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Atmospheric Chemistry and Physics Discussions

Interactive comment on "An extended Kalman-filter for regional scale inverse emission estimation" *by* D. Brunner et al.

D. Brunner et al.

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We would like to thank the referee for the numerous comments which helped clarifying several points and improving the manuscript (referee comments in italics).

General comment:

I would like some more general statements about how this work fits in with previous papers that have tried to infer emissions and how much further we still have to go. I would like a better feeling of where we are in our ability to infer estimates from country- and continental-scale regions. You state how big interannual emission changes must be to "see", but with your analysis can you provide a better general feeling of how much we should believe the country specific and European estimates that you have

provided? Are we to the point now where we should believe your estimates more than the reported ones? As you rightly point out in the introduction, emissions estimates may become important to validate a future climate agreement. Can we do that now using your approach? Is it just a matter of a higher density of observation locations? I realize some of these answers cannot be answered by your current work, but any general summary information you can provide along these lines could be very helpful to those not directly in the inverse modeling field, but who want to use the results.

These are all very good but at the same time difficult questions which we can only partially address in the framework of this study. In the conclusions we will add a reference to the recent publication of Manning et al., "The challenge of estimating regional trace gas emissions from atmospheric observations", Phil. Trans. R. Soc. A (2011) 369, 1943–1954, doi:10.1098/rsta.2010.0321, which tackles these questions in a more general way.

Our ability in estimating emissions top-down is advancing due to better transport models and inverse estimation approaches, and uncertainties associated with these methods are increasingly well understood. However, properly quantifying these uncertainties is still a major challenge. In Section 4.1., for example, we note that the sensitivity analysis presented there does not cover potential biases in the transport model. Such biases are very difficult to quantify as there are too few experiments like the ETEX tracer release experiment to test transport models. High altitude stations like Jungfraujoch have the advantage that they can "see" (are sensitive to) a large domain but transport simulations are complicated by the complex topography and the fact that the smooth orography in the model is usually well below the true station altitude. To better address this important issue of potential biases we will add a few lines at the end of Sect. 4.1 as described in detail in our answer to the review of Christoph Gerbig.

Yes, we believe that a denser network of stations would allow us to estimate European emissions on a country-by-country basis to a degree required to be useful as a complement to, and independent verification for, bottom-up estimates in international agreements. A denser station network would also greatly improve our understanding of

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transport biases for certain station types (e.g. coastal stations versus stations inland, stations in flat terrain versus elevated stations, measurements at the surface versus measurements from tall towers, etc.). Such an analysis requires redundant information: individual regions need to be "seen" by several stations, but unfortunately this is not possible with the sparse halocarbon network in Europe. The benefit of a denser network regarding country-specific emission estimation was demonstrated (for methane) in the study of Villani et al. (2010) already referenced in our manuscript. A further important step towards reliable top-down emission estimation is multi-model-inversions as currently undertaken in the framework of the EU project INGOS. Comparing the results from different models is imperative to better understand and constrain model uncertainties. The approach presented in this paper will contribute to a multi-model inversion study of European halocarbon emissions in INGOS.

To address the points raised by the referee at least partially, we will add, in addition to the extended discussion of potential biases in Sect. 4.1, the following lines at the end of the conclusions section:
"A posteriori 1sigma uncertainties of country-specific annual emissions were typically in the range of 10-20% for HFC-125, 15-30% for HFC-152a and 20-40% for HCFC 141b (Table 5). A sensitivity analysis for HFC-125 indicated that country-specific emissions vary by less than 25% for different setups of the inversion including varying a-priori distributions and uncertainties, the application of a land-sea mask, and the addition of the station Monte Cimone in Italy. These uncertainties do not yet account for potential biases in the transport simulations which can be caused by several factors (Manning et al. 2011b). A particularly relevant uncertainty specific for mountain sites is the choice of particle release height. We determined the optimal release height for Jungfraujoch to be around 3000 m a.s.l. for the model configuration used, but higher or lower release heights within the range of uncertainty would lead to up to 20% higher or lower emissions, respectively.
Reported country emissions were often outside the uncertainty range of the a posteriori emissions. Together with the fact that for selected countries the agreement between

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estimated and reported values varies strongly between different compounds, this provides clear evidence for inconsistencies in the bottom-up estimates that can be readily identified by top-down methods.
The method presented here will be compared with other top-down approaches in the framework of the EC FP7 project INGOS. Such a multi-model approach is crucial to improve our understanding of model uncertainties and the ability to derive country-specific estimates. A robust quantification of uncertainties is needed to further strengthen top-down methods as complementary approach to the bottom-up emission estimates (Nisbet and Weiss, 2010)."

Specific comments and minor comments:

For the minor comments below, my wording suggestions are just that. Feel free to use other wording that is better.
Page, line 29196,18 – change to "higher and lower than reported emissions in different countries" Done

29196,21 – replace "ban" with "controls in developed countries". Ban implies a complete phaseout, which has not yet occurred.
Changed as suggested.

