# Determination of the atmospheric lifetime and global warming potential of sulphur hexafluoride using a three dimensional model

Kovacs et al.

We thank the reviewers for their comments. These comments are reproduced below in *italics*, followed by our responses in **bold red**.

## **Reviewer 1**

The following comments rely mainly on the presentation of the results of a work of good quality. The redaction has to be revised to bring more numerical precision with the aim of the justification of the conclusions.

## **General Comments**

All along this paper, there is a need of more precision: References, numerical values, in particular when conclusions are given. Very often, only overall appreciation is given, without documented justification.

The authors make the hypothesis that the reader has "under hands" their previous papers including necessary information. In any case, it should be necessary to provide some very short reminders for not totally familiar reader with the subject and the publications of the authors.

*The figures are of poor qualities; it is especially difficult to read included numerical values.* 

## We have replotted the figures.

## **I Introduction**

#### **General comments:**

This introduction is too long and has to be restructured. Organization of the paper with key study points should be clearly and shortly given.

### See responses to specific comments below.

The sentence of lines 106-107: "The definitions of these radiative terms are discussed in detail in our recent publication Totterdill et al. (2016)" should be associated with an Appendix where definitions of <u>ALL</u> the radiative terms, as used all along the paper, should be recalled.

## OK. We have added Appendix A which defines the relevant radiative and climate metrics used.

Sub-paragraphs included in section: 2. (Methodology), and section 3. (Results) are misplaced or too much developed at this stage (Introduction) of the paper, i.e.: *Lines 60 to 75* should be associated with lines 311 to 334. *Lines 76 to 101* should be associated with lines 261 to 286. *Lines 151 to 161* should be associated with lines 179 to 185.

We thank the reviewer for this suggestion but we feel that the present arrangement of material is logical and appropriate. We note that Reviewer 2 found the paper 'clear and well written' as it is.

### Other comments

Line 46 Surface measurements show that SF6 increased by about 7%/year during the 1980s and 1990s Line 47 (Geller et al., 1997; Maiss and Brenninkmeijer, 1998).

No information available during the 1990's-2016's??

Yes, this statement was outdated. We have added a reference to Dlugokencky et al. (2016) and updated the %/year increase rate.

*Lines 64 and 65: Precise units for the quantities in (E1).* 

The equation is general. AoA will have the same units as t, and can be any measure of time. We do not need to give specific units.

Lines 151 and 152: Quote Kovacs et al. (2016) and give a short reminder on D region and its altitude, in particular.

OK. We have added the following sentence: "The D region is the lowest part of the ionosphere, extending from about 60 to 85 km. It is characterized by the appearance of cluster ions (e.g. proton hydrates  $H^+$ .( $H_2O$ ) $_n$ , where  $n \le 6$ ) and negative ions (e.g.  $O_2^-$ ,  $CO_3^-$  and  $NO_3^-$ ) rather than free electrons. These species predominate because the atmospheric pressure is high enough to facilitate the three-body attachment of ligand species like  $H_2O$  to positive ions, and electrons to neutral molecules."

### Section 2 Methodology

### 2.1 WACCM 3D Model

#### **General Comments**

Paragraphs of this section to be associated with those of the introduction are identified above (may be non-exhaustive list).

See response above. We have left the order of material unchanged.

Some complementary brief reminder on the "Lyman- $\alpha$  photolysis", as used in WACCM, should be given.

We already give information on the Lyman-a scheme in WACCM by giving the fluxes used and the value for the SF6 cross section.

#### Other comments

Lines 162 to 178: The discussion is not enough numerically documented. It relies mainly on short (non-numerical/appreciation) comments from poor quality numerical values implemented on the figures. More information should be given on the plotted values, with possibly short information tables. Please, be so kind as to revise.

## We have added in some references to numbers in the plots.

Lines 185 to 187: "WACCM was run for the period 1990-2007, and the first five years were treated as spin-up. For the analysis the monthly mean model outputs were saved and later globally averaged for the lifetime calculations"

Those lines would be better placed after line 120. Why this restricted chosen time period?

The first paragraph of Section 2.1 gives general information about the model. Specific information about the experiments performed (number of tracers, duration) is given in this paragraph (old line 179 onwards). Therefore, we feel that this sentence is in the correct place. WACCM is quite expensive to run and so we chose a time period which was sufficient to investigate the SF6 lifetime over a whole solar cycle. The period from mid 1990s onwards was optimum for comparison with observations (e.g. MIPAS).

## 2.2 Infrared absorption spectrum 1 and radiative forcing

Lines 190-192: "Previous quantitative infrared absorption spectra of SF6 have been compared in Hodnebrog et al. (2013) (their Table 12). There are differences of ~10% between existing integrated cross section estimates, and the measurements cover different spectral ranges."

