Dr Claire Reeves
ACP Editor

Dr Philippe Ricaud
Météo-France
GAME/CNRS UMR 3589
42, Avenue G. Coriolis
31057 Toulouse
France
Phone: +33-5 61 07 96 09
Fax: +33-5 61 07 96 10
Email : philippe.ricaud@meteo.fr

Object: Revision of the manuscript acp-2014-235

Toulouse, 29 July 2014

Dear Dr Reeves,

Please find attached the revised 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 the reviewers.

Firstly, we would like to thank the two anonymous reviewers for their very fruitful comments, together with the comments from Dr Georgoulias. We have replied in detail below to all the questions and comments. We have considerably modified the manuscript in order to clarify the motivations and the main outcomes of the study. We hope the revised manuscript meets the high scientific standard of the ACP journal to be accepted for publication.

Secondly, in order to reply to several comments related to the motivations of our study, we have modified the title to explicitly focus on the processes studied. The title has thus 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 reviewers

Version 14, 29 July 2014

Firstly, we would like to thank the two anonymous reviewers for their very fruitful comments, together with the comments from Dr Georgoulias. We have replied in detail below to all the questions and comments. We have considerably modified the manuscript in order to clarify the motivations and the main outcomes of the study. We hope the manuscript meets the high scientific standard of the ACP journal to be accepted for publication.

Secondly, in order to reply to several comments related to the motivations of our study, we have modified the title to explicitly focus on the processes studied. The title has thus 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.

Short Comments from A. K. Georgoulias

ageor@auth.gr

Received and published: 17 April 2014

Since this interesting study is focused on the region of Mediterranean Basin and the authors do part of their analysis separately for the Western and Eastern part, I suggest that they should include the following paper in their citation list. To our knowledge this is the only paper dealing with tropospheric methane from satellites in the region.

Georgoulias, A.K., Kourtidis, K.A., Buchwitz, M., Schneising, O., Burrows, J.P.: A case study on the application of SCIAMACHY satellite methane measurements for regional studies: the Greater Area of Eastern Mediterranean, Int. J. Remote Sens., 32(3), 787-813, doi:10.1080/01431161.2010.517791, 2011.

→ We were not aware about this paper. We thank Dr Georgoulias to send a short comment relevant to our discussion. The reference of the paper has indeed been inserted in the revised manuscript for three main reasons.

a) Indeed, this paper deals with tropospheric methane in the region of the Mediterranean Basin and a sentence has been inserted in the introduction.

> Total columns of CH_4 as measured by SCIAMACHY over land and the Eastern Mediterranean from 2003 to 2004 show latitudinal and seasonal variations that cannot be attributed to volcano eruptions (Georgoulias et al., 2011).

b) The paper is also based on the SCIAMACHY measurements of CH_4 in the NIR domain above land. Thus, we have considered a sentence related to the capabilities of the NIR measurements, compared to the SWIR and TIR measurements (see replies to the reviewer #1's comments).

c) This paper finally presents some interesting results on CH_4 from space in the vicinity of the Mediterranean Sea, but only over land and essentially over the East of the Mediterranean. The paper states that the seasonal evolution of the total columns of CH_4 as measured by SCIAMACHY in 2003 and 2004 has an obvious maximum in August above the Greater Area of the Eastern Mediterranean. The authors do not deal with the interpretation of this maximum, that at least cannot be attributable to any eruptions from mud volcanoes. Being given that the sensitivity of the SCIAMACHY CH_4 total columns covers the vertical domain 1000-200 hPa from the vertical structure of the averaging kernels presented in Buchwitz et al. (2005), we note that 1) this maximum localized in August is consistent with our study, and 2) the impact of the Asian Monsoon Anticyclone on the CH_4 fields in the mid-to-upper troposphere cannot be ruled out. This point has been underlined in the discussion section.

One new paragraph has been inserted in the discussion section together with the reference to Buchwitz et al. (2005) and to Georgoulias et al. (2011).

> Finally, Georgoulias et al. (2011) present some interesting results of CH_4 from space in the vicinity of the Mediterranean Sea, but only over land and essentially over the Eastern Mediterranean. The authors found, from the total columns of CH_4 as measured by

SCIAMACHY in 2003 and 2004, an obvious maximum in August that could not be attributed to any volcano eruptions although this area hosts a significant number of geological formations that could potentially contribute to the total CH_4 burden. Being given that the sensitivity of the SCIAMACHY CH_4 total columns covers the vertical domain 1000-200 hPa from the vertical structure of the averaging kernels presented in Buchwitz et al. (2005), we note that 1) this maximum localized in August is consistent with our study, and 2) the impact of the AMA on the CH_4 fields in the mid-to-upper troposphere cannot be ruled out.

Buchwitz, M., de Beek, R., Burrows, J. P., Bovensmann, H., Warneke, T., Notholt, J., Meirink, J. F., Goede, A. P. H., Bergamaschi, P., Körner, S., Heimann, M., and Schulz, A.: Atmospheric methane and carbon dioxide from SCIAMACHY satellite data: initial comparison with chemistry and transport models, Atmos. Chem. Phys., 5, 941-962, doi:10.5194/acp-5-941-2005, 2005.

Georgoulias, A.K., Kourtidis, K.A., Buchwitz, M., Schneising, O., Burrows, J.P.: A case study on the application of SCIAMACHY satellite methane measurements for regional studies: the Greater Area of Eastern Mediterranean, Int. J. Remote Sens., 32(3), 787-813, doi:10.1080/01431161.2010.517791, 2011.

In addition, it would be nice if the authors added in their discussion a number of ground-based studies that were conducted in the area. It has to be highlighted that the greater Mediterranean area hosts a significant number of geological formations that could potentially contribute to the total methane burden.

→ This point has been dealt in point c) above.

Anonymous Referee #1

Received and published: 11 May 2014

General comments:

1. The authors mentioned:

a. The IASI methane profiles have not been validated and therefore, not operational (p.9983, l.29)
b. The number of daily total column of methane is highly variable.
c. The different height of maximum sensitivity: IASI at 8 km (p.9983, l. 26); AIRS at 9-12 km (p. 9984, l. 23).

The authors should comment on these 3 issues and their respective influence on the results obtained.

→ Below are the replies regarding the three points.

a) As we state in the manuscript (p. 9984, l. 1), "the methane products [for IASI] are experimental products, routinely generated for demonstration and evaluation." It is one of the outcomes of the present paper to point out the quality of this satellite product, since the seasonal variation of the East-West difference in total columns of CH_4 from IASI as delivered by EUMETSAT is consistent with theoretical results. We have inserted a sentence in that direction in the conclusions.

> Despite the fact that IASI CH_4 data are not operational, the seasonal variation of the East-West difference in total columns of CH_4 from IASI as delivered by EUMETSAT is consistent with theoretical results and measurements from AIRS and IASI.

b) We agree the number of daily total column of methane is highly variable for IASI. But we can also present the same argument for the vertical profiles of AIRS, together with TANSO-FTS on GOSAT. For that main reason we have considered monthly-averaged data into bins of 1°x1°. If we consider, over one year, the number of IASI pixels (total columns) used in each of the Western or Eastern box, we can find values ranging from 30,000 to 80,000 depending on the month considered, with a median value that can be approximated to 40,000 that is consistent with our statement p. 9995, l. 1. The values obtained for AIRS are approximately a factor 5 less than the ones obtained for AIRS. For TANSO-FTS on GOSAT, the number of pixels available in each of the boxes are much more reduced, by a factor 10 compared to IASI, mainly due to the weak quality of the calibrated spectra (L1B data) that has impacted on the quality of the vertical profiles (L2 data in version 0.1) for which only pixels with Degrees of Freedom of Signal greater than 0.6 were selected. Consequently, GOSAT monthly-averaged data appear to be slightly noisier in our analyses (see e.g. Fig. 8) compared to AIRS monthly-averaged data. This important point has been discussed in section 4.3 by inserting a new paragraph. Note that IASI total columns are not and cannot be directly compared with AIRS or GOSAT profiles in our analysis. We have thus inserted a new paragraph.

> We have also to remind that statistically the number of spaceborne measurements used in our analysis (see section 2) is ~5 times greater in IASI compared to AIRS, ~30 times greater in AIRS compared to GOSAT. Consequently, GOSAT monthly-averaged data appear noisier than AIRS monthly-averaged data. Note that IASI

> total columns are not and cannot be directly compared with AIRS or GOSAT profiles in our analysis. Nevertheless, although IASI data are not operationally produced, the IASI E-W seasonal variation is very consistent with the E-W seasonal variation as deduced from all other datasets. The monthly random error attributed to the E–W IASI CH_4 is about 0.1%, much less than the observed peak-to-peak yearly variation. We estimate that the AIRS monthly random error attributed to the E–W CH_4 is twice greater than the one calculated for IASI, and that the GOSAT monthly random error is about 5 times greater than the ones calculated for IASI. We discuss in the next section the origin of the summer peak in the E–W seasonal variation.

c) IASI, AIRS and TANSO-FTS on GOSAT are all instruments measuring CH_4 in the TIR domain, but not in the same bands. Basically, TIR measurement sensitivity is in the middle troposphere. In the literature, some pieces of information are given in order to have a broad idea of the vertical sensitivity of the measurements that depends on several key parameters mentioned in the manuscript (surface emissivity, surface temperature, thermal contrast at the surface), together with the location (latitude) of the pixel considered and the time of the day.

For GOSAT (Saitoh et al., 2012), the CH_4 averaging kernels peak at 10 km with a sensitivity, defined as the full-width at half-maximum of the averaging kernels, from 5 to 15 km. But this averaging kernel is evaluated for measurements performed in the tropical Pacific Ocean within a box 10°N-35°N and 140°E-150°E. For IASI (Razavi et al., 2009), the tropical CH_4 averaging kernels are centred at 10 km with a sensitivity from 5 to 15 km, consistently with GOSAT. At mid-latitudes, the CH_4 averaging kernels are centred at 8 km with a sensitivity from 4 to 14 km. For AIRS (Xiong et al., 2008), the tropical CH_4 averaging kernels are centred at 200 hPa (~11 km) with a sensitivity from 500 to 70 hPa, consistently with GOSAT and IASI. At mid-latitudes, the CH_4 averaging kernels are centred at 300 hPa (~9 km) with a sensitivity from 700 to 100 hPa, consistently with IASI.

In conclusion, the values attached to the vertical sensitivity of the three instruments at mid-latitudes are all consistent to each other. We have outlined this point in the revised manuscript.

In section 2.1, we have defined the vertical sensitivity of the TIR measurements as:

> Therefore, the vertical sensitivity of the TIR measurements, defined as the full-width at half-maximum of the averaging kernels from the optimal estimation method (Rodgers, 2000), over the sea is consistent during day and night and concentrated in the mid-troposphere.

In section 2.1.1, we have clarified the vertical sensitivity of IASI measurements.

> At mid-latitudes, the vertical sensitivity of the total column CH_4 is peaking in the mid-troposphere at ~8 km from 4 to 14 km (Razavi et al., 2009) and, in the tropics, at ~10 km from 5 to 15 km.

