

## Response to Review 2

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We thank the referee for her/his thoughtful comments and suggestions for improvements. We revised the manuscript critically and think that the manuscript has significantly improved after the comments and suggestions have been considered.

In the following, we respond to individual comments. Original remarks of the referee have been enclosed in quotation marks, using an *italic* font. Responses are given below each comment and are marked by "Answer" in a ***bold italic*** font.

### Major comments:

*"1) I think it would be useful to compare the magnitude of the stratospheric aerosol variations due to QBO to that of seasonal variability, annual variability, and volcanic influences. For instance, how much stronger is QBO than seasonal or annual (e.g. tape recorder) variations on stratospheric aerosol? Does the QBO phase impact aerosol properties more than recent volcanic eruptions in the lower stratosphere? upper stratosphere? This could be included in the abstract and some discussion and particularly the conclusions."*

**Answer:** Focus of our manuscript is the examination of QBO influences on aerosol microphysics. It is not our attempt to explain the time-series of tropical stratospheric aerosol in the recent past, which is largely contaminated by volcanic material, in particular beyond 2003 (e.g. Neely et al., 2013). Therefore, it is important to relate the strength of the QBO modulations to the strength of other variabilities or modulations which may affect the analysed aerosol properties. In this respect we agree to the referee. In the revised manuscript we add figures showing the comparison between the amplitudes of the annual cycle and the QBO. Also referring to Review #1, we will revise Section 3.2.1. where we compare our modelled SAD to observations (CCMI data set) and consider estimates of statistical significances and discuss the influence of the volcanic signature in the CCMI SAD in greater detail. That is, of course, an important point, which will be mentioned also in the abstract and the conclusions.

*"2) the recent SO<sub>2</sub> observations by Hopfner et al which included the contributions of QBO phase is a very relevant comparison to your model. It would be useful to conduct a more detailed comparison between the ranges observed by Hopfner et al and your model, provide a more detailed description of this dataset in your introduction and/or section 2.2, and compare seasonal, annual and QBO-induced variability between the model and dataset."*

**Answer:** We agree with the referee. In the revised manuscript we consider an analysis of MIPAS SO<sub>2</sub> (Hoepfner et al., 2013) in a consistent manner, as done for the model and the observation-based CCMI SAD data set.

*"3) There are many places in the paper where you provide qualitative terms like "strongly", "more or less", "substantially smaller", "QBO effect exceptional", "indicates to a certain extent", "heavily influenced" and "rather in-phase" etc. It would be useful to provide more quantitative terms such as x % larger or smaller in x region."*

**Answer:** In the revised manuscript we will avoid using those qualitative terms and consolidate our interpretation by referring to relative quantities where appropriate.

*"4) There are also many places in the paper with grammatical errors and typos. I've tried to list most of them in my specific comments but please double check other places."*

**Answer:** We thank the referee for several corrections and comments on typographic and grammatical problems.

*"5) How might contributions from aerosols that you don't include in these simulations convolute your analyses? Please provide some discussion around that. For instance meteoritic dust contributes significantly to upper stratospheric aerosol (see Neely et al., 2011), and recent volcanic eruptions (Vernier et al 2011) and other aerosol species such as carbon (Murphy et al 2007) contribute to lower stratospheric aerosol."*

**Answer:** With respect to volcanic material, we cannot neglect that it may leave an imprint in the inferred signatures of the CCMI SAD, a merged SAGEII/CALIOP data set. For the model the relationships are even more clear - there is no direct influence of volcanos considered, as we state clearly in our manuscript and the companion paper Hommel et al., 2011. As mentioned above, and also in our answers to Reviewer #1, in the revised manuscript we will improve the discussion about the imprint of volcanos in the CCMI SAD. However, we like to emphasise that the focus of our paper is the QBO-aerosol microphysics relationship and not an analysis of the stratospheric aerosol record as observed in the recent past. In our opinion it is necessary to first understand how aerosol processes are affected by the QBO, which is the dominant natural forcing in the lower tropical stratosphere, before the effects from the modulation by precursors with volcanic origin are separated. In this respect our work can be seen as one necessary step towards an in-depth understanding of the lower stratospheric aerosol behaviour as observed in the recent past.

