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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Technical Note: Formation of airborne ice crystals in a wall independent reactor (WIR) under atmospheric conditions" *by* E. Fries et al.

Anonymous Referee #2

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This paper presents a new instrument to nucleate and grow ice crystals in the laboratory under atmospheric conditions. It aims at experiments to study the uptake of organics during nucleation and growth in an environment which is supersaturated with respect to ice. The paper describes the setup and presents some experimental and calculated data to characterize the instrument for later applications as described above. The technique described here is is principle interesting and worth publishing but there is important data missing. I therefore recommend this paper for publication after major revisions have been made. My main concerns relate to the way in which the relative humidity profile is calculated and the conclusions made thereafter about possible ice nucleation and growth in the chamber which seems to be the main purpose of the

experiment.

General remarks of technical or scientific nature:

Introduction

first paragraph, especially lines 10-15: I guess what you are trying to emphasize here is the fact, that most previous works have been carried out under equilibrium conditions and not for growing ice particles. I would try to be more specific here so that this fact is highlighted better and clearer to understand.

p. 13020 line 6: could you specify what kind of wall effects you mean? I guess adsorption of organics/hydrocarbons? line 16: I am not sure if it is obvious to the reader what you mean by gas and nucleation scavenging and what the difference here is and what kind of consequences this has on the interpretation of experimental results. Do you really mean scavenging of gases during the nucleation or more during growth which would be something different.

Experimental setup:

lines 21/22: One could ask why the chamber was setup in the cold chamber at all if the effect is unwanted?

p. 13021, lines 1-4: If only the effectiveness of removing particles is of importance or has been measured here I would only speak of a filter and filter efficiency. I guess the system is later intended to also remove organics from the flow before a controlled amount of hydrocarbons is added to the flow for the scavenging experiments. However this is nowhere mentioned and as the setup is described now and depicted in figure 1a this removal devices does not make a lot of sense to me, except for the dryer and filter. Did you also measure how well organics were removed?

This paper has one weak point which is the humidity profile within the chamber. To begin with, a saturation ratio or a supersaturation normally refers to either ice OR water. It should therefore be clearly mentioned (e.g. by indexes) to which phase you are

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referring to. Without this knowledge the whole discussion about possible ice nucleation and growth is inconclusive to the reader. Calculations of relative humidity profiles: It is not clear to me, which temperatures were used to calculate the saturation ratios reported in table 1. At least the saturation vapor pressure C_sat should change by more than a factor of two if one goes from -14°C to -6°C along the center of the reactor and this does not seem to be the case for the calculations summarized in table 1. Two more columns with the temperatures at the center and wall being used in the calculations would be helpful (and maybe two more for the radius and velocity of the jet, see below). If certain temperatures (how many?) from figure 4 have been averaged it should be mentioned and discussed under which assumptions this has been done. The data in table 1 predicts where ice crystals may form and how quickly they grow, so the temperatures being used here should only represent the part of the airflow in which the aerosol particles from the inlet air stream are located. In other words, table 1 should represent the conditions along the trajectories of particles in the inlet air stream, respective temperatures should be used therefore. If the radius of the jet is assumed to be rather big, the variance of temperature and relative humidity and therefore the homogeneity within the jet should also be discussed. The choice of these temperatures (and therefore the radius and the velocity of the jet) is crucial and should be discussed. Furthermore, a measurement of relative humidity at a few points would be very helpful to support these calculations. A supersaturation wrt. ice is required to nucleate ice on the dust particles supplied by the aerosol generator. By looking at the data in table one, one can see that this is the case only for the second half of the reactor (if S refers to ice). If S does not refer to ice but rather to water, one definitely needs a reference temperature to obtain the ice supersaturation. S_ice is the relevant value for ice nucleation. Furthermore, the temperature in the jet stream has climbed to around -10°C at this point. Mineral dust is very likely to nucleate ice only at high supersaturations at this temperature and possibly only a small fraction of all particles. If you expect (or need) a significant fraction of the dust particles to nucleate ice (in the deposition freezing mode) you need temperatures below -25°C at least. At warmer temperatures

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there is very likely only immersion freezing being active above water saturation. But you then have to expect water droplets to form in parallel and these are very likely outnumbering the ice crystals formed. Depending on the true conditions there might also be a large fraction of unactivated particles present in the reactor. I am not sure if these droplets or unactivated dust particles and their surfaces will interfere with any experiments that are planned later to study the scavenging of hydrocarbons on ice. The different modes of heterogeneous ice nucleation should be discussed in the context of the conditions produced in the chamber. Conclusions should include the likelihood of ice and droplet formation and the consequences for the planned experiments. Consequently, the value Q_m,i can only represent a potential ice growth rate and only there where ice crystals are present and supersaturated conditions are found. But even then, the effective growth rate is also dependent on the ice particle number concentrations. If only a few ice crystals are present, the diffusion of water vapor to their surfaces limits the mass growth rate in the chamber. If the number concentrations are large, then a depletion of water vapor and therefore also a decrease in supersaturation will be the consequence. Are the authors aware of this fact and the underlying theory (capacitor analogue theory of ice growth)?

