Responses to Anonymous Referee #1

This paper proposes an analysis of the global and regional methane cycle for the past 40 years using long-term simulations of a chemistry-transport model, forced by state-of-the-art emissions and sinks and with analysed meteorology (after 1997). It presents the impact of the different regions and processes on the atmospheric observed changes at a subset of surface atmospheric stations measuring methane and for the different important periods of the methane cycle: the pre-1990s growth, the 2000s stagnation and the post 2006 renewed growth.

The paper is an important and useful piece of information about the methane cycle for the past four decades. One original point is about the balanced analysis of both sources and sinks, whereas most studies on the global methane cycle focus on emissions only. Another (related) interesting point is the analysis of the underlying processes of the OH trend found in the paper. The outline is clear and it is well-written. The figures are supporting the text. I suggest publication in ACP after accounting for the following comments and questions

➢ *We thank the reviewer for a thorough evaluation with useful suggestions helping to improve the manuscript.*

General comments

1/ The rather crude extrapolation done for the emissions after 2008 limits the analysis of 2007-2012 period. My suggestion is to re-run the last period of the 40 years with less anthropogenic-source-increasing scenario the prescribed one is clearly not adapted to the observations (and now rather well documented). If too long, this solution should be replaced by more acknowledgements in the text that the conclusions about this part should not be taken with caution.

➢ *The Edgar 4.2 inventory we used was recently extended with gridded anthropogenic methane emissions up to 2010 and emissions per country basis up to 2012. A simplified approach was used and updates for other key emission components (NOx, CO, NMVOCs) are not available yet. Our applied natural methane emission inventory lack data after 2009 (wetland emissions drive much of the inter-annual variability). Therefore, the results from a rerun for the 2009-2012 period would still be hampered by considerable uncertainty. Instead of rerunning the model we have added a comparison and discussion of anthropogenic methane emission estimates over the period 1990-2015 containing our extrapolation of the Edgar data, the Edgar update and ECLIPSE emissions in the Supplement. Though we had already stated in several places in the main manuscript that the conclusions for the 2009-2012 period are uncertain, we agree with the reviewer comment and now acknowledge this more strongly throughout the manuscript.*

2/The paper is too long to my opinion with too many figures and no real synthesis at the end of each section (e.g. the interesting lifetime sections need synthesis and conclusions). It leads to hide and diffuse a bit too much the important results of the paper to my opinion. In particular, I suggest a substantial reduction of section 3.3. Please provide a section with more synthetiszed text and only few stations that are characteristic of the different regions, to support the conclusions of the text for the main regions. Else, the reader gest a bit lost in the large amount of local to regional results provided. Other stations can go in the supplementary with their detailed analysis. Else it is too dense

➢ *We have made changes in the manuscript in accordance with the suggestions. The text is shortened in several places, especially section 3.3, where also the number of compared stations are reduced. Now, a short synthesis is added at the end of the sections as suggested by the Reviewer to improve the clarity and highlight the important findings. This is done for the lifetime section (section 3.5) and a summary of the two sections 3.3 and 3.4 is given at the end of section 3.4. Some more text is also added to the Introduction to emphasis the motivation for the various analysis made in the Results section.*

3/The "tracer" analysis is interesting but the main text should include the minimum to understand what is done, which is not the case (see specific comments)

We agree, and we have now included more information in the introduction (first paragraph) of section 3.3: "In the Supplement, we explain how the CH_4 mole fraction can be split into two components: A quite uniform background component and an inhomogeneous recently emitted component. The latter is advected and mixed, and when achieving a good mixing (after 1-2 months) it is converted into the background component. We show how the use of a 1-month e-folding fictitious tracer (Total tracer) is valid as a proxy for the inhomogeneous component. The CH_4 surface emissions act as the sources for the tracer. In the Supplement we use the continuity equation for the CH_4 mole fraction (CH_4 model) as starting point and further arguments to derive the following approximation:

$$<CH_4 \text{ model}> - [<CH_4 \text{ model}>] = B \times (<\text{Total tracer}> - [<\text{Total tracer}>]) + \text{Residual} \quad \text{(Equation 1)"}$$

In the last part of the paragraph we also added some additional information on the terms in equation 1.

4/In many places, the text should be more precise (see specific comments) and avoid redundancies (e.g. MCF & OH changes in several places)

➢ *We have removed some redundancies and shortened the text. We think this now provides a more concise and precise analysis.*

Specific comments

Abstract, last sentence : "In our analysis. . . ." Please provide more precise results in these relations.