With respect to particulate matter other than sulphate dominated (liquid) particles, we agree to the referee that other studies indicated their relevance for the stratospheric aerosol burden, AOD, radiative forcing etc. But, as said before, we focused on sulphate aerosols, and did not considered other species than H<sub>2</sub>O and H<sub>2</sub>SO<sub>4</sub> or even other particulate matter because sulphate clearly dominates the stratospheric aerosol mass. Our attempt is to reduce the complexity of the system as far as possible without losing physical meaning. In our opinion this is a common procedure in atmospheric science global model studies. It is beyond the scope of our study to show a complete picture of particulate matter dynamics in the lower stratosphere. Each process and each substance additionally considered would imply a much higher complexity in the relationships to be analysed and making an interpretation very complicated. Another, more technical aspect shall be noted here: we used a computationally relatively expensive aerosol scheme (sectional approach with 35 bins). Extending the scheme to other aerosol classes and mixing states would increase the computational demand beyond a reasonable level. For more complex model studies, e.g. in-detail studies on volcanic effects, other less expensive aerosol schemes are favoured - and succeeded already when coupled to the same host model as used here (e.g. Niemeier et al. 2009).

In this respect we like to mention that this paper presents work in progress. Until now, not much has been published about the detailed mechanisms of the QBO-aerosol microphysics relationship. So far those relationships have been either indicated from aerosol extinction/backscatter observations or have been shown for very few quantities on much shorter time-series from models (Brühl et al., 2012). To address this wide field of lower stratospheric aerosol processes, this study is a first step towards a deeper understanding by utilising a system of reduced complexity.

*"6) The discussion of ozone on p16256 is confusing. Are you presenting any of your ozone results here? If not, it seems risky to compare your model's aerosol extinction to observations of ozone and make conclusions regarding the relative changes."*

**Answer:** We do not show ozone. And, we have to correct the reviewer, we do not show aerosol extinctions either. Our attempt here was to point out that the QBO-aerosol relationship is nothing special related to aerosol exclusively. Instead most of the mechanism's exist for most of the trace constituents in the lower stratosphere. We have chosen the example ozone because the ozone-QBO relationship is the best explored and discussed in a variety of articles. We will carefully revise the section and make clear why ozone is simply an analogy. Although the magnitude of the QBO modulations in the mixing ratios of aerosol and ozone is approximately similar, we cannot prove in our study whether it arises from the same mechanisms which dominate the modulations.

*"7) I find it a little concerning that your section 3.4 Microphysical processes ignores coagulation and sedimentation. Aerosol microphysical processes occur together in complex ways, and for instance coagulation and sedimentation can alter the rates of condensational growth and evaporation. Perhaps you could devote some discussion as to the caveats of your approach in section 3.4."*

**Answer:** We thank the referee for mentioning this point. During the time of manuscript preparation we critically examined whether it makes sense to describe potential QBO effects in sedimentation and coagulation. We did not diagnosed both processes in a way making such a comparison meaningful (stated on page 16262 lines 23-26). We decided to withdraw the two sections about coagulation and sedimentation. Let us explain in a few words the reasons: Sedimentation, for instance, is diagnosed in terms of the sedimentation velocity for each aerosol bin that has been defined in the microphysics scheme and as an accumulated flux at the surface. Both parameters are not suitable to examine QBO effects. Since we did not have an appropriate measure to quantify coagulation, the process has not been diagnosed during model integration. An offline diagnostics also does not seem possible, because it cannot separate the competing size distribution shaping processes from each other. This could be achieved by sensitivity studies, switching on/off the microphysical processes, for instance, but such experiments have not been conducted so far and are subject of future research.

