

Interactive comment on “Impact of the eruption of Mt. Pinatubo on the chemical composition of the stratosphere” by Markus Kilian et al.

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We thank referee 2 for the quick report. Here are our replies to the comments:

- *General Comments: The paper tries to separate the effects of aerosol heating and aerosol chemistry on ozone. It is, however, an odd concept to nudge temperatures and winds to observations in the region where temperature and dynamics changes due to aerosol should be analysed (page 5 and 6). This must have consequences for the results. To get some feeling for the introduced artifact it would be good to perform a sensitivity simulation with nudging only up to about 100hPa to get the tropospheric wave forcing but the unperturbed effects of calculated radiative heating due to Pinatubo aerosol on stratospheric dynamics, QBO nudging*

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based on the Singapore data might be on for this case. The manuscript might be published after revision.

Reply: Thank you for your feedback. However, we disagree that we use an “odd” concept to study the Mt. Pinatubo eruption with nudged simulations. This concept has explicitly been selected as appropriate due to the following reasons:

Nudging (to ERA-interim analysis data) is applied to the prognostic variables temperature, divergence and vorticity (-> horizontal wind field), and the logarithm of the surface pressure. We apply this nudging in the spectral space by omitting the nudging of wave-zero of the temperature, thus we do not correct temperature biases, implying that the absolute temperature can evolve. Moreover, the nudging is applied as low-normal mode insertion, i.e. down to the synoptic scale only, with comparably long relaxation times. The nudging is applied such that the large(r than synoptic) scale patterns correspond to those of ERA-Interim, but not the absolute temperature. This means that the synoptic situation is that of ERA-Interim, whereas sub-synoptic variations can evolve freely. Such as for instance the influence of the volcanic cloud on the vertical velocity and the temperature profile.

The effect of nudging on the results was already discussed in detail in a previous study on the Mt. Pinatubo eruption by Löffler et al. (2016, their sections 2.3 and 5, see also their supplement). In that study the scientific focus was on the change of water vapour due to the eruption. During the review process of the study the question came up, how the nudging of temperatures might influence the results. This point is indeed important, as the water vapour change due to the eruption strongly depends on the temperature distribution at the cold point and hence on how the aerosol heating of the volcanic cloud is represented in a nudged simulation. As part of that study, an additional set of sensitivity simulation pairs (one simulation with (VOL) and one without volcanic eruption (NOVOL)) with prescribed monthly average chemistry (to save computing time) was performed to address the sensitivity of the model results to the nudging procedure. The

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simulation pair we are interested in (called QF – quasi free running simulation) nudged only the (logarithm) of the surface pressure (and prescribed SST/SIC) to study the effect of omitting the nudging of temperature, divergence and vorticity on the results. The presented results are all differences between a simulation with volcano (VOL) and without (NOVOL) in the specific model configuration, i.e. for our case the QF pair. This previous sensitivity study does not exactly use the nudging height as proposed by the referee, but represents a simulation where the model is nearly free-running, but does not deviate too far from the actual synoptic situation due to the surface pressure nudging. The effect of the volcanic eruption on the temperature in different heights in the stratosphere can be seen in the Figure S2 below (taken from the supplement of Löffler et al., 2016). The temperature change (VOL-NOVOL) for QF is overestimated compared to the nudged simulations and, moreover, it appears noisy. However, the development of temperature is similar to the nudged simulation pairs (FC-full chemistry and RE-prescribed chemistry). For more details we refer to Section 2.3 and Section 5 (page 6557) of Löffler et al. (2016).

Using nudged simulations has several advantages over free-running simulations to study the impact on the chemistry: The temperature response is closer to observations, which is important, as ozone chemistry is temperature dependent. The results appear less noisy. Our (nudged) simulation pair (VOL and NOVOL) are similar with respect to the synoptic situation, so the effect of aerosol heating on subgrid-scale chemistry and transport of ozone can be contoured more clearly. This would be more difficult, if one allows the synoptic situation to evolve freely. Moreover, for such a concept a large set of ensemble simulations is necessary (see for instance Aquila et al., 2013).

