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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Laser filament-induced aerosol formation" *by* H. Saathoff et al.

Anonymous Referee #1

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The authors present in their manuscript a description of a series of experiments in which laser filaments were introduced into an aerosol chamber, and the resulting particle formation was studied using several instruments. In the abstract, the authors introduce as their main findings

1. a quantification of particle formation rates in the plasma volume

2. the observation that particle formation increases exponentially with the concentration of water vapour

3. the increase of the particle yield by number for the addition of trace gases (SO2, a-pinene, toluene, NH3?)

4. the increase of particle mass with the addition of a-pinene

5. that particle formation is efficiently supported by acids produced photo-ionization

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of both major and minor components of the air (examples given are N2, NH3, SO2, organics)

In their conclusions, the authors additionally draw attention to the following findings:

6. oxygen addition reduces the particle production rate due to its high electron scavenging efficiency, which in turn reduces the plasma reactivity

7. nitrogen species do not cause significant increase in particle formation compared to Argon

8. growth and particle formation are dependent of different physio-chemical processes

9. dilution of the laser plasma affected particle production rates except in the case of ambient air

10. laser-generated particles “homogeneously nucleate water close to water saturation”

Regarding these points, I have the following comments:

1. The quantification of the particle formation rate is done assuming either a. All particles are formed in the plasma volume and transported to the measurement inlet without additional particle formation or losses on the way

b. Particle formation rates, as they are given now, should be understood as the overall formation rate in the vicinity of the ionized region, and can then be scaled by this ionized region.

Case a. seems rather unlikely to me; case b. is more likely, but as the formation rates were calculated from a single point in the chamber without mixing, quantitative reproduction of these experiments will be quite hard. This is not a problem inside this study (as the experiments were performed with fairly identical setups), but should maybe be mentioned for the benefit of anyone wishing to reproduce the results herein. Also, if I understood correctly, the particle formation rate was calculated by dividing

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the total increase of particle number by the total experiment time. This does not take into account the losses during the experiments, which at these very high rates could be quite substantial due to both self-coagulation and wall losses. The authors should at least indicate some of these uncertainties in the description of the derivation of the formation rates, if no estimate of the size of the uncertainty is given.

2. The observation of particle number (and mass in some cases) increasing with increasing water vapour concentration is very clear and a nice result. However, as it has become quite evident recently, small amounts of impurities can be the cause of particle formation in chambers; do the authors have an estimate whether the purified water could contain impurities such as amines in significant amounts (leading to vapour concentrations of the order of 106-108 #/cc)?

3. This seems clear, qualitatively

4. Ditto

5. This statement is quite vague. Was there direct proof of the acids supporting the nucleation, or is this more of a (well-justified) speculation? If not direct evidence of the acids exists, I would suggest that this is reworded to reflect that particle formation is supported by compounds produced in the plasma reaction products of the specified trace gases.

6. This is clear, again qualitatively

7. I agree with this, too

8. This statement is again quite vague (true, of course, but not really a conclusion). It could be better to state the relevance of the participation of some specific vapors to each process, which has been the topic in recent discussion of particle formation and growth.

9. This is actually something that I find very interesting. The actual mechanism that causes the particle formation rate dependence on the fan activity is not really clear

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to me. As particle formation is strongly non-linear, the dilution effect that the authors propose is easy to accept as one reason. As it is presented now, the fan effect is in my eyes a major reason that makes quantification of the particle formation uncertain. This could be highlighted more in the article. For future experiments, understanding the dilution effect, both in the particle phase and the gas phase, is a point that should be addressed with much care.

10. To my knowledge, homogeneous nucleation refers to nucleation directly from the gas phase, so the phrase 'particles homogeneously nucleate water' does not really make sense. If heterogeneously is used instead of homogeneously, the sentence would make sense and the statement would be ok.

Additionally, the discussion on atmospheric nucleation rates in comparison with the rates obtained in these experiments seems out of place for this manuscript. The experiments presented here are valuable for researchers trying to understand particle formation from the gas-phase species present in the atmosphere, and these types of experiments are a good tool for understanding the processes involved in particle formation. However, a direct comparison of the formation rates is not really useful in my opinion, first, because of the non-uniform formation process in the chamber (including the fan effects), and because the exact situation occurring in the experiments does not really occur in the atmosphere.

In general, very interesting paper with a lot of substance and many very interesting results, that are understandably quite qualitative. Clarifying the qualitative nature of the paper at the relevant sections should suffice for publication, as well as answering the points raised in reference to some of the conclusions. If these are done, I think that this study will be valuable to the atmospheric research community. In my opinion, the quality of the results here is very good, but due to the very complexity of the phenomenon that is being studied, the presentation of the results, especially the quantification, should be done with care. I do think that the revisions needed are not very big (I will suggest minor revisions), but they are important.

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I have a few specific comments, given below:

P 29856: l7: reults-results

P29861, line 2: what is meant by ‘larger particles seemed to be less stable’? Is there some observed break-up of particles, or does the concentration fluctuate? Clarify.

P 29867, l 20: I would reformulate this as ‘laser filaments generate new particles that grow to sizes of 3 to 130 nm during the experiment’, as this more exactly reflects the nature of the process

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Interactive comment on Atmos. Chem. Phys. Discuss., 12, 29851, 2012.

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