We will carefully revise Sec. 3.4 and refer to the two processes in an appropriate way.

### Specific items:

Basically, many of the other comments the referee listed under 'Specific items' refer to linguistic problems of the manuscript. We have considered carefully each of the remaining comments in the revision. We very much appreciate the valuable suggestions of the referee.

The more content specific comments are answered below:

*"Abstract: Please quantify the relationship between QBO and the anomalies. Instead of saying that the aerosol load is "predominately influenced by QBO-induced anomalies...", please state the relationship (easterly-phase causes xxxx to happen). Instead of saying "large impacts are seen" quantify the percentage change from one QBO phase to the other."*

**Answer:** We agree with the referee and rephrase the abstract.

*"p16244 line26: change "is influenced by" to "may be influenced by" (since for example Neely et al 2013 found very little contribution of asian aerosol to the stratospheric aerosol)"*

**Answer:** This is correct. The sentence has been rewritten.

*"p16345 line 15: Please provide more details with regards to "These problems are addressed in the current study". perhaps something along the lines of: "In this study we propose to quantify the contributions of QBO to changes in stratospheric concentrations of background aerosols and their precursors.""*

**Answer:** We agree with the referee. We will change the text accordingly.

*"p16246 line 19, 21: Add English et al., 2013 citation to the sentence describing Pinatubo studies using size-resolved models, and Campbell et al 2014 to the list of citations for background aerosol."*

**Answer:** We thank the referee for pointing us towards the Campbell et al. paper, which we didn't consider in the manuscript so far. In order to complete the list of models resolving the size of stratospheric aerosol, in the revised manuscript we will also refer to volcanic studies and cite e.g. the English et al. paper.

*"p16246 line 22: Be more specific in the "In this study" sentence, describing that you are mainly focusing on the impacts of QBO on stratospheric dynamics and aerosol."*

**Answer:** We agree with the referee and make the sentence more clear in the revision.

*"p16246 line 24: In addition to geoengineering, this work is valuable to understand the contributions of QBO and natural variability to recent observed changes to stratospheric aerosol."*

**Answer:** This is true, we will consider this suggestion.

*"p16248 line 19: 39 levels is somewhat coarse to capture stratospheric dynamics. Have you conducted any studies to determine whether the vertical resolution is sufficient to capture stratospheric processes?"*

**Answer:** QBO nudging greatly improves the representation of stratospheric dynamics. Without QBO nudging, tracer transport in the vertically coarse 39 layer model has strong deficits which arise mainly from too strong upwelling (see also Giorgetta et al., 2006). Our model configuration has been tested against the free-running, the QBO not reproducing, model version in some more detail. Results were published as a technical note in Hommel (2008; in German only). As shown there, the behaviour of the water vapour tape recorder was greatly improved in the QBO-nudged model, being in good agreement with the vertically much higher resolved 90 layer version of the host model. That gave us the confidence that we found an appropriate setup up to conduct our aerosol experiment. We didn't test the 90 layer version of the model, simply because during the time we conducted the studies we did not have the computational resources to perform a companion long-term integration with the higher resolved version coupled to this expensive microphysics scheme. It is true that technical advances have not stopped in the meantime. And, like other middle-atmosphere GCMs, also our host model underwent development, and the capacities of computational facilities increased rapidly. Nowadays, one could perform further studies on the subject with a free-running (i.e. no nudging) and higher resolved model, and also coupled to chemistry (see Neely et al. 2013; Dhomse et al., 2014).

We will carefully revise our manuscript in order to discuss potential deficits that may be associated with the model's vertical resolution.

