

1 Reply on Reviewer Comment 1

Dear Reviewer,

We sincerely thank you for your extensive review and optimistic outlook on the usefulness of our dataset. In the following we address all comments and how they shaped the updated version of our manuscript.

Q: Line 2: how was “most widely used” determined?

A: We determine “most widely used” based on the available literature regarding total production volume. But as data is limited and the landscape of used PFAS is shifting rapidly we attenuated it as “some of the most widely used”

Q: Line 50: do you plan to include other POP’s in this inventory eventually? Otherwise shouldn’t it be named something specific to PFAS?

A: As the numerical framework is in theory not PFAS specific and specifically build to be able to support other organic pollutants we settled on this name and plan to extend it if time permits. We agree that the name could be improved, yet during the review phase we already gathered a small user community and would not like to confuse people by renaming it at this time.

Q: Line 57: technically no PFAS have been “outlawed” at least in the US (I’m not sure about elsewhere)—PFOA and PFOS were voluntarily phased out by companies

A: To our knowledge, the Stockholm Convention banned the use of PFOA and PFOS which the US signed but has not ratified (see UNEP, POPS/POPRC.1/9 and decisions SC-9/12 and SC-4/17, as well as the congress Executive Report 108-8 - CONVENTION FOR INTERNATIONAL CARRIAGE BY AIR (TREATY DOC. 106-45))

As the legal specifics are not relevant at this point in the text, we changed “outlawed” to “phased-out”

Q: Line 77: just to clarify, all of the species except HFPO-DA and Adona are currently or soon to be regulated in the EU?

A: At the state of writing the European Commission published Proposal COM/2022/540 regarding guidelines to address PFAS pollution in groundwater and drinking water including upper limits for the sum of concentrations for the named 24 PFAS. This includes HFPO-DA and Adona.

Q: Table 1: suggest adding CAS numbers as well

A: That is a very good suggestion. We added CAS numbers to the manuscript.

Q: Line 110: why just Swedish airports? Can you take the emissions from these 2 references and apply them to airports globally?

A: This decision was based on the excellent quality of the two given studies that provide total estimations not typically given for studies measuring near pollution sources like airports. The set standards of the ICAO regarding firefighter training exercises, the main source for PFAS pollution of airports, led us to value spatial consistency over the approximation errors that we expect of this approach. At least in theory fire fighting exercise frequency should depend on airport size and be similar in ICAO all member countries.

Q: Table 2 is a bit hard to read as is—suggest left-justifying the text, or adding borders for the boxes maybe?

A: We left-justified the text and added borders.

Q: Line 164: do you have a reference for the statement that PFAS are typically polymerization aids? My understanding is that the monomers (e.g. TFE, HFPO, etc) are often emitted at large quantities as well. But I think the following statement is true regardless if the emission in question is a monomer or a processing aid?

A: A reference has been added. The scientific effort has been focused so far on the polymerization aids, while the monomers and resulting fluoropolymers might be of high concern as well (Lohmann et al. 2020). As the two statements in this sentence are actually independent of each other they have been split up.

Q: Line 202: GenX is now produced by Chemours (which was formerly DuPont), I think DuPont did make it before they spun off Chemours in 2015 though

A: That is absolutely correct, thank you for pointing out that error. We changed that in the manuscript accordingly.

Q: Line 204: this sentence is confusing as worded, both GenX and Adona can be used to make both PFOA and PFNA? I think either “both” should be replaced with “they”, or “respectively” needs to be removed

A: Yes, the way it is worded is confusing, we replaced “both” with “they”.

Q: Line 213: this is a bit weird to give percentages without giving the base number for PFOS—can you give the base number here or tell the reader where that base number can be found in the manuscript or SI?

A: The absolute numbers have been added.

Q: Line 277: what percent of airports is this? What defines “relevant” classes?

A: This meant all but the small civilian airports as indicated two sentences later, that has now been moved up to clear up the confusion.

Q: Line 278: what is the “huge disparity”? Much more emissions at which type of airport and ballpark/percentage how much more?

A: Thank you for this suggestion, we indicate in the text now, that the approximated emissions in military airports have been show to be able to reach three times the mass of comparable civilian airports.

Q: Line 279: why aren’t large civilian and military airports considered?

A: They are handled as described two paragraphs before, we added “As large civilian and military airports are handled individually...”

Q: Line 302: should this be “the entire world”?

A: The groups are meant to be exclusive, but the title “Consumers” suggests otherwise. We changed that to “non-producing Consumers”

Q: Section 2.4.2: Can you provide a map with the location of each of the sites?

A: We added a map with the location of each of the sites in section 2.4.1

Q: Section 2.4: emissions are distributed equally between identified sources—can you comment on how realistic this is and how different allocations between individual sources might impact modeling results?

A: As the largest sources with fluoropolymer production are accounted for individually (and making up for example up to 80

Q: Line 386: PFAS were first produced sometime in the 1930’s-1940’s, suggest changing “the whole time span since the first production” to something like “the main time span of PFAS emissions, from early production in the 1950’s to present day”

A: To keep the consistency we changed it to “industrial scale production”.

Q: Figure 1 (and all subsequent figures): maybe this is not the final figure, but could you please increase and bold font size, increase marker size, and increase line thicknesses?

A: Done

Q: Line 407: do you need “seemingly” here? Since you show the exponential curve with it?

