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Atmospheric Measurement Techniques Discussions

Interactive comment on "Humidity effects on the detection of soluble and insoluble nanoparticles in butanol operated condensation particle counters" *by* Christian Tauber et al.

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We appreciate the thoughtful comments by referee 2. For discussion purposes we would like to respond to the general points raised.

***Summary:** The paper demonstrates a humidity dependence on NaCl activation using a butanol CPC, where increasing humidity decreases the activation cut-size, while showing no such dependence with Ag particles. The measurement was performed with both continuous flow CPCs and the SANC. Due to the strong humidity-dependence of some particles, it is believed that ambient measurements could be activated at smaller sizes than their laboratory-controlled equiv-*

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alent and care should be taken in assuming constant cut-sizes for butanol-based CPCs in ambient measurements of unknown composition.

These results seem significant and interesting, but I think it could be reorganized to better integrate, introduce, and motivate each experiment's contribution to the objective. Some experiments are not fully motivated of their potential significance until the results section.

The abstract/intro is missing a significant section of the paper regarding charge effect. Poor humidity sensor selection for a paper on targeting humidity effects. Sensor uncertainties need to propagate through to resulting figures (e.g. Figure 4).

A paragraph in the introduction for the charge effect will be added in the updated manuscript. The shrinkage measurements in Figure 4 were conducted with SHT75 sensors, with an accuracy of +/- 1.8% and with a HMI38 Humidity Data Processor with a HMP35E probe, with an accuracy of +/- 2.0%. For the sake of readability uncertainties were not included in Figure 4 in the final version of the manuscript. The corresponding uncertainties are discussed in the figure caption instead.

Would a working fluid such as Fluorinert counteract the humidity dependence of NaCl activation, or would the NaCl still uptake water and nucleate more easily with humidity?Practical solutions and guidelines for scientists would be helpful.

These compounds consist of carbon chains with fluorine atoms instead of hydrogen atoms. These molecules have a low dipole moment. Due to the spherical structure and very electronegative atoms on the outside the overall surface of the molecule is slightly negatively polarized. As a result, a shift in the cut-off diameter can be expected by using a mixture of Perfluorcarbonates and water. The overall polarity of the working fluid would almost exclusively be based on the dipole moment of water molecules. In fact, the interaction between the NaCl seeds and the water molecules will be persistent if only the working fluid gets changed. To remove the influence the carrier gas should

be kept dry. In case this cannot be done properly, a possible influence of the relative humidity should be kept in mind, especially at measurement sites located near the sea.

Major Comments: The citations provided, on nucleation for example (P2 L5), should credit the work from previous authors. Also, heterogeneous nucleation mechanisms needs to include more sources than Wang et al. 2013. In general, the sources need to be expanded to include the major pioneers of the subjects. If the experiments were already performed with propanol in Schobesberger et al., why would a different behavior with butanol be expected? The difference between the papers, aside from changing the working fluid, should be highlighted.

We acknowledge the well-justified comment on the citations of heterogeneous nucleation mechanism and will include them in the updated manuscript. We were not expecting a different behavior. Initially the temperature dependence found by Schobesberger et al. (2010) was used to explain the enhanced detection efficiencies for NaCl found during the laboratory measurements. During the measurements it was found that the relative humidity dependence was stronger than the temperature dependence. Schobesberger et al. (2010) focused on the temperature dependence and compared it to different theoretical approaches like Fletcher theory. In this work we focused on the deviating activation behavior of butanol-based CPCs. The deviating activation behavior is caused by the temperature settings and by increased RH levels.

Figure 1 does not include control system mechanism for RH. The paper focuses on humidity as a primary variable, but not much detail was provided on how it was varied and controlled. Is RH controlled with a feedback loop to account for transients through-out the experiment? If RH is the main variable of interest, its introduction to the system and control should not be glossed over.

Figure 1 will be moved into the supplemental materials. In the revised manuscript it will be replaced by the figure shown below. The carrier gas was humidified by passing it through a diffusion type humidifier. The relative humidity of the carrier gas was

monitored and recorded throughout every measurement. The deviation of the recorded values was always below +/- 1% which is below the sensor accuracy.

Honeywell sensors mentioned are crude (accuracy +/- 3.5%, hysteresis 3%, repeatability +/- 0.5%, etc.). These uncertainties need to be reflected in the figures.

The humidity measurements were conducted with two different humidity sensors. The cut-off diameter measurements were conducted with the Honeywell sensors. Due to the accuracy of +/- 3.5% the minimal RH difference between two measurements was 10%. The shrinkage measurements were conducted with SHT75 sensors with an accuracy of +/- 1.8%, hysteresis 1%, repeatability +/- 0.1%. In the final version of the manuscript the error bars were not included in Figure 4 for improved readability. Uncertainties will be discussed in the figure caption.