In section 2.1.2, we have clarified the vertical sensitivity of AIRS measurements.

> At mid-latitudes, the most sensitive layer of AIRS channels to CH_4 is at 300 hPa (~9 km) with a vertical sensitivity from 700 to 100

hPa (Xiong et al., 2008), and, in the tropics, at 200 hPa from 500 to 70 hPa consistently with the IASI TIR measurement sensitivity.

In section 2.1.3, we have clarified the vertical sensitivity of GOSAT TANSO-FTS TIR measurements.

The TIR measurements from Band 4 (5.5–4.3 μm) provide vertical profiles of CH_4 along 7 vertical levels (Imasu et al., 2007) by using the optimal estimation method with a vertical sensitivity in the tropics peaking at 10 km (higher than at mid-latitudes) from 5 to 15 km (Saitoh et al., 2012), consistently with the vertical sensitivity of IASI (Razavi et al., 2009) and AIRS (Xiong et al., 2008) in the tropics.

2.

a. The MOCAGE calculated profiles seem to be not consistent with the seasonally averaged profile obtained from AIRS and GOSAT for 2010 (Fig. 8) neither consistent with the modeled profiles for JJA 2009 (Fig. 3).

b. The calculated methane concentration differences between EMB and WMB by CNRM-AOCCM and LMDz-OR-INCA are not very consistent regardless of the IPCC scenarios (Fig. 10).

The manuscript would benefit much if the authors include a section while discussing these large discrepancies while explaining their possible reasons.

→ Below are the replies regarding the two points.

a) “The MOCAGE calculated profiles seem to be not consistent with the seasonally averaged profile obtained from AIRS and GOSAT for 2010 (Fig. 8).” This is related to the impact of the vertical resolution of the TIR measurements vs. model data. The model data resolution can be degraded by using the averaging kernels attached to the measurements. This is explained in detail in the reply to the point 27) of the reviewer #2’s comments.

“(…) neither consistent with the modeled profiles for JJA 2009 (Fig. 3).” We have carefully checked the Figures 1-8 (new Figures 1-6) since, as noted by the reviewer, differences were coming from the use of different runs of MOCAGE. They are now consistently produced considering the same run of MOCAGE. Compared to the previous version, Figure 6 (Figure 8 in the previous version) shows amounts of CH_4 in the lowermost troposphere from MOCAGE of about 1700-1740 ppbv, much less by 150-200 ppbv than the ones from GOSAT (and also from surface measurements (not shown) at Lampedusa, Italy and Negev Desert, Israel). Note that part of this point is also discussed in the replies to the reviewer#2’s point 27. Global models are known to underestimate mixing ratios of trace species largely due to coarse horizontal resolution and large uncertainties in estimated surface emission. Also note from new Fig. 10 that LMDz-OR-INCA surface CH_4 is about 1720-1750 ppbv in the Mediterranean, consistently with MOCAGE.

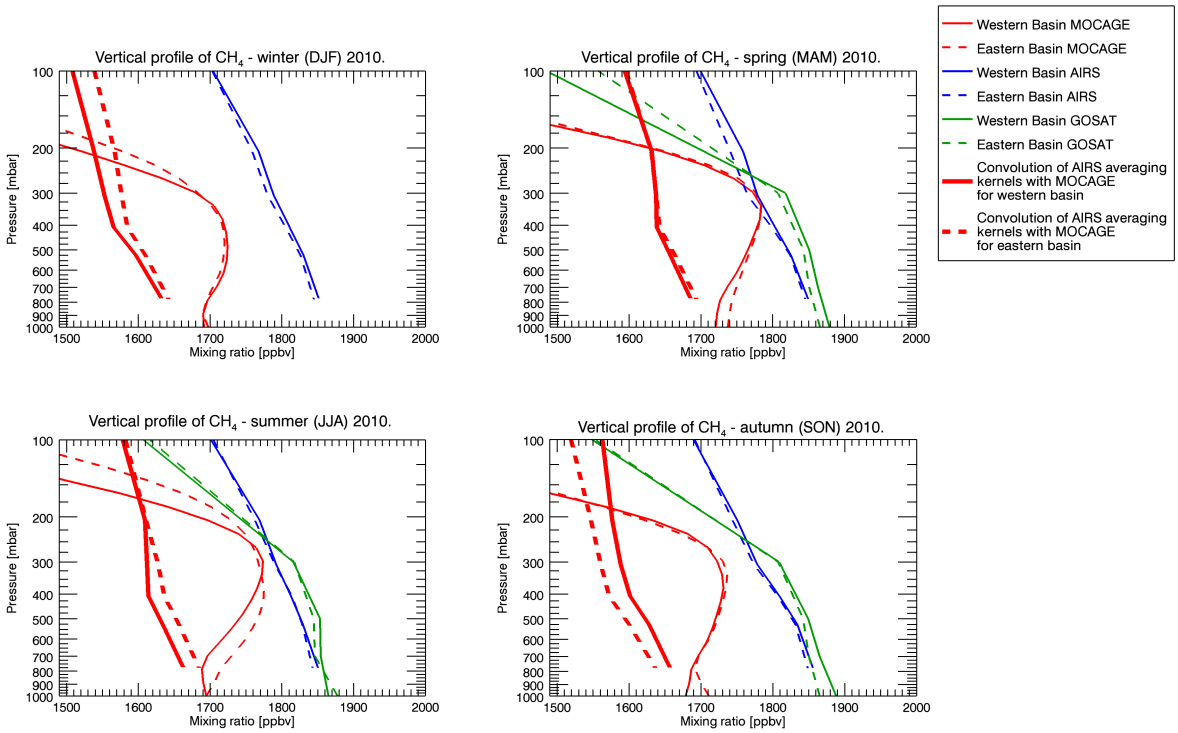


Figure 6. *(From top to bottom and from left to right) Seasonally-averaged vertical profiles of CH_4 as measured by AIRS (blue lines) and GOSAT (green lines), and as calculated by MOCAGE (thin red lines) over the Eastern (dashed lines) and Western (solid lines) MBs in winter, summer, spring and autumn 2010. Also shown are the seasonally-averaged MOCAGE profiles convolved with the AIRS averaging kernels (thick red lines) for the four seasons over the Eastern (dashed lines) and Western (solid lines) MBs.*

We have thus modified the section 4.2 by considering and discussing the CH_4 low bias between MOCAGE (and overall global models) and the satellite data. We have inserted a sentence relative to the low-to-mid vertical profiles of MOCAGE.

> Separately, whatever the season considered, the MOCAGE low-to-mid tropospheric CH_4 is low biased compared to the measured profiles by ~150-200 ppbv.

We have inserted a sentence relative to the surface CH_4 of MOCAGE.

> Near the surface, the amount of CH_4 is about 1700-1750 ppbv for MOCAGE, and is on average less than the CH_4 GOSAT data by about 150-200 ppbv. (…) Consequently, the amount of surface CH_4 in the MOCAGE run for 2010 is actually low biased by about 150-200 ppbv (8-10%) but is very consistent with the LMDz-OR-INCA surface data of ~1725-1750 ppbv over the Mediterranean (Fig. 10).

And we have discussed the reasons why the model CH_4 is less than observations.

> Convolved MOCAGE CH_4 profiles are now consistent with AIRS CH_4 profiles whatever the season considered but a systematic low bias of ~150-200 ppbv (8-10%) between AIRS and MOCAGE convolved

profiles is observed. This might be due to the fact that no a priori information contributes to the convolved profile. This is also due to the overall underestimation of CH_4 by global models. Indeed, due to coarse horizontal resolution and large uncertainties in the estimated surface emissions, tropospheric CH_4 lifetimes, e.g. evaluated by the multi-model intercomparison project ACCMIP, are about 5-13% lower than observation estimates (Naik et al., 2013; Voulgarakis et al., 2013).

We have added two references.

Naik, V., Voulgarakis, A., Fiore, A. M., Horowitz, L. W., Lamarque, J.-F., Lin, M., Prather, M. J., Young, P. J., Bergmann, D., Cameron-Smith, P. J., Cionni, I., Collins, W. J., Dalsøren, S. B., Doherty, R., Eyring, V., Faluvegi, G., Folberth, G. A., Josse, B., Lee, Y. H., MacKenzie, I. A., Nagashima, T., van Noije, T. P. C., Plummer, D. A., Righi, M., Rumbold, S. T., Skeie, R., Shindell, D. T., Stevenson, D. S., Strode, S., Sudo, K., Szopa, S., and Zeng, G.: Preindustrial to present-day changes in tropospheric hydroxyl radical and methane lifetime from the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP), Atmos. Chem. Phys., 13, 5277-5298, doi:10.5194/acp-13-5277-2013, 2013.

Voulgarakis, A., Naik, V., Lamarque, J.-F., Shindell, D. T., Young, P. J., Prather, M. J., Wild, O., Field, R. D., Bergmann, D., Cameron-Smith, P., Cionni, I., Collins, W. J., Dalsøren, S. B., Doherty, R. M., Eyring, V., Faluvegi, G., Folberth, G. A., Horowitz, L. W., Josse, B., MacKenzie, I. A., Nagashima, T., Plummer, D. A., Righi, M., Rumbold, S. T., Stevenson, D. S., Strode, S. A., Sudo, K., Szopa, S., and Zeng, G.: Analysis of present day and future OH and methane lifetime in the ACCMIP simulations, Atmos. Chem. Phys., 13, 2563-2587, doi:10.5194/acp-13-2563-2013, 2013.

b) Discussion of the results shown in Fig. 10 (new Fig. 9).

We have detailed the interpretation of the differences seen by the models in the CH_4 E-W seasonal variation regarding the different IPCC scenarios, focussing on the layers 200 and 100 hPa.

It is not obvious to understand why the E-W seasonal variation at 200 hPa is positive in summer for RCP 4.5 and not for the other RCPs (except RCP 8.5 in August). The horizontal distribution of CH_4 calculated by the two models at 200 hPa (Fig. 11) drastically differs but local maxima are centred within the AMA. A zonally-symmetric structure showing a strong South-North gradient in CH_4 is modelled by CNRM-AOCCM with maxima in the tropics (1800 ppbv) and minima at high latitudes (1700 ppbv) and a local maximum centred within the core of the AMA with values greater than 1807 ppbv elongated towards two axis: 1) South-East Asia and 2) Middle East and EMB. The CH_4 field calculated by LMDz-OR-INCA considering the 4 scenarios also shows two maxima over Northern India and over North-East Asia but the horizontal distribution is not zonally-symmetric due to a zonally-asymmetric CH_4 surface field. In all the scenarios considered, the CH_4 maxima within the AMA range from 1710

to 1750 ppbv with increasing RCPs from 2.6 to 8.5. An elongated tongue of enriched CH_4 enters the EMB. More precisely, we can argue that in RCPs 2.6, 6.0 and 8.5, the primary maximum of CH_4 is located northward at 50°N, 135°E (CH_4 values greater than 1720, 1730 and 1750 ppbv, respectively) although it is a secondary maximum in RCP 4.5 (CH_4 values less than 1720 ppbv). Through long-range transport, this mid-latitude maximum is transported Eastward within a band 40°N-50°N enriching CH_4 in the WMB and producing a E-W minimum in summer for RCPs 2.6, 6.0 and 8.5. Since there is a North-South gradient with a maximum in the South for CNRM-AOCCM, CH_4-depleted air masses reach the WMB although CH_4-enriched air masses from the AMA reach the EMB producing a systematic peak in summer, consistently with RCP 4.5.

