Response to reviewer #1 comments to manuscript:

Curci, G., Palmer, P. I., Kurosu, T. P., Chance, K., and Visconti, G.: Estimating European volatile organic compound emissions using satellite observations of formaldehyde from the Ozone Monitoring Instrument, Atmos. Chem. Phys. Discuss., 10, 19697-19736, doi:10.5194/acpd-10-19697-2010, 2010.

R = Reviewer's comment
A = Authors' reply

The authors thank reviewer for his/her careful revision and thoughtful comments which we believe improved our manuscript. In particular, we extended analysis of results with a country level breakdown of emissions, for ease of comparison with other studies, and improved justification for the study. In the related revision process we also found a bug in our routines related to calculation of continental totals of averages: emission monthly totals in Table 2 and Figures 5 and S2, and column averages in Table 1 are corrected in the current version of the manuscript. In view of new analysis developed in section 4.2 and 4.3, we also revised to a more conservative statement the final comments on the robustness of Bayesian inversion. Detailed response to your comments are enclosed below.

R1.1) It would be great to read more about how this study fits in with previous literature. How is it an improvement or innovation upon what came before.

A1.1) We agree. A comparison with previous work is missing in our results section. We now added a new Table with emissions split to country scale and expanded section 4.2 to compare other estimates of European isoprene emissions. Please see also A1.20.

R1.2) Throughout the paper, it is remarked that OMI-observed HCHO over Europe is close to detection limit, especially in the colder months. This naturally raises the question if it is appropriate to use OMI data for such a study. It would be helpful to read a much stronger justification for the study. One also can't help noticing that the best measurements are over sunny, cloudless regions. The paper does not, however, address the possible bias coming (or not) from that.

A1.2) Here the question is twofold. (1) Regarding justification of the work, we meant to provide the reader with evidence of usefulness of HCHO satellite observations in the introduction (two paragraphs from P19699-L32 to P19701-L11). Probably the key messages were not clear enough and they are now stated explicitly at the end of each paragraph. (2) On the potential bias coming from cloud screening of satellite scenes we referred to previous work and used a suggested threshold for cloud fraction of 0.2 (Palmer et al., 2006; Millet et al., 2006, 2008). Typically low clouds increase measurement sensitivity, while high clouds decrease the sensitivity. The threshold of 0.2 for OMI data it is a good compromise between minimizing the impact of clouds on retrieval and retain enough data for analysis. For present work, we now recalculated monthly averages of OMI data with a cloud fraction threshold of 0.4 and verified that, on average, HCHO columns differ by 0.1-0.2 × 10^{15} molecules/cm^2 at the expense of an increased error of 0.4-0.9 × 10^{15} molecules/cm^2.

R1.3) I found the organization of the paper a bit confusing. For example, in section 2, I expected to see model validation. It was not intuitive to me that it would be in section 3.2. Meanwhile, section 4.2 devoted first three paragraphs to errors and not emissions. Perhaps the order in that section could be reversed?

A1.3) We moved model validation against EMEP surface measurements at the end of section 2. Regarding section 4.2 the choice of presenting errors first is motivated by the fact that they are the base for the *inversion of emissions. However, we agree the presentation of results starting from error is counterintuitive and we swapped presentation of Figure 4 and 5, with related changes to text throughout section 4.2. This should make the paragraph more readable.*

R1.4) As I am sure the authors do as well, I wish there was more data to compare the model and results to. It seems that EMEP measurements are not at all useful in evaluating the inversion results. Are there any other data? Maybe even a quick comparison with SCIAMACHY could be informative. The authors point out that the Bayesian framework necessarily yields equal or smaller a posteriori errors. Therefore, if there is any other data to include in the study, it would be of great help.

A1.4) We have been looking for additional data useful to the study even before first submission, but unfortunately found that from EMEP is only dataset available. Ground based DOAS measurements were reported in the Netherlands, where however correction to emissions are also very small (e.g. see Figure 4 or Table 3). In the conclusion we recommend future work over some interesting places (e.g. France, Italy) in combination with a complete suite of in situ measurements, such as the ACCENT-VOCBAS campaign in Italy (Fares et al., 2009).

R1.5) p 19703, l. 1-2: What is the implication of the model not including the latest isoprene chemistry. Does this introduce a bias? Which way? Again, one wonders about all details since data is close to detection limit.

A1.5) The implication of not including the latest isoprene chemistry is already discussed in the paper, just in the following lines (please see next question).

R1.6) p 19703, l. 2-7: I'm confused if all those issues listed are related.

A1.6) This is the answer to previous question on new isoprene chemistry. The mechanisms proposed for "pristine" atmosphere are expected to have negligible effect, while the mechanism proposed by Hofzumahaus et al. (2009) is still very uncertain and needs supporting measurements to be constrained (Pugh et al., 2010). We modified this sentence in order to make clear the logical connection with isoprene chemistry issue.

R1.7) p 19704, l. 1: Please define CCDs

A1.7) CCD is the acronym of Charge-Coupled Device, now explicitly stated in the text. It is a well-known electronic device consisting of an ordered grid of "binary" elements (e.g. photoelectric devices) which may be used to reconstruct a 2-D image in a certain wavelength range. A common application is the imaging sensor of digital cameras.

