Responses to the **Reviewer 1** comments

Comment: In their manuscript, the authors present results from collecting PM10 samples from a scaled-down activated sludge treatment tank and analyzing the samples for several emerging and legacy PFAS. The study was conducted in October 2023 and repeated in March 2024 in the UK, and domestic sludge from a regional WWTP was used. Overall, the manuscript is well written and appropriately structured, and it fills a gap in the literature regarding the study of aerosolization of PFAS during WW treatment. However, there are a few issues that the authors should address.

Response: We would like to sincerely thank Reviewer 1 for his/her important comments and for recognising the significance of our study in addressing the knowledge gap regarding the aerosolisation of PFAS during wastewater treatment. We appreciate the reviewer's acknowledgment that the manuscript is well-written, appropriately structured, and fills the gaps in the literature. Our detailed responses to the reviewer's comments are provided below in blue. The new text added to the revised manuscript is shown below in italics.

Major issues:

- From the brief description in the abstract and the slightly more extensive description in the Methods section (L. 107-112), the setup of the scaled down (what is the scale?) in conjunction with the "parent WWTP" is unclear and I strongly suggest adding more detail, including the flowrate of the sludge that is supplied to the smaller tank and how it is connected to the main WWTP.

  Response: The authors thank the reviewer for the comment. We have a strict non-disclosure agreement (NDA) with the wastewater treatment company where we collected the samples. The NDA imposes restrictions on us to add details such as the scale of the smaller tank in comparison to the main tanks, the flow rate of the sludge, and how the scaled-down tank is connected to the main WWTP. However, we strongly believe that even without this information, our results still provide critical information on PFAS emissions, particularly for legacy PFAS that remain in wastewater streams despite their ban, highlighting their persistence in sewage processing and their distribution beyond expected environments, including partitioning between water and aerosol phases in sewage processing.

  In the conclusion of the abstract and also in the Results & Discussion section, the concerns associated with aerosolization of PFAS from WWTP remains vague and should be summarized more explicitly, given the persistence of PFAS in the environment and their ability to travel long distances. As other (industrial) sources become more restricted, the focus likely has to shift to sources like WWTPs. Please add this discussion to the manuscript.

  Response: We sincerely appreciate the reviewer's insightful comment. However, our study primarily focuses on quantifying airborne PFAS concentrations within the WWTP atmosphere and was therefore submitted as a measurement report. The study aims to demonstrate that WWTP processes have the potential to introduce PFAS into the atmosphere, even when treating domestic wastewater. A comprehensive assessment, including concurrent measurements of PFAS levels in both wastewater and air, would be necessary to fully characterise the extent and discuss the implications of PFAS aerosolisation from WWTPs.

  We believe that our study serves as a foundation for future research on PFAS emissions from domestic waste sources. Future studies should incorporate simultaneous monitoring of PFAS in wastewater, particulate matter, and the gaseous phase to enhance our understanding of emission pathways and associated risks.

We have now added a limitation section to address the above comment: "*Future research should consider simultaneous characterisation of wastewater PFAS levels alongside PM measurements to improve understanding of the relationship between airborne PFAS emissions. Expanding the range of monitored PFAS beyond the 15 fully validated targets in our study, particularly including neutral PFAS such as FTOHs and FOSEs, would enhance understanding their role in the WWTP aerosolisation. Additionally, incorporating gas-phase sampling would be valuable in assessing the potential partitioning of PFAS into the gaseous phase, further refining our understanding of their atmospheric behaviour*." (lines 326-331 page 13 of the revised main text).

Nonetheless, as stated above, our findings provide critical insights into how domestic wastewater treatment can facilitate PFAS distribution in unexpected environmental compartments, particularly through partitioning between water and aerosol phases during sewage processing.

- In conjunction with the previous comment, in L. 83 the authors state "…a knowledge gap exists regarding the atmospheric fate of PFAS…", which I suggest rephrasing as "…a knowledge gap exists regarding the release of PFAS into the atmosphere…" to better fit the context of this manuscript.

