

Black: referee's comments red: authors' answers

First, we want to thank the referee for the detailed analysis of our paper.

For the details, please look into the paper with keeping track of changes.

The manuscript submitted by Zhou et al. presents measurements and trends of SF₆ in the atmosphere using remote sensing techniques with ground-based FTIR spectrometers on Reunion Island. These are supplemented with analysis of measurements from in-situ instruments and space-borne sensors. This work is very significant and I recommend its publication after a few items below have been addressed/clarified.

General Comments

Column averaged dry mole fractions of SF₆: To readers not familiar with this method, elaboration of this procedure is necessary. How are the dry air total columns measured/obtained? Since the XSF₆ trend would depend on this, it would be important to establish and/or show the robustness of the dry air total column measurements.

We added the definition of XSF₆ in the text. The uncertainty of the total column of dry air is discussed in the text, which is less than 0.1%.

Pseudo-lines: Please explain/justify the use of pseudo-lines, e.g. how are they derived? Do ACE and MIPAS use pseudo-lines as well?

Added in the text.

Pseudo-lines were produced by G.C. Toon (NASA-JPL) by fitting all the laboratory spectra simultaneously, which include mean intensities and effective lower state energies on a 0.005 cm⁻¹ frequency grid. These artificial lines at arbitrary positions do not represent transitions of molecules. Instead, their line-widths and intensities are fitted to the laboratory spectra such that the pseudo-line list allows to simulate the measured spectra. We use the Pseudo-lines for the FTIR retrievals, because it allows us to get a better fitting.

ACE uses HITRAN 2004 (http://www.ace.uwaterloo.ca/ACE-FTS_v2.2/ACE-SOC-0011-1D-ACE-FTS_ascii_fileformat_for_v2.2.pdf)

MIPAS uses HITRAN2000 (Haenel et al., 2015).

Smoothing error: From my understanding, the smoothing error can be evaluated if there is an independent measurement of the true profile, an estimate of its variability and covariance matrix (e.g. Barrett et al., 2002 did this for Ozone,

<http://onlinelibrary.wiley.com/doi/10.1029/2001JD001298/abstract>). Though I understand that following this would be difficult for SF₆ at the site, would the authors please comment on this?

As there is no real measurement available (e.g. balloon sondes). We apply the following method to get the systematic and random uncertainties of SF₆.

Systematic uncertainty:

The SF₆ is constantly increasing during last two decades, with the annual growth of ~3.0%. As the SF₆ a priori profile is fixed and scaled to the in-situ measurement in the year of 2009, we assumed that there is no systematic error for 2009, but 3% for 2010 and 2008; 6% for 2011 and 2007; 9% for 2012; 12% for 2013; 15% for 2014 and 2004; 18% for 2016. After taking the number of measurements as the weighing function, the mean value is about 5%. Therefore, we apply 5% as the systematic uncertainty of SF₆.

Random uncertainty:

We calculated the covariance matrix from the MIPAS measurements between 2002 and 2012 at Reunion Island. The standard deviation is about 30%, therefore we apply this value as the random uncertainty of SF₆.

On the use of the retrieval window and improved fits: What criteria is used to determine a “better fit”? For example, in Fig. 2. (right panel), the residuals at 948.0 cm^{-1} seem to indicate that something is not fitted well.

The root mean square (RMS) of the fitting residual is applied to be the key parameter to check whether the fitting is better or not. The RMS is the root mean square of the difference spectrum (obs-fit).

In Fig.2 (right panel), the RMS is 0.0015, reprinting the noise level, while the intensity of the SF6 absorption line is 0.03. Such a fitting ($0.0015/0.03 = 5\%$) is close to the retrieval random uncertainty (6.7%; see Table 2), therefore, we think the fitting is good enough to retrieve SF6.

Specific comments

Instrument and ILS: A mention of the spectral resolution used in the actual retrieval and the goodness of the ILS would be helpful to readers. For example, in Section 2.1., it was mentioned that the max OPD of the instrument is 250 cm, is this the same OPD used for SF6 measurements at both sites?

Added in the text.

The spectra of $700\text{-}1400 \text{ cm}^{-1}$ at St Denis and Maïdo are recorded with the same settings. Two maximum optical path difference (MOPD) of 82 and 125 cm are operated to gather the interferogram of the direct solar radiation, and then the interferogram is transformed to a spectrum with the spectral resolution of 0.010975 and 0.0072 cm^{-1} through a fast Fourier transform (FFT) algorithm.

Page 6 line 10 “balloon measurements” appears here for the first time. I think this should be defined earlier in the manuscript. I assume these are sondes, right?

Yes, they are balloon sondes.

Table 2, Please elaborate on how the systematic and random errors under “SF6 Spectroscopy” were calculated.

Added in the text.

Technical Comments

Page 2, line 2: I suggest to change “SF6 emissions reduced” to “SF6 emissions decreased” Page 2, line 17: “aboard a shuttle”: please specify which one, if possible.

Page 4, line 19: “In order to extract as much as possible information” -> “In order to extract as much information as possible”

Page 4, line 28: Consider rephrasing this sentence: “That means when we use the FTIR retrievals, only the total column should be trusted instead of the profile.”

All corrected.

References:

Haenel, F. J., Stiller, G. P., von Clarmann, T., Funke, B., Eckert, E., Glatthor, N., Grabowski, U., Kellmann, S., Kiefer, M., Linden, A., and Reddmann, T.: Reassessment of MIPAS age of air trends and variability, Atmos. Chem. Phys., 15, 13 161–13 176, doi:10.5194/acp-15- 13161-2015, 2015.