➢ *We added a sentence with information on the key meteorological and chemical factors behind these relations in the revised version: "In our simulations, the atmospheric CH_4 lifetime decreases by more than 8 % from 1970 to 2012, a significant reduction of the residence time of this important greenhouse gas. Changes in CO and NO_x emissions, specific humidity, and ozone column drive most of this, and we provide simple prognostic equations for the relations between those and the CH_4 lifetime."*

P30898, l7 : It should be mentioned that Bousquet et al. provides optimized emissions against atmospheric observations. However, using only their natural+BBG do not guar-antee that the atmospheric evolution will be matched as anthropogenic emissions are taken from EDGAR. This should be précised at some point.

➢ *We have now included text about this issue in the comparison of global mean surface methane (section 3.2)*

P30898, l15 : As EDGAR is already suspected to have too large emissions and trends (e.g. Bergamsachi et al 2013), the extrapolation after 2007 is probably enhancing even more the issue. EDGAR have released their data until end 2012 now. Can you compare your extrapolation with their data and eventually acknowledge differences ? Ideally, It would be necessary to redo the end of the period with more realistic anthropogenic emissions accounting for trends more in line with IIASA ECLIPSE or EPA or at least with the latest EDGAR. I do not request it but this issue should be mentioned at this early stage of the paper and discussed later in the text.

➢ *This last EDGAR update was done using a simplified (fast track) approach and updates for other key emission components (NOx, CO, NMVOCs) are not available yet. Our applied natural methane emission inventory lack data after 2009 (wetland emissions drive much of the inter-annual variability). Hence, the results from a rerun for the 2009-2012 period would still be hampered by considerable uncertainty. ECLIPSE emissions are only available on 5-year intervals and extrapolating them to annual intervals would also be a simplification. A scenario is used by ECLIPSE to estimate 2015 emissions based on 2010 data, which would add uncertainty if this inventory was used. Another difficulty with using ECLIPSE emissions is that the first year of the inventory is 1990 while we start our simulations in 1970. Instead of rerunning the model, we have added a comparison for anthropogenic methane emissions over the period 1990-2015 containing our extrapolation, the Edgar update, and ECLIPSE emissions in the Supplement. We now also compare the regional EPA inventory with the EDGAR inventory in our analysis for North America in section 3.3.*

P30899, l8 : the collapse of former USSR should be mentioned here.

➢ *This is now mentioned.*

P30899, l15 : why not applying BP statistics in your standard ? It seems more conser- vative than the simple extrapolation of EDGAR.

➢ *The BP approach, made for methane emissions only, and perturbing key methane emission sectors, was used as a simple sensitivity test on some of the potential impact of the financial crisis on the methane evolution since this is not captured in our baseline extrapolation. BP statistics for gas production, oil and coal consumption were used to scale relevant methane emission from oil, gas and coal production. If this approach were to be used to set up a consistent baseline emission inventory, these BP factors should also be used to scale emissions of other compounds (e.g., CO and NOx). This would be complicated and will introduce uncertainties as it is less clear how to use the BP factors to scale emissions for road traffic, power plants etc., which constitute a substantial share of the emissions for these compounds. Therefore, a simpler extrapolation based on changes for previous years was preferred as baseline since it can be used in a consistent way for all compounds.*

P30900, l25 : how do you "drive" the model ? Nudging ? which variables ? which relaxing time?

➢ *In the manuscript we refer to Søvde et al. (2012) for model details to avoid the article becoming too long. In the manuscript we had written: "The Oslo CTM3 simulations were driven with 3-hourly meteorological forecast data from the European Centre for Medium-Range Weather Forecasts (ECMWF) Integrated Forecast System (IFS) model (see Søvde et al. (2012) for details). We now added the following information: "These data are 36-hours forecasts produced with 12 hours of spin-up starting from an ERA-Interim analysis at noon on the previous day".*

P30901, l1 : why not using ERA-I product instead of recycled meteorology ? It would have allowed to study the impact of varying meteorology on you results for the full period ?

➢ *The IFS forecast fields are used since they provide additional meteorological fields needed in our transport scheme that are not available from reanalysis products. An additional advantage of using IFS is the availability of 3rd hourly meteorological fields compared to 6 hourly for ERA.*

Figure 2. Very interesting figure indeed. I ma surprised not to see more the effect of Pinatubo eruption on the loss ? Can you comment ? Also, the period after 2008 is hard to fully analyse because of the crude hypotheses on emissions changes. Again, if possible it would be good to update emissions and re run the last years to draw more robust conclusions. But I leave the option to the decision of the authors.

