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Atmospheric Chemistry and Physics Discussions



# *Interactive comment on* "Atmospheric observation-based global $SF_6$ emissions – comparison of top-down and bottom-up estimates" *by* I. Levin et al.

**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. 1: The title perhaps should be changed, as in its present form, consisting of two statements, it is not optimal. "Observations" perhaps should be reserved for remote sensing, whereas here we deal with GC measurements. The first statement is too cryptic, and on its own illegible. The second statement suggests the reader that comparisons are made between two different types of estimates in general. Maybe: "Using



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measurements of atmospheric SF6 for calculating its emissions, and a comparison with bookkeeping attempts to estimate its emissions"

We have changed the title to

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

Rev. 1: The lifetime of SF6 is given as being between 800 and 3200 years. In the abstract it is ïĆż 3000 years. Is 3000 years an estimate of the most probable value?

We will put the range (800-3200 years) of estimated lifetimes into the Abstract and cite the two different estimates in the text, as well as the IPCC-adopted number, which is 3200 years.

Rev. 1: Page 26655. Line 23 "practically", perhaps one can simply give the % that accumulates. "Practically" is a bit sloppy for a scientific paper, unless there is no other way to quantify a process.

Changed to "more than 96-99%" (as a lifetime of 800-3200 years would correspond to a loss of 4-1%).

Rev. 1: Line 12-13. List the countries given here in order of emission size.

We only want to make clear here which countries belong to Annex I, starting actually with the Annex II (industrialized) countries. As we do not look at individual country emissions at all, we do not see the point to sort them in order of emission size.

Rev. 1: Page 26658. This statement must either be changed or substantiated. The more people work on emission scenarios using electrical light, computers and the like, the higher SF6 emissions will be, be it by a very small amount. Moreover, the statement compares "changes" with "inventory" (inventory changes?). Or do the authors suggest that the emission scenarios are partly based on the atmospheric measurements? I suggest to email Sir Dr. Edgar and ask him. Either give proof, or better, drop the



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statement.

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. 1: Page 26662. Line 5. The model used is fairly adequate for this long lived gas, although the gas has concentrated sources (electrical light intensity around the globe may be a measure?) and is increasing at a substantial relative rate. A question is to what degree one can get "really independent validation". Perhaps the model, because of its structure, a priori cannot get transport correctly simulated. SF6 in fact has been used to validate transport in more complex models as the authors state themselves.

We suggest an "independent" tracer, i.e. 85Krypton which, contrary to SF6 or light intensity, has only well-defined point sources. However, this tracer is only applicable in a model with higher spatial resolution, not in GRACE. This is discussed further down in the same section of the original manuscript.

Rev. 1: Line 16. Here the authors mention high resolution models, but state that these must be "very well validated". Various ECMWF models are astoundingly good in forecasting many physical atmospheric properties. Such would not be possible if transport were not highly correct. GCM Models using "nudged" transport may be suitable.

We agree that the global models get better and better in correctly modeling atmospheric transport. Still we think that they need to be tested with tracers that have very different source distributions and characteristics, such as e.g. 85Krypton (large single point sources) and SF6 (more area-like source distribution)

Rev. 1: Page 26662. Line 21-21. "Suggest . . .may be too low" perhaps better "Indicates ..may be too low" or alternatively "Suggest. . ...are too low".

Changed the phrasing into "suggests . . . are too low".



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Rev. 1: Line 25-26. Why is it implausible that Non-Annex 1 SF6 emissions per electricity production (sic) are 2 up to 4 times higher than for Annex 1 countries? Note that pollution in some Non-Annex 1 countries for a range of production processes is much higher, so why not for SF6 spillage?

SF6 in this sense is not a “pollutant” but rather a by-product of modern energy distribution. Therefore it cannot really be compared to “ordinary” pollutants which are co-emitted with e.g. CO2 during burning of coal or during traffic.

Rev. 1: Page 26663. Line 10. “top-down verification” Perhaps once should simply write verification by atmospheric measurement. The “top down” and “bottom up” revolutionary jargon has confused e.g. oceanographers because it sounds upside down to them (they want bottom-down approaches), and also in New Zealand it has different meanings I believe. Further.. “A dense network of high precision atmospheric observations”. It seems to me that a project like this CARIBIC that uses passenger aircraft may be a way to get regularly, many atmospheric observations from across much of the globe”. It also helps to solve issues of transport (in models). The AGAGE network installs automated mass spectrometers which give information on many trace gas species, but is in practice rather expensive of course. This development also shows how the flask networks have evolved from only CO2 to presently many species, and at times even on line.

We changed “Top-down” to “Verification by atmospheric measurement”.

It is correct that CARIBIC provides a lot of interesting measurements covering large parts of the globe. However, these data are mainly from the upper troposphere, far away from the sources. It will be difficult to use them for inverting emissions on the country scale.

Rev. 1: Page 26663. Line 16. “gases with well known sinks such as SF6, and other fluorinated and chlorinate compounds”. This statement in not formulated carefully enough. Perhaps replace “and” by “or”. Or drop the first “and”. Moreover the reviewer



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notes that the sink of SF6 is not well known because, as the authors state, its lifetime is 800 to 3200 years. Better write “defined”, or gases with well known or negligible sinks. Question from the reviewer: what are these gases, one or two examples?

We changed the phrasing accordingly. The group of gases was mentioned already in the last sentence. We think that it does not add to clarity if individual species were named here.

Rev. 1: Final comments. When considering the effort in obtaining the measurement results presented by the authors, the application of 3D models is warranted. It does not seem to be difficult to include a virtually conserved tracer in model runs. The role of the monsoon in inter-hemispheric transport and the effect from using surface measurements (in particular Alert, Arctic Haze) will be better parameterized then. Also, this would show whether the model used is adequate. If space allows a figure could perhaps show the fluxes between the boxes. It would give the reader some insight in transport. The authors give potential methods to better calibrate their model (Page 26662, ...”A possible tracer for transport validation may be 85Kr”). But is it really the heart of the matter? It is then mentioned that 85Kr after all is not suitable. It may well be that when this model is “calibrated” for one tracer (with a given lifetime, rate of increase, and source distribution) it is not for another tracer.

We feel that an additional figure showing the (air) transport between GRACE boxes will not be very helpful, but, as suggested by Reviewer 2, we have now included the range of hemispheric residence times used in the sensitivity study (Figure 3b). We already mention that only for the very coarse grid GRACE model 85Krypton is not suitable as transport tracer because e.g. large emissions are located close to the boundaries of the GRACE boxes and the modeled concentration distribution in GRACE changes very much depending on where the emissions are located. This problem is not valid for a higher resolution model e.g. as that used by Jacob et al., 1987 or the one by Levin and Hesshaimer, 1996.



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