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## Interactive comment on "Atmospheric observation-based global SF<sub>6</sub> emissions – comparison of top-down and bottom-up estimates" by I. Levin et al.

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We wish to thank the reviewer for his/her kind and helpful comments. We will change the editorial and wording corrections as suggested; for the more substantial concerns and questions, our replies are given below.

Rev. 2: The authors have succeeded in writing their story down in an accessible, credible, and attractive way and I recommend publication of this paper provided that two points of criticism are dealt with adequately in the final version.

Rev. 2: (1) On page 26658 the authors state that the the latest EDGAR version has used some of the atmospheric mixing ratio information already to come up with emis-C11305

sion totals, but to what extent is "not clear". This is very unsatisfactory given that this paper continues after that to compare EDGAR and UNFCCC emissions to atmospheric derived ones and hint at which product might better reflect the reality of the independent atmospheric approach. It seems to me that it would not be hard to provide the readers clarity on how much atmospheric information went into EDGAR4. A simple call to John van Aardenne at the Joint Research Center would probably suffice. I would like to see this issue resolved.

We have now received the information from Jos Olivier who has made the EDGAR SF6 estimates: He confirmed that atmospheric observations were included in the global total of the EDGAR estimates and we will thus include this information in a revised manuscript.

Rev. 2: (2) In the comparison of simulated and observed SF6 distributions it is shown that the UNFCCC emission based values tend to underestimate the Alert-Neumayer gradient. Accounting for a 25% uncertainty in N-S exchange overcomes some of this deficit, and it is suggested that further sensitivity might lie in the N-S distributions of the assumed SF6 emissions with non-Annex I country emissions generally closer to the equator. I would like to know how large that sensitivity is, which one could calculate with some simple model runs. For instance, would the Alert-Neumayer gradient be simulated more accurately with the UNFCCC scenario if one did not assume all the non-reported emissions to be in non-annex I countries but also 10% in annex 1 countries themselves? Or even better: could one construct a scenario that divides the non-reported emissions between annex I and non annex I countries such that the emissions/GWH are more reasonable? How would that scenario do in Figure 3?

When evaluating our measurements and comparing them with reported emissions we indeed made a large number of such sensitivity tests. At that time (autumn 2008) we did our separation with Annex II and non-Annex II countries (Annex I includes all Annex II countries as well as Eastern Europe, Turkey and some small Western European countries); the difference to Annex I countries in reported SF6 emissions is less

than 5%). Emission scenarios that were able to reproduce the observed north-south difference required for example an increase of Annex II emissions by 50-75 % (see Figure 1 below). So, it is certainly possible to construct (many) scenarios that fit the observations but the spatial resolution of our data basis and the GRACE model are not adequate to pick a "most accurate one". Therefore we decided to skip this part of the discussion from the paper.

Rev. 2: Title: The current title is a little bit hard to interpret as the words "atmosphericbased", "top-down" and "bottom-up" are not uniquely defined across research fields. I suggest to select a title that more strongly reflects the main message of the paper, as reflected best in the last sentence of the abstract.

We will change the title to

"The global SF6 source inferred from long-term high precision atmospheric measurements and its comparison with emission inventories"

Rev. 2: abstract: I suggest to add a sentence on the disparity between emissions/GWH in the UNFCCC reported emissions, and use it to already in the abstract make a statement about the importance of long-term monitoring as a verification mechanism.

We will add the sentence: This suggests a strong under-estimation of emissions in Annex I countries and underlines the urgent need for independent atmospheric verification of Greenhouse Gases emissions accounting.

Rev. 2: page 26661: Could you please add the inter-hemispheric exchange time of GRACE in the standard and 25% scenarios in units of yr-1.

The mean inter-hemispheric exchange time in GRACE is 0.95 years. To account for a possible bias in the way we determined inter-hemispheric mixing in GRACE, it was varied for the sensitivity test in Figure 3b between 0.71 and 1.19 yrs.

Supplementary Material:

## C11307

Rev. 2: Section 3.3 describes a very detailed and complicated method to construct long-term time-latitude-altitude distributions of SF6 to estimate the global burden, as well as the growth rate. I can see why the burden (=emissions) needs to be estimated taking into account the latitudinal+tropospheric+stratospheric gradient. But for the emissions increase (=growth rate) one might take the more conventional method of taking the time derivative of a representative background site. How much does the third column of Table 1 in the main text deviate from a simple site-derived growth rate?

The differences from the "more conventional method", i.e. taking the growth rate of a background station are approximated by the differences between growth rates in Figure 1 of the manuscript. They are largest during periods of large changes (i.e. between 1995 and 2000). Taking a site in mid southern latitudes for this estimate, e.g. Cape Grim, would be best to meet the global growth rate, however, it will still not account for strong changes happening elsewhere.

Rev. 2: Did you consider comparing your derived vertical/time structure of SF6 to the available observations from the NOAA aircraft program that has collected many SF6 samples between 0-6 km since 2002?

No, because the TROPOSPHERIC vertical gradient of SF6 in background air is only very small due to the fast mixing. However, these data may well be used to verify vertical mixing in high-resolution models. Important for estimating the global SF6 burden is the stratosphere where mixing ratios strongly decrease due to the slow vertical mixing in the stratosphere.

Rev. 2: In section 3.4 a comparison is shown between emissions (inverse growth rates) derived from several different networks/measurements of SF6. I have seen such a graph previously presented by Brad Hall from NOAA ESRL and was surprised to see the large spread in the temporal behavior of each network. For completeness, I would like to see the growth rate figures themselves for each network, with the NOAA ESRL record included in its entirety, i.e., flask and continuous records up to 2009.

We want to leave the comparison of our source estimates (Table 2) with those from NOAA and AGAGE data to the groups that measured these data. We decided not to use unpublished and potentially not rigorously calibrated/corrected measurements from other groups in the present work, although they may be publicly available. We have seen NOAA results being presented at the WMO meeting in Jena last September (http://www.bgc-jena.mpg.de/service/iso\_gas\_lab/IAEA-WMO2009/Posters/Hall\_poster\_2009\_WMO.pdf) and AGAGE results at the British Royal Society Meeting in February 2010 (http://royalsociety.org/Greenhouse-gases-in-the-Earth-system-setting-the-agenda-to-2030/) and these emission estimates, although partly more variable, essentially confirm our findings.

Caption Figure 1:

Observed (red dashed line) and modeled (black and blue lines) north-south difference of tropospheric SF6. The black line was obtained with an emission scenario where Annex II emissions were increased by 50% while for the scenario resulting in the blue line Annex II emissions were increased by 75%.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 26653, 2009.