In the lower stratosphere (100 hPa, Fig. 9), all the model outputs are consistent to each other showing an annual oscillation, with a wide maximum in summer (60-80 ppbv) and a wide minimum in winter (20-35 ppbv). This is apparently surprising keeping in mind that both models significantly differ from the surface (see Fig. 10) to ~500 hPa. But, in the WMB, the 100-hPa pressure corresponds to 420-K potential temperature both in summer (Fig. 2) and in winter (Fig. 4) whilst, in the EMB, it corresponds to 390 K in summer and 400 K in winter, namely closer to the tropopause in summer than in winter. Consequently, whatever the model considered, the E-W CH_4 seasonal variation at 100 hPa a) is always positive and b) shows a peak in the summer period. We note that the summer peak in E–W seasonal evolution from the middle to the upper troposphere has also been observed and calculated by considering other constituents like CO and O_3 (not shown). This is the main topic of a forthcoming paper.

Specific comments:

1) p. 9979, l.3-24: I strongly suggest to insert a Table which includes all these details on the several platforms, their time of operations and the measured species.

→ A new Table 1 has been inserted according to the reviewer's comments.

Table 1. Nadir-viewing instruments having the capabilities to actually measure long-lived species in the troposphere. Please, refer to the text for the acronyms.

Platform	**Instrument**	**Operation time**	**Wavelength**
ADEOS-1	IMG	1996-1997	TIR
ENVISAT	SCIAMACHY	2002-2012	NIR
Aura	TES	2004-date	TIR
GOSAT	TANSO-FTS	2008-date	SWIR & TIR
AIRS	Aqua	2004-date	TIR
MetOp-A	IASI	2008-date	TIR
MetOp-B	IASI	2012-date	TIR
MetOp-C	IASI	Expected in 2016	TIR

2) p. 9979, l. 23-28: The authors are encouraged to describe briefly the benefits and drawbacks of other measurements as done for TIR and SWIR channels.

→ We have added a sentence related to the capabilities of the NIR measurements, essentially over land.

> In the NIR, analyses are essentially restricted to areas over land because the retrievals over sea are considered less reliable due to fairly low surface albedo of water, which results in low signals thus low signal-to-noise ratios (Georgoulias et al., 2011).

Reference to Georgoulias et al. (2011) already inserted (see Short Comments from Georgoulias).

3) p. 9982, l. 23-25: The location of the statement on NCEP/NCAR Reanalysis is not appropriate, pls place it in another place along the text.

→ We indeed moved this statement to P. 9989 L. 14, and re-wrote a new sentence as:

> On Figures 2 and 4, the MOCAGE CH_4 fields are superimposed with 1) the wind fields from ARPEGE analyses and 2) the cold point tropopause pressure fields provided by the National Oceanic and Atmospheric Administration (NOAA) National Centers for Environmental Prediction (NCEP)/National Center for Atmospheric Research (NCAR) reanalyses, all these data being averaged over the same period.

4) p. 9991: Correction of the title of Section 4.1 instead of “Global” I suggest the following title : “Methane spatial distribution over the MB”

→ Done.

5) p. 9989, l.1-15: This part of the text should be moved to p. 9990 after l. 7.

→ Since we develop our discussion of the atmospheric conditions controlling the CH_4 spatial distribution on both, Figs. 1-6 and already published literature, we absolutely need to present first the Figures. So we did not change the order of the paragraphs as proposed by the reviewer.

6) p.9990, l. 18-20: The interpretation here is wrong. The subsidence is caused mainly by the Subtropical High positioned over the EM rather than the cooler SST as compared to the land surface temperature.

→ We agree with the reviewer that the strong subsidence observed over the MB is not due to the surface temperature difference between the sea and the continents. We have rewritten the entire sentence.

> A systematic subsidence is present over the MB (Fig. 2) whatever the longitudinal bin considered due to the presence of semi-permanent subtropical high pressure systems which are centred over the tropical deserts. More precisely, in the WMB, the descent is caused by the presence of a high pressure cell (Fig. 1) whilst, in the EMB, it is coming from the Hadley cell that is further displaced over the Northern Africa producing a downward branch in the area 30°N-35°N.

7) p. 9989: The title of Section 3 is too vague and recommended to be changed by: “Atmospheric conditions controlling the spatial distribution of methane”

→ Done.

8) p. 9994: I suggest to replace the title of Section 4.3 to: “ The east-west seasonal variations -measured and calculated differences”.

→ Done.

9) Replacing some parts of the text. Please add the text from p. 9995 l. 10-28 to p. 9996 l. 1-23 to Section 4.3.

→ Done.

10) p.9996 l. 23: I strongly suggest changing the title “Discussion” to “Contribution of the Asian Monsoon Anticyclone” before l. 23.

→ Done

11) p.9999 l. 18-26: The text in these lines was already mentioned. Please omit it.

→ Text removed.

12) p. 9999 l. 4: Please change to :” assess the spatial variability of methane over the EMB and to attribute the variability to differing synoptic and global scales..”

→ Done.

13) p. 9981 l. 8: Please change to : “ attribute the variability to different processes at both, synoptic and global scales..”

→ Done.

Anonymous Referee #2

This study presents results of a comprehensive analysis of atmospheric methane distributions over the Mediterranean Basin in the troposphere using both satellite measurements and model simulations. Multiple instruments with varying measurement technique as well as global chemistry transport model and chemical climate models are utilized in the analyses. I found the contents of this study fairly presented and the general subject of this work has scientific significance. However, the overall structure of the paper seems to be rather poorly constructed. Below are my comments for the authors may take into consideration for improving clarity of this manuscript.

General Comments:

1.What is the motivation of this work? Apart from the satellite retrieval issues, why the Mediterranean Basin is important? Why are you looking at methane total column data from IASI and methane profiles from AIRS? Why did you include GOSAT data even though there are only few good measurements available? Why are you using model outputs from three different models? Is the purpose of this work to present model intercomparison? The selection of all the method and data used in the study has to be justified, preferentially in the introduction.

→ A detailed response is presented below.

The introduction has been more focussed on the Mediterranean Basin underlining:

a) the results already obtained and presented in literature considering CH_4 but also other constituents and aerosols (including new references to Lionello, 2012; Giorgi and Lionello, 2008; Cros et al., 2004; Ladstätter-Weißenmayer et al.,, 2003; Scheeren et al., 2003):

> The Mediterranean Basin (MB) is located in a transitional zone between subtropical and mid-latitudes regimes (Lionello, 2012), highly sensitive to climate change. To illustrate, simulations tend to show a pronounced decrease in precipitation (2000-2100), especially in the warm season (Giorgi and Lionello, 2008), and Lionello (2012) reported on an observed summer West-East dissymmetry in precipitation (1979-2002). In terms of anthropogenic pollution sources, the MB is at the confluence of three continents, Europe, Africa and Asia. 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) or forest fires (e.g. South East of France, Corsica, Portugal, and Greece) is still not well understood, especially on the O_3 and CO budgets. Besides these regional sources, polluted air masses may originate from Asia during the summer monsoon period, Africa through the Hadley cell and upper level anticyclone and North America through the westerlies. The "Expérience sur Site pour COntraindre les Modèles de Pollution atmosphérique et de Transport d'Emissions" (ESCOMPTE) campaign (June-July 2001) in southeastern France aimed to characterize the summer time pollution events over there (Cros et al., 2004). The goal of the Mediterranean Intensive Oxidant Study (MINOS) campaign (July-August 2001) in the eastern Mediterranean was to measure long-range transport of air pollution and aerosols from South East Asia and Europe towards the MB (Ladstätter-Weißenmayer et al.,,

2003; Scheeren et al., 2003). They have demonstrated the importance of coastal and synoptic transport mechanisms on the variability of constituents but were not adapted to assess the budgets of O_3, CO and long-lived species.

Cros, B., Durand, P., and Cachier, H.: An overview of the ESCOMPTE campaign, Atmos. Res, 69(3-4), 241-279, 2004.

Giorgi, F., and Lionello, P.: Climate change projections for the Mediterranean region, Global and Planetary Change, 63(2), 90–104, doi:10.1016/j.gloplacha.2007.09.005, 2008.

Ladstätter-Weißenmayer, A., Heland, J., Kormann, R., von Kuhlmann, R., Lawrence, M. G., Meyer-Arnek, J., Richter, A., Wittrock, F., Ziereis, H., and Burrows, J.-P.: Transport and build-up of tropospheric trace gases during the MINOS campaign: comparison of GOME, in situ aircraft measurements and MATCH-MPIC-data, Atmos. Chem. Phys., 3, 1887–1902, 2003.

Lionello, P. (Ed.): The Climate of the Mediterranean Region: From the past to the future. 592p, Elsevier, 2012.

Scheeren, H. A., Lelieveld, J., Roelofs, G. J., Williams, J., Fischer, H., de Reus, M., de Gouw, J. A., Bolder, M., van der Veen, C., and Lawrence, M.: The impact of monsoon outflow from India and Southeast Asia in the upper troposphere over the eastern Mediterranean, Atmos. Chem. Phys., 3, 1589–1608, 2003.

and b) the motivation of the present work:

> The aim of the present paper is to assess the variability of CH_4 in the mid-to-upper troposphere between the East and the West of the Mediterranean Basin and to attribute the seasonal variability of the East-West gradient to different processes at both, synoptic and global scales depending on the season and the altitude layer considered. We will study in detail the impact of the summer-time long-range transport of CH_4 from Asia to the Eastern MB through the Asian Monsoon Anticyclone. Since we have already underlined that measurement and modeling of the tropospheric CH_4 distribution are challenging, we will adopt a climatological approach and will use a wide variety of space-borne measurements and model outputs to verify that they give consistent results.

In order to support the general description of the processes operating during the summer season on a global scale and affecting the CH_4 distribution in the Eastern Mediterranean, we have draw a schematic Figure (Figure 12) that represents the different processes. (1) Trapping of lower tropospheric pollutants in the Asian monsoon. (2) Updraft of pollutants in the Asian monsoon up to the upper troposphere. (3) Build-up of pollutants within the Asian monsoon in the upper troposphere. (4) Large-scale re-distribution of pollutants by the Asian Monsoon Anticyclone to the Middle East and North Africa in the upper troposphere. (5) Build-up of pollutants through descent down to the middle troposphere above the Eastern Mediterranean Basin. This Figure has been inserted in the discussion, section 5.

