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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Analysis of global methane changes after the 1991 Pinatubo volcanic eruption" *by* N. Bândă et al.

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General remarks:

We would like to thank the referee for the helpful comments. Guided by the comments of both referees, we made some major changes to the manuscript, that are listed below:

- Both referees commented on the need to use both steady state and transient simulations. The motivation of using both steady state and transient simulations was to show the difference between the instantaneous source-sink imbalance and its effect on transient concentrations. However, since this seems to increase the complexity of the paper and is not necessary for the final conclusions, we

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decided to remove the results of the steady-state evolution. Therefore the purple plot in Fig. 3 and Fig. 5a of the old manuscript are removed.
In view of these changes, section 3.2.2 of the old manuscript was also removed, and the discussion of the concentrations and growth rates was grouped in one single section.

- Both referees asked for more validation of our model with observations. We therefore included a validation using methyl chloroform (MCF) measurements for the period 1988 to 2005. We model offline MCF concentrations using time-varying OH fields from a transient run of the column chemistry model. We added a new figure in which the modelled and observed global mean concentrations of MCF are presented.

- The comparison of MCF concentrations with measurements shows that surface concentrations are too sensitive to the emissions. We believe this is related to a difference in the sampling of the model and the observations. Observations are taken in remote areas, away from the emissions, and are considered to be representative of the global burden. In contrast, in our model, emissions are put in the surface gridbox where the surface sampling is done. We find that modelled tropospheric mean MCF concentrations represent well the observed global means. Therefore we switch from a surface sampling to a tropospheric mean sampling of the model. This has little effect on our conclusions.

- We retuned the model in order to obtain a better comparison of our modelled budgets to more complex 3D models, and in order to obtain a good comparison between observed methane concentrations and the modelled tropospheric means, rather than the modelled surface concentrations.

  – The latitudinal band used for atmospheric profiles and stratospheric ozone was changed from 20°N to 30°N.

ACPD
12, C10207–C10220, 2012

- The vertical diffusion coefficients were increased to improve the vertical gradients in modelled concentrations.
- The CO yield from NMVOC was decreased from 0.5 to 0.35, to have a better comparison of the global CO production from NMVOC with 3-dimensional GCMs.
- The amount of lightning NO_X was changed from 5 Tg yr^{-1} to 6.3 Tg yr^{-1}, in order to be consistent with Huijnen et al. (2010), as done for other natural emission values.

Following these changes we obtain a stratospheric inflow of ozone of 240 Tg yr^{-1} and a more realistic ozone mixing ratio of 148 ppb near the tropopause.

- We followed the suggestion of referee # 2 to introduce a table in which we summarise sensitivities of our model and those found in other studies. To increase clarity of the text, we also present these in a separate subsection.
- We removed the isoprene effect from the previous Fig. 4, and included it instead as a sensitivity in the model validation section.
- We regrouped previous Figs. 5b and 6a in a single figure. Previous Fig. 6b becomes Fig. 7.

Replies to general comments:

"It should be pointed out more clearly that a quantification of the processes influencing the methane concentration is of limited significance with the strongly simplified model they are using. The manuscript rather presents a sensitivity study - which is very interesting in itself and a prerequisit for a study using a more sophisticated model system."

ACPD
12, C10207–C10220, 2012

Our study is indeed a sensitivity study that uses a simplified approach. To make this clearer, we mention the fact that this is a sensitivity study in the revised manuscript.
In the abstract: 'We present a sensitivity study of the effects that these processes had on CH4 concentrations.' instead of 'We quantify the effects that these processes had on methane concentrations'.
In the introduction: 'In this sensitivity study we will use a simplified tropospheric column chemistry model.' instead of 'In this study ...'
In the discussion section: 'Since our study is an idealised sensitivity study, there are many reasons that may explain differences between the modelled and the observed growth rate.' In the conclusions: 'Although we acknowledge that model is simplified and difficult to apply in a globally-averaged fashion,...'

"In general, the paper is well structured and well written. The sensitivity studies are systematically evaluated. However, it is not stated clearly enough which simulation represents the base line for the evaluation."

