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Object: Second revision of the manuscript acp-2014-235

Toulouse, 16 September 2014

Dear Dr Reeves,

Please find attached the second revision of the manuscript initially entitled "**Variability of tropospheric methane above the Mediterranean Basin inferred from satellite and model data**" by Ph. Ricaud and co-authors, together with the replies to the comments from one reviewer.

We hope the revised manuscript meets the high scientific standard of the ACP journal to be accepted for publication.

Please note that the title of the article has been changed from:

> **Variability of tropospheric methane above the Mediterranean Basin inferred from satellite and model data**

to:

> **Impact of the Asian Monsoon Anticyclone on the Variability of mid-to-upper tropospheric methane above the Mediterranean Basin**

We hope this change in the title of the manuscript will be acceptable in the review process of the ACP journal.

Sincerely,

Philippe Ricaud

Replies to the reviewer

Version 16, 16 September 2014

Review of "Variability of tropospheric methane above the Mediterranean Basin inferred from satellite and model data" by P. Ricaud et al.

Overall, the revised manuscript has been significantly improved from the previous version. In particular, the inclusion of trajectory model simulations adds credibility to the correlation between the methane variations and the circulation. However, I think the overall structure of this manuscript can be improved even further by putting a little more work. Below are my suggestions for the authors may take into consideration.

1. I think using measurements from more than one satellite and multiple model simulations are definitely beneficial to this type of work. However, one still wants to know how those satellites products are selected for this work among others and how using three different model simulations are beneficial to the comparisons. Did the authors include AIRS upper tropospheric profiles because IASI does not provide vertical profiles of methane? Are GOSAT data considered to be useful to this work even with fewer measurements with higher noise? Is one of the research goals to validate the satellite products? This can be added in the section starting from L97.

→ We have clarified this point by inserting a new paragraph after presenting all the spaceborne instruments able to detect tropospheric CH_4 and the different types of models that take into account CH_4. This paragraph replaces the two sentences starting after L. 153.

> We have collected the maximum of information available from satellite measurements and model results in order to study the variability of tropospheric CH_4 over the MB and to assess the processes driving this variability. We have thus built a wide dataset combining all these pieces of information keeping in mind that 1) it is out of the scope of the present paper to perform a validation of satellite products, 2) all these datasets have their own strengths and weaknesses, and 3) the more data we gather, the better the statistics are and furthermore, the dataset consistency can be better assessed. Regarding space-borne measurements, we have considered tropospheric columns of CH_4 from IASI over the period 2008-2011, and upper tropospheric CH_4 profiles from AIRS and GOSAT over the periods 2008-2011 and March-November 2010, respectively. Regarding the models, we have considered three types of chemical models to calculate CH_4 variability in the mid-to-upper troposphere. The MOCAGE (Josse et al., 2004) chemical transport model (CTM), constrained by the ARPEGE meteorological analyses, should a priori give CH_4 vertical profiles more realistic than climate models over a specified period despite the fact that, due to the long lifetime of CH_4, the short spin-up period (3 months vs. 12 years of lifetime) may impact its distribution. On the other hand, chemical climate models (CCMs) as LMDz-OR-INCA (Hauglustaine et al., 2004; Szopa et al., 2013) from the Laboratoire des Sciences du Climat et de l'Environnement (LSCE) and CNRM-AOCCM (Huszar et al., 2013) from Météo-France are run over a much longer period (greater than 10 years) than MOCAGE and should be more adapted to study the climatological variability of CH_4 over the MB. The LMDz-OR-INCA is mainly dedicated to the tropospheric CH_4 profiles since it takes into account the major surface processes that can drive the CH_4 variability in the entire troposphere depending on the inventory scenarios (see section 2.2.3). The CNRM-AOCCM is mainly dedicated to the upper tropospheric-stratospheric CH_4 profiles because it has a detailed description of the

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stratosphere and should better describe the processes impacting the CH4
variability in the upper troposphere-lower stratosphere. The 3 models are
thus complementary in the study of the CH4 variability in the mid-to-
upper troposphere over the MB.
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2. What are the similarities and differences between the three models used in this study? And again why those three models are selected and used in this study? I think comparing results from two different CTMs, for example, would be simple to understand. But when I look at the results from three different models, e.g., CTM, GCM, and CCM, I am not sure how to interpret their differences. Aren't the results from the model simulations expected to be different? Section 2.2 includes detailed description of the three models. But I think it would be more useful to know what to expect from those model simulations. Is one model supposed to be better in simulating methane in the troposphere because of better chemistry modules, dynamics or resolution, etc.?

