RE: Point-by-point response to the Editor's comments

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Title: "Reaction kinetics and multi-sulfur products formation of sulfur-containing volatile organic compounds with OH radicals"

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Dear Prof. Jason Surratt,

Thank you very much for your careful evaluation of our revised manuscript.

We have carefully rechecked the whole manuscript and Supplemental Information and corrected some expressions of "*low* NO_x". Besides, we have also expanded Section 2.2 to clarify the collection, transportation, storage, and handling of the freshwater algae samples. We further discuss the possible influence of O_3 reactions at the air-liquid interface during the subsequent oxidation stage and explain that a direct mass-spectrometric "sniff" measurement of the original water sample was not conducted.

In the point-by-point response below, we address each of your comments in detail, specifying the corresponding revisions implemented in the manuscript and SI.

We thank you again for your time and consideration!

Yours sincerely,

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Editor Comment 1

Line 26: change "low NO_x" to "low-NO_x conditions." Note this mistake was made in many other places throughout the text.

Response:

We have changed "under low NO_x" to "under low-NO_x conditions" at the indicated location in the Abstract. We also carefully checked the entire manuscript and Supplemental Information and corrected the other occurrences of "low NO_x".

Editor Comment 2

Line 49: change "particulate sulfate ," to "particulate sulfate,"

Response:

We have removed the extra space before the comma at the indicated location.

Editor Comment 3

Collection details of the water samples. It remains unclear to me in the main text how the water samples were collected, transported and stored before being separated into 12 petri dishes that were placed in the chamber. Can the authors clarify what exactly the water was collected into, how much total water was collected, whether the water was filtered or had turbidity removed before its use, and if there was any concerns of how large concentrations of O3 would affect what VOCs you would measure? For the latter, could gas-phase O3 in the chamber react at the air-liquid interface of your petri dishes changing what VOCs would off gas? Meaning, could ozonolysis reactions happen with the bacteria or other biological materials releasing more high volatility organics from the water? I think it would be interesting to know how the chamber VOC profile compares to a "sniff" test of the water sample (without being in the chamber) by the CIMS.

Response:

Thank you for raising these important questions. We agree that the previous version did not provide sufficient details regarding the collection, transportation, storage, and handling of the freshwater sample. We have therefore expanded the statements to clarify these procedures.

Approximately 10 L of algae-containing lake water was collected using a clean 12 L polyethylene terephthalate (PET) drinking-water bottle. The sample was promptly transported to our laboratory and kept in the laboratory at approximately 21 °C for approximately 2 d before the chamber experiment. No filtration or other turbidity-removal treatment was applied before use. The untreated algae-containing water was directly distributed among 12 petri dishes, providing a total exposed liquid surface area of 0.213 m^2.

After the petri dishes containing the water samples were placed in the chamber, the chamber was thoroughly flushed with zero air at a flow rate of 15 L min^{-1} for 2 h. After the zero-air flushing was completed, no additional gas was introduced, and the water samples were kept under static conditions in the chamber for 18 h at approximately 21 °C to allow the release and accumulation of VOCs. After this static period, the VOCs released from the samples were measured by Vocus-PTR-LToF-MS for approximately 30 min before O_3 was introduced. Only after completion of the pre-oxidation VOC measurement, O_3 was introduced into the chamber to a concentration of approximately 90 ppb, and followed by illumination with two 253 nm UV lamps to initiate •OH-oxidation of VOCs. Therefore, the pre-oxidation VOC profile shown in Figure 4 was obtained in the absence of O_3 and was not affected by the injection of O_3.

We agree that, during the subsequent O_3/UV oxidation stage, gas-phase O_3 could potentially react with reactive dissolved constituents or biological material at the air-liquid interface. Such interfacial reactions could, in principle, consume certain reactive compounds or generate additional volatile oxygenated products. Because these processes were not independently quantified in our experimental design, their possible contribution to the species observed during the oxidation stage cannot be completely excluded. However, this possible influence applies only to the subsequent oxidation-stage measurements and not to the pre-oxidation VOC profile shown in Figure 4, which was measured before O_3 addition.

A direct mass-spectrometric “sniff” measurement of the original water sample or the headspace of the collection container outside the chamber was not conducted. Therefore,

a direct comparison between the chamber VOC profile and the immediate headspace composition of the collected water is not available. We have clarified this limitation in the revised manuscript. The profile shown in Figure 4 represents VOCs released from the untreated water sample and accumulated in the chamber during the static incubation period, rather than an instantaneous measurement of the original sample headspace. Parallel direct-headspace and chamber measurements would be valuable in future studies.

Representative revision in the manuscript, Lines 179-190:

"Approximately 10 L of algae-containing lake water was collected using a clean 12 L polyethylene terephthalate (PET) drinking-water bottle. The sample was promptly transported to the laboratory and kept in the laboratory at approximately 21 °C for approximately 2 d before the chamber experiment. No filtration or other turbidity-removal treatment was applied before use."

"During the emission characterization process, water samples were divided into 12 petri dishes (with a total exposed liquid surface area of 0.213 m^2) and placed in the chamber. The chamber containing the water samples was then flushed with zero air at a flow rate of 15 L min^{-1} for 2 h. After the flushing was completed, no additional gas was introduced, and the water samples were kept under static conditions in the chamber for 18 h at approximately 21℃ to allow the release of VOCs (Figure S2b). After this static period, the VOCs released from the water samples were measured by Vocus-PTR-LToF-MS for approximately 30 min before O_3 was introduced into the chamber."