

## ***Interactive comment on “Model simulations of atmospheric methane and their evaluation using AGAGE/NOAA surface- and IAGOS-CARIBIC aircraft observations, 1997–2014” by Peter H. Zimmermann et al.***

**Anonymous Referee #1**

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### General comments

The discussion paper of Zimmermann et al. presents an analysis of the global budget and trends of atmospheric CH<sub>4</sub> for the period 1997-2014, using the EMAC atmospheric chemistry general circulation model. As such the study contributes to the highly controversial discussion on the drivers of the renewed increase of atmospheric CH<sub>4</sub> observed since 2007, and is well within the scope of ACP. However, there are several significant limitations of the study, which limit the conclusions that can be drawn from the presented results.

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(1) The study uses only a very limited number of atmospheric stations. In fact, only one single NOAA station (MLO) has been used (in addition to the 5 AGAGE stations). These 5+1 stations cover only the latitude range between 53°N and 41°S. It is not clear, why the authors do not use any data from the comprehensive NOAA ESRL global cooperative air sampling network (nor from the second NOAA station with continuous CH<sub>4</sub> measurements at Barrow, Alaska). The very limited set of stations used in this study limits the information that can be obtained on the CH<sub>4</sub> emissions at continental scale.

(2) 4 of the 6 stations used in this study are coastal sites (MHD, THD, RPB, CGO). Using such data requires that the model can properly simulate synoptic scale variability (e.g. change between marine and continental air masses). The EMAC model, however, is a general circulation model, and - as described in the paper - nudged to ECMWF meteorology only in the free troposphere (apart from surface pressure). Therefore, the capability of the EMAC model to simulate synoptic variability is probably worse compared to offline atmospheric transport models which are directly driven by analyzed meteorological fields. Good model representation of the continental stations, however, is essential for the study, since the interhemispheric gradient is derived as "as the difference between average CH<sub>4</sub> mixing-ratios at the northern stations MHD (53°N) and THD (41°N) and the southern station CGO (41°S)" (lines 302-304) - and the interhemispheric gradient derived in this way is used to optimize the contribution from the "tropical wetland source (SWA)" and "landfill-, coal-, gas-, and oil (FOS)" emissions. Related to the concern of the potential limitations of the EMAC model to simulate the synoptic variability is the fact that the study uses "Monthly mean mixing - unfiltered with respect to local pollution events" (lines 205-206) measurements, which are compared to monthly mean model output. Especially for the 4 coastal sites, it would be more appropriate to use hourly (or daily) observations. If the EMAC model cannot properly simulate these sites, the use of monthly mean values for the comparison is likely to result in biased results.

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(3) Unfortunately, the study investigates only 2 scenarios to analyze the recent CH<sub>4</sub> trend: (1) scenario "TRO" with additional emissions from the tropical wetlands, and (2) scenario "SHA" with additional emissions from the North American shale gas drilling sites. However, further hypotheses have been proposed in the literature, including increasing CH<sub>4</sub> emissions from agriculture and waste sectors [Saunois et al., 2017; Schaefer et al., 2016], and decreasing CH<sub>4</sub> emissions from biomass burning [Saunois et al., 2017; Worden et al., 2017]. While the decreasing  $\delta^{13}\text{CH}_4$  observed in the atmosphere points to an increasing microbial sources (including both wetlands and anthropogenic microbial sources), Saunois et al. [2017] and Schaefer et al. [2016] concluded that among the microbial sources agriculture and waste sectors are more important than natural wetlands. This hypothesis is also supported by statistical data which suggest a significant increase of global CH<sub>4</sub> emission from enteric fermentation and manure by 10 Tg CH<sub>4</sub> yr<sup>-1</sup> between 2000 and 2011 ([Saunois et al., 2017], Fig. S12). The magnitude of the estimated decrease in biomass burning is smaller (estimated to be 3.7 ( $\pm 1.4$ ) Tg CH<sub>4</sub> per year from the 2001–2007 to the 2008–2014 period [Worden et al., 2017]), but plays an essential role for the  $\delta^{13}\text{CH}_4$  budget and to reconcile the different hypotheses about the recent CH<sub>4</sub> increase.

