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Jason D. Surratt
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Title: Measurement report: Biogenic VOC emissions profiles of Rapeseed leaf litter and their SOA formation potential
Author(s): Letizia Abis et al.
MS No.: acp-2021-135
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Dear Jason,

We are hereby submitting our revised manuscript entitled “Biogenic VOC emission profiles of rapeseed leaf litter and its SOA formation potential” by Abis et al.

We noted that 2 reviewers out of 3 were recommending the publication of our manuscript while the second reviewer raised again some concern about the repeatability of the starting conditions of our experiments.

As it can be seen in the point-to-point answer, we clearly show that the initial conditions were comparable for all set of experiments. This clearly address the concerns raised by this reviewer. We are therefore confident that an intercomparison between the chosen conditions is meaning full (bearing in mind the obvious variability of biological matter)

I believe that this revised version now meets ACP’s criteria as a measurement report, and hope it will be accepted.

Yours sincerely,

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Editor's comments

One of the major concerns remaining that this reviewer had is that you have not provided any context for how the emissions are changing over time from this single litter sample completely independent of your ozone and UV treatments. This reviewer strongly indicated that you surely have some "initial" emission data, which you could provide for each treatment trial just to give some sense of how that baseline emission value is changing.

> We are showing below by answering to reviewer 2, that our starting conditions were comparable for all type of experiments performed here due to the storage conditions of the leaf litter sample.

The main remaining issue for this reviewer is regarding the need for replicates. The lack of replicates makes it difficult to make meaningful conclusions about the observations. However, there are several reasons why the paper could be considered suitable for publication anyway.

> Thanks for these comments. We also noted that 2 out of 3 reviewers recommended publication as is. We show below that the experimental conditions were reproducible and for each condition, and despite the limited number of samples each experiment was repeated twice. This is not a perfect situation but it is as good as possible for such type of experiments.

Reviewer 2

The revised manuscript has addressed some of the concerns raised previously, but critical flaws remain in data interpretation and presentation. Consequently, I cannot recommend this paper for publication.

General Comments

Any description and discussion of the results needs to make it abundantly clear that differences in VOC emission behavior observed between the different experimental conditions cannot be directly compared because the same leaf litter sample was used throughout, and emissions will change over time as litter decomposes. I understand seasonal constraints on these types of experiments very well, but the interpretation of results needs to be provided within an appropriate context. These 3 different conditions can be presented as 3 entirely separate trials, highlighting that the same litter is being used throughout so the BVOC emissions were changing over time completely independent of the experimental condition imposed. The emissions at the beginning of each condition were likely very different even before the UV, O3, or UV_O3 was applied, so making comparisons about how O3, UV, or UV_O3 influence BVOC emission behavior on leaf litter that was 2 days old at the start of one condition, 9 days old at the start of another condition, and 15 days old at the start of another condition isn't meaningful (I'm guessing on the ages because that information was not provided clearly). The interpretation and discussion of results is still very focused on making comparisons between the different conditions instead of looking at each condition as a

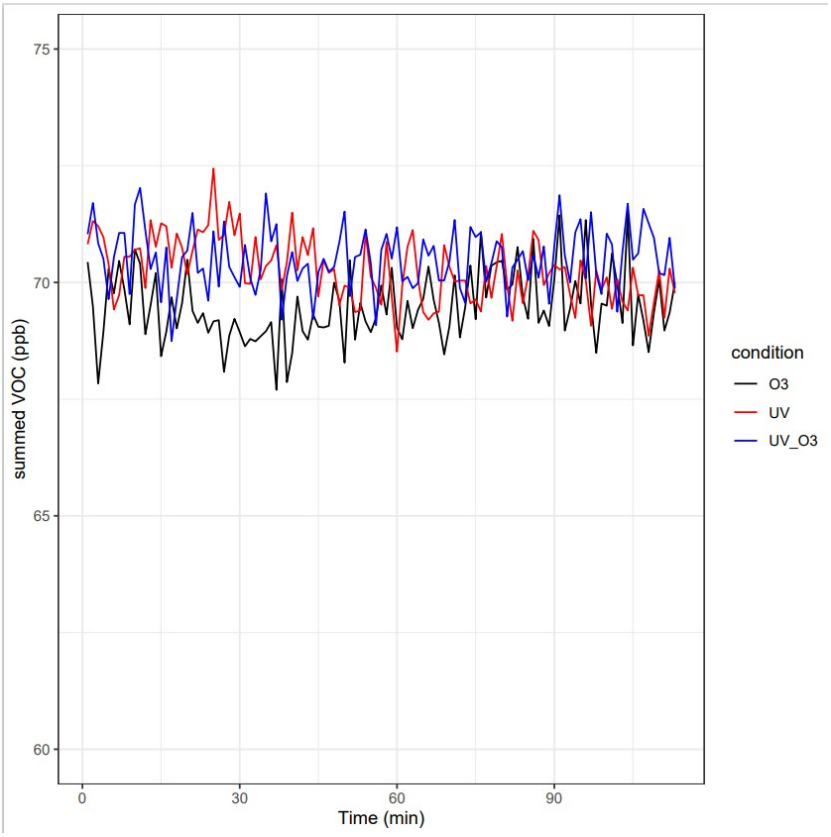
completely separate trial using litter with very different starting characteristics at the beginning of the experiment.