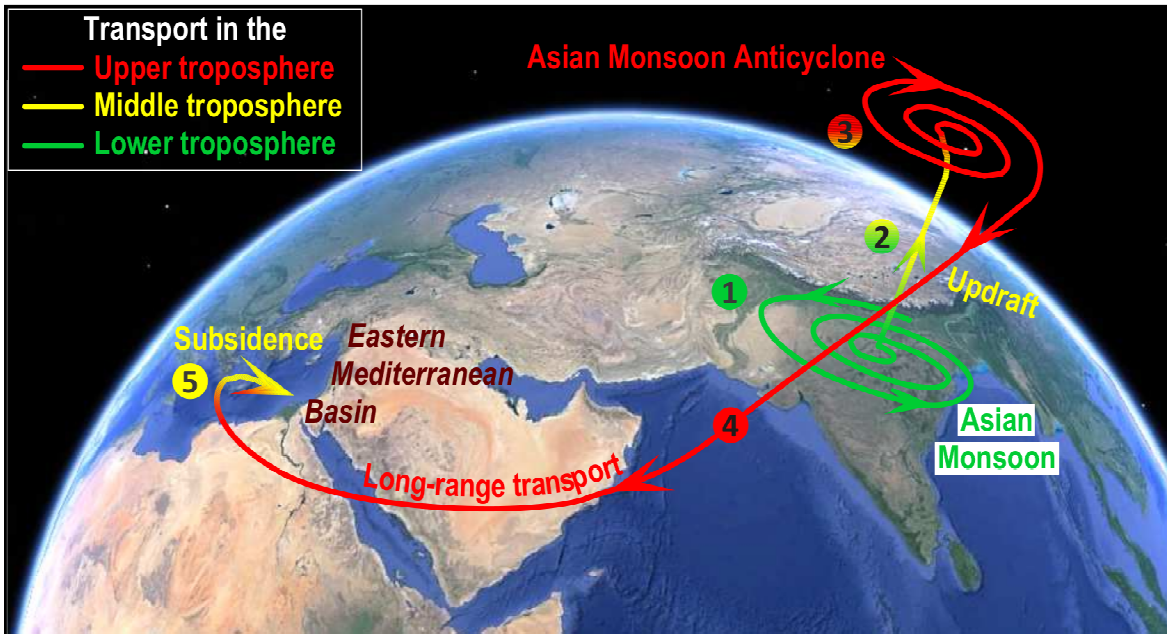


Figure 12. *Schematic representation of the processes impacting the mid-to-upper tropospheric pollutants, including CH_4, above the Eastern Mediterranean Basin in summer (July-August). (1) Trapping of lower tropospheric pollutants in the Asian monsoon. (2) Updraft of pollutants in the Asian monsoon up to the upper troposphere. (3) Build-up of pollutants within the Asian monsoon in the upper troposphere. (4) Large-scale re-distribution of pollutants by the Asian Monsoon Anticyclone to the Middle East and North Africa in the upper troposphere. (5) Build-up of pollutants though descent down to the middle troposphere above the Eastern Mediterranean Basin.*

We have also modified the title to explicitly focus on the processes studied. The title has thus 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

2.What is the main goal of this study? The authors seemed to have their main focus on the satellites and model description rather than new findings about methane climatology and transport. If the goal of this paper is to describe the data and the model, there is not much exciting science to be claimed. If the authors' intention was to focus on the methane climatology, the overall structure of this paper has to be reconsidered.

→ See point 1. above and detailed description of the main goal of the study.

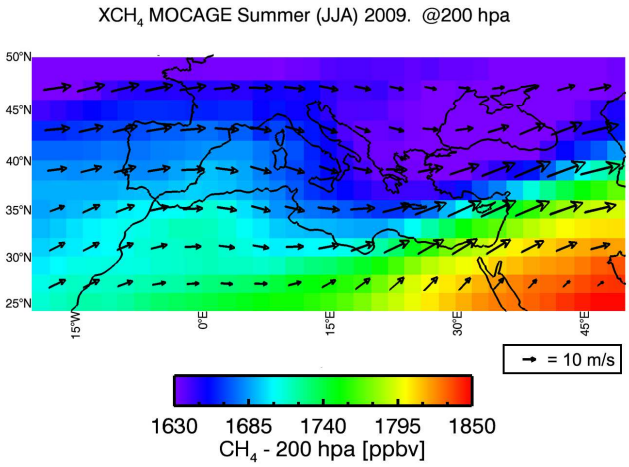
3.The background of methane climatology, seasonal variability in the troposphere including what has been done or what has not been done (e.g., previous literature), why measuring methane from space is important but difficult has to be clearly mentioned. Each figure containing methane distribution has to

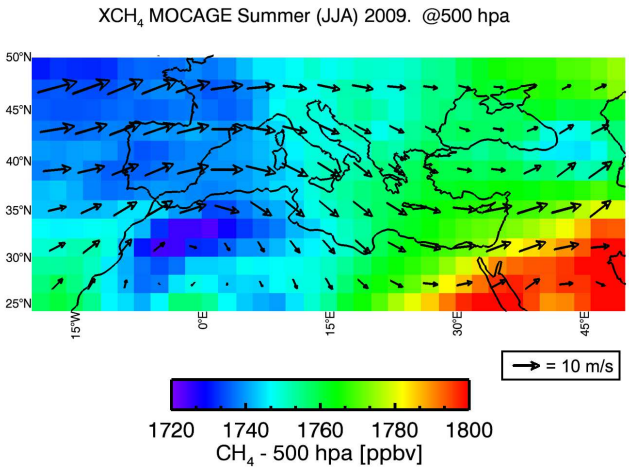
have its own clear point, separately. In my opinion, section 3 seems to be the most important part of the paper but the inclusion of the figures are all lumped together, which makes it hard to follow the authors' explanation.

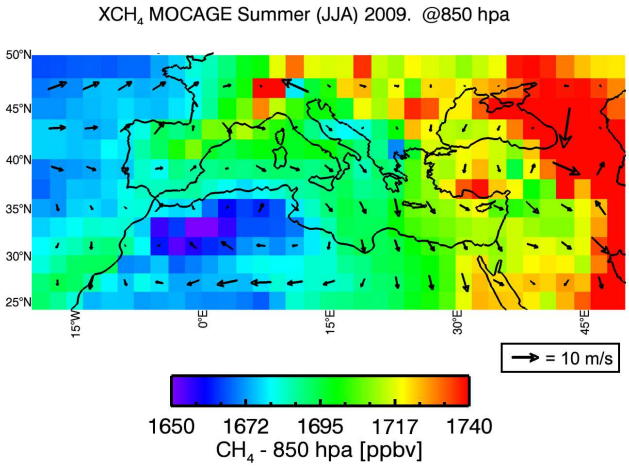
→ We have removed Figures 3 and 6 (north-south transect at different longitudes and two seasons) that did not bring too much information in order to focus on our main topic, the impact of the AMA on the mid-to-upper tropospheric CH_4 in the EMB in summer. Sections 4 and 5 are now as important as section 3 since the long-range transport is discussed in detail.

4.There seems to be lack of supporting evidence or explanation showing strong connection between methane distributions and meteorology (transport). I recommend the authors only include the wind fields when they are needed and showing clear correlation with the tracer fields. For example, Fig. 1 is a very busy plot with many arrows. Either removing the horizontal grid lines or change the color of them to gray would make the arrow look more dominant.

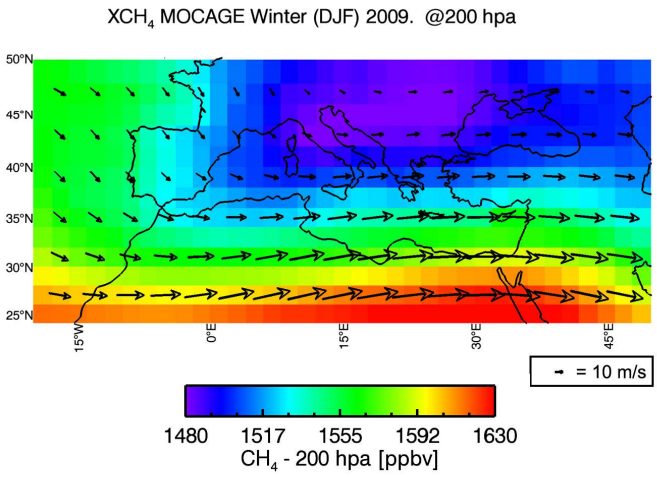
→ Firstly, we have modified Figures 1 and 4 (new Figures 1 and 3) by removing horizontal and vertical grid lines. We carefully checked that the presence of the wind arrows did not overload the incriminated Figures. We have removed from the revised manuscript the Figures 3 and 6 that did not bring new information.

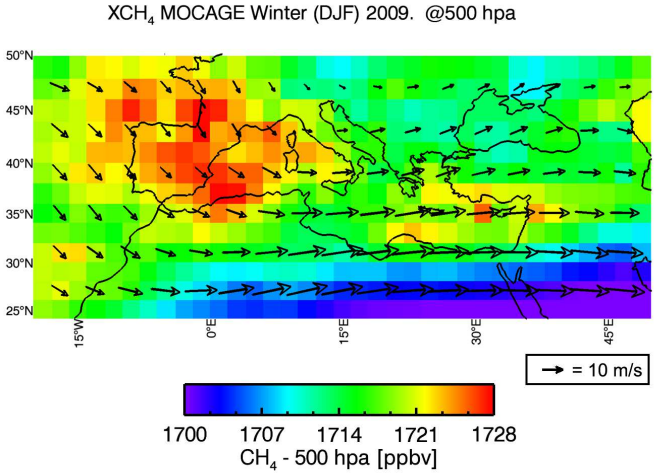

XCH4 MOCAGE Summer (JJA) 2009. @200 hpa
50°N
45°N
40°N
35°N
30°N
25°N
15°W
0°E
15°E
30°E
45°E
= 10 m/s
1630 1685 1740 1795 1850
CH4 - 200 hpa [ppbv]

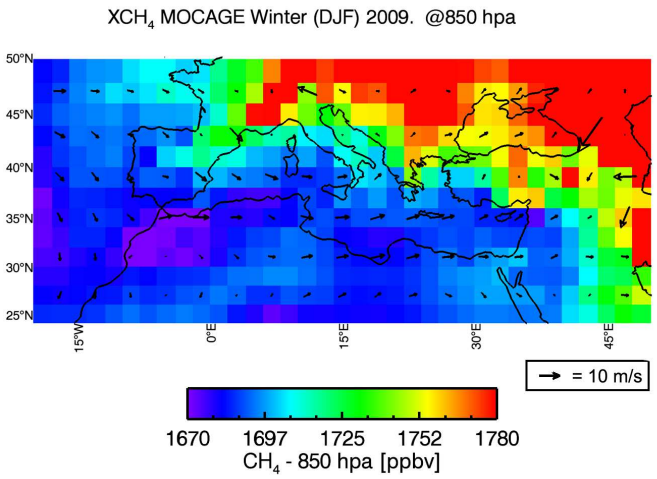

XCH4 MOCAGE Summer (JJA) 2009. @500 hpa
50°N
45°N
40°N
35°N
30°N
25°N
15°W
0°E
15°E
30°E
45°E
= 10 m/s
1720 1740 1760 1780 1800
CH4 - 500 hpa [ppbv]


XCH4 MOCAGE Summer (JJA) 2009. @850 hpa
50°N
45°N
40°N
35°N
30°N
25°N
15°W
0°E
15°E
30°E
45°E
= 10 m/s
1650 1672 1695 1717 1740
CH4 - 850 hpa [ppbv]

New Figure 1.