Based on these general comments, I recommend to thoroughly revise the study, analyzing in more detail the capability of EMAC to simulate synoptic scale variability, to use a more comprehensive set of surface observations, and to include additional scenarios (in particular including the increase of CH<sub>4</sub> emissions from agricultural sources).

Further specific comments:

Abstract, line 21: I would suggest to replace "atmospheric CH<sub>4</sub> calculations" by "atmospheric CH<sub>4</sub> concentrations" or "atmospheric CH<sub>4</sub> dry air mole fractions"

Abstract, line 24: "rescaling of individual emissions with proportional effects on the corresponding inventories": it is not clear what is meant here with "inventories" as compared to the "emissions".

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Abstract, line 27: "all-station mean dry air mole fraction of 1792 nmol/mol": reference time period should be given (is this the 2000-2005 period mentioned in the following sentence, or the 1997-2006 period mentioned earlier?).

Abstract, line 38: "The coefficient of determination of R<sup>2</sup> = 0.91 indicates even higher significance than before 2006": This could be partly due a larger range of concentrations values (and the given RMS is slightly higher than before 2006, indicating rather slightly poorer agreement).

Abstract, line 40-41: "...indicating that the model reproduces the seasonal and synoptic variability of CH<sub>4</sub> in the upper troposphere and lower stratosphere." The analysis in the paper shows also clear limitations to simulate the variability in the lower stratosphere. This should be mentioned also in the abstract.

Introduction, lines 45-46: "and its concentration has been growing by about 1%/y since the beginning of the Anthropocene in the 19th century (Crutzen, 2002)": I would suggest to add further references for the atmospheric CH<sub>4</sub> increase.

Introduction, lines 47-50: I would propose to present here mainly the most recent estimates of the radiative forcing. If the authors want to include also the older estimates, they should briefly explain the reasons for the large differences in the estimates. Furthermore, the given values "0.57 Wm<sup>-2</sup> (direct 0.44Wm<sup>-2</sup>, indirect 0.13Wm<sup>-2</sup>)" are not consistent with the given [Dlugokencky et al., 2011] reference (where higher values are reported).

Introduction, lines 52-53: "...in 2007 the CH<sub>4</sub> increase resumed unexpectedly (Bergamaschi et al., 2013)": Include here the primary references reporting the CH<sub>4</sub> increase from the measurements ([Dlugokencky et al., 2009; Rigby et al., 2008]).

Introduction, lines 72-73: " Schaefer et al. (2016)... raising concern about the contribution from rice production versus wetland emissions". It should be mentioned here that Schaefer et al. (2016) conclude that the increase could be largely explained by

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increase of CH<sub>4</sub> emissions from ruminants (see also my general comment (3)): " Inventories report increased annual agricultural emissions over the 2000-2006 average of 12 Tg by 2011; dominated by ruminants (21, 23). This can largely account for the post-2006 [CH<sub>4</sub>]-growth, estimated at 15-22 Tg/a (30). Also, India and China's dominance in livestock-emissions (23) and S.E. Asian rice cultivation are consistent with the location of the source increase (13)."

Introduction, line 77: "Further, it was concluded that fossil fuel related sources had decreased". It should be stated explicitly who concluded this (it is not clear if this refers only to the [Schwietzke et al., 2016] or to both papers discussed here).

Model Setup, lines 117-118: "...operational analysis data of the European Centre for Medium-range Weather Forecasting (ECMWF) (van Aalst et al., 2004).": Why did the authors use operational analysis data and not the reanalysis (which should be superior in terms of consistency over time, which is essential for any trend analysis)?