Thank you for the positive evaluation. Indeed the base simulation was not clearly defined. Now we add a sentence in the Sect. 2.4 that states which is the baseline simulation. 'In the first set of simulations, we define the base simulation as the 1990 equilibrium situation.'
Additionally, we make a more clear the distinction between the two sets of simulations performed.

"Furthermore, the need of having both, steady-state and transient simulations, should be motivated in the beginning."

This issue has been addressed above in the general remarks.

Replies to specific comments:

ACPD
12, C10207–C10220, 2012

"P18030, L9: To really quantify the effects of the processes a more sophisticated model appraoch would be needed. With a simplified, one-dimensional model this is rather a sensitivity study."

This has been corrected in the revised manuscript.

"P18031, L28: It should be metioned that the higher interannual variability found by Prinn was for an earlier time period, ie. pre-1995."

The reviewer is correct. We have included this in the revised manuscript: 'In a previous study, Prinn et al. (2005) found an interannual variability in tropospheric OH of 7 to 9% for the period 1978 to 2004. '

"P18032, L6: State what was emitted?"

Corrected 'the eruption emitted about 18 $\pm$ 4 Tg' to 'the eruption emitted about 18 $\pm$ 4 Tg SO_2'.

"P18040, L3-11: List more clearly the simulations performed. Some more details form the reference simulations are needed. Which simulations are shown in Fig. 3? Why are these simulations starting at methane concentrations considerably higher than the observed value in 1890? Does the 'base simulation' contain anthropogenic CH4 emissions, which vary in time, like those shown in Fig.3?"

The "base" simulation is the equilibrium state in 1990. (we add the sentence 'In the first set of simulations, we define the base simulation as the 1990 equilibrium situation.') Therefore the 'base simulation' has fixed emissions.

The simulations in Fig. 3 of the old manuscript start with the equilibrium condition obtained using emission values for the year 1890. These concentrations are higher than the observed ones because the atmosphere was likely not in equilibrium in 1890. The anthropogenic emissions of methane were already increasing in 1890, leading to

ACPD

12, C10207–C10220, 2012

an increase in methane concentrations. Therefore the equilibrium methane concentration obtained using the emission values for the year 1890 is higher than the observed transient one.

"P18040, L20-21: That 'Surface concentrations fall well within the range of observations' is not at all shown in this section. But this would be required to allow the reader to judge this statement. Please add a comparison with observations here."

For the validity of this study it is important that the CH_4 lifetime and concentrations are well represented. This is shown in Fig. 3. In the revised manuscript, we have added a comparison of modelled MCF with observations, as a validation of OH concentrations. For the other species in the model, it is important to have representative global burdens and budgets, in order to obtain a realistic CH_4 lifetime and correct model sensitivities. For the purpose of this sensitivity study, we do not consider it essential to compare CO, O_3, and NO_X concentrations to observations. It is also difficult to compare our global column to profiles of short-lived species due to their large spatial variability. The profiles for these species are shown for general interest only. The sentence 'Surface concentrations found by our model fall well within the range of observations.' has been removed from the manuscript.

"P18041, L20-28: The comparison with budgets from other studies needs to be evaluated more critically. Differences in some components are substantial, eg. for ozone stratospheric exchange. Consequences for the validity of this study should be discussed."

Following the retuning of the model described above, we obtain a stratosphere-troposphere exchange of 241 $Tg\,yr^{-1}$ ozone. This is still lower than the range of three-dimensional global transport models. We note, however, that transport from the stratosphere to the troposphere occurs mainly in the extratropics (Gettelman et al., 2011), while the tropics are dominated by transport of air from the troposphere into the

ACPD
12, C10207–C10220, 2012

stratosphere. We use here tropical profiles for temperature and stratospheric ozone, which might not represent well the global stratosphere-troposphere exchange. We find that the effect of stratospheric ozone depletion in our results is dominated by the effect through radiation. Therefore uncertainties in the stratospheric inflow are believed to have a small impact on our results.