Again, please, add complementary precision

The review paper Hodnebrog et al. (2013) does not discuss the precision of the IR spectra measurements, nor is it comprehensively covered in the underlying papers. Therefore, it is difficult to assess. Precision also varies greatly with wavelength and methodology and generally assumed to be better than systematic bias between measurements. It is the spread of previous integrated cross section measurements that we deem important to consider here, so feel adding to the text would only confuse the reader and we choose to leave as is.

## **Section 3 Results**

### 3.1 Global distributions of SF6 from WACCM simulations

## **General comment**

From lines 214 to 260 revision should occur to include more numerical precision in the given conclusions, which very often only stem from figure displays (Fig. 3 and Fig. 5, in particular), with sparse or missing included numerical values.

We have added in numerical values throughout this section.

#### Other comment

*Line 234: Figure 5 shows the zonal mean annual mean SF6 distribution: which unit?* 

The units of pptv are stated in the figure caption.

## 3.2 Atmospheric lifetime

## General comments (already made).

In any case, the definition of "Atmospheric lifetime", should be given earlier in the text, as already suggested.

Complementary definition to provide, as well: "partial lifetime" and "overall lifetime".

Atmospheric lifetime has been defined when first used in Section 1. In Section 3.2 we explain the terms partial and overall lifetimes when first used.

#### Other comment

Lines 300-302: "Finally, if we do not include the electron scaling factor to reduce the electron density below 80 km due to negative ion formation, then the SF6 lifetime decreases to 776 years (not shown), which is similar to the value obtained by Morris et al. (1995)". Please, document this sentence: "then the SF6 lifetime decreases to 776 years (not shown), which is similar to the value obtained by Morris et al. (1995)."

We have added the value obtained by Morris et al. to the text.

## **Section 3.4 Radiative Efficiency and Forcing**

The references for SF6 cross-sections, as provided in lines 370-372, "To determine the radiative efficiency and global warming potential of SF6, integrated cross-sections were taken from the GEISA: 2011 Spectroscopic Database (Varanasi, 2011), the HITRAN 2012 Molecular Spectroscopic Database (Rothman et al., 2012)" should be quoted differently. The following necessary modification in the text should be made, i.e.:

To determine the radiative efficiency and global warming potential of SF6, integrated cross-sections were taken from two public Molecular Spectroscopic Databases, i.e.:

- GEISA-2009/2011 (Jacquinet-Husson et al., 2011): data of Varanasi (2001) and Hurley (2003);
- HITRAN 2012 (Rothman et al., 2012): data of PNNL (Pacific Northwest National Lab) IR Database, Sharpe et al. (2004)

The references should be updated accordingly, i.e.:

• Jacquinet-Husson, N., Crepeau, L., Armante, R., Boutammine, C., Chédin, A., Scott, N.A., Crevoisier, C., Capelle, V., Boone, C., Poulet-Crovisier, N., Barbe, A., Campargue, A., Chris Benner, D., Benilan, Y., Bézard, B., Boudon, V., Brown, L.R., Coudert, L.H., Coustenis, A., Dana, V., Devi, V.M., Fally, S., Fayt, A., Flaud, J.-M, Goldman, A., Herman, M., Harris, G.J., Jacquemart, D., Jolly, A., Kleiner, I., Kleinböhl, A., Kwabia-Tchana, F., Lavrentieva, N., Lacome, N., Li-Hong, Xu, Lyulin,

O.M., Mandin, J.-Y, Maki, A., Mikhailenko, S., Miller, C.E., Mishina, T., Moazzen-Ahmadi, N., H.S.P. Müller, A. Nikitin, J. Orphal, V. Perevalov, Perrin, D.T. Petkie, A. Predoi-Cross, Rinsland, C.P., Remedios, J.J., Rotger, M., Smith, M.A.H., Sung, K., Tashkun, S., Tennyson, J., Toth, R.A., Vandaele, A.-C., J. Vander Auwera, J.: The 2009 edition of the GEISA spectroscopic database. J. Quant. Spectrosc. Radiat. Transfer 112, 2395–2445, 2011; http://cds-espri.ipsl.fr/etherTypo/?id=950

- Varanasi P., 2001: Private communication
- Hurley, M.D., 2003: Private communication.
- Sharpe, S.W., Johnson, T.J., Sams, R.L., Chu, P.M., Rhoderick, J.C.: Gas-Phase Databases for Quantitative Infrared Spectroscopy, Appl Spectrosc. 58(12), 1452-1461, 2004.

OK. The references and text have been updated. The personal communications will need to conform to the journal regulations.

Table 4 should be revised accordingly, as well

## OK.

Line 384: Varanasi (2001): please correct Ref. in the text; no other comparison data available?

## OK, changed to 2001.

*Lines 412-414: References should be corrected as in lines 370-372 (see above).* 

### OK, done.

Lines 423-424: Give reference and numerical values for: "somewhat higher than previously published values"

This text follows on from the previous sentences which compare the values in Table 6. We have added 'the' to make it clearer.

Line 434: Same comment as above for: "shows a significant dependence on the solar cycle"

OK. At this line we have inserted the magnitude of this dependency