Supplementary information for:

The impact of SF_6 sinks on age of air climatologies and trends

Sheena Loeffel[1], Roland Eichinger[2,1,5], Hella Garny[1,2], Thomas Reddmann[3], Frauke Fritsch[1,2], Stefan Versick[4], Gabriele Stiller[3], and Florian Haenel[3]

[1]Deutsches Zentrum für Luft- und Raumfahrt (DLR), Institut für Physik der Atmosphäre, Oberpfaffenhofen, Germany
[2]Ludwig Maximilians Universität, Institut für Meteorologie, Munich, Germany
[3]Karlsruhe Institute of Technology, Institute for Meteorology and Climate Research, Karlsruhe, Germany
[4]Karlsruhe Institute of Technology, Steinbuch Centre for Computing (SCC), Karlsruhe, Germany
[5]Charles University, Department of Atmospheric Physics, Faculty of Mathematics and Physics, Prague, Czech Republic

Correspondence: Sheena Loeffel (Sheena.Loeffel@dlr.de)

S1 Atmospheric Model

S1.1 Submodel SF6

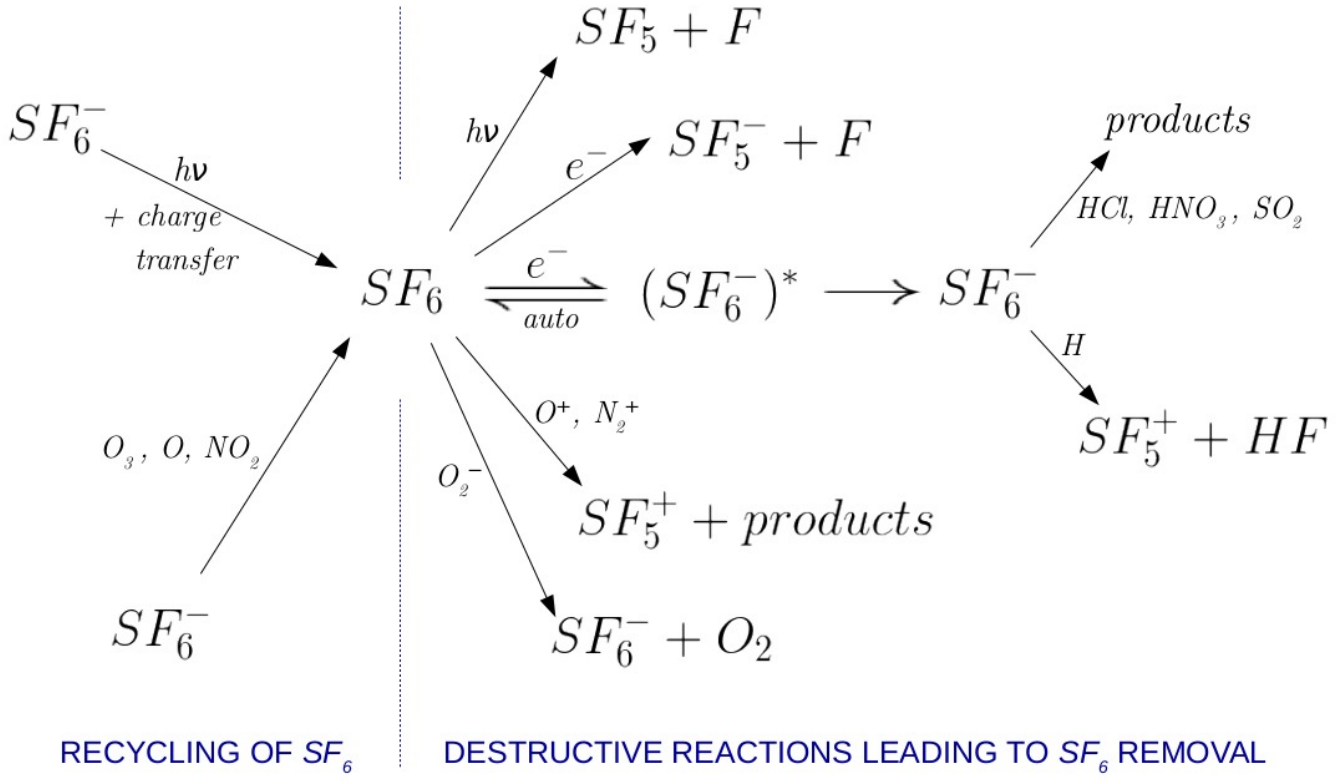

Figure S1. Reactions of SF_6 in the mesosphere, following the schemes put forward by Reddmann et al. (2001) and Totterdill et al. (2015). The left hand side depicts the reactions leading to the recycling of SF_6, whereas the right hand side illustrates the chemical degradation schemes of SF_6 in the mesosphere.

S2 Results

S2.1 SF_6 mixing ratios

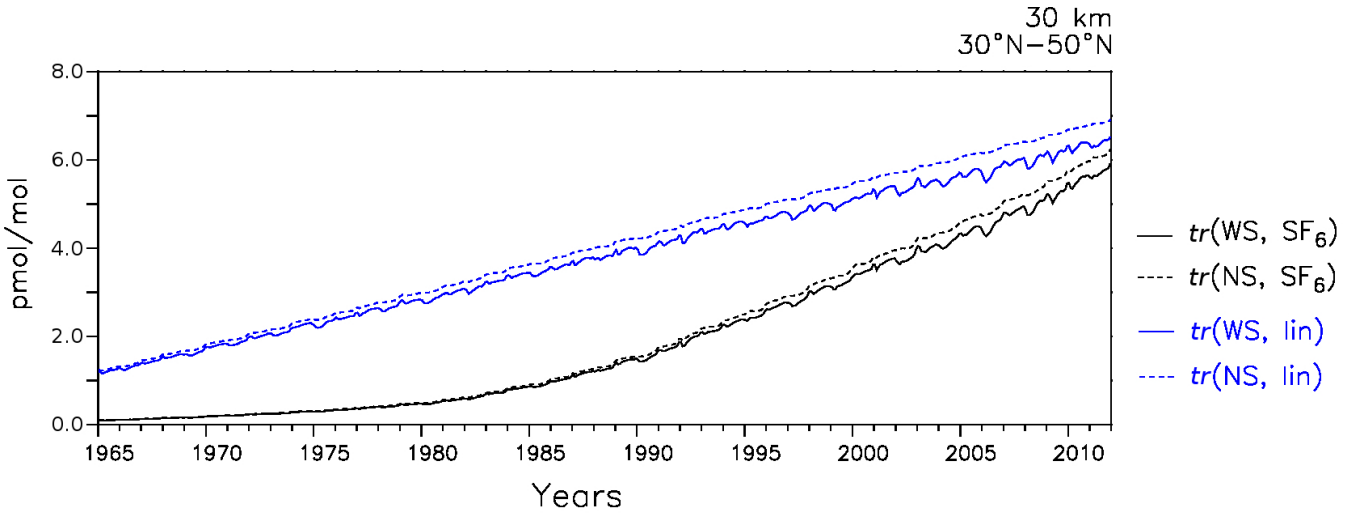

Figure S2. Timeseries of modelled SF_6 averaged over 30°N-50°N at 30 km. The tracers with mesospheric sinks are shown as solid lines, those without sinks as dashed lines. Tracers with non-linear and linear boundary conditions are depicted in black and blue, respectively.