Model Setup, lines 119-122: " the nudging method is applied in the free troposphere, tapering off towards the surface and tropopause, so that stratospheric dynamics are calculated freely, and possible inconsistencies between the boundary layer representations of the ECMWF and ECHAM models are avoided.": This might be an advantage in terms of self-consistency of the model physics, but may lead to deficiencies to simulate the synoptic-scale variability also in the boundary layer. As outlined in my general comment (2), the capability to simulate the synoptic-scale variations observed at the surface stations needs to be further analyzed (as this is essential to properly simulate the coastal stations used in this study).

Model Setup, line 127: "photolysis": Is this relevant in the EMAC model domain ?

Model Setup, lines 146-147: "Natural wetland emissions are based on Walter et al. (2000) and Fung et al. (1991).": These are different wetland inventories - which one has been used in this study ? Furthermore, the Walter et al. (2000) reference is missing.

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Model Setup, lines 153: "GFED statistics": The specific GFED version number should be mentioned.

Model Setup, lines 154: "EDGAR2.0 database (Olivier, 2001)": Why has this old version of the EDGAR database been used, and not more recent versions?

Model Setup, lines 161: "yearly differences in the 20 Tg/y biomass burning": I would suggest to replace "yearly" by e.g. "inter-annual".

Model Setup, lines 179-180, "The negative flux distribution has a pronounced seasonal cycle in phase with the emissions": which emissions are meant here?

Observations used for model verification, line 190: Maybe replace "verification" by "validation" (however, there is indeed not a consistent use of these terms in the scientific literature)

Observations used for model verification, lines 199-205: The calibration scales used should be mentioned, including potential differences between the NOAA and AGAGE scales.

Observations used for model verification, lines 199-205: Why has only this very limited set of atmospheric stations been used (see general comment (1))?

Observations used for model verification, lines 205-207: "Monthly mean mixing - unfiltered with respect to local pollution events - are compared to respective monthly averaged model samples...": Why did the authors use monthly mean values, and not hourly or daily averages (see general comment (2)) ?

Observations used for model verification, lines 209ff: which calibration scale has been used for the CARIBIC CH<sub>4</sub> measurements?

Simulation results, lines 229-230, "spin-up simulations and scaled to match the 1997 station measurements", and lines 254-255 " For initialization, a global methane distribution pattern for January was created iteratively in several spin-up cycles and finally

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rescaled to Jan. 1997 station measurement data": The spin-up and scaling should be described in more detail (but best in section with model description): how long is the spin-up, which emissions have been used (probably the same as for the period 1997-2006?)? Did you just scale the calculated 3D fields? If so, there would be some inconsistency between the applied emissions and the concentrations (which may also explain why the simulated CH<sub>4</sub> concentrations still increase between 1997 and 2000).

Simulation results, lines 265-269: "The linear dependency between source strength and atmospheric abundance...", and lines 286-289: "The integrated model CH<sub>4</sub> masses exactly match the mass calculated": this has already been discussed before.

Simulation results, line 310: "... fossil group of categories comprising landfill-, coal-, gas-, and oil (FOS)": CH<sub>4</sub> from landfills are (usually) not fossil, but primarily from relatively recent carbon.

Simulation results, line 346: "In contrast to the monthly average station data, the CARIBIC individual methane observations...": The station data - as provided to users - are hourly data. See also general comment (2).

Simulation results, lines 365-366: "...suggesting that the vertical resolution of the model grid is not optimal to resolve the fine structure in the tropopause region.": Probably this is not only due to coarse vertical resolution, but also due the vertical CH<sub>4</sub> gradient in the stratosphere.

Simulation results, line 369: "according to the definition in Sect. 3.2 (Fig. S3)..." I assume this should be Fig. S2?

Simulation results, lines 386-387: "Figs. 12a and b...logarithmically scaled": the figures seem so use a linear scale. Furthermore, the figures show concentrations, while the figure caption states "Assumed additional emissions..." (Should be rephrased to e.g. "Impact of assumed additional emissions...").

