

We thank the referee for the comments.

Here the comments are listed (black) with our reply (red, italics).

General comments

The discussion paper of Zimmermann et al. presents an analysis of the global budget and trends of atmospheric CH₄ 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 CH₄ 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.

Following the referee's advice, most points of criticism have been taken into account and new simulation runs have been performed:

(1) The study uses only a very limited number of atmospheric stations.

Indeed only a limited amount of stations were used in the comparison. Following the referee's advice, the simulation was repeated using 16 NOAA stations and CGO (AGAGE):

Code	Station Name	Country	Lat °	Lon °	elevation / m
ALT	Alert	Canada	82.45	-62.51	190
ASC	Ascension Island	UK	-7.97	-14.40	85
AZR	Terceira Ile., Azores	Portugal	38.77	-27.38	19
BRW	Barrow, Alaska	USA	71.32	-156.61	11
CGO	Cape Grim, Tasmania	Australia	-40.68	144.69	94
CRZ	Crozet Island	France	-46.43	51.85	197
EIC	Easter Island	Chile	-27.16	-109.43	47
GMI	Mariana Islands	Guam	13.39	144.66	0
HBA	Halley Station,	Antarctica, UK	-75.61	-26.21	30
MLO	Mauna Loa, Hawaii	USA	19.54	-155.58	3397
RPB	Ragged Point	Barbados	13.17	-59.43	15
SEY	Mahe Island,	Seychelles	-4.68	55.53	2
SHM	Shemya Island, Alaska	USA	52.71	174.13	23
SMO	Tutuila, Am. Samoa	USA	-14.25	-170.56	42
SPO	South Pole	USA	-89.98	-24.80	2810
ZEP	Ny-Alesund, Svalbard	Norway, Sweden	78.91	11.89	474

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 53oN and 41oS. 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 CH₄ measurements at Barrow, Alaska). The very limited set of stations used in this study limits the information that can be obtained on the CH₄ 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).

We thank the referee for pointing this out.

In the new simulation all stations are “clean air” sites and filtered wrt synoptic scale pollution (except CGO).

We also revised the text in Ch. 3.1:

“The data provided (Dlugokencky et al., 2018) are filtered with respect to synoptic scale pollution events. We take advantage of 16 stations about fairly equally distributed over the globe (Fig. 2a) and remote from the major emission areas to ensure comparability with the model results which are not filtered. For the same reason, in case of Cape Grim, Australia (41° S, 145°) we refer to the unfiltered AGAGE records (Prinn et al., 1978, 2013). At all stations monthly mean mixing-ratios are compared to respective monthly averaged model samples. “

Our actual model resolution “T106” with a grid size of 125 km at the Equator is able to capture also synoptic scale events; however the fifteen NOAA stations that we used are remote from the main source regions. CGO is influenced by air from the Australian continent but the model is able to reproduce eventual pollution events.

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.

Indeed, we operate the EMAC GCM/CCM in "nudged" mode, i.e. by Newtonian relaxation towards ECMWF meteorology. The nudging (of divergence, vorticity, temperature (excluding global mean) and logarithm of surface pressure) is applied in spectral space, however (by so-called low normal mode insertion) only down to the synoptic scale. Thus, the meteorological sequence of ECMWF is reproduced by EMAC on the synoptic scale, whereas the variability on sub-synoptic scale is determined by the model physics (e.g. convection etc.).

Thus, the synoptic variability should not be an issue here.

Good model representation of the continental stations, however, is essential for the study, since the is derived as "as the difference between average CH₄ mixing-ratios at the northern stations MHD (53oN) and THD (41oN) and the southern station CGO (41oS)" (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.

Following the referee's advice, in the new simulation we are using an improved interhemispheric gradient definition and define:

$$\Delta NH/SH = \text{avg}(ALT, ZEP, BRW) \text{ minus } \text{avg}(CGO, CRZ, HBA, SPO).$$

Furthermore the contributions of all (10) emission categories are now explicitly considered in the optimization procedure.

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.

As mentioned above, the sites have changed in the new simulation.

Consistently with the measurements, the model now samples at every time step and averages at the end of every month (sub-model SCOUT).

(3) Unfortunately, the study investigates only 2 scenarios to analyze the recent CH₄ 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 CH₄ emissions from agriculture and waste sectors [Saunois et al., 2017; Schaefer et al., 2016], and decreasing CH₄ emissions from biomass burning [Saunois et al., 2017; Worden et al., 2017].