29198, 12-15 – since technology has already changed, consider rewording to "Without further regulation, their continued growth in the atmosphere led to a non-negligible contribution to radiative forcing equivalent to 7-12% of the radiative forcing of CO2 by the year 2050 in a scenario in which it was assumed that developing countries replace HCFCs in the same manner as developed countries have." or something to this effect.
Thanks for this good suggestion – changed.

29198, 18 – ozone assessments have used 1- and 12-box models as well to infer emissions.
Reference to WMO ozone assessment added.

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29198, 22 – clarify whether you mean all these studies or just the 3-D ones
Just the 3-D ones are addressed. We rearranged the sentence to make this clearer.

29198, 26 – replace 'lain down in' with 'provided by' or 'contributed by'
Ok, done

29198, 27 – change 'data' to 'dataset'
Done

29199, 26 – add "(" before "Keller"
Done

29201, 12 – change 'as fire extinguisher' to 'in fire extinguishing equipment'
Done

29201, 14 – insert 'a' before 'solvent'
Done

29203, 12 – replace 'angles' with 'directions'
Done

29205, 18 – I would like a little more explanation about the separation between the emissions contribution and the background. For example, you could say a little here about what contributes to the background. Also, clarify what you mean by assuming persistence for the emissions. Clearly emissions change over time for the various grid boxes. How is that consistent with the assumption of no change in emissions from time t-1 to time t. What time frame is the derivative of the background assumed to be fixed?
We will add the following sentence to explain the background: "The idea of including the background concentrations in the state vector is that an observed concentration can be decomposed into a (large scale) background plus a contribution from recent emissions as covered by the transport model simulation, in our case from the previous 5 days". Concerning the temporal evolution of emissions: If we had a priori information on how the emissions are changing with time we could easily build this information into

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the prediction model. By using a persistence model, however, we simply assume that we do not have such information and the best prediction, in that case, is that emissions remain constant. The error term in equation (1) nevertheless allows emissions to change with time such that the simulated concentrations more closely follow the observed ones. The larger this error term, the more rapidly the estimated emissions adjust to the observations. To clarify this we will add the following sentence: "When it is not known a priori how emissions are changing with time, a persistence model provides the best prediction for the next time step."

29210, 26 – change '10%' to '10th'
Agreed, this makes more sense.

29212, 26 – since North America is out of your domain, how does the inversion deal with this? The episodes would be too abrupt to be dealt with by the background term, wouldn't they be?
Yes, that's correct. However, we do not think that this would significantly compromise the emission estimation. During such an event the transport model would simulate near-zero sensitivity to European emissions and therefore the assimilation would have no means to falsely attribute the elevated concentrations to European emissions but rather try to adjust the background.

29214, 20 – clarify whether just temporally correlated or also spatially
Added "temporally" to make this clear.

29214, 19-22 – how would the answers have differed if the noise were added to the measurements (at the 2 or 3 sites) themselves? If you have not done this, this would be another important aspect of the problem, in order to understand the importance of imperfect observations at Mace Head and Jungfraujoch.
Such an investigation would not fit to this section but could indeed be valuable to characterize the importance of imperfect observations. We refrain from adding such an analysis for two reasons: First of all, measurement uncertainties are significantly

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smaller than transport model uncertainties and therefore not a limiting factor, at least for the species considered in this study. Second, the main aim of this paper is to present and demonstrate a new inversion method rather than to analyze specific measurement data sets such as those from Jungfraujoch and Mace Head.

29215, 6-29 – it might be helpful to have a figure that shows the spatial pattern of the retrieved emissions for one of the cases that doesn't work quite so well. With total RMS numbers, it is not possible to tell whether there is a systematic spatial pattern to the error. For example, are the errors largest where the emissions are largest? If a single pattern cannot be representative, perhaps some text could provide some more detailed information.
We will add two panels in Fig. 7 showing the emission distributions for cases 3 and 7b in the year 2010. The panels will be displayed to the right of the corresponding country emissions and panels 7b and 7d. The figures are indeed a good illustration of the increasing difficulty to reproduce the original emission distribution with increasing noise.

29215, 12 – remove 'a' before 'Gaussian' 29215, 14 – insert 'the' before 'case'
Done

29216, 7-10 – this a very important, well-made point. It is mentioned in the conclusions, but it might be worth giving a bit more emphasis in the conclusions.
We will add the following sentence at the end of the first paragraph in the conclusions: "A realistic description of these errors is crucial for optimal results and reliable a posteriori uncertainties."

29216, 17 – add 's' to 'emission'
Done

29217, 9-11 – it is hard to evaluate the comparison in this figure. Other options would be to show one curve and then another of the difference (percent or absolute), or keep the current figure and then include one shorter time period in an expansion inset. I

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leave it to the authors' judgment to determine the best approach.
This is a good suggestion. We will add panels zooming into the months Nov 2009 – Feb 2010 to the right of the existing figures.

29217, 15-16 – I do not see any discussion of the correlation numbers "with background" that are shown in the figure.
The following sentence will be added: " When including both the AR(1) term and the background concentrations, the correlations between simulated and measured concentrations (r with bgnd) are increased to 0.95 for both Jungfraujoch and Mace Head since a considerable fraction of the observed variance is due to the long-term increase which can easily be traced by the inversion."