We will add the paragraph (item 3 from above) to the description of our methodology (section 2.3) to the revised manuscript.

We agree, however, that our concept is not appropriate to study the effect of the volcanic eruption on the global dynamical system, as it has been studied by Graf et al. (1993) and this was not our intention, either. Although we thought that we have men-

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tioned that in our manuscript, it was obviously not clear or detailed enough. We will also refer to our sensitivity simulation with respect to nudging as described by Löffler et al., 2016 (see their Supplement and the open review discussion).

2 Specific comments: Page 1, line 5: The use of CCMI slang hides the problem with nudged temperatures up to 10hPa.

Problems with "specified dynamics" and/or "T42L90MA"? The used wording corresponds to the standard expressions used in modeling and is further explained in the main text. We already discussed the influence of temperature nudging further above (page 1).

- *Page 1, line 10f: Separate between aerosol and ozone heating (see section 4.1), please reword for clarity.*

Reply: Thank you for this hint. We will clarify the difference between the volcanic heating by the aerosol and the secondary heating effect due to the ozone increase in the revised manuscript. We will add the following explanation to section 4.1:

"The strongest heating due to absorption of solar and terrestrial infrared radiation by volcanic aerosols and by the increase of ozone due to transport occurs in the middle stratosphere of the tropics (Figure 4b)."

- *Page 2, line 8: Don't forget to mention the terrestrial infrared.*

Reply: We will mention the terrestrial infrared radiation in the introduction of the revised manuscript as suggested.

- *In the introduction references to earlier studies with EMAC on Pinatubo are missing.*

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Reply: We forgot to cite the study by Brühl et al. (2015), who presented an inter-comparison between observations and model simulations of the stratospheric sulfur cycle and its relation to radiative and dynamical processes. We will add the reference to the revised introduction.

- *Page 4, line 6: Is the assumed distribution monomodal or how many modes are included?*

Reply: Thank you for your comment. The assumed distribution is a single-mode log-normal aerosol size distribution (Revell et al., 2017). We will add this information to the revised manuscript.

- *Page 5, line 17: 2011 is perturbed by the medium size volcanic eruption of Nabro and therefore not background. Where are the data from? SAGE died in 2005.*

Reply: Thank you for your critical question. Indeed, you are right, the aerosol data of our NOVOL simulation originate from the CCMI dataset, more precisely from CALIPSO (2006-2012) and not from SAGE (which terminated operation in 2005) (Diallo et al. 2017; Revell et al., 2017). The definition of a “background” aerosol distribution refers to a period, when volcanic activity and related aerosols are less present. We selected the year 2011 because no strong eruption occurred in the decade before. Nevertheless, the medium size volcanic eruption of Nabro in 2011 is indeed included in the CCMI dataset. In comparison with other eruptions Nabro just emitted 1 Tg SO₂ (Pinatubo: 20Tg) into the atmosphere up to heights of 14 km. The tropopause height at 15° N is at 17 km and thus above the volcanic plume. Therefore we assume that only a negligible amount of sulphate aerosols were emitted into the stratosphere in that year. We will correct this point in section 2.2 and discuss this in our revised manuscript.

- *Page 7, line 18: More details please, NIR, IR. Transport effect on ozone heating?*

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Reply: Thank you for your comment. The assumed distribution is a single-mode log-normal aerosol size distribution (Revell et al., 2017). We will add this information to the revised manuscript.

- *Page 9, section 4.2: Why is 1993 not addressed? In this year were the largest effects on total ozone in midlatitudes in observations but also in Fig. 5. It would be also good to compare with observations here (e.g. TOMS).*

Reply: You are right. Overall, 1993 shows the strongest decrease of the ozone column after the eruption. So from the point of view of the observations, this might be the most interesting year to analyze. The separation of the effects, however, shows that the chemistry effect, resulting in strong ozone depletion, has the largest impact already in 1992. Nevertheless, most of our analysis is represented as a time series, thus the year 1993 is already considered in the presentation of the results. We will compare the simulated total ozone in the midlatitudes with the TOMS observations in the revised section 3. Thank you for the hint.