*"p16249 line 20: What are prescribed "climatological" oxidant fields? Do they include the variations in stratospheric concentrations due to QBO? If not, it would be useful to quantify how much they vary between different phases of QBO, and how that might impact your model results."*

**Answer:** We thank the referee for this objection. Here, we refer to the climatological means of monthly mean oxidant mixing ratios derived from a long-term integration of the chemistry climate model MESSy (Jöckel et al., 2005). More details are given in Hommel et al., 2011. We will revise the paragraph carefully in order to make clear that we coupled sulphur chemistry to the aerosol module, which is not an interactive full chemistry scheme. Therefore, the oxidant fields are needed.

*"p16249 line 28: why does it take 6 years to reach steady state? stratospheric lifetime is typically a year or 2."*

**Answer:** This is true. But the model needs a few years longer to achieve equilibrium state because it was initialised based on the climatological mean zonal mean SAGEII volume density (University of Oxford retrieval, Wurl et al., 2010). More details on the basic experiment design are given in Hommel et al. 2011. It shall be noted that more details about the spin-up procedure are described in the technical note of Hommel (2008; in Germany only). Alternatively, one could initialise the model from scratch, i.e. no aerosols initialised. Then the aerosol is formed from the emission and it will need more than a decade until they have been well mixed in the atmosphere (we estimated 1.5\*maximum of mean age of air in the stratosphere is the required minimum spin up time for such a case).

*"p16250 line 4: Describe the specific "aerosol forcing data set" you are referring to. Extinctions? SAD?"*

**Answer:** Thank you for pointing this out. In the revised manuscript, it is now stated that the forcing data set consist of SAD.

*"p16253 lines 14+: this paragraph could use more citations."*

**Answer:** We agree with the referee and consider additional citations.

*"p16257 lines 1-4: It is important to take what into account? condensational growth? Is this more important that coagulation? It seems that several microphysical properties are important."*

**Answer:** We thank the referee to point this out. We rephrase the paragraph in order to state more clearly, that biases between model and remotely sensed integrated aerosol size quantities may be easily introduced, when in the calculation of the model parameters the model's aerosol size range is not adopted to the detection range of the instrument.

*"p16257 line 5: please quantify "strongly depends". For instance something like "including particles smaller than xx nm increases SAD by xx %""*

**Answer:** We agree with the referee and rephrase the paragraph.

*"p16257 lines 14-15: I don't believe that larger particles evaporate at higher rates than small particles. As large particles start to evaporate they become small particles. Please clarify."*

**Answer:** We agree with the referee. This is misleading and has been removed from the manuscript.

*"p 16257 line 22: do you mean greater than 0.005 um instead of less than?"*

**Answer:** This is correct, it is a typo.

*"p16258 lines 12-20: how does your modeled SAD compare to SAGE when you cutoff particle size smaller than the detection limit of SAGE?"*

**Answer:** The bias to SAGEII would be even more pronounced when more fine mode aerosols are considered in the SAD integral of the model. In our companion paper we show this effect for the effective radius compared to measurements from the University of Wyoming optical particle counter (Hommel et al., 2011; Fig. 13).

*"16259 line 18: negatively biased to what kind of observations – satellite or aircraft? Satellite observations have known biases as you've stated but aircraft observations are more reliable"*

**Answer:** This sentence refers to SAGE II observations based on the climatologies provided by the University of Oxford (Wurl et al, 2010) and NASA AMES (Bauman et al., 2003a,b). The sentence has been rewritten.

*"p16261: the paragraph discussing nucleation should probably go before the current preceding paragraph which discusses other microphysics. References"*

**Answer:** We are not entirely sure what the referee means. In our opinion also the nucleation process depends on the water content and the stratospheric temperature, i.e. two key aspects which are mentioned in the introductory paragraph of Sec. 3.4, intended to introduce the chapter about the modelled QBO impact on aerosol microphysics. In our opinion, also nucleation is a microphysical process, therefore we like to keep the section's structure. Appropriate references will be added.