  Response: As suggested, the original text was changed to "*Consequently, a knowledge gap exists regarding the release of PFAS to the atmosphere, particularly their association with* PM_{10} *aerosols, during the treatment of domestic wastewater, especially under conditions of vigorous aeration*." (lines 83-85, page 3 of the revised main text).

- Please provide the LODs and LOQs for this study in the SI.

  Response: The LODs and LOQs for this study have been included in the SI as Table S5.

- As part of the discussion in L. 179-189 it should be stated that PFAS levels were not measured in the sludge itself, which makes any explanation for the observed trends highly speculative. This fact is touched upon in L. 222-224 and again in L. 246-251, but needs to be mentioned earlier and more explicitly.

  Response: As suggested, the following sentence has been added to the Section 3.1 of the main text: "*It should be noted that the concentrations of PFAS in the wastewater were not measured in our study*." (line 201-202, page 9 of the revised main text). This is also now mentioned in the limitation section: "*Future research should consider simultaneous characterisation of wastewater PFAS levels alongside PM measurements to improve understanding of the relationship between airborne PFAS emissions. ….*" (lines 326-331 page 13 of the revised main text).

- I suggest moving the three sentences from L. 246-251 to L. 181, because the discussion provided in these sentences is important for context at this earlier point.

  Response: We thank the reviewer for this comment. This has been now addressed in Section 3.1 of the main text: "PFBA is one of the most volatile PFAS observed in our study. Further, short chain PFAS such as PFBA and PFBS are reported to have lower aerosolisation tendencies compared to long chain compounds (e.g., PFOS and PFOA) (Johansson et al., 2019; Pandamkulangara Kizhakkethil et al., 2024). Despite being volatile and having low aerosolisation tendency, the presence of PFBA in the PM_{10} aerosols at considerable concentrations in our study could potentially be due to the presence of high levels of PFBA in the sewage during the sampling period." (lines 188-193, page 9 of the revised main text).

- 190-200: The authors are discussing different general sources of the detected PFAS, but are not clear about how these PFAS might have been transferred into the sewage sludge. Are the authors aware of any of those industries in their sampling region? What about levels of PFAS in the local drinking water supply, which likely makes up a large portion of the wastewater? Laundry water may also be a source, as PFAS have been detected in dryer lint.

  Response: As detailed in the manuscript, the WWTP treats domestic wastewater, with no known nearby industries that could serve as a potential point source of PFAS. Regarding the drinking water as source of PFAS, we are not clear about this question. We agree with the reviewer that laundry water may be one of the many sources of PFAS in the domestic wastewater. This has been now included in Section 3.1 of the main text: "*Laundry water could potentially be one of the sources of PFAS in the sewage since the WWTP receive a major portion of the sewage from households* (Clara et al., 2008)." (lines 213-214, page 9 of the revised main text).

- 225-232: The discussion of the diurnal trends seems incomplete: The only clearly observable trend appears to be that a larger number of PFAS are present above LOD/LOQ during the night compared to the day. Why is that? If a compound is detected during the day, its level is ballpark similar to the corresponding night measurement. PFOA appears to be the only exception with clear spikes on some days. Maybe people do more of their laundry in the evening, run the dishwasher, produce more PFAS containing WW? Please revise this section.

  Response: Thank you for this comment. The sewage in the AS tank is mixed and contains wastewater from both the day and the night-time periods. As detailed in Section 2.2, the scaled-down tank receives and processes sewage from the main tanks, which is already primary treated. Therefore, the influence of diurnal household activities could not be captured in our samples. One of the potential reasons for the diurnal variations of PFAS concentrations could be the shorter sampling duration of the daytime samples (collected between 10.00 AM-3.00 PM) in comparison that of to the night samples (collected between 3.00 PM and 10.00 AM the following day). The MiniVol™ tactical air sampler does not allow automatic filter change requiring us to manually replace filters within the facility's access hours. The shorter sampling duration of day samples could lead to lower PFAS mass load in the samples and thereby <LOD values. This information has been now included in Section 3.1: "*The shorter sampling duration of day samples compared to the night samples likely led to a lower aerosol mass load on the filters, resulting in several PFAS mass loads below the LODs, which could explain the observed diurnal differences in PFAS concentrations*." (lines 248-250, page 10 and 11 of the revised main text).