➢ *Reduced emissions are implicitly included in the natural CH_4 emission inventories, but*

changes in meteorology (temperature, water vapor, etc.) and volcanic SO_2 and sulphate aerosols in the stratosphere, are not accounted for in the simulations and that is the reason why there is too small effect of Pinatubo on the loss. These issues are discussed later in the text, in section 3.4.

30903, l27: Can you give at least the relative importance of Chlorine and 01D loss in your study here ?

➢ *We only calculated the total loss in these simulations and not the contributions from individual compounds. In previous studies performed with this model, the loss from chlorine and O^1D were in the order 5-10 %, in agreement with findings in other studies in the literature.*

Figure 4 needs attention. I suggest to add a panel below the evolution of the global mixing ratio representing the atmospheric growth rate (derivative of the model and obs mixing ratios) for observations and model (as done by NOAA on its website (Dlugo- kencky classical double panel figure). This would reveal more clearly the model goods and weaknesses.

➢ *Growth rates are shown in figures 11-14 and discussed in detail in section 3.4. We agree that it would provide useful information also in Figure 4 but we decided to limit these discussions to figs. 11-14 and section 3.4 to avoid repetitions and increasing the number of figures*

Supplementary, S3. It should be mentioned that " ' " refers to time fluctuations and " * " refers to longitudinal fluctuations. What is the impact of this rather technical treatment of the 18 tracers compared to simply using their relative weight as passive tracers emitted 1 month and stopped ?

➢ *This is now mentioned in the Supplement. The technical discussion in the Supplement explains why we can use simple passive tracers with e-folding lifetime of one month as proxies for recent methane changes. Emitting tracers for one month and then stop would only give the influence from various sectors for that specific month and not capture sectors/regions responsible for trends in methane concentrations.*

P30905, l9. It is unclear and not straightforward how equation 1 comes from the text the supplementary (S3). This paragraph should be clarified for the reader to have enough information in the main text. I suggest to phrase in simple words what equation 1 represents. You want to represent the contribution of all the different tracers at different stations after removing seasonal cycle (<>) and north/south differences ([]). It would help the reader to have things written with words at this stage. - B is not clearly defined.

- "if some prerequisites discussed in thesupplementary are met. Âż : please be more precise

here, unclear.

➢ *We agree. We have added new text and included information from the Supplement in the main text to make these issues clearer. The new introduction in section 3.3: "In the Supplement, we explain how the CH_4 mole fraction can be split into two components: A quite uniform background component and an inhomogeneous recently emitted component. The latter is advected and mixed, and when achieving a good mixing (after 1-2 months) it is converted into the background component. We show how the use of a 1-month e-folding fictitious tracer (Total tracer) is valid as a proxy for the inhomogeneous component. The CH_4 surface emissions act as the sources for the tracer. In the Supplement we use the continuity equation for the CH_4 mole fraction (CH_4 model) as starting point and further arguments to derive the following approximation:*

$<CH_4$ model$>$ – $[<CH_4$ model$>]$ = B x (<Total tracer> - [<Total tracer>]) + Residual (Equation 1)

Where [ ] denotes longitudinal mean along a whole terrestrial parallel and < > denotes annual running mean. We are interested in the inter-annual variation of CH_4, so we have carried out annual running means to remove the strong seasonal cycle. The subtraction of longitudinal means on each side of Eq. 1 removes the influence of differences in lifetimes (the mean lifetime of CH_4 is around 9 years, whereas the mean lifetime of the Total tracer is 1 month). B and Residual are constants (or almost constant), if the prerequisites discussed in the supplement (S3, last paragraph) are met. We expect B to be near or equal to 1, and Residual to be small. If B and Residual were exactly constant, the Pearson linear correlation coefficient between $<CH_4$ model$>$ – $[<CH_4$ model$>]$ and <Total tracer> - [<Total tracer>] would be exactly equal to 1. The tracer approach then gives valuable information on the contribution to CH_4 variation from recent regional-local emission or transport changes.

P30905, l18 : "recent regional-local emission or transport changes Âż : as you remove the longitudinal mean, would not it be only (or mostly) East/west changes that you can analyse ? Please be more precise here.

