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Interactive comment on "Global and regional emissions estimates for HCFC-22" *by* E. Saikawa et al.

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Thank you, Prof. Krol, for your comments on our paper. We appreciate your time to read our manuscript and give us these comments. Below please find our answers to your questions.

> I find the paper well written and only have some major comments on the treatment of the prior and posterior errors. Also, I miss prior and posterior statistics on the stations. I think this information should be included.

Thank you for this comment. We calculated the root-mean-square deviation (RMSD) of differences between the monthly-average observed mixing ratios and the modeled mixing ratios using both prior and posterior emissions. We provide these values in

Supplementary Table S1 in the revised manuscript.

Major points:

> In Table 2 posterior global emissions are presented for the global and the regional inversions. Very striking are the larger posterior errors in the regional inversions on the global numbers. Since more data are used in the inversions I would expect a larger error reduction. In combining regions, errors should be calculated taking into account the full co-variance matrix. This leads to a reduction of the error on the global scale compared to regional scales, which is perfectly logical, because the combined observations constrain global emissions better than emissions in individual regions. It seems that the errors on the regional scale are simply added to calculate the global error. Since there are correlations between the regions, this is a wrong procedure.

Thank you for pointing out a mistake in our error calculation. We have recalculated the errors taking into account the full co-variance matrix and we now provide those numbers instead in the revised manuscript. The revised error values are 24.1, 23.3, 22.6, 23.4, and 26.1 for 2005 to 2009, respectively, as listed in Table 2 and 3 of the revised manuscript. These error reductions from the regional inversions are smaller than the values we calculate from a coarse-resolution global inversion. The reason for this is because we include measurements with pollution events for the regional inversions, and the standard deviations at some of these stations are much larger than those at background stations used in the global inversion.

> Likewise, the prior error settings in the regional and global inversion should be consistent. Globally, an error of 40% is chosen. If you would split this up in two (uncorrelated) regions with equal emissions and also errors of 40%, the error on the global scale becomes larger than 40%: sqrt(402 + 402) = 56.6%. Error in non-equal regions (in terms of emissions) should be transformed in Gg/yr of course to arrive at an error in Gg/yr on the global scale.

Thank you for this comment. We have set up the prior error in the regional inversion

consistent with the global inversion by making the prior error as follows, assuming no correlation between the regions: 20% for North Asia, Europe, and Oceania (developed countries); 40% for South America and Middle East/Africa (developing countries); 60% for the 4 regions in the U.S. (placed higher uncertainty as these are further disaggregated regions within a country), and 90% for Article 5 Asia (this region has the largest estimated emissions and there is uncertainty associated with the estimate). We have modified the text in the revised manuscript on p. 11 as follows to incorporate this change:

For the regional inversion, we assume 20% uncertainty for our prior values for the emissions from developed countries, 40% uncertainty for South America and Middle East/Africa, 60% uncertainty for the 4 regions within North America, and 90% uncertainty for Article 5 Asia. This range is justifiable as there have been higher uncertainties in HCFC-22 emissions, especially in recent years after the increase in consumption and production in developing countries. In addition, assuming the emissions are uncorrelated among the regions, the total in the regional inversion is consistent with the 40% uncertainty in the global inversion.

> In the global inversion, only background observations are used to derive global emissions. In doing so, "pollution" events are screened out from the measurements. On the model side, the model is sampled over the ocean, to prevent sampling on continental emissions. However, if the wind blows from the continent, some pollution events in the model cannot be prevented. Ideally, model and measurements should be "cosampled". The authors should proof that their model is "pollution-event free" to allow a sound comparison to screened observations.

Thank you for making this point. We have conducted another global inversion including the pollution events and included it as a Supplementary Figure 1 in the revised manuscript. As you can see, there is very little difference between the two sets of results. We modify the text as follows on p. 18 to include this inversion result:

In addition, we conducted another inversion including the pollution events (see Fig. S1) and the results are similar to what we found excluding the pollution events.

Minor points:

> Title: emissions estimates: replace by "emission estimates". Also at some other places.

We have replaced them with “emission estimates” everywhere in the manuscript.

> page 18246, line 22. I suggest: Due to its short lifetime "for an ozone depleting compound".

Thank you for this suggestion. We have changed as you suggested.

