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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

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.

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- 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:

"First, large amounts of particulates with a short atmospheric lifetime were injected into the atmosphere after the eruption, but these are neither included nor mentioned in this study. "

Because of their lifetime of only a few days, direct particle emissions from the eruption are not believed to have had a significant global impact (Guo et al., 2004; Niemeier et al., 2009). We mention them in the revised manuscript.

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"Second, while the authors try to account for changing anthropogenic CH4 emissions, the economic collapse of the Soviet Union in 1992 was not specifically mentioned, despite that it was believed to abruptly affect CH4's budget, especially in the northern hemisphere."

We did mention the possibility that changes in anthropogenic emissions might have had an impact on the methane growth rate in the years following the eruption. Additionally, we performed simulations where we included these changes, as given by the EDGAR4.1 inventory. We found that indeed part of the reduction in methane growth rate between 1991 and 1993 can be attributed to anthropogenic emissions and this was mentioned in the original manuscript. The economic collapse of the Soviet Union in 1992 might have been the reason for these changes in anthropogenic emissions, and indeed this was not mentioned. We rectify this in the revised paper.

"Third, they do not consider if this large modeled result is consistent with observations of other species whose loss is predominantly by OH."

We want to thank the reviewer for making this suggestion. MCF has been used in many studies to determine OH concentrations. In the revised manuscript we include a simulation of MCF concentrations for the period 1988 to 2005. We find a good agreement between measured concentrations and modelled tropospheric means. Since MCF is mainly destroyed by the reaction with OH, this shows that our modeled OH concentration is reasonable.

A validation of our results for the post-Pinatubo period (1991-1995) with MCF is more difficult, and therefore not included in the manuscript. Using MCF inversions, Bousquet et al. (2005) find a 10% decrease in OH between the periods 1991 and 1992, and a subsequent recovery. Prinn et al. (2005) and Krol and Lelieveld (2003) find variations of $\pm$ 10% in OH from one year to another in the same period. We find a 5% increase in OH concentrations in 1993 compared to 1991, which falls in the range of estimates in the aforementioned studies. However, uncertainties associated both with the measure-

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ments and with the emissions of MCF in this period make it difficult to make a reliable comparison on a timescale of 1-2 years.

Some additional insight about OH concentrations can generally be found by looking at CO concentrations, since CO is mainly lost by the reaction with OH. Granier et al. (1996) find decreases in CO concentrations in the period 1990 to 1993 at many stations around the world. Stratospheric ozone loss has been shown to partly explain this decrease (Granier et al., 1996; Yurganov et al., 1999). Using a 3D model, Granier et al. (1996) find a decrease of 3.2% in CO lifetime between 1989 and 1993 due to stratospheric ozone loss. They conclude that a combination of sources and sinks would explain the observed decrease in CO, including the effect of stratospheric ozone change. The estimated lifetime decrease in CO is not inconsistent with our results. However, since the CO lifetime is only two months, CO concentrations show large variability throughout the troposphere. The scarcity of data in the early 1990s makes it difficult to distinguish between the possible scenarios. Therefore a validation using CO concentrations in our one-dimensional approach is not included in the paper. This will be done when performing this experiment in a three-dimensional model.

The paper needs editing to reduce excess words and improve clarity of the discussion.

The discussion sections have been largely restructured.

"Since it is the transient response to the eruption that can be compared with observations, why focus so much on the steady state response? "

This issue has been addressed above in the general remarks.

"Chemical symbols are used for many species, so why not CH4? "

We replaced 'methane' with 'CH_4' throughout the text.

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"What other data are available that could corroborate these results? "

This has been addressed above.

" Figures should be enlarged, and bolder lines and larger fonts used."

We did our best to improve the quality of the figures.

Replies to specific comments:

"P18030,L13: this is vague; what is the effect of stratospheric O3 depletion?"

We replaced 'the overall effect of the eruption on the methane growth rate is dominated by the effect of stratospheric ozone depletion' with 'the overall effect of natural processes after the eruption on the CH_4 growth rate is dominated by the reduction in CH_4 lifetime due to stratospheric ozone depletion'.