We do not discriminate the latter sectors in this study. It has been our intension to test the sensitivity of the station- and flight records to the hypothetical TRO and SHA emission assumptions.

Biomass burning emissions are not considered in the trend phase.

We take advantage of GFED4.1s which includes agricultural waste burning. No persistent decrease is obvious in this dataset (Fig. 1 at the end of this document.)

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.

It is not possible to resolve the latter sectors in the current model setup. The referee's advice will be subject to further investigation also considering observations beyond 2016.

This hypothesis is also supported by statistical data which suggest a significant increase of global CH₄ emission from enteric fermentation and manure by 10 Tg CH₄ yr⁻¹ 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 (± 1.4) Tg CH₄ 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 CH₄ 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,

Following the referee's advice we revised the study and performed new simulations.

and to include additional scenarios (in particular including the increase of CH₄ emissions from agricultural sources).

It has been our aim from the beginning to test how well the taken assumptions (in form of given emission datasets of 10 categories) can explain the observations. Realizing a significant underestimation from 2007 on, the two hypothetical scenarios TRO and SHA have been included for sensitivity testing of the observed trend.

Further specific comments:

Abstract, line 21: I would suggest to replace "atmospheric CH₄ calculations" by "atmospheric CH₄ concentrations" or "atmospheric CH₄ dry air mole fractions"

Ok: "The atmospheric CH₄ 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".

New formulation: "... rescaling of individual emissions with proportional effects on respective source segregated methane abundances."

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?).

New formulation: ". . . the 2000 – 2005 observed all-station mean dry air mole fraction of 1780 nmol/mol could be reproduced within an RMS = 0.40 % . . ."

Abstract, line 38: "The coefficient of determination of $R^2 = 0.91$ 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).

In the new simulation including 16 ground stations we had to update the statistics:

"We explored the contributions of two potential causes, one representing natural emissions from wetlands in the tropics "TRO", and the other anthropogenic (e.g. shale gas fracturing) emissions in North America "SHA". Based on the acceptance of the no-trend period emission distribution, with the Solver we estimated annual 19.4 TRO and 5.7 Tg/y SHA contributions, respectively, to optimally fit the trend (RMS = 0.55 % / $R^2 = 0.88$)."

Abstract, line 40-41: "...indicating that the model reproduces the seasonal and synoptic variability of CH₄ 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.

Following the referee's advice we included the following statement:

"The coefficient of determination R^2 implies that the model reproduces the seasonal and synoptic variability of CH₄ in the UTLS. Regression analysis however indicates evident underestimation of the calculated CH₄ variability, suggesting that the vertical resolution of the model grid is not optimal to resolve the fine structure in the tropopause region."

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 CH₄ increase.

Following the referee's advice we included: "Clais et al. 2013"

Reference: Ciais, P., C. Sabine, G. Bala, L. Bopp, V. Brovkin, J. Canadell, A. Chhabra, R. DeFries, J. Galloway, M. Heimann, C. Jones, C. Le Quéré, R.B. Myneni, S. Piao and P. Thornton, 2013: Carbon and Other Biogeochemical Cycles. In: Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change [Stocker, T.F., D. Qin, G.-K. Plattner, M. Tignor, S.K. Allen, J. Boschung, A. Nauels, Y. Xia, V. Bex and P.M. Midgley (eds.)]. Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA.

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⁻² (direct 0.44Wm⁻², indirect 0.13Wm⁻²)" are not consistent with the given [Dlugokencky et al., 2011] reference (where higher values are reported).

Following the referee's advice we included:

"The resulting factor of 2.5 increase in the global abundance of atmospheric methane (CH₄) since 1750 contributes 0.5 Wm⁻² to total direct radiative forcing by long-lived greenhouse gases (2.77 Wm⁻² in 2009), while its role in atmospheric chemistry adds another approximately 0.2 Wm⁻² of indirect forcing (Dlugokencky et al., 2011) Etminan et al, (2016) presented new calculations including the impact of the shortwave forcing and found that the 1750-2011 radiative forcing is about 25% higher (increasing from 0.48Wm⁻² to 0.61Wm⁻²) compared to the value in the Intergovernmental Panel on Climate Change (IPCC) 2013 assessment."

Reference: Etminan, M., G. Myhre, E. J. Highwood, and K. P. Shine (2016), Radiative forcing of carbon dioxide, methane, and nitrous oxide: A significant revision of the methane radiative forcing, Geophys. Res. Lett., 43, 12,614 – 12,623, doi:10.1002/2016GL071930.

