Final Response, Ralf Sussmann, Karlsruhe Institute of Technology, Garmisch, 13 April 2012.

We thank both referees for their efforts and very interesting comments which helped to significantly improve the paper. We thereafter present our point to point reply.

I) Response to Referee#1

Ad 1: The interference errors discussed in Sussmann et al. (2011) had been shown to cause a seasonal bias which is of systematic nature. Therefore, the number 0.14 % refers to "interference errors". The number 0.3 % refers to precision, i.e., other, random-type errors.

To clarify we reformulated (p 30760, l 23) to: "… eliminate H2O/HDO-CH4 interference errors down to the ≈0.1% level (i.e., 0.14 % for the wettest test site and 0.10 % and 0.02 %, respectively for the two dryer test sites)." Note this paragraph has been shifted to Section 2 in the revised manuscript.

Ad 2: Our idea of validating (via trend behavior) the finding in Sussmann et al. (2011) that no significant interference errors are present in MIR_GBM v1.0 had been based on the following assumption: if the neighboring sites Zugspitze (2964 m) and Garmisch (743 m) have the same water vapor trend in per cent per decade, the absolute increase in columnar water vapor per decade would be higher for Garmisch compared to Zugspitze. Knowing that the HDO/H2O-CH4 interference errors increase approximately linearly with absolute H2O (or HDO) column (see Sussmann et al., 2011, Fig.6) this would mean that the Garmisch methane trend should be more impacted by such water vapor trend compared to Zugspitze. The resulting artifact would be a difference in methane trends between Garmisch and Zugspitze - if interference errors were present.

We now calculated the absolute magnitude of such artifact in methane trends using realistic numbers for the water vapor trend (e.g., 0.79 mm per decade for Zugspitze, see Sussmann et al., ACP 2009). Unfortunately, it turned out that the magnitude of the resulting artifact, i.e., the difference in methane trends between Garmisch and Zugspitze is always insignificantly small (i.e., in the order of or less than 0.1 %), even for a non-optimum retrieval strategy (e.g., retrieval strategy using HITRAN 2008 and MW12345 with a strong HDO/H2O-CH4 interference error of -2.97 % for Garmisch, see Table 5 in Sussmann et al., AMT 2011). In other words, we found that methane trend behavior is no good proxy for the existence versus non-existence of interference errors, whereas the latter can dominate the seasonal cycle of methane as shown in Sussmann et al. (AMT 2011). The simple reason for this is that a typical decadal increase of total-column water vapor (e.g., 0.79 mm per decade for Zugspitze, see above) is much smaller than the typical intra-annual (summer-winter) variability (e.g., max - min = 12 mm for Zugspitze).

As a consequence we cancelled the text about validation of MIR-GBM via trend behavior (1st paragraph of Sect. 3.2) and removed related statements throughout the text.

Ad 3. This text has been removed in response to the previous point.

Ad 4: We would like to keep the trend change points and significance intervals in the paper by the following reasons:

i) In our case the division into 3 time periods has been performed in agreement with dedicated trend change points defined and used in earlier work. To clarify, we added the following sentence (p 30762, l 25): "The division into 3 time periods has been performed in agreement with dedicated trend change points defined and used in earlier work for methane (Dlugokencky et al., 2003; 2009; Angelbratt et al., 2011)."
ii) We agree that there is an intrinsic paradox with linear trend analyses because the investigator inevitably influences the trend result by "artificially" defining the boarders of the time period to be investigated. This touches a general science-philosophy issue: any specific formulation of a question impacts (i.e., reduces) the solution space for possible answers; obviously the solution to this paradox is not to stop asking questions.
iii) In spite of the dependence of a trend on the chosen time period it is necessary to perform a significance analysis of the trend result because otherwise the derived number for the trend would be without any meaning (if there is zero information on significance).
iv) Referee#2 has criticized the opposite, namely he criticized missing significance intervals for the SCIAMACHY trend, and we agree to add these.
v) The numbers we give for the significance intervals are not "wrong"; we think they add valuable information; anyway we see no benefit of dropping these numbers.

Ad 5: We agree that OH concentrations do not explain all CH4 loss by OH radicals. The whole section about OH has been re-written, stating more precisely the recent results about OH changes.

Ad 6: We agree that a discussion on the causes of the changes of atmospheric methane would require much more analyses and that this is beyond the scope of this paper. We largely reduced the part of the paper devoted to the "after 2009" causes.

II) Response to Referee#2

GENERAL COMMENTS

“In Table 2 zonal mean estimates are given for SCIAMACHY. It is possible that its data coverage does not allow, for example, a Europe-specific estimate. An attempt in that direction would nevertheless be useful.”

