

Review comments on "Sea spray aerosol as a newly identified source of atmospheric gas-phase perfluoroalkyl carboxylic acids" by Aggarwal et al.

General Comments:

This paper presents an interesting idea and addresses a state-of-the-art problem concerning the role of sea spray aerosol (SSA) in the atmospheric transport of perfluoroalkyl carboxylic acids (PFCAs). The dataset, combining field observations from the Eastern North Atlantic with controlled laboratory studies using HR-ToF-CIMS, is highly useful and provides the first evidence for the unconsidered pathway.

However, in its current form, the manuscript reads more like a short letter paper than a full regular article suitable for ACP. While the core finding is impactful, the manuscript needs a major expansion, particularly regarding the mechanistic proof of the conceptual schematic. The data show that gaseous PFCA release depends on low pH, but the paper must do more to prove the mechanistic chain. I recommend a major expansion to provide a more robust quantitative and mechanistic analysis.

Specific Comments:

1. Introduction/Context (Line 58-61):

The manuscript states that previously reported SSA transport has been considered a "main pathway." Since SSA itself is a form of emission from the ocean surface, this phrasing is confusing. I suggest rewriting these two sentences to clarify the distinction between direct emissions from point sources and SSA. The key is to clearly show what the challenge is.

2. Data/References (Line 331):

The manuscript cites "oceanic levels ($\sim 35 \text{ g kg}^{-1}$)" for salinity. Please provide a reference to support this baseline number.

3. Real-World Applicability (Line 351-352):

The conclusion that "both high salinity and low pH are required for gas-phase PFCA release" raises a critical question regarding the environmental relevance of this mechanism. I am curious: are the specific combinations of high salinity and low pH required for this process actually common in the real oceanic environment?

- Suggestion: add figures or analysis to address this. Specifically, the manuscript would be significantly strengthened by clarifying whether this mechanism is a major, frequent process in the real atmosphere, or if it is a minor, idealized pathway that may only occur under rare conditions. If it is the latter, the mechanism might be interesting but of limited use for solving the real-world problem. The authors need to argue convincingly for the

environmental significance of their findings, perhaps by estimating the frequency of such conditions in the Eastern North Atlantic based on their field data.

4. Conceptual Schematic (General):

The conceptual schematic is very nice and visually explains the proposed process (SSA formation → drying → pH decrease → gas-phase partitioning). However, it currently functions as a hypothesis rather than a proven conclusion. It lacks a robust link to the data.

- Suggestion: I recommend adding more analysis to make the schematic "solid." For example:

- Can the authors use their data to estimate the percentage of total PFCAs that is in the gas phase as a function of pH?

- Can the field observations be compared directly to the laboratory results to validate that the proposed mechanism occurs under natural conditions?