S2.2 Age of air climatologies

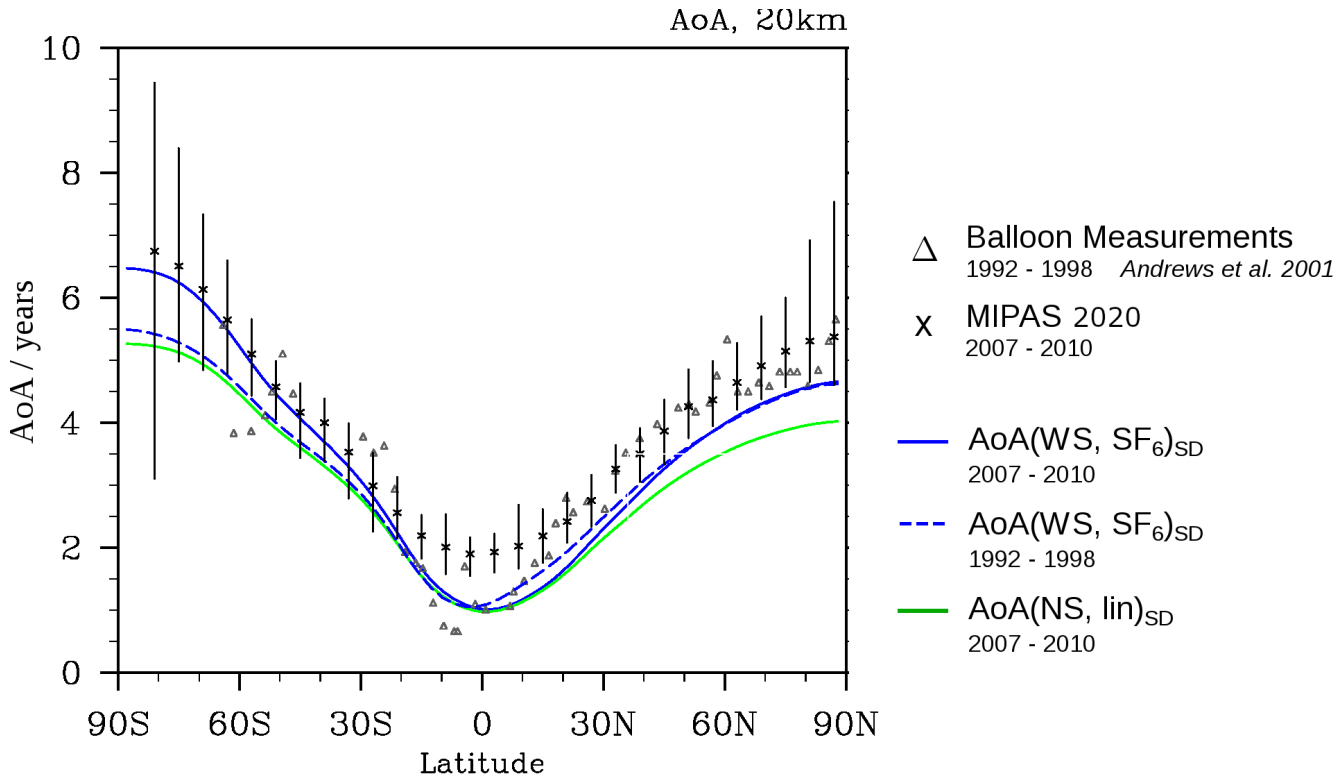

Figure S3. Latitudinal distribution of AoA at 20 km. Modelled AoA from EMAC simulations SD(WS, SF_6) for 2007 - 2010 and 1992 - 1998 are shown in blue as solid and dashed lines, respectively. The solid green line depicts modelled AoA from SD(NS, lin). Black crosses show MIPAS AoA, which consists of a complete data set for the time period 2007 - 2010. The spread of MIPAS AoA is depicted as error bars. Balloon flights spanning the period 1992 - 1998 are shown as triangles.