29217, 26 – I do not recall reading whether these are 1-sigma or 2-sigma error bars.
Good point: The figure shows 1-sigma uncertainties. This information will be added.

29218, 14-16 – does this statement mean that there are plants in this part of France that manufacture HCFC-141b, and we just don't know how much they emit? Please clarify.
This was indeed not well formulated and will be replaced by the following: "France is known to have been one of the main producing countries of HCFC-141b in Europe. The large emissions from France are likely related to HCFC-141b release from these production plants for which a priori emissions were not available."

29218, 22-24 – can you provide an explanation of why this occurred?
This is explained further below (P29219, Lines 4-8).

29219, 4-7 – I find this statement confusing. If it is accurate, some additional explanation about the retrieval outside the European domain would be very helpful. Specifically, why do Southern Hemisphere values matter to your retrieval?
The sentence will be changed to "HFC-152a has a strong north-south gradient due to its relatively short lifetime and dominant sources in the Northern Hemisphere (HFC-152a is about 50% lower in the Southern Hemisphere) and probably has significant

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vertical gradients. Air masses originating from low latitudes are therefore associated with lower background values than air masses from high latitudes."

29219, 6-7 – I am also confused by this sentence. Doesn't it either reproduce the variability, or it doesn't because it attaches a large uncertainty. But it doesn't reproduce the variability by assigning a large uncertainty does it?
Assigning a large uncertainty to the (simulated) background indeed has the effect that the simulated background closely tracks the observed variations.

29219, 9-11 – quantify the size of the effect on emissions. Also, perhaps add more explanation about implications for conclusions. It seems this could be an important sensitivity.
Yes, this is an important sensitivity. The following sentence will be added on p29219: "As a preliminary solution we reduced the background uncertainty by a factor of three which leads to a smoother background and in turn to higher emissions: European emissions are on average enhanced by about 20% suggesting that the treatment of the background variability represents a major uncertainty in our HFC-152a emission estimates." We will also mention this sensitivity in the conclusions.

29219, 24-26 – can you add error bars to the increases? Otherwise, we cannot tell if the inversion estimates are consistent with the reported values or not.
No error bars are available for the reported values but we will add error bars to our estimates.

29220, 13-18 – this section needs clarification because it initially seems you are talking about a comparison with the Manning estimates.
This could easily be misunderstood, indeed. We have added a line break and slightly changed the following sentence to make clear that we are talking about our own results.

29220, 20-22 – but shouldn't this sen sitivity be reflected in the size of the error bar? In that case, there should be agreement within the error, or twice the error bar.
Yes, in principle this should be reflected by the error bar but our experience shows that

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emissions in regions close to the borders of the inversion domain are particularly sensitive to different inversion configurations (e.g. small changes in the inversion domain) and this sensitivity is often exceeding the estimated uncertainty.

29221, 4 – this could be read as Italy reports 0 emissions. Perhaps removing 'any' would help? The wording in the abstract and conclusions is clear.
Changed as suggested.

29223, 8-11 and general about entire section 4.1 – after reading this and the comment about the land-sea mask on Italy, I am left wondering how accurate the Italian estimates really are. Some additional discussion seems needed here, particularly to convince the reader that the 15-20% value for your ability to infer country emission estimates is accurate. Also, do you expect similar impacts for -152a and -141b for all these sensitivity cases.
Yes, we expect similar impacts for the other trace gases.
Note that the 15-20% value only refers to our ability to detect relative differences between two selected years. These numbers should not be confounded with an absolute uncertainty. It is reasonable to assume that biases introduced for example by the release height issue (see response to Christoph Gerbig's comments) will be similar in different years. Thus, even if our estimates were systematically biased high or low it would still possible to detect relative changes by 15-20%.

29223, 27 – change 'much' to 'many'
Done

29224, 6 – perhaps consider adding 'arising from our implementation of the FLEXPART model' or something similar to the end of the sentence.
This is a general statement not specific to our FLEXPART implementation and therefore we don't see the need to extend this sentence.

29224, 21-23 – this information would be useful in section 4.1
We prefer keeping the sentence there since otherwise we would have to duplicate the

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information.

29226, 11 – I believe this range arises from the 15% in the end of section 3.2. The range should be given there to be consistent with the conclusions. If it is not this straightforward, more explanation of the 15-20% range should be given before the conclusions.
Yes, this conclusion is indeed based on the results of Section 3.2. The last sentence in Sec. 3.2 was adjusted correspondingly.

29226, 22 – 'ban' – same comment as for abstract
Done.

29236 – make clear that cases 1-7 are synthetic and the HFC cases are 'real' in the caption. This helps if one looks at the table without reading through the corresponding text.
Thanks, good suggestion. This information will be added to the table caption.

29248 – in the top 4 panels, the lowest 2 values are both 0.01. I know why, but you may wish to change it so the bottom number is 0.007.
We will change the color scale to a true exponential scale to better highlight the large dynamic range of the estimated emissions.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 29195, 2011.