105 °E, 15-45 °N) including the Tibetan Plateau in July–August, for each year between 2006 and 2009. Thomason and Vernier (2013) confirmed the existence of the ATAL, beginning in 1999 with the Stratospheric Aerosol and Gas Experiment (SAGE II) satellite extinction coefficient data, while they stated that “there is no evidence of an ATAL in the SAGE II data prior to 1998”.

The ASM anticyclone is a typical circulation pattern in the UTLS during the Northern Hemisphere summertime, and the deep convection that leads to the ASM anticyclone plays a key role in the transport of pollutants from the boundary layer to the UTLS and their trapping within the anticyclone (Gottelman et al., 2004; Randel and Park, 2006; Park et al., 2007; Park et al., 2008; Randel et al., 2010). Satellite observations have shown that the concentrations of tropospheric trace gases (including H<sub>2</sub>O, CO, CH<sub>4</sub>, [NO](#), [NO<sub>2</sub>](#), C<sub>2</sub>H<sub>6</sub>, C<sub>2</sub>H<sub>2</sub>, [NO](#), [NO<sub>2</sub>](#), N<sub>2</sub>O, and HCN) are higher within than outside the anticyclone during the ASM period (Kar et al., 2004; Park et al., 2004; Li et al., 2005; Randel and Park, 2006; Park et al., 2007; Park et al., 2009; Xiong et al., 2009; Randel et al., 2010). Observations of trace gases like hydrogen cyanide (HCN), [being](#) a tropospheric pollutant [produced in from](#) biomass burning, reveals a combined influence of the ASM and human activities on chemical constituents in the UTLS region (Randel et al., 2010). Although the ATAL presence has been confirmed by measuring the aerosol optical properties, it remains a challenge to identify specific chemical components from satellites. General circulation models coupled with chemistry have been used to study the chemical composition and formation of the ATAL (Li et al., 2005; Fadnavis et al., 2013; Neely et al., 2014; Yu et al., 2015; Gu et al., 2016; Lelieveld et al., 2018).

Li et al. (2005) revealed the enhancement of anthropogenic aerosols within the ASM anticyclone by global chemical transport model (GEOS-Chem) simulation, and proposed that the ASM anticyclone could “trap” anthropogenic pollutants emitted and lofted from India and southwest China. Fadnavis et al. (2013) [investigated the transport of aerosols in the UTLS during the ASM using an aerosol chemistry climate model \(ECHAM5-HAMMOZ\), and found simulated](#) persistent maxima in black carbon (BC), organic carbon (OC), sulfate and mineral dust aerosols [in the UTLS](#) within the [ASM](#) anticyclone [in the UTLS throughout the ASM using an aerosol-chemistry-climate model \(ECHAM5-HAMMOZ\)](#) and showed that [boundary layer pollution is the source of this layer of UTLS aerosols layer is originated from the pollution sources in the](#)

boundary layer. Neely et al. (2014) studied the ATAL and its possible origin using an aerosol microphysical model coupled to a chemistry and climate coupled model (WACCM3). They argued that “the ATAL is most likely due to anthropogenic emissions, but its source cannot solely be attributed to emissions from Asia”. According to Neely et al. (2014), Chinese and Indian SO<sub>2</sub> emissions from China and India make about a contribute ~30% contribution of to the sulfate aerosol extinction in the ATAL during volcanically quiescent periods. Yu et al. (2015) explored the composition and optical properties of the ATAL using a sectional aerosol model coupled with a climate model (CESM1) coupled by a sectional aerosol model and found that “the ATAL is mostly composed of mixed organics and sulfates”. Gu et al. (2016) performed model simulations with GEOS-Chem, and their simulations showed elevated sulfate, nitrate, sulfate, ammonium, OBC and OC-BC in the UTLS over the Tibetan Plateau and ASM region throughout the summer as well. Gu et al. (2016) concluded that “nitrate aerosol is simulated to be of secondary importance near the surface but the most dominant aerosol species in the UTLS over the studied region”. Above all, the models could consistently simulate an enhancement of aerosols in the UTLS within the ASM anticyclone, but there is disagreement among these model results with respect to which chemical component(s) make a dominant contribution to and what are the most important source areas of the ATAL.

The Tibetan Plateau plays a critical role in the troposphere-to-stratosphere transport of water vapor and air pollutants during throughout the ASM (Zhou et al., 1995;Fu et al., 2006;Lelieveld et al., 2007;Yang et al., 2014;Yu et al., 2017). Zhou et al. (1995) proposed that the Tibetan Plateau might be an important pathway for the transport of pollutants in eastern Asia from the lower troposphere into the stratosphere during summertime while exploring the causes of ozone valley over the Tibetan Plateau. Subsequent model simulations and satellite data analyses confirmed this assumption and indicated that deep convection over the Tibetan Plateau and its southern slope is “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 acts as “a well-defined conduit”, where strong convection lofts air parcels in the boundary layer, mostly from the plateau and India/southeast Asia, into the ASM anticyclone (Bergman et al., 2013). A recent model study indicated that the ASM anticyclone serves as “an efficient smokestack”, venting aerosols with a substantial amount of organic and sulfates to the UTLS (Yu et al., 2017). While most studies have focused on the transport of pollutants originating from south and southeastern Asia to the UTLS over the Tibetan Plateau and within the ASM anticyclone, the influence of natural sources such as mineral dust and their interactions with pollution on the formation of ATAL has not been well addressed. The Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations (CALIPSO) and Multi-angle Imaging SpectroRadiometer (MISR) satellites detected the summertime dust aerosol plumes over the northwestern Tibet, most probably originating from the Taklamakan desert and lofted from the surface to an altitude of about 9 km around the source region (Huang et al., 2007;Huang et al., 2008;Xia et al., 2008). Further analysis of the CALIPSO and MISR satellite data indicated that “aerosols appear to be more easily transported to the main body of the Tibetan Plateau across the northern edge rather than the southern edge” and “dust is found to be the most prominent aerosol type on the Tibetan Plateau” (Xu et al., 2015). A 3-day case study using the WRF mesoscale model coupled with a dust module showed that deep convection in early summer over the Tibetan Plateau can inject dust aerosols into the stratosphere (Yang et al., 2014). The contribution of these dust plumes to



the total aerosols in the UTLS over the Tibetan Plateau and associated transport and transformation processes needs to be thoroughly investigated.

Volcanic eruptions are a large source of aerosols in the UTLS and can ~~significantly impact~~ influence the global climate significantly (Robock, 2000; Vernier et al., 2009; Solomon et al., 2011; Bourassa et al., 2012; Zhuo et al., 2014). From the global-scale viewpoint, the effects of volcanic eruptions on stratospheric aerosols mainly arise not from the injected ash but from the secondarily formed aerosols in the plumes; the former is composed of large size particles that are rapidly removed by gravitational sedimentation, while the latter are composed largely of dilute sulfuric acid droplets formed by the oxidation of injected SO<sub>2</sub> and have a long residence time in the atmosphere, typically one year or more in the stratosphere (Junge et al., 1961; Brock et al., 1995; Solomon et al., 2011). The Nabro volcano, located at the border between Eritrea and Ethiopia, northeastern Africa (13.37° N, 41.70° E, 2218 m a.s.l.), erupted on 12-13 June 2011, injecting approximately 1.3-2.0 Tg of SO<sub>2</sub> into the UTLS; the SO<sub>2</sub> and aerosol plumes with little amounts of ash were detected by different instruments onboard several satellites in a few weeks after the eruption (Bourassa et al., 2012; Clarisse et al., 2012; Bourassa et al., 2013; Fromm et al., 2013; Theys et al., 2013; Vernier et al., 2013; Clarisse et al., 2014; Fairlie et al., 2014). The eruption of Mt. Nabro in June 2011 ~~is has been considered as~~ the largest single injection of SO<sub>2</sub> to the UTLS ~~since~~ after the eruption of Mt. Pinatubo in June 1991 until now, and the dispersion of its plume to East Asia provides an actual case for examining the combined effects of the volcanic eruption and ASM anticyclone on the UTLS aerosols over the Tibetan Plateau and the ATAL (Fairlie et al., 2014).

In the present study, we investigate the chemical composition and source regions of aerosols in the UTLS over the Tibetan Plateau during the ASM, using the ECHAM/MESSy Atmospheric Chemistry (EMAC) model (Jöckel et al., 2006; Jöckel et al., 2010). The adopted model resolution is T106L90, which is finer than that typically used in previous studies (e.g., T42L31, ~~T42L90~~ or T63L90), allowing the model to reproduce the processes affected by the topography more realistically. Our model simulations cover the period from January 2010 to December 2012, for which SO<sub>2</sub> plumes from twenty-seven volcanic eruptions, including the Nabro eruption, had been detected by satellite and were implemented into the model. In addition, all other natural and anthropogenic sources of gases and aerosols, such as on-line calculated dust emissions, are taken into account. Comprehensive aerosol microphysical and gas/aerosol partitioning processes as well as a full chemical mechanism for both the troposphere and stratosphere implemented in EMAC are included in our simulation. Our study focuses on the seasonal mean spatial distributions of aerosol chemical components and associated optical properties in the UTLS during the ASM in the three simulation years. A detailed description of the model settings is given in Sect. 2, and the results and discussion are presented in Sect. 3. A summary of the main findings and conclusions is given in Sect. 4.

## 2 Model description and setup

The EMAC model is a chemistry-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 and predict atmospheric processes from the troposphere to middle atmosphere and their interactions with land and oceans. For this study, we used version 2.52 of EMAC, which includes the MESSy submodels describing various chemical, physical and dynamical processes in detail (Jöckel et al., 2005; Jöckel et al., 2010; Jöckel et al., 2016). EMAC has been extensively used at a range of spatial resolutions and evaluated against site, aircraft and satellite measurements of trace gases and aerosols in both the troposphere and stratosphere (Jöckel et al., 2006; Lelieveld et al., 2007; Pozzer et al., 2007; Pringle et al., 2010; Tost et al., 2010; Astitha et al., 2012; Brühl et al., 2012; Pozzer et al., 2012; Brühl et al., 2015; Eichinger et al., 2015; Pozzer et al., 2015; Jöckel et al., 2016; Tsimpidi et al., 2016; Abdelkader et al., 2017; Gottschaldt et al., 2017; Karydis et al., 2017; Bacer et al., 2018; Brühl et al., 2018; Gottschaldt et al., 2018; Khosrawi et al., 2018; Klingmüller et al., 2018; Lelieveld et al., 2018). The model spectral resolution used in this study is T106L90, which corresponds to a horizontal grid resolution of approximately  $1.125^\circ \times 1.125^\circ$  and 90 vertical layers extending from the surface to an altitude of 0.01 hPa (~80 km) with a vertical resolution of about 500m in the tropopause region. In this study, the model simulation was performed for the years 2010–2012. One year spin-up simulation was performed followed by three years of normal simulation for the period 2010–2012, and results from the latter are presented and discussed below.

EMAC simulates chemical reactions in the gas-phase ~~species~~ online through the Module Efficiently Calculating the Chemistry of the Atmosphere (MECCA) submodel (Sander et al., 2011). MECCA calculates the concentration of a range large number of gaseous species and radicals, including ~~aerosol-various gaseous~~ precursors ~~species of aerosols~~, such as SO<sub>2</sub>, NH<sub>3</sub>, nitrogen oxides (NO<sub>x</sub>  $\equiv$  NO + NO<sub>2</sub>), volatile organic compounds (VOCs), and dimethyl sulfide (DMS), and major oxidant species like OH, O<sub>3</sub>, H<sub>2</sub>O<sub>2</sub>, and NO<sub>3</sub>. 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 in a delta-two-stream method. The chemical mechanism used in this study is primarily based on stratospheric chemistry used by Brühl et al. (2015) plus VOC chemistry reported by Taraborrelli et al. (2012).

Aerosol microphysics and conversion between gaseous and aerosol specieses ~~gas/conversion~~ aerosol partitioning are treated by the Global Modal-aerosol eXtension (GMXe) module (Pringle et al., 2010), which uses 7 interacting lognormal modes (M7) to describe the ~~aerosol-size distributions~~ s of aerosols in the nucleation mode and hydrophilic and hydrophobic Aitken, accumulation and coarse modes. The aerosol composition ~~within each mode~~ is uniform in size within each mode (internally mixed), while it can vary ~~between different modes~~ from one mode to another (externally mixed). Each mode is represented ~~in terms of by~~ the number concentration ~~and~~, the number mean radius ~~together with and~~ its geometric standard deviation (Pringle et al., 2010).

Within the GMXe module, the nucleation schemes of Vehkamäki et al. (2002) was used, which calculates the nucleation of new particles as a function of temperature (for a range of 190 K < T < 305.15 K), relative humidity (for a range of 0.01% < RH < 100%) and the concentration of sulfuric acid (H<sub>2</sub>SO<sub>4</sub>). The primary OC aerosol is assumed to be emitted by 65% as hydrophilic and 35% as hydrophobic (Pringle et al., 2010). Following Pringle et al. (2010) and Pozzer et al. (2012),

secondary OC particles are directly emitted as primary OC. The organic aerosol formation and chemical aging ~~are~~ can be calculated by the ORACLE submodel ~~in EMAC~~ (Tsimpidi et al., 2014). ~~However, and~~ in this work the partitioning of secondary organic ~~aerosol-compound~~ between the gaseous and ~~particulate-aerosol~~ phases is not considered, ~~and the ORACLE module was not used in the simulation. The aging that leads the conversion of hydrophobic BC and OC to hydrophilic ones is realized by the collisions with hydrophilic particles and the condensation of inorganic gases such as sulfuric acid (Pringle et al., 2010 and references therein).~~

~~The inorganic aerosol composition-chemistry~~ is simulated by the ISORROPIA-II thermodynamic equilibrium model (Fountoukis and Nenes, 2007), with ~~updates as discussed in the work of latest version introduced by~~ Capps et al. (2012). ISORROPIA-II calculates the gas/liquid/solid equilibrium partitioning of the  $\text{Na}^+$ ,  $\text{K}^+$ ,  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{NH}_4^+$ ,  $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$ ,  $\text{Cl}^-$ , and  $\text{H}_2\text{O}$  aerosol system. ~~Calcium, Potassium, sodium, calcium and magnesium are considered-treated as chemically-chemically active composition of mineral dust and are assumed to crustal species existing in the form of 14 mineral salts (Ca(NO<sub>3</sub>)<sub>2</sub>, CaCl<sub>2</sub>, CaSO<sub>4</sub>, KHSO<sub>4</sub>, K<sub>2</sub>SO<sub>4</sub>, KNO<sub>3</sub>, KCl, MgSO<sub>4</sub>, Mg(NO<sub>3</sub>)<sub>2</sub>, MgCl<sub>2</sub>, NaHSO<sub>4</sub>, Na<sub>2</sub>SO<sub>4</sub>, NaNO<sub>3</sub>, NaCl) and four ions in the aqueous phase (Ca<sup>2+</sup>, K<sup>+</sup>, Mg<sup>2+</sup>, Na<sup>+</sup>). ISORROPIA-II solves for the equilibrium state by considering the chemical potential of the species and minimizes the number of equations and iterations required by considering specific compositional regimes.~~ To account for kinetic limitations, ~~ISORROPIA-II calculates the process of gas/aerosol partitioning is calculated in by~~ two stages (Pringle et al., 2010). ~~In the first stage, assuming that condensation is diffusion limited it calculates the amount of the gas-phase species that is able to kinetically condense onto the aerosol phase within the model time step is calculated, assuming diffusion limited condensation. In the second stage, ISORROPIA-II redistributes the derived mass is redistributed between the gas and the aerosol phase, with an assumption of assuming instant equilibrium between the two phases. In ISORROPIA-II aerosol liquid water (ALW) of the simulated aerosol compounds is determined based on the Zdanovskii-Stokes-Robinson (ZSR) relationship (Stokes and Robinson, 1996) of the simulated aerosol compounds, with the uptake limited by a grid box mean relative humidity of 95% of grid box mean relative humidity.~~

~~The loss of gas phase species to the aerosol through heterogeneous reactions of species on aerosol surfaces is calculated-are simulated~~ using the MECCA\_KHET submodel (Jöckel et al., 2010). MECCA\_KHET calculates the tropospheric heterogeneous reaction rates as mass transfer rates based on the aerosol surface density from GMXe (Jöckel et al., 2010). For the stratosphere the Multiphase Stratospheric Box Model (MSBM), which is coupled to MECCA\_KHET, takes the input from GMXe to calculate the heterogeneous reaction rates on polar stratospheric cloud (PSC) particles and on stratospheric aerosols (Jöckel et al., 2010; Kirner et al., 2011).

Aerosol extinction is calculated online by the AEROPT submodel based on Mie theory using pre-calculated look-up tables for the six aerosol components, including black carbon (BC), organic compounds (OC), mineral dust (DU), sea salt (sea spray), water-soluble species (WASO), i.e., all other water soluble inorganic species (e.g.,  $\text{NH}_4^+$ ,  $\text{HSO}_4^-$ ,  $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$ ), and ~~aerosol liquid water (ALW)~~ in the Aitken, accumulation, and coarse modes.

The uptake of  $\text{SO}_2$ ,  $\text{HNO}_3$  and  $\text{NH}_3$  and the aqueous-phase oxidation of  $\text{SO}_2$  by  $\text{H}_2\text{O}_2$  and  $\text{O}_3$  in cloud ~~droplets~~ are calculated by the SCAV submodel (Tost et al., 2006a; Tost et al., 2007a). The ~~removal-scavenging~~ of gases and aerosols

~~through by~~ wet deposition is calculated by the SCAV submodel (Tost et al., 2006a), ~~and their removal through~~ dry deposition is calculated by the DRYDEP submodel (Kerkweg et al., 2006), ~~and The SEDI submodel with a first-order trapezoid scheme is used to calculate~~ the sedimentation of aerosols ~~is calculated by the SEDI submodel~~ (Kerkweg et al., 2006) ~~using a first-order trapezoid scheme~~.

We used an emission inventory of the Representative Concentration Pathways scenario 8.5 (RCP8.5), namely ~~DLR-~~ MACCcity emission inventory, for fossil fuel combustion and biomass burning emissions in our simulation (see Jöckel et al. (2016) and references therein). The RCP8.5 global emission inventory has a horizontal grid resolution of  $0.5^\circ \times 0.5^\circ$  at monthly intervals and vertical distributions as described in Pozzer et al. (2009), and it is a “reasonable” choice for anthropogenic emissions over the period from 2000 to 2010 (Granier et al., 2011; Pozzer et al., 2015). The monthly cycle of the RCP8.5 emissions in 2010 are used in this study. The natural emissions of non-methane hydrocarbons (NMHCs) and  $\text{NH}_3$  are based on the Global Emissions Initiative (GEIA) database (Bouwman et al., 1997).  ~~$\text{NO}_x$  produced by lightning is calculated online and distributed vertically based on the parameterization of Price and Rind (1992)~~. The  $\text{NO}$  ~~emissions from~~ soils ~~emissions~~ are calculated online according to the algorithm of Yienger and Levy II (1995). Carbonyl sulfide (OCS) (Montzka et al., 2007) and other long-lived source gases are constrained by observed ~~monthly zonal average~~ surface volume mixing ratios ~~(in monthly zonal average value)~~, as ~~done~~ in Brühl et al. (2015). The oceanic dimethyl sulfide (DMS) emissions are calculated online by the AIRSEA submodel (Pozzer et al., 2006). ~~Emissions of s~~Sea spray aerosol ~~emissions~~ are ~~estimated from the~~ offline ~~using the~~ monthly emission data set of AEROCOM (Dentener et al., 2006), with a composition fraction of 55%  $\text{Cl}^-$ , 30.6%  $\text{Na}^+$ , 7.7%  $\text{SO}_4^{2-}$ , 3.7%  $\text{Mg}^{2+}$ , 1.2%  $\text{Ca}^{2+}$ , and 1.1%  $\text{K}^+$  (Seinfeld and Pandis, 2006).