XCH4 MOCAGE Winter (DJF) 2009. @200 hpa
50°N
45°N
40°N
35°N
30°N
25°N
15°W
0°E
15°E
30°E
45°E
= 10 m/s
1480 1517 1555 1592 1630
CH4 - 200 hpa [ppbv]


XCH4 MOCAGE Winter (DJF) 2009. @500 hpa
50°N
45°N
40°N
35°N
30°N
25°N
15°W
0°E
15°E
30°E
45°E
= 10 m/s
1700 1707 1714 1721 1728
CH4 - 500 hpa [ppbv]


XCH4 MOCAGE Winter (DJF) 2009. @850 hpa
50°N
45°N
40°N
35°N
30°N
25°N
15°W
0°E
15°E
30°E
45°E
= 10 m/s
1670 1697 1725 1752 1780
CH4 - 850 hpa [ppbv]

New Figure 3.

→ Secondly, in order to deal with the comment related to "lack of supporting evidence or explanation showing strong connection between methane distributions and meteorology (transport)", we have performed back-trajectory calculations over a long time period (10 years) in order to study the origin of air masses reaching the Eastern Mediterranean Basin according to the season and the pressure level considered.

We have thus inserted a new Figure 8 corresponding to the climatological six-day back-trajectories from the point at 33° N, 35° E located in the Eastern Mediterranean Basin (red filled circle) calculated from the British Atmospheric Data Centre trajectory service (http://badc.nerc.ac.uk/community/trajectory/) from 1st July to 31st August from 2001 to 2010 every 12 hours at 850 (red line), 700 (orange line), 500 (green line), 300 (blue line) and 200 hPa (yellow line). The position of the gravity centre of each distribution at each level is represented every 24 hours by a star. Data from ECMWF archive (2.5 degree/pressure levels) are used in the calculation.

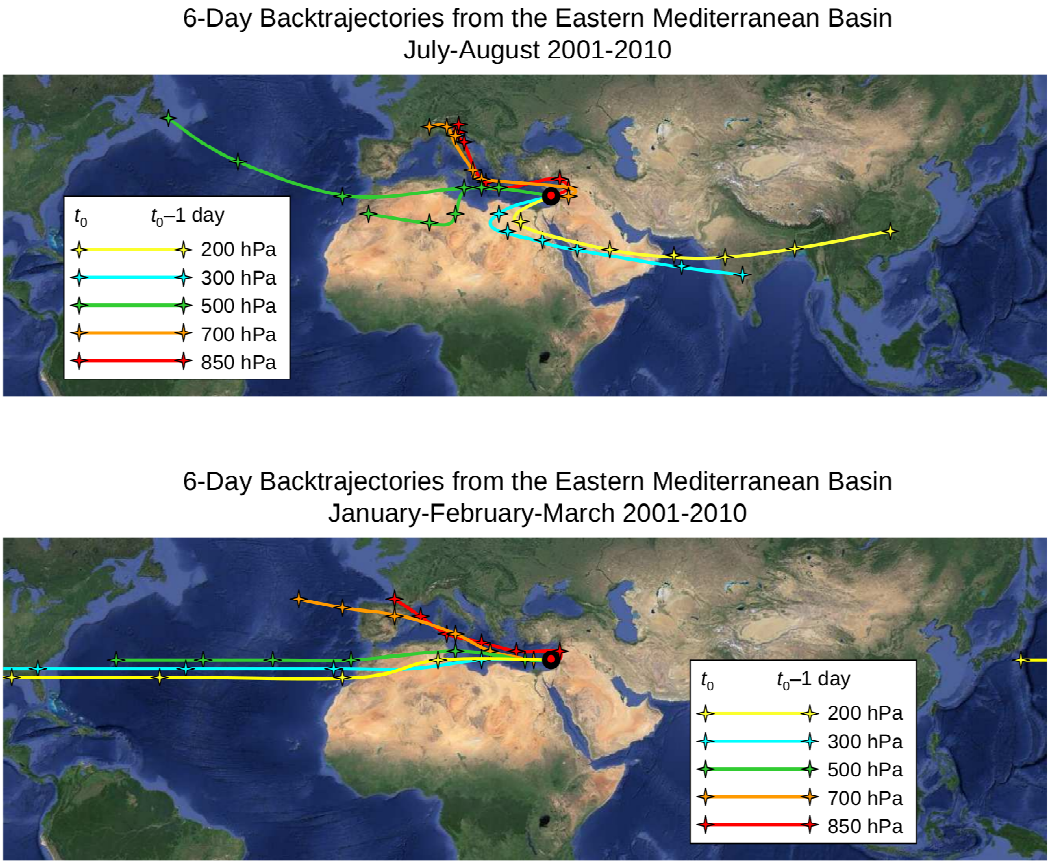


Figure 8. *(Top) Climatological six-day back-trajectories from the point at 33° N, 35° E located in the Eastern Mediterranean Basin (red filled circle) calculated from the British Atmospheric Data Centre trajectory service (http://badc.nerc.ac.uk/community/trajectory/) from 1st July to 31st August from 2001 to 2010 every 12 hours at 850 (red line), 700 (orange line), 500 (green line), 300 (blue line) and 200 hPa (yellow line). The position of the gravity center of each distribution at each level is*

represented every 24 hours by a star. (Bottom) Same as top, but calculated from 1st January to 31st March 2001-2010.

This new Figure is a synthesis of the back-trajectory distributions of the position of the air masses from the point at 33° N, 35° E located in the Eastern Mediterranean Basin (red filled circle) calculated by the BADC trajectory service at the considered 6 pressure levels from July-August 2001-2010. Examples for the pressure levels of 200, 500 and 850 hPa are shown in Figs. R1, R2, and R3, respectively. The methodology has been first used over the Dome C (Concordia) station in Antarctica and presented in Ricaud, 2014.

Ricaud, P.: Variabilités de la vapeur d'eau et de la température troposphérique mesurées par le radiomètre micro-onde HAMSTRAD au Dôme C, Antarctique. Partie II : Résultats scientifiques, La Météorologie, 85, 35-46, 2014. DOI: 10.4267/2042/53749.

This Figure undoubtedly shows that air parcels reaching the EMB during the Asian monsoon period of July-August from 2001 to 2010 are originated from Asia in the upper troposphere, from Northern America and Northern Africa in the mid-troposphere and from Europe in the low troposphere.

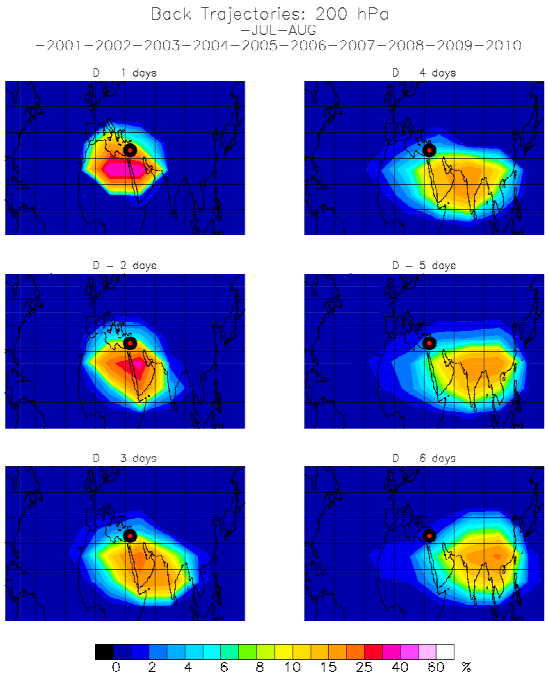


Figure R1. *(From left to right, and from top to bottom) Back-trajectory distribution of the position of the air masses from the point at 33° N, 35° E located in the Eastern Mediterranean Basin (red filled circle) calculated from the British Atmospheric Data Centre trajectory service (http://badc.nerc.ac.uk/community/trajectory/) from 1st July to 31st August from 2001 to 2010 every 12 hours at 200 hPa after 1, 2, 3, 4, 5 and 6 days.*

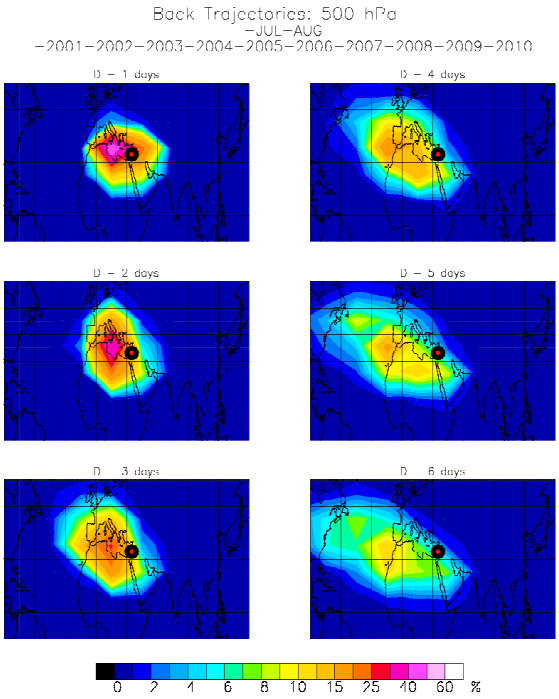


Figure R2. *Same as Fig. R1, but at 500 hPa.*

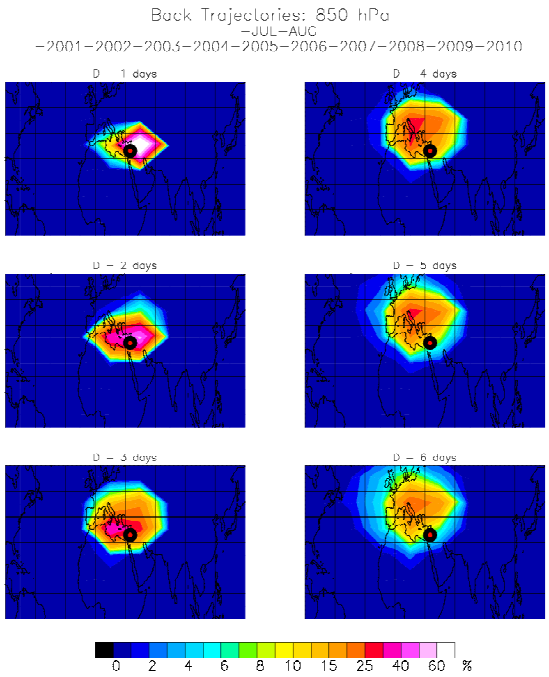


Figure R3. *Same as Fig. R1, but at 850 hPa.*

The same analysis has also been performed concentrating on the winter (JFM) 2001-2010 period (Figure 8) at 200, 500 and 850 hPa as shown in Figs. R4-R6, respectively. The same Figure also shows that in winter (and all other seasons but summer, not shown) air parcels reaching the EMB are

originated from the West (Europe, Atlantic Ocean, North America, Pacific Ocean) whatever the pressure level considered from 850 to 100 hPa.

