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Interactive comment on "Can we explain the observed methane variability after the Mount Pinatubo eruption?" *by* N. Bândă et al.

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Replies to general comments:

We would like to thank the referee for the positive feedback.

Please note that some small changes were made to Table 1 and Figs. 5 and 6 due to a mistake we found in the implementation of HYMN emissions. The main results and conclusions of the paper are not affected.

"However, some of the background for this work could still be covered more clearly. Patra et al 2014 is being cited in the section on potential sources of uncertainty, and that review paper raised major questions about the extent to which atmospheric chemistry

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models can reproduce the spatial distribution of OH, particularly the ratio of Northern Hemisphere to Southern Hemisphere concentrations. Given this level of structural uncertainty in atmospheric chemistry models, it would be useful to include a short comparison of the TM5 model, as used here, with others used elsewhere."

Figure 6b of the paper shows that our OH is higher in the Northern Hemisphere, as found by other chemical models as well. From methyl-chloroform observations, Patra et al. (2014) find that OH fields with smaller North-South gradient are more realistic. We mention this issue in the Discussion section of the paper as one of the sources of uncertainty in our results.

The OH fields in TM5 are presented in Huijnen et al. (2010), including a validation with methyl chloroform that shows a good performance against observations. A comparison of the OH fields in TM5, EC-Earth and Spivakovsky et al. (2000) is also shown in van Noije et al. (2014). Presenting additional validation and comparisons to other studies is beyond the scope of this paper.

We added in the Method section: "The performance of TM5 in simulating atmospheric chemistry is presented in Huijnen et al. (2010) and van Noije et al. (2014). The conclusions of these studies remain valid with the current setup."

"Also while the Patra et al 2014 paper was focussed on the spatial distribution of OH, the earlier paper by Montzka et al 2011 (Small Interannual Variability of Global Atmospheric Hydroxyl. Science 331, 67-69. 2011) was a review of how different tracers for OH can bring in some constraints on the extent to which this has varied over 1985 - 2007. A brief comparison between that and the results of this paper would be helpful in the conclusion."

We thank the reviewer for this suggestion. The variability we find for the CH_4 sink by reaction with OH of 8 Tg/yr, or 1.6%, in the years 1990 to 1996 agrees well with the conclusion of Montzka et al. (2011) that OH concentrations are quite stable in the atmosphere, with a variability of about 2% over the period 1985 - 2007. Our results do not support the earlier estimates for the global OH variability of 7-9% by Prinn et al. (2001) and Bousquet et al. (2005).

We included in the discussion: "The 1.6% IAV we find for the CH_4 loss by reaction with OH supports the conclusion of Montzka et al. (2011) that OH concentrations are buffered against atmospheric perturbations, having an IAV of about 2%. The large OH inter-annual variations, often exceeding 10%, previously found for the 1990s using methyl chloroform observations are not produced in our chemistry-transport simulations (Prinn et al., 2005; Bousquet et al., 2005)."

Replies to specific comments:

"It would be helpful if the introduction included a reference to the general summary of the CH4 budget given in Table 6.8 of Ciais et al, 2013 (Chapter 6: Carbon and Other Biogeochemical Cycles, Working Group I Contribution to the IPCC Fifth Assessment Report, Climate Change). That is a more recent assessment than Denman et al 2007 and clearly shows the very large discrepancies that still exist between bottom-up and top-down budget analyses, and therefore the relevance of this work."

We cite Kirschke et al. (2013), which is the original source of the table in the IPCC Fifth Assessment Report. The IPCC TAR (Denman et al., 2007) is cited as a reference for the conversion factor between CH_4 emission and CH_4 global atmospheric concentration, which was not re-evaluated in later assessment reports.

We added in the introduction: "with large discrepancies between bottom-up and top-down estimates of CH_4 sources and sinks."

