

Interactive comment on „Atmospheric observation-based global SF₆ emissions – comparison of top-down and bottom-up estimates” by Levin et al.

Anonymous Referee #3

Sulfur hexafluoride (SF₆) is a trace greenhouse gas of essentially anthropogenic origin, having high global warming potential and long residence time in the atmosphere. The authors used a 30-year record of SF₆ concentration in the atmosphere to derive the history of global, surface emission rates of this gas. They used the so-called top-down approach where the global surface emission rates of SF₆ are equal to the first temporal derivative of the global atmospheric inventory of this gas derived from atmospheric observations (measurements of atmospheric mixing ratios at ground-based stations supplemented by vertical profiles). The central assumption here is that no significant atmospheric sinks of this gas exist. Although this approach has already been used in the past by several authors to derive global emission rates of SF₆, the present attempt is by far the most comprehensive one. The quality of the presented observational data set is remarkable. The authors went to great pains to make sure that the presented data are consistent and their uncertainties are well-quantified.

The central problem tackled in the paper is presented in Fig.2 where SF₆ emission rates inferred from the global inventory changes of this gas are compared with the bottom-up estimates based on the EDGAR database and the UNFCCC inventory. The inferred global emission rates of SF₆ compare rather well with the bottom-up estimates based on EDGAR database for the entire period involved in the comparison (1978-2005), although small differences can be observed for some years (e.g. the period between 1986 and 1989 for which EDGAR suggests continuous growth of emissions while the inferred emission rates stagnate). This general agreement between the top-down and bottom-up estimates provides a strong argument in favor of the methodology adopted within EDGAR. If this is the case, then one may argue that the share of the global SF₆ emission rates between the Annex I and non-Annex I countries, as derived from EDGAR database and showed in Fig.2, may also be trusted. It indicates that in 2005 the Annex I countries were responsible for ca. 60% of the global emissions of SF₆. Surprisingly, the SF₆ emission rates reported under the UNFCCC by the Annex I countries appear to be lower by a factor of ~2 when compared to those derived from EDGAR database!. If one believes in UNFCCC reporting, than in 2005 the Annex I countries emitted only ca. 30% of the global emissions while for the rest the non-Annex I countries would be responsible Which numbers are then correct?

This “politically loaded” question was not fully resolved within the paper. The authors tried to get some insight into the spatial distribution of SF₆ sources through a 2D multibox transport model (GRACE model). However, as they admit in the text, inherent uncertainties associated the model used (coarse spatial resolution, treatment and calibration of atmospheric transport in the model) prevent them from unequivocally resolving the issue. One may ask if there is any way out of this dilemma. Is there any chance for an independent verification of SF₆ emission rates reported by Annex I countries under UNFCCC ?. A state-of-the-art, high-resolution general circulation model with realistic transport and atmospheric mixing schemes seem to be the right way to proceed. However, as correctly pointed out by the authors, a more dense observational network would be required in this case.

To conclude, the paper is of high significance and definitely worth of publishing in ACP. It demonstrates convincingly that the global emission rates of SF₆ can be independently validated via atmospheric observations. It is seldom to see the power of the top-down method

demonstrated so convincingly, applied to the problem of global significance and, at the same time, of high political relevance. The work also demonstrates the potential of the top-down approach in the regional context, provided that adequate observation network is in place. Below are listed some specific comments which might assist the authors in preparing the final version of the paper.

Page 26655. Line 15. It would be good to give the number for global warming potential of SF₆.

Page 26657. Line 21-24. It would be desirable to show (in the Supplementary Material) those two meridional profiles of SF₆ collected in 1990 and 1993 over the Atlantic Ocean. The authors announce them on page 26656 as belonging to the database discussed in the paper, but then did not show them.

Page 26658. Lines 6-10. It would be worth to mention here that there is a natural production of SF₆ in the Earth's crust. This was proved by measurements of SF₆ concentrations in groundwater originating from specific lithologies (mainly silicic rocks). Thus, one may expect SF₆ emissions into the atmosphere via volcanic exhalations in some areas and/or via groundwater usage. Those emissions are most probably quite low but they should be mentioned here just for completeness. Line 13. In fact the inferred global emission do not increase steadily. As seen in Fig.2 and in Table 2, they decrease between 1987 and 1989. Line 21-22. I suggest to remove the sentence "...but it is not clear to us how independent the DGAR (2009) emissions inventory is from observed atmospheric mixing ratio changes" or to clarify this issue.

Page 26660. Line 25-29. This "almost perfect agreement" with observations can be misleading as the EDGAR adjusted annual totals were derived with the help of atmospheric data...

Page 26663. Line 5. The reference (GDP, 2008) is not listed in the list of references.