- I recommend adding a Limitations section, which should include at a minimum the following considerations: only one location was sampled during limited time frame, the sewage sludge was not analyzed to investigate the relationship between PFAS concentrations in PM10 and in the sludge, PM10 concentrations were not measured nor was the PM10 further characterized, and neutral PFAS (e.g., FTOHs, FOSEs) were not included in the analysis.

Response: A limitation section has been now included in the main text: "*Future research should consider simultaneous characterisation of wastewater PFAS levels alongside PM measurements to improve understanding of the relationship between airborne PFAS emissions. Expanding the range of monitored PFAS beyond the 15 fully validated targets in our study, particularly including neutral PFAS such as FTOHs and FOSEs, would enhance understanding their role in the WWTP aerosolisation. Additionally, incorporating gas-phase sampling would be valuable in assessing the potential partitioning of PFAS into the gaseous phase, further refining our understanding of their atmospheric behaviour*." (lines 326-331 page 13 of the revised main text).

Minor issues:

- I understand that this manuscript has been submitted as a measurement report, but does that categorization have to be part of the title? The "Measurement report" part is missing from the title in the SI, so at least the title should be made consistent.

  Response: The requirement of the journal for SI title is as follows "**Supplements will receive a title page added during the publication process including title ("Supplement of"), authors, and the correspondence email. Therefore, please avoid providing this information in the supplement**.". Therefore, the title of the SI has been now removed.

- Throughout the manuscript, I suggest replacing several instances of "aerodynamic size" with "aerodynamic diameter", including in the abstract.

  Response: This has been now addressed in the main text (lines 14, 70, 280, 281, and 294 in pages 1,3, 11, 12, and 12, respectively, of the revised main text).

- 114: Define "near" – how far from the aeration tank was the sampler installed?

  Response: The sampler was placed at a distance less than 0.2 m from the aeration tank. This information has been now included in Section 2.3 of the main text: "The MiniVol™ tactical air sampler (Air Metrics, United States of America) used for PM_{10} sampling was installed near the aeration tank *(<0.2 m from the aeration tank) with the sampling head slightly above the rim of the tank (10 cm above).*" (lines 114-115, page 4 of the revised main text).

- 118: Define "day" and "night" – what were the time frames? And why was sampling so much shorter during the day compared to the night?

  Response: We thank the reviewer for this important comment. Day sampling started after 10:00 AM and finished before 3:00 PM, while night sampling began after 3:00 PM and ended before 10:00 AM the following morning. As mentioned in our response to another question above, the MiniVol air sampler does not allow for automatic filter changes at a specified time and thus filters were manually changed during the allowed access hours to the facilities. Due to these limitations, the sampling duration for the daytime samples was shorter than that of the nighttime samples.

  The 'day' and 'night' have been now defined in Section 2.3 of the main text: "*PM_{10} samples were collected separately during day (between 10.00 AM and 3.00 PM) and night (between 3.00 PM and 10.00 AM the next day).*" (lines 118-119, page 4 of the revised main text).

- 128: Regarding the importance of sampling artifacts, especially for PFAS, please consider Chang et al. (2024), "Indoor air concentrations of PM2.5 quartz fiber filter-collected ionic

PFAS and emissions to outdoor air: findings from the IPA campaign" (https://doi.org/10.1039/D4EM00359D).

Response: The suggested reference is now cited in Section 2.3 of the main text "It is important to note that the use of GFF and quartz fiber filters (QFF) during PM sampling has been reported to cause positive sampling artefacts, such as the adsorption of gas-phase organic compounds (*Chang et al., 2025*; Turpin et al., 1994)." (lines 129-130, page 5 of the revised main text).

- 131: Delete "slightly".

  Response: The word 'slightly' has been removed.

- 139: What were the filters prewashed with?

  Response: The filters were prewashed with 15 mL of methanol as described in Kourtchev et al. (2022). This has been now addressed in Section 2.4 of the main text: "*The methanol extracts were then filtered through a prewashed with methanol (3 times with 5 mL) 0.45 µm PTFE syringe filter.*" (line 140-141 page 6 of the revised main text).

