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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Impact of Isolated Atmospheric Aging processes on the Cloud Condensation Nuclei-activation of Soot Particles" *by* Franz Friebel et al.

Anonymous Referee #1

Received and published: 26 August 2019

General comments

This paper discusses the aging of soot particles by ozone oxidation in an environmental chamber. Aging times of up to 12 hours were achieved, and the data analysis follows recently published work by the same author to interpret results from the continuous-flow stirred tank reactor chamber. Speaking of that prior work (Friebel and Mensah, 2019), it seems that this paper uses the exact same dataset. The findings are consistent with prior published results on CCN activation of soot particles; namely that soot with a higher fraction of organic carbon (OC) activates at a shorter oxidation time than soot with less OC. Activation times are strongly dependent on temperature, but not on

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Discussion paper

relative humidity (up to 75%).

With regards to the data analysis, it seems there is alot of repeat material from the previously published work and this paper, including an entire figure. Is this really necessary? I realize that some overview of the method is a good thing, especially with a method so new and not really used before; but even with the extended discussion in the current paper I feel like I need to read the other publication to fully grasp what is happening. So, I recommend shortening and/or removing some of the repeat information and improving the description of activation time.

This paper also applies the experimental results to a global aerosol-climate model (ECHAM6.3-HAM2.3) by altering parameters that determine the aerosol number concentration of CCN-active particles. In all cases, the modeled CCN and CDNC increase, with varying amounts of increase in different locations around the Earth. The increase makes sense, because it seems like the current model does not consider soot aerosol to be CCN-active at all; thus, any parameterization which does make these particles CCN-active should increase the overall number. The spatial pattern of CCN and CDNC increases is explained as larger increases where soot particle loading is high and/or pre-existing CCN concentrations are low, such that competition for water vapor is minimized.

With regards to the modeling results, my remaining question is what is the big-picture conclusion? It seems two activation times were used in the parameters, reflecting the results from two kinds of experiments in the CSTR, but which result is likely closer to truth? What does a 30-100% increase in CDNC mean for long-term climate projections?

Somewhat related to my modeling questions above, one key improvement that needs to be made to the manuscript is relating miniCAST soot to ambient soot. Because you emphasize the global model results at the end of the paper, the question becomes how well does your experimental protocol mimic what would happen in the atmosphere?

One key element to that is how well does your soot match what is found in the atmosphere? Simply stating that the miniCAST has been used for many studies is not enough. Consider the following references, in addition to providing more details from the papers you already cite:

Le, K., Pino, T., Pham, V., Henriksson, J., Török, S., Bengtsson, P. (2019). Raman spectroscopy of mini-CAST soot with various fractions of organic compounds: Structural characterization during heating treatment from 25°C to 1000°C Combustion and Flame 209(), 291-302. https://dx.doi.org/10.1016/j.combustflame.2019.07.037

Moore, R., Ziemba, L., Dutcher, D., Beyersdorf, A., Chan, K., Crumeyrolle, S., Raymond, T., Thornhill, K., Winstead, E., Anderson, B. (2014). Mapping the Operation of the Miniature Combustion Aerosol Standard (Mini-CAST) Soot Generator Aerosol Science and Technology 48(5), 467 479. https://dx.doi.org/10.1080/02786826.2014.890694

Marhaba, I., Ferry, D., Laffon, C., Regier, T., Ouf, F., Parent, P. (2019). Aircraft and MiniCAST soot at the nanoscale Combustion and Flame 204, 278-289. https://dx.doi.org/10.1016/j.combustflame.2019.03.018

Ess, M., Vasilatou, K. (2018). Characterization of a new miniCAST with diffusion flame and premixed flame options: Generation of particles with high EC content in the size range 30 nm to 200 nm Aerosol Science and Technology 53(1), 1-44. https://dx.doi.org/10.1080/02786826.2018.1536818

Overall, the paper needs some more polish before it is ready for publication.

Specific comments

Do not duplicate information from the figure captions to the body of the paper. You only need to give descriptions of the figure colors, etc. once. In the body of the paper, focus on presenting what the figures tell us (not how they tell us).

There is also some duplication with regards to the experimental differences between 2016 and 2018. Polish the writing a bit.

Section 5 - How accurate is your SMPS for diameter? Is 3 nm really significant and measurable? I don't believe that it is because at the end of Section 2.1, you mention that the mode diameter of CBW particles is 90 nm after a pre-mix chamber even though you are size-selecting them to be 100 nm. How do they get smaller? I believe you also said that denuding the particles made no difference.

The experiments which vary by a factor of 2 when all else is supposed to be equal seems very interesting and concerning to me. What else could be different besides the temperature? What was the total particle concentration for these two experiments? Only one experiment is shown in Fig 2; would be nice to see the rest in a Supplemental Section. Is there any basis in the literature for just 4 degrees C to cause that big of a change in CCN number concentration?

Figure 5 - is "statistical none-significance" a common term? I am not familiar.

Are the terms "homogeneous ozone oxidation" and "heterogeneous ozone oxidation" commonly used? I've not seen it said this way before. I'm familiar with homogenous and heterogenous nucleation, and I can guess at what these terms mean; but I wonder if there is a better or more precise way of saying these concepts or not.

Section 6.2 - Better describe what it means to be "statistically significant" in your modeling.

Table 3 is not referenced at all in the text.

Figure S1 isn't really discussed.

Technical corrections

There are many cases where "since" was used but "because" is the proper word to use.

Do no use “this” or “it” as a noun in a sentence; too vague.

In many places, “particle and VOC free” should be “particle- and VOC-free” for clarity.

Do not start a sentence with a number, e.g. “65 % of ...”

The date of each experiment is irrelevant for anyone not a part of the study; consider naming the experiments A, B, C, etc.

pg 3 line 16 - “The similar results” needs to be reworded

pg 6 line 10 - missing a space in “humiditywere”

pg 11 line 22 - should be “represents”

pg 12 line 12 - missing space in “Table1”; “lower” should be “shorter”

pg 13 line 7 - parenthesis are messed up

pg 14 line 1 - missing comma after “Figure 4”

pg 14 line 4 - missing comma after “case”

pg 14 line 20 - What does “very” theoretical approach mean?

pg 15 line 7 - missing a period

pg 17 lines 27, 30 - some extra commas and periods here

pg 21 line 12 - “less” could be “faster”

pg 21 line 16 - no comma after “Both”

pg 21 line 29 - When is “shortly”?

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