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Atmospheric Chemistry and Physics Discussions

Interactive comment on "A new analytical inversion method for determining regional and global emissions of greenhouse gases: sensitivity studies and application to halocarbons" *by* A. Stohl et al.

A. Stohl et al.

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We thank reviewer 1 for the thorough review of our paper. Below, we have repeated specific points of the reviewer in italics, followed by our responses in upright font shape.

The greatest limitation of the work so far is that the error in a priori emissions estimates is not propagated through to estimate uncertainty in the a posteriori emissions. This weakens the conclusions that can be drawn from the summary tables 4-7. In addition, it seems somewhat dangerous to quote the emissions estimated per country (Tables 5-7) and their change between 2005 and 2006 without obtaining a rigorous uncertainty estimate for each number. For example, can we really conclude that German

HFC-134a emissions increased from 2005 to 2006 (Table 4) or are the figures indistinguishable given the uncertainty? Furthermore, as stated in the text, the attribution to each country is uncertain, especially when emissions are close to borders. In the case of Germany are the emissions really half the level derived by UNFCCC or are the a posteriori estimates for this country too uncertain to tell?

We fully agree with the reviewer's assessment that the missing uncertainty estimates in the output is currently the biggest limitation of our method. We shall work on implementing an estimation of the uncertainty of the a posteriori emissions, but this will take more time. It should also be considered that, unfortunately, a formal error propagation is not the ultimate answer towards estimating uncertainty and the significance of the emission adjustments. The fact that we do not have good uncertainty estimates for the a priori emissions and the "observation" error is a major limiting factor. A further complication is that these errors are not normally distributed, as we have shown for the observation error. Thus, a formal error propagation may give wrong and possibly misleading results. For these reasons and because we are currently lacking a formal error propagation scheme, we have tried to give the reader an understanding of the uncertainty by performing a series of sensitivity studies. The uncertainty problem is not yet solved and even if we are going to add a formal estimate in the future, this would not replace such other approaches.

We address the point regarding the reporting for individual countries in our response to the next comment.

On p.19088, l.20 the authors subjectively estimate errors in the emissions. How were these numbers estimated? You need to justify them. I recommend quoting large uncertainties which you feel would safely encompass the true value and then use your future work to reduce the uncertainty and estimate it more precisely. These numbers need to be quoted in Tables 4-7 so that readers do not draw misleading conclusions.

The uncertainties were derived based on the results of the sensitivity studies. In the

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paper, we have added the remark "...from the range of results obtained in the sensitivity experiments we subjectively estimate...". Based on the sensitivity studies, the 20-30% errors are a comfortable uncertainty range. However, we have also added the remark: "For smaller regions (e.g., individual countries), errors may be larger."

In the caption of Table 5, we have added: "Estimated uncertainties of the a posteriori values are generally 20-30% but may be somewhat larger for countries with substantial emissions close to borders with other countries (e.g., Germany). Uncertainties in the relative changes from the year 2005 to 2006, however, are likely to be only about 20% since the geometry of the observation network has not changed between the years."

New support for our country-derived values also comes from a recent study of Millet et al. (2009), now cited in the paper, who reported emission values based on a completely independent data set (aircraft measurements) and method (based on ratios to carbon monoxide) that agree, for all species, within the combined uncertainty ranges with our results. Regarding the specific case of Germany, we have mentioned in the text that figures for this country are somewhat more problematic because of the large emissions occurring close to Germany's borders, such that the attribution of emissions to Germany and neighboring countries is often ambiguous. We still think that the relative changes from the year 2005 to the year 2006 are realistic and informative.

At no point do you justify the use of 20 day backward simulations. What motivated this choice? Would you use longer simulations if you had the resources? At what point does trajectory length become irrelevant?

At the end of section 3, we have added the following explanation:

"The choice of the 20 d length of the backward simulations was motivated by the fact the value of every additional simulation day decreases rapidly with time backward. This has three reasons:

1) Since we were using data from surface stations, the total emission sensitivity in the

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footprint layer per day of backward calculation is largest shortly before the arrival of the air at the receptor. Before, particles may have resided above the boundary layer, in which case they do not contribute to the emission sensitivity.

2) Due to turbulent mixing and convection, the volume (or area) over which emission sensitivities are distributed grows with time. This makes it more and more difficult with time to extract information on individual emission sources (this is a consequence of the second law of thermodynamics). In other words, the emission contributions from various regions become more and more well mixed and start forming the baseline.

3) Model errors also grow with time.