~~Convection is parameterized by the submodel CONVECT, in which the~~ Tiedtke (1989) ~~scheme with modifications by~~ Nordeng (1994) ~~is set as default~~ (Tost et al., 2006b) ~~and used in this study. This scheme was developed for resolution T63. As discussed in Brühl et al. (2018), it overestimates vertical transport in the UTLS for T42 and underestimates it for T106. The  $\text{NO}_x$  emissions from lightning activity are calculated online using the submodel LNOX (Tost et al., 2007b). In this study we apply the parameterization developed by~~ Grewe et al. (2001), ~~which links the flash frequency to the updraft velocity. Here flash frequency obtained by this parameterization is scaled by a factor of 0.0695 for T106L90 simulations. In this study global emissions of  $\text{NO}_x$  from lightning are simulated to be 7.9, 6.7 and 6.3 Tg (N)  $\text{a}^{-1}$  for the years 2020, 2011 and 2012, respectively, falling into the range of 2–8 Tg (N)  $\text{a}^{-1}$  suggested by Schumann and Huntrieser (2007). The parameterisation schemes for convection (Tiedtke, 1989; Nordeng, 1994; Tost et al., 2006b) and lightning (Grewe et al., 2001) used in this study have been evaluated with simulations at coarser resolution (Tost et al., 2007b; Lopez, 2016; Gottschaldt et al., 2018) and the agreement is particularly noticeable for the ASM anticyclone region (Gottschaldt et al., 2018).~~

Dust emissions are calculated online with an advanced dust flux scheme, ~~which was~~ developed by Astitha et al. (2012) and updated recently by Klingmüller et al. (2018). The emission scheme combines meteorological parameters, which are derived online from the EMAC model, with soil properties including the ~~soil~~ clay fraction ~~of the soils~~, the rooting depth and the vegetation area index, which are prescribed at monthly intervals based on the latest satellite observations (Astitha et al., 2012; Klingmüller et al., 2018). The dust particles ~~are considered~~ ~~asturn to be~~ mobilized in the air when the wind friction

velocity exceeds a threshold value, which is determined by the ~~soil particle size distribution and~~ soil texture classification ~~and soil particle size distribution~~. ~~Considering the regional differences in the soil particle size distribution,~~ ~~t~~The emission scheme does provide an explicit variant (DU\_Astitha2) ~~to additionally accounts for regional differences in the emitted soil particle size distribution~~ based on soil characteristics ~~in every of each model~~ grid cell (Astitha et al., 2012). In the present study, we chose an alternative variant (DU\_Astitha1), which assumes that the dust particles emitted from different soils have the same size originally. Compared to DU\_Astitha2, the DU\_Astitha1 scheme can achieve competitive results with reduced complexity and has been used in recent studies as well (Abdelkader et al., 2015; Abdelkader et al., 2017; Klingmüller et al., 2018). We used the DU\_Astitha1 in combination with new ~~input data set comprising~~ land cover classification, vegetation, clay fraction and topography ~~data set, based on up-to-date observations,~~ which has been implemented by Klingmüller et al. (2018) ~~based on up-to-date observations and can well represent to account for~~ the rapid changes of deserts and semi-arid regions in recent decades. Emissions of individual crustal species are ~~calculated~~ ~~estimated~~ as a constant fraction of ~~general~~ mineral dust emissions, and this fraction can vary with the different dust source regions (Karydis et al., 2016). In this study, spatially uniform mineral dust composition is assumed, with a crustal species fraction of 1.2% Na<sup>+</sup>, 1.5% K<sup>+</sup>, 2.4% Ca<sup>2+</sup>, and 0.9% Mg<sup>2+</sup> in the dust (Sposito, 1989).

As in the work of ~~Brühl et al. (2015) and~~ Brühl et al. (2018), the volcanic SO<sub>2</sub> plumes detected by the Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) satellite were used to account for the volcanic SO<sub>2</sub> emissions in the simulation of this study. MIPAS (Fischer et al., 2008) is a limb sounder on board the Envisat sun-synchronous polar orbiting satellite ~~Envisat~~, which was launched on 1 March 2002 and lost contact with the ground on 8 April 2012. Global distributions of SO<sub>2</sub> ranging from the upper troposphere to 20 km altitude have been retrieved from the thermal radiation (685–2410 cm<sup>-1</sup>) emitted by the atmosphere as measured by MIPAS ~~(Höpfner et al., 2015),~~ and 3-D fields of volcanic SO<sub>2</sub> plumes in the UTLS with a temporal resolution of 5 days were derived ~~from 3-dimensional data fields of MIPAS~~ (Höpfner et al., 2015) ~~with a temporal resolution of 5 days~~. Since MIPAS cannot detect very fresh plumes as the field of view is obscured by ash, in most cases two to six subsequent 5-day periods are used for extrapolation of the initial spatial SO<sub>2</sub> distribution. As shown in Fig. 1a, there were about 27 explosive volcanic eruption events occurring over the period of January 2010 through March 2012. The highest SO<sub>2</sub> mixing ratio of >1 nmol mol<sup>-1</sup> (or ppbv) was detected in the Nabro eruption plume. As can be seen in Fig. 1b, peak ejections generally occurred at altitudes of about 16-17 km, i.e., around the thermal tropopause (defined by the World Meteorological Organization (WMO)). The volume mixing ratios of the volcanic SO<sub>2</sub> plumes from MIPAS were added to the simulated background SO<sub>2</sub> mixing ratios at the ~~time of the eruption~~ time by using the restart technique implemented in EMAC. Due to limitations of MIPAS data, no volcano eruption is considered in our simulation for the period after March 2012. There were two large major volcanic eruptions occurring in 2012, namely Soputan (124.73 °E, 1.11 °N) on 18 September 2012 and Copahue (288.8 °E, 37.86 °S) on 22 December 2012, with the amounts of ejected SO<sub>2</sub> being 1-2 orders of magnitude smaller than that of Nabro, as reported by Mills et al. (2016). These two volcanic eruptions are not included for the simulations of this study. The biases of the MIPAS SO<sub>2</sub> data were estimated to be within 50 pmol mol<sup>-1</sup> (or pptv) in case of the volcanically enhanced concentrations (Höpfner et al., 2015). Detailed information on the name,

time, location and the amount of injected SO<sub>2</sub> for individual eruption can be found in the work of Bingen et al. (2017) and (Brühl et al., 2018) and references therein.

In the simulation, the meteorology was nudged by Newtonian relaxation towards the European Centre for Medium-Range Weather Forecasts (ECMWF) operational re-analysis data for the years 2010–2012. Temperature, vorticity, divergence and surface pressure are ~~The prognostic variables~~ nudged towards the realistic meteorological conditions ~~(i.e., the re-analyses)~~ are temperature, vorticity, divergence and surface pressure, and the nudging weights are chosen such that the mesosphere and upper stratosphere and the boundary layer are not directly influenced (Lelieveld et al., 2007). The nudging was exerted with maximum weights at the model levels from 37 (~ 10 hPa) to 93 (~706 hPa), leaving the highest thirty (upper middle atmosphere) and the lowest three (boundary layer) model levels free (apart from surface pressure). as used by Jöckel et al. (2016). The chemical initial conditions for this model study were provided by the results from a previous simulation of EMAC T42L90 (Brühl et al., 2015) for all gases and aerosols, except for NMHCs which were from an EMAC T106L31 simulation (Pozzer et al., 2012). ~~One year spin-up simulation was performed followed by three years of normal simulation for the period 2010–2012, and results from the latter are presented and discussed below.~~ Pozzer et al. (2012) used the same modules (e.g., GMXe and ISORROPIA-II for aerosols) as in this study but different emission data (e.g., dust emissions from AEROCOM) in the T106L31 simulations of tropospheric aerosols for the years 2005–2008. By comparing the model output with observations from different measurement networks and satellite remote sensing instruments, they found that the main spatial and temporal atmospheric distribution of sulfate, ammonium and nitrate aerosols were well reproduced in general, but there was an underestimation of aeolian dust emissions in the dust outflow regions (Pozzer et al., 2012). Klingmüller et al. (2018) develop an advanced dust emission scheme, with the updated land cover classification, the inclusion of the topography factor and the modification of the sandblasting efficiency function. They performed the T106L31 simulations for the year 2011; by comparing with the aerosol optical depth (AOD), dust concentrations and deposition results from various observational platforms, they concluded that the update significantly improves agreement with the observations (Klingmüller et al., 2018). With respect to aerosol and stratospheric chemistry, the model setup in this study is similar to those used in Brühl et al. (2015, 2018), the latter of which (Brühl et al., 2018) used the T106L90 resolution for a 1-year sensitivity test.

### 3 Results and Discussion

#### 3.1 General features

The ASM circulation is characterized by cyclonic flow and convergence in the lower troposphere, and a strong anticyclone and divergence in the UTLS (Krishnamurti and Bhalme, 1976); its structure is primarily a response to diabatic heating associated with deep convection (Hoskins and Rodwell, 1995). Known as the “sensible heat ~~bumpump~~”, the Tibetan Plateau modifies monsoon circulation (Wu and Zhang, 1998) and even modulates large-scale atmospheric circulations over the Northern Hemisphere (Zhao and Chen, 2001; Zhou et al., 2009). The strength and position of the ASM anticyclone can be



represented by different parameters, such as the circulation stream function (Randel and Park, 2006; Park et al., 2007), geopotential height (Bergman et al., 2013; Pan et al., 2016; Li et al., 2017) or potential vorticity (Garny and Randel, 2013; Ploeger et al., 2015). In this study we use the pressure deviation ( $dP$ ) as a measure, defined as the difference of pressure,  $P$ , at each model grid relative to their regionally averaged value over 20–140 °E and 10–45 °N,  $P_{avg}$ , at the same altitude. We used the  $dP$  value of 1 hPa (and 1.3 hPa) to plot the contour lines. This selection is arbitrary, but it appears to work well as demonstrated below. Note that the ASM circulation is not steady but exhibits substantial intra-seasonal variability, with northward propagation during the onset phase of the monsoon and westward propagating features during the mature phase (Krishnamurti and Ardanuy, 1980; Yasunari, 1981). There is eddy shedding to the east and to the west of the ASM anticyclone. Here we focus on the seasonal averaged characteristics associated with the ASM.

Figure 2 shows EMAC simulated horizontal distributions of  $dP$  overlaid by the horizontal wind field,  $O_3$  and  $CO$ , averaged for July–August of 2010, 2011 and 2012, at altitudes of 15, 16, 17 and 18 km, respectively. It can be seen that the general characteristics of the ASM anticyclone are well represented by EMAC. The anticyclone is very strong in the upper troposphere (at 15 km), and it is still clearly visible in the lower stratosphere (e.g., at 18 km) with the core area shifting northerly with increasing altitude. As found in early studies (e.g., Highwood and Hoskins, 1998), the ASM tropopause is relatively high, with a maximum around 17 km (corresponding to about 92–95 hPa) over the area of approximately 30–120 °E and 20–35 °N, including the Iranian Plateau to southern Tibetan Plateau. The air masses in the ASM anticyclone are characterized more by tropospheric than stratospheric properties. This feature has been well reproduced by EMAC using  $O_3$  ~~for as a~~ stratospheric tracer and  $CO$  ~~for as a~~ tropospheric tracer, as ~~demonstrated also done in previous other model studies~~ (e.g., Gettelman et al., 2004; Park et al., 2007; Park et al., 2009; Barret et al., 2016; Santee et al., 2017). It was shown recently that EMAC realistically simulates reactive gases and radicals within the ASM anticyclone by comparing the simulation results with aircraft measurements (Lelieveld et al., 2018).

As a general overview, we show the simulated global distributions of the tropospheric ~~column~~ burden of various aerosol components, including BC, OC, dust, sea spray, sulfate, nitrate, ammonium and ALW for July–August ~~2011–2012~~ in Fig. 3 (the same global distributions of these aerosols are presented in Figs. S1 and S2 for the years 2010 and ~~2012~~2011). The geographic distribution patterns of the column integrated aerosol properties are similar to those given in the work of Pringle et al. (2010), who also used EMAC but with a different grid resolution (T42L19) and for an earlier simulation period (2001–2002). However, the concentration levels of anthropogenic aerosols, especially sulfate, over the Middle East, India and northeast China, are significantly higher in our simulations due to increasing emission trends over the period of 2001 to 2010 (Granier et al., 2011). The concentration levels of dust aerosols from our simulation are similar to those of Pringle et al. (2010), but there are some differences in the fine structure, probably due to different meteorological conditions for different periods and/or an improved scheme used in our simulation (Klingmüller et al., 2018).



### 3.2 Aerosol extinction

In situ measurements of aerosols in the UTLS over the Tibetan Plateau have been very sparse. Tobo et al. (2007a, b) firstly observed relatively high number concentrations of sub-micron size aerosols (with radii of 0.15–0.6  $\mu\text{m}$ ) near the tropopause region (between about 130 and 70 hPa) during the ASM period ~~using-with~~ a balloon-borne optical particle counter at Lhasa, China in 1999. He et al. (2014) found a 3–4 km thick aerosol layer in the UTLS peaking at an altitude of 18–19 km (1–2 km above the tropopause), by measuring the aerosol extinction vertical profiles ~~of the aerosol extinction coefficient with using~~ a micro-pulse lidar at Naqu, China in July–August 2011. Recently, Yu et al. (2017) further revealed a widespread enhanced aerosol layer extending from several kilometres below the tropopause up to 2 km above the tropopause within the ASM anticyclone, by measuring the vertical profiles of particle surface area density using an optical particle spectrometer in balloon soundings from Kunming, China, in August 2015. By balloon-borne measurements, Brunamonti et al. (2018) found the maximum aerosol backscatter occurring at the cold-point tropopause, revealing the thermodynamically significant levels of the ATAL. Figure 4a shows EMAC simulated temporal variation in the vertical profile of aerosol extinction over Naqu in August 2011, which coincided with the period of aerosol vertical profile measurements at Naqu reported by He et al. (2014). It can be seen that EMAC predicts persistently enhanced aerosol abundance between 17–20 km altitude, with maxima occurring at 18–19 km; aerosols near the tropopause have a higher variability in the upper troposphere than in the lower stratosphere. A comparison of the model simulated monthly mean aerosol vertical profile with lidar measurements of He et al. (2014) is presented in Fig. 4b. The aerosol vertical profiles from measurements are shown to have a high variability, especially at high altitudes (i.e., 18–19 km), and simulated aerosol extinction coefficients tend to be at the lower end of the measured range at 18 km altitude and downward.

The ATAL was found by detecting the aerosol optical properties in the UTLS during the ASM (Vernier et al., 2011; Thomason and Vernier, 2013). In this study we also investigate the geographic distributions of model simulated aerosol optical properties together with the associated chemical composition. Figure 5 shows EMAC simulated aerosol extinction coefficients ( $K_e$ ) at altitudes of 15, 16, 17 and 18 km, averaged for July–August of 2010, 2011 and 2012. Unexpectedly, the ATAL, characterized by extensively enhanced aerosol concentrations within the ASM anticyclone as shown by Vernier et al. (2011) and Thomason and Vernier (2013), was not fully reproduced by our simulation. Only over the Tibetan Plateau can the maxima of  $K_e$  in the horizontal direction be found, most clearly at 15–16 km altitude (in the upper troposphere) and still visible at 17 km (in the lower stratosphere) in 2010 and 2012. Although the absolute  $K_e$  values at 15 km altitude over the Tibetan Plateau are at the same levels in 2011 as those in 2010 and 2012, the enhancement of aerosols is partly shielded by higher  $K_e$  values outside the anticyclone (especially at higher latitudes) in 2011. Aerosols outside the anticyclone increased dramatically from the upper troposphere to the lower stratosphere, and as a result even a relative minimum of  $K_e$  can be seen within the ASM anticyclone at 17 km in 2011. Previous studies have shown that the Nabro volcano eruption in 2011 has a large impact on stratospheric aerosols over East Asia and the Northern Hemisphere (Sawamura et al., 2012; Uchino et al., 2012). The high  $K_e$  values outside the anticyclone in 2011 can be attributed to the effect of the Nabro volcano eruption,

which has been taken into account in our model simulation (see Fig. 1 and Sect. 2 above). Note that this “anti-ATAL” phenomenon was also detected by the CALIPSO satellite for July-August 2009 (see Fig. 1d in the work of Vernier et al. (2011)), when a few months earlier the eruption of Sarychev volcano had injected a large plume of SO<sub>2</sub> above the tropopause (Vernier et al., 2011).

5 Mineral dust, WASO and ALW compounds are the dominant aerosol components contributing to aerosol extinction, and BC, OC, and sea spray ~~(SS)~~ have very small (1–2 orders of magnitude lower)  $K_e$  values in the UTLS within the ASM anticyclone (see Figs. S3, S4 and S5). Figures S6, 6, and 7 show the percent contributions of mineral dust, WASO and ALW to the total  $K_e$  values at altitudes of 15, 16, 17 and 18 km in July–August of 2010, 2011 and 2012. The contributions from BC, OC, and ~~SS~~ sea spray are not shown due to their small values (less than 5% over the Tibetan Plateau). By comparing the  
10 chemical aerosol patterns with those of the total  $K_e$  shown in Fig. 5, we can see that the broad maxima of aerosol extinction at 15–16 km over the Tibetan Plateau are caused firstly by mineral dust, which contributes approximately 30–60%, and secondly by WASO and associated ALW, which contribute comparably with about 10–40%. There are two maxima in the absolute contribution of WASO to the total  $K_e$  in the upper troposphere within the anticyclone in both 2010 and 2012: the first occurring in the eastern part of the anticyclone over the (eastern) Tibetan Plateau, accompanied by a maximum in the  
15 ALW part of  $K_e$ , and the second in the western part over the Iranian Plateau with no maximum in the extinction by ALW.

High aerosol extinction outside the ASM anticyclone was caused by both WASO and ALW, with WASO making a predominant contribution (typically 50–70%) to the north of the anticyclone and ALW contributing dominantly (up to 60–80%) to the south of it. In contrast to the absolute  $K_e$  from mineral dust, which is at the same level in the three years, the absolute  $K_e$  contributed by WASO as well as ALW is much higher in 2011 than in 2010 and 2012, especially in the lower  
20 stratosphere (e.g., at 17 km), leading to the “anti-ATAL” phenomenon as pointed out above. [The pronounced Nabro eruption of SO<sub>2</sub> in 2011 can enhance sulfuric acid and sulfate aerosols in the UTLS, and increase the contributions of WASO and ALW to the aerosol in the UTLS, especially in the lower stratosphere and outside the ASM cyclone, as shown and discussed in the following section.](#)

### 3.3 Ionic aerosols in equivalent

25 As described in Sect. 2, ionic aerosol composition, including Na<sup>+</sup>, K<sup>+</sup>, Ca<sup>2+</sup>, Mg<sup>2+</sup>, NH<sub>4</sub><sup>+</sup>, SO<sub>4</sub><sup>2-</sup>, HSO<sub>4</sub><sup>-</sup>, NO<sub>3</sub><sup>-</sup>, and Cl<sup>-</sup> in the liquid phase and their salts in the solid phase, are calculated by the ISORROPIA-II thermodynamic equilibrium model implemented in EMAC. Investigation of these ionic species is helpful for understanding the sources and formation pathways of the aerosols. Figures S7, 8, and 9 present EMAC simulated water soluble ions in the accumulation mode at altitudes of 15, 16, 17 and 18 km for July–August of 2010, 2011 and 2012. Note that (1) the accumulation mode is considered here since  
30 nearly all extinction (>99%) is caused by aerosols in this mode; (2) we use “equivalent” (eq) rather than “mole” (mol) or “gram” (g), to express the amount of ions, considering that it may be used more easily to explore the possible formation of a chemical compound from different cations and anions (e.g., 1 mol NO<sub>3</sub><sup>-</sup> = 1 eq NO<sub>3</sub><sup>-</sup>, and 1 mol SO<sub>4</sub><sup>2-</sup> = 2 eq SO<sub>4</sub><sup>2-</sup>; (3) Cl<sup>-</sup> is not shown here due to its low concentrations (e.g, two orders of magnitude smaller than that of NO<sub>3</sub><sup>-</sup>) in the investigated

regions; (4) for simplicity,  $\text{Na}^+$ ,  $\text{K}^+$  and  $\text{Mg}^{2+}$  together with  $\text{Ca}^{2+}$  in the mineral dust are accounted for as  $\text{Ca}^{2+,*}$  in the equivalent amounts since  $\text{Ca}^{2+}$  is a typical ionic tracer for mineral dust (Ma et al., 2003). Note that these four ionic species are also contained in sea salt and generally  $\text{Na}^+$  is taken as a tracer for sea salt. However, mineral dust appears to be a dominant source and also for all these crustal species in the UTLS over the Tibetan Plateau since they are well correlated with the dust plume in the UTLS over the Tibetan Plateau and the concentrations of  $\text{Ca}^{2+}$  are much higher than those of  $\text{Na}^+$  there.