Regarding the counting efficiency experiment (P4), each TSI CPC can have a unique counting efficiency based on laser age, optics cleanliness, alignment, etc. Characterizing the effect of CPC deltaT on nucleation effectiveness using 3 different CPCs does not seem substantial enough, as each CPC could have a different counting efficiency at the same temperature. It would have been preferable to compare one CPC and repeat the experiment 3 times, so the additional factors mentioned above are constrained. Otherwise, it would need to be stated that the 3 CPCs have been normalized to one another for each of the deltaTs (or proven operation is identical).

In total about 400 detection efficiency curves were recorded. To rule out any dependence on the individual activation behavior of different CPCs, measurements involving one single CPC set to all three temperature settings one after another, and measurements involving three CPCs that were all set to one specific setting, have been performed. Based on these measurements the maximum error linked to the individual activation behavior was calculated to be at the highest at 8.1% (P. J. Wlasits, 2019). Figure 2 and 3 are chosen as exemplary measurements for different temperature set-

tings and CPCs at the same T settings (C. Tauber et al., 2019).

The charge effect isn't addressed in the abstract or introduction/background and seems significant to the NaCl activation. The inclusion of this experiment is good, but it feels like it came only at the result of Section 3.1, and didn't have its own proper introduction to motivate why it's included.

We thank the reviewer for making us aware of the leak in the introduction section. In the final version of the manuscript we will add an additional paragraph on the charge dependence to the introduction.

Minor Comments: P1 L14-22: This paragraph seems irrelevant to the focus of the paper goals and losing it would not detract from the message. The overview seems a little vague, when it would benefit with a background more targeted to the objective.

We will remove this paragraph and add an introduction paragraph on the charge dependence / effect.

Commas must be added in sentences using passive voice. This is done with some, but not all sentences and must be corrected throughout.

We will review the manuscript to correct that.

Simplify wordy sentences throughout: e.g. P2 L4: "This process is called nucleation and arises..." could simplify to "This process of nucleation arises..."; and P2 L18.

This change will be included in the updated manuscript.

P2 L7: Remove "a" to make CCN plural to agree with your verb and that you used "nuclei" instead of "nucleus", i.e. "...nanometer-sized particles contribute as cloud condensation nuclei (CCN)."

This change will be included in the updated manuscript.

P2 L15-17: You describe deliquescence and efflorescence without using the term for the mechanism.

This will be explained in the updated manuscript.

P2 L 20: Subject-verb agreement; "One technique...is CPCs", not "are".

This change will be included in the updated manuscript.

Interactive comment on Atmos. Meas. Tech. Discuss., doi:10.5194/amt-2019-23, 2019.

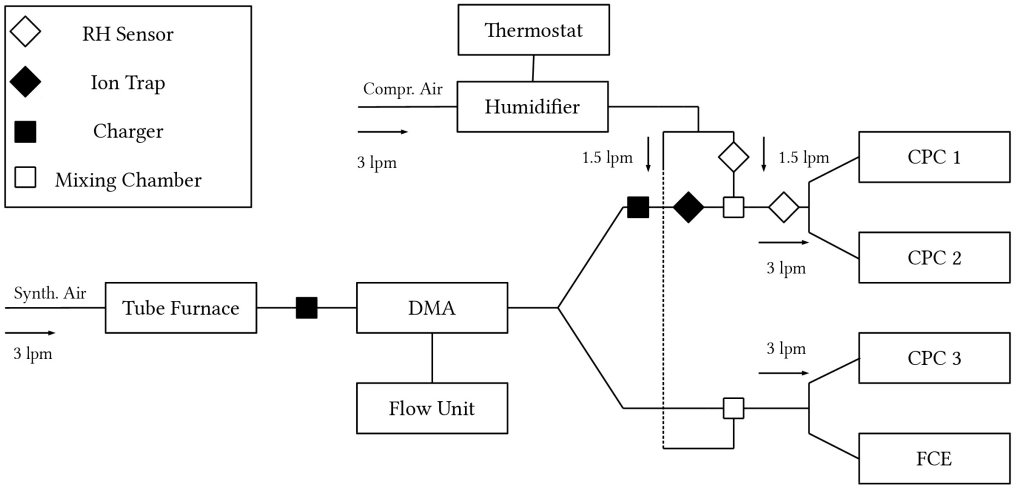

Fig. 1. The experimental setup for evaluating the relative humidity dependent counting efficiency of continuous flow type CPCs (TSI 3776 UCPC), which was measured by operating a Faraday Cup Electrometer (FCE)