- *Page 10, line 1: This appears to be in contradiction to Fig. 4.*

Reply: Thank you for your comment. This might be a misunderstanding. The differences between the effects (chemistry and heating effect) are additive: the combined effect (VOL-NOVOL) is the sum of the heating effect (VOL-CVOL) and the chemical effect (CVOL-NOVOL). This can be shown for total ozone, but also for the temperature in Figure 4. Due to a remark from reviewer1 we eliminated the word “linear” in line 2 of Page 10 and hope that this clarifies this misunderstanding.

- *Page 10, line 31: You may mention PSCs here.*

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Reply: Thank you very much for your feedback. We will refer here to the formation of PSC's and give a reference to section 4.4.1 in the revised manuscript.

- *Page 12: There are several ways to separate the catalytic ozone destruction cycles. You may discuss the meaning of HO₂ + NO for aerosol perturbed lower stratospheric ozone.*

Reply: It is not really clear to us, what the referee suggests here. The reaction HO₂ + NO → OH + NO₂ is a gas-phase reaction which affects the interconversion between HO₂ and OH, in other words their ratio at all altitudes. Their ratio is controlled by the temperature dependent reaction rate and through the concentrations of O₃ and NO. In the lower stratosphere the reaction HO₂ + O is negligible, and HO₂ + O₃ is predominant. Since NO decreases in the winter 1991 in the lower stratosphere in the tropics and in spring 1992 in higher latitudes, more HO₂ is available to react with O₃ instead of with NO. This leads to an increase of the meaning of the HO_x cycle for the lower stratosphere.

We have added this explanation into section 4.4 of the revised text.

- *Figures 10 and 11 might be merged, as well as Figs. 12 and 13. It should be also better to show a common pressure level in the lower stratosphere in tropics and extratropics (Figs. 9 to 13) instead of 30 and 20hPa. What is included in Ox? Please define, the Chapman cycle alone cannot explain the curves for the lower stratosphere.*

Reply: Indeed, it is a good idea to merge Figures 10 and 11, and Figures 12 and 13 into one figure, respectively. We will modify the manuscript accordingly. However, we are very much hesitating to show common pressure levels in Figures 9-13. The vertical levels were selected due to the results displayed in Figures 6-8, because at 20 and 70

hPa the strongest ozone decrease/increase could be found and therefore these levels appeared to be most interesting to analyze. Yet, we will motivate this in the revised manuscript.

Our study, includes all relevant Chapman equations in the Ox cycles such as the production of ozone via $O_2 + hv \rightarrow O + O$ and $O + O_2 \rightarrow O_3$, as well as the ozone depletion via $O_3 + hv \rightarrow O_2 + O$ and $O + O_3 \rightarrow 2O_2$. Note that the photolysis rates in our study are unaffected by the volcanic aerosol. Hence, the photolysis of ozone might be overestimated.

We do not understand the last point you addressed, that “the Chapman cycle alone cannot explain the curves for the lower stratosphere”. Nowhere, we claimed that the Chapman cycle alone explains the curves for the lower stratosphere. In Figures 11 and 13 we find the ClOx, BrOx and HOx cycles being important in reducing ozone, when the NOx cycle is reduced by the volcanic aerosols.

- *Page 17, line 27: There should be also PSCs lower down, at least to 80hPa.*

Reply: You are right. The data in Figure 14 are the summed volume of PSCs down to 80 hPa. We will clarify this in the revised manuscript.

- *Section 4.5: H2O is very sensitive to uncertainties in the parameterized satellite data and gap filling in the lowermost stratosphere, please discuss. This section is difficult to understand.*

Reply: Thank you for your feedback. We agree that the uptake of water vapour by aerosols is sensitive to the aerosol surface area density and consequently affected by uncertainties in the satellite data and gap filling of the CCMI dataset. We will mention this uncertainty in the discussion of the revised manuscript.