We agree and added to Section 3.2 a SCIAMACHY trend analysis including a significance characterization using the same (bootstrap) technique as for the FTIR. To obtain a Europe-specific estimate we used a 1000-km selection radius around the Zugspitze. We found agreement of the Zugspitze FTIR trend with the trend derived from SCIAMACHY data within the 95 % confidence intervals of the trends. In case a 1000-km pixel selection radius around the Zugspitze is used, the trend confidence interval is narrower for the FTIR trend analysis compared to SCIAMACHY, whereas for a loosened pixel selection criterion (1000-km half-width latitudinal band centered round Zugspitze) the SCIAMACHY trend significance interval becomes narrower.

“It would be interesting to know how near IR and mid IR compare, regarding both the size and the uncertainty of the derived trend estimates.“

We agree but this had been the subject of another study by Forster, F. et al.: First intercalibration of column-averaged methane from the Total Carbon Column Observing Network and the Network for the Detection of Atmospheric Composition Change, Atmos. Meas. Tech. Discuss., 5, 1355-1379, 2012. In this study no significant difference of MIR and NIR CH4 trends has been found.

“It is mentioned that a water vapor trend has been observed. However, it is unclear what would be the size of the error in CH4 depending on the mid IR retrieval strategy. Without this information it is impossible to judge the value of comparing Garmisch and Zugspitze.”

See our answer to Referee#1 above: Ad 2.

“The discussion about what is known about the CH4 growth rate seems to be rather out of proportion ….In particular the meaning of the second paragraph of page 30765 is unclear.”

As suggested also by Referee#1, we removed the precipitation and temperature analyses performed after 2009 as a more complete analysis would be necessary that is beyond the scope of this paper.

“Else, the part starting with “According to different possible interpretations” (page 30766) is really too vague”

We removed this sentence.

SPECIFIC COMMENTS

“Page 30759, line 6: GWP is dependent on more factors such as lifetime of the molecule and, more importantly in this context, the position of absorption lines in the spectrum. Chemically, CH4 and CO2 differ by much more than only symmetry. CO2 has strongly saturated absorption lines, but outside the atmospheric window. This, in my opinion is as important as the symmetry argument raised here.”

We fully agree and added the window argument.

“P30759, line 18: The papers by Rigby and Montzka actually do point to a possible role of OH, however, it is difficult to judge the value of MCF measurements in recent years because of its decreasing abundance in the atmosphere. This, however, does not exclude a possible role of OH.”

The OH analyses have been moved to the discussion section. We agree that the statement about OH was not clear. We rephrased the analyses in the discussion section pointing a possible role of OH in the recent atmospheric CH4, as found by Montzka and Rigby. However, we think that the decrease of MCF in the atmosphere since 1998 makes it easier to use this proxy to infer OH changes because, as stated by Montzka, it relies less than before 1998 on the magnitude of emissions, as they have been largely reduced. Therefore, as long at MCF concentrations in the atmosphere are larger than the measurement precision (of about 2 ppt), we think it is a reliable proxy to infer OH variations.

“Page 30760, line 7: The suggestion is made that a single FTS could replace several in situ measurement sites. The sensitivity of surface sites to fluxes and their representation of larger scales vary greatly from site to site. Therefore, I would have been surprised if Olsen and Randerson made such a claim, which indeed I was unable to find (Olsen is spelled without ‘h’).”

We reformulated: “Several studies have explored how column observations would complement surface networks and help to further reduce uncertainties associated with sources and sinks in atmospheric inversions (e.g., Olsen and Randerson, 2004 or Bergamaschi et al., 2009, and references therein).”

“Page 30760, line 16: A reference is needed for the 5% error.”

We added the reference for the 5 % error. Note this passage was moved to Section2.

“Page 30764, line 26: The fact that OH doesn’t show a high variability in CTMs doesn’t
necessarily mean that this possibility can be excluded in the real world.”

An ensemble of evidences suggest that OH changes are limited from one year to the next, and much smaller than the one inferred for the 1990s by atmospheric inversions (8-10%). The ensemble is made of different approaches: recent atmospheric

inversions (for the 2000s), CCMs and CTMs. We completely re-wrote the OH section and tried to make more clear these different lines of evidences.

Additional changes (p 30765, “In this context …. Decline of the atmospheric growth rate during 1990-2006”): We removed this paragraph on the analysis of the constant period 1999-2006 as the paper focuses on the recent positive anomaly.

End of response.