

General comments:

As already pointed out by the initial reviewers, the authors present a novel framework for modelling transport and chemical transformation of PFAS, a tracer group of particular concern because of its potential hazardousness and its long persistence in the environment. In the revised manuscript, all major concerns have been answered appropriately. Description of hybrid coupling to CAM-Chem background concentrations has been improved significantly, but should also be reflected shortly in the abstract to avoid confusion for the first-time reader. The authors also assessed sources of uncertainty and variability in a new comprehensive appendix on sensitivity studies and their statistical analysis which adds valuable information for the interested reader.

In general, this is a pioneer study on atmospheric PFAS modelling and merits publication in GMD after some minor revision, even if the current limitations of the setup and the lack of observations leads to discrepancies in model evaluation that leave the reader feeling somewhat unsatisfied.

Specific comments:

Introduction:

Page 2, line 23: Reference “Schymanski, 2023” is missing in the reference section.

Page 2, line 34; Reference “Environment and Canada, 2022” must be changed. Issuing authority was Environment and Climate Change Canada. Please also update the entry in the reference list. You could think of adding the Web link: <https://www.canada.ca/en/environment-climate-change/services/management-toxic-substances/list-canadian-environmental-protection-act/long-chain-perfluorocarboxylic-acids.html>

Page 2, line 54: ... a wider PFAS speciation (**including PFCAs and FTOH precursors**) is included in Table A1.

Page 3, line 66: Can you give any reference for your previous modelling efforts?

Model Description:

Page 4, lines 111-113: Please refer to the Tables B1 and B2 here.

Page 4/5, lines 119-125: Again, please refer to Tables B1 and B2.

Page 5, line 126: Please mention that these are monthly mean values (except OH).

Page 5, line 132: ”... (further described **in** Fig. 1)”.

Page 6, lines 184-185: “Although” This is not a full sentence.

Model Setup:

Page 9, line 240: What is meant by “un-forced by further observations”? Is there data assimilation involved?

Page 10, line 259: “ ... boundary conditions are provided **for** each restart ...”

Page 10, line 275: Reference “Young, 2012” is missing in the reference section.

Page 10, line 278: I guess you mean “Thackray and Selin (2017)”.

Page 11, line 301: Change Reference “Will, 2005” to “Will and Kälin, 2005”.

Page 11, line 318: Please refer to “UNEP, 2024” instead of “Report, 2024”.

Page 12, line 322: Please stick to Copernicus style for units: $\text{pg}/\text{m}^3 \rightarrow \text{pg m}^{-3}$. Please change accordingly throughout the manuscript including appendices.

Page 12, line 331: Categorization into urban and background stations is not shown in Table A2. Please omit or add respective information. I would strongly propose not to use any urban stations for global model comparison because of their limited spatial representativity. Have you done so?

Model Evaluation:

Page 14, lines 392-393: “On the other hand AFFF emissions are estimated by airport size handling the 60 biggest air ports in the world ~~are handled~~ individually.”

Page 21, lines 469-470: Besides the larger variation in emission sources, the discrepancies could also be attributed to stronger variation of weather patterns in the mid latitudes, which are not fully reflected in your emissions as well as chemical reactants.

Appendix A:

Table A1: There are only 3 compounds which are marked with an * (used for this study). In the main manuscript it reads like all compounds are being used. Please clarify.

Table A2: “Kjeller, UK” must be changed to “Kjeller, Nor”.

Table A2: Please stick to the same number of decimal places. Two decimal places may be sufficient here.

Appendix B:

Table B3: Reference “Endo et al. (2023)” must be marked d), not e).

Appendix D:

Figures D1-D3: You are not referring to these Figures anywhere in the manuscript. Thus, I can’t infer any connection to the main manuscript. If you need these Figures, establish a clear reference. Moreover, the Figure captions should be self-explanatory.

References:

On several occasions, dois are given in the form <https://doi.org/https://doi.org/...> . Please shorten.

Page 51, line 782: Reference “Canada, C.H., 2010” must be changed. Issuing authority was Environment Canada. You could think of adding the Web link:
<https://www.canada.ca/en/environment-climate-change/services/canadian-environmental-protection-act-registry/historical/substances-lists/draft-screening-assessment-perfluorooctanoic-acid-salts-precursors.html>

Page 51, line 786: Change web link to: <https://doi.org/10.1016/j.envint.2024.108717> .

Page 54, lines 871-872: The zenodo publication is missing here.

Page 54, lines 882-883: It seems like you want to refer to the following publication: OECD (2018), Summary report on the new comprehensive global database of Per- and Polyfluoroalkyl Substances (PFASs), OECD Series on Risk Management of Chemicals, OECD Publishing, Paris,
<https://doi.org/10.1787/1a14ad6c-en> .

Page 54, lines 891-892: Name of this publication is: “ICON Tutorial 2024”.

Page 54, lines 901-902: Issuing authority is the Stockholm Convention on Persistent Organic Pollutants (POPs). The Reference name should reflect this by using something like “Stockholm Convention on Persistent Organic Pollutants, 2022” or “POPs Review Committee, 2022”.

Page 56, lines 950-952: The correct DOI is: https://doi.org/10.1007/978-3-642-15825-4_65 .