Introduction, lines 52-53: "...in 2007 the CH₄ increase resumed unexpectedly (Bergamaschi et al., 2013)": Include here the primary references reporting the CH₄ increase from the measurements ([Dlugokencky et al., 2009; Rigby et al., 2008]).

Following the referee's advice we included these references:

"The resuming upward trend after 2007 (Dlugokencky et al., 2009; Rigby et al., 2008, IPCC 2014) is not fully understood: data analysis (Nisbet et al., 2016, Worden et al., 2017) and inverse modelling studies (Bergamaschi et al., 2013) indicate..."

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 increase of CH₄ 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 [CH₄]-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)."

We inserted in the paragraph before:

“Schaefer et al. (2016) by means of $^{13}\text{C}/^{12}\text{C}$ (CH_4) data and a box model concluded that fossil fuel related emissions are a minor contributor to the renewed methane increase, compared to agricultural emissions dominated by ruminants.”

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).

We updated this paragraph:

“As mentioned above, Schaefer et al. (2016) showed that “after 2006, the activation of biogenic emissions caused the renewed CH_4 rise”, raising concern about the contribution from rice production versus wetland emissions, and Schwietzke et al. (2016), based on reassessment of data of the $^{13}\text{C}/^{12}\text{C}$ ratio of CH_4 from fossil sources, conclude that the assumed global fossil fuel CH_4 emissions need a major upward revision of 60-110 %. In other words, it was found by both authors that the combined fossil CH_4 sources (1985-2002) must have been much stronger (factor of 2), at the expense of microbial sources. Further, it was concluded that fossil fuel related sources had decreased.”

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)?

Following the referee's advice, we use the ERA-Interim coefficients in the new simulation.

Reference:

Dee, D. P., et al.: The ERA-Interim reanalysis: configuration and performance of the data assimilation system, Q. J. Roy. Meteor. Soc., 137, 553–597, doi:10.1002/qj.828, 2011.

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).

see above - and no coastal stations anymore except CGO which is simulated at $R^2 = .93$ (Fig. 1 at the end of this document)

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

Because the model domain reaches up to 1 Pa we mentioned that for completeness, even the effect is small. “Removal of CH_4 by photolysis becomes important only in the mesosphere.”

(T. Röckmann et al., The isotopic composition of methane in the stratosphere:

*high-altitude balloon sample measurements, Atmos. Chem. Phys., 11, 13287–13304, 2011
doi:10.5194/acp-11-13287-2011)*

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.

We updated this paragraph:

“Natural wetland emissions are based on Walter et al. (2000), fossil sources based on EDGARV2.0 and remaining sources as compiled by I. Fung et al. (1991).”

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

The new simulation is based on GFEDv4s.

Reference: Randerson, J. T., G.R. van der Werf, L. Giglio, G.J. Collatz, and P.S. Kasibhatla. 2018. Global Fire Emissions Database, Version 4, (GFEDv4). ORNL DAAC, Oak Ridge, Tennessee, USA. <https://doi.org/10.3334/ORNLDAAC/1293>

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?

We did not update this dataset because this category contributes just 2.6 % of total CH₄ and because the dataset serves as a priory assumption for the optimization procedure.

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

We follow the referee's suggestion.

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

In the new simulation the “negative flux” approach is replaced by a “deposition velocity” parametrization which was not yet ready for the old paper version.

We updated the respective paragraph:

“The MESSy sub-model “DDEP” simulates dry deposition of gas phase tracers and aerosols (Kerkweg et al. 2006). For our CH₄ budget modeling the deposition velocity was derived for a fixed atmospheric-methane mixing ratio of 1800 nmole/mole (Spahni R. et al., 2011, Ridgwell et al., 1999) and is scaled correspondingly. The deposition has a pronounced seasonal cycle in phase with the wetland emissions and depends on soil temperature, moisture content and the land cultivation fraction and varies from 2.4 Tg in January to 4.0 Tg in July.”

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)

We follow the referee's suggestion.

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

NOAA standard scale (Dlugokencky et al., 2005)

ALE/GAGE calibration procedure (Prinn et al., 2000)

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

As mentioned above, now 16 NOAA stations

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)) ?

As mentioned above, consistently with the measurements, the model now samples at every time step and averages at the end of every month (sub-model SCOUT).