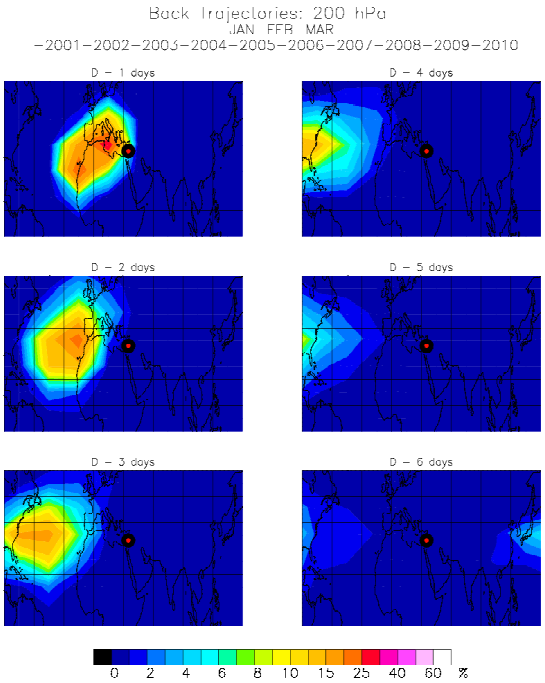


Figure R4. *(From left to right, and from top to bottom) Back-trajectory distribution of the position of the air masses from the point at 33° N, 35° E located in the Eastern Mediterranean Basin (red filled circle) calculated from the British Atmospheric Data Centre trajectory service (http://badc.nerc.ac.uk/community/trajectory/) from 1st January to 31st March from 2001 to 2010 every 12 hours at 200 hPa after 1, 2, 3, 4, 5 and 6 days.*

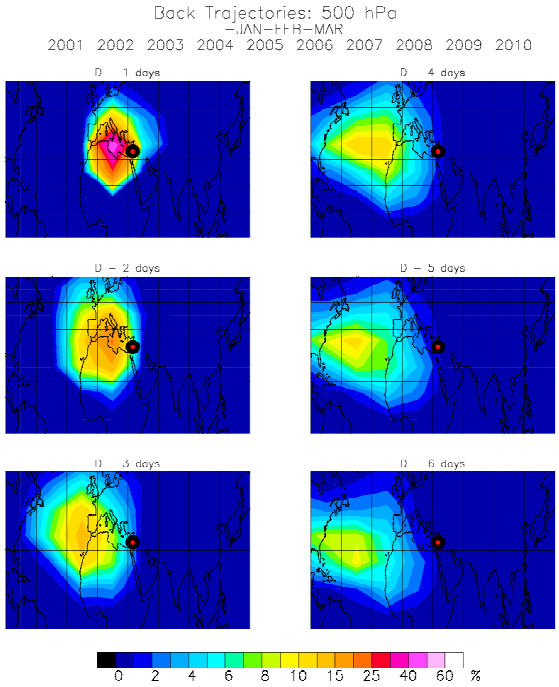


Figure R5. *Same as Fig. R4, but at 500 hPa.*

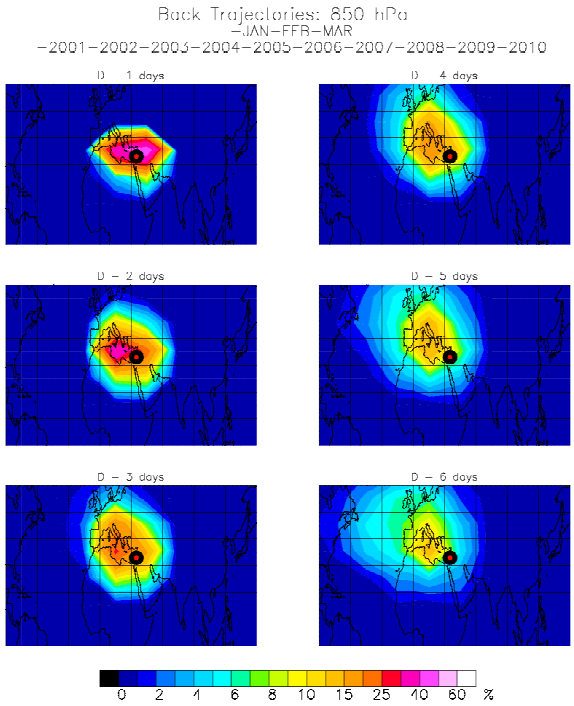


Figure R6. *Same as Fig. R4, but at 850 hPa.*

We have thus inserted a new paragraph that presents the study related to the origin of air masses in summer (July-August 2001-2010) and winter (January-March 2001-2010) above the EMB together with the associated Figure 8 (combination of Figures 8a and b).

> 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 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 position of the gravity centre of each distribution at each level is represented every 24 hours by a star on Figure 8. Data from European Centre for Medium-Range Weather Forecasts (ECMWF) archive (2.5 degree/pressure levels) are used in the present calculation. The methodology has been first used over the Dome C (Concordia) station in Antarctica and presented in Ricaud (2014). We have also performed the same analysis but for the winter period from 1st January to 31st March 2001-2010 (Fig. 8). Figure 8 undoubtedly shows that air parcels above the EMB during the Asian monsoon period of July-August from 2001 to 2010 are originated: a) from Asia in the upper troposphere, b) from Northern America and Northern Africa in the mid-troposphere and c) from Europe in the low troposphere. The same Figure also shows that in winter (and all other seasons but summer, not shown) air parcels above the EMB are originated from the West (Europe, Atlantic Ocean, North America, Pacific Ocean) whatever the pressure level considered from 850 to 100 hPa.

We have added the new reference Ricaud (2014) in the Reference list.

> Ricaud, P.: Variabilités de la vapeur d'eau et de la température troposphérique mesurées par le radiomètre micro-onde HAMSTRAD au Dôme C, Antarctique. Partie II : Résultats scientifiques, La Météorologie, 85, 35-46, doi: 10.4267/2042/53749, 2014.

Specific Comments:

1. P9977, L28 – we can conclude -> we conclude

→ Done.

2. P9978, L2 – Is this true for the all seasons?

→ Yes, but in the lower troposphere, not in the middle troposphere. We modified the sentence into (also in the conclusion):

> In the lower troposphere, the local sources of emission in the vicinity of the MB mainly affect the CH_4 variability.

3. P9978, L13 –Does the net impact refer to net radiative impact?

→ Yes, we modified the text accordingly.

4. P9978, L24 – very variable -> variable

→ Done.

5. P9978, L25 – Beside this -> Besides this

→ Done.

6. P9978, L25 – P9979, L2 – The meaning of this sentence is not clear. What do particularities and differences mean?

→ We have removed the incriminated sentence.

7. P9979, L28 – In parallel. . . (This can be a new paragraph).

→ Done.

8. P9980, L3 – Acronym (ACCMIP) should be mentioned.

→ Done.

9. P9980, L6 – recent studies (Ricaud et al., 2009) – only one study?

→ We can note indeed some campaigns like the HIPPO campaigns during which airborne in situ measurements of N_2O have been done. But regarding space-borne N_2O tropospheric measurements, YES, this is the only study presented so far. But the incriminated sentence has been removed to mainly concentrate on CH_4.

10. P9980, L12 – What is the time period of ChArMEx?

→ The first phase of the ChArMEx project is 2010-2015. This has been clarified in the manuscript.

11. P9980, L14 – proposed by France – Does this mean it's only proposed or it's being conducted as well?

→ Indeed, proposed and conducted by France. Text modified accordingly.

12. P9982-, section 2 - The model and data description can be shortened by keeping the information only needed for this study. Currently, there is too much general information in section 2.

→ We have considerably shortened section 2 by 2 pages.

13. P9982, L3 – This sentence can be rewritten. 'Surface emissivity on the sea is relatively smaller in magnitude and spatially uniform compared to the one over land'.

→ Done

14. P9982, L10 – The meaning of 'somewhat consistent' is vague.

→ We removed the term “somewhat”.

15. P9982, L24 – Does this mean NCEP/NCAR reanalysis?

→ Yes indeed, this means NCEP/NCAR reanalyses. The text has been modified accordingly.

16. P9983, L12 – Brief explanation of ‘feed-forward artificial neural network’ will be helpful.

→ We have simplified the presentation of the retrieval method of IASI pixel into:

> The retrieval algorithm for CH_4 is based on the neural network theory adapted from Turquety et al. (2004).

17. P9983, L24 – associated to -> associated with

→ Done.

18. P9984, L3 – Roughly, how many profiles are contaminated by cloud and excluded (per day or per region)?

→ We cannot answer this question. For IASI, only cloud-free pixels are retrieved. For GOSAT, only CH_4 retrievals with DOF greater than 0.6 are provided. For AIRS, only cloud-free pixels are retrieved. It is beyond the scope of this paper to go back to the calibrated spectra (Level 1 data) in order to check whether clouds interfere within the line of sight of the measurements.

To us, the most important point is to know how many pixels (profiles or total columns) are actually used in each box when considering monthly averages because this really affects the statistics thus the random noise associated to the mean. This is related to the point 20) below for which we give a detailed response (reply of the point 1.b from the reviewer#1’s comments).

19. P9985, L4 – gases research -> Does this mean that this satellite is a research satellite?

→ Yes indeed, GOSAT is a research satellite, not an operational satellite.

20. P9985, L25 – How many profiles are used in each bin?

→ See the reply of the point 1.b) from the Reviewer #1’s comments.

21. P9986, L24 – It is not clear if the emissions used in the model run are yearly or monthly averages.

→ The emissions used in the model run can be either yearly or monthly averages but, for CH_4, these are monthly averages. We modified the text accordingly.

> More precisely, the CH_4 surface emissions are monthly averages and split into anthropogenic sources taken (…).

22. P9987, L13 – I wonder why convection is not included in this study and what this mean to the results presented here?

→ This is an important point that requires a detailed answer.

As it is mentioned in the manuscript, the chemistry in this version of the model is computed down to the 560 hPa level while for higher pressures the mixing ratios of a number of species (namely N2O, CH4, CO, CO2, CFC11, CFC12, CFC113, CCl4, CH3CCl3, CH3Cl, HCFC22, CH3Br, H1211, H1301, Ox, O3, Cly, Bry, NOy) are relaxed towards evolving global mean surface abundances (see SPARC (2010) for the ozone depleting substances and greenhouse gases, and the CNRM-CCM technical documentation for the other compounds). Explicit wet deposition of chemical species is not considered in this version of the model, and neither are convective and turbulent transport (see Teyssèdre et al. (2007); Michou et al. (2011); Morgenstern et al. (2010) for further details)..