*"p16261 line29: nucleation mode does not prove BHN occurs as other processes such as ion-mediated nucleation may occur. perhaps state that it suggests BHN is occurring."*

**Answer:** We agree with the referee and rephrase the paragraph.

*"p16262 line 21: "vapour contents" is not a common way to describe the thermodynamics. Perhaps use the words "supersaturation of h2so4 and water, which depends on temperature and vapor concentration..."*

**Answer:** We agree with the referee and rephrase the paragraph.

*"p16263 line 9: What do you mean by "can amount to 50%"? under which circumstances?"*

**Answer:** We will rephrase the sentences and more precisely describe the result.

*"p16263 line 16: what are the units of time-averaged molecule concentration transferred? seems like time should be on the denominator, but this is not noted in Fig. 9."*

**Answer:** We diagnosed, as in Hommel et al. (2011), the H<sub>2</sub>SO<sub>4</sub> molecule concentration that is transferred between the two phases. Units are molecules per cm<sup>3</sup>. This quantity is directly comparable to the 'normal' sulphate concentrations of the liquid and gas phase. Diagnosing this transferred concentration directly helped us a lot to understand how the model behaves, e.g. how the size distribution is shaped in the presence of steep gradients. Diagnosing a rate would have made sense if we would have had other data to compare. But in literature we did not find comparable mass transfer rates of sulphate under representative stratospheric conditions (in contrast to nucleation rates), so that we tried to retain control over the modelled mass transfer process on the molecular level (incl. extensive mass balancing). To our knowledge, also deviations of microphysics process fluxes due to the QBO have not been published so far.

*"p16264 line 21: does warmer T also explain the changes in saturation vapor pressure above 20 hPa?"*

**Answer:** We are not entirely sure what the referee means. The saturation vapour pressure is modulated by the QBO in an almost linearly manner, under the assumption that no additional gain or loss due to mass transfer occurs.

*"p16265 lines 14-19: could the temperature biases affect modeled nucleation and growth in addition to evaporation as you've noted?"*

**Answer:** The referee raises a good point. Nucleation should be affected, yes. Condensational growth theoretically also, but we assume the effect is more critical for evaporation. Because this process limits the upper tail of the Junge layer. And, as we mentioned in our manuscript, this upper tail varies due to the QBO by up to 5 km. Thus, potential temperature biases should affect especially those processes which occur there. We will rephrase the paragraph, pointing out the importance for the other processes.

*"p16266 line 18: how does QBO "interfere" with the annual cycle?"*

**Answer:** We will consider additional citations here, rephrase the paragraph and describe in brief the relationships.

*"p16267 line 27: add Campbell et al 2014 citation."*

**Answer:** We thank the referee for pointing us towards the Campbell et al. paper. We like to state that we did not have knowledge about its existence during time of manuscript preparation. We will cite it in the revised manuscript.

*"p16268 lines 16-25: A more direct comparison between your model SO<sub>2</sub> and Hopfner et al would be useful. How do each SO<sub>2</sub> vary between QBO phases? how do so<sub>2</sub> annual and seasonal variations compare? "*

**Answer:** As mentioned above, we will add an consistent analysis of MIPAS SO<sub>2</sub> (Hoepfner et al., 2013).

*"p16269 lines 17-21: this reasoning is not clear to me. To me, aerosols in the lower stratosphere seem strongly driven by transport from upper troposphere, but aerosols in the middle stratosphere are more driven by OCS oxidation. Please clarify your reasoning."*

**Answer:** The sentence "Together with ... indicates ... to a certain extent ..." refers to the sentence before - or in other words to anomalies in the Aitken mode number density. Which are largely in-phase with SO<sub>2</sub> and H<sub>2</sub>SO<sub>4</sub> vapour anomalies up to the 30 hPa pressure level. Additionally, we find also in-phase anomalies in the nucleation mode, and obviously in the nucleation rate, around 50 hPa, which suggest that not all of the Aitken mode aerosol in this region has been formed in the free troposphere. Whether the H<sub>2</sub>SO<sub>4</sub> vapour at this altitude comes from OCS oxidation or SO<sub>2</sub> is not entirely clear from our model, because we used prescribed OCS mixing ratios (due to the absence of appropriate emission data during time we performed the experiment).