If CFD software like FLUENT is available, a simulation of the chamber to estimate relative humidities along particle trajectories would be very useful. Instead of figure 3 which does not include a lot more information than is already presented in figure 4 I suggest to show the results of the humidity calculations (once the issues mentioned above are resolved) in an extra graph. They are crucial for the formation and growth of ice. The same arguments are true for the residence time estimated on page 13026, line 19 (which assumptions are made regarding the jet diameter and the corresponding velocity). Do you expect turbulent mixing at the entrance into the chamber. The sharp temperature increase from -20 to -15°C within a few centimeters suggests that. In addition, the residence time cited from DeMott et al. 2003 refers to steady state conditions in contrast to your experiment where temperature and relative humidity increase. Furthermore, this time is NOT the time needed to form ice crystals but the time needed to

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grow them to large enough sizes so that they can be distinguished from aerosols in the optical particle counter of the CFDC instrument. As a last point here: If the chamber is up and running, including the particle generator: Have you carried out proof of concept experiments to see if you can produce ice crystals the way you expect them to form? It seems that you have everything set up to do so. Do you see any ice crystals at the end of the chamber e.g. with the particle spectrometer you used to measure the size of the particles produces with the particle generator?

line 27: I am not sure what you mean by caloric gain here. The fact that moist air has a greater heat capacity than dry air? Could it be that the upper two lines in figure 4 represent mainly the trajectories of the inlet air stream and the others a forced convective cell driven by warm temperatures at the wall and a cold temperature of the jet stream plus the frictional force of the jet stream velocity against the surrounding air within the reactor? Is this additional force being accounted for in the calculations? Since all temperatures in figure 4 merge together with increasing reactor depth this could be simply heat exchange between the jet stream and the convective circulation. The jet stream very likely widens when it enters the chamber and narrows at the exit. Again, trajectories of the jet stream air in figures 2 and 4 would help in the interpretation of the data.

Typos and writing style:

Abstract

line 19: ...were kept ..... ?' one of these two words is too much. Recc: ....stayed well below... line 21: ...was calculated to BE 40s. line 22: delete "also" lines: 24-27: This paragraph can only be understood after reading the full paper. I recommend to leave it out or be more specific. The purpose of the removal device is not explained. BTW: You only talk about of removal efficiency regarding aerosols so far, so just "filter efficiency" would work as well and causes less confusion.

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p. 13019 line 9: remove "s" after hydrocarbons line 14: "Therefore" suggest a conclusion of the previous sentence which does not seem to be the case.

Experimental setup: p. 13021, line 14: Please start the sentence: The air stream..... with words connecting it to the sentence before so it makes more sense: Like: By this, ...

line 15 (and somewhat more often in the text): You now call the wetted wall a "physical wall" instead of virtual wall before. I leave it up to you but my feeling is that this is just a "wetted wall" and nothing more. Each wall is physical somehow so I do not understand why this is highlighted here.

p. 13022, line 10: Within a .... first series of experiments.....

p. 13023 lines 11-18. This is a very long sentence and hard to follow. Could you break it up into smaller one perhaps?

p. 13024 lines 14 + 20 (and many more times in the manuscript and also figure captions): If you write ... in A different reactor depth you are referring to ONE specific other position. I guess you want to say ...as a function of ... or .... AT different reactor depthS .....

p. 13025 line 9: replace ambient by sample air? line 22: ...influencing....

p. 13026 line 22: either THIS experimental condition or THESE experimental conditionS

p. 13027 line 1: did you mean SUPERsaturated air masses? Lines 9+21: see comment for p. 13024 lines 14 + 20.

p. 13028 lines 19/20: You are contradicting yourself here. In some other places (e.g. 13026, lines 18/19) you correctly state that supersaturated conditions are needed to form ice. Here you speak of ice formation in section h1 where you have only subsaturated conditions. No ice will form or even grow here! If it was there it will even

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evaporate! Taking into account these arguments, does the estimate of 40s residence time where ice crystals can grow still hold?

13030 line 16: ...particle generation is (?) on (?) needed.

13031 lines 6-9: If would be nice if the authors could briefly mention somewhere, how these scavenging experiments will be performed (e.g. with which additional equipment attached to the setup). Otherwise one has no clear idea of how the described setup is planned to be used for the proposed scavenging experiments. The reader has therefore no possibility to judge or confirm if the described experiment is suitable for these experiments or not.

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 13017, 2008.