In consideration of the 20 year's simulation period and the >8 years lifetime of methane are comparing monthly means.

Observations used for model verification, lines 209ff: which calibration scale has been used for the CARIBIC CH₄ measurements?

We inserted:

"The calibration is carried out using NOAA Methane WMO scale (Dlugokencky et al., 2005) For further information . . ."



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 and finally 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?

We introduced in

Ch. 2, Model Setup:

"Using a priori emission estimates, an initial CH₄ distribution was derived in the course of several spin-up simulations repeated until a steady state global CH₄ mass has settled over the years 1997 through 2006."

Ch. 2.2.1 Methane emissions:

"The GFED biomass burning statistics include agricultural waste burning events. Biomass burning emissions are inter-annually variable and the 1997 emission was 2.4 times as high as the 1998-2015 average (Fig. S1c)."

4.1 The period 1997 through 2006

“For initialization, a global methane distribution pattern for January was created as mentioned above and ensures a balanced annual average global CH₄ mass over the entire period with inter-annually constant sources and sinks up to deviations caused by variations in biomass burning.”

Ch. 4.1.1 NOAA/AGAGE stations

“The initial distribution, which is the result of a long term simulation, does not precisely reflect the special Jan. 1997 situation, but obviously overestimated starting values at northern hemispheric stations level down in the course of the first six months. The Solver cannot improve this because it acts on the whole biomass burning series and not on individual years.”

If so, there would be some inconsistency between the applied emissions and the concentrations (which may also explain why the simulated CH₄ concentrations still increase between 1997 and 2000).

Figs. 1 and 5 in the new paper version (cf. Fig. 1 for CGO at the end of this document) reveal an increase also in the between 1997 and 2000. NH-enhanced values level down within six months and are caused by the anomaly in GFED biomass burning.

Simulation results, lines 265-269: "The linear dependency between source strength and atmospheric abundance...", and lines 286-289: "The integrated model CH₄ masses exactly match the mass calculated": this has already been discussed before.

New formulation:

“The linear dependency between source strength and atmospheric abundance is reflected in the model’s partial differential equation system and allows the redistribution . . .”

Simulation results, line 310: "... fossil group of categories comprising landfill-, coal-, gas-, and oil (FOS)": CH₄ from landfills are (usually) not fossil, but primarily from relatively recent carbon.

In the new simulation that is no longer an issue: all categories are considered individually.

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).

The usage of the sub-model SCOUT, consistently with the measurements, samples at every time step and averages at the end of every month.

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 CH₄ gradient in the stratosphere.

The comment of the referee is not in contradiction to our conclusion: The tropopause region with the declining vertical CH₄ gradient cannot be properly resolved due to the coarse vertical resolution (~500m) of the hybrid model grid at this altitude.

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

According to the referees comment, in the updated version, Fig. S3 now is correctly referred as Fig. 2b

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...").

According to the referees comment Fig. 12 is now Fig. S2 in the updated version and log scaled. We rephrased to "Impact of assumed additional emissions . . ."

Simulation results, lines 391-392: "...upper estimate from Bergamaschi et al. (2013) of 22 Tg CH₄ yr⁻¹ 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.

*Following the referee's advice we avoid a citation because the 28 Tg CH₄/y in the new version this has to be considered just as an upper limit for the Solver:
"We used an upper limit emission of 28 Tg CH₄/yr to be added in order to fit the upward trend."*

Simulation results, lines 395-396: "Both scenarios perfectly reproduce the observed CH₄ trend...": I would suggest to avoid the term "perfectly".

We avoided 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 (CH₃CCl₃) (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 CH₃CCl₃ based estimates still show variations on the order of +/- 3%, which is equivalent to a variability of the OH sink of +/- 17 Tg CH₄ yr⁻¹. Furthermore, the recent papers of and Turner et al. [2017] demonstrated the potential significant impact of variations in OH on the trend and inter-annual variability of CH₄. I suggest to include the references to the two papers.

Following the referee's advice we refer to Turner et al. (2017) in the new text. :

"Turner et al. (2017) based on numerical analyses find that a combination of decreasing methane emissions overlaid by a simultaneous reduction in OH concentration (the primary sink) could have caused the renewed growth in atmospheric methane. However they cannot exclude rising methane emissions under time invariant OH concentrations as a consistent solution to fit the (rising) observations."

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 $_NS$ 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.