"P18042, L14-28: Given the large overestimation of sensitivity factors of CH4 and OH to changes in the ozone column compared to estimates from more sophistcated 3-D CTMs, the question arises wether the results of this study is sinificant at all. At least the authors point out that their results should be interpreted with care. Please state why the study should nevertheless give meaningful insights."

We agree that our sensitivity has a range of uncertainty. However, it is difficult to draw a definite conclusion about the quality of the sensitivity of our model to changes in ozone column by comparison to the studies of Fuglestvedt et al. (1994); Camp et al. (2001), because of differences in the setup. This sensitivity is larger in the tropics, where CH_4 is mostly oxidised, and smaller in the extratropics. In our model, the tropical ozone columns and atmospheric profiles used imply that our sensitivity is representative for the tropics. Contrarily, the ozone column changes in these studies are driven by the extra-tropics. For example, Fuglestvedt et al. (1994) look at the effects of stratospheric ozone on OH and CH_4 lifetime. Their global mean ozone changes are dominated by the high southern and northern latitudes. The tropical ozone changes found in Fuglestvedt et al. (1994) are of the order of 2% (Fig. 6), while the global mean change is 4.51% (Table 6). This also induces large changes in OH at high latitudes. However, these will have less impact on the CH_4 lifetime.

After Pinatubo, changes in stratospheric ozone occurred in the tropics and in the extra-tropics with similar magnitudes (Chipperfield et al., 2003). The perturbations in OH and CH_4 concentrations due to stratospheric ozone changes after Pinatubo are comparable to those found by other studies (Bekki and Pyle, 1994; Wang et al., 2004). Therefore

ACPD
12, C10207–C10220, 2012

the sensitivity of our model to stratospheric ozone is not unrealistic.

Based on the above we replaced the comparison of our ozone sensitivity to the studies of Fuglestvedt et al. (1994); Camp et al. (2001) with a comparison to Bekki and Pyle (1994). They find a 6% increase in OH due to a 6% decrease in ozone column, while we find a 6.4% OH increase following the same perturbation in ozone column. Note that in the later study, the sensitivity of OH to changes in ozone column is evaluated on a timescale of a few years. In order to represent this short timescale, we evaluated the sensitivity in our model while keeping methane concentrations fixed. The sensitivity obtained using this procedure is about 30% lower than the one including the long-term feedback of CH_4 concentration on its own lifetime.

The above issues are mentioned in the revised manuscript.

"P18043, L21: Why does an underestimation of the methane lifetime lead to an overestimation of atmospheric concentrations."

Thank you for pointing this out. It should have been written 'overestimation' instead of 'underestimation'.

"P18043, L22-23: Why should it be expected that the model is able to represent the temporal evolution following a volcanic eruption if it is not able to represent the temporal evolution of the concentration in the century before? Please explain."

The evolution of CH_4 presented in Fig. 3 of the old manuscript is only based on the changing anthropogenic emissions. It shows the extent to which our model can be used to reproduce CH_4 concentrations. In the real atmosphere, there are regions with low NO_X concentrations, sensitive to NO_X emissions, and regions of high NO_X concentrations, which are less sensitive to NO_X emissions. Because of our representation of the global atmosphere in a single-column model we are not able to reproduce this separation, and we only have one regime. Additionally, we use a fixed CO yield from

ACPD
12, C10207–C10220, 2012

NMVOC. However, this value is known to vary with NMVOC species and with pollution levels. Therefore we expect it to change on a centennial scale. Processes not included here, such as changes in stratospheric ozone, temperatures and possible trends in natural and biomass burning emissions may cause an additional offset. Therefore we are not able to represent well the CH_4 concentrations at the centennial scale. Note, however, that there are no advanced 3D models that are able to reproduce the methane changes since 1890 with "free-running" methane concentrations.

We consider that we have reasonable sensitivities to CH_4 emissions, ozone and temperature by choosing to work with a tropical profile for temperature, water vapour and photolysis rates. The model also represents well the global budgets and lifetimes, except for ozone, where it is more representative of tropical budgets. Therefore we consider that the model is able to represent reasonably well the sensitivity of CH_4 concentrations to the perturbations following the volcanic eruption.

These issues are mentioned in the revised paper.