> page 18247, line 9 ... Montzka (2009) ...I re-read the sentence a couple of times, but it remained unclear. Rephrase.

We have rephrased the sentences as follows on p. 5 in the revised manuscript:

In contrast to the developed countries, which were required to gradually phase-out HCFCs, developing countries have now become major consumers and producers of these species. For example, Montzka et al. (2009) calculated that 79% of the reported annual global total of HCFCs production (consumption) in 2006 (UNEP, 2007) is from the developing countries. Furthermore, this production (consumption) value is equivalent to what was reported as the global total during the 1990s.

> page 18252, line 5. Emissions inventory ...replace by.... emission inventory

Thank you for this. We have replaced it by “emission inventory.”

> page 18254, top:specify how lifetime is calculated and provide the lifetimes for OH and O1D loss also separately

Thank you so much for this suggestion. We calculate the lifetime as the ratio of the annually averaged global burden of HCFC-22 by the removal rate due to OH and O1(D).

We have quantified the lifetime of HCFC-22 by OH and O1(D) separately and they are approximately 12.2 and 600 years, respectively. We rephrased the sentence as follows on p. 12 of the revised manuscript:

The lifetime of HCFC-22, calculated by the ratio of the annual total global burden to the loss rate calculated in the chemical transport model, is approximately 12 yr, which is consistent with the current estimates of its lifetime (Montzka et al., 2011). The lifetime of HCFC-22 due solely to OH or O1(D) is approximately 12.2 or 600 years, respectively, illustrating the importance of HCFC-22 due to OH as we expect.

> page 18254, l13: For both inversions... Sentence unclear

We rephrased it as follows on p. 13 of the revised manuscript:

For both inversions, when there were measurements from multiple different networks, we combine the datasets to create monthly averages and standard deviations at a site using the number of measurement-weighted averages.

> page 18255, Please specify that matrices W and S are chosen diagonal

We inserted that they are diagonal on p. 14 in the revised manuscript.

> On page 18257 it is stated that "Because of the difference in the number of measurements in a month between high-frequency observations and weekly flask measurements, this error is approximately three to ten times lower for high-frequency observations, compared to that associated with NOAA and AGAGE flask measurements". I could easily think of a time series for which the standard error of continuous measurements is larger, e.g. a large day-night difference, pollution events etc. In contrast, flasks are normally sampled under baseline conditions. Please provide examples for typical stations

Thank you for this comment. Standard errors become smaller for high-frequency measurements when compared with the flask measurements at the same site, and this is not the case if we compared standard error of high-frequency measurements at a very

polluting site to that of flask measurements at a background site because of the reason that you mention above. Since this sentence caused confusion we rephrased it as follows on p. 15 in the revised manuscript:

Because of the difference in the number of measurements in a month between high-frequency observations (every 2-hours) and weekly flask measurements, this error is approximately three to ten times lower for high-frequency observations, compared with the error associated with NOAA and AGAGE flask measurements at the same site. Even when we assume a 10-hour serial correlation for the AGAGE in situ measurements (resulting in approximately 70 uncorrelated measurements in a month), it does not affect the inversion results in any substantial way (the largest difference being approximately 2% change in optimized emissions).

> On page 18259:These 12-box estimates are shown in 3-yr averages, and thus they are much smoother than our estimates. I directly wonder why? Remove the 3-yr average to allow for a fair comparison

We have now included the 12-box estimates without the 3-yr averages and present them in Figure 3 and Supplementary Figure S1 in the revised manuscript.

> Page 18260: at least for years between 1995 and 2009. I see in figure 3 only green symbols up to 2004.

The green symbols are the bottom-up estimates by the previously published study. What is referred to here is our priors (red line in Figure 3), rather than the green symbols. We rephrased the sentence as the following to make this clearer on p. 19 in the revised manuscript:

The result reconfirms the need for further research, but it also indicates that the consumption-based emission estimates that we created here (both “raw” and polynomial fit priors) give good approximations for HCFC-22 emission trends, at least for years between 1995 and 2009.

> Please find a way to make the error bars in figures 6-10 more readable.

Thank you for this suggestion. We have modified the figures so that the error bars are more readable. Please see Figures 6a-e in the revised manuscript.

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 18243, 2012.