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Atmospheric Chemistry and Physics Discussions

Interactive comment on "History of atmospheric SF_6 from 1973 to 2008" *by* M. Rigby et al.

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The authors present estimates of global and hemispheric SF6 emissions for the period of 1973-2008, based on globally distributed tropospheric observations of this trace gas from the AGAGE and NOAA networks as well as from archived air samples. Including also non-background measurements for the years 2004 to 2008 they also estimate emissions for eight large continental regions in order to identify the source region responsible for the large increase of global emissions in this latter time period. The authors compare their global emissions with bottom-up emission estimates from the EDGAR V4.0 data base as well as official emission data reported by 39 industrialized countries to the UNFCCC. They come to the conclusion that (1) it is likely that these reported numbers to UNFCCC are too low, and (2) that Asian countries (e.g. China and India) are most probably responsible for the large increase of emissions in the last 5 years.

Only a few months ago there was a paper published in ACP by Levin et al. which, based on a completely independent data set of SF6 background observations starting in 1978, came essentially to the same conclusions as Rigby et al. in the present work. At this point the present work is just a confirmation by independent data.

But what is really new in the present work is that Rigby et al. use a more powerful modeling framework than Levin et al. that potentially allows them to resolve SF6 emissions from defined regions (large continental areas). Also, at least for the second part of their investigated time period, they base their results on a much larger data set, including stations over the continents, as well as quasi-continuous records at AGAGE and a few other sites. Still, Rigby et al. are not able to pin down the emissions in the large continental regions and provide the irrefutable proof that the increase of global SF6 emissions after about 2000 is mainly happening in the newly developing countries in Asia and that emissions reported to UNFCCC by industrialized countries are significantly too low. One might argue that the authors used a modeling framework that was in-adequate for answering the question they posed. In fact in their discussion they state that even a higher resolution transport model would be required for regional emission validation, in addition to more high-frequency monitoring sites over the continents. Again, this had already been concluded by Levin et al.

In September 2010 the process of independent state greenhouse gas inventories for the 2008 reporting year, the first of the Kyoto Protocol's five-year commitment period starts. Atmospheric observations could play an important role in this process. The reader would thus have expected to see now, after the paper by Levin et al. which was dealing mainly with global emissions, significant progress in the top-down method. This means, a more dedicated modeling framework should have been applied that would be able to "digest" ALL the continuous observations now available at the AGAGE sites, and not only deploying a subset measured during supposedly background conditions, as done in the present study.

One of my additional concerns relates to the authors' estimate of the uncertainty of

EDGAR emissions (global totals as well as the spatial distribution) which was assumed to be only 10%: What is the basis of this assumption and to what extent are the inverse model estimates by Rigby et al. possibly biased by this (small) a-priory uncertainty of EDGAR emissions? I guess that this concerns not only the regional inversion but also the hemispheric and possibly even the global results (Figure 2b). I understand that it is not easy to determine uncertainties of bottom-up emissions inventories, and I suggest that the authors contact EDGAR people to confirm the 10% uncertainty as well as its probable range (for the global total but also for the spatial distribution).

My third concern is dealing with the accuracy of model transport: The authors state that they can well reproduce background observations when using EDGAR emissions (page 13529, line 21). But if these emissions are biased then model transport is biased too. It is unclear to me how transport model uncertainties have been taken into account in the hemispheric but also in the regionally inverted emissions.

In summary, although the present study presents some progress compared to the earlier Levin et al. paper, it is disappointing that Rigby et al. were not able to really proof the earlier suggestions made by Levin et al. which eventually warrants a follow-up paper. What is needed instead would be the application of a more appropriate modeling framework which could take real advantage of the wealth of the new continuous SF6 measurements.

All in all, if a revised version is envisaged, it will be mandatory to substantiate the results by addressing the concerns mentioned above. To do so, following specific points should be considered as well:

Abstract:

Line 8: What makes this data set so unique? Please substantiate.

Last two sentences: These main findings (i.e. still assumptions) are not new but were first expressed by Levin et al., 2010. Adequate acknowledgement to these suggestions

made in this earlier work thus needs to be given, either here in the Abstract or at least in the Introduction.

Introduction:

Page 13523, line 18: Reference should be given here to the - to my knowledge first - application of SF6 in modeling studies which is the TransCom2 paper by Denning et al. Tellus B, 1999.

AGAGE measurements:

Page 13525: The measurement repeatability of the different data sets should be given at some place. The second half of the page presenting the comparison results is confusing. I suggest listing these results in a Table.

Page 13526, line3: What were the explicit criteria to reject/accept data from the NH air archive (why should polluted air which is not really representative for a large region be archived)?

NOAA measurements:

Page 13527, line 25: I cannot find “materials of the surface network” be mentioned in the manuscript.

Intercomparison:

Page 13529, line 1: Please give a number of the agreement (what means “well” in this context?).

Global and hemispheric emissions:

Page 13535, line 28: It is possible to receive information from the EDGAR scientists on how many “top-down” results from atmospheric observations were included in their estimates (and what the uncertainties of the distribution actually are, see general comments).

Continental emission estimates:

Page 13537, line 8ff: What is the temporal resolution of the meteorological fields driving model transport? Lines 12-13: This sentence is unclear: If I want to “extract” pollution events I would need higher than weekly resolution.

Page 13538, lines 8-11: Where are these uncertainty reductions shown? Lines 16-19: This is unclear: Why can’t we be sure to see regional pollution events (or the regional influence) at a (flask) sampling site in the middle of a continent?

Figure 2 (global inferred emissions):

What must have happened in the respective industrial applications to make GLOBAL SF6 emissions suddenly increasing by 20% (i.e. from 2001 to 2003) and then suddenly decreasing by 10% in the next year (2004)? This source variation is not visible in the estimates made by Levin et al., 2010. Are the authors sure that this strange behavior is not based on a measurement/calibration artifact? I think the global source estimates by Levin et al. show higher emissions in 2008 than in 2007. Please check.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 13519, 2010.