"P18044, L6: the term 'base simulation' should be introduced already at the end of Sect. 2, see comment above."

This has been corrected in the revised manuscript.

"P18046, L21ff: It is not immediately clear where the statements concerning the changes in OH come from. Where is this shown?" (referring to Fig. 5 - 'We find a positive methane growth rate due to decreased OH for about one year after the eruption')

We find a negative perturbation in CH_4 emissions in response to the eruption, and the temperature effect on the rate is small. Therefore a positive growth rate can only be related to a decrease in OH. This has been clarified in the revised manuscript.

ACPD
12, C10207–C10220, 2012

"P18047, L3: It is not easy to extract this number from Spahni et al.(2011). Could you please explain from which part of the paper you have extracted this number." (referring to the sentence: 'This is similar to the bottom up estimate of about 5 Tg yr^{-1} in Spahni et al. (2011).' - about changes in wetland emissions)

This number is taken from Figure 8a. in Spahni et al. (2011) by visual inspection. Corrected to 'This is similar to the bottom up estimate of about 5 Tg yr^{-1} shown in Spahni et al. (2011) Fig. 8a'

"P18047, L9: The reference (Spahni et al., 2011) refers probably only to the first part of the sentence." (referring to the sentence 'CH_4 emission changes also depend on spatial and temporal changes in soil moisture and precipitation, which have also been observed after the eruption (Spahni et al., 2011).')

The reference also refers to the second part of the sentence. Spahni et al. (2011) show in Figure 8 the soil moisture and precipitation used to force the vegetation model. This data is based on observations (CEP and CRUNCEP).

"P18047, L21-23: In this sentence it is not unambiguous what a lower growth rate is. Hence the argumentation is not completely clear."

Since this is no longer obvious after retuning the model, we removed this statement. ('The growth rate found with our column chemistry model is generally lower than the observed one')

"P18047, L27-29: The statement of 'a general correspondence' is overoptimistic. This should be assessed more critically."

We reformulated to 'we find a similar range of values for the modelled and observed growth rates in the years 1991 to 1996'.

ACPD
12, C10207–C10220, 2012

"P18049, L20: 'remarkable comparison' should be rephrased to be more modest even in view of the simplicity of the model."

Following comments from both referees, the sentence has been removed.

"P18049, L24-26: This statement is certaily true. However the model used in this study is highly simplified and does not fullfil the stated requirements. This should be recalled here in the conclusions." (referring to 'The dominating effects are those through tropospheric photolysis rates, with ozone depletion having the largest effect. This shows the importance of stratospheric-tropospheric couplings, and that a good representation of stratospheric chemistry is needed in order to model accurately methane concentrations.')

Our column chemistry model is coupled to the stratospheric ozone through the radiation model TUV. The simplicity of the model is recalled at the beginning of the next paragraph.

Reply to technical corrections:

"P18038, L12: add a reference for 5 TgN of NOx from lightning."

Changed from 5 to 6.3 TgN yr^{-1} of NO_X from lightning and included a reference to Huijnen et al. (2010).

"P18034, L10 ... observed d13C ..."

Corrected.

"P18039, L10-12: add here again the reference (Guo et al., 2004)"

Corrected.

ACPD

12, C10207–C10220, 2012

"P18039, L13: explain briefly what 'GISS data' are"

We added the sentences 'For the aerosol optical thickness, we use the Goddard Institute for Space Studies (GISS) monthly averaged values (Hansen et al., 2005), based on SAGE II satellite data. For surface temperature we use GISS analysis data (Hansen et al., 2010).'

"P18041, L17 ... possibly due to ... "

Corrected.

"P18043, L6: replace 'this study' by Etheridge et al. (1998)"

Corrected.

"P18048, L4 ... differences ... "

Corrected.

"P18051, L18 ... tropical troposheric ... "

Corrected.

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ACPD

12, C10207–C10220, 2012

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ACPD

12, C10207–C10220, 2012

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Interactive comment on Atmos. Chem. Phys. Discuss., 12, 18029, 2012.

ACPD

12, C10207–C10220, 2012

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