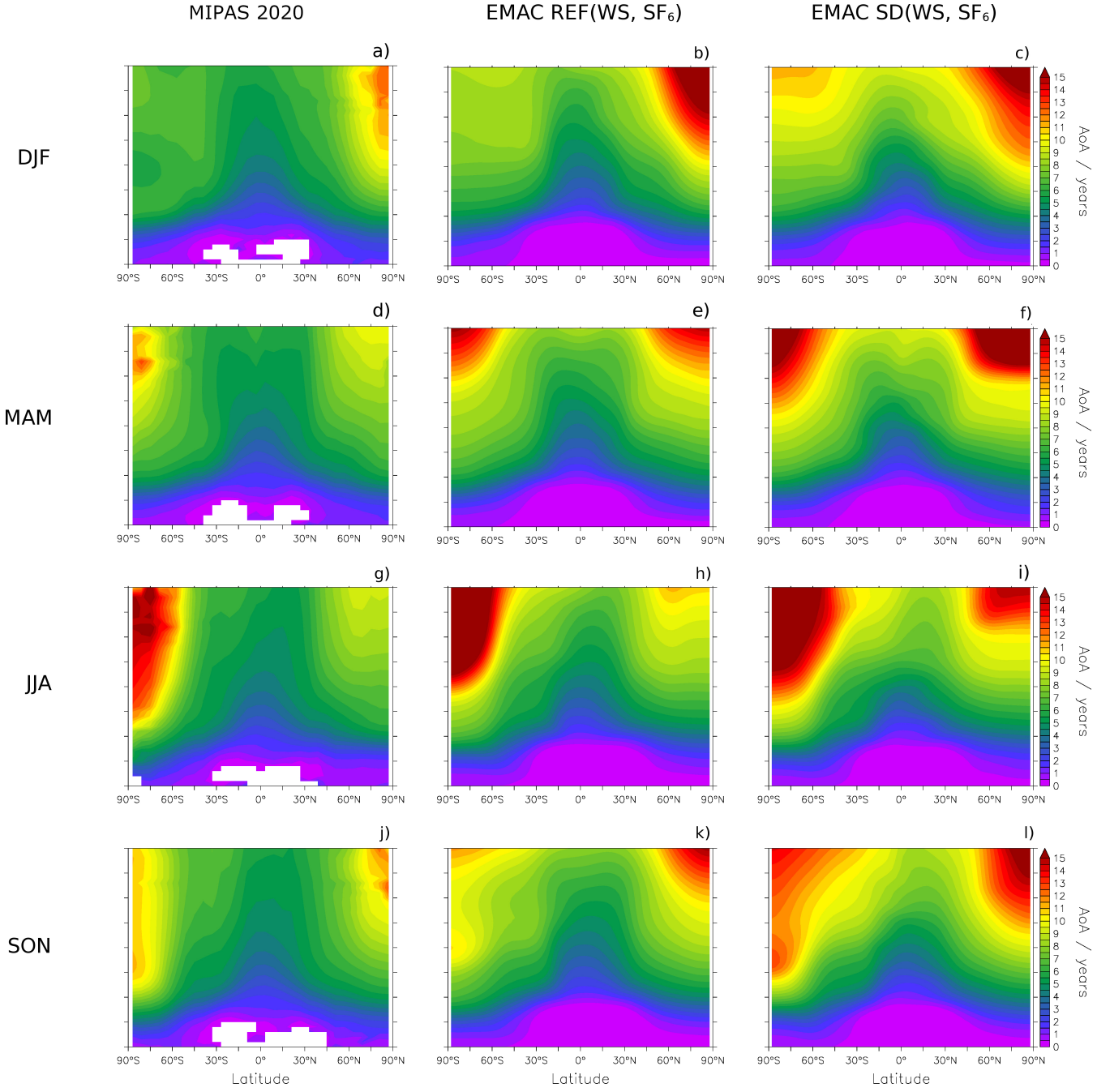

Figure S4. 2007 - 2010 seasonal means of AoA from MIPAS and from the simulations REF(WS, SF_6) and SD(WS, SF_6). a-c show seasonal means for December-January-February (DJF), d-f March-April-May (MAM), g-i June-July-August (JJA), and j-l September-October-November (SON).

