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Interactive comment on "Estimates of European emissions of methyl chloroform using a Bayesian inversion method" *by* M. Maione et al.

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This paper presents evidence of ongoing methyl chloroform (MCF) emissions from Europe in the period 2002-2012. In fact, it is found that a region in France (SEF) contributes significantly to the MCF emissions, even on the global scale. The authors combine measurements at three European stations with inverse modeling and derive most likely emissions. They include sensitivity analysis to estimate the error in their estimates. The main text is well written and presents, as far as I can see, a balanced analysis of what inversions of this kind can and cannot do. Still there are a number of open issues that need further clarifications (see below). This holds specifically for the Appendices, which were rather messy and contain a number of annoying errors.

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Nevertheless, the paper provides a valuable contribution to the ongoing debate about MCF emissions and provides useful insights in the inverse techniques that are used. The identification of the SEF emission region is quite noteworthy.

Major comments

I have some doubts about the results presented in figure 4. If I understand the method correctly, you assign uncertainties to the prior emissions that are a fraction of the emissions themselves. This implies that with zero or very small emissions the assigned uncertainty is small (page 8221). Also, the resolution at which the emissions are derived varies. In a region with low sensitivity (e.g. Norway) the resolution should be about 36x36 degrees (page 8218). Figure 4 (and the accompanying text) shows that emissions are projected in Norway, exactly at the spots where "emissions to soil and water" are reported. And these were not included in the prior inventory, so I guess that the prior emissions are very low. The same holds for Madrid, Barcelona and hotspots in Northern Africa. Yet, the posterior emissions presented in figure 4 (right panel) show "hotspot" emissions rather far away from the measurement locations that are not in the prior (e.g. Madrid). This implies a tremendous skill of the model to pinpoint emissions in places where the prior has low values (with an uncertainly that is proportional to this low emissions). I cannot believe that inversions are capable to accomplish this, although also Appendix B (figure B1) shows that emissions are derived at the coast of Norway. So, I do not believe that the prior emission map in Figure 4 (left) and the described uncertainty was the basis for the posterior map in Figure 4b. So I would like to see more analysis. What station constrains the Norway emissions? What was the resolution of the emission grid in Norway, Spain, and Portugal?

Another major issue is the posterior fit with the observations. The information is presented in Appendix A1.4. The relevant section is: "Given that the sources regions are scarce... being CMN with r2,ba = 0.5." In general, these results seem to be influenced by the fact that both the model and the observations correctly predict the general decline in MCF over the years. This naturally leads to a high correlation with has nothing

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to do with the inversion. So, I think the analysis should be done on de-trended data. As a result, I would expect a very small r2. What strikes me also is the small improvement of the r2eb compared to r2ea. Since this is the signal that drives the inversion, it would be instructive to present timeseries at the various stations (prior and posterior) just to see the skill of the model. Statistics do not look promising though. In Appendix A1.4 it is also written " the low values (of r2eb) obtained at MHD in this analysis confirms how scarcely is MHD affected by polluted air masses, despite the presence of numerous UK sources declared by the E-PRTR inventory". The correlation is not determined by the low number of data-points, but by the skill of the model to reproduce enhancements above the background. This has nothing to do with "scarcely". So, in general, I think Appendix A1.4 should be rewritten (text is not as well written as the main text), and more clearly present how the observations above the baseline drive the emission increments. After all, these are the data that drive the whole story in the main text.

Minor comments

Page 8211, line 4. I think it should be stressed here that the amendments to the Montreal protocol were much more stringent than the original protocol.

Page 8212, line 6. Same issue.

Page 8212, line 13: longitudinal? Do you mean latitudinal?

Page 8213, line 20: on going. Should be one word (ongoing).

Page 8214, line 11: given the fact that the other stations project emissions in Norway, I find it hard to believe that ZEP does not "see " these emissions. Thus, I find it rather strange that $\frac{1}{4}$ of the SOGE network is not used in this analysis. At least it would be interesting to present the background concentration of ZEP in this paper.

Page 8214, line 22: Awkward sentence. Consider: ..and m/z values of .... are selected for detection and ...of MCF.

Page 8215, line 19: Space is missing I guess (SA-6RIX)

Page 8216, line 22: consider "points" after data.

Page 8217, line 24: "analytical". This is in conflict with the iterative procedure described under (iii) at line 27.

Page 8218, line 10: I think this does not reflect an outlier in the model simulation, but the fact that the model is not able to adequately represent a certain measurement (i.e. a representation error). By the way, in the remainder of the paper, I have not seen a quantification of the number of outliers, and their influence on the inversion.

Page 8218, line 25: I suggest to replace "but" by "and".

Page 8218, line 26: "is needed". I suggest something like: "The emissions at large distances from the measurement locations cannot be resolved at high spatial resolution".

Page 8219, line 3: At the start of this paragraph, it would be useful to write something about the reason why this second method is needed. Later on it becomes more or less clear, but this is the location to highlight this. Also mention here that you can estimate time varying emissions with the point source analysis (now it is written later).