As shown in Figs. S7, 8, and 9, the levels of  $\text{NO}_3^-$ ,  $\text{NH}_4^+$ , and  $\text{Ca}^{2+,*}$  are enhanced at 15–16 km altitude (in the upper troposphere) within the ASM anticyclone in all three years, and the enhancements are still visible at 17 km (in the lower stratosphere) in 2012. Similar to the WASO contributed  $K_e$  in 2010 and 2012 (Figs. S3 and S5), there are two maxima of  $\text{NO}_3^-$  and  $\text{NH}_4^+$ , one in the eastern part of the anticyclone (over the eastern Tibetan Plateau), and another in the western part (over the Iranian Plateau and the surrounding area). A maximum of  $\text{Ca}^{2+,*}$  is found over the Tibetan Plateau, coinciding with that of the dust contributed  $K_e$  shown in Figs. S3–S5. In contrast,  $\text{SO}_4^{2-}$ ,  $\text{HSO}_4^-$ , and  $\text{H}^+$  are found to be higher outside the anticyclone than inside, mainly due to increased stratospheric sulfate aerosols by the volcanic eruptions, especially in 2011 when the Nabro eruption occurred (Fairlie et al., 2014). The ASM anticyclone not only traps tropospheric pollutants inside but also efficiently to some extent blocks intrusions of stratospheric ozone and aerosols from outside. It should be noted that here we only investigate a seasonal mean aspect of the ASM anticyclone. Due to the dynamical instabilities of the ASM anticyclone, entrainment of stratospheric tracers does occur frequently, approximately twice a month (Gottschaldt et al., 2018).

The maximum concentrations of  $\text{NH}_4^+$  and  $\text{NO}_3^-$  are shown to occur in the same areas and at the same levels, indicating the existence of  $\text{NH}_4\text{NO}_3$  in the upper troposphere (at 16 and 17 km) and even in the stratosphere (e.g., at 17 km in 2012) within the ASM anticyclone. The areas of high  $\text{NH}_4^+$  are shown to be wider spread than those of high  $\text{NO}_3^-$  within the anticyclone, suggesting that some  $\text{NH}_4^+$  ions are in the form of  $(\text{NH}_4)_2\text{SO}_4$  and partly in  $\text{NH}_4\text{HSO}_4$  (e.g., at 17 km in 2011) in the corresponding areas. The levels of nitrate ( $\text{NO}_3^-$ ) and sulfate ( $\text{SO}_4^{2-}$ ) within the anticyclone can be either similar or rather different from each other, depending on the altitude and simulation year. For example, higher  $\text{NO}_3^-$  than  $\text{SO}_4^{2-}$  is simulated at 15 km altitude within the anticyclone, in particular over the western area of the Iranian Plateau; at 17 km, the simulated  $\text{NO}_3^-$  concentrations within the anticyclone are still higher in 2012, but are much smaller than those of  $\text{SO}_4^{2-}$  in 2010 and 2011. Due to the effect of the Nabro eruption, higher  $\text{SO}_4^{2-}$ ,  $\text{HSO}_4^-$  and  $\text{H}^+$  concentrations are found in the year 2011 than in 2010 and 2012 over the ASM region and the whole northern hemisphere, most profoundly in the lower stratosphere. The distribution patterns of  $\text{SO}_4^{2-}$  and  $\text{HSO}_4^-$  indicate that considerable fractions of sulfate aerosols can enter into the anticyclone from outside, mainly through the northern and eastern edges; these sulfate aerosols might be quite acidic in the lower stratosphere, as indicated by high  $\text{H}^+$  concentrations, resulting in a deficiency of  $\text{NO}_3^-$  in the aerosol phase within the anticyclone (e.g., at 17 km in 2011).

In addition to ammonia, the reaction with mineral dust can also provide a pathway for nitrate formation due to neutralization by the crustal cations such as  $\text{Ca}^{2+}$  (Ma et al., 2003). An investigation of the fine distribution structures of

$\text{NO}_3^-$ ,  $\text{NH}_4^+$ , and  $\text{Ca}^{2+,*}$  over the Tibetan Plateau shows that large residuals of  $\text{NO}_3^-$  minus  $\text{NH}_4^+$  exist in some areas (e.g., at 15–16 km in 2010 and 2012), where the maxima of  $\text{Ca}^{2+,*}$  occur as well. This finding indicates that multiphase reactions of gaseous nitric acid ( $\text{HNO}_3$ ) have taken place on the surface of mineral dust, leading to the formation of  $\text{Ca}(\text{NO}_3)_2$  or analogs in the upper troposphere over the Tibetan Plateau. Considerable amounts of  $\text{Ca}^{2+,*}$  are also found in the lower stratosphere (e.g., at 17 km in 2010 and 2011) within the ASM anticyclone; however, these  $\text{Ca}^{2+,*}$  cations should exist in the form of  $\text{CaSO}_4$  (gypsum) or analogs since  $\text{NO}_3^-$  is deficient. As demonstrated in previous studies, lightning  $\text{NO}_x$  clearly dominates the  $\text{NO}_x$  budget from the tropopause to 100 hPa below it, and its emissions are much stronger in the Tibetan part (Gottschaldt et al., 2018). Lightning  $\text{NO}_x$  also plays a central role in sustaining upper tropospheric OH concentrations over the monsoon (Lelieveld et al., 2018). In our simulations, lightning  $\text{NO}_x$  emissions within the ASM anticyclone (20–140 °E and 10–45 °N) are very intensive, with estimated values of 1.1, 0.5 and 0.9 Tg (N)  $\text{a}^{-1}$  in the years 2010, 2011 and 2012, respectively. High  $\text{NO}_x$  and OH concentrations are in favour of  $\text{HNO}_3$  formation via the reaction  $\text{NO}_2 + \text{OH}$ , which may provide an important source of nitrate within the anticyclone. Tost (2017) found 60% of aerosol nitrate between 500 hPa and the tropopause being produced from lightning in the ASM and its outflow under the present climatic condition. In addition to the available nitric acid, other factors such as neutralising ions (e.g.,  $\text{Ca}^{2+}$ ,  $\text{NH}_4^+$  and  $\text{SO}_4^{2-}$ ) and temperature can also influence the amount of nitrate aerosols in the ATAL. How these factors and associated processes affect nitrate aerosols in the UTLS over the Tibetan Plateau will be investigated thoroughly in a future study.

### 3.4 Aerosol mass concentrations

Figures S8, 10 and 11 present EMAC simulated mass concentrations of aerosols in the accumulation mode, including specifically BC, OC, mineral dust, sulfate, nitrate, ammonium, and ALW at altitudes of 15, 16, 17 and 18 km, averaged for July–August 2010, 2011 and 2012. For simplicity, the the crustal species  $\text{Ca}^{2+}$ ,  $\text{Na}^+$ ,  $\text{K}^+$  and  $\text{Mg}^{2+}$  are not shown individually, considering that these ionic species within the ASM anticyclone they are originate predominantly from attributed to mineral dust, having the same constant relative fractions as their emission sources do (see Sect. 2 above) and cannot be easily distinguished from dust in chemical filter sample analysis. Sulfate encompasses both  $\text{SO}_4^{2-}$  and  $\text{HSO}_4^-$ , the latter at relatively low concentrations, and is thus expressed as  $\text{SO}_4^{2-}$  as well. Other species such as sea spray components are not shown due to their extremely low mass concentrations in the UTLS over the Tibetan Plateau. The geographical distribution features of major aerosols in the UTLS over the Tibetan and within the ASM anticyclone have been presented by their extinction or ionic species in Sects. 3.2 and 3.3. Here we use aerosol mass concentrations for two purposes: (1) to further quantitatively evaluate the relative importance of different aerosol components over the Tibetan Plateau; (2) to make a comparison of EMAC simulated major chemical components with previous studies, where mass concentrations were widely used.

It is shown that mineral dust, sulfate and nitrate are three major components of dry aerosols in the UTLS within the ASM anticyclone. Ammonium is not mentioned here considering that it can exist in the form of sulfate or nitrate, as discussed in Sect. 3.3. Over the Tibetan Plateau mineral dust is simulated to be a dominant component in the UTLS, except for the lower stratosphere in 2011 when the Nabro eruption increased sulfate aerosols dramatically, which is in agreement with

measurements by He et al. (2014). Li et al. (2005) predicted an enhancement of aerosols within the ASM anticyclone using GEOS-Chem, which considered only BC, OC and sulfate aerosols. Neely et al. (2014) only considered sulfate aerosols in their model study of the ATAL using WACCM3. They demonstrated that if moderate volcanic activity was included in WACCM3, aerosol extinction from sulfate could be simulated to be higher outside than inside the anticyclone (see Fig. 5 of Neely et al. (2014) ), in good agreement with the simulation results from this study.

Fadnavis et al. (2013) simulated aerosols in the UTLS during the ASM using ECHAM5-HAMMOZ (with the resolution T42 corresponding to  $2.75^\circ \times 2.75^\circ$  in the horizontal dimension), into which M7 (the same as in the present study) had been implemented to describe the size distribution of aerosols, including BC, OC, sulfate, sea salt and mineral dust, but without considering nitrate (which has been included in EMAC and used in the present study). Of all aerosol types around the tropopause within the ASM anticyclone, mineral dust was simulated to occur at highest concentrations, followed by sulfate (see Fig. 1 of Fadnavis et al. (2013)). This result is in fair agreement with the order of major components derived in this study, although nitrate was not considered in the work of Fadnavis et al. (2013). Fadnavis et al. (2013) predicted a maximum seasonal mean dust aerosol concentration of  $\sim 30 \text{ ng m}^{-3}$  at 110 hPa, much lower than the value ( $>100 \text{ ng m}^{-3}$ ) at 16 km over the Tibetan Plateau as simulated in this study (Figs. S8, 10 and 11), possibly related not only to the fact that the updated EMAC dust emission scheme takes the topography into account, which has been shown to enhance the emissions from basins and valleys, in better agreement with observations (Klingmüller et al., 2018), but also to the higher resolution used in this study. The geographic distribution pattern of sulfate from our simulation is quite different from that of Fadnavis et al. (2013); ~~since most likely due to the fact that the volcano eruption effect~~ volcanic sulfate was most likely not considered in their simulation.

Yu et al. (2015) explored the composition and optical properties of the ATAL using CESM1 (with  $2^\circ$  horizontal resolution) coupled with a sectional aerosol microphysics model including primary emitted organics, secondary organics, dust, sea salt, black carbon, and sulfate. Yu et al. (2015) predicted an aerosol enhancement mainly composed of mineral dust extending from the surface up to 13 km above Africa. However, although mineral dust was included in their simulation, Yu et al. (2015) suggested the ATAL (between 100–230 hPa) to be mostly composed of organics (roughly 60%) and sulfate (roughly 40%) by mass given 2010 emissions (See Fig. 3 of Yu et al. (2015)). Our simulation results show that OC has a much smaller contribution to the ATAL than either dust or sulfate (or nitrate), in contrast to Yu et al. (2015). Similar to the comparison with Fadnavis et al. (2013) mentioned above, the different dust emission scheme and model resolution used by Yu et al. (2015) might lead to significant discrepancy of dust concentration in the UTLS within the ASM anticyclone compared to this study. Moreover, Yu et al. (2015) did not take into account stratospheric volcanic emissions and nitrate aerosols in their simulation, which might underestimate the contribution of inorganic aerosols to the ATAL with respect to this study.

Gu et al. (2016) performed model simulations using GEOS-Chem (with ~~a horizontal resolution of~~  $2^\circ$  latitude by  $2.5^\circ$  longitude horizontal resolution), which ~~has a fully coupled treatment of~~ considers detailed tropospheric chemistry ~~and~~

~~coupled with~~ aerosols including ~~BC, OC,~~ sulfate, nitrate, ammonium, ~~BC, OC,~~ sea salt and mineral dust. They showed elevated sulfate, nitrate, ammonium, BC and OC in the UTLS over the ASM region ~~throughout the~~ summer and concluded that nitrate ~~aerosol~~ is the most dominant aerosol ~~species composition~~ in the UTLS over the studied region. In the work of Gu et al. (2016), concentrations of mineral dust were simulated to be 5.0–7.0 ng m<sup>-3</sup> over the studied region and contribute less than 5.0% to total aerosol mass at 100 hPa. It turns out that there is a large discrepancy between Gu et al. (2016) and this study regarding the contribution of mineral dust to the enhancement of aerosols over the Tibetan Plateau and ASM region. Similar to Gu et al. (2016), we also simulate higher nitrate at 16–18 km within the ~~ACM-ASM~~ anticyclone in one of the simulated years, i.e. 2012, after March when volcanos are considered as quiescent arbitrarily in our simulation due to limited MIPAS data. However, the distribution patterns of nitrate in the UTLS are rather different between the two studies. Firstly, we found two nitrate maxima, located over the eastern and western parts of the anticyclone, respectively, which was not simulated (or shown clearly) by Gu et al. (2016). Secondly, an elongated, very high nitrate belt at the northern edge of the anticyclone around 40°N, as shown in Fig. 7c of Gu et al. (2016), is not reproduced by our simulation. Note that persistent and widespread more acidic conditions outside the anticyclone, as simulated in this study by including the volcanic SO<sub>2</sub> eruption effects, do not favor the presence of ammonium nitrate that was reported by Gu et al. (2016).

### 3.5 ~~Dust~~ Transport of dust, ammonia and water vapor

~~As shown above~~ According to our EMAC simulations, mineral dust aerosols in the accumulation mode make a major contribution to the total aerosol extinction in the UTLS with the ASM anticyclone, contributing predominantly to the maximum  $K_e$  at 15–16 km over the Tibetan Plateau. Here we investigate the spatial distribution of these dust aerosols to understand from where they originate. Figure 12 presents EMAC simulated dust mass concentrations in the accumulation mode at different altitudes from the surface to 18 km, averaged for July–August ~~2011~~ 2012. Although there are some small differences in the fine structure of the mass concentration distribution, the seasonally averaged transport patterns of dust over the Tibetan Plateau and the surrounding area are the same in 2010 and ~~2012~~ 2011 (see Figs. S9 and S10) as in ~~2011~~ 2012. The geographical distribution of mineral dust at the surface shows that the emission sources are widely spread, e.g. from northern Africa, the Arabian Peninsula and northern India, to western China and Mongolia. There is a broad maximum of dust surface concentration at the northern edge of the Tibetan Plateau, located at the northern slope of the middle range (77–86 °E) of the Kunlun Mountains running from west to east, to the north at the Tarim Basin. The enhanced dust aerosol concentrations from different source areas are clearly visible at 6 km, and their regional impacts can still be seen at 8 km altitude. At 10 km altitude, the enhancement of dust aerosols over the Tibetan Plateau are still remarkable, with a lofted dust plume from the local emission source in the northwest of the Tibetan Plateau. At altitudes above 10 km, up to 16 km, enhanced dust aerosols persist over the Tibetan Plateau, with the maximum shifting eastward and then southward under the influence of anticyclonic circulation. The ~~spatial distribution~~ plume transport pattern ~~of that connects the maximums of~~ mineral dust aerosols over the Tibetan Plateau to the surface indicates that dust aerosols in the UTLS over the Tibetan Plateau are predominantly from emission sources at the northern Tibetan Plateau and its northern slope areas.

The distribution pattern of dust near the surface on the Tibetan Plateau and the nearby areas shown above (e.g., in Fig. 12) is generally similar to that of the tropospheric column concentration (e.g., Fig. 3). However, the position of maximum values for the latter shifts slightly to the north, where larger amounts of dust in the total column may be attributed to the Taklamakan Desert in the Tarim Basin than the central Tibetan Plateau. Satellite data analysis by Xu et al. (2015) has also shown high aerosol optical depth (AOD) values in the same area to the north of the Tibetan Plateau (see Fig. 2 in the paper of Xu et al. (2015)). Note that higher AOD to the south than the north of the Tibetan Plateau shown by Xu et al. (2015) might be attributed to much greater contributions of anthropogenic aerosols from India; moreover, only clear days are considered for satellite data products, which tends to affect the seasonal statistics more for aerosols in the southern Tibetan Plateau, where there are more clouds and precipitation associated with the ASM (Zhao et al., 2018).

Our simulation results support the arguments of previous studies (Huang et al., 2007; Xia et al., 2008; Xu et al., 2015) in that mineral dust aerosols are the dominant aerosol type on the Tibetan Plateau and they are most likely transported to the main body of the Tibetan Plateau across the northern edge. Here we emphasize that compared to the transport from the interior of the Taklamakan Desert in the Tarim Basin, direct emissions of dust aerosols from local sources in the northern Tibetan Plateau, i.e. at the northern slope (around 4-5 km altitude) of the middle Kunlun Mountains, are more likely to contribute to the maximum. The dust layers detected by satellite have been reported to extend up to 8–10 km altitude over the northern Tibetan Plateau in summer (Huang et al., 2007; Liu et al., 2008; Xu et al., 2015). Our simulation shows that the maximum of dust at 10 km altitude is still located just over the strongest source area at the surface, agreeing with satellite measurement results and indicating that dust aerosols are lofted directly to the upper troposphere along the northern slope of the Tibetan Plateau. At 10 km altitude and above, the position of maximum dust aerosols travels along the anticyclonic circulation, mainly within its core area.

~~The mineral dust emission scheme used in the present study was validated by comparing the dust AOD (DAOD) from EMAC simulation of the year 2011 at T106 resolution with various satellite and ground-based measurements (Klingmüller et al., 2018). This advanced scheme, with the updated land cover classification, the inclusion of the topography factor and the modification of the sandblasting efficiency function, has been demonstrated to significantly improve agreement between model simulation and the observations (Klingmüller et al., 2018).~~ Brühl et al. (2018) showed ~~that high sensitivity of~~ mineral dust reaching the UTLS ~~is sensitive~~ to model resolution, owing mostly ~~due~~ to the differences in convection top height and overshooting convection in the ~~convection~~ parametrizations. ~~It is concluded that e~~ Compared to T63L90 (with 1.88° horizontal resolution in longitude and latitude, and 90 vertical layers), which fits best to the observations (including the profile shown in Fig. 4) as for which the convection parameterization was developed (Tiedtke, 1989), EMAC tends to overestimate stratospheric DAOD with simulation at lower resolution (e.g., T42L90), and underestimate it at higher resolution (e.g., T106L90) (Brühl et al., 2018). We find that the aerosol extinction levels in the UTLS within the ASM anticyclone of this study are systematically underestimated with respect to the results of Brühl et al. (2018) with T63L90. For example, the maximum monthly mean  $K_e$  at 16 km over the Tibetan Plateau from the T63L90 simulation is about 3 times larger than that from the T106L90 simulation. ~~However, in addition to~~ Such difference might partly be attributed to the



difference in simulated mineral dust, but also it is most likely ~~other types of aerosols and their gaseous precursors, especially those from south Asia, could be underestimated as well,~~ due to less efficiently convective transport of anthropogenic aerosols and their gaseous precursors in the T106L90 simulations compared to T63L90 simulations. It should be noted that deep convection events occur much less frequently over the northern part of the Tibetan Plateau than the southern part of it, to the latter pollution from South Asia tends to accumulate (Lelieveld et al., 2018). ~~Therefore, using T63L90 rather than T106L90 should not affect our results on the relative contributions of mineral dust to the total  $K_e$  in a qualitative sense. Instead, our~~ Figures S11, S12 and 13 present the geographical distributions of specific humidity,  $q_v$ , at various altitudes from the surface to 18 km over the ASM region from our EMAC simulations, averaged for July–August in the years 2010, 2011 and 2013, respectively. It is shown that deep convection of moisture occurs over the southern edge of the Tibetan Plateau, with the maximum  $q_v$  attained from 10–11 km altitude upward. This is similar to the reanalysis results from previous studies, e.g., Xu et al. (2014), who found the most significantly relative enhanced  $q_v$  at 250–300 hPa over the same area. The geographical position of the mineral dust maximum in the upper troposphere over the Tibetan Plateau is apparently different from that of the moisture maximum, indicating that different pathways for the transport of aerosols and their gaseous precursors from the lower troposphere to the UTLS over the Tibetan Plateau.

Figures S13, S14 and 14 present the geographical distributions of gaseous ammonia ( $\text{NH}_3$ ) at various altitudes from the surface to 18 km over the ASM region from our EMAC simulations, averaged for July–August in the years 2010, 2011 and 2013, respectively. In addition to northern India, which is located to the southwest of the Tibetan Plateau, an enhancement of surface  $\text{NH}_3$  is also found in the central main body of the Tibetan Plateau, although the maximum  $\text{NH}_3$  mixing ratio in the latter area ( $\sim 10$  ppbv) is about half of that in the former area ( $\sim 20$  ppbv). It is shown that surface ammonia can be more efficiently convected and transported to the UTLS from the Tibetan Plateau than from the Indian region, leading to high levels of  $\text{NH}_3$ , e.g., about 0.1 ppbv at 17 km altitude. Such large amounts of lofted ammonia accumulate within the ASM anticyclone, providing the basic gas favourable for the formation of nitrate aerosols (especially in the years 2010 and 2012 when the volcanic eruption effect is relatively small with respect to 2011), as discussed in Sect. 3.3. ~~Our~~ EMAC simulation at a relatively high resolution (i.e., T106L90) ~~resolution~~ reveals clearly the important role of ~~source~~ emissions and the orographic forcing and deep convection transport of in transport of mineral dust and gaseous precursors like  $\text{NH}_3$  transport on-over the Tibetan Plateau ~~in the ATAL formation. While T63L90 has been preferred to the studies recently, such as quantitative assessments of stratospheric aerosol radiative forcing (Brühl et al., 2018).~~ T106L90 with improved convection parameterization is suggested to investigate the transport of aerosols and their gaseous precursors associated with complex topography ~~or and~~ finer structure of the anticyclone.

