

Dear Reviewer #3,

Thank you for your minor corrections; all of them general and specific (listed below) have been solved, and you will find the details marked in bold below the corresponding correction.

We would like to thank the reviewer for their thoughtful, honesty, and constructive assessment of our work. We particularly appreciate the recognition of this paper as a pioneer study in the field of atmospheric PFAS modeling.

On the general comments, Referee 3 mentioned that:

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

Regarding the specific suggestion for the abstract, we have added a concise description of the hybrid coupling to the CAM-Chem background concentrations to ensure clarity for first-time readers:

This study presents the ICON-ART (ICOsahedral Non-hydrostatic model framework with Aerosol and Reactive Tracers) PRIEST (PFCA Reactions In Earth System Transport) model extension, developed to simulate the transport and transformation of Per- and Polyfluorinated Substances (PFAS) in the atmosphere. While the ICON-ART framework was developed to simulate atmospheric physics and chemical composition, the newly developed PRIEST extension incorporates additional gas-phase and aqueous physics, along with chemical reaction mechanisms, to model the transport, transformation, and deposition of Perfluoroalkyl Carboxylic Acids (PFCA) precursors. Therefore, the model includes 22 aqueous-phase reactions that depend on liquid cloud water and temperature. The aqueous-phase processes represent the adsorption of precursors in water droplets, with variable absorption rates. The model follows a hierarchical initialization, starting with the emissions, followed by aerosols, chemistry, and finally removal. To efficiently represent the atmospheric oxidative environment, PFAS chemistry is implemented in a semi-offline framework forced by prescribed monthly mean oxidant fields from CAM-chem. A simple parameterization of the OH radical is implemented to improve the simulation of PFCA precursors. The global model results ($\approx 105 \text{ km}^2$ grid resolution and 6 hours temporal resolution) show the capability of the model system to simulate regional and global variations of PFCA concentrations and their deposition processes. The results reveal an overestimation of atmospheric concentrations observed in Europe and an underestimation in East Asia. These differences are mainly related to the coarse spatial model resolution and the uncertainties arising from the underlying emissions model. In conclusion, ICON-ART PRIEST represents a significant step

forward in simulating the atmospheric fate of PFCAs precursors and their transformation products by integrating an enhanced chemical mechanism into the ICON-ART framework that couples both gas-phase and aqueous-phase processes, and also with the incorporation of a detailed temporally resolved PFAS emission inventory.

Specific comments by reviewer #3:

We are very thankful you pointed out some specific parts in our manuscript which should be corrected and/or improved. We also appreciate and agree with your comments addressed these individually as follows.

Introduction:

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

Now Page 2, line 24: Reference “Schymanski, 2023” has been added in the reference section as.

“Schymanski, E. L., Zhang, J., Thiessen, P. A., Chirsir, P., Kondic, T., and Bolton, E. E.: Per- and Polyfluoroalkyl Substances (PFAS) in PubChem: 7 Million and Growing, *Environmental Science and Technology*, 57, 16 918–16 928, <https://doi.org/10.1021/acs.est.3c04855>, 2023.”

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-climatechange/services/management-toxic-substances/list-canadian-environmental-protection-act/longchain-perfluorocarboxylic-acids.html>

Now page 2, line 35; reference states “Environment and Climate Change Canada, 2023”

Also, it was updated in the reference list as:

Environment and Climate Change Canada: Toxic substances list: long-chain perfluorocarboxylic acids (LC-PFCAs), their salts and precursors, <https://www.canada.ca/en/environment-climate-change/services/management-toxic-substances/list-canadian-environmental-protection-act/long-chain-perfluorocarboxylic-acids.html>, 2023.

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

The change was made on Page 2, lines 55 – 56: A table with a wider PFAS speciation (Including PFCAs and FTOH precursors) is included in the Table A1.

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

Now Page 3, line 66 – 68 reads as: “This extension improves the representation of PFCA precursors’ transformation, transport, and scavenging within the atmosphere building upon other authors previous modeling efforts (Thackray and Selin, 2017; Thackray et al., 2020; Wallington et al., 2006).”