➢ *Mostly, but in some cases additional information can be extracted, i.e. discussion of contributions to trends at Ushuaia and Cape Grim where we discuss how the tracer results indicate that long-range transport from other latitudes are decisive.*

P30905, l20 : I agree with the argument of time/space coverage, but the R2 argument is a bit weak. For stations with poor model performances, it is critical to study them and analyse why the model fails. The different tracers can bring information on this. I strongly suggest to add an analysis for such stations (if existing) with some text & hypotheses for the causes of low performances. Else it gives the impression that the authors have (a bit) chosen the stations at "their convenience". (p30906, l 15-16 is too short on this aspects)

➢ *The stations chosen for comparisons are based on the objectively defined station selection criteria given in section 2.3. An additional constrain is employed in the tracer study (section 3.3). As explained in section 3.3, only stations where the coefficient of determination between <CH_4 model> – [<CH_4 model>] and <Total tracer> - [<Total tracer>] is higher than 0.5 are used for this analysis. We agree that analysis of the different tracers could give some information on the causes at stations with poor model performance but it would not provide the complete picture. Revealing the causes would to our opinion be the topic of a separate study/paper covering runs with different model resolutions (reviewer agrees that coarse resolution likely is a major cause) and sensitivity studies on other possible explanation factors (transport, chemistry, deposition, etc.).*

P30905, l20 and P30906, l14-15 : What do you exactly correlate (deseasonlized totals, full signals, ..)? This is a bit confusing. It should be precised in the text.

➢ *We think the general clarifications made in section 3.3 now make this easier to grasp. For the first coefficient the text says: "the Pearson linear correlation coefficient between <CH_4 model> – [<CH_4 model>] and <Total tracer> - [<Total tracer>]". This correlation is for annual running means as < > (defined earlier in the same paragraph) denotes annual running mean. For the second coefficient it is written: "In general the model reproduces the seasonal and year to year variations very well with high coefficients of determination, R^2, for most stations," ,so this is for the full seasonal (monthly signal) as seen in the top panels of Fig. 6-10.*

Fig 6-10 : Using the marine boundary layer latitudinal synthesis from NOAA to get [<observation>], you could probably compute <observation> - [<observation>] as well and compare to the same model term. Did you try this ? It would worth trying.

➢ *This is an interesting suggestion. Due to sparse coverage of stations in many regions the width of the latitudinal bands in the NOAA latitudinal synthesis is much larger than the width we use in the tracer approach. Unfortunately, the tracer approach is no longer valid if we extend the widths of the latitudinal bands. Moreover, the NOAA marine boundary layer latitudinal synthesis is only valid above the oceans, whereas most of the Earth parallels contain also a significant fraction of land*

P30907, l21 : "This indicates that the contribution to CH4 from regional emissions are small and that long-range transport from other latitudes is decisive Âż. I do not fully agree as Cape Grim is one of the only site where, the B(tracer – mean(tracer)) term explains the growth after 2000. Please provide explanations in the text.

➢ *B(tracer – mean(tracer)) + the Residual explains the difference CH4-mean(CH4) (see eq 1) and* ***not*** *the trend in CH4. See equation 1 in the manuscript.*

P30908, l19 : Keybiscane analysis. This requires attention. Is the coal increase from EDGAR reliable ? Can you cross this increase with EPA inventory and see whether this is consistent or not ?

➢ *The increase in coal emissions from 2003 to 2008 is almost 12 % in the EDGAR inventory. An increase of 28 % is found from 2005-2010 in the EPA inventory. We have added this information in the manuscript.*

P30909, l3 : "i.e. other locations at the same latitudes have a larger trend in CH4. Âż Pleas be more precise here. As Europe also shows reductions the blame is probably on Asia as shown by following figures.

➢ *We agree and now point to Asian emissions as the likely cause.*

P30912, l1 : for Minamitorishima, I do not understand why B(tracer – mean(tracer)) term is constantly decreasing. With the pattern of individual emission change (mostly increase). Please provide explanations in the text.

➢ *This means that the latitudinal mean tracer grows more than the tracer at this specific site. Since this is a background station in the ocean some of the signal from the strong emission increase at the continent disperses before reaching the station. This station also has a relatively strong influence from regional natural emissions. The tracer from this source decreases somewhat over time whereas the latitudinal mean tracer for this source shows a smaller trend. (To shorten section 3.3 the comparison for this station is now removed from the manuscript.)*

P30913, l28. I think there is now a majority (if not a consensus yet) to agree that OH variations inferred for the 80s/90s from MCF are too large (e.g. Montzka et al 2011). I would be more clearly state this point that wetland variations are most probably overestimated in Bousquet et al., 2011 for this period.

➢ *We agree and have changed the text in accordance with the suggestion from the reviewer.*

P30914, Pinatubo analysis. OH changes are not mentioned in this analysis whereas it probably explained a lot of the changes. Why so ? Ir it because "changes in meteo-

rology (temperature, water vapor, etc.) and volcanic SO2 and sulphate aerosols in the stratosphere Âż are not accounted for ? You should at least specified their expected impact on methane through OH changes (reduction).