## 4 Conclusions

We have investigated the chemical components and their sources of aerosols in the UTLS over the Tibetan Plateau during summer, within the ASM anticyclone, using the atmospheric chemistry general circulation model EMAC at T106L90 resolution. ~~A relatively fine model grid resolution and an advanced dust flux scheme in EMAC improve the dust simulation~~

~~over the Tibetan Plateau which has a complex topography. The use of SO<sub>2</sub> mixing ratios from MIPAS satellite measurements enhances the model's ability in simulating volcano eruption plumes in the UTLS.~~ The model simulation performed for the period from January 2010 to December 2012, and for this study the seasonally (July–August) averaged results are analyzed, focusing on the spatial distribution characteristics of aerosols extinction and chemistry at different altitudes around the tropopause. Model evaluation against lidar measurements over the central Tibetan Plateau shows that EMAC can reproduce the basic vertical distribution feature of summertime aerosols in the UTLS over the Tibetan, although simulated aerosol extinction coefficients tend to be at the lower end of the measured range.

Our simulation results show that over the Tibetan Plateau, there is a maximum in aerosol extinction in the upper troposphere for all three investigated years (at 15–16 km in 2010–2012), and even in the lower stratosphere (at 17 km in 2010 and 2012) except for 2011, when the Nabro eruption occurred (in June). It is found that the maximum aerosol extinction over the Tibetan Plateau can be attributed mainly to mineral dust aerosols (with a contribution of 30–60% at 15–16 km), followed by water soluble aerosols and associated liquid water (each making a comparable contribution of 10–40% at 15–16 km), and these three components also dominate aerosol extinction in the UTLS within the ASM anticyclone. In contrast to the absolute  $K_e$  from mineral dust, which is at the same level in the three years, the absolute  $K_e$  contributed by water-soluble species (WASO) and aerosol liquid water (ALW) is much higher in 2011 than in 2010 and 2012, especially in the lower stratosphere (e.g., at 17 km). The enhancement of aerosols around the tropopause within the ASM anticyclone in 2011 is less pronounced, due to shielding by high background sulfate aerosols outside the anticyclone. We even find an “anti-ATAL” phenomenon characterized by a relative minimum of aerosol extinction in the lower stratosphere (e.g., at 17 km in 2011) due to the strong volcano effect. This indicates that the ASM anticyclone not only efficiently traps tropospheric pollutants inside but also blocks the intrusions of stratospheric ozone and aerosols from outside.

It is shown that mineral dust, sulfate and nitrate are three major components of dry aerosols in the UTLS within the ASM anticyclone. The mineral dust and nitrate concentrations decrease from the upper troposphere to the lower stratosphere, and vice versa for sulfate due to greater volcano influence in the lower stratosphere than in the upper troposphere. The levels of NO<sub>3</sub><sup>-</sup>, NH<sub>4</sub><sup>+</sup>, and crustal cations like Ca<sup>2+</sup> are strongly enhanced in the upper troposphere (at 15–16 km altitudes) within the ASM anticyclone, and the enhancements persist in the lower stratosphere during volcanically quiescent times (e.g., at 17 km in 2012). In contrast, sulfate aerosol (SO<sub>4</sub><sup>2-</sup> and HSO<sub>4</sub><sup>-</sup>) concentrations in the lower stratosphere are found to be generally higher outside the anticyclone than inside, especially in 2011, due to volcano eruptions occurring outside the ASM area. We find two maxima of NO<sub>3</sub><sup>-</sup> and NH<sub>4</sub><sup>+</sup> in the UTLS within the ASM anticyclone, one over the east of the Tibetan Plateau, and another over the Iranian Plateau and its surrounding area. A maximum of crustal cations is found to be located over the Tibetan Plateau, coincided with that of mineral dust. Our ionic aerosol composition analysis reveals the existence of Ca(NO<sub>3</sub>)<sub>2</sub> or analogs (from Mg<sup>2+</sup>, for example) in the upper troposphere over the Tibetan Plateau due to multiphase reactions on mineral dust, and substantial amounts of NH<sub>4</sub>NO<sub>3</sub> within the ASM anticyclone. In the stratosphere these cations are most likely to exist in the form of CaSO<sub>4</sub> (or analogs) and (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> (partly as NH<sub>4</sub>HSO<sub>4</sub>) since NO<sub>3</sub><sup>-</sup> is expelled from the aerosol

under acidic conditions, especially when the impact from volcano eruptions is large. We find considerable influence of stratospheric aerosols in the anticyclone from its surroundings, mainly through the northern and eastern edges.

We find that dust aerosols in the UTLS within the ASM anticyclone are predominantly from the emission sources in the northern Tibetan Plateau and its northern slope areas. Compared to the interior of the Taklamakan Desert in the Tarim Basin (located to the north of the Tibetan Plateau), the areas at the northern slope of the middle Kunlun Mountains (located in the northern Tibetan Plateau) are more likely to be a predominant source of dust aerosols over the Tibetan Plateau. Our simulations indicate that dust aerosols accumulated in these source areas can be lofted directly to the upper troposphere along the northern slope of the Tibetan Plateau, up to an altitude of at least 10 km. At 10 km altitude and above, lofted dust aerosols are transported eastward and then southward under the influence of anticyclonic circulation and most of them entrain into the core area of anticyclone. [We also find that the emissions and convective transport of ammonia in the central main body of the Tibetan Plateau contribute significantly to the enhancement of gaseous NH<sub>3</sub> in the UTLS over the Tibetan Plateau and entire ASM anticyclone area.](#)

Compared with previous model studies, the EMAC model version we used for this study appears to be the most complete in the treatment of aerosol sources, microphysics and chemistry. [We find a larger dust contribution to the ATAL after updating the dust emission scheme. Our results are also consistent with the recently reported observation of enhanced nitrate \(Höpfner et al., 2019\), which appears to be missing in previous studies. Both the ATAL and Anti-ATAL characteristics are well simulated by our model in general.](#) Nevertheless, further model improvements are desirable, e.g., of convection processes, which at T106L90 might underestimate the transport of pollution to the UTLS from the boundary layer to some extent. While recent field campaigns have been carried out to measure aerosols and trace gases in the UTLS in the southern and/or western parts of the ASM anticyclone (Lelieveld et al., 2018; Vernier et al., 2018), intensive measurements over the Tibetan Plateau are still sparse and strongly recommended (Zhao et al., 2018). This study highlights the important role of the northern Tibetan Plateau in the emissions and transport of dust aerosols to the anticyclone. We argue that the UTLS over the Tibetan Plateau acts as a well-defined conduit, not only for pollutants but also for natural aerosols and gases. Interactions of these natural and anthropogenic aerosols and gases in the UTLS are intricately connected and need further investigations.

## **Code and data availability**

[The Modular Earth Submodel System \(MESSy\) is continuously further developed and applied by a consortium of institutions. The usage of MESSy and access to the source code is licensed to all affiliates of institutions, which are members of the MESSy Consortium. Institutions can become a member of the MESSy Consortium by signing the MESSy Memorandum of Understanding. More information can be found on the MESSy Consortium website \(<http://www.messy-interface.org>, last access: 16 July 2019\). The ECHAM climate model is available to the scientific community under the MPI-M Software License Agreement \(<https://www.mpimet.mpg.de/en/science/models/license>, last access: 16 July 2019\). The simulations results analyzed here are archived at CAMS, Beijing, and available on request.](#)

## Author contributions

JM, XX, XZ, KA, and JL designed the study. AP, CB, VK, KK, and HT provided the EMAC and submodel code. JM performed the model simulations with support from BS, CB and AP. QH provided the aerosol vertical profile data from lidar measurements. BC, YJ, NL, and PY gave assistance in dust source identification and meteorological data analysis. JM prepared the manuscript with contributions from all co-authors.

## Competing interests

The authors declare that they have no conflict of interest.

## Special issue statement

This article is part of the special issue “Study of ozone, aerosols and radiation over the Tibetan Plateau (SOAR-TP) (ACP/AMT inter-journal SI)”. It is not associated with a conference.

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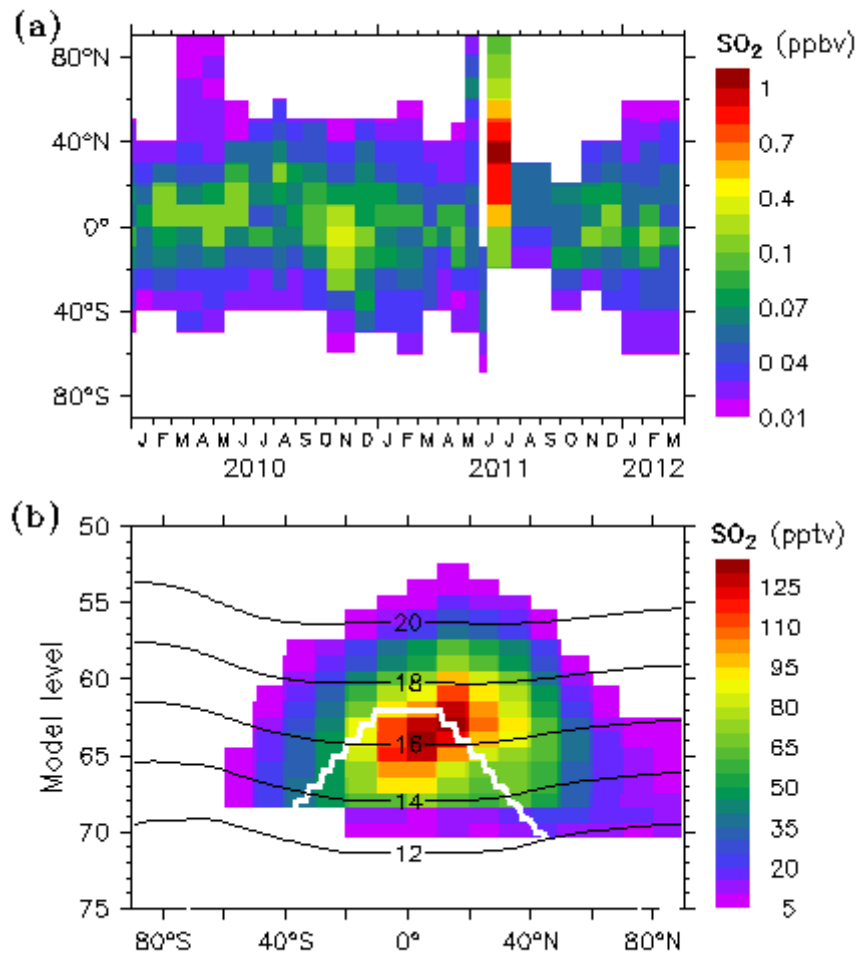
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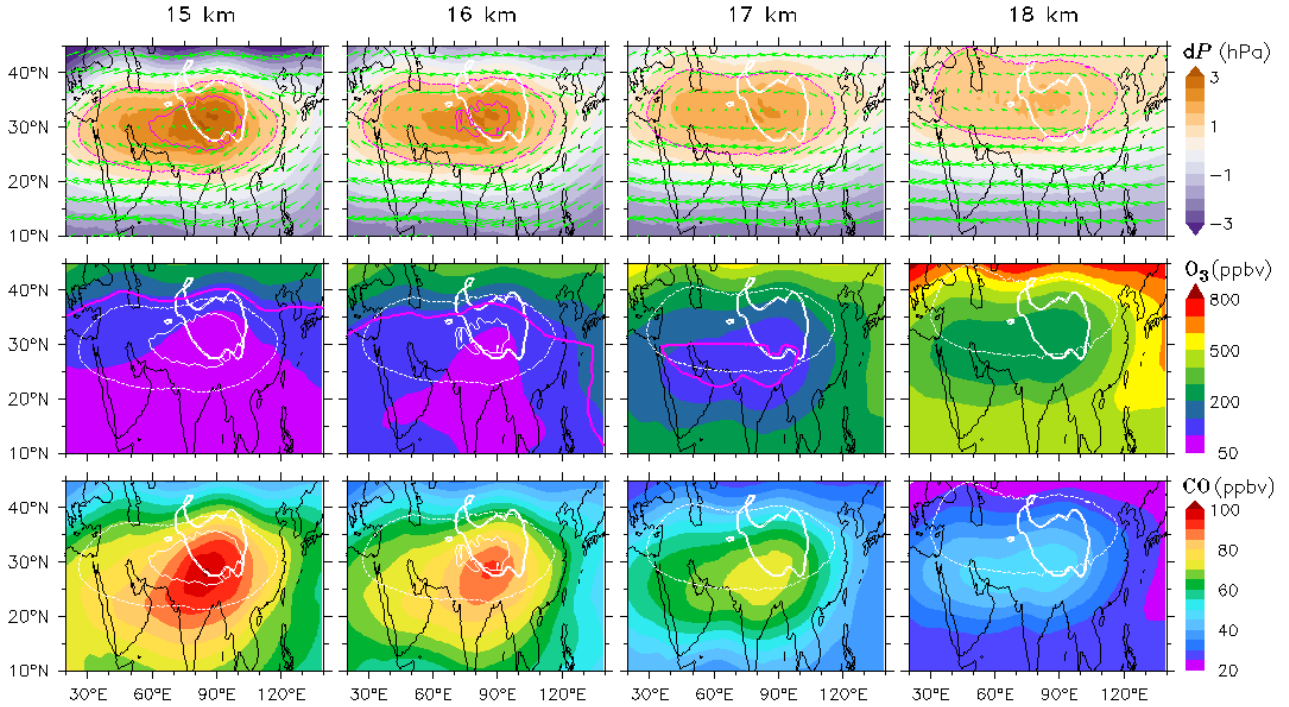


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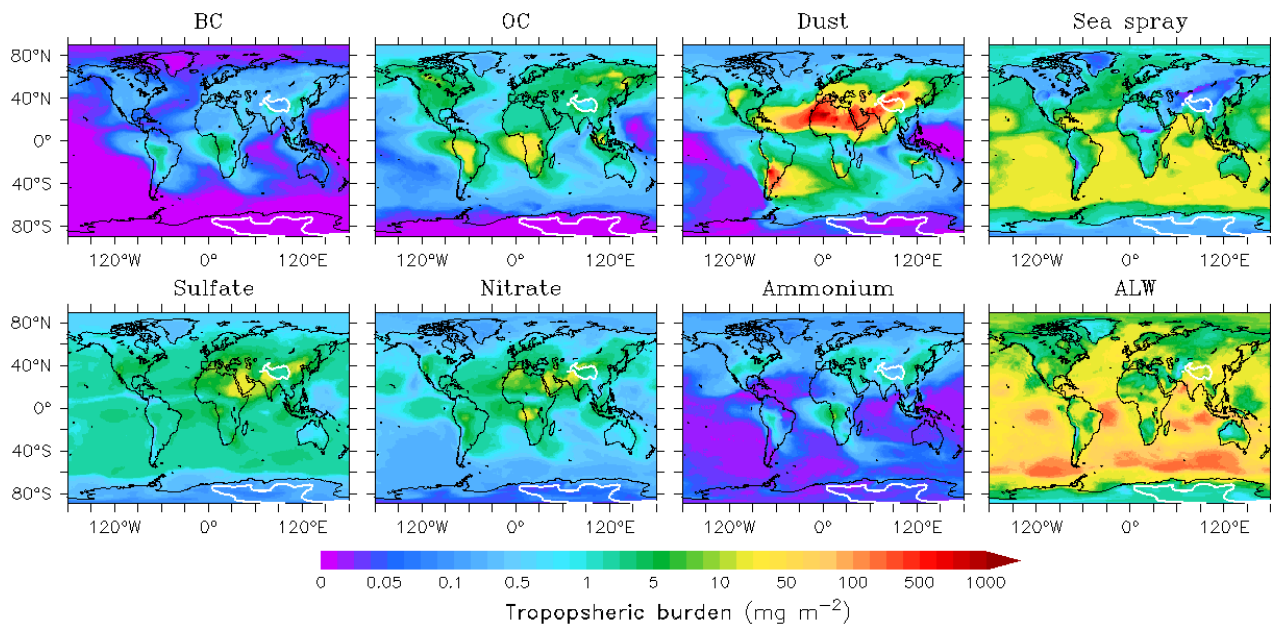
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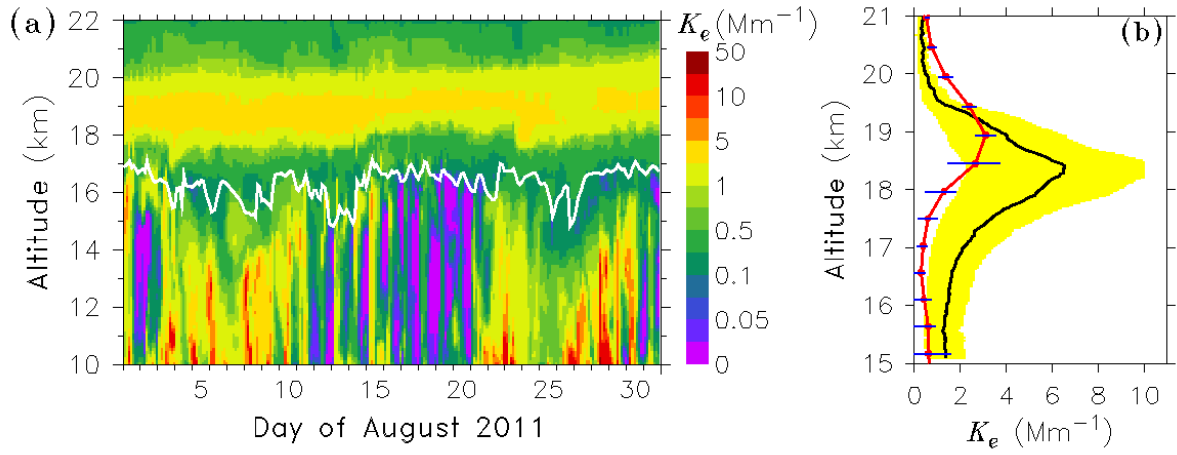
**Figure 1.** MIPAS observed SO<sub>2</sub> in the volcano eruption plumes during January 2010 - March 2012, projected onto the EMAC model grid and averaged in the zonal direction for the plot. **(a)** Latitudinal distribution of model level 60–68 (approximately 14–18 km [a.s.l.](#)) averaged SO<sub>2</sub> (in [nmol mol<sup>-1</sup>](#) or ppbv), as a function of volcano event time. The grid width for each event represents the time interval between the previous and next event. **(b)** Latitudinal and vertical distribution of SO<sub>2</sub> (in [pmol mol<sup>-1</sup>](#) or pptv), averaged for all volcano events. Black lines are altitude contours (in km [a.s.l.](#)), and the white line highlights the WMO tropopause. The blank refers to the areas where the volcano effects on SO<sub>2</sub> were not detected by MIPAS.