The new simulation now considers 16 stations– and optimization procedure upgraded

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

New formulation:

"The slopes of the linear trend lines $0.32x$ (CARIBIC) and $0.31x$ (EMAC) over time where $x =$ number of months . . ."

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

New formulation:

"the influence of the lower stratosphere is stronger, leading to reduced linear slopes together with comparably less R^2 of 0.59 and 0.72 (Fig. 14). "

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

A co-author is taking care for this and his findings will be incorporated in the conclusions as soon as possible.

Conclusions and Outlook, lines 497-499: "In view of the additional global CH_4 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.

Following the referee's advice we included in

4.2 Simulating the recent methane trend - first paragraph:

"Between 2007 and 2013 the slope appears nearly linear (Fig. 1), and the discrepancy can be removed by assuming an additional constant CH_4 source for this period. We find that after 2013 an additional increment is necessary to fit the trend."

and in

5 Conclusions and Outlook:

"A 2nd order polynomial extrapolation predicts steady state after 13 years, assuming that the emissions remain unchanged, which does not seem realistic in view of the observed development after 2013/14 (Fig. 1). "

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 gen. com. (3)).

Following the referees comment we formulated:

“Two possible additional methane sources, shale gas extraction (SHA) and tropical wetlands (TRO) have been investigated, that could cause the resuming methane growth since 2007. We showed that a methane increase of 25.47 Tg/y in 2007 and subsequent years, of which 69 % from TRO and 20 % from SHA, can optimally explain the recent CH₄ trend until 2013.”

As mentioned above, realizing a significant underestimation from 2007 on, just the two hypothetical scenarios TRO and SHA have been included for sensitivity testing of the observed trend.

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

The methane emissions are based on (Houweling et al. 2006)

The “burning”-part of the GAMEs dataset is replaced by the GFEDv4s statistics (Randerson et al., 2018) in addition to biofuel combustion emissions from the EDGAR2.0 database (Olivier, 2001).

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

Following the referees comment we reduced the number of figures.

Figure 1: How were the data fitted?

The data have been fit by linear regression – now formula inserted for better interpretation.

Figure 3: Explain the meaning of the individual red circles

Figure no longer used

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

Figure no longer used because of non-linear Solver approximation in the new version.

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

In the new version “Figure 3 at the end of this document” includes 16 stations and interpolating lines left out.

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

"dashed line": Text updated.

Figure 7: What is the meaning of the colors?

now “Figure S6” no more color shift

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.

It turned out to be useful, because in the beginning we were misguided by some Africa flights in a way that we tended to blame to low model mixing ratios to convection problems. This graph finally demonstrated the methane increase and the effect on Africa flights which started in 2009.

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

In the new version “Figure 10” includes 16 stations - interpolating lines in this graph are helpful to visually associate the points to a category. (cf. Fig. 4 at the end of this document)

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

The spatial coverage of the flights changing over time used to be depicted in Fig.S6, which is now Fig. S7 in the new paper version.

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

Fig. 17 is now Fig. 15 in the new version and the legend is updated (cf. Fig. 15 copy at the end of this document).

References

Dlugokencky, E. J., L. Bruhwiler, J. W. C. White, L. K. Emmons, P. C. Novelli, S. A. Montzka, K. A. Masarie, P. M. Lang, A. M. Crotwell, J. B. Miller, and L. V. Gatti, Observational constraints on recent increases in the atmospheric CH₄ burden, *Geophys. Res. Lett.*, 36(L18803), doi: 10.1029/2009GL039780, 2009.

Dlugokencky, E. J., E. G. Nisbet, R. Fisher, and D. Lowry, Global atmospheric methane: budget, changes and dangers, *Phil. Trans. R. Soc. A* 369(1943), 2058-2072, doi: 10.1098/rsta.2010.0341, 2011.

Rigby, M., R. G. Prinn, P. J. Fraser, P. G. Simmonds, R. L. Langenfelds, J. Huang, D. M. Cunnold, L. P. Steele, P. B. Krummel, R. F. Weiss, S. O'Doherty, P. K. Salameh, H. J. Wang, C. M. Harth, J. Mühle, and L. W. Porter, Renewed growth of atmospheric methane, *Geophys. Res. Lett.*, 35(L22805), doi:10.1029/2008GL036037, 2008.