A: Changed.

Q: Line 454: why might POPE be underestimating the time lag?

A: There is no explicit time lag included for emissions by disposal sites. In reality PFAS should slowly leach depending on environmental conditions and product. As this is hard to accurately estimate this has been included in the scaling, therefore allowing a maximum time lag of 1 year which is probably too low. This explanation has been added to the manuscript.

Q: Line 471: this sentence is confusing to me, I think maybe “Apart” is not the correct word to use? And the wording of “has to be captured by POPE”? Are you trying to say something like “Consumption of PFAS-containing products grows as the number of people and their income grows, which is captured by POPE”?

A: No, we want to state, that increased PFAS-products use has two distinct parts, general wealth increase and commonly available products getting substituted by PFAS containing products, whereas

the second part has not been accounted for up to this point in the methodology. But it is stated confusingly and has been completely rephrased as “With technological advancement, PFAS containing products grow more prevalent as a share of all consumer expenses”

Q: Line 571: see previous comment, I’m not sure you can definitively say 1950 was the first year PFAS were emitted to the environment. Could alter this sentence to say something like: “are, for the most part, temporally complete.”

A: We assume, that the emissions before that time falls well below our model uncertainty, which we now state accordingly.

Q: Line 578: do you have a reference for this being the largest PFAS emitter? I’m not sure that is the case?

A: It is up to this point the largest in North America based on the production data that we have a hold of, but any hints at missing sites are very much appreciated.

Q: Line 629: the sentence starting “Also the general trend. . .” is confusing to me, could you please re-word it for clarity?

A: Indeed, that has now been rephrased to “The expected changes due to an increase in emission load relative to the total watermass follow the expectation

Q: Figure 9: suggest spelling out “Modeled” on the x-axis, and if possible reversing the legend (not necessary, may be too difficult, but would be nice)

A: We changed this according to the reviewer’s suggestions.

Q: Line 795: see previous comments, suggest changing “beings with the first production of PFAS in 1950” to something like “begins with the year 1950”

A: Also taken care of, thank you.

Thank you further for the corrections and suggestions, which helped to increase the quality of our manuscript.

2 Reply on Reviewer Comment 2

Dear Reviewer,

Thank you for your comments, they are all very much appreciated and helped us tremendously in the improvement of our manuscript.

Q: “The only major concern I have is the evaluation of uncertainties from various data sources. The authors should discuss more about how the uncertainties are considered in the method, and how the uncertainties from the original datasets influence the key conclusions from the results.”

A: To address your major concern another chapter ”speciation” has been added. The corresponding section in the results has also been expanded to explain how we expect the uncertainties from used datasets and their interaction affects results and how they are incorporated into our upper bound and lower bound scenarios.

For your specific comments:

Q: Title: I recommend adding spatial and temporal resolution in the title.

A: We agree with the reviewer and changed the title of the manuscript to “POPE: an Annual Global Half Degree Emission Inventory for PFAS 1950-2020”

Q: Abstract: The journal requires a link to the data repository in the abstract, which is currently missing. Since I don’t have access to the data, I cannot evaluate the data quality beyond what is presented in the manuscript.

A: That link has been added: <https://doi.org/10.5281/zenodo.12783504>

Q: L89: Please add more information on the downscaling process.

A: As also our other reviewer had questions on this and that is one of the main sources of the mentioned uncertainties an extended explanation is now also found in the ”speciation” chapter.

Q: L110: why Swedish airports? How representative are they?

A: As stated in the answer to our other review we assume that based on ICAO standards, they are closely comparable to other airports in country group one, and a good first order estimate for all

other airports. As we are aware, that AFFF use might still vary wildly, and different environmental standards are at play, the well researched swedish airports are a baseline scenario, where we expect and account for the large uncertainty of this assumption in our upper and lower bounds.

Q: L119: please be more specific about what these datasets are.

A: The formatting was a bit irritating in this list, as this is the headline for the following datasets. To avoid confusion, the listing symbols have been removed from the headlines

Q: L372: ‘caused by’ – ‘from’

A: Changed

Q: L531-532 ‘As . . . estimates’: this sentence is not clear to me, please rephrase.

A: It is indeed irritating and has now been rephrased to: “POPE relies for the biggest emission sector: fluoropolymer production, on the inventory Wang et al.. Therefore both are expected to yield similar results.”

Q: Figure 6: if ‘Asia’ represents ‘Asia excluding Japan’, please be clear about it.

A: Thank you, that has been changed

Q: L581: is it concluded from Figure 5 or elsewhere? I couldn’t find it.

A: This is indeed not shown in the figure and rather an observation from the underlying data which has now been clearly stated.

Q: L681-683 ‘A fraction . . . the USA’: why is it? Could you explain more?

A: As the fraction of the emissions that is distributed top-down rather than bottom up is conserved, small scale industrial production and consumer use “compete” for their relative contribution as an emission source. As the methodology to account for industrial production is based on population and GDP in Asia whereas it is based on datasets like the EPRTTR and FRS in country group one, this in theory may lead to a shift of emissions from country group two to country group one when the industrial fraction of the per person emissions in country group two is underestimated.

Q: Figure 8: please increase the limit of y-axis. Currently the whisker-bars of Haw-River and Ohio-River are out of the plot.

A: As to not reduce the readability of the plot, the upper limit of the whisker bars is now given in the text and caption.