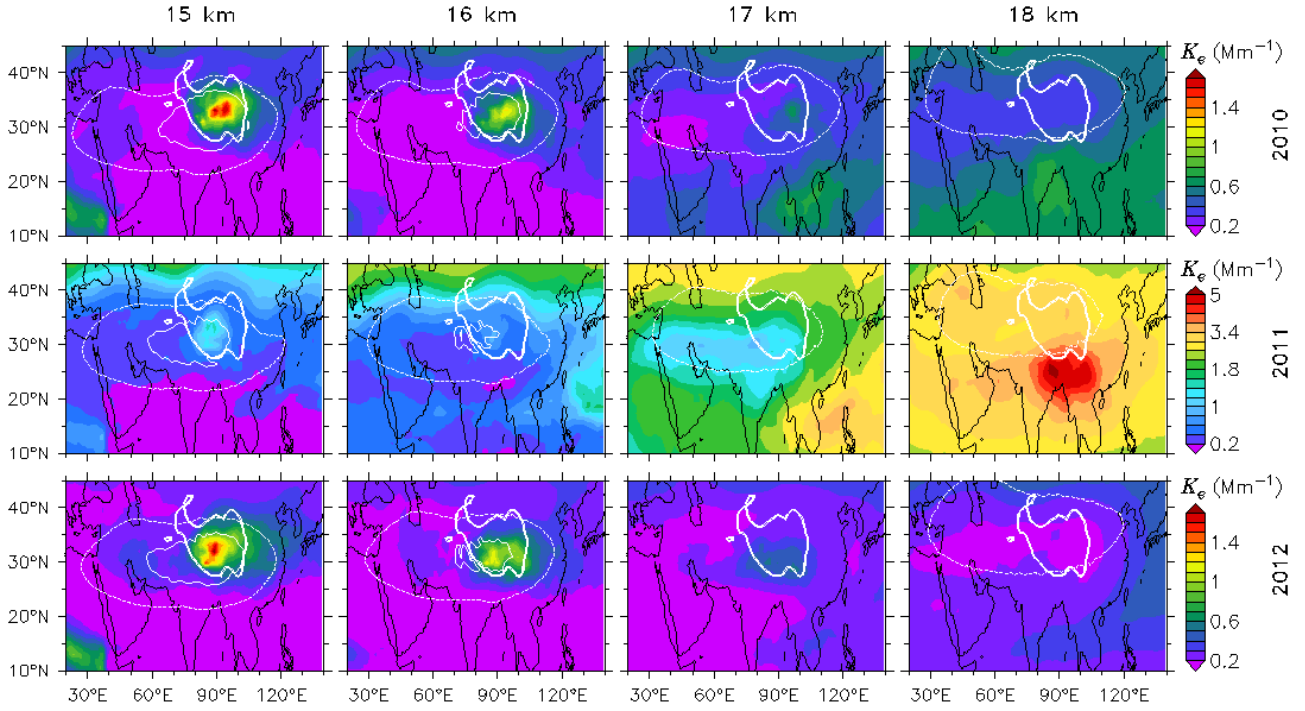
**Figure 2.** Pressure deviation,  $dP$ , in units of hPa (**top**),  $O_3$  in units of  $\text{nmol mol}^{-1}$  or ppbv (**middle**) and CO in units of  $\text{nmol mol}^{-1}$  or ppbv (**bottom**) at altitudes of 15, 16, 17 and 18 km [a.s.l.](#) (**first to forth column**), averaged for July–August of 2010, 2011 and 2012, from EMAC simulations. Pressure deviation, i.e.  $dP = P - P_{avg}$ , refers to the difference of pressure,  $P$ , at each model grid relative to the regionally averaged value,  $P_{avg}$ , at the same altitude, with  $P_{avg}$  values of 132.1, 111.4, 93.6 and 78.7 hPa at 15, 16, 17 and 18 km altitudes, respectively. Green arrows indicate the wind field (**top**), and purple solid lines the position of the WMO tropopause (**middle**). Purple (**top**) and white (**middle** and **bottom**) thin dashed lines (and also solid lines) are the pressure deviation contour of 1 hPa (and 1.3 hPa), highlighting the anticyclone area (and its core area, applicable to the 15 km panels). Thick white lines refer to the terrain height contour of 3 km, highlighting the Tibetan Plateau area.



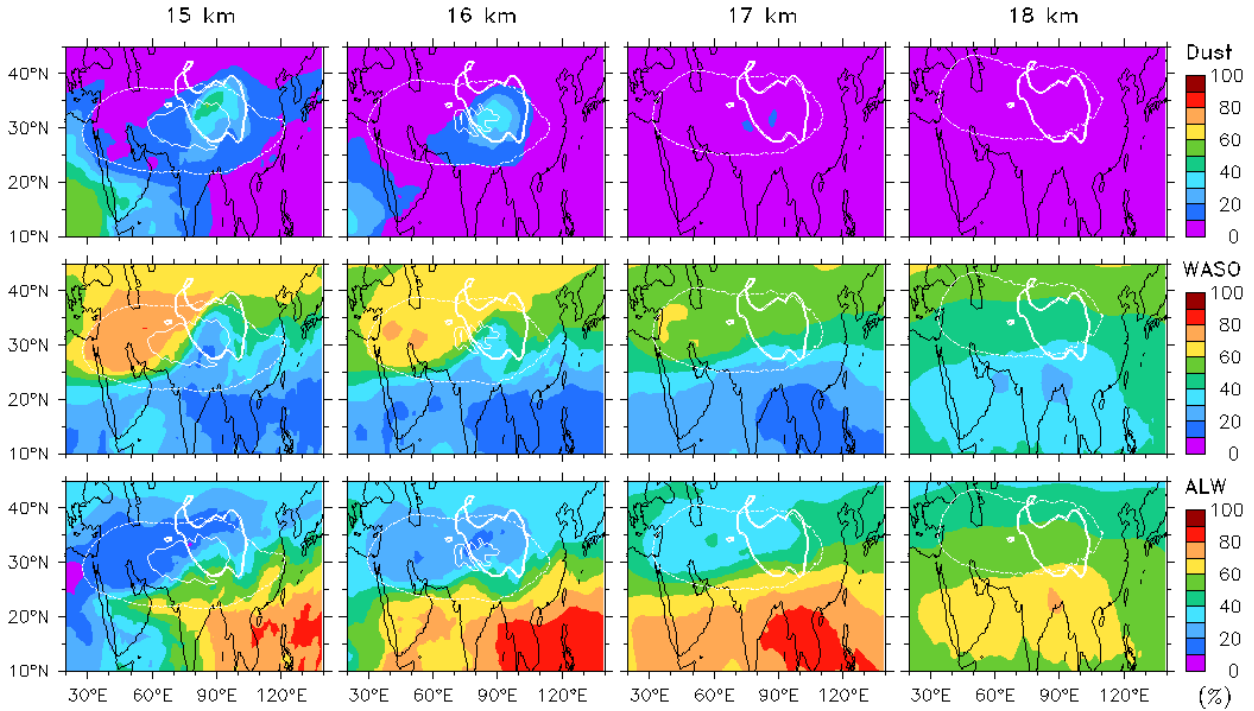
**Figure 3.** EMAC simulated global distributions of the tropospheric burden (in units of  $\text{mg m}^{-2}$ ) ~~of the column concentrations~~ of black carbon (BC), organic compounds (OC), mineral dust, sea spray, sulfate, nitrate, ammonium and aerosol liquid water (ALW), averaged for July–August [2011](#)[2012](#). White lines refer to the terrain height contour of 3 km, highlighting the Tibetan Plateau area in the northern hemisphere.



**Figure 4.** (a) Time series of the vertical profile of aerosol extinction coefficient [at 550 nm wavelength](#),  $K_e$ , in unit of inverse of megameter ( $\text{Mm}^{-1}$  [or  \$10^{-6} \text{ m}^{-1}\$](#) ) over Naqu at three-hour intervals in August 2011, as simulated by EMAC. Thick white line refers to the WMO tropopause altitude in unit of km [a.s.l.](#) (b) Comparison of the model simulated vertical distribution of  $K_e$  in the UTLS region with lidar measurements at Naqu. Red and blue lines refer to the monthly mean values and standard deviations derived from the model. Black line and yellow shaded area are the mean values and standard deviation ranges of measurement data.

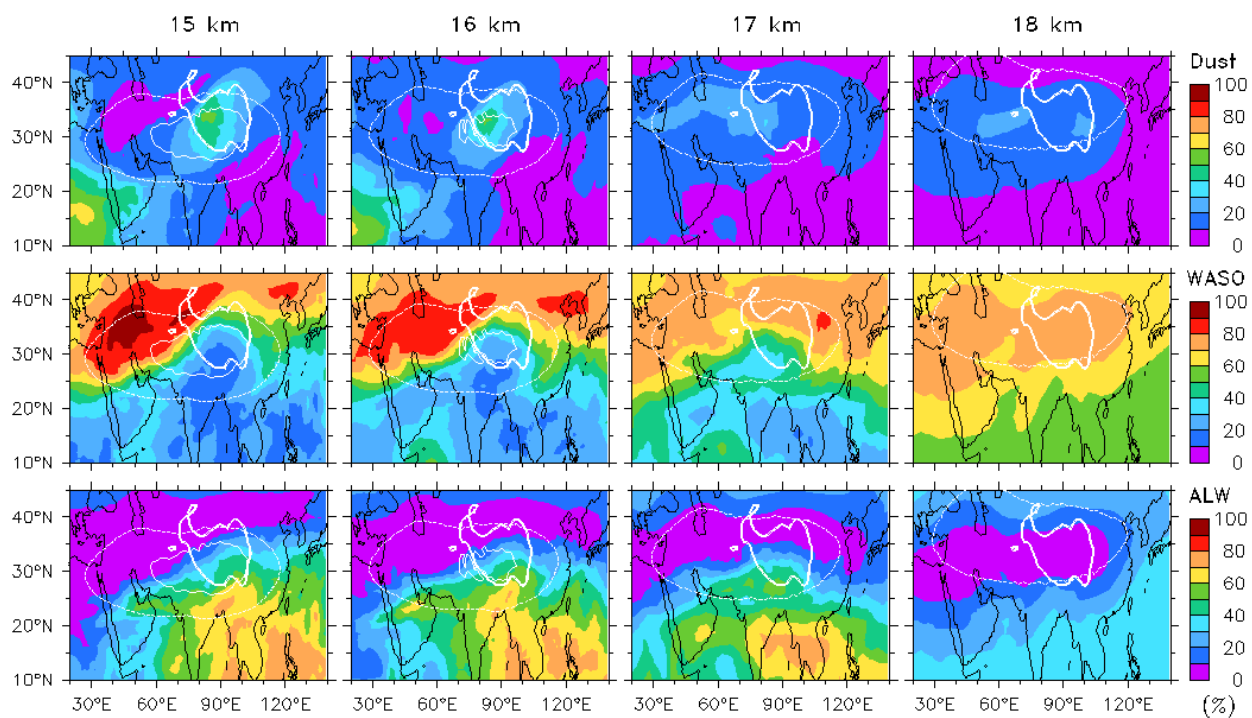


**Figure 5.** EMAC simulated aerosol extinction coefficients at 550 nm wavelength,  $K_e$ , in units of  $\text{Mm}^{-1}$  (i.e.,  $10^{-6} \text{ m}^{-1}$ ) at altitudes of 15, 16, 17 and 18 km a.s.l. (first to forth column), averaged for July–August of 2010 (top), 2011 (middle) and 2012 (bottom). Thin white lines indicate the anticyclone area, with the same index as used in Fig. 2. Thick white lines refer to the terrain height contour of 3 km, highlighting the Tibetan Plateau area. Note that a different colour bar scale is used for  $K_e$  in 2011 from that in 2010 and 2012.

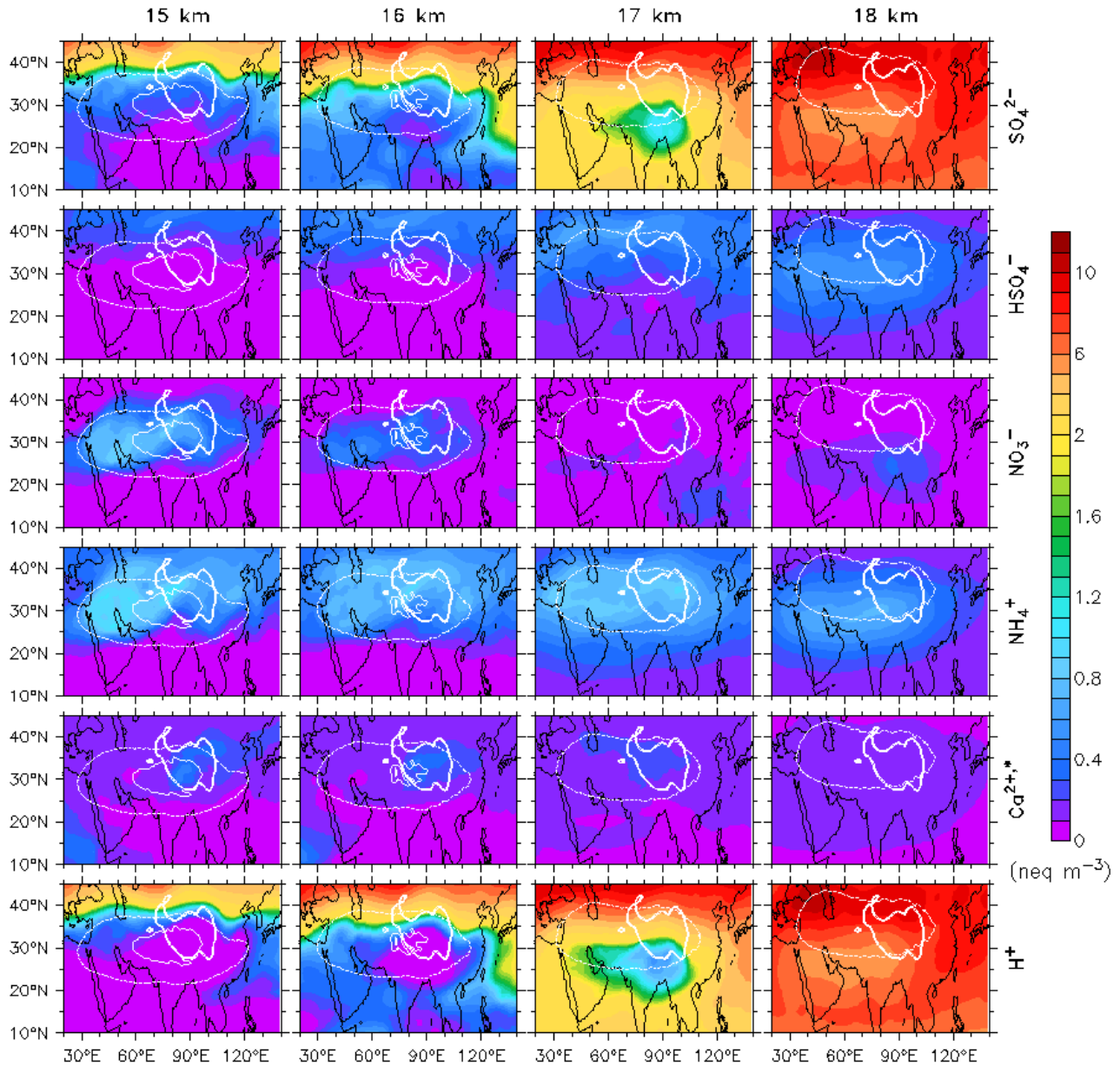


**Figure 6.** Percent contributions of mineral dust (**top**), water-soluble species (WASO) (**middle**) and aerosol liquid water (ALW) (**bottom**) to the total aerosol extinction coefficient at altitudes of 15, 16, 17 and 18 km a.s.l. (**first to fourth column**), during July–August 2011. Thin white lines indicate the anticyclone area, with the same index as used in Fig. 2, and thick white lines highlight the Tibetan Plateau area.

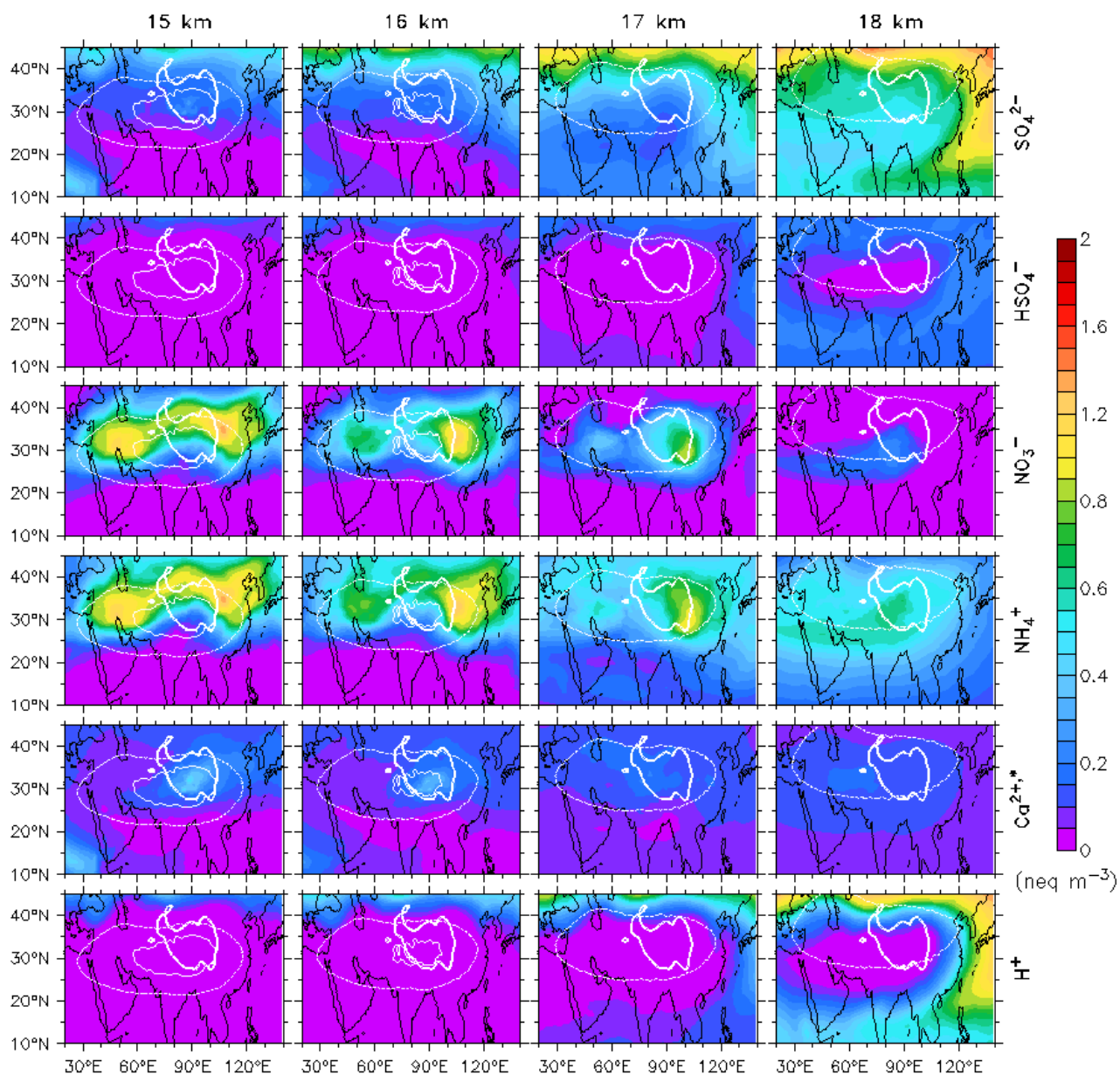




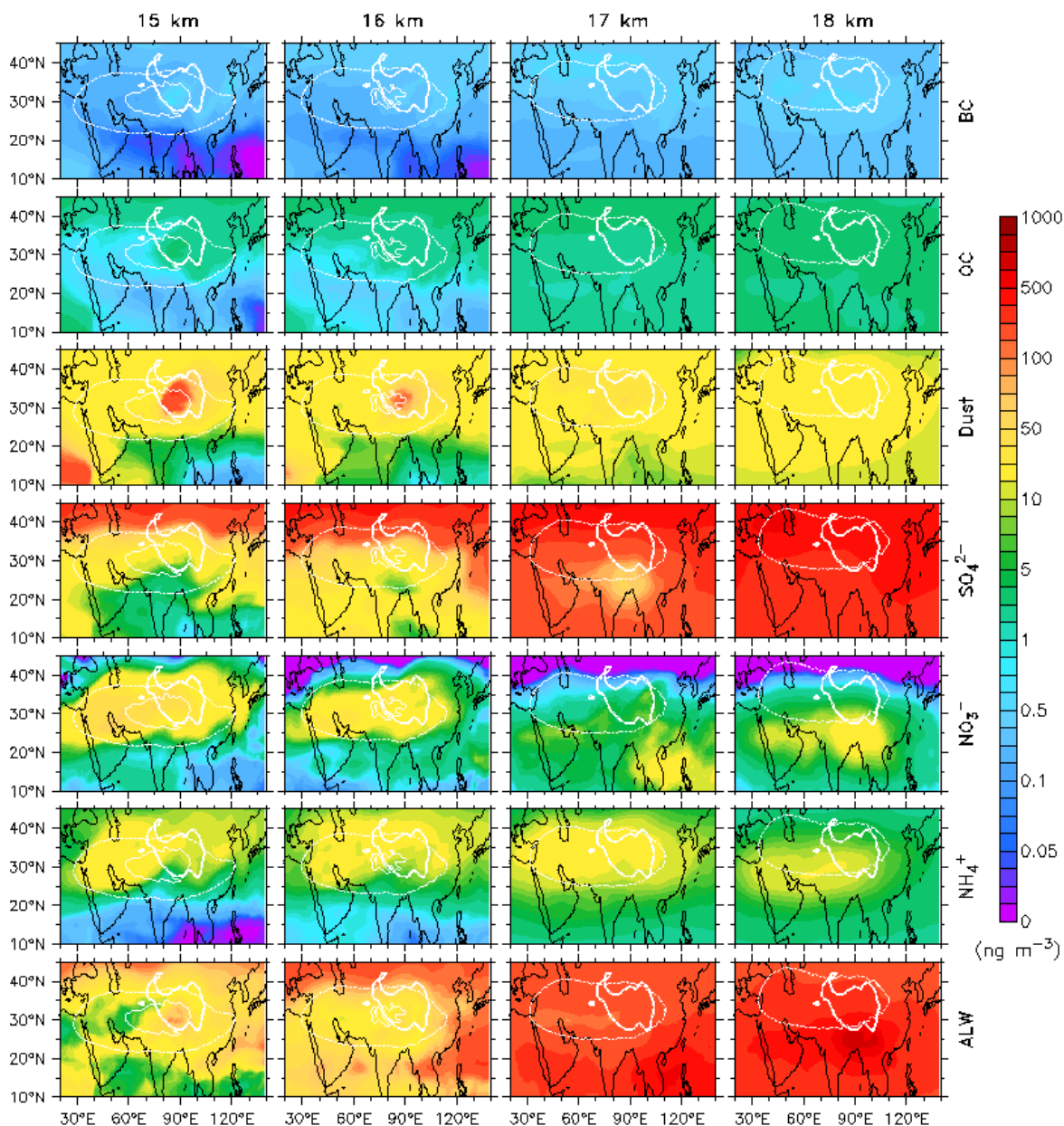
**Figure 7.** Same as Fig. 6, but for the year 2012.