P18032,L6: Tg of what?

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

L8: photolysis of what?

'Decrease in tropospheric photolysis' has been replaced by 'decrease in ozone photolysis in the troposphere'.

"P18032,L26: and a shorter lifetime for anything else whose predominant loss process is reaction with OH. "

Since our focus is on the effects on methane concentrations, we do not consider this addition necessary.

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P18033,L17: Soden et al. found...

Corrected.

"P18033,L21: much of the O(1D) produced does not react with water, so I would not say it is immediate. " (referring to 'Less water vapour in the troposphere would imply less OH formation from ozone, because of the intermediate reaction of O(1D) with water.')

OH is only formed when O(^1D) reacts with water vapour, so a decrease in water vapour would lead to less OH formation. We rephrased as: "because the reaction between O(^1D) and water becomes less likely".

"P18034,L8-10: The 13C data responsible for the feature that this claim is based on are not consistent with other data from high southern latitudes. "

The reviewer is right. We added this statement in the revised manuscript.

"P18037,L12: what is AUTO software? "

The simulations of steady-state evolution using the AUTO software have been removed in the revised manuscript. Therefore we also remove the reference and description of this software.

"P18038,L24-25: why distribute SO2 globally rather than only in the tropics, consistent with observations? Were sulfate aerosols produced as SO2 decayed to get the timing correct for their impact on photochemistry? Is the increase in AOD from sulfate aerosols only? Were particulates from the eruption considered? "

We try to simulate a global mean perturbation, that represents both the tropics, where SO_2 was present, as well as the subtropics, where no SO_2 was present. Since we do not represent different latitude bands in our "global" column model, we can only study

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a global perturbation. Currently we are performing a similar study with a 3D CTM.

Observed stratospheric AOD are used in the model. Translating SO_2 decay to AOD would need a sophisticated aerosol model, which can account for aerosol growth. This is beyond the scope of this paper.

Because of their short lifetime, particles emitted are not considered to have had a significant global impact (Guo et al., 2004; Niemeier et al., 2009). They might have had a localised impact on the observed AOD in the first few weeks after the eruption. We use here observed monthly and globally averaged AOD, so we consider the emitted particles to have a small impact on the input data.

"P18039,L26: why use a Q10 at the lowest end of the range found in most field studies. "

The temperature sensitivity of wetland emissions is highly variable between plant species and environmental conditions. For this reason, in global vegetation models, the Q10 factor is computed at every grid cell. Estimating a global mean is very difficult.

We choose here a global Q10 value of 2 because most measurements and inverse modeling results show values at the lower range of the spectrum. Using this value, our estimate of the maximum change in wetland emissions compares well with other bottom-up estimates (Bekki and Pyle, 1994; Spahni et al., 2011).

"P18040,L1: What is the MEGAN-like dependence? "

Since our model overestimates the sensitivity to isoprene emissions, we include isoprene only in the model validation in the revised manuscript, and not any more in the results. We no longer calculate the change in isoprene emissions, but apply a fixed perturbation of 9%. Therefore the description of how they are calculated has been removed.

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"L9:"forcing of the eruption" is vague. "

We replaced it by 'natural forcings after the eruption'.

"L18: The description of Fig 2 is unclear. How are the values plotted in Fig 2 been averaged? Annually and globally? Where is it shown that the model falls within the range of observations? "

The values in Figure 2 have not been averaged. They are the result of the column chemistry model, which does not have spatial or seasonal variation, but which we consider to be representative for annual global mean concentrations.

In the revised manuscript, we have added a comparison of modelled methyl chloroform 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.

"L24: Lightning produces NOx, it does not emit it. Where is it produced? "

We assume that NO_X is produced by lightning throughout the model column. This has been clarified in the revised manuscript.

"L25-26: These statements about O3 are vague and unclear unless you already know where O3 in these regions comes from. "(referring to 'Ozone in the boundary layer is produced from NOx and hydrocarbons, while higher up it is mostly determined by the

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stratospheric boundary condition.')

The sentence has been removed.

"P18041,L4: emissions of O3?"

