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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.

Anonymous Referee #1

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Review of AMT submission "amt-2020-172" by Romanov et al. titled "Regularities of new particle formation and evolution of existing atmospheric aerosol particles in a large (3200 m3) isolated volume"

Significance: The study introduces a large environmental chamber facility "Large Aerosol Chamber (LAC) RPA ("Typhoon")" located in Obninsk, Russia. It has been previously used for cloud physics investigations, and now, is considered to be used for studies of atmospheric new particle formation. In this publication several observations of particle dynamics inside the LAC are described, presumably due to the want to include this very large chamber setup into the international collection of atmospheric

simulation chambers serving the atmospheric community. I certainly recognize the need for various different platforms for studying atmospheric transformations, and generally, the larger the facility, the rarer it is. This chamber is unique already for being so massive, and thus could perhaps supply key insight into atmospheric particulate matter formation, permitting to study the nucleation processes with minimal influence from prevailing surfaces. However, there are several practical issues that should be resolved before I could advocate its utilization within the community. Moreover, the current level of presentation is not in line with the common norm of AMT articles, and thus I suggest major revisions to the structure and context, before I can propose this article to be accepted for publication in AMT.

Major scientific concerns: 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.

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. In connecting this to your presented results, you imply a 15 nm/hour growth rate in a very large chamber without background particles and without oxidants? (Well of course there are

oxidants such as OH and Criegee radicals, as the chemistry is initiated by ozonolysis reactions – yet none of this is discussed in the paper, which is another problem). This is a very large growth rate and needs a large fraction of condensing species present, and a steady source of small seed particles. I guess what I’m really trying to say here is: 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.

To sum it up: this is an interesting and rather unique facility for studying atmospheric aerosol dynamics at sizes above 15 nm, with the current instrumentation and description. I don’t see the ability to study new particle formation without significant further studies and serious updating of the major instrumentation.

Specific suggestion: I strongly recommend trying to measure the aerosol precursor pool of the chamber – what are the main gaseous molecules inside of the chamber after filling? Especially what are the SO2 and O3 concentrations in your experiments – as these gases are most likely needed to induce the first steps of the particle formation

in a dark chamber. For hydrocarbons and their oxidation products, a proton transfer reaction mass spectrometer (PTRMS) and gas chromatograph (GC) would suffice to get an idea about the constituents oxidizing and ultimately forming particulates. For the actual in-situ aerosol precursors you would need to have an atmospheric interface equipped chemical ionization MS applying some of the clustering reagent ions (e.g., NO3- and I-). Like it’s said in the abstract “The mass concentration of newly formed particles depends on the concentration of precursors.” Absolutely! And thus, it would be important to know about them to make statements about new particle formation vs secondary mass growing on top of small existing particulates.

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. I am not one and thus I cannot perform this task. Currently, I am not 100% sure I understand the text exactly as it was supposed to, and I presume my non-nativity was part of the reason why I could understand the text quite well already.

Few specific comments: I did not dwell on the minor problems within the text as I found the text has to be revised significantly before it can be considered for publication in AMT. However, I have picked here just a few issues which could be fixed already at this stage. Somewhat uncommon terminology is used throughout the text. Some examples: *“conversion gases” seems uncommon. Maybe the wording you are looking

for is “trace gases”? *What is “skewness”? *What is the meaning of the sentence: “The value ras $\approx$ 2 defines the analytical description of the size distribution as the gamma distribution.”? *“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? *It wasn’t clear to me where this came up from: “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?

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