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Atmospheric Measurement Techniques Discussions

Interactive comment on "Regularities of new particle formation and evolution of existing atmospheric aerosol particles in a large (3200 m^3) isolated volume" *by* Nikolay Romanov et al.

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The authors express gratitude to the Reviewer for useful comments. Below are our responses to the Reviewer's comments:

1 Comment #1

1.1 Reviewer comment

15 nanometers can hardly be discussed as "new particle formation". A lot needed to happen that the particles could reach this size range. Also, it is clear that the particle formation, if there is such, is initiated already when filling the chamber, as atmospheric particle growth rates very seldom reach $15 - 30$ $nm/hour$, which seems to be needed to explain the current observations. Or is the whole observation just secondary organic aerosol growth on top of already present small particles? With this instrumentation we will simply not know.

1.2 Response

Our experiment was as follows. The Large Aerosol Chamber with air inside was plugged hermetically. You can see in Fig.3 particle number concentration of filled air was 10^4 particles per cm^3. After that, the air inside the Chamber was filtered so that no particles greater than 10 nm (technical characteristic of HEPA (HEPA, 2019)) remained inside the Chamber. After that, we began to measure the particle size spectrum inside the Chamber with the help of a particle size spectrometer mounted outside the Chamber. The particle number concentration of all detected particles was less than 50 particles per cm^3. We observed the increase of particles with sizes more than 15 nm 3 hour after the start of the measurements (particle number concentration increased to a maximum of 10^4 particles per cm^3). After that, we filtered the air once more so that no particles greater than 10 nm remained inside the Chamber and started measurements, but no increase of the particles' concentration was observed during 300 hours of probing. We concluded that observed new particles of 15 nm scale had originated from precursors due to the gas-to-particle conversion or had been condensed on sur-

faces of particles smaller than 10 nm (both had been contained in the air) before the second filtering, and we did not observe the increase after the second filtering because almost all precursors and particles smaller than 10 nm had been already involved into the process of gas-to-particle transformations and had been swept out as condensation centers of new particles by the second filtering. Such low concentration (almost zero), reached after the second filtration, was observed in many experiments. This low concentration value remains almost constant for more than ten days.

2 Comment #2

2.1 Reviewer comment

It seems strange to have instruments inside the chamber facility if you are attempting to measure such a fragile process as gas-phase nucleation. I do recognize that this might have been needed for the cloud formation experiments which I don't possess expertise, but to study new particle formation (=nucleation) in a chamber with instruments inside seems strange indeed. For me it is very difficult to see how these machines inside the chamber would not influence the particle formation experiments. I have to ask: In what other chambers studying nucleation of nanoparticles there are instrumentation inside the chamber? Again, nucleation is a very fragile process indeed. To the same background influence, one has to ask what does the "ship paint" contain that has been used to paint the inner walls? Is this a possible source of nucleation precursor chemicals to the chamber? Commonly investigations go through a lot of trouble in ensuring the walls behave inert. Yet, I definitely agree on what is said about Teflon not being an optimal choice. I do not know what this "ship paint" is composed of, but it seems like a rather questionable surface for conducting NPF studies -which are notoriously influenced by even very minor concentrations of certain chemicals. According to Figure 1, the chamber is literally full of potential particle sources, and I'm not sure if you could ever decide

with the current instrumentation if there's new particle formation due to them or not.

2.2 Response

The instruments inside the Chamber were turned out and were not enabled in our experiment. After the Chamber had been purified with the help of an internal HEPA filter (in Fig.3 the time interval $t_2 - t_3$), the concentration of aerosol particles decreased by 3 orders of magnitude and remained low (about $30 - 40$ particles per cm^3) during the next 300 hours. This testifies against the chamber walls (including its "ship paint") and inactive instrumentation mounted inside the chamber as potential sources of precursors or condensation centers.

The measurements provided in the one point of LAC, far from any equipment and wall. As shown in Fig.2, air velocity enough small to influence the experiments. The huge volume of LAC allows to research particle evolution in almost natural conditions.