**Figure 8.** EMAC simulated water soluble ionic aerosols in the accumulation mode, including specifically  $\text{SO}_4^{2-}$ ,  $\text{HSO}_4^-$ ,  $\text{NO}_3^-$ ,  $\text{NH}_4^+$ ,  $\text{Ca}^{2+,*}$ , and  $\text{H}^+$  (first to sixth row) in units of nano-equivalent per cubic meter of air ( $\text{neq m}^{-3}$ ), at altitudes of 15, 16, 17 and 18 km a.s.l. (first to fourth column), averaged for July–August 2011. Note that besides  $\text{Ca}^{2+}$ ,  $\text{Na}^+$ ,  $\text{K}^+$  and  $\text{Mg}^{2+}$  are also accounted for in  $\text{Ca}^{2+,*}$ . Thin white lines indicate the anticyclone area, with the same index as used in Fig. 2, and thick white lines highlight the Tibetan Plateau area.

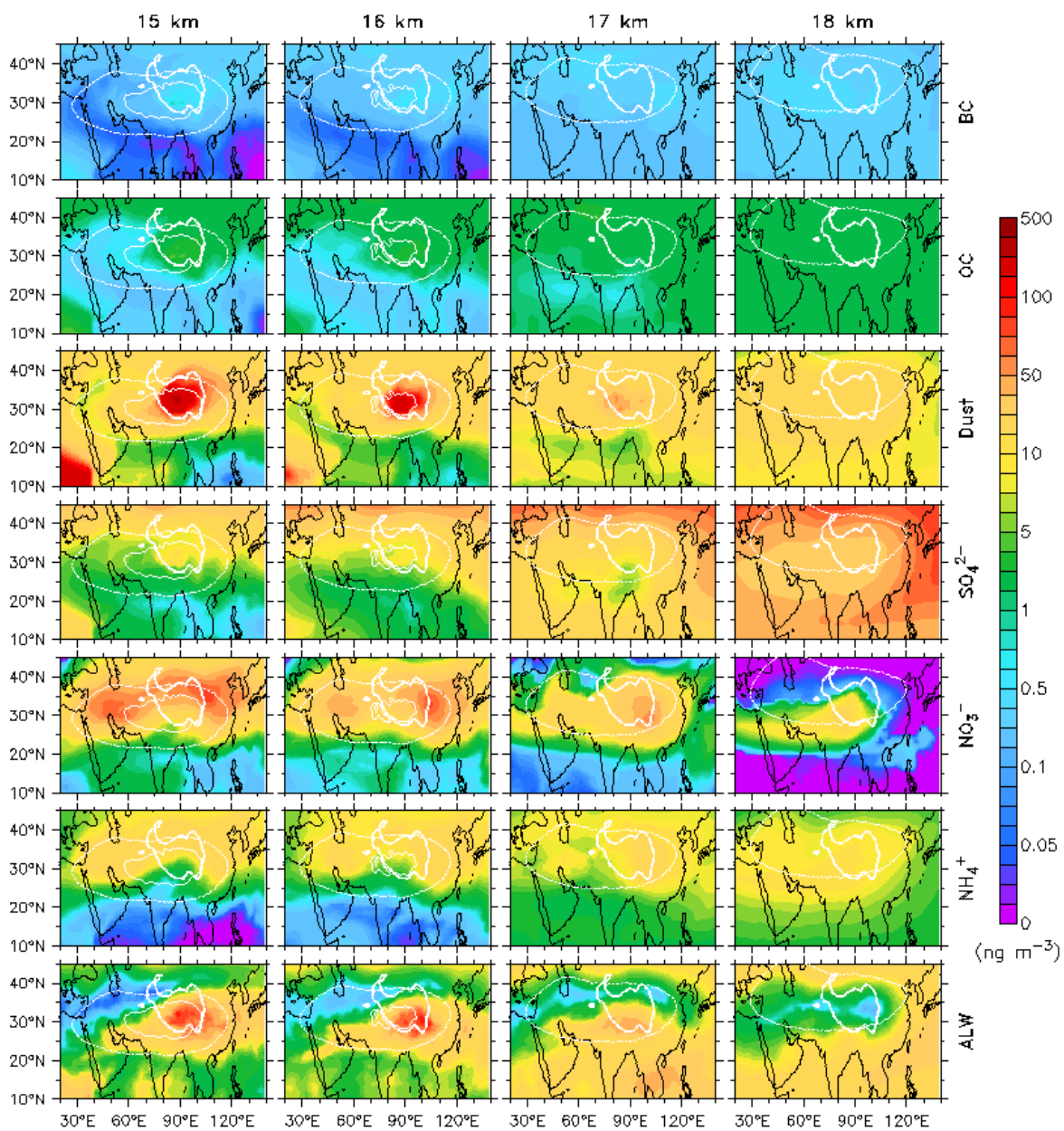


**Figure 9.** Same as Fig. 8, but for the year 2012 with a different colour scale.

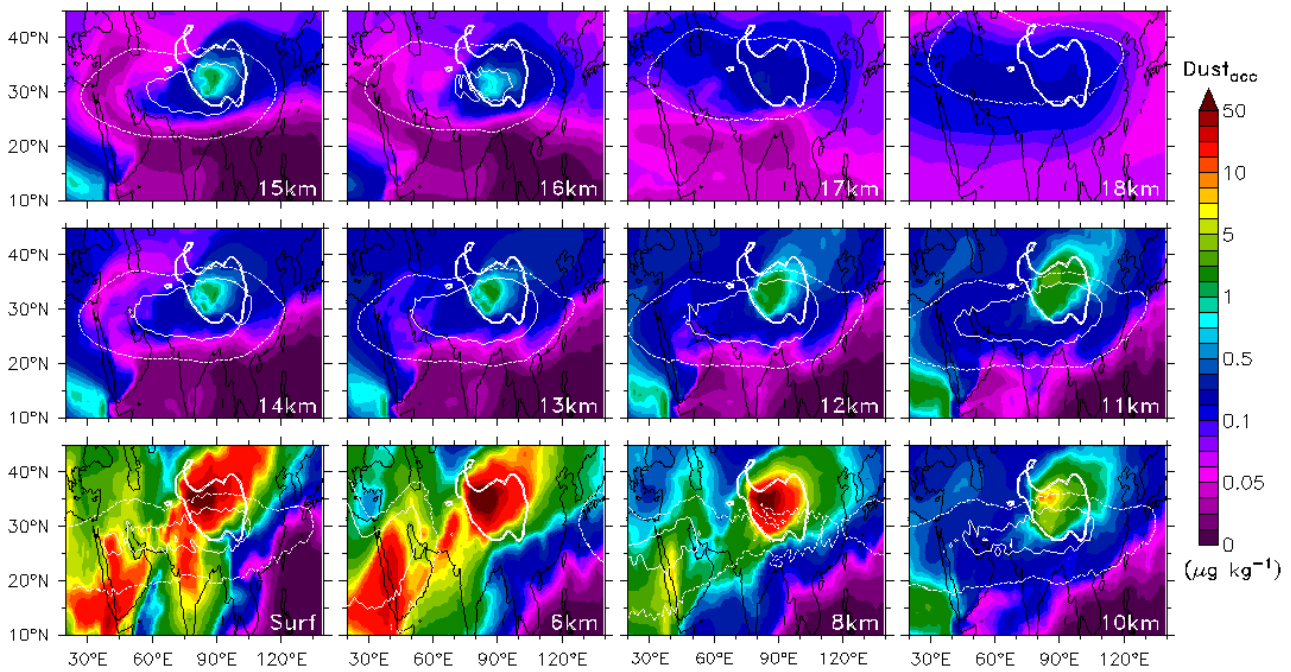


**Figure 10.** EMAC simulated mass concentrations of aerosols in the accumulation mode, including BC, OC, mineral dust, sulfate ( $\text{SO}_4^{2-}$  plus  $\text{HSO}_4^-$ , denoted as  $\text{SO}_4^{2-}$ ), nitrate ( $\text{NO}_3^-$ ), ammonium ( $\text{NH}_4^+$ ), and ALW (first to seventh row) in units of nanogram per cubic meter of air ( $\text{ng m}^{-3}$ ), at altitudes of 15, 16, 17 and 18 km [a.s.l.](#) (first to forth column), averaged for July–August 2011. Thin white lines indicate the anticyclone area, with the same index as used in Fig. 2, and thick white lines highlight the Tibetan Plateau area.

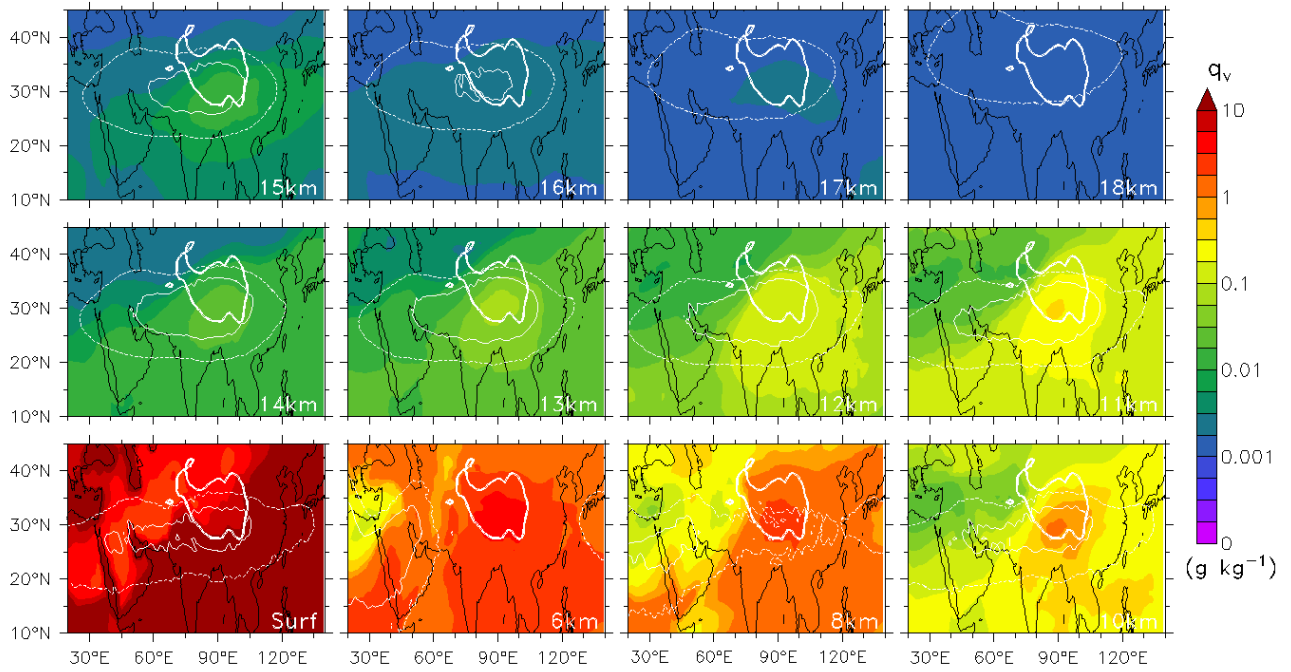




**Figure 11.** Same as Fig. 10, but for the year 2012 with a different colour scale.

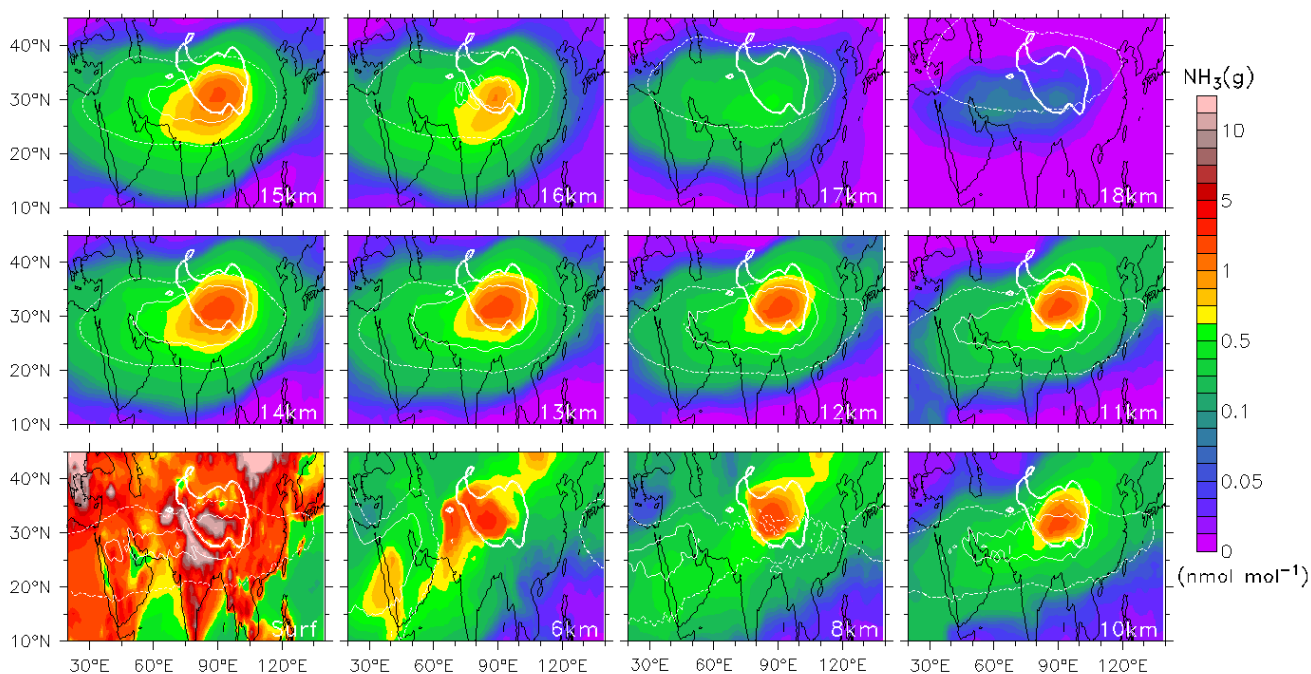


**Figure 12.** EMAC simulated dust mass concentrations in the accumulation mode,  $Dust_{acc}$ , in units of microgram of dust per kilogram of air ( $\mu g\ kg^{-1}$ ) (note that a molecular weight of 40.08 is used for dust), at selected altitudes, i.e. at the surface, 6 km, 8km, 10 km, 11 km, 12 km, 13 km, 14 km, 15km, 16 km, 17 km and 18 km [a.s.l.](#), averaged for July–August [2011](#)–[2012](#). Thin white lines indicate refers to the contour of pressure deviation, as defined in Fig. 2, and thick white lines highlight the Tibetan Plateau area.



**Figure 13.** EMAC simulated specific humidity,  $q_v$ , in units of gram of water vapor per kilogram of air ( $\text{g kg}^{-1}$ ), at selected altitudes, i.e. at the surface, 6 km, 8km, 10 km, 11 km, 12 km, 13 km, 14 km, 15km, 16 km, 17 km and 18 km a.s.l., averaged for July–August 2012. Thin white lines indicate refers to the contour of pressure deviation, as defined in Fig. 2, and thick white lines highlight the Tibetan Plateau area.





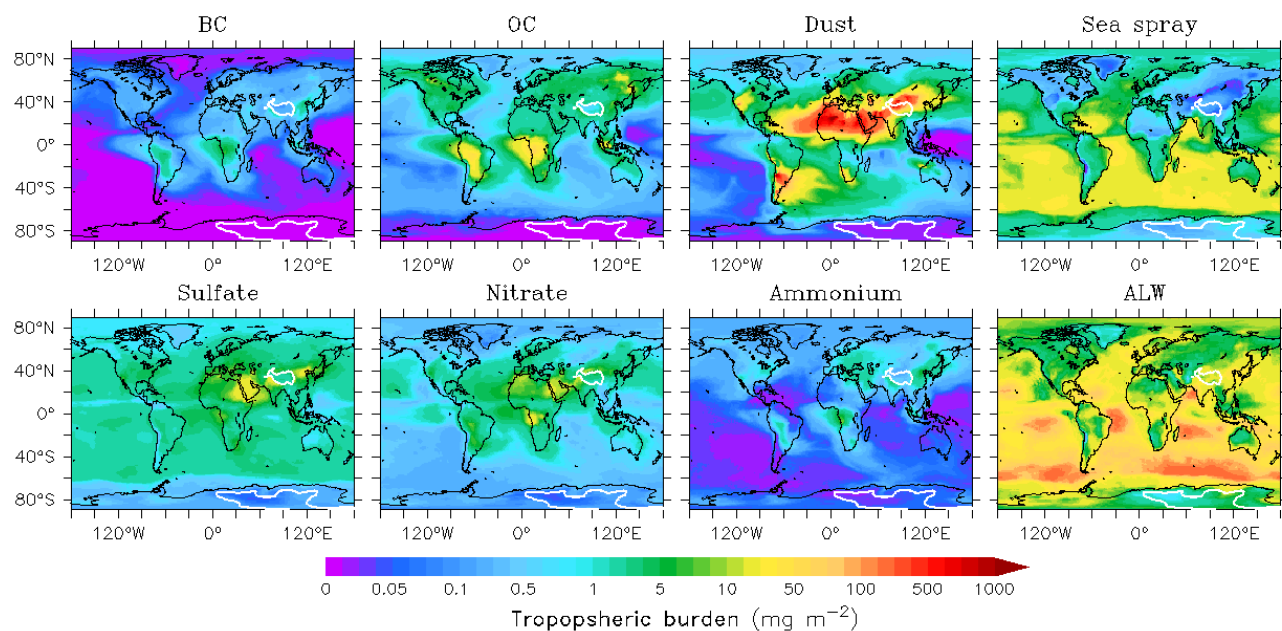
**Figure 14.** EMAC simulated ammonia in the gasphase,  $\text{NH}_3(\text{g})$ , in units of nanomole of ammonia per mol of air ( $\text{nmol mol}^{-1}$ ) or parts of ammonia per billion of air in volume (ppbv), at selected altitudes, i.e. at the surface, 6 km, 8 km, 10 km, 11 km, 12 km, 13 km, 14 km, 15 km, 16 km, 17 km and 18 km a.s.l., averaged for July–August 2012. Thin white lines indicate refers to the contour of pressure deviation, as defined in Fig. 2, and thick white lines highlight the Tibetan Plateau area.