Rigby, M., S. A. Montzka, R. G. Prinn, J. W. C. White, D. Young, S. O'Doherty, M. F. Lunt, A. L. Ganesan, A. J. Manning, P. G. Simmonds, P. K. Salameh, C. M. Harth, J. Mühle, R. F. Weiss, P. J. Fraser, L. P. Steele, P. B. Krummel, A. McCulloch, and S. Park, Role of atmospheric oxidation in recent methane growth, *Proceedings of the National Academy of Sciences*, 114(21), 5373-5377, doi: 10.1073/pnas.1616426114, 2017.

Saunio, M., P. Bousquet, B. Poulter, et al., Variability and quasi-decadal changes in the methane budget over the period 2000–2012, *Atmos. Chem. Phys.*, 17(18), 11135-11161, doi: 10.5194/acp-17-11135-2017, 2017.

Schaefer, H., S. E. Mikaloff Fletcher, C. Veidt, K. R. Lassey, G. W. Brailsford, T. M. Bromley, E. J. Dlugokencky, S. E. Michel, J. B. Miller, I. Levin, D. C. Lowe, R. J. Martin, B. H. Vaughn, and J. W. C. White, A 21st century shift from fossil-fuel to biogenic methane emissions indicated by ¹³CH₄, *Science*, doi: 10.1126/science.aad2705, 2016.

Turner, A. J., C. Frankenberg, P. O. Wennberg, and D. J. Jacob, Ambiguity in the causes for decadal trends in atmospheric methane and hydroxyl, *Proceedings of the National Academy of Sciences*, 114(21), 5367-5372, doi: 10.1073/pnas.1616020114, 2017.

Worden, J. R., A. A. Bloom, S. Pandey, Z. Jiang, H. M. Worden, T. W. Walker, S. Houweling, and T. Röckmann, Reduced biomass burning emissions reconcile conflicting estimates of the post-2006 atmospheric methane budget, *Nature Communications*, 8(1), 2227, doi: 10.1038/s41467-017-02246-0, 2017.

Interactive comment on *Atmos. Chem. Phys. Discuss.*, <https://doi.org/10.5194/acp-2017-1212>,

2018.

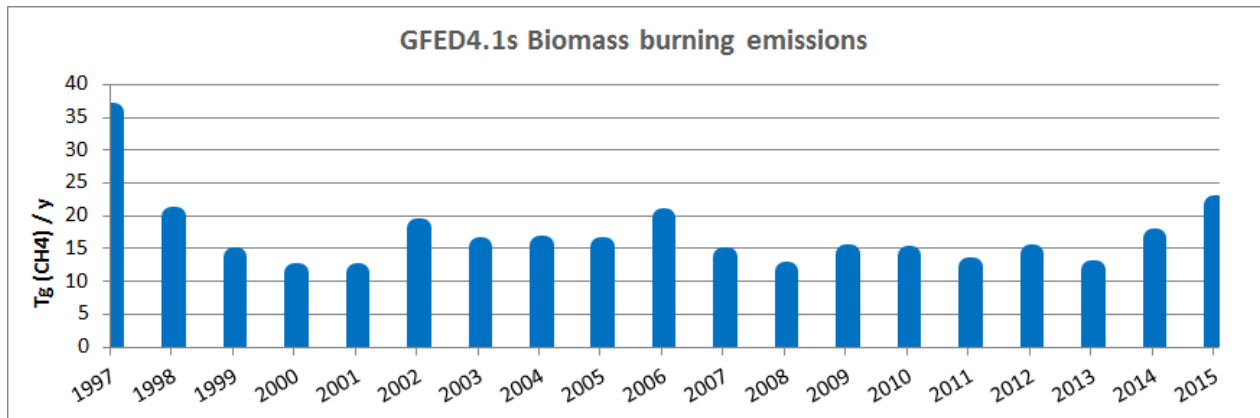
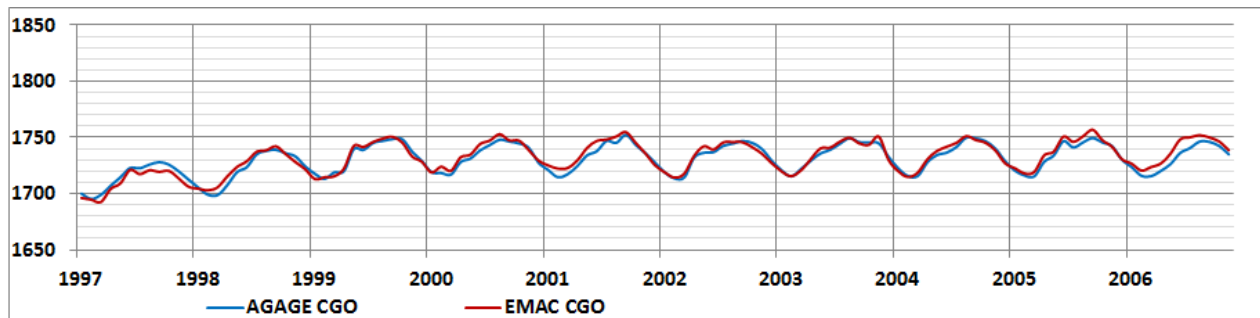


Figure 1: The integrated annual amount was calculated $5.7e12$ g higher by the solver.