On the other hand, the computational cost of the model calculations per day of simulation even increases slightly with time because the convection scheme must be called for a growing number of grid cells. All this suggests a relatively early termination of the backward calculation. Model experiments show that for most stations a duration of about 5 d is sufficient to explain most of the concentration variability. The extra 15 d add relatively little to the concentration variability and, thus, longer simulations than 20 d would not result in a better reconstruction of emission sources. Notice, however, that the baseline as defined below depends on the duration of the simulation: longer simulations result in a lower baseline, as more emissions are directly accounted for."

p.19069, l.20: Use either Ny-Alesund or Zeppelin.

Right. We have changed everything to Zeppelin.

p.19073, l.11: Why use a stepwise linear segments of 31 days long? Is there a relation to the 20 day trajectory length? What are n1 and n2?

In section 4.3, we have added: “Notice that the 31 d averaging interval is a compromise between the desired capacity to describe temporal variations of the baseline and the need to limit the number of unknowns in the inversion. Notice also that this interval is not related to the 20 d duration of the backward calculations.”

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n_1 is the number of 31-day intervals, n_2 is the number of emission grid boxes, $n = n_1 + n_2$ is the total number of unknowns in the inversion.

p.19073: Although you could refer to this method as Bayesian, once all the approximations to the PDFs (i.e., normally distributed, uncorrelated errors etc) have been made it looks like a 3D-VAR algorithm to me, boiling down to a least squares cost minimisation problem. It would be worth adding some references to the literature on data assimilation for numerical weather prediction (you appear to be using the standard notation of that literature).

The different approaches to inverse problems are all related and, can be shown to be equivalent under suitable assumptions. The literature we were actually using has already been mentioned. We are now adding one more sentence at the end of the first paragraph in section 4.1: "For a more detailed discussion of chemical data assimilation and inverse modelling, see **?** and **?**."

p.19081: Why refer to the zero prior emissions assumption as Tikhonov regularisation? On its own this is a rather meaningless statement that would baffle unfamiliar readers unless connected to the relevant literature where Tikhonov regularisation is used.

We have removed the remark about the Tikhonov regularization.

p.19094, l.14: A time-lag between emission and reported consumption is used to explain unexpected results for HCFC-22. Why not adjust for a two-year time-lag and repeat the inversion to show that you can obtain more consistent results?

The two-year time lag seems to be compatible with the inversion results but also seems to be a "logical" explanation (i.e., intuitively, we would expect some delay). Unfortunately, we have no a priori information on the actual time lag between consumption and emission and using the a posteriori results to change the a priori would be circular reasoning. Therefore, we have refrained from adjusting for any time lag in the a priori. However, this point should be kept for possible future refinements.

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p.19095, l.1: You point to the large increase in emissions for Central America resulting from inversion in Fig.11. This also appears for the other two halocarbons, together with a strip of very high increases down the western flank of the Andes. Given that Fig.1 shows a very low SRR in this region, how much of this would you consider to be realistic? Is there a consistent problem with the method along the high orography of Central and South America?

The topography is probably somewhat problematic for the model calculations, however it does not alone explain why emission increases occur. One problem is probably that a priori emission uncertainties are chosen too high for this region, allowing relatively large changes to be made even though the overall emission sensitivity is low.

p.19095, l.6: Expand LPDM here to help the readers.

Done.

p.19096, l.5: In my opinion, these are the most important conclusions to draw in this paper. Given that resources are limited, where would you set up new stations in the network (in priority order) and how much would they have the potential to improve emissions estimates?

We agree that these are important conclusions and we think that we have given clear recommendations how stations should be sited to be of maximum use for inversion studies. We have also listed the regions lacking constraints most urgently: Africa, large parts of South America, India, Indonesia and northern Australia and give specific recommendations by having added the sentences:

"Most urgent, however, would be stations in tropical areas where constraints are currently missing almost entirely (see Fig. 1): probably on the Maldives to constrain emissions from India, at the Pacific and Atlantic coasts of northern South America to constrain emissions in South America, at the Cape Verde islands to constrain emissions in Africa, or in northeastern Australia to constrain emissions in Northern Australia

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and Indonesia. However, before deciding on a location, backward model calculations should be done specifically for candidate sites to find out which options would yield the best overall improvement of the global emission sensitivity distribution (Fig. 1). In this context, it could be interesting to make use of backward transport calculations carried out routinely by the Comprehensive Nuclear Test Ban Treaty Organisation (CTBTO/PrepCom) for its global network of radionuclide monitoring stations (Wotawa et al., 2003; Becker et al., 2007. This network is also interesting because at these sites the necessary infrastructure is already available."

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