

## ***Interactive comment on “Hydrochlorofluorocarbon and hydrofluorocarbon emissions in East Asia determined by inverse modeling” by A. Stohl et al.***

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We thank reviewer 2 for the comments on our paper. We repeat the specific points raised by the reviewer in italic font, followed by our responses in normal font.

*- Given the long life times and the global nature of most of the emissions, I wonder how 1) background concentrations have been taken into account, and 2) what the associated errors are. On page 2094 it is mentioned that results are only marginally sensitive to 'including' data from Mace Head and Trinidad Head, but indeed a large part of the measured concentration must derive from Asian emissions.*

Since we use the method exactly as in Stohl et al. (2009) where an extensive description (Section 4.3: The baseline definition) can be found, we do not want to extend the

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method description in the present paper. This would be highly repetitive of the Stohl et al. (2009) study.

Monthly baseline values for every station are estimated by the method as part of the optimization process. The baseline is defined as the measured value minus the contribution of emissions from the past 20 days. In practice, a priori baseline values are defined based on the lowest measured values in a month minus the simulated contributions during the last 20 days for these cases, and they are subsequently adjusted by the algorithm to improve agreement between the model and the measurements. Thus, while a large part of the measured concentration even at Mace Head is probably originally from Asian emissions, Asia would contribute mostly to the baseline concentrations at Mace Head, which bear no (or little) regional information. Information on regional emissions is extracted with our method solely from the enhancements above the baseline and, in the case of Mace Head, these enhancements are mainly due to European and North American emissions.

The largest source of error arises from the fact that the baseline is determined only on a monthly basis. Thus, episodic variations of the baseline are neglected. In East Asia, this can occur when air from the Southern hemisphere is transported to the station. During such events, the measured concentrations can be even slightly below the monthly baseline. However, the enhancements above the baseline during pollution episodes are sufficiently large that this error source does not invalidate the results. It is difficult to quantify the influence of the baseline calculation on the final results but we estimate that the associated systematic error is below 10%. Nevertheless, this is an area of active development work where future improvements of the method are planned.

*- p. 2126 and section 2.1 what exactly is given in the foot-print sensitivity in Figure 1. Is this the average of the footprint for 4 stations?*

Yes, this is the average footprint sensitivity for the four stations.

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- p. 2096 the authors loosely mention 'excellent' agreement among various measurements techniques, but can they explain what that means, and how much the impact on the inversion would be.

We have added the sentence: "Intercomparisons between the NIES and the AGAGE standards performed at Hateruma during the year 2008 yielded ratios of 0.987, 1.006, 0.999, 0.995, 1.010 and 1.003 for, respectively, HCFC-22, HCFC-141b, HCFC-142b, HFC-23, HFC-134a and HFC-152a."

-p. 2096 Explain better the methodology of removing negative emissions.

Again, an entire subsection (4.2 Positive definiteness) is devoted to that in Stohl et al. (2009). We see no point in repeating that information but we will extend the sentence describing this to read: Finally, negative emissions which can occur in the solution are removed in an iterative scheme where the uncertainty of the concerned emissions is decreased step by step, therefore binding them more closely to zero.

-p. 2096 information on meteorological dataset and resolution should be given here (now in 3.1). Do the 1377 'boxes' correspond to 1x1 degree?

Information on the meteorological data set (from ECMWF) is given at the beginning of section 2.2. We have added the resolution of the meteorological data (1 × 1).

Regarding the emission grid, we have added: "The inversion is performed for a variable-resolution emission grid. Emission grid boxes in the vicinity of the stations have a maximum resolution of 1 × 1, whereas further away the resolution is lower." Again, details on how the variable-resolution grid is constructed can be found in Stohl et al. (2009).

p. 2097 Explain why a distribution of emission by population is a good proxy, especially in the context that there are limited production facilities. In fact it is mentioned on p. 2102 that this is unrealistic. Is there any temporal variation assumed? What are the a-priori errors of the emissions: gridbox errors, I didn't understand 'at least 100% of

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#### *global emissions'*

The distribution according to population is probably not a good proxy, especially for species that are emitted mainly during production processes (e.g., HFC-23). Unfortunately, no emission inventories are available for most species (and for those species where an inventory, EDGAR, is available, the distribution appears also to have been made according to population). Information on production facilities and their respective emissions are also unavailable, incomplete or confidential. The distribution according to population is, therefore, the best one can do. A uniform distribution, for instance, would be even worse.

We have assumed that emissions are constant throughout the year. This is probably not a bad assumption, since neither production nor consumption of halocarbons should vary with season.

The assumed a priori emission errors were not well described. This is now changed to read: "We assume that all the a priori emission fluxes have an uncertainty of 50%. To allow the inversion algorithm to make reasonably large adjustments even in grid cells with small (or zero) a priori emissions, we prescribe a minimum uncertainty of at least 100% of the global mean emission flux."

-p. 2100 correlations are determined for 3 hourly data? I would call the correlations great.

Yes, the correlations are determined for 3 hourly data. We have added that information. Some of the correlations are good but bear in mind that these are relatively simple substances to simulate, for which emissions can be assumed to be nearly constant, which have no complicated natural sources, and for which we can ignore loss processes. Therefore, a reasonably good model performance can be expected.

-Section 3.2 There are various pieces of information in the results section that I think would better fit in the introduction or emission sections. E.g. what the use of the various

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*emission is.*

We have actually tried this before but it didn't read well. One problem is that the emission information used is different for every species. Therefore, describing all that together in the Introduction or emission section would require subsections for the various species. Otherwise this would not be clearly structured. This would extend the length of the article. More importantly, the a priori emission information used is important for the inversion results, so that we think it is best to not separate the two. Otherwise, the reader would have to switch between sections all the time to know which emission information has been used for a particular species. We prefer to keep the current structure.

*-Conclusions section: To me the errors on the inverted emissions (in the order of 10%) seem rather small. Can the authors comment on how/whether model errors are included as well?*

A detailed description of the error characterization is given in section 4.6 (Observation-related uncertainties) of Stohl et al. (2009). The "observation errors" contain both the measurement errors and the (larger) model errors. They are determined according to error statistics based on the a priori model results and are subsequently refined. However, the reported errors only characterize quantifiable stochastic errors. In addition, there may be systematic errors such as resulting from systematically too low or too high boundary layer heights, etc. Thus, the total errors may be larger than reported. However, given the rather successful model simulations, we do not expect uncertainties of 50% or larger.

We have slightly extended section 2.4 (Uncertainties) to better explain that the reported uncertainties do not include possible systematic errors.

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