→ Part of this issue is already discussed in the point 1). Regarding the outputs from the models, a detailed comparison and some explanations of the differences obtained among the models considering different future emissions that have participated to the ACC-MIP intercomparison program together with CNRM-AOCCM are presented in a manuscript in preparation.

Ricaud, P., N. Jaidan, P. Huszar, S. Szopa, M. Michou, L. El Amraoui, J.-L. Attié, R. Zbinden, and D. Hauglustaine, Chemical climate evolution above the Mediterranean Basin, manuscript in preparation, 2014.

They have also been presented and discussed in two posters.

Ricaud, P., N. Jaidan, P. Huszar, D. Saint-Martin, M. Michou, L. El Amraoui, J.-L. Attié, B. Josse, V. Marécal, R. Abida, R. Zbinden, D. Hauglustaine, S. Szopa and the ACCMIP modelers, Chemical Climate Evolution above the Mediterranean Basin, SPARC Conference, Queenstown, New Zealand, January 2014.

Ricaud, P., N. Jaidan, L. El Amraoui, J.-L. Attié, R. Zbinden, M. Michou, P. Huszar, S. Szopa, D. Hauglustaine, J. Warner, T. August, and R. Imasu, Chemical Climate Evolution above the Mediterranean Basin, IGAC conference, Natal, Brazil, September 2014.

We can nevertheless try to give some explanations of the expected differences between the three models. The differences in the outputs of LMDz-OR-INCA regarding the 4 RCPs were already discussed in section 5 starting L. 592. We have also to underline that the three models behave consistently when their outputs are compared to observations considering either total columns or upper tropospheric CH_4 (Fig. 7) by showing a maximum in summer in the seasonal variation of the E-W.

The differences in the CH_4 evolutions over the MB from the 3 models might be due to:

a) Dynamics.

CTMs forced by analyzed meteorological fields might represent the atmosphere at a time t or over a period of few months more accurately than the meteorological fields provided by the GCMs. This is particularly important for the long-range transport and the impact of the Asian Monsoon Anticyclone over the Eastern MB. Due to the impact of the subsidence over the Eastern MB, stratospheric processes also need to be present. This is the case for the CNRM-AOCCM model but not for the LMDz-OR-INCA model.

b) Chemistry.

Both LMDz-OR-INCA and MOCAGE models contain a detailed tropospheric chemistry module. CNRM-AOCCM has a stratospheric module that is also used in the troposphere down to 500 hPa. Consequently, in terms of chemistry, the CH4 profiles in the troposphere should be better represented in LMDz-OR-INCA and MOCAGE than in CNRM-AOCCM.

c) Surface CH_4 emissions.

This is probably the key parameter to expect strong impact on the CH_4 fields calculated by the models. Indeed, the LMDz-OR-INCA model contains a very detailed vegetation model taking into account 4 different RCP scenarios and should give the best representation of climatological tropospheric CH_4, although MOCAGE relaxes CH_4 surface amounts to monthly averaged data. CNRM-AOCCM does not take into account surface CH_4 but rather relaxes to zonally symmetric CH_4 amounts following the A1B climate scenario and should provide a relatively consistent representation of CH_4 in the UTLS.

d) Spin-up period.

MOCAGE has been run over several years by considering a spinup period of 3 months that is rather short compared to the lifetime of CH_4 (~12 months). The two other CCMs were run over a very long period from 1980 to 2100 and, consequently, in these two runs, CH_4 can be considered to be in equilibrium but not in MOCAGE.

In conclusion (as presented in the point 1 and in the new version of the manuscript), we state again that the three models have their own strengths and weakness and they are very complementary for studying the time evolution of CH_4 over the MB.

3. Section 3 (L313~): I think it would be easier for the readers to understand the results if the figures are individually presented here instead of describing them all together. As a reader, I would like to be able to exactly follow what the authors are referring to, in each figure, which will also help understand results in a big picture. For example, which figures do I have to look at when I read L325-329?

→ As strongly recommended by the reviewer, we have systematically referred any comment/discussion to the associated Figure in the section 3.

4. L39: ~12 years and is supposed to be well mixed.

→ Done.

5. L52-54: This sentence can be rewritten for clarity. Also, at the end of the sentence, ..critical issue was to evaluate 'something…'.

→ We have clarified the sentence into:

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During the last decades, the impact and the role that atmospheric
trace gases play in climate and air pollution changes have been
the source of major concerns.
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6. L54: Full acronym for IPCC has to be given here.