➢ *The theoretical background refers to literature studies and discuss the overall effect of*

emission and OH changes to compress the text. We agree that we could provide a bit more information and have now adjusted the text to distinguish those perturbations only affecting OH and how this changes methane.

Figure 12 : Why coal and gas are largely positive in the southern hemisphere for this period ? Please comment on that in the text

.

➢ *Instead of showing relative numbers for growth, the figure is now changed to show absolute numbers. This gives a better measure of the individual sector's contribution and makes it easier to see where emission changes occur. The coal and gas signal is no longer as striking as for relative numbers, therefore we include them under the "anthropogenic" umbrella in the text: "Both model and measurements have the strongest growth (Fig. 12) in the Southern Hemisphere, which had large wetland emissions in 1998 (Bousquet et al., 2006;Dlugokencky et al., 2001). In the model, slowly rising anthropogenic emissions in the Southern Hemisphere also seems to contribute (Fig. 12b-f)."*

P30918, l7 "Much of this is due to intensification of oil and shale gas extraction in the US and coal exploitation in China Âż. Are gas emissions from gas extraction in the US increase in EDGAR4.2 ? I am not sure this inventory accounts for the shale gas for instance. Please precise.

➢ *The EDGAR inventory does not fully take into account the increase in shale gas exploitation. However, in this discussion in the manuscript we refer to a number of studies in literature of which some take into account US shale gas extraction.*

P30919, l7-9 : "who attributes much of the recent increase in total emissions to wet- lands Âż I suggest to add Âń for the period 2007-2009 Âż as Bousquet et al study does not cover the most recent years (since 2010).

➢ *We agree. The sentence now says: "Our natural emissions are from Bousquet et al. (2011) who attributes much of the 2007-2008 increase in total emissions to wetlands."*

P30921-22 : please provide a conclusion to the literature analysis performed about OH changes. There might not be a consensus but it is worth summarizing where we are at the end of the part.

➢ *In accordance with the suggestion we have now added a few concluding remarks.*

P30922 : "An increase in NOx emissions increases global OH as long as it takes place

outside highly polluted regions Âż : what happens in Asia so ? It is important to estimate the impact of such highly polluted regions on you conclusions about OH impacts in this paper. Please provide at least hypotheses.

➢ We have now added the following text in section 3.5: "Of particular importance is large increases in OH over Southeast Asia, mainly due to strong growth in NO_x emissions. From 2000-2010 the modelled tropospheric OH column increase by 10-20 % over China and India (not shown)."

P30924 : Are these two equations to represent methane lifetime very dependent of your model ? It would be important to assess somehow the genericity of these equations as it may be useful for other scientists in the communitog.

➢ *We think the key message from this finding is that simplified equations can be used to hindcast or project methane lifetime for similar types of perturbation studies. This should be of interest for other scientists in the community. Regarding model genericity it is a bit difficult to answer. To our knowledge it is only Holmes et al. (2013) and Dalsøren and Isaksen (2006) who have performed similar analysis on equations for methane lifetime. Since we study different time periods and both emissions and meteorology are perturbed in our simulations this probably explains much of the difference compared to these studies. In general, if multi-model studies perform identical emission perturbations we expect the slope and intercept of the equations to be model dependent due to numerous differences in applied chemistry schemes, transport schemes etc. However, we think that the OH affecting parameters forming the equations should be the same. If the parameters are different it would be a reason for detailed investigation of differences in model representation of OH chemistry.*

P30925, l19 : Âń that our applied emission inventories are reasonable Âż i suggest to rephrase : that our applied emission inventories and computed transport and chemistry are reasonable.

➢ *We agree and included the suggested addition to the text.*

P30925, l27 The model overestimates the growth in all regions, in particular in Asia after 2006

➢ *We now make it clear that this is after 2006.*

P30926, l28 : ". . . model results after 2009 due to lack of comprehensive emission inventories Âż. Edgar4.2, although not perfect as noticed in the paper has released data until 2012. There is also IIASAS and EPA having projections for the next years. I would rephrase suggestions that inventory should improve and account for consistent suggestions

that Asian emissions are overestimated in EDGAR.

- *For the global inventories the 2008-2012 EDGAR projection is based on a simplified approach and the step from 2010 to 2015 in ECLIPSE is scenario based. We therefore think it is correct to state that comprehensive emission inventories for recent years are lacking.*