

Declining, seasonal-varying emissions of sulfur hexafluoride from the United States point to a new mitigation opportunity

Lei Hu et. al.

Overview

This paper describes how emissions of SF₆ have been estimated over the US using atmospheric observations and atmospheric transport modelling. These estimates have been compared to the reported inventory both at country and state level. The decline in US emissions of SF₆ is seen in both the inverse modelling and the inventory, although the magnitudes differ, especially in the early years of the comparison. Through strong interaction with the US EPA several mitigation opportunities have been identified.

The paper is well-written and the results are presented very clearly. I have some questions and comments that are detailed below.

Comments

P3, L128: A supplement table listing the stations used and their inlet height / aircraft heights is necessary I think.

P4, L145: So in option 2 were all particles used regardless of whether they exited the US or not? It implies 500 values were used but also says it just uses particles that left the domain.

P4, L173: Particles released from the inlet height of the observation? Up to what height is the surface sensitivity modelled (surface to model BL or a fixed height)?

P4: 500 particles seems remarkably small to me, very different to the many 1000s other modelling systems use. It would be extremely helpful to understand the impacts of this decision on the US estimates. A sensitivity analysis of some sort would be very valuable, or point to a reference where this has been done and add a sentence as to the results.

P4, L180: The comparisons in the paper use EDGAR v6. Please explain the logic behind using v4.2 as the prior.

P5, L186: Are all off-diagonal elements of the transport uncertainty matrix zero? i.e. are all the observation-model uncertainties assumed to be time and space independent? For flasks every two days a long way apart physically this is okay, but not for the observations in the same vertical profile in aircraft data? How was this handled?

P5, L189: There has been no discussion on inversion grid resolutions. Uncertainty calculations from different grids - how are they combined to estimate region and US emission sigma?

P5, L194: Are cross, non-diagonal elements of the uncertainty matrix considered?

P5, L194: This formula implies that all of the inversions are independent. They are to an extent, e.g. different meteorology, but equally there are lots of dependence, e.g. the baseline options have strong correlations. The delta S term is interesting, I can see why it is added but wonder if a better idea would be to combine them all in one step. E.g. create 1000 estimates from each inversion and then estimate a mean and a std directly? I think the method described here will create uncertainties that are too small, although hard to be very sure given the delta S term.

P6, L231: How was this trend and uncertainty calculated? Do these uncertainties account for the reduction in uncertainty over time?

P18, Fig 3: Are the uncertainties using the different transport models calculated using 6 inversion each? I assume so but best to specify

P18, Fig3: Is this the inversion grid used to estimate the emissions in all cases? It would be good to explicitly show the inversion grid and to describe how it is defined.

Supp. P2, Fig S2: Why not just use 2008-2010 for both? It looks unusual that 2009-2010 are not plotted. Is it because 2009 looks so very different? This should be shown.

Minor Text Comments

The United States is described as ‘U.S.’ and ‘US’ - Please be consistent throughout the paper.

P2, L72: “after emissions” – remove as unnecessary or change to “after emission”

P2, L73: Add “by the U.S.”

P2, L74: “accurate estimates of the magnitude and distribution of SF6 emissions”

P2, L82: I do not like the use of ‘developed (ing) countries’, I prefer ‘Annex 1’ and ‘non-Annex 1’ countries.

P2, L83: “(including China, one of the largest SF6 emitting countries)”

P3, L104: The same list of papers are repeated several times, maybe this should be reduced to just the key paper after they are first mentioned.

P3, L116: Manning et al 2021, ACP is a more appropriate reference as it is peer-reviewed.

P4, L150: Please re-word the sentence beginning “Because there was ...back for 10 days”

P4, L155: Consider “.. to populated areas, defined as summed ...”. I was very unclear where the “less than 0.1 ppt (pmol ..)” came in?

P4, L 174: “which has fewer vertical levels than NAMs.” – please define ‘fewer’

P5, L189: I don’t think the word ‘ensembles’ is useful here.

P5, L218: Maybe this EPA report does not need to be cited so much, it has been cited 3 times in the last two paragraphs? Or can the citation be condensed in length?

P6, L267: Reference (16)? What is this?

P7, L278: Consider replacing ‘through’ with ‘up until’

P7, L306: ‘lbs’ – replace with ‘mass’

P9, L410: Add the word ‘reporting’ e.g. ‘from other reporting countries’

P11, L499: Extra line break

P12, L523: “CO₂” – incorrect formatting

P13, L552 and L592: “SF₆” – incorrect formatting

P14, L594: No year associated with Rodgers C.D.

P16, L680: Change “difference” to “different”