One has to note that state-of-the art CCMs rarely consider tropospheric chemistry (particularly the non-methane hydrocarbon chemistry, NMHC) because of computer resources (among the 18 models of CCMVal-2 only 3 represented tropospheric chemical reactions, see Morgenstern et al. (2010)). The chemical scheme we use is fully convenient for the study of all the processes within the stratosphere, the UTLS and down to the middle troposphere. This scheme has been evaluated in a large number of publications as the CCMVal-2 effort was aimed at assessing CCMs performances, both individually and collectively, among 17 other CCMs models. The evaluated processes cover radiation, stratospheric dynamics, transport in the stratosphere, stratospheric chemistry, UTLS, natural variability of stratospheric ozone, long-term projections of stratospheric ozone, and the effects of the stratosphere on the troposphere. A number of CCMVal-2 related publications appear in Michou et al. (2011).

The choice of no considering tropospheric chemistry (especially NMHC chemistry) is scientifically coherent with not considering the meteorological processes that occur in the middle and lower troposphere: namely, i.e. dry deposition, wet deposition, diffusion and convection. We exclude these chemical/physical processes from our simulations in the sake of computing time, vital in climate modelling where transient simulations are performed. This way of taking into account the lower troposphere is common among the CCMVal-2 CCMs (see SPARC (2010)).

The impact of non considering the above-mentioned processes on the distribution of atmospheric constituents in UTLS needs to be investigated further. This is indeed one of the by-products of the present analysis. Finally, we can indeed mention that, in the lower stratosphere, CNRM-AOCCM and LMDz-OR-INCA give consistent results presented in the updated version. A new paragraph deals with this issue (see reply to the reviewer#1's point 2b).

> In the lower stratosphere (100 hPa, Fig. 9), all the model outputs are consistent to each other showing an annual oscillation, with a wide maximum in summer (60-80 ppbv) and a wide minimum in winter (20-35 ppbv). This is apparently surprising keeping in mind that both models significantly differ from the surface (see Fig. 10) to ~500 hPa. But, in the WMB, the 100-hPa pressure corresponds to 420-K potential temperature both in summer (Fig. 2) and in winter (Fig. 4) whilst, in the EMB, it corresponds to 390 K in summer and 400 K in winter, namely closer to the tropopause in summer than in winter. Consequently, whatever the model considered, the E-W CH_4 seasonal variation at 100 hPa a) is always positive and b) shows a peak in the summer period. We note that the summer peak in E–W seasonal evolution from the middle to the upper troposphere has also been observed and calculated by

considering other constituents like CO and O_3 (not shown). This is the main topic of a forthcoming paper.

Huszar, P., Teyssèdre, H., Cariolle, D., Olivié, D. J. L., Michou, M., Saint-Martin, D., Senesi, S., Voldoire, A., Salas y Melia, D., Alias, A., Karcher, F., Ricaud, P., and Halenka, T.: Modeling the present and future impact of aviation on climate: an AOGCM approach with online coupled chemistry, Atmos. Chem. Phys., 13, 10027-10048, doi:10.5194/acp-13-10027-2013, 2013.

Michou, M., Saint-Martin, D., Teyssèdre, H., Alias, A., Karcher, F., Olivié, D., Voldoire, A., Josse, B., Peuch, V.-H., Clark, H., Lee, J. N., and Chéroux, F.: A new version of the CNRM Chemistry-Climate Model, CNRM-CCM: description and improvements from the CCMVal-2 simulations, Geosci. Model Dev., 4, 873–900, doi:10.5194/gmd-4-873-2011, 2011.

Morgenstern O., Giorgetta, M. A., Shibata, K., Eyring, V., Waugh, D. W., G. Shepherd, T., Akiyoshi, H., Austin, J., Baumgaertner, A. J. G., Bekki, S., Braesicke, P., Brühl, C., Chipperfield, M. P., Cugnet, D., Dameris, M., Dhomse, S., Frith, S. M., Garny, H., Gettelman, A., Hardiman, S. C., Hegglin, M. I., Jöckel, P., Kinnison, D. E., Lamarque, J.-F., Mancini, E., Manzini, E., Marchand, M., Michou, M., Nakamura, T., Nielsen, J. E., Olivié, D., Pitari, G., Plummer, D. A., Rozanov, E., Scinocca, J. F., Smale, D., Strahan, S., Teyssèdre, H., Toohey, M., Tian, W., and Yamashita, Y.: Review of present- generation stratospheric chemistry-climate models and associated external forcings, J. Geophys. Res., 115, D00M02, doi:10.1029/2009JD013728, 2010.

SPARC CCMVal, SPARC CCMVal Report on the Evaluation of Chemistry-Climate Models: edited by: Eyring, V., Shepherd, T. G., and Waugh, D. W., SPARC Report No. 5, WCRP-132, WMO/TD-No. 1526, available at: http://www.atmosp.physics.utoronto.ca/SPARC, 2010.

Teyssèdre, H., M. Michou, H. L. Clark, B. Josse, F. Karcher, D. Olivié, V.-H. Peuch, D. Saint-Martin, D. Cariolle, J.-L. Attié, P. Nédélec, P. Ricaud, V. Thouret, R. J. van der A, A. Volz-Thomas, and Chéroux, Atmos. Chem. Phys., 7, 5815-5860, 2007.

23. P9991, L15-21 – The meaning of this sentence is not clear. Multiple shorter sentences with clear key point rather than one long sentence will be desired.

→ We have changed the incriminated sentence into:

Due to its long lifetime (~12 years), CH_4 is considered as a well-mixed species in the troposphere. Nevertheless the CH_4 spatial distribution over the MB in summer (JJA) 2009 shows some gradients both in the East-West and the North-South directions. Indeed, in the middle troposphere (inferred from the sensitivity of the IASI total columns) and in the upper troposphere (200-260 hPa), an East-West gradient is observed in the model and satellite data of ~60 ppbv (~4%) in total column and ~30-150 ppbv (~2-9%) in mixing ratio. A North-South gradient is also detected in the MOCAGE and AIRS data but not in the IASI data set. Therefore, there is systematically a maximum of CH_4 from the middle to the upper troposphere in the East of the MB compared to the West.

24. P9991, L21-25 – Long-range transport from Asia is not convincing unless backward trajectory model or something equivalent is used.

→ A new Figure 8 has been inserted. See all the points presented and discussed above.

25. P9992, L25 – Does this mean MOCAGE is sampled like AIRS (horizontally) as well?

→ No, AIRS and MOCAGE are not initially sampled consistently. The MOCAGE vertical profile closest to an AIRS pixel has been convolved with the AIRS averaging kernel attached to the corresponding pixel. Note, for one particular day, the AIRS averaging kernels attached to the AIRS maritime pixels over the Mediterranean Basin are all very consistent to each other. We have finally monthly averaged the convolved MOCAGE profiles and the AIRS profiles within the same boxes (East and West) consistently.

26. P9993, L4 – very consistent -> consistent

→ Done.

27. P9993, L5-7 – I don't think the difference between AIRS and MOCAGE is only related to a-priori. Global models known to be underestimate mixing ratios of trace species largely due to coarse horizontal resolution and large uncertainties in estimated surface emission.

→ This is a good remark that cannot indeed be ruled out. This issue is dealt in detail in the replies to the reviewer#1's point 2a.

28. P9994, L20 – E-W maximum -> maximum in E-W gradient?

→ All over the manuscript, we defined the "East minus West difference" as the term E-W. So, the "E-W maximum" means a maximum in the "East minus West difference". This implies 1) a maximum in the East-West gradient and 2) the amount of CH_4 is greater in the East than in the West.

29. P9994, L26-28 – Why the amplitude of seasonal cycle is consistent even though the absolute values are different?

→ In fact, the amplitude of the seasonal cycle is almost twice greater in the satellite measurements (~25 ppbv) than in the model data (~15 ppbv). Why? We can try to explain the difference in amplitude between satellite and model in the seasonal evolution of E-W by: a) regarding the comparison technique, there is a broader vertical domain in the measurements compared to the model data, b) regarding the processes in summer, we may have less CH_4 trapped in the Asian Monsoon Anticyclone redistributed towards the Eastern Mediterranean Basin in the models compared to the measurements, c) regarding the processes in winter, we may have too much CH_4 brought over the Mediterranean Sea to the East compared to the West producing a too smooth E-W gradient in the models compared to the measurements. We have modified the incriminated sentence and have inserted some elements of interpretation.

> The peak-to-peak amplitude of the E–W seasonal variation is almost twice greater in the satellite measurements (~25 ppbv) than in the model data (~15 ppbv). This represents a ~1.5-2.0% variation of CH_4 in the E–W over the entire year. The difference in amplitude between satellite and model in the seasonal evolution of E-W may be due to: a) the comparison technique. There is a broader

vertical domain in the measurement data compared to the model data; b) regarding the processes in summer, we may have less CH_4 trapped in the Asian Monsoon Anticyclone redistributed towards the EMB (see section 5) in the models compared to the measurements; c) regarding the processes in winter, we may have too much CH_4 brought over the Mediterranean Sea to the East compared to the West producing a too smooth E-W gradient in the models compared to the measurements.

30. P9996, L19 – We can note -> We note

→ Done.

31. P9996, L22 – issue -> topic or subject

→ We changed to “topic”.

32. P9998, L17 – non-zonally-symmetric -> zonally-asymmetric

→ Done.