Page 8219, line 23: "While individual emission values are noisy, their average can substantially reduce this noise". Individual data points cannot be noisy. I would describe the method (as I understand it) as follows: The PSA method takes each individual measurement above the baseline, and determines the emission that is needed in a predefined source region (X) to reproduce this measurement exactly. Due to inaccuracies in transport and other numerical errors, this normally results in a noisy emission time series that, upon averaging in time, provides an estimate of the emission in region X. (see major comments).

Page 8220, line 6: mixing ratio enhancements above the baseline. I would use either "mixing ratio enhancements" or "mixing ratios above the baseline". Also the caption of figure 2

Page 8221, line 1: "emissions to soil and water". Not clear to me what is meant exactly.

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Please clarify.

Page 8221, line 10: "leading to", consider, "i.e. 50% to 500% of the prior emission estimate. "

Page 8221, line 11: "The emission variability and correlations between measured and a posteriori modelled data substantially increased over this range of values". Unclear sentence. I think the intention is to say that you try to balance between (i) enough flexibility in emissions to allow adjustments that fit the observations better (ii) not too much flexibility because this might lead to over-fitting of the observations and noisy and unrealistic emissions (e.g. negative emissions). I think this message should be better phrased here.

Page 8221, line 14: I think "changes" should be "improvements".

Page 8221, line 17: "allowing" should be "obtaining".

Page 8221, line 25: points (ii) and (iii). See major comment.

Page 8222, line 4: within SEF are included. I suggest: "SEF includes"

Page 8222, line 10: We can extend. ... I suggest: We extend (to reflect your choice).

Page 8223, line 10: not impacted. ... I suggest: not severely impacted

Page 8223, line 26: cannot be excluded. I think "is likely" better reflects the situation.

Page 8224, line 6: MHD, meanwhile. I suggest full stop: ..MHD. Meanwhile . . .

Page 8225, line 6: (except MHD). I suggest to use. ..., but not for MHD.

Page 8225, line 7: The error estimates for JFJ and MHD are unrealistically low. How are these number derived? Later on it is suggested that the error bars reflect the scatter in the individually bi-hourly derived estimates. I cannot imagine such small scatter, given e.g. the uncertainty in transport modelling.

Page 8225, line 19: "This is expected as the inversion results are bound towards a priori

emissions that are clearly too low for the SEF area. This leads to a low bias also in the obtained a posteriori emissions". I disagree with this sentence. If you ascribe all the measured concentration enhancement to emission from the SEF region (as done with PSA), you will overestimate the SEF emissions (i.e. you project emissions elsewhere also to this region). So I think that both the Bayesian and PSA analysis are biased, but the latter more obviously.

Page 8226, line 19: Conclusions. I suggest, "Discussion and Conclusions", since some discussion is provided, e.g. on non-reported emissions.

Page 8227, line 2: "With a less accurate a prior field". I have not read this in the main paper. It is presented in Appendix B, but no reference to this section can be found.

Page 8227, around line 13: I find this hard to believe. See major comments.

Page 8227, around line 15: "It is thus shown". I think this is not convincingly shown in the paper. Maybe there are "signs" of emissions from Norway, but omitting ZEP from the analysis is strange then. Which station picks up the signal, and what is the resolution at which the emissions are calculated? The reported errors are anyhow 100%, so "shown" is way to strong.

Page 8227, line 26: "even higher". See remark above.

Page 8228, line 13: grids. Should be grid.

Page 8228, line 15: R_p. How defined? I understand to varied the grid on which emissions are derived (between 2000 and 6000?, how many simulations?). This gives a mean emission and variations. But what is meant with "maximum error"?

Page 8228, line 20: I think "comprised" can be removed. Does R_p now relate to the variance between the different scaling factors or to the variance calculated on the different emission grid (between 2000 and 6000)? Also, in table A1 I cannot find the reported values for the SEF region. Under which header?

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Page 8229, line 1: I thought the analysis was done only for 2008 and 2009?

Page 8229, line 5: In which column?

Page 8229, line 12: For the other regions the averaged Rp is around 40% (Table A1). Table A1 is about SEF.

Page 8229, line 14: Figure A2 is a mystery. The caption mentions 5 regions, while the number of points is much larger. Also, the numbers in the text do not seem to correspond with the figure. So I doubt if the correct figure is shown.

Page 8229, line 20-21. Again the values in the text do not correspond to table A1.

Page 8230, line 9. "The relative error reduction $1-Ea/Eb$ (see Table A2) for CMN and JFJ were -0.18 and -0.17, respectively, and for MHD was -0.42, showing that the two mountain stations are more influenced by the sources present in the study domain". I do not see from these numbers that the mountain stations are more influenced by the sources. I would expect actually a larger error reduction at CMN and JFJ if emissions are updated, because there is more to gain.

Page 8232, line 4-5. Note here that since the emissions are homogeneous, also the assigned errors are homogeneous. I find it rather surprising that specific hotspots are retrieved in this inversion (see also major comment #1). For instance, Greece, northern Africa and Norway. These areas are quite distant from the stations, so transport errors will be considerable. Again, at which resolution have these inversions been done?

Table 1: unit (%)? Should that not include a time unit?

Table A2: units are missing in general.

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 8209, 2014.