Model Description:

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

Now page 4, lines 113 – 115: “The key focus was extending the simplified gas-phase reaction mechanism (Tables B1 and B2) from fluorotelomer precursors to PFCAs (Thackray et al., 2020). This is achieved by implementing both gas-phase reaction mechanisms and aqueous-phase chemistry (Table B3)”

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

Now Page 5, lines 121 – 125: “To study PFCAs in ICON-ART, we integrated aqueous-phase chemistry (Table B3) into ICON-ART based on the simplified gas-phase (Tables B1 and B2) PFCAs reaction mechanism built by Thackray et al. (2020). Originally, the mechanism accounted for 31 gas-phase and two photolytic reactions. It calculates the degradation of 8:2 FTOH, 8:2 FTO, and 8:2 FTI (where 8:2 represents the 8 fluorinated carbons, and a 2 carbon ethyl alcohol group in the molecule) as the main commonly emitted fluorotelomer PFCAs precursors (Prevedouros et al., 2006; Wang et al., 2014).”

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

Now page 5, line 130: “provided as monthly means for all reactants except OH.”

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

Now page 5, line 136: “...(further described in Fig. 1)”.

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

Now page 7, lines 187 – 189: “Thus, a compound with a high Henry’s constant favors the air, while a low value indicates it readily dissolves in water; however, it does not consider the timescale over which equilibrium is achieved”.

Model Setup:

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

Now page 9, line 245: "... unforced by further observations along the run-time period, meaning no data assimilation is applied".

Page 10, line 259: "... boundary conditions are provided for each restart ..."

Now page 10, line 265 has been corrected to: "... boundary conditions are provided for each restart ..."

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

Now page 10, line 281: Reference has been corrected to "(Young and Mabury, 2010)".

Corresponding to in the reference list to page 58, lines 1003 – 1005: "Young, C. J. and Mabury, S. A.: Atmospheric Perfluorinated Acid Precursors: Chemistry, Occurrence, and Impacts, vol. 208 of Reviews of Environmental Contamination and Toxicology, pp. 1–109, Springer New York, New York, NY, ISBN 978-1-4419-6879-1, https://doi.org/10.1007/978-1-4419-6880-7_1, 2010.1005"

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

Now page 10, line 283: corrected as you mentioned accurately to "Thackray and Selin (2017)".

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

Now page 11, line 307: Corrected to "Will et al., 2005".

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

Now page 12, line 322 – 323: Corrected to "(UNEP, 2024)".

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

Now page 12, line 327: Corrected to pg m^{-3} . Also, in the whole manuscript.

Model Evaluation:

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

Now page 14, lines 397 – 398 has been changed to: "On the other hand AFFF emissions are estimated by airport size handling the 60 biggest airports in the world 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.

Now page 21, lines 475 – 478 reads as: “As a general observation, the model tends to have larger discrepancies over mid-latitude regions. This uncertainty could be associated with the presence of a greater number and larger emission sources. Furthermore, the discrepancies may also be attributed to stronger variations in mid-latitude weather patterns, which are not fully reflected in the emission’s model and 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.

In page 9, lines 233 – 235 we now specify the compounds used for this study: “We apply a recently developed emission inventory (Simon et al., 2025) for four groups of PFAS (PFCAs, PFSA, FTOHs, and PFECAs) with 22 species, including different chain-lengths (Table A1), that are globally distributed on a 0.5 degree grid for each year from 1950 till 2020. Note that only the compounds marked with a * in Table A1 are used for this study”.

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

“Kjeller, UK” has been changed to “Kjeller, Nor” in table A2.

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

In table A2, all the decimal places are now established to three.

Appendix B:

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

Reference “Endo et al. (2023)” has been now marked with the label d).

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.

