Letter to the editor

Dear Editor, with this letter, the questions raised in the second round of review have been addressed. Here the comments are written in bold, followed by the reply. The overall text has been strongly revised, and therefore it was not feasible to add an annotated manuscript.

REFEREE #1

The manuscript by Zimmermann et al. presents findings of a modeling study trying to identify the source strength of CH4 from different sectors, so that the ECHAM5/EMAC/MESSy modelling system properly reproduces the measured CH4 concentrations throughout a 20 year period. In the manuscript the model setup is explained and the results are presented, supported by an abundance of figures.

We thank the referee for reading and comment the second version of our manuscript.

There are a few minor points that I would like to see addressed before final publication on ACP:

1 - The authors should mention a reason why they are not using information for the inter-annual variability of CH4 emissions. It is clearly stated that inter-annually constant natural and anthropogenic emissions are used, when there have been regional - and global, changes within the period they are simulating (e.g. see "Global trends of methane emissions and their impacts on ozone concentrations" (doi:10.2760/820175)). The do give an explanation on why they do not use inter-annually changing OH concentrations, supported by references, but not for the emissions. If they choose not to include these changes there should be a short assessment explaining how they believe this affects the results.

In our approach we adopted the geographical and seasonal distribution of a given set of emissions and assume their amounts to be constant over the “no-trend period” 1997 through 2006, which we see as permissible with regard to the long CH4 lifetime. We agree that this is an important assumption, and we underline this issue in the conclusions.

2 - The sentence in lines 139-141 is repeated 10 lines later (149-151). One of the two should be changed/removed.

The text has been completely rewritten for better readability.

3 - P4 L152: The authors state that the model properly represented the years 1997-2006 after multiple spin-ups. Up to this point in the manuscript there is no mention of any kind of emission optimizing. This means that the model, with inter-annually constant emissions, without any emission optimization taking place, captured both the increasing concentrations of the years 1997-2000, and the stagnated period of 2000-2006. If this is the case, I would assume that the model was not yet in equilibrium, since there should be a linear relation between emissions and CH4 concentrations.

We agree with the referee and we would like to mention that the non-equilibrium in 1997-200 is due to strong biomass burning event which does influence the following years due to the long methane lifetime (see for example Granier et al. (2011, doi: 0.1007/s10584-011-0154-1).

EDITOR'S REVIEW

The manuscript uses a large number of ground based and aircraft data to evaluate the EMAC methane simulations and to attribute the observed methane trends to changes in specific emission changes.

However, the manuscript lacks of clarity in terms of clear hypothesis, key message and associated uncertainties. The added value compared to earlier studies has to be clearly stated.

The abstract needs to be restructured: 1. Key question, 2. Use of EMAC and observations, 3. Assumptions made for modeling, 4. Key results and added value compared to earlier studies (to what the no-trend period is due and to what the trend-period). Also it misses some quantitative information on the findings, e.g. (lines 28-30) by how much fossil fuel is reduced and tropical wetlands and rice paddies are increased in the posteriori emissions?

Dear Editor, following the main comment we decided to rewrite the manuscript to make it more readable and easier to follow. Not only the abstract has been reformulated, and all the repetitions have been removed.

Furthermore, there are a number of hypotheses made for this work that require to be clearly spelled out, justified and discussed with regard to the uncertainty they introduce to the conclusions of this study (as also pointed by the other reviewer). This has to be done early in the paper in section 2 where the model set up is explained. An appropriate place would be section 2.2., which could be 'Model set up and assumptions made'. These assumptions are mainly:

1. the no-interannual variability of OH radical and thus the use of a prescribed OH radical concentration, which also implies no chemical feedbacks and linearity in the chemical destruction of CH4. The use of the prescribed OH is mentioned in the model set up while references that can partially support such choice are coming later in page 12 (lines 440-444). No discussion is really made on the uncertainty introduced in the results from such assumption.

2. the constant interannual CH4 emissions for the a priori scenario

3. testing different changes in specific sources emission intensity but assuming the constant geographic distribution per source.

4. Neglecting a change in the soil sink? When optimizing for the emissions (or I misunderstood?)

We have summarized these assumptions in the conclusion of the manuscript where we think it would be more appropriate.

The model description is also unclear at several places. For instance

1. How the prescribed OH field has been derived (line 146 or later on line 212)?

2. How the steady-state global CH4 simulation for the years 1997-2006 has been performed? (lines 152-153

3. How many and which online simulations of EMAC have been performed ?

4. And how many and which 'solver' fits have been performed?

5. What kind of constrains are imposed to the 'Solver'

All these informations have been added to the manuscript.

a. which constrains are exactly used in this study?

b. for which case studies is the Solver used? (A Table could be very illustrative for this).

c. As currently written, the reader has the impression that multiple (online) EMAC simulations have been performed and the figures presenting the model versus observations show the results of such simulations.

d. In lines 321-324 you discuss tolerance intervals; which are used for the various sources studied here?

All these points have been now clearly addressed in the manuscript.

6. The tagged simulations (are online simulations?) have to be described in section 2 and not refer to them mainly in section 4 (results). Also give the simple equation (sum of tagged species) to explain how you calculate the total CH4 concentration.

In addition, there are several repetitions throughout the manuscript that have to be removed.

At several places text from other publications is used with quotation marks, for instance lines 82-92, or without, for instance lines 441-443. Such text has to be re-written in author's proper words with appropriate reference to the original study.

Done and quotations have been removed.

Furthermore, there are several long sentences difficult for the reader that can be broken down in 2 or 3 shorter ones and increase readability of the text (for instance lines 21-24).

As the entire text has been revised, we do not have a point by point answer to the specific comments. The main revisions of the text are:

The abstract has been completely rewritten, highlighting the main conclusion of this work. While the introduction remains mostly untouched, the section on the numerical model (Sect.2) has been re-structured to give a better overview of the emissions and the numerical work performed in our study, with special focus on the tagging and error minimization procedure. While the observations description has not changed, the further sections (4 and 5) where drastically reduced, by removing all the redundant information and trying to be clear and concise. We hope in this way to have increased the manuscript readability and to facilitate the editor's work in this way.