- Tables S3 and S4 mention that the results were blank corrected. Please add a brief description about the process to the Methods section.

  Response: We thank the reviewer for this important comment. This has now been included in Section 2.4 of the main text: "*The maximum concentration value of the PFAS detected in the field blanks and baked filter blanks were subtracted from the PFAS concentrations detected in the samples.*" (lines 163-165, page 6 of the revised main text).

- Section 2.5: Were field blanks collected?

  Response: Field blanks were collected during the campaign. This was mentioned in Section 2.3 line 125 of the main text in the original submission. For more clarity this has now been modified to "*Two types of blanks were used to evaluate possible PFAS contamination from handling the filters. These include: 1) baked filters (BF) and 2) baked filters placed in MiniVol® air sampler and collecting air above the AS tank at 10 L min^{-1} for 2 min (field blanks, FLDB).*" (lines 125-128, page 5 of the revised main text).

  The blank values including the field blank values have now been added to the SI as Tables S6 and S7.

- 208-209: The sentence about the pH value is difficult to understand. Please rephrase.

  Response: The sentence was revised to "*The pH of the contaminated water has been reported to influence the atmospheric transfer of PFAS*". (lines 222-223, page 10 of the revised main text).

- 218: Period missing after "concentration".

  Response: the period (.) has been added.

- 218-219: Rain water may also dilute the wastewater, depending on the PFAS sources.

  Response: This has been now included in Section 3.1 of the main text: "*Additionally, dilution of PFAS levels in the sewage due to rainfall could also affect the airborne PFAS concentration*." (lines 233-234, page 10 of the revised main text).

- 252 and L. 257: It is my understanding that Weinberg et al. measured PFAS in TSP and not “estimated” the concentrations. “Estimated” indicates theoretical/modeling results. Please revise.

  Response: The word ‘estimated’ is now replaced with ‘*measured*’.

- 261: Why the parentheses around “up to 1.31 pg m-3”?

  Response: The parentheses have been removed.

- 307: Replace “could potentially represent” with “likely represent”.

  Response: Done.

- 320: Replace “filed” with “field”.

  Response: “filed” has now been replaced with “*field*”.

- SI: I recommend including the full author list in the SI instead of using “Jishnu Pandamkulangara Kizhakkethil et al.”.

  Response: The requirement of the journal for SI title is as follows “**Supplements will receive a title page added during the publication process including title ("Supplement of"), authors, and the correspondence email. Therefore, please avoid providing this information in the supplement.**”. Hence, the author information has been now removed from SI.

- SI: Please include table and figure captions in the table of content.

  Response: The table and figure captions have been included in the table of contents of the SI.

- SI: Please list CAS RNs or other unique identifiers in Table S1 together with the PFAS name and abbreviation.

  Response: The CAS RNs for the PFAS in the EPA 533 PAR mix have now been included in Table S1 of SI.

- SI: Please indicate clearly in the captions of Tables S3 and S4 that none of the other targeted PFAS were detected in any sample. If any were detected at least once, I recommend including the concentration in the provided tables.

  Response: We thank the reviewer for this comment. This has now been addressed in the captions of Tables S3 and S4 of SI: “*Other targeted PFAS amenable to the method but not listed in the table were either below the method's LOD or not detected in any of the collected samples*”. (lines 62 and 76, page 4 and 5, of revised SI).

References

Clara, M., Scharf, S., Weiss, S., Gans, O., and Scheffknecht, C.: Emissions of perfluorinated alkylated substances (PFAS) from point sources—identification of relevant branches, Water Sci. Technol., 58, 59-66, https://doi.org/10.2166/wst.2008.641, 2008.

Weinberg, I., Dreyer, A., and Ebinghaus, R.: Waste water treatment plants as sources of polyfluorinated compounds, polybrominated diphenyl ethers and musk fragrances to ambient air, Environ. Pollut., 159, 125-132, https://doi.org/10.1016/j.envpol.2010.09.023, 2011.