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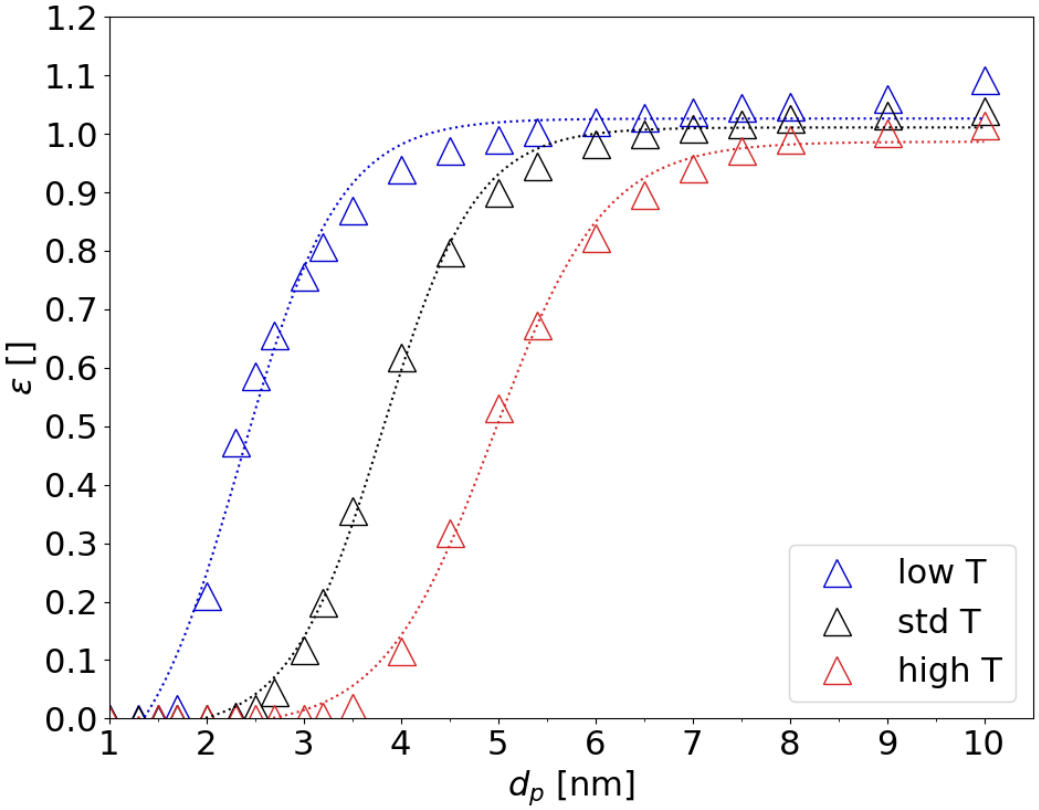

Fig. 2. NaCl positive with 10% RH, all curves were recorded with the same CPC.

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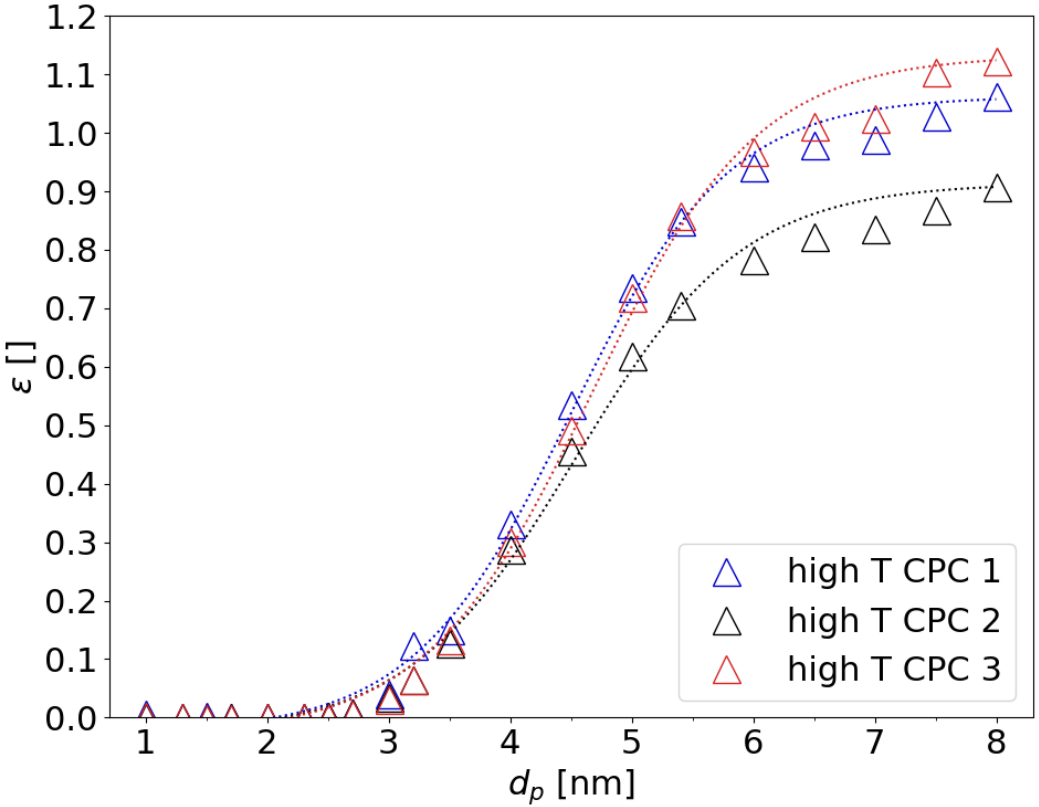

Fig. 3. NaCl negative with 0% RH, all curves were recorded with different CPCs but at constant temperature settings.

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