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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.

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We would like to thank Christoph Gerbig for the positive remarks and constructive comments. Below we present a point-by-point reply (referee comments in italics).

General Comments:

The authors correctly state that the estimates are subject to bias errors in transport, and that this is not accounted for in the estimates. May be this should be discussed a bit in context of the uncertainties stated in the paper for the emissions.
We agree that this important point should receive some more attention. We will add the following sentence at the end of section 4.1:
"Significant biases can be introduced for example by errors in the estimation of bound-

ary layer heights and corresponding vertical mixing (Gerbig et al. 2008). This would be most problematic in cases with strong local sensitivities and emissions, which in addition might undergo strong diurnal variations, but it is probably less relevant for the remote sites and more constantly released compounds considered here. A potentially important source of biases specific for mountain sites is associated with the choice of release height. The study of Folini et al. (2008) investigated the sensitivity of transport simulations for Jungfraujoch to different parameters in a mesoscale Lagrangian particle dispersion model and concluded that the choice of release height had the largest impact on the results. Consistent with their study we find a major reduction of the footprints by on average 40% when the release height is increased by 580 m from our preferred height of 3000 m to the true station altitude of 3580 m. Estimated HFC-125 emissions increase accordingly by approximately 40% when releasing particles from 3580 m. Assuming that the optimal release height is known only within a range of $\pm$300 m, the uncertainty in emissions associated with this parameter can roughly be estimated to $\pm$20%. A detailed sensitivity study for this critical parameter is beyond the scope of this manuscript but will need to be addressed in the future."

Specific comments

Pg 29196 Ln 16: "Kyoto protocol" you probably mean UNFCCC
Yes, changed.

Pg 29204 Ln 7-11: Selection of release altitude: the authors should comment on how the amplitude of simulated CO depends on the release altitude. I would assume that not just the correlation coefficient changes, but also the slope of modeled vs. observed enhancements above background.
Yes indeed. The correlation coefficient was not the only measure we investigated but also root mean square errors and the amplitude of variations in simulated as compared to measured CO. These measures gave essentially the same answer but less clearly than the correlation coefficient. We tested this not only for 2008 but additionally also for 2009 and again found the best results for a release height of 3000 m. Instead of

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elaborating on this issue in this section we will simply add a line “The sensitivity to this choice will briefly be discussed in Section 4.1.” and provide a more detailed analysis in Section 4.1 as described above.

Pg 29210, Ln 17: May be to the reader with less experience with Kalman filtering the authors can comment on the multiple use of the same observations in three iterations of Kalman filtering, and how this impacts the resulting uncertainties in the emission estimates. Are those posterior emission uncertainties still realistic, or do they keep decreasing with each iteration?

No, the posterior emission uncertainties do not keep decreasing due to the prediction equations (4) and (5) which increase the uncertainty of the state as it moves from time $k-1$ to k. This is an important difference from a Bayesian inversion which indeed deserves more attention. In Sect. 4 we will add a new figure (Fig. 8) illustrating the evolution of the HFC-125 emission uncertainties (based on the trace of matrix P_k) together with the following text:

“In order to demonstrate the effect of the three iterations applied (see also Sect. 2.4) on the estimated uncertainties, Fig. 8 shows the evolution of the mean relative uncertainty of the HFC-125 emissions. The simulation starts with a uniform a-priori uncertainty of 200%. After the first iteration, i.e. after assimilating the observations of the period 2006-2010 for the first time, the average uncertainty is reduced to about 62% on average. During the second iteration it is slightly reduced further to 58% but remains constant thereafter, indicating that the uncertainty has essentially reached an equilibrium level already after the first iteration. Using the same observations repeatedly does thus not lead to unrealistically low uncertainties because in equilibrium the error reduction due to the assimilation (Eq.12) is exactly compensated by the error increase due to the state prediction (Eq.5).”

Pg 29210, Ln 27: “The respective initial uncertainties are set to 0.1% and 0.001% of the background concentration.“ This is unclear, does the 0.1% apply to the background, and the 0.001% to the trend? Then with a zero initial trend this would not really make

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sense.
We have rephrased the sentence to: “The respective initial uncertainties are set to 0.1% of the background concentration and to a 0.001% change of the background concentration per 3 hours.”

Pg 29211 Ln 24: Here the first reference to a table is to Table 4, so the tables should be reordered accordingly
Since the table better fits to section 4 we would like to keep the order of tables. We will remove the reference to Table 4 at this place and simply refer to Section 4 instead.

Pg 29217 Ln 20: replace "parameters settings“ with "parameter settings“
Changed.

Pg 29219 Ln 10: is the prior to posterior increase in RMS differences in table 3 (HFC-152a for MHD) related to the choice of a factor three smaller than optimal background uncertainty?
No, this result turned out to be due to a numerical instability. A closer investigation of the issue showed that this instability is related to the design of the prediction error covariance matrix Qk (Eq. 15 and 16). Instead of representing the spatial correlation by a Gaussian distribution it makes much more sense to describe this relation by an exponential decay: $\mathbf{q}^{\mathrm{e}}_{k,ij} = \eta^2_{\mathrm{e}} \cdot e^{-d_{ij}/d_{\mathrm{s}}}$ where d_s is the length scale. With this change the results for HFC-152a are significantly improved and robust. We have therefore decided to change equation (16) to the above formula and to repeat all simulations which entailed major adaptations: All figures 4-10 and tables 2-6 had to be regenerated and the text had to be adjusted at several instances to reflect these changes. This had not only a positive effect on the HFC-152a simulations but also on some other results, in particular those for HCFC-141b. We consider the new results as a significant improvement. The main conclusions of our study remain unchanged.

Pg 29221 Ln 28: add "in“ between "model“ and "Derwent“
Changed.

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Pg 29221 Ln 28: Why was the impact of the background prediction error not included in the sensitivity analysis, given that it has an impact on the magnitude of the estimated emissions?
Different from HFC-152a, results for HFC-125 were quite insensitive to different settings for the background prediction error. Therefore, this was not further investigated here.

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