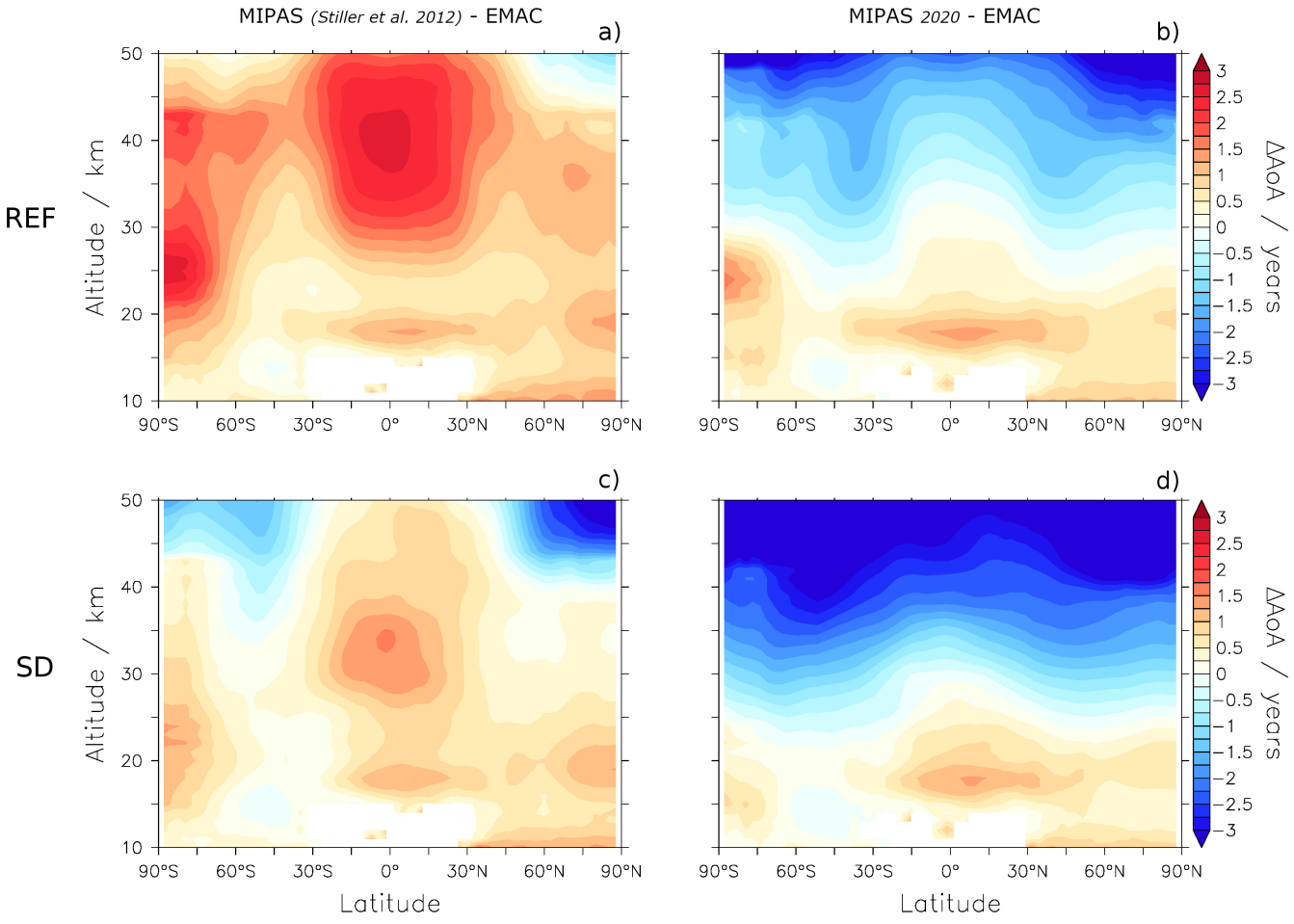

Figure S5. Modelled AoA subtracted from MIPAS (Stiller et al., 2008, 2012; Haenel et al., 2015) (a,c) and the MIPAS AoA using new SF_6 absorption spectra (Harrison, 2020; Stiller et al., 2020) (b,d). Values averaged over the region 30°N-50°N and 2007-2010.