33. P9999, L25 – ‘somewhat consistent’ is a vague description.

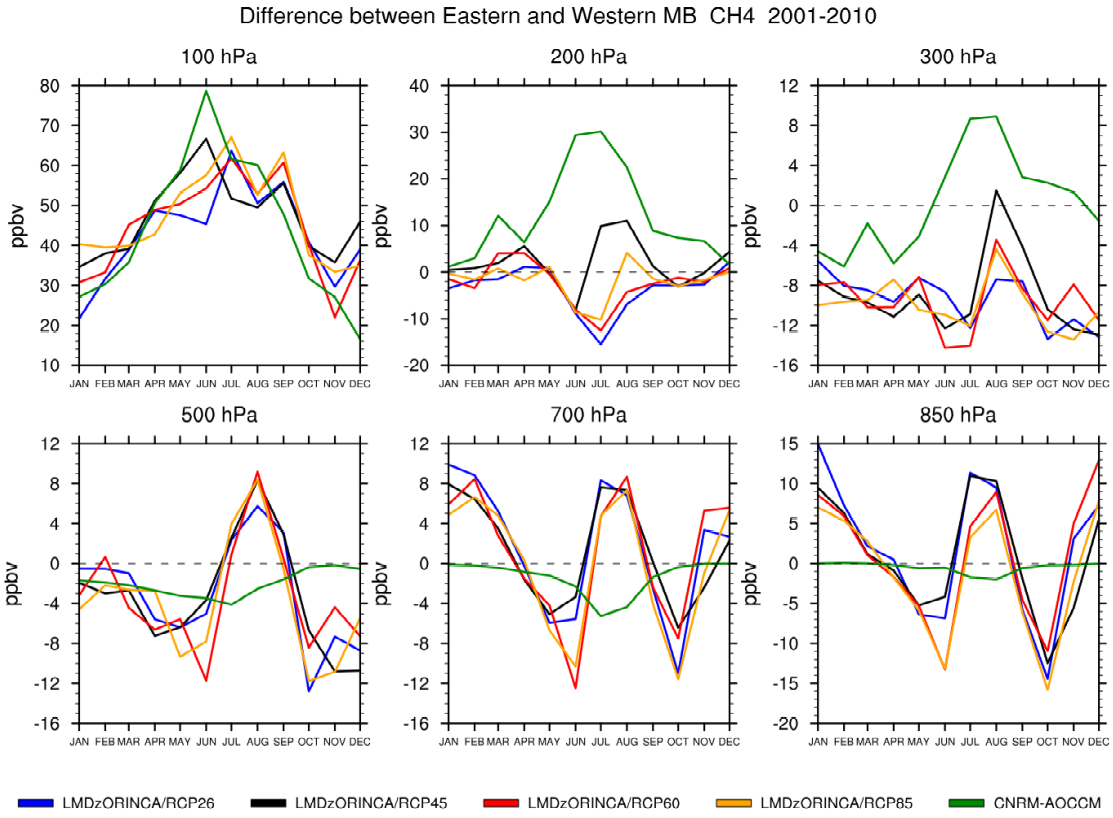
→ This paragraph has been removed from the conclusion. See replies to Reviewer #1.

34. P10000, L17 – we can -> we

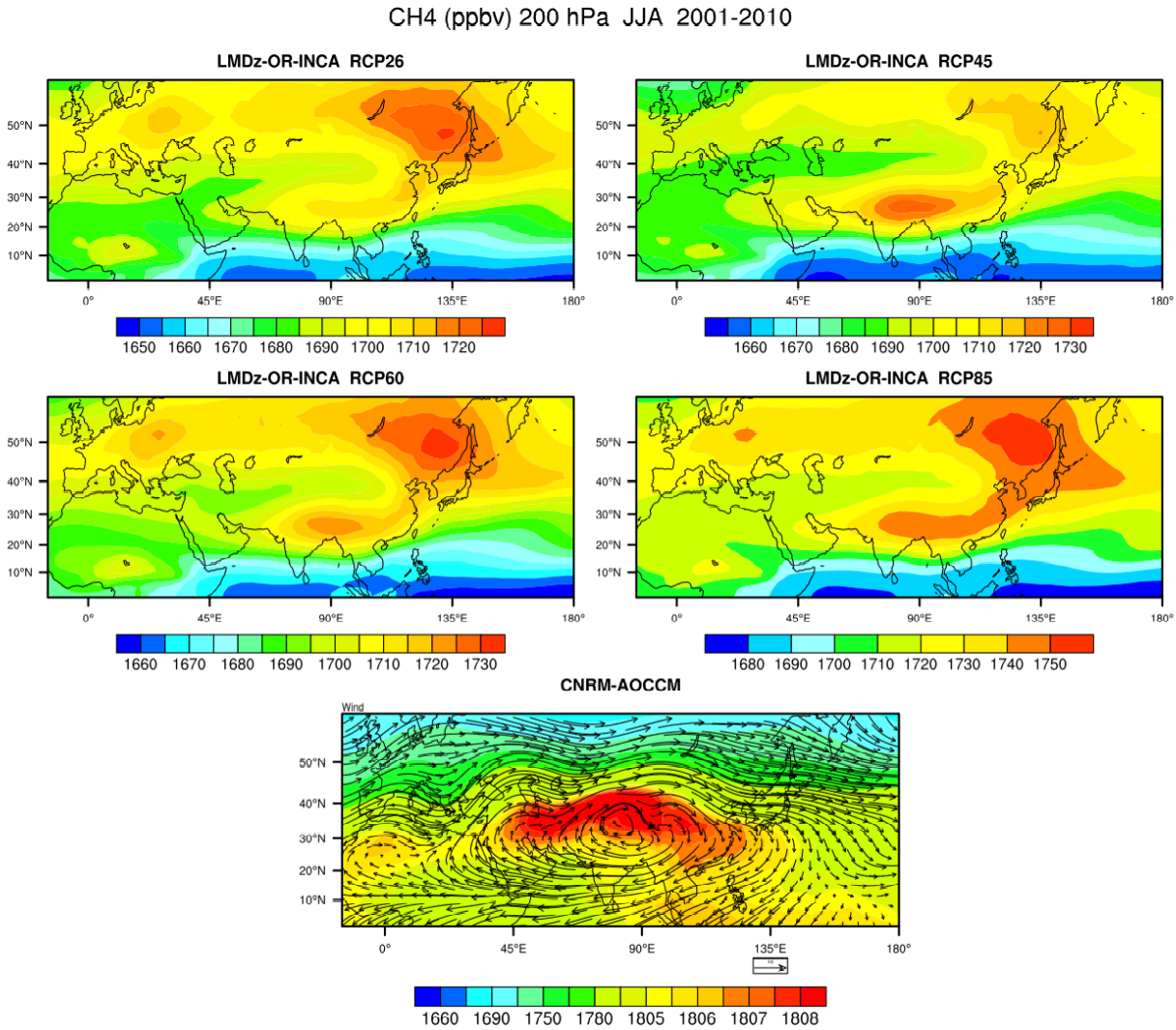
→ Done.

35. Fonts size for the figure titles and color bars has to be bigger than the one currently used.

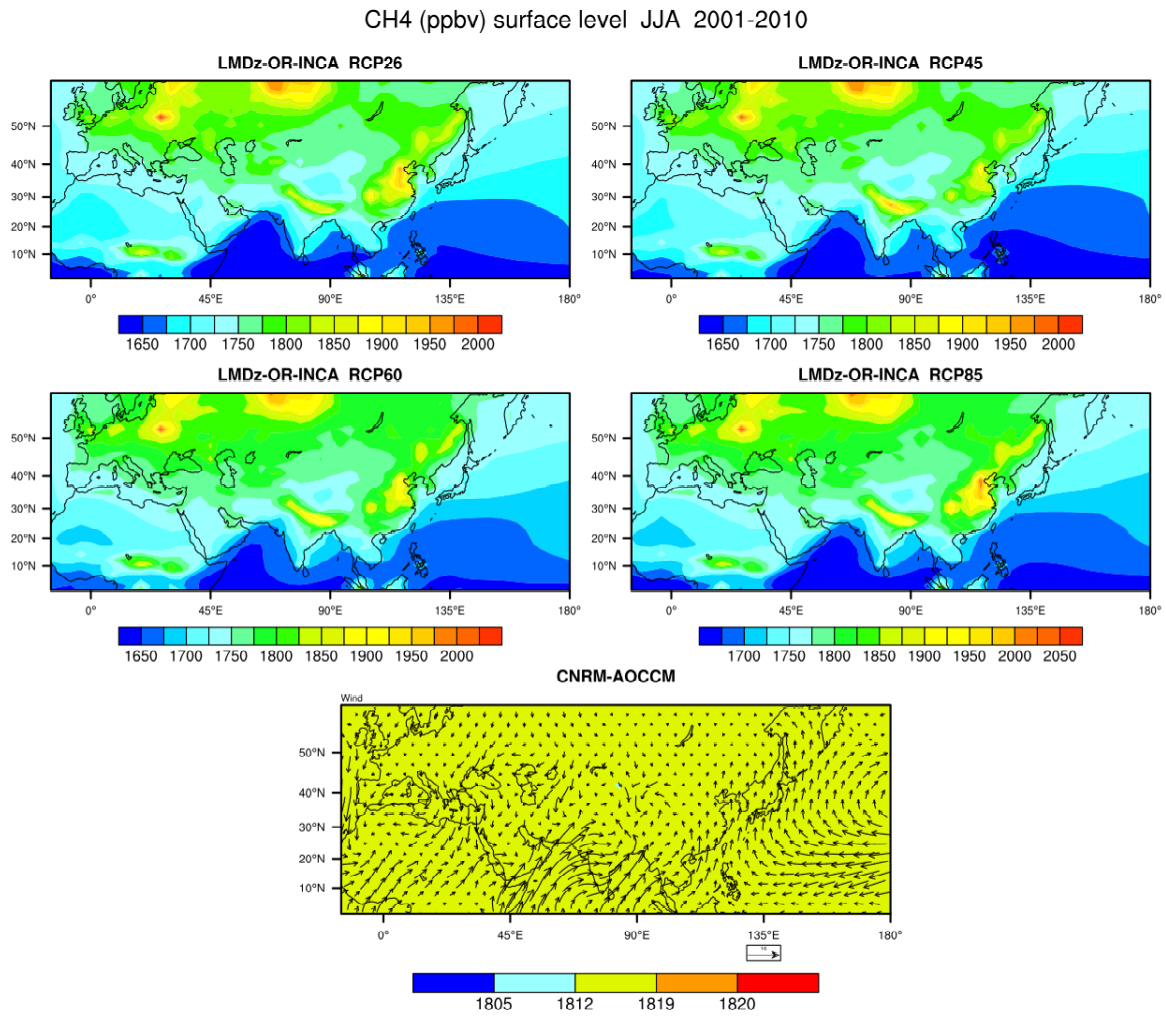
→ We have updated the Figures 10, 11 and 12 according to the reviewer’s comments.



New Figure 10 (Figure 9 in the revised manuscript).



New Figure 11 (Figure 11 in the revised manuscript). *Fields of CH_4 as calculated by the CNRM-AOCCM model (bottom) and the LMDz-OR-INCA model (top and centre) considering the 4 IPCC scenarios (RCPs 2.6 (top left), 4.5 (top right), 6.0 (centre left) and 8.5 (centre right)) at 200 hPa averaged over the summer season (JJA) and the climatological period 2001-2010. Superimposed to the CNRM-AOCCM CH_4 fields (bottom) is the wind field at 200 hPa averaged over the same period. Note that the range of the colour scale changes for each figure and that the colour scale for the CNRM-AOCCM model (bottom) is non linear.*



New Figure 12 (Figure 10 in the revised manuscript). *Fields of surface CH_4 as calculated by the CNRM-AOCCM model (bottom) and the LMDz-OR-INCA model (top and centre) according to the 4 IPCC scenarios (RCPs 2.6 (top left), 4.5 (top right), 6.0 (centre left) and 8.5 (centre right)) averaged over the summer season (JJA) and the climatological period 2001-2010. Superimposed to the CNRM-AOCCM CH_4 fields (bottom) is the wind field at the surface averaged over the same period. Note that the range of the colour scale changes for each figure and that the surface CH_4 for CNRM-AOCCM (bottom) is constant.*