Simulation results, lines 391-392: "...upper estimate from Bergamaschi et al. (2013) of

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22 Tg CH<sub>4</sub> yr<sup>-1</sup> as a first guess": It should be mentioned that the estimate of Bergamaschi et al. (2013) is for a different time period (2007-2010), compared to the 2007-2014 period used in this study.

Simulation results, lines 395-396: "Both scenarios perfectly reproduce the observed CH<sub>4</sub> trend...": I would suggest to avoid the term "perfectly".

Simulation results, lines 400-401: "Changes in the removal rate of methane by the OH radical have not been seen in other tracers of atmospheric chemistry, e.g. methyl chloroform (CH<sub>3</sub>CCl<sub>3</sub>) (Montzka et al., 2011; Lelieveld et al. 2016) and do not appear to explain short-term variations in methane.": I do not agree with this statement. Although Montzka et al. [2011] derive only small interannual variability their CH<sub>3</sub>CCl<sub>3</sub> based estimates still show variations on the order of +/- 3%, which is equivalent to a variability of the OH sink of +/- 17 Tg CH<sub>4</sub> yr<sup>-1</sup>. Furthermore, the recent papers of Rigby et al. [2017] and Turner et al. [2017] demonstrated the potential significant impact of variations in OH on the trend and interannual variability of CH<sub>4</sub>. I suggest to include the references to the two papers.

Simulation results, lines 423-424: "This shows that, when the SHA emissions are located away from the North America, no fraction is found that could minimize simultaneously the  $\Delta$ NS and RMS": Given the very limited number of stations (see general comment (1)) and the question how well coastal / regional stations are simulated by the EMAC model (see general comment (2)), the question is, if this finding is really significant / robust.

Simulation results, line 437: "linear trend lines 0.32x (CARIBIC) and 0.31x (EMAC)": units are missing

Simulation results, lines 451-452: "the tropopause influence is stronger": probably also the influence of the lower stratosphere.

Conclusions and Outlook, lines 488-496: Would be useful to expand the conclusions,

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including a discussion / summary of the novel aspects of this study, the uncertainties of the results and limitations of the study. Furthermore, it should be summarized, how the results from this study compare with the existing literature studies.

Conclusions and Outlook, lines 497-499: "In view of the additional global CH<sub>4</sub> source since 2007, a source – sink equilibrium has not yet established after the 8 years of emissions considered. A 2nd order polynomial extrapolation predicts steady state after 13 years, assuming that the emissions remain unchanged.": This scenario seems quite hypothetical, and global emissions (including their latitudinal distribution) remaining constant over 13 years relatively unlikely.

Conclusions and Outlook, lines 497-499: "Nevertheless, the degree of freedom in the choice of sources is limited,...": Taking into account also uncertainties in the spatial (and temporal) distribution of emissions, a very large number of emission scenarios is possible - while only 2 scenarios were investigated in this study (see general comment (3)).

Table 1: references should be given for the individual a priori emission estimates.

Figures - general comment: The number of figures seems very large - several of them could be put in the supplementary material.

Figure 1: How were the data fitted?

Figure 3: Explain the meaning of the individual red circles

Figure 4a: "Zero" point (a priori emissions) should be indicated

Figure 4b: What is the meaning of the curves (interpolation) between the individual stations?

Figure 5: "dashed line": The figures seem not to show any dashed line.

Figure 7: What is the meaning of the colors?

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Figure 11b: Is the shown average for all CARIBC flights as function of time really very useful? Probably the spatial coverage of the flights is also changing significantly over time.

Figure 13b: What is the meaning of the curves (interpolation) between the individual stations?

Figure 16: (as for Figure 11b): How is the spatial coverage of the flights changing over time?

Figure 17: Legend needs to be explained. Which curves are for which period?

#### References

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