"The long paragraph over lines 58 - 112 would be clearer if it were reorganised into separate paragraphs summarising the previous treatments of sources, then of sinks, and then factors such as cross tropopause transport, rather than merging these together

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as currently done."

As the reviewer suggested, we have restructured the description of processes affecting CH_4 in the early 1990's (previously lines 51-72) into three paragraphs, presenting the effects of the eruption on CH_4 emissions, the effects of the eruption on the CH_4 sink and other effects, respectively. However, many of the studies covered in the literature review that follows (previously lines 73-108) present both source and sink effects. Therefore they are organised in two paragraphs, describing bottom-up process studies and, respectively, attempts to explain the CH_4 growth rate.

"It would also read a bit more clearly if a new paragraph started on what is now line 108 with "In the present . . . " as that would specifically bring out the focus of this paper."

We followed the reviewer's suggestion to start a new paragraph.

"At some point it would be useful for the paper to say whether the approach is to exclude CH4 removal by soil processes, and by reaction with tropospheric chlorine, or to treat these as static over the period 1991 - 1995. Relative variations in these would have to be large to explain much of the atmospheric CH4 variations in this period, however, soils involve a mix of methanogenic and methanotrophic processes and the net flux to the atmosphere can vary across a range of 10 - 40% of what is actually being produced (e.g. Le Mer, J. & Roger, P.; Production, oxidation, emission and consumption of methane by soils: A review. European Journal of Soil Biology 37, 25-50, 2001)."

CH_4 removal by tropospheric chlorine, which accounts for about 5% of CH_4 removal, is not included in our model. The nudging procedure would compensate for mismatches between measured and modelled background CH_4 concentrations because of not including this removal process. However, we do not expect significant year-to-year variations in this CH_4 sink category.

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In all simulations, the CH_4 soil sink from the LPJ inventory was included in the form of monthly two-dimensional uptake rates per ppb CH_4. LPJ provides CH_4 emissions and CH_4 soil sink strengths by taking into account methanothrophic and methanogenic soil processes. As shown in Table 1 of the paper, the variability in the soil sink is less than 2 Tg/yr, and counteracts the variations in CH_4 emissions from wetlands rather than providing additional variability. The variability in the CH_4 soil sink is included together with CH_4 emissions in the difference between 'NoPinS' and 'FixWetl'. As this was not clearly specified in the text, we changed Section 2.2 line 221 from: "In the 'FixWetl1' simulation we used constant CH_4 emissions from wetlands from the year 1990." to "In the 'FixWetl1' simulation we used constant CH_4 emissions from wetlands and CH_4 soil uptake rates from the year 1990."

"Section 2.1 is clearly written and shows why this work is an extension of what has been published previously."

We thank the reviewer for the positive remark.

"The way in which tropopause variability and cross tropopause transport is treated can be very relevant for the period after the Pinatubo eruption. While section 3.1 mentions that a tropopause based on Lawrence et al 2001 is used, it is not clear to what extent that would differ from a more detailed treatment of its seasonal and latitudinal variations such as the NCEP/NCAR reanalysis project (Kalnay et al., 1996 Bull Am Met Soc and the continual updates at ftp://ftp.cdc.noaa.gov/Datasets/ncep.reanalysis.dailyavgs/tropopause)."

Meteorological fields in TM5, including vertical transport airmass fluxes, are used from the ECMWF ERA-Interim reanalysis data. The tropopause definition in the ECMWF model includes detailed treatment of its seasonal and latitudinal variations. The tropopause definition from Lawrence et al. (2001) is a simple latitudinal-dependent pressure level, which gives a tropopause level of 85 hPa at the equator and 300 hPa at

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the poles. This definition is only used here to calculate and plot CH_4 budget terms integrated over the troposphere, and is not used in driving the CH_4 transport between the troposphere and stratosphere. Lawrence et al. (2001) shows that only about 8% of the global CH_4 loss by reaction with OH occurs in the upper troposphere and stratosphere, in the region above 250 hPa in the tropics and above 500 hPa in the extratropics. Therefore, a possible error of the order of tens of hPa in the tropopause level used for our CH_4 budget calculations would have little effect on the results.