R1.8) p 19705, l. 1-4: This is where I start to wonder if there could be a clear-sky bias.

A1.8) As anticipated in A1.2, the issue is now treated in the paragraph where we introduce Table 1 (P19705-L12-19). We put a new Table S1 in online supplement, which is similar to Table 1, but calculated applying a threshold of cloud fraction in OMI data of 40% instead of 20%. Comments are added to the text after P19705-L19.

R1.9) p 19705, l. 7: replace "of" with "as"

A1.9) Text changed.

R1.10) p 19706, l. 20-22: Is upper troposphere feature captured by CHIMERE model, considering that its vertical extent is only 200hPa?

A1.10) No, the feature is not captured by CHIMERE, as is not captured by other CTMs in general (e.g. Kormann et al., 2003). However, considering the average profile reported by Kormann et al. (2003) and an exponential decrease of air density with height the contribution of the HCHO layer above 10 km to the total HCHO column is less than 5%.

R1.11) p 19707, l. 13-14: This is where I again wonder if there is a clear-sky bias (due to photochemical production of HCHO).

A1.11) Here the reviewer calls into cause the clear-sky only observations as possible reason to explain the large underestimate of model HCHO column over sea. However, we believe this is not likely the case, since we are comparing with observation with model output sampled at same time and location of satellite overpass. Thus, if the model reproduced dynamical and photochemical processes correctly, it would not have this large bias, because it would have accounted for photochemical production with clear sky conditions.

R1.12) p 19708, l. 5-10: It’d be educational to see what’s "very small bias" and "poor" correlation for HCHO. Please provide more numbers.

A1.12) Right, we now put numbers in.

R1.13) p 19709, l. 5-6: How do we know that regional biases are due to incorrect emissions? How about chemistry and transport errors?

A1.13) This is an anticipation of analysis of HCHO production over Europe illustrated in next section. The statement is now clarified.

R1.14) p 19709, l. 8: remove "on"

A1.14) Text changed.

R1.15) p 19709, l. 18: "column background of 55-85%": What is this a percentage of? I was confused.

A1.15) It is the percentage of total column provided by oxidation of CH4 and other long lived VOCs, and considered a “background” column. According to analysis by Dufour et al. (2009) this background contribution ranges 55-85% throughout the continent. The sentence is now rephrased for clarity.

R1.16) p 19711, l. 19-20: If it is a linear relationship, it doesn't mean that you can neglect transport (think CO, for example); please clarify your assumptions that allow you to neglect transport. I'm thinking it's a short lifetime.

A1.16) Right. Actually "neglect transport" was referred to "local", but it was not clear as written. We rephrased the sentence, pointing out the two assumptions (neglect transport and linear relationship) and letting the reader have the details on subsequent paragraphs.

R1.17) p 19712, l. 9: remove "s" at the end of "two-elements"

A1.17) Text changed.

R1.18) p 19713, l. 19-20: The sentence "without proper "guidance" the inversion..." sounds quite suspicious. Could you be more explicit about the "guidance"?

A1.18) The sentence is now removed because ambiguous and not very useful.

R1.19) p 19713, l. 21-23: I don't understand why you would use daily OMI data to constrain monthly emissions in this inversion setup. Isn't it much more computationally expensive without much scientific gain? Perhaps the scientific gain is not transparent to me. Please clarify.

A1.19) Monthly averages, as also pointed out by e.g. Dufour et al. (2009), are needed to decrease observational error. In our framework this can be done in two ways: or making an observational vector with N daily observation over the month, or reducing the observational vector y to a scalar and reducing the average observational error by the square root of N. We actually did both and obtained pretty much the same results, but choose to present the first approach because it gave generally more "pieces of information". Moreover, the difference in terms of computational cost is minimal. 99% of CPU cost in this work is calculation with forward model (CTM) to estimate K and b, which is the same in any case.

R1.20) Section 4.2: It would be nice to see absolute emission numbers rather than only percentage changes. Future studies that might not use MEGAN might have an easier time comparing results. In fact, it would be great to have a table, like Table 2 or Table S1, with a breakdown by large source countries. Looking at the maps, it's clear there are some inhomogeneities in the inversion results that could be averaged out in the whole-Europe numbers.

A1.20) As anticipated in A1.1, this is a very good point. We added Table 3 with breakdown of continental emissions to country scale. Results with MEGAN are compared to results using Derognat et al. (2003) emissions and to estimated given by Simpson et al. (1999). Moreover the section 4.2 is now expanded with comparison with other work on European isoprene emissions.

R1.21) p 19718, l.3: Although it would be nice to account for nonlinearities in the problem, the adjoint model framework would not be the solution. The adjoint is just a more computationally efficient tool for the same linear approach. Only the Ks could be updated with each iteration. Unless this is a different point and I am missing something. Please clarify or remove.

A1.21) We agree, and removed the sentence.

R1.22) p 19720, l. 4: remove "s" at the end of "depends"

A1.22) Text changed.

R1.23) p 19720, l. 5: I do not understand the phrase: "uncertainty is introduced by estimation of inversion parameters".

A1.23) We changed "is introduced" with "is related to".