A new paragraph has been added in page 29, lines 641 – 650: “An analysis of the modeled versus observed air concentrations (pg m^{-3}) reveals, compound-specific biases in the model’s predictive performance. For PFNA, the model shows a general tendency toward overestimation, particularly for European sampling sites; though most predictions remain within a single order of magnitude of the observations (Fig. D2). Contrariwise, the model systematically underestimates

PFOA concentrations across all evaluated global regions (Africa, Americas, East Asia, and Europe), with the majority of data points clustering between the 1:5 and 1:10 underestimation boundaries, indicating a consistent negative bias that may stem from missing emission sources, overly rapid simulated atmospheric deposition or unaccounted weather phenomena non captured by the model (Fig. D1). Finally, the predictive accuracy for 8:2 FTOH do not describe a clear pattern compared to the PFCAs; the data exhibits large variance, with multiple predictions for both the Americas and Europe falling well outside the factor-of-ten agreement band, showing significant current limitations in accurately capturing the air concentrations of this volatile precursor (Fig. D3)”.

The three figure captions in Appendix D have been updated to become more explanatory as requested:

“Figure D1. Comparison of observed versus modeled PFOA concentrations (pg m^{-3}) by region on a log-log scale. The solid line represents 1:1 agreement, with dotted lines indicating deviations by factors of 2, 5, and 10”.

“Figure D1. Comparison of observed versus modeled PFNA concentrations (pg m^{-3}) by region on a log-log scale. The solid line represents 1:1 agreement, with dotted lines indicating deviations by factors of 2, 5, and 10”.

“Figure D1. Comparison of observed versus modeled 8:2 FTOH concentrations (pg m^{-3}) by region on a log-log scale. The solid line represents 1:1 agreement, with dotted lines indicating deviations by factors of 2, 5, and 10”.

References:

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

All the problems with dois in the whole document have been solved

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-environmentalprotection-act-registry/historical/substances-lists/draft-screening-assessment-perfluorooctanoicacid-salts-precursors.html>

Now page 53, lines 837 – 840 have been updated to: “Environment Canada: Draft Screening Assessment Perfluorooctanoic Acid, its Salts, and its Precursors, Tech. rep., <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>, 2010”

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

Now the complete reference is located in page 52, lines 803 – 805: “Coperchini, F., Greco, A., Croce, L., Teliti, M., Calì, B., Chytiris, S., Magri, F., and Rotondi, M.: Do PFCA drive the establishment of thyroid cancer microenvironment? Effects of C6O4, PFOA and PFHxA exposure in two models of human thyroid cells in primary culture, *Environment International*, 187, 108 717, <https://doi.org/10.1016/j.envint.2024.108717>, 2024.805”

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

Now page 55, lines 896 – 897, the zenodo doi have been added: “Meza-Landero, H. A., Bruckert, J., Petrik, R., Simon, P., Vogel, H., Matthias, V., Bieser, J., and Ramacher, M. O. P.: ICON-ART PRIEST extension code, <https://doi.org/10.5281/zenodo.16615113>, 2025.”

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> .

Now in page 55, lines 907 – 908 have been updated to: “OECD: Summary report on the new comprehensive global database of Per- and Polyfluoroalkyl Substances (PFASs), OECD Series on Risk Management of Chemicals, OECD Publishing, Paris, ISBN 9789264884434, <https://doi.org/10.1787/1a14ad6c-en>, 2018.”

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

Now in page 55, line 916 – 917, name of this publication has been updated: Prill, F., Reinert, D., Rieger, D., and Zängl, G.: ICON Tutorial 2024, Deutscher Wetterdienst, https://doi.org/10.5676/DWD_pub/nwv/icon_tutorial2024, 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”.

Now in page 57, lines 963 – 965: “Stockholm Convention on Persistent Organic Pollutants (POPs): POPRC-17/6: Long-chain perfluorocarboxylic acids, their salts and related compounds, Tech. rep., Persistent Organic Pollutants Review Committee (POPRC.17), <https://www.pops.int/TheConvention/POPsReviewCommittee/Meetings/POPRC17/Overview/tabid/8900/ctl/Download/mid/25875/Default.aspx?id=77&ObjID=31061>, 2022.”

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

Now in page 57, line 981: The DOI have been updated to
“https://doi.org/10.1007/978-3-642-15825-4_65”

Once again, we are grateful for your careful reading and suggestions, which have helped us sharpen the discussion of uncertainties and make the model’s limitations and next steps much more transparent.

Warm regards,

Hiram Meza

on behalf of all co-authors