Replaced 'increases in emissions' by 'increases in emissions of ozone precursors, CO and CH_4'.

"L17: "be due to"? "

Corrected.

"P18042,L1: I suggest summarizing the sensitivities used in this study in a table and comparing them with those determined from other studies. "

We thank the reviewer for the suggestion. A table has been included in the revised manuscript.

"L7: Hasn't the sensitivity of CH4 burden to emissions been assessed since the SAR? " (referring to 'which falls within the range of 1.2-1.6 found in the Second Assessment Report of the IPCC')

The reviewer is correct. In the revised manuscript, we include a comparison of our sensitivity of CH_4 burden to CH_4 emissions with the ones found in IPCC Third Assessment Report (Prather et al., 2001) and in Voulgarakis et al. (2012). These studies give similar estimates to the ones in SAR, finding a best estimate of 1.4 and a range of 1.23 to 1.69, respectively.

"L19: Cape Grim "

Corrected.

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"L27-30: if you believe the sensitivity of atmospheric CH4 to stratospheric O3 is likely too large, why proceed? Is there evidence from other species (e.g., CO or CH3CCl3) that the decrease in CH4 lifetime resulting from stratospheric O3 destruction could be this large? "

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 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

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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,L5: How did you put measurements from Etheridge et al. and globalview on the same scale? "

This was explained in the text. We used the measurements at the stations Alert and South Pole and applied the same procedure as used in Etheridge et al. to calculate the global mean.

"L7: Alert belongs to Environment Canada, not NOAA. "

The data used is from the NOAA Globalview project. So although the station does not belong to NOAA, the data used does. This was clarified in the revised text.

"L21: Using a CH4 lifetime that was too short would increase the sink and should make atmospheric CH4 too low, not too high. "

Thank you for pointing this out. 'Underestimation' should have been 'overestimation'.

"P18045,L7: before comparing growth rates, modeled and observed atmospheric CH4 abundance should be compared in a figure for 1980 or 1990 to present. It is impossible to tell from Fig 3 how well they compare. Also, a more detailed description of how growth rates were calculated for model results and observations is required. It is hard to see how the growth rate in Fig 6b was calculated from the transient curve in Fig 3. "

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

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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 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.

"P18047,L24: The impact of the strong El Nino in 1997 to 1998 is not shown on the figure, so why discuss it? "

The discussion about El Nino in 1997-1998 has been removed in the revised manuscript.

"P18048,L4-5: So you assume that you have all the impacts you've considered exactly right, so any differences between model and observations result from processes not included here or interpolation procedures? "

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We do not claim these are the only reasons for mismatch. However, they do constitute a major source. Additional sources of uncertainty have been mentioned in the next paragraphs, including uncertainties in model parameters, and the fact that we use a one-dimensional model. These paragraphs have been restructured to make the discussion clearer.

"L9-11: how would interpolation procedures explain differences between model and observations? "

Interpolation can smooth out sudden changes in the growth rate, such as the one related to the injection of SO_2. We expect this effect to be overestimated by our model by assuming an instant homogeneity if the plume around the globe. However, the converse is also true, that sudden changes are smoothed out by the interpolation. This is mentioned in the revised paper.

"L12-14: I do not understand "comparing the sum of the "Pinatubo all" and "Anthrop" curves to the growth rate evolution when including both Pinatubo forcings and changes in anthropogenic emissions, we can find the role of second order effects". Isn't the sum of the "Pinatubo all" and "Anthrop" curves the growth rate including both Pinatubo forcings and changes in anthropogenic emissions? "

Including both processes is not the same as adding them up. When we include both processes and allow them to interact, we also account for possible nonlinear effects.

"L28: this uncertainty seems rather small. " (referring to 'We estimate therefore a maximum error of 15% in the modelled growth rate due to the model setup.')

This is strictly related to the parameters in the model, and not to the fact that it is a one-dimensional model. The discussion on uncertainties has been restructured to make it clearer.

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"P18049,L20: in what way is this comparison "remarkable"? "

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

"L21-22: this conclusion was apparent based on previous studies before this study. "

We have included references to such studies.

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