Dear Dr Benoit,

Thank you for posting additional responses to the reviewers (on the 26th of March). I also would like to thank you for performing additional experiments with varying HRMS instrument parameters. I suggest these results are to be added into Supplementary Information (SI).

With regards to your response that "*you observed that the change in acquisition mass range (50-750 to 50-450 m/z) had an effect only on the quantitative aspect of the results, without changing the set of chemical molecules identified."*

This is well expected, as the ion response depends on the RF amplitude applied to the c-trap (in Orbitrap). This effect strongly depends on the first mass of the mass range (e.g. *m/z* 50 in your case). The lower the starting mass, the lower the RF amplitude. With a low RF amplitude, higher masses are not trapped/transmitted so efficiently. As a rule of thumb, the c-trap can catch a mass range of "starting mass - (starting mass*15)". I expect this difference to be substantial when comparing *m/z* 50-750 vs *m/z* 150-750. So, I fail to understand the reason for comparing data from scan ranges of *m/z* 50-750 to *m/z* 50-450 and I am not surprised at all that your observed differences at those conditions were minor.

I either suggest performing additional experiments with applying the following scan ranges *m/z* 50-750 vs *m/z* 150-750 and adding your results to SI as a proof of concept or adding a statement in the text with a caveat for comparison of results from various publications. So that the reader, who is not familiar with HRMS technique, can understand that the molecular formulae that are only present in your study (or vice versa) could also be due to the differences in the MS acquisition parameters.

With regards to your following response:

"The HESI source was used only to compare the chemical formulas of different works. It is preferably adapted to a wide range of mass detection. Given the set of experimental differences you recalled, one might have expected several distinct sets of chemical formulas that are usually well visualized with Kendrick or van Krevelen type graphical tools. But, it is a continuity and an important similarity of data that we observed. The comparison is only qualitative and the range of masses studied 50-750 is relatively small. The differences observed in HESI are often due to a bad optimization of the experimental conditions. A bad pot ential difference, a too high injection quantity, a temperature not adapted to the studied elements, an initially not properly cleaned ionization chamber, a too high nitrogen flow,... Unfortunately, even with bad settings, the ionization can occur giving false results. We gave a lot of attention to all these parameters and compared our results to those obtained in APCI. Given our sensitivity, we did not find any qualitative difference. Concerning the existence of compounds through accretion reactions, it is difficult to exclude it definitively, but our observations did not allow us to highlight its presence."

It is not a matter of 'bad' or 'good' settings (as you mentioned in your responses), it is to what compounds your system was optimised to. I realise you are comparing molecular formulae, but those formulae are determined from ions which transmission is affected by the MS instrument settings. Again, I don't see any problem if you use the same system and the same HRMS settings to compare your numerous experiments; however, when comparing them with external data, such caveat needs to be stated in the paper as ACP is not a Mass Spectrometry specialised journal and such caveat is not apparent for those who do not use this technique.

I agree with your statement: "*We cannot exclude that some new compounds could be associated with unrealistic compounds compared to the atmosphere,….*" However, I also think this needs to be emphasised in your manuscript, especially considering the two reviewers' comments on the relevance of the presented work to the real atmospheric conditions.