3 Comment #3

3.1 Reviewer comment

I don't see why this paper claims to report new particle formation, as the measurement had no techniques capable of extracting this information. Yet, the study of aerosol dynamics in forming the larger particles seems certainly worthy of reporting! However, that steps out of my key knowledge, and some better qualified person should judge on the novelty of the results. So, the last line of the current abstract "These results may be used for the development of aerosol evolution models" is to my mind, absolutely to the point.

3.2 Response

In our paper, we describe the experimental study of the evolution of aerosol particles in the presence of a source of condensable products and propose an analytical description of asymptotic coagulation spectra of aerosol particles. To us, the obtained experimental data provide evidence that gas-to-particle conversion processes take place in the Large Aerosol Chamber. Certainly, we cannot specify a mechanism of new particle formation in the Chamber without the gas composition (ozone, nitrogen oxides, hydrocarbons, etc.) analysis, which becomes the subject of our forthcoming publications.

The article presents the evolution of ultrafine particles with a size of more than 15 nm, including NPF, under almost natural conditions, to the size of cloud condensation nuclei, which is very important for assessing the effect of submicron particles on meteorological processes in the atmosphere.

4 Comment #4

4.1 Reviewer comment

Major formatting concerns: Currently the level of presentation is a bit awkward, and the manuscript in parts reads more of a historical documentary, or a thesis, than a scientific article. There are too much description of previous works and platforms, which are not really needed to describe the current facility or the obtained results. For a historical description a review article would be more suitable format, yet then also the journal would probably have to change. To my mind AMT is reserved for introducing and describing new methodologies and instrumentation, and as such, this facility would fit the description. Obviously, also some background is needed from the previous work to understand the context, yet this should be done briefly enough to not confuse the

main topic of the work. The language also has to be improved substantially before this draft is really ready for submission. Ideally a native English speaker should edit the text first.

4.2 Response

The referee drew attention to the increased volume of chamber descriptions. This section has been expanded on the advice of an APC referee. The main argument is that the materials of Russian studies are poorly represented in the English-language literature and are not known to a wide range of specialists. In particular, the search for literary sources that we referred to for the description of the equipment used caused difficulties for the reviewers. Undoubtedly, AMT has extensive experience in analyzing the informative attractiveness of article materials. And, if the editorial board finds the shortest material, we will revise the article, considering all the recommendations.

5 Comment #5

5.1 Reviewer comment

"conversion gases" seems uncommon. Maybe the wording you are looking for is "trace gases"?

5.2 Response

The term "conversion gases" denotes the gas components that participate in gas-to-particle conversion; not all "trace gases" do participate in the process.

6 Comment #6

6.1 Reviewer comment

What is "skewness"?

6.2 Response

Skewness refers to asymmetry of statistical distribution. If the curve is shifted to the left or the right, it is skewed. Skewness can be quantified as a representation of the extent to which a given distribution varies from a normal (statistically symmetric) distribution. A normal distribution has zero skewness while a lognormal distribution, for example, exhibits some degree of right-skew. See, for example, (Teegavarapu, 2019).

7 Comment #7

7.1 Reviewer comment

The large volume of the chamber allows eliminating wall influence on particle sedimentation." To my mind "sedimentation" specifically relates to gravity, and is thus different from wall deposition

7.2 Response

Of course, it is a typo. The sentence should be: Large chamber volume allows for avoiding significant particle wall deposition and floor sedimentation.

8 Comment #8

8.1 Reviewer comment

This allows concluding that aerosol particles greater than 200 nm *have a life-time of more than five days, while particles smaller than* 15 nm*, not more than five hours. "While I understand the comment on* 200 nm *particles, I did not understand how did you come up with* 5-hour *lifetime for smaller than* 15 nm *particulates if they were never measured?*

8.2 Response

Life-time for 200 nm particles was evaluated from experimental data to be more than 5 days, this estimate is close to that reported by various authors, for example (Kreidenweis, 1999). The Figure 8 (Fig. 8a Fig. 8d) shows that during the time interval between the two first measurements of particle size spectra (which is 5 hours in average) the concentration of 15 nm particles decreases by an order of magnitude.

References

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