S2.3 Apparent age of air trends

Table S1. EMAC AoA trends at 30 km averaged over 30°N-50°N. Values are provided for the two relevant simulations REF(WS, SF_6) and SD(WS, SF_6), and are shown for two scenarios: excluding the QBO (no QBO), and using the QBO from the respective EMAC simulation (QBO (EMAC)).

Simulation	2002-2011 Trend (years/decade)	
	No QBO	QBO (EMAC)
REF(WS, SF_6)	0.23 ± 0.12	0.22 ± 0.12
SD(WS, SF_6)	0.52 ± 0.13	0.50 ± 0.13

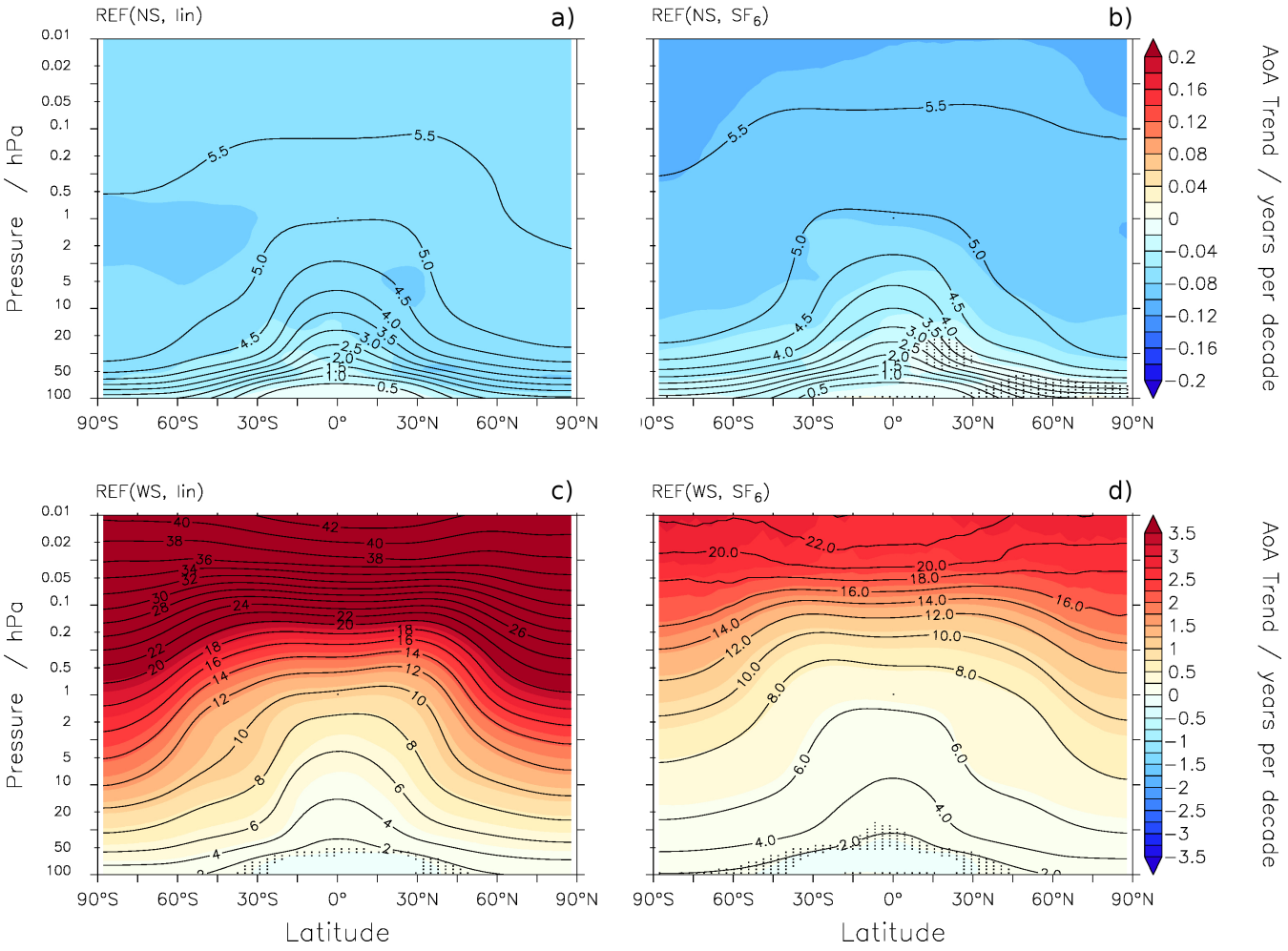

Figure S6. AoA trend from the reference simulation in colours calculated over 1965- 2011: AoA from the idealised tracer [AoA(NS, lin_{REF}] is shown in a), AoA from SF_6 emissions without sinks is shown in b). The bottom row shows AoA with mesospheric sinks, from the linear tracer (c) and non-linear tracer (d). Contours show the AoA climatology (years), averaged over 1990-2011. Dotted regions indicate where the trend is not significant, i.e. the statistical significance of the trend is below the 5% threshold. Note that the results for the AoA from the idealised tracer are statistically significant on the 95% level throughout the presented region

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, Atmospheric Chemistry and Physics, 15, 13 161–13 176, https://doi.org/10.5194/acp-15-13161-2015, https://www.atmos-chem-phys.net/15/13161/2015/, 2015.

Harrison, J. J.: New infrared absorption cross sections for the infrared limb sounding of sulfur hexafluoride (SF6), Journal of Quantitative Spectroscopy and Radiative Transfer, 254, 107 202, https://doi.org/https://doi.org/10.1016/j.jqsrt.2020.107202, http://www.sciencedirect.com/science/article/pii/S0022407320304453, 2020.