*Figure 2: AGAGE station CGO, Cape Grim, Tasmania:
Observations (blue) vs simulation (red) – $R^2 = .93$*

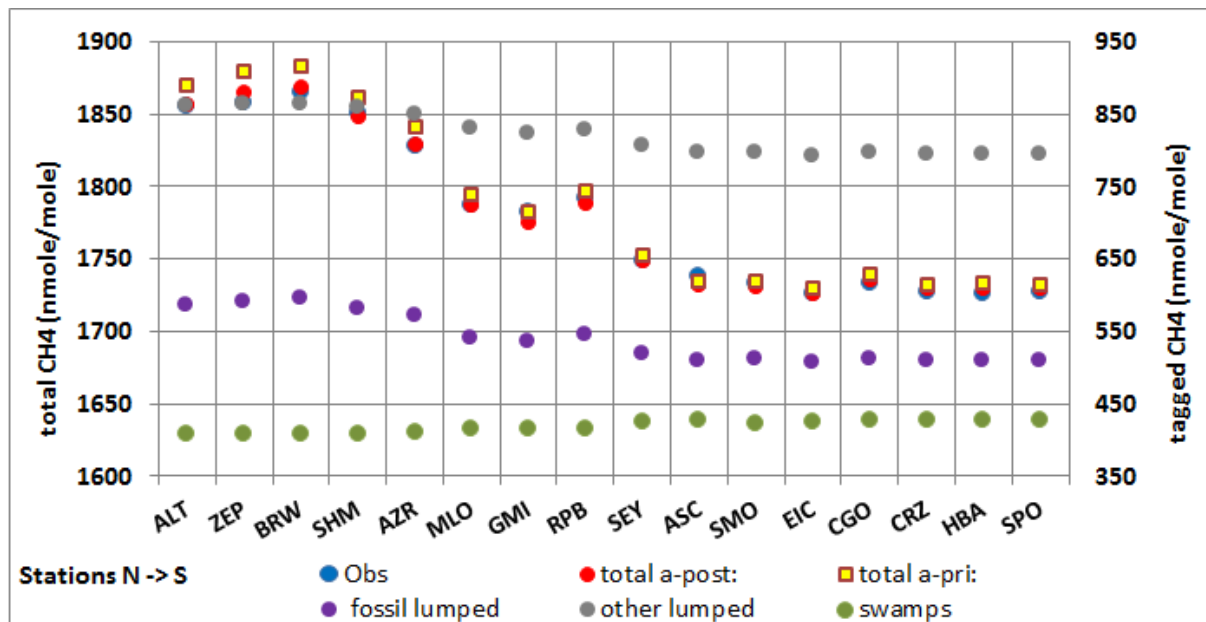


Figure 3: Optimization of calculated ground station CH₄ mixing-ratios towards observation (blue) in north-south direction by scaling the tagged emission contributions (now Fig. 6 in paper).