*Supplement of*

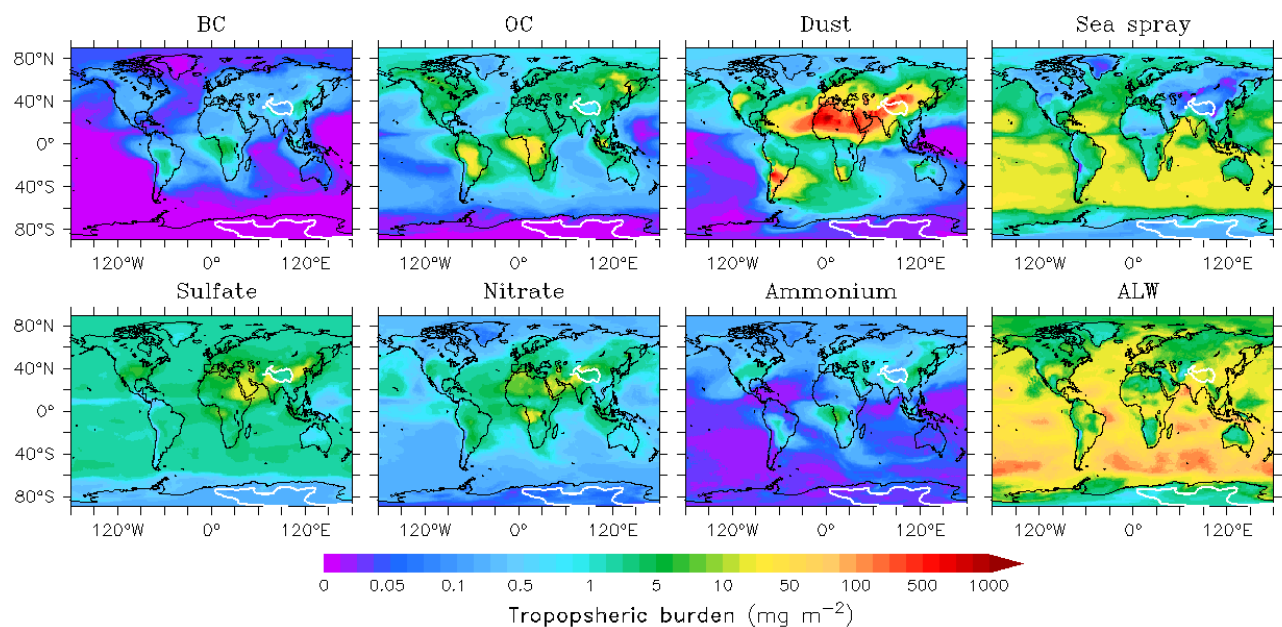
**Modelling the aerosol chemical composition of the tropopause over the Tibetan Plateau during the Asian summer monsoon**

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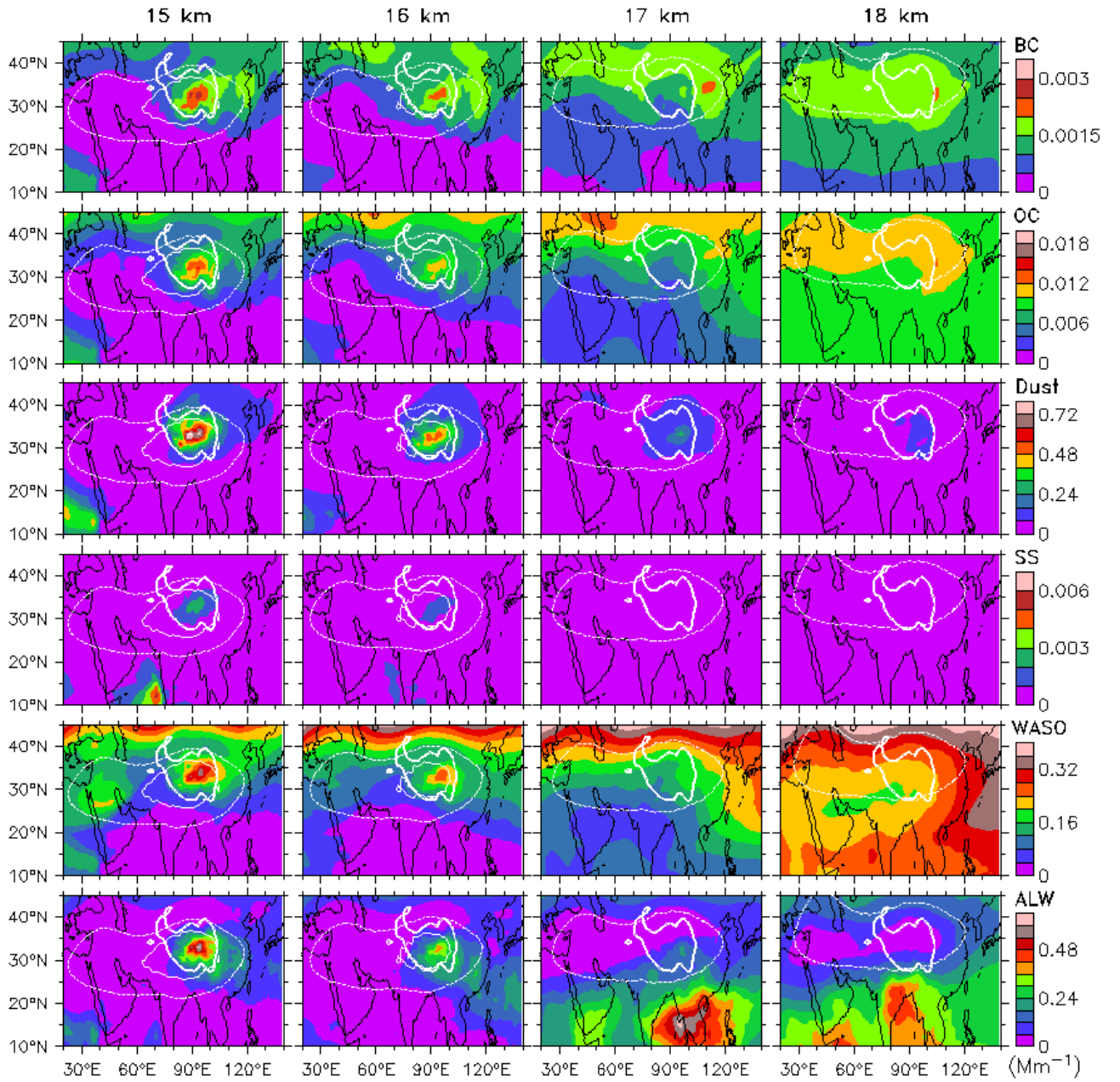
*Correspondence to:* Jianzhong Ma (majz@cma.gov.cn)



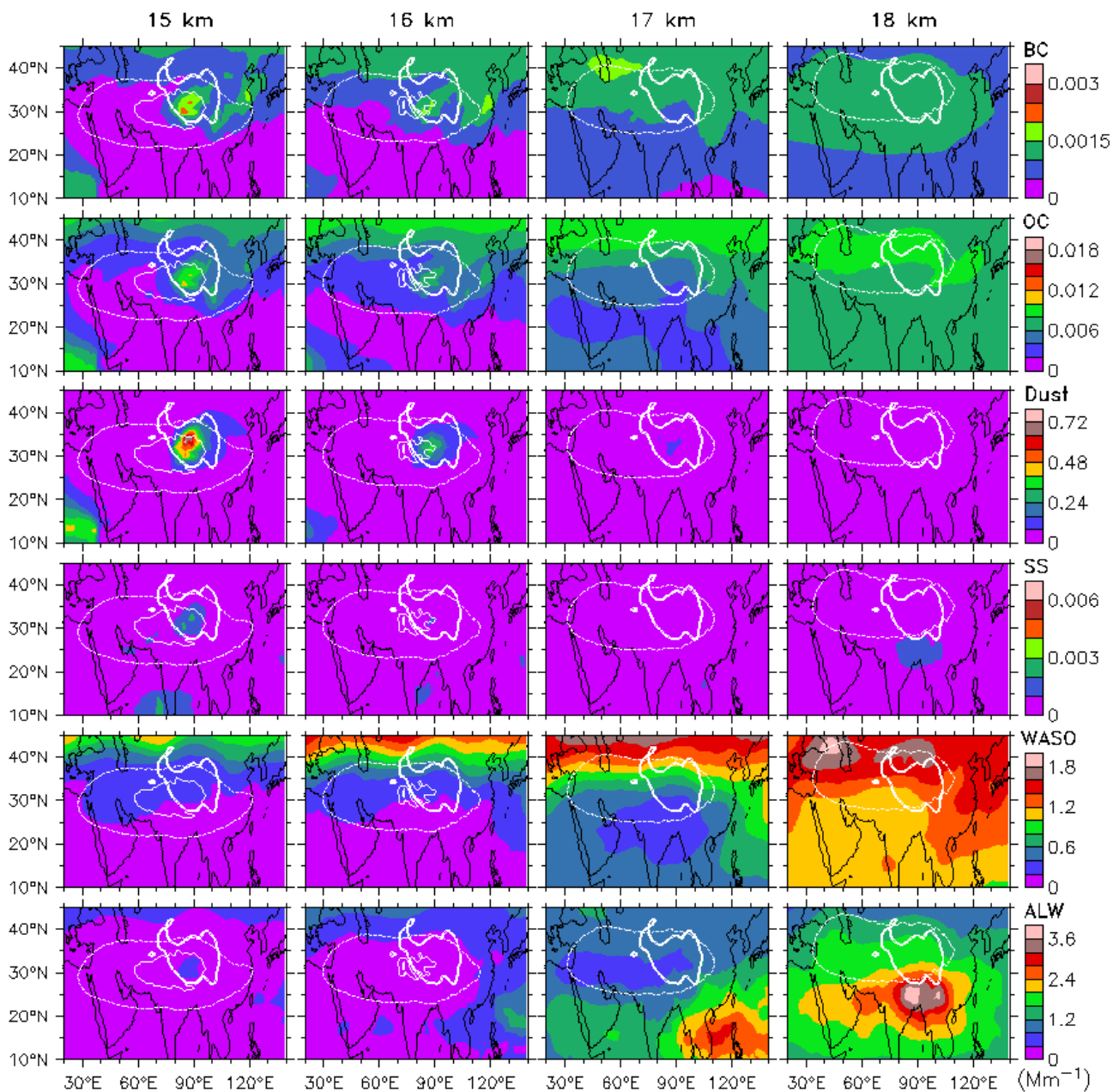
**Figure S1.** Same as Fig. 3, but for the year 2010.



5 **Figure S2.** Same as Fig. 3, but for the year [2011](#).

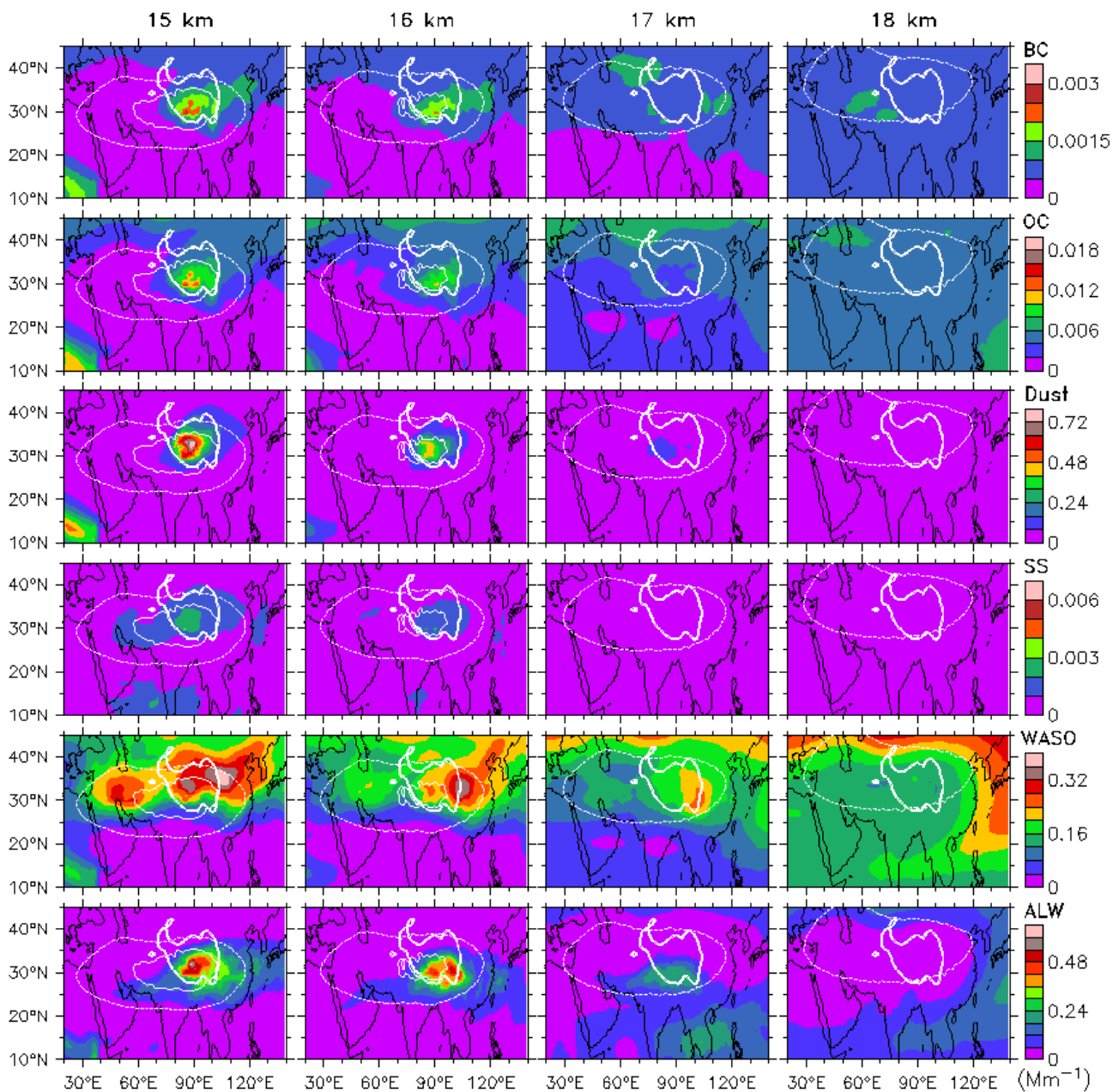


**Figure S3.** EMAC simulated aerosol extinction coefficients at 550 nm wavelength,  $K_e$ , in units of  $\text{Mm}^{-1}$  (1  $\text{Mm} = 10^6$  m), attributed to BC, OC, dust, sea spray (SS), WASO (i.e.,  $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$  and  $\text{NH}_4^+$  ions) and ALW (**first to sixth row**), at altitudes of 15, 16, 17 and 18 km a.s.l. (**first to fourth column**), averaged for July–August 2010. Thin white lines indicate the anticyclone area, with the same index as used in Fig. 2, and thick white lines highlight the Tibetan Plateau area.

