

Atmos. Chem. Phys. Discuss., author comment AC2 https://doi.org/10.5194/acp-2022-554-AC2, 2022 © Author(s) 2022. This work is distributed under the Creative Commons Attribution 4.0 License.

## **Response to Referee #2**

Spiro D. Jorga et al.

Author comment on "New particle formation and growth during summer in an urban environment: a dual chamber study" by Spiro D. Jorga et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2022-554-AC2, 2022

(1) This paper uses a dual chamber set up to investigate new particle formation in Greece. The dual chamber provides a unique way to study perturbations in order ensure results are actually the product of the perturbation. By introducing ammonia in only one of the chambers, the authors are able to conclude that ammonia is one component limiting new particle formation in this area. I think the writing is clear, the conclusions made are reasonable, and the limitations of the study are discussed appropriately. I think it fits well within the scope of ACP and would recommend it for publication subject to a few comments/revisions.

We appreciate the positive assessment of our work by the reviewer. Our answers (in regular font) follow each comment of the reviewer (in italics).

(2) This work could be improved by more clarity on what is novel here. Ammonia being a key component of new particle formation has been reported before so is the novel aspect of this paper the dual chamber experiments? The location being studied? The findings appear to be the same as that in Pikridas et al (2012). A follow-up study now 10 years later may be warranted, but I think whatever the motivation is, it needs to be made clearer.

The two major new advances of this work are first the use of a new experimental technique that allowed us to test the hypothesis of Pikridas et al. (2012) which was based on circumstantial evidence (the ratio of ammonium to sulfate in the particles and the wind trajectories) and second the results of the experiments that strongly support the hypothesis. An additional contribution is a technique to estimate the nucleation and growth rates from these data even without measurements of sub-10 nm particles We have followed the advice of the reviewer and we have made clearer the motivation of our work and stressed further the novel aspects of our work.

**(3)** How confident are you in the ammonia mixing ratios stated? Do you account for ammonia's significant wall losses? Do you expect desorption from the walls to impact later experiments?

The reported levels of ammonia in the chamber have been based on the volume of the chamber and the amount of liquid ammonia injected. It is possible that those levels could be lower due to losses of ammonia to the walls during the injection and the experiment. We tried to minimize those losses with the use of a heated line and the flushing of the line with additional air after the injection of ammonia. Desorption of ammonia from the walls was tested with blank experiments the following day from a perturbation experiment. The system was filled with ambient air, with no addition of ammonia and the system response was tested. In all the blank experiments we did not observe any nucleation in the perturbed chamber due to the previously addition of ammonia. In addition the flushing of the chambers at the beginning and at the end of each experiment minimized the potential desorption. We have added that information in the revised version of the manuscript acknowledging the uncertainty of the stated ammonia concentrations.

(4) I appreciate the authors' attempt to not draw any sweeping conclusions, but I think some broader context could be given. Since you are using a box model, what minimum ammonia concentration is necessary to evoke NPF under the conditions investigated here? How does that compare to ammonia concentrations in the study and any actually measured in the area? This does not need to become a modeling paper, simply a brief discussion of this may help ground this work more in atmospherically relevant conditions.

The used box model does not direct include the ammonia concentration or a nucleation parameterization but is used instead to provide estimates of the nucleation and growth rates that are consistent with the measurements. New particle formation occurred even at the lowest ammonia levels (20 ppb) used in these experiments. One of the reasons that we used relatively high ammonia levels in these experiments was to make sure that ammonia would not be the limiting reactant for nucleation. So the reviewer does make a good point for future experiments that could use much lower ammonia concentrations and determine the ammonia concentration thresholds. Right now we can conclude that this threshold is less than 20 ppb and also that the 20 ppb is much more than the average ambient ammonia levels in the area. This brief discussion has been added to the paper.

**(5)** Figure 1: Is the delay in the appearance of particles from when ammonia was introduced and the lights were turned on just due to not being able to measure the smallest particles? This seems to align with the average GR given.

The observed delay of the  $N_9$  particle number concentration, in the perturbed chamber is attributed to the time is needed for the particles to grow to larger sizes that were observed by our available instrumentation. Using this observation of particles we were able to calculate the average initial growth rate in our experiments which may be a little lower than the actual growth rate if there was a small delay in nucleation after the turning on of the UV lights. This point is now explained better in the paper.

(6) Lines 222-224: Does this refer to both particle wall losses and gas wall losses?

The correction refers to only particle wall losses occurring to the chamber walls and the sampling lines. We have revised this sentence to make this point clear.

(7) Line 239: Is this "initial growth rate" from the time you first see the particles at 9 nm

or extrapolated back to ~1 nm?

We now clarify that this is initial growth rate refers to the period between the start of the experiment and the time the particles appear at 9 nm size. It is actually the average growth rate during this initial period of the experiment.

(8) Line 278-280: The sentence beginning with "Unfortunately, we did not..." is confusing.

We have rewritten this sentence.

(9) Lines 308-310: The sentence starting with "Taking into account..." is also confusing.

We have rephrased this sentence.

**(10)** *Line 320: How good is this assumption? Is the condensation sink changing much during these experiments?* 

This is a necessary assumption given the available measurements. This constant rate is in practice an average rate for the estimated duration of the event. The change in condensation sink during the experiments was modest (reduction 10-30%) however other important parameters like the concentration of the species participating in the nucleation process were probably changing at the same time. We have some discussion of this point in the manuscript.

(11) Line 48: This sentence seems out of place here.

We have removed this sentence from the manuscript.

(12) Line 179: I think this is the only spelling of "sulphuric" like this.

We corrected the word to "sulfuric" to match the rest of the manuscript

(13) Line 221: I don't think this is a full sentence.

There was a typo in the sentence that has been corrected.

(14) Figure S1: Caption I think has "t=oh'' instead of "t=0 h''

Corrected to "t=0 h".

(15) Figure S3: There is a typo in the caption.

We have corrected the typo in the caption of the figure.