→ We have modified the text as follow.

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Intergovernmental Panel on Climate Change (IPCC, 2007)
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7. L61-62: these long-lived greenhouse gases, e.g., CH4, N2O and CO2, account for.

→ Done.

8. L69: To illustrate, global (or regional) model simulations.

→ Done.

9. L72: asymmetry in precipitation over the MB?

→ Done.

10. L76: References to the O_3 and CO budgets needed here. I also wonder how relevant of those two species to this study.

→ The O_3 and CO budgets are affected by CH_4 through complex reactions with NOx (Dentener et al., 2005). We have modified the text and added some references.

> The impact of these distinct continental sources such as from manufactures and densely populated coastal areas (e.g. Marseille, Barcelona, Athens, Tunis, Cairo, Genoa or Roma) (Kanakidou et al, 2011; Im and Kanakidou, 2012) or forest fires (e.g. South East of France, Corsica, Portugal, Greece) (Cristofanelli et al., 2013) is still not perfectly understood, especially on the O_3 and CO budgets in which CH_4 interplays through complex reactions with nitrogen oxides (NOx) (Dentener et al., 2005).

Kanakidou, M., Mihalopoulos, N., Kindap, T., Im, U., Vrekoussis, M., Gerasopoulos, E., Dermitzaki, E., Unal, A., Kocak, M., Markakis, K., Melas, D., Kouvarakis, G., Youssef, A. F., Richter, A., Hatzianastassiou, N., Hilboll, A., Ebojie, F., von Savigny, C., Ladstaetter-Weissenmayer, A., Burrows, J., and Moubasher, H.: Megacities as hot spots of air pollution in the East Mediterranean, Atmos. Environ., 45, 1223–1235, 2011.

Im, U. and Kanakidou, M.: Impacts of East Mediterranean megacity emissions on air quality, Atmos. Chem. Phys., 12, 6335-6355, doi:10.5194/acp-12-6335-2012, 2012

Cristofanelli, P., Fierli, F., Marinoni, A., Calzolari, F., Duchi, R., Burkhart, J., Stohl, A., Maione, M., Arduini, J., and Bonasoni, P.: Influence of biomass burning and anthropogenic emissions on ozone, carbon monoxide and black carbon at the Mt. Cimone GAW-WMO global station (Italy, 2165 m asl), Atmos. Chem. Phys., 13, 15-30, doi:10.5194/acp-13-15-2013, 2013.

11. L76-79: Needs references here.

→ We have modified the text and added one new reference.

> Besides these regional sources, polluted air masses may originate from Asia during the summer monsoon period (Randel and Park, 2006), Africa through the Hadley cell and upper level anticyclone (Ziv et al., 2004; Liu et al., 2009) and North America through the westerlies (Christoudias et al, 2012).
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> Christoudias, T., Pozzer, A., and Lelieveld, J.: Influence of the North Atlantic Oscillation on air pollution transport. Atmos. Chem. Phys., 12, 869-877, doi:10.5194/acp-12-869-2012, 2012.

12. L81: Names of specific locations instead of 'over there'.

→ The sentence has been modified.

> The Expérience sur Site pour Contraindre les Modèles de Pollution atmosphérique et de Transport d'Emission (ESCOMPTE) campaign (June-July 2001) aimed to characterize the summer time pollution events in the vicinity of Marseille, France (Cros et al., 2004).

13. L97-98: What are the challenges to measure and simulate long-lived species in the troposphere specifically?

We removed this sentence because the difficulty of measuring and modelling long-lived species strongly depends on the considered greenhouse gas: N_2O, CO_2 or CH_4. Since our article mainly focus on CH_4, we present in the next paragraphs all the spaceborne sensors and some models than can potentially give an insight in the variability of CH_4 above the Mediterranean Basin.

14. L99-100: This statement is not entirely true. Recent measurements of CH4, N2O and CO2 from HIPPO show large hemispheric asymmetries in their global distributions (Wofsy et al., 2011).

→ Yes, you are right. To avoid any confusion between the different molecules and the aim of the present paper, we have removed this sentence (see point 13).

15. L118: Table 1 summarizes…Also references to each instrument can be added to Table 1.

→ Done.