In our perspective, we found a reasonable chain of process modulations here, and do not rule out that fine mode aerosols are transported through the TTL. We will carefully revise this section in order to avoid misunderstandings.

*"p16269 line 26-27: add "and so2 measurements (Hopfner et al 2013)."*

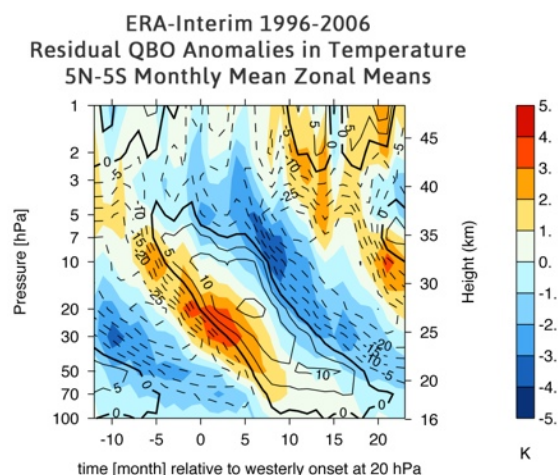
**Answer:** Considered in the revised manuscript.

*"p16271 lines 24-27: the assumption that condensation and evaporation occur concurrently seems risky. I would suggest that you analyze your instantaneous model output to determine whether this is true, or change your discussion of it."*

**Answer:** We explicitly state "... in the time average ...", at the beginning of the criticised sentence, line 24. It is a fact that during an output time step (6 hours), which is considerably longer than an integration time step (15 minutes), both processes can be diagnosed within the same grid cell. This has to bear in mind when the QBO signatures in the two processes are analysed. Because equally phased anomalies may overlap partially in the residual composites, although condensation and evaporation compete on the process level in the model. Also we like to mention that the output is time-averaged, and not instantaneous. At the beginning we also had some doubts and revised the results several times (also results of the companion simulation, Hommel et al., 2011, where it is similar but less pronounced). We have no indication that this is an error in the diagnostics or the model itself - instead for us it is very plausible mechanism and a straightforward model behaviour.

*"Fig. 2. Why is there a sharp gradient in ERA-interim at 15 hPa?"*

**Answer:** We are not entirely sure what the referee means. We think she/he refers to a horizontal line of colour shades in Fig. 2 a around the 15 hPa pressure level, representing the QBO induced temperature anomaly in the ERA-Interim reanalysis. We revised the data and it seems that the referee refers to a visual impression that arises from the so-called 'blockfill' technique of our post-processor, which fills the area between the data points with their colour-coded cell averages. Below, we reproduced the figure without using the 'blockfill' technique. Instead, here it has been linearly interpolated between the data points. No exceptional vertical gradients in the temperature anomalies can be found.



*"Fig. 3: I thought your control simulation had prevailing easterly winds? why are there some non-dotted lines (e.g. westerlies)? Also "Ratio" is mis-spelled in the title"*

**Answer:** We thank the referee to point this out. It should be noted in Sec. 3.1.1, and also in Sect. 2.3 'Meteorology' that in the CTL experiment westerly winds of the semi-annual oscillation (SAO) penetrate down from the mesosphere to the 30 hPa pressure level. Fig. 1c is showing these westerlies in reddish colours, enclosed by a continuous black contour line, marking the zero zonal mean wind. The same zonal wind contour lines overlay the aerosol mixing ratio in the CTL experiment, Fig. 3a. We will rephrase respective paragraphs and describe this model feature. However, the prevailing wind regime in the CTL experiment is easterly (see also Giorgetta et al., 2002).

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