Reddmann, T., Ruhnke, R., and Kouker, W.: Three-dimensional model simulations of SF_6 with mesospheric chemistry, Journal of Geophysical Research: Atmospheres, 106, 14 525–14 537, https://doi.org/10.1029/2000JD900700, 2001.

Stiller, G., Harrison, J., Haenel, F., Glatthor, N., K. S., and von Clarmann, T.: Improved global distributions of SF6 and mean age of stratospheric air by use of new spectroscopic data, in: EGU General Assembly 2020, Online, 4–8 May 2020, EGU2020-2660, https://doi.org/10.5194/egusphere-egu2020-2660, 2020.

Stiller, G. P., von Clarmann, T., Höpfner, M., Glatthor, N., Grabowski, U., Kellmann, S., Kleinert, A., Linden, A., Milz, M., Reddmann, T., Steck, T., Fischer, H., Funke, B., López-Puertas, M., and Engel, A.: Global distribution of mean age of stratospheric air from MIPAS SF_6 measurements, Atmospheric Chemistry and Physics, 8, 677–695, https://doi.org/10.5194/acp-8-677-2008, https://acp.copernicus.org/articles/8/677/2008/, 2008.

Stiller, G. P., Clarmann, T. v., Haenel, F., Funke, B., Glatthor, N., Grabowski, U., Kellmann, S., Kiefer, M., Linden, A., Lossow, S., and Lopez-Puertas, M.: Observed temporal evolution of global mean age of stratospheric air for the 2002 to 2010 period, Atmospheric Chemistry and Physics, 12, 3311–3331, https://doi.org/10.5194/acp-12-3311-2012, 2012.

Totterdill, A., Kovács, T., Gómez Martín, J. C., Feng, W., and Plane, J. M. C.: Mesospheric Removal of Very Long-Lived Greenhouse Gases SF6 and CFC-115 by Metal Reactions, Lyman- Photolysis, and Electron Attachment, The Journal of Physical Chemistry A, 119, 2016–2025, https://doi.org/10.1021/jp5123344, https://doi.org/10.1021/jp5123344, pMID: 25647411, 2015.