Platform	**Instrument**	**Operation time**	**Wavelength**	**References**
ADEOS-1	IMG	1996-1997	TIR	Clerbaux et al. (1998)
ENVISAT	SCIAMACHY	2002-2012	NIR	Buchwitz et al. (2000)
Aura	TES	2004-date	TIR	Worden et al. (2012)
GOSAT	TANSO-FTS	2008-date	SWIR & TIR	Yokoto et al. (2009)
Aqua	AIRS	2004-date	TIR	Xiong et al. (2008)
MetOp-A	IASI	2008-date	TIR	Hilton et al. (2012)
MetOp-B	IASI	2012-date	TIR	
MetOp-C	IASI	Expected in 2016	TIR	

16. L136-139: References are needed here.

→ The incriminated sentence summarizes the points discussed in the previous sentence containing several references. We have modified the text accordingly.

> From these references, we note the impact of 1) the different meteorological regimes and 2) the seasonal variabilities of the emissions of atmospheric constituents, e.g. CO emitted from fires in summers, produces a seasonal variation in all the constituents.

17. L169-170: The datasets -> Datasets, The satellite data -> Satellite Data.

→ Done.

18. L171-173: This sentence can be rewritten, something like, 'Our study uses CH4 measurements from there different sensors and only the pixels measured over the Mediterranean Sea are considered due to larger systematic biases over land.'

→ The text has been modified accordingly.

> Our study analyses CH_4 measurements from three different spaceborne TIR sensors (IASI, AIRS and GOSAT) and take into account only the pixels measured over the Mediterranean Sea due to the larger systematic biases over land.

19. L176-177: This sentence can be rewritten, something like, 'The amplitude of diurnal cycle is larger over land than over the sea.'

→ Done.

> The amplitude of diurnal cycle, and its spatial variability, is larger over land than over the sea.

20. L184-185: Thus by applying temporal and geographical averages…

→ Done

21. L322-324 (also L979): by (from) the NCEP/NCAR reanalyses…

→ Done

22. L480: What does 'broader vertical domain' mean?

→ Text has been modified.

> These results suggest that the difference in amplitude between satellite and model in the seasonal evolution of E-W may be due to: a) the comparison technique, the vertical resolution of the models is much better than the vertical resolution of the satellite observations (…).

23. L483-485: What is the supporting evidence for those statements?

→ We have clarified this point.

> c) regarding the processes in winter, since westerlies are mainly present over the MB in the mid-to-upper troposphere (Figs. 3 and 8), we may have too much and/or too rapidly CH_4 transported over the Mediterranean Sea to the East compared to the West, leading to a too smooth E-W gradient in the models compared to the measurements.

24. L499-501: I'm not sure how to interpret the meanings of three 'evolution' in one sentence.

→ We have rephrased the sentence.

> As stated in sections 3 and 4, interpreting the E–W CH_4 seasonal variation along the vertical requires to consider the distribution of CH_4 over the Asian continent because of the importance of long-range transport.

25. L538: What is the resolution of the trajectory model?

→ From the BADC trajectory service, we have used the ECMWF-ERA40 with a 2.5° x 2.5 ° horizontal grid. The text has been slightly clarified and reorganized (see point 26).

26. L544: Does the position of gravity center mean a maximum in probability distribution function (PDF)?

→ Yes indeed. The text has been modified accordingly.

> In order to analyze the climalogical impact of the AMA onto the EMB, we have calculated (Fig. 8) the climatological six-day back-trajectories from the point at 33° N, 35° E located in the EMB (red filled circle on Fig. 8) based on the British Atmospheric Data Centre (BADC) trajectory service (http://badc.nerc.ac.uk/community/trajectory/) from 1st July to 31st August (summer convective period) from 2001 to 2010 every 12 hours at 5 different pressure levels: 850 and 700 hPa (lower troposphere), 500 hPa (middle troposphere), and 300 and 200 hPa (upper troposphere). The BADC trajectories were derived from 40-year (ERA40) re-analysis (2.5°x2.5°/pressure levels) produced by the European Centre for Medium-Range Weather Forecasts (ECMWF). The position of the gravity centre of each distribution (i.e. the maximum in the probability distribution function) at each level is represented every 24 hours by a star on Figure 8. This methodology has been firstly used over the Dome C (Concordia) station in Antarctica (Ricaud, 2014).

27. L612: consistent with each other.

→ Done.

28. Figure 12: This is a very nice diagram. However, I am not sure if the relative size and magnitude of the low -pressure center at the surface and the high-pressure system in the upper troposphere are the same. I think the upper tropospheric anticyclonic center has to be larger than the cyclonic center at the surface.

→ We agree, and the figure has been modified accordingly (see below).