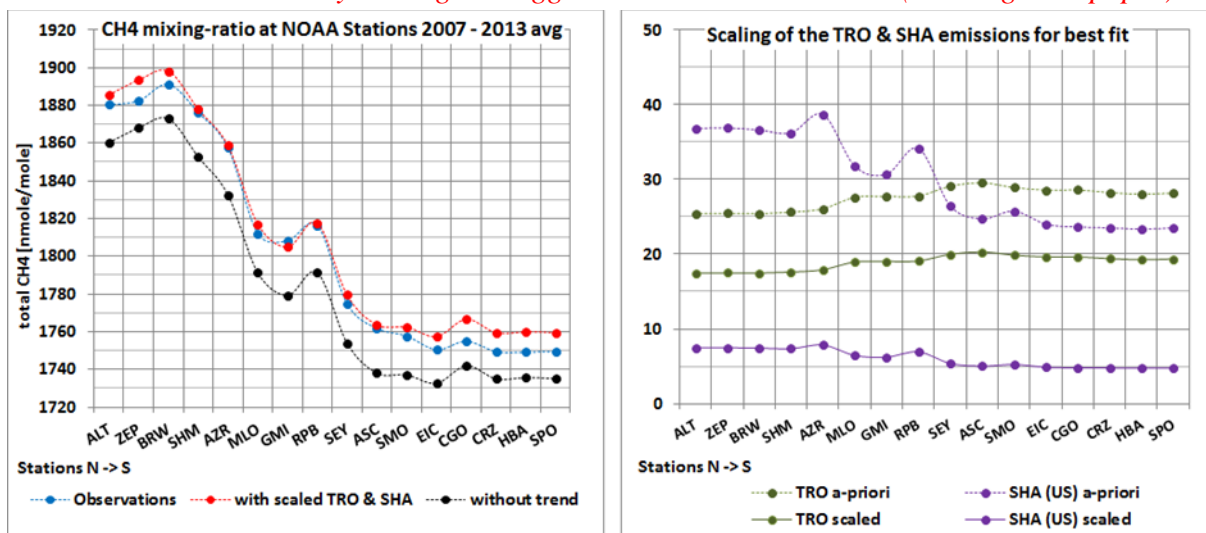


Figure 4: Scaling TRO and SHA emission fractions to fit the all-station observations within smallest RMS:

Left: Observations (blue) and total calculated CH₄ without- (black), and with (red) trend period emissions (solid lines right panel).

Right: A-priori estimates (dashed) and solver-scaled (solid) TRO (19.44)- and SHA (5.74 Tg/y) emissions for trend years. (now Fig. 10 in paper).

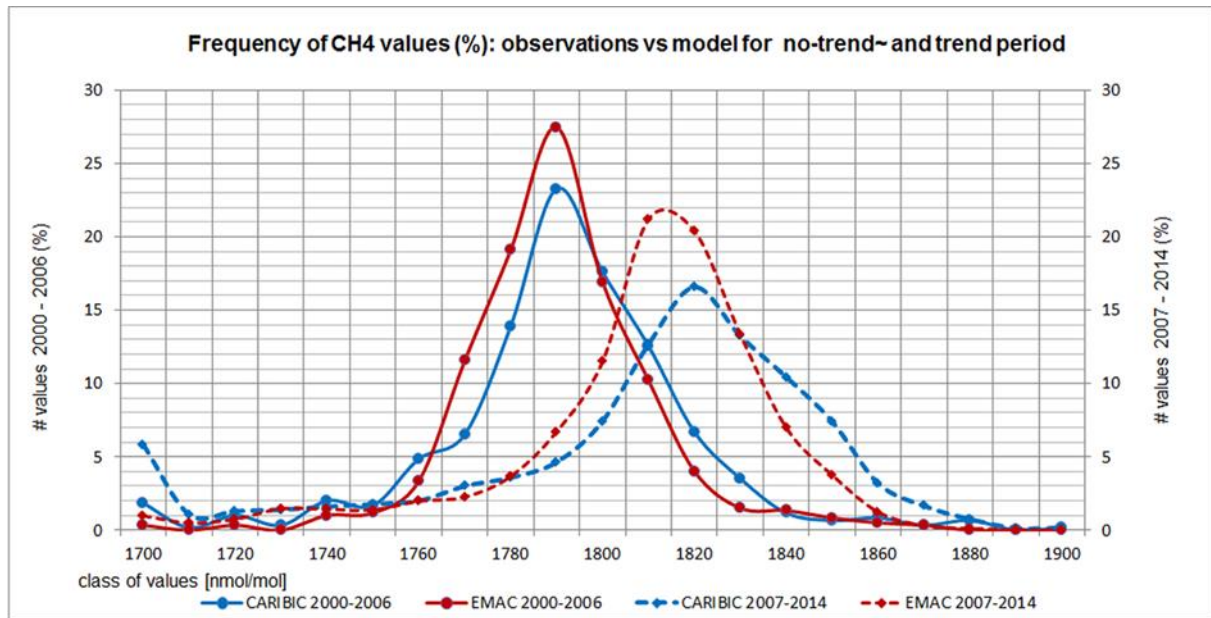


Figure 15copy: Frequency spectrum of CARIBIC observed and EMAC simulated CH₄-mixing-ratios separately plotted for the years 2000-2006 and 2007-2014.