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- *Page 21, line 3: Here the artificial heating/cooling from nudging can cause artifacts.*

We apply nudging in the spectral space by omitting the nudging of wave-zero of the temperature, thus we do not correct temperature biases, implying that the absolute temperature can evolve. Moreover, the nudging is applied as low-normal mode insertion, i.e. down to the synoptic scale only, with comparably long relaxation times. The nudging is applied such that the large(r than synoptic) scale patterns correspond to those of ERA-Interim, but NOT the absolute temperature. This means, that the synoptic situation is that of ERA-Interim, whereas sub-synoptic variations can evolve freely, such as for instance the influence of the volcanic cloud on the vertical velocity and the temperature profile. We will extend the discussion and display the results from Löffler et al,(2016) on the comparison of a nudged and free-running simulation of the Mt. Pinatubo eruption.

- *Page 22, line 9ff: Here the sensitivity study without temperature nudging between 10 and 100hPa should be discussed. This paragraph is too tentative now.*

We will discuss and refer to our results from the quasi-free running simulation as presented and discussed by Löffler et al. (2016) in the revised manuscript. In that study the effect of nudging on the results of the Mt. Pinatubo eruption was already presented. We agree that this aspect is so important that it should be addressed in this study, too.

- *3 Technical corrections Page 1, line 8: better "background" here.*

Reply: Yes, you are right we will reword this.

- *Page 1, line 23: "photolysis of O2".*

Reply: Thank you, we added O2.

- *Page 2, line 16: "reduction" instead of "loss".*

Reply: Yes, corrected.

- *Page 3, line 27: Is this the correct meaning of the acronym? There are several versions around in the literature, also it differs from the abstract.*

Reply: We will use the following description for our model: Version 2.51 of the European Centre for Medium-Range Weather Forecasts-Hamburg (ECHAM)/Modular Earth Submodel System (MESSy) Atmospheric Chemistry (EMAC) model. We will correct it in the revised manuscript.

- *Figures 1 and 2: Better use a logarithmic color scale instead of the scale with the arbitrary jump by one order of magnitude at $10\mu\text{m}^2/\text{cm}^3$.*

We do not really understand your criticism. The shown figure of the aerosol surface density has a logarithmic scale except for the range < 1 because these values are showing the background aerosol and are not important at this point. We think, that our representation is an appropriate way to highlight the volcanic aerosol plume.

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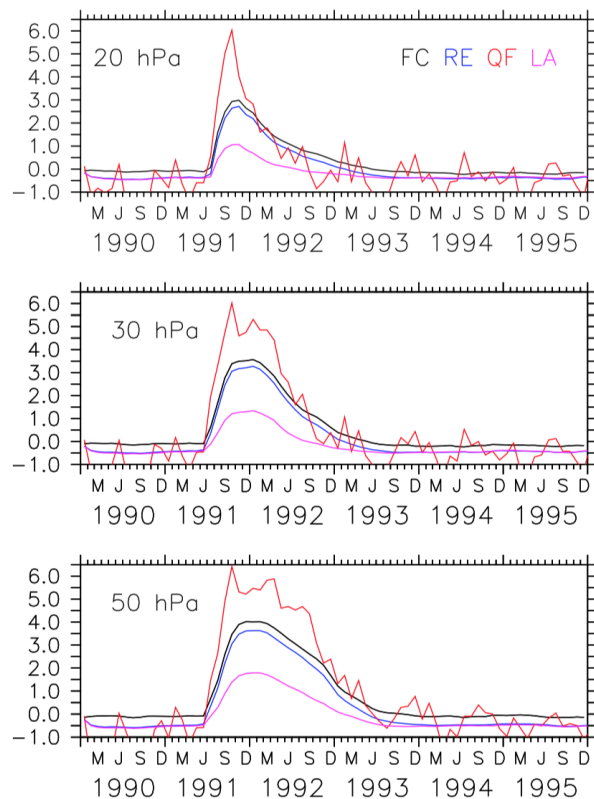


Figure S2: Temperature [K] differences (VOL-NOVOL) for the tropics (5°S-5°N), zonally averaged after the June 1991 Mount Pinatubo eruption for 20 hPa (upper panel), 30 hPa (middle panel) and 50 hPa (lower panel). The different simulation pairs are coloured as labelled in the upper panel.

Fig. 1.