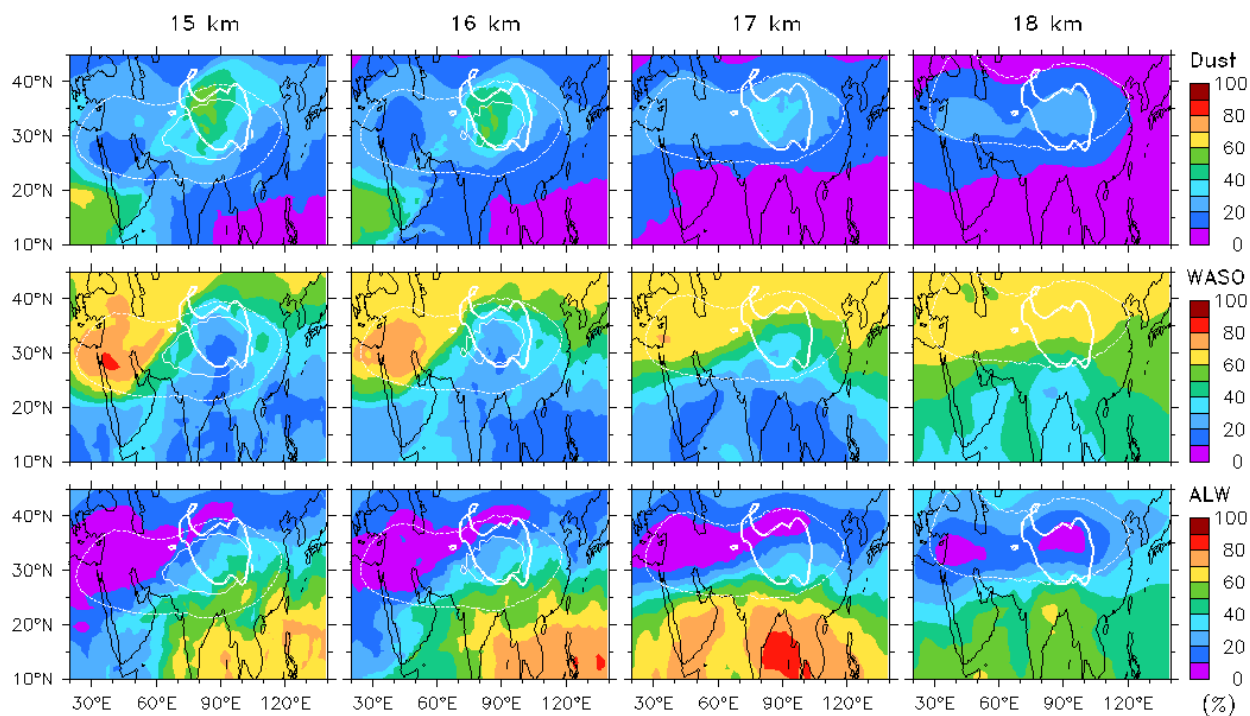


**Figure S4.** Same as Fig. S3, but for the year 2011.



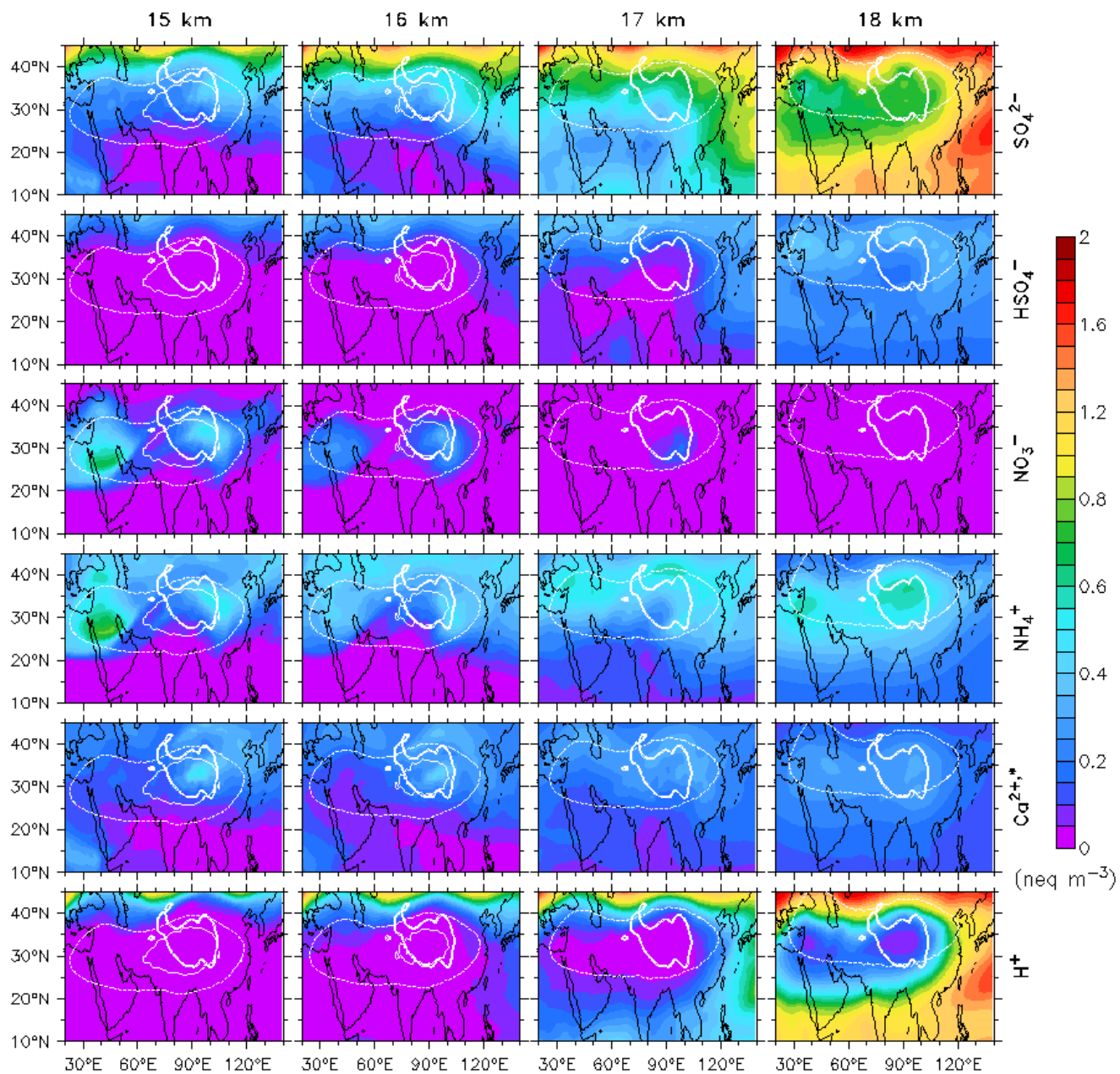


**Figure S5.** Same as Fig. S3, but for the year 2012.

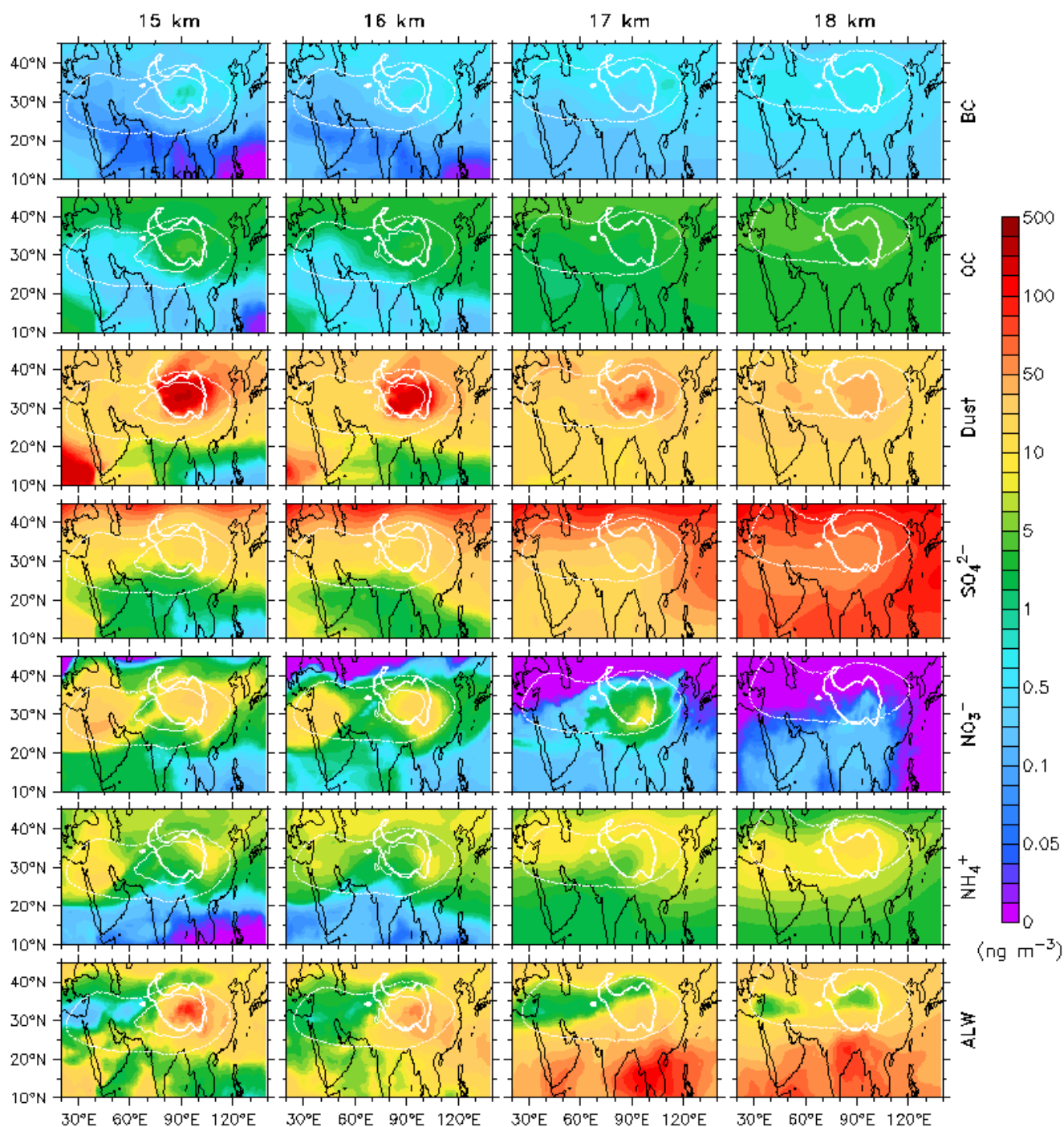


**Figure S6.** Same as Fig. 6, but for the year 2010.

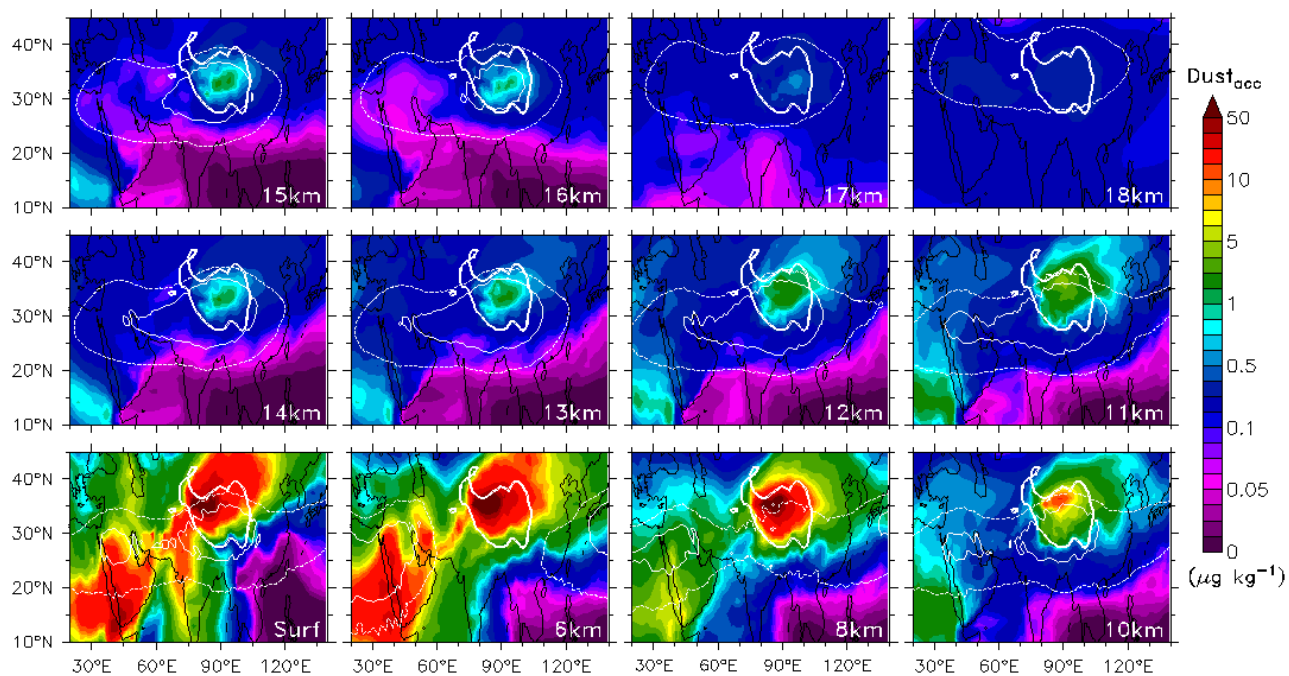




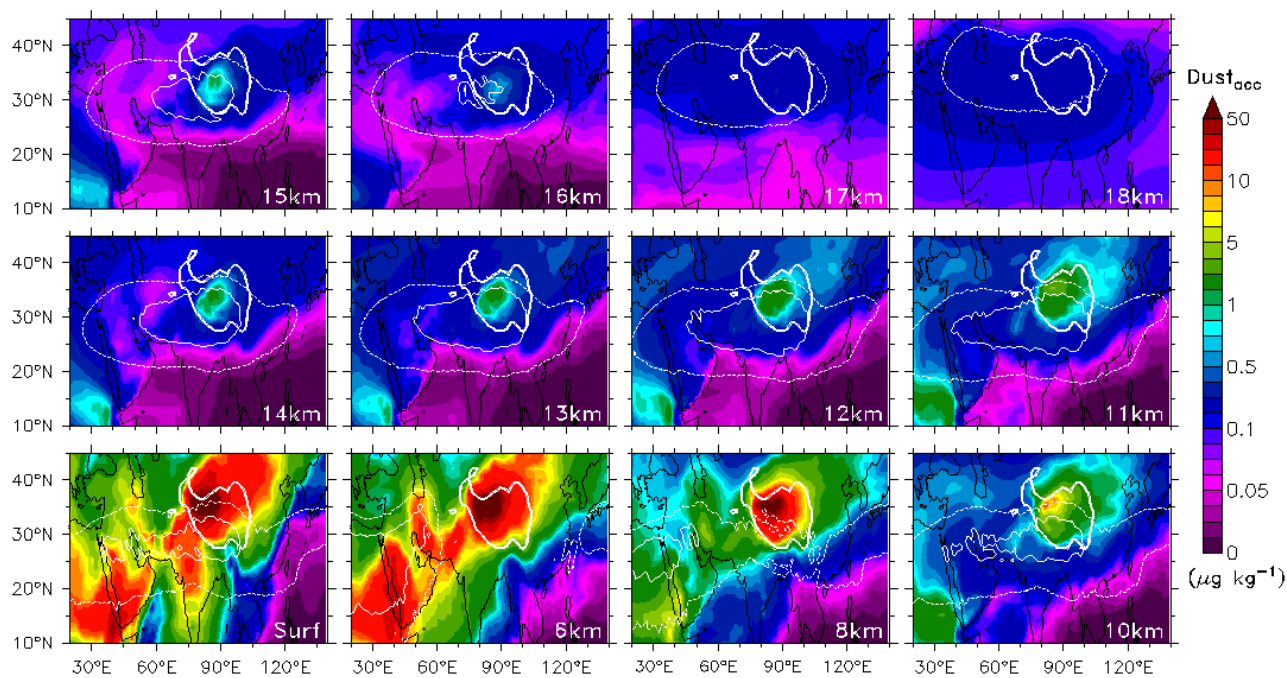
**Figure S7.** Same as Fig. 8, but for the year 2010.



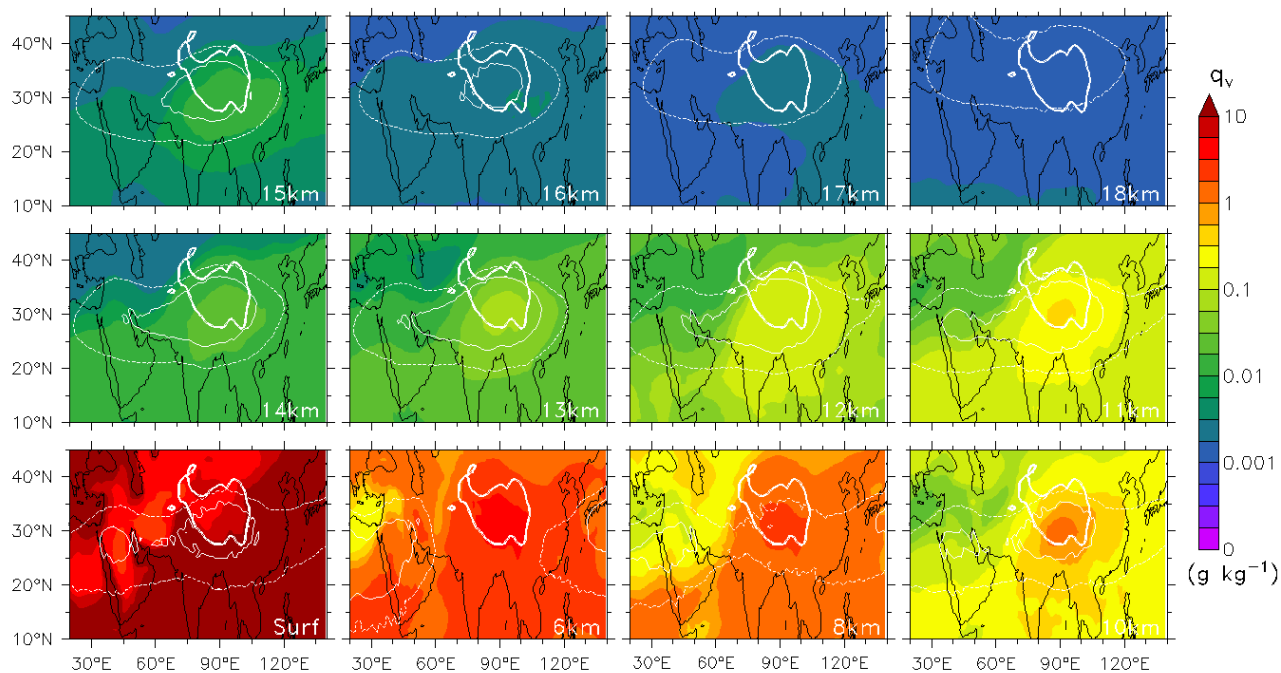
**Figure S8.** Same as Fig. 10, but for the year 2010.



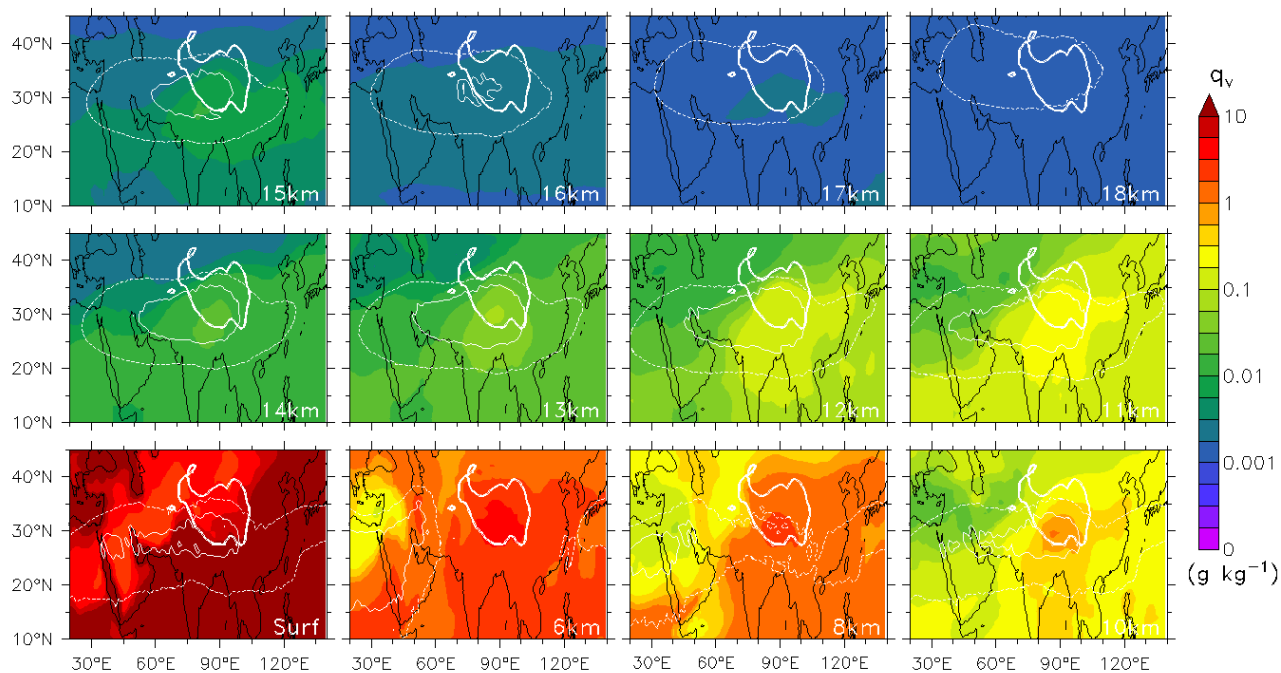
**Figure S9.** Same as Fig. 12, but for the year 2010.



**Figure S10.** Same as Fig. 12, but for the year [2011](#).

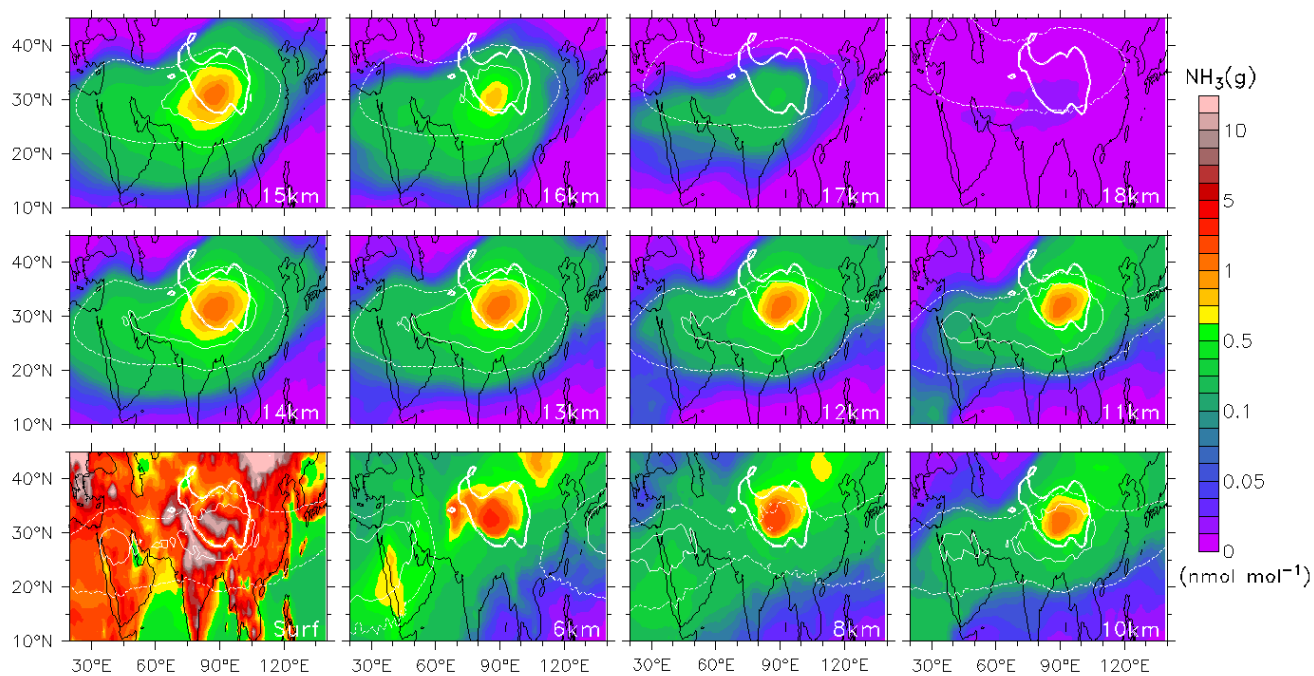


**Figure S11.** Same as Fig. 13, but for the year 2010.



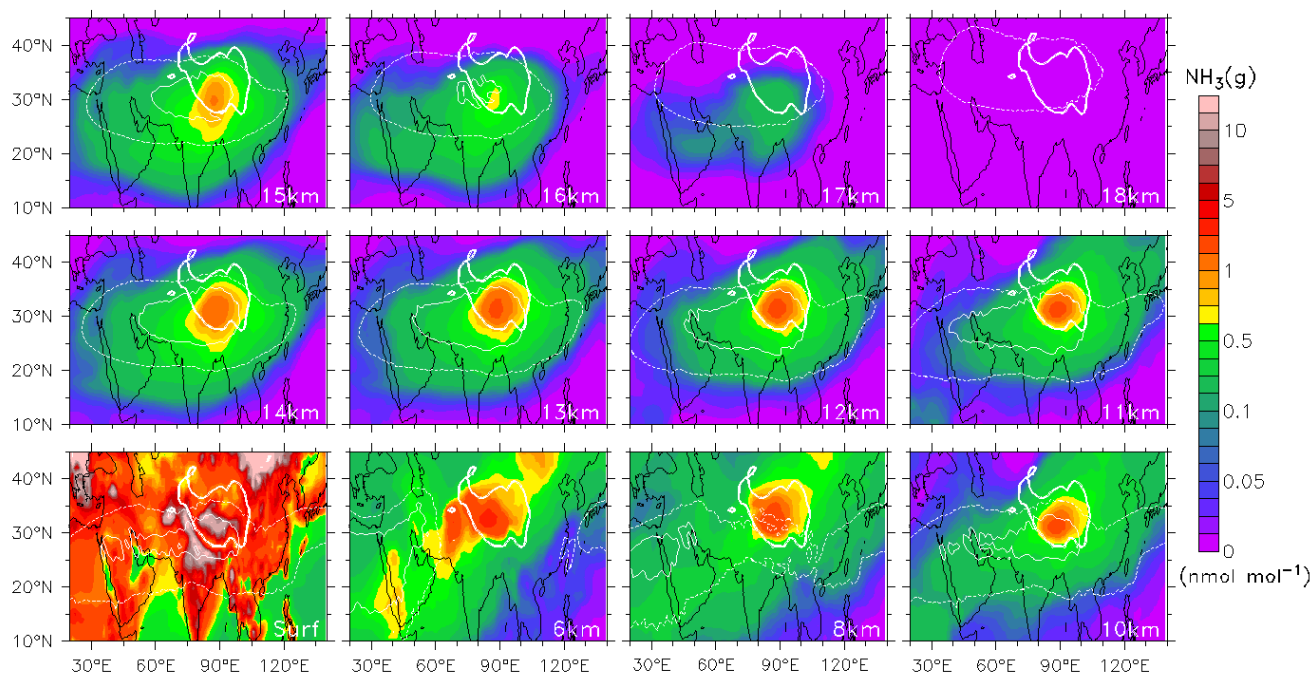
**Figure S12.** Same as Fig. 13, but for the year 2011.





**Figure S13.** Same as Fig. 14, but for the year 2010.





**Figure S14.** Same as Fig. 14, but for the year 2011.