We clarified in the main paper line 314: "These differences are integrated up to a tropopause level defined here as [. . .] Lawrence et al. (2001)." instead of "The tropopause level is defined here as [. . .] Lawrence et al. (2001)."

"In section 2.2 it would be useful to know how the altitudes for the Mauna Loa and Niwot Ridge sites are being dealt with as they are outside the lowest 2 km of the atmosphere being used from TM5. For example there is a significant seasonal cycle in the differences between mole fractions for Cape Kumukahi at the surface and Mauna Loa at 3397 m. Similarly no sites are being used over the 0-30° S region and again there are quite different seasonal cycles for the mole fractions at Samoa and Ascension Island. However, these differences are being reflected to some extent in Figure 4 where differences between the GLOBALVIEW analysis and for the Pacific sites is shown."

The five stations used in constructing the latitudinal average CH_4 mixing ratios were chosen such that they are in remote locations, representing background CH_4 concentrations. The latitudinal mean CH_4 constructed by interpolating the monthly mean mixing ratios at these locations, is then compared to the modelled surface dateline mean CH_4. Therefore we do not take into account the station locations or altitude.

We acknowledge that this procedure has some shortcomings. An alternative would have been using the GLOBALVIEW, which has a more complete set of CH_4 stations. However, some of the stations that are included in GLOBALVIEW might have been affected by local emissions. Furthermore, additional processing is applied to gap fill and homogenise station data in GLOBALVIEW. Figure 4 shows that the two observational estimates of CH_4 growth rate agree well, except for the year 1991, where GLOBALVIEW shows an increase in CH_4 growth rate, and then a decrease, while our "Background5" estimate shows a continuous decrease. An inverse modelling system would be needed to make the best use of the measurement data available and to make a more robust estimate of the required adjustment in CH_4 emissions. This would take into account the station locations and altitudes, as well as latitudinal transport beyond 10° latitude bands.

We have added a paragraph at the end of the Discussion section on this: "The latitudinal nudging term needed to correct the mismatch between modelled and observed CH_4 mixing ratios and presented in Sect 3.2 is calculated from measurements at five remote stations. Some uncertainty exists in these terms due to possible observational uncertainty and shortcomings of the nudging procedure. An indication of the observational uncertainty is given in Figure 4, where two observation-based estimates of the global mean CH_4 growth rate variations are shown. The GLOBALVIEW data uses a more complete set of stations, but might contain measurements affected by nearby emissions. Furthermore, additional processing is done to gap-fill and homogenise the station data. The two estimates are in good agreement except for the year 1991, where they differ by about 4 ppb/yr. Some uncertainty also exists in the timing and location of the missing emission variations given by the nudging term. Our nudging procedure is in general able to capture the global growth rate variations. However, because the nudging corrects the amount of CH_4 in the zonal band where the mismatch occurs, this procedure does not account for sub-monthly transport between zonal bands. An inverse modelling setup would be needed to exploit all available measurements to better resolve the sources of mismatch."

"Section 3.1 gives a good summary but the paragraph from lines 375 to 406 could be clearer if it were broken into two that covered what was similar between ORCHIDEE and LPJ, and then what was different."

We followed the reviewer's suggestion to split this paragraph in two.

"Line 383 has a repetition of words."

We removed the first instance of "the growth rate" on line 383.

"Figures 6a and 6b are not referred to in the text."

A reference to these figures was meant on line 448. This typo was corrected in the revised manuscript.

"The summary in Section 4 is well organised. The conclusion in Section 5 is clear but as noted earlier it would be useful to make some comments on the differences between this analysis and that of Montzka et al, 2011."

We thank the reviewer for the positive remarks. See the above replies to general comments.

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