The main concern raised here by this reviewer is that the starting conditions of each of our experiments were different and therefore those cannot be intercompared. This is not correct.

To clarify this, we now added (and corrected) our manuscript with the following information:

To avoid inhomogeneous samples in terms of the decomposition stage, all of the leaves were cut directly from the stems but making sure that they were falling or about to fall. Overall, 3 kg of leaves were collected from different plants in the field (field area around 1 km2). The rapeseed litter used for the measurements was made of leaves at the beginning of senescence. The leaf samples were stored for at -20 °C. The sampled litter was reused for all the measurements, throughout the experimentation, defrosting just the fraction of sample needed for the experiment. At the beginning of each experiment, the leaves had visually the same aspect and identical mass to volume ratio (as an indirect metric of their decomposition). In addition, the VOCs were monitored during the stabilisation of the experimental conditions and showed identical patterns.

To demonstrate that intercomparison is indeed possible, we plotted below the total VOCs measured for 2 hours prior the start of each condition tested here (i.e., in the dark, prior to ozone injection, prior to switching on lights in presence of ozone). It can clearly be seen that those starting conditions are in the same range. We therefore assumed that the following evolution is due to the actual experimental plan.



PCA analysis: the authors state that the VOC emission profiles of the different conditions were “strongly different”. First, the time/decomposition component cannot be separated out from the experimental condition component. We need to see what the emissions looked like at the start of each trial BEFORE the experimental condition was imposed (essentially a comparison of the "initial emissions" for each condition). Second, PCA does not provide any measure of statistical significance and cannot be used to make any statements about the strength of difference between samples. You can use a PCA to show clustering, but statistical significance has to be measured with a different approach, such as an analysis of variance (ANOVA).

> We agree with the reviewer that that no statistical differences can be highlighted with the PCA. However, the PCA can be used to visualize possible differences between VOC emissions profiles. For this reason, we changed the sentence as follow:
>
> > “The PCA shows that the VOC profiles emitted during the UV condition were separated from the VOC profiles emitted from the UV_O_3 and O_3 conditions”
>
> We clarified, with the text above, the comparison of the initial emissions.

Figures 4, 5 and 7 are not publication quality at the standard generally seen in ACP.

> We now improved the quality of these figures by increasing the resolutions for figure and changing the layout of figure 7. For the PCA plot (figure 4), we believe that this standard presentation is adequate, but we nevertheless increased slightly its quality.

Now that it is clear the same leaf sample was used for each condition, there are other ambiguities in the methods section that need to be clarified. In Section 2.1 on “sample collection”, how many leaves/branches were collected? From how many different plants? It says a “random sampling method” was used, which made more sense before it was clear that the same set of leaves was used throughout the experiment. What does “random sampling method” mean in this context? Were a certain number of leaves collected from a random sampling of plants? Or were X number of leaves randomly sampled from the same plant? This is still unclear.

> To clarify this, we now added (and corrected) our manuscript with the following information (identical to above):
>
> > *To avoid inhomogeneous samples in terms of the decomposition stage, all of the leaves were cut directly from the stems but making sure that they were falling or about to fall. Overall, 3 kg of leaves were collected from different plants in the field (field area around 1 km2). The rapeseed litter used for the measurements was made of leaves at the beginning of senescence. The leaf samples were stored for 2 days at -20 °C for the first sample used, 7 days for the second sample used and 15 days for the third sample before measurement. The sampled litter was reused for all the measurements, throughout the experimentation, defrosting just the fraction of sample needed for the experiment. At the beginning of each experiment,*

the leaves had visually the same aspect and identical mass to volume ratio (as an indirect metric of their decomposition). In addition, the VOCs were monitored during the stabilisation of the experimental conditions and showed identical patterns.

Now that it is clear the same sample was used for each condition, Table 1 should indicate the dates for each condition. The leaves would have been decomposing over time (and their emissions changing as a result). It would be useful to have some sense for the "age" of the leaf litter during each condition.

As the initial conditions were comparable and no decomposition occurring due to the storage conditions, we do not believe that adding the dates of experiments is a useful information.

Section 2.4 Experimental Set-up should make is explicitly clear that the same leaf litter is being used for all three experimental conditions studied. In my opinion, there is still some ambiguity about that as written.

We now believe that the text above clearly answers also to this comment.