

Authors' response to the Referee 1 comments on manuscript titled “ *Versatile Aerosol Concentration Enrichment System (VACES) operating as a Cloud Condensation Nuclei (CCN) concentrator. Development and laboratory characterization.*” submitted to AMT 25th February 2019.

The authors would like to thank the reviewer for the helpful comments. Our responses are given in blue after each of the comments (bold characters). In the revised MS, changes concerning the comments of the reviewers are indicated in red. For a better understanding the representation of the virtual impactor in Fig. (1) was modified. Small editorial changes (such as corrections of typos, use of symbols instead of full text, etc.) are not reported here, as they are irrelevant for the scientific content of the paper.

However, based on the results presented I have some reservations how well it actually works. Although this is more like an introductory manuscript intended to demonstrate that VACES has some potential to operate as CCN counter, I would still like to see more evidence. Now the results are presented only for NaCl, which is a natural choice as it is well characterized material, but results with other particle types with lower hygroscopicity would be needed also.

In the revised MS, results from activation curves of 100 nm and 150 nm Polystyrene Latex particles (PSL) which have a less hygroscopic behaviour are added (see Table 2 and 3; page 20). The maximum experimental enrichment factor obtained from the PSL measurements is over 16 which is very similar to the experimental enrichment factor obtained for NaCl particles. The experimental enrichment factor for 100 nm PSL particles at different ΔT is incorporated in Fig. 2 (page 16) and shows the correct performance of the new CCN-VACES performance also for less hygroscopic particles. Some recent publications were referenced that provide a detailed description of VACES operation and demonstrate its potential as particle concentrator (page 10).

Zhi N., Moore, K.F., Polidori, A., and Sioutas, C. “Field Validation of the new miniature Versatile Aerosol Concentration Enrichment System (mVACES)”. *Aerosol Science and Technology*, 40 (12), 1098-1110, 2006

Saarikoski S., Carbone S, Cubison MJ, Hillamo R, Keronen P., Sioutas C., Worsnop D.R, Jimenez J.L. “Evaluation of the performance of a particle concentrator for on-line instrumentation”. *Atmospheric Measurement Technologies*, 7, 2121-2135, 2014

Zhao, Y., Bein, K.J., Wexler, A.S., Misra, C., Fine, P.M. and C. Sioutas, C. “Using a Particle Concentrator to Increase the Hit Rates of Single Particle Mass Spectrometers”. *Journal of Geophysical Research*, 110 (D7): Art. No. D07S02, 2005

Based on the presented data it is really difficult to estimate if the instrument accuracy is high enough for determining the CCN activity.

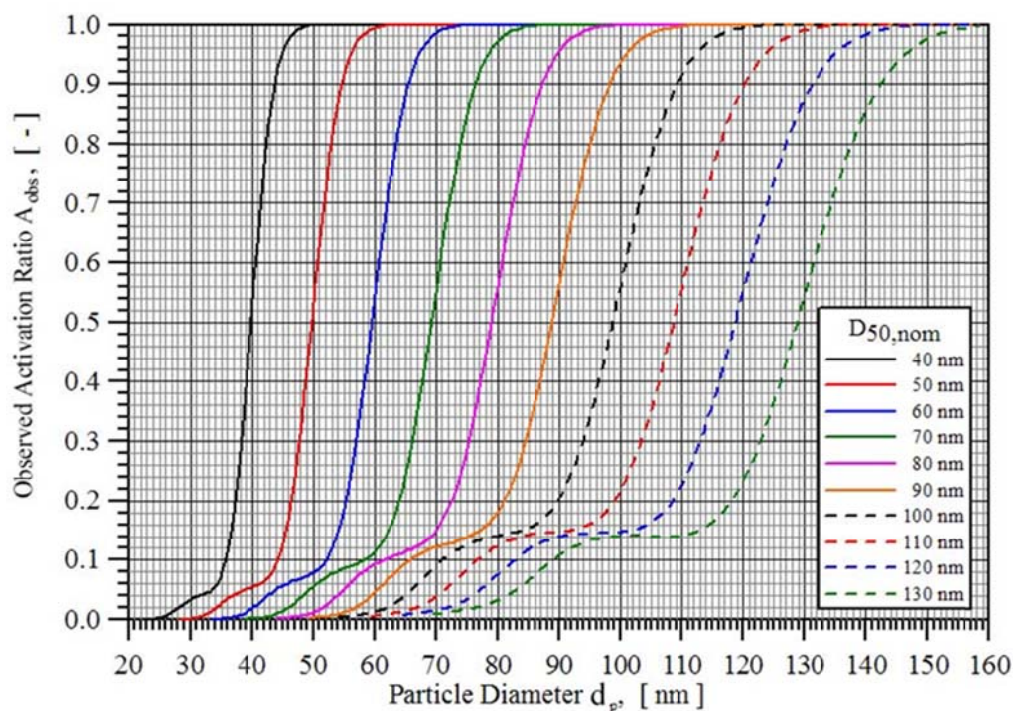
The activations curve as well as their reproducibility (under stable ambient conditions) are a clear evidence of the proper functioning of the new CCN-VACES. We do suggest, however, that the system should be calibrated by recording an activation curve with test aerosol particles previous to a measurement series. If the modified VACES had not functioned properly, the enrichment factors would not have been comparable to studies (Kim et al., 2001a).

1) The instrument is described nicely, but more information could be added how long it actually takes to conduct a full scan of particle size and different temperatures. Also it is said that original VACES is modified to get into lower supersaturations, this could be elaborated more also.

A more detailed paragraph has been incorporated in the manuscript. Modifications from the original VACES are explained in (page 4, lines 19-25). More information on the measurement procedure has also been added (page 7, lines 3-5; page 7, lines 15-23).

2) There is always quite a high fraction of activated particles. Have the authors accounted for the multiple charging of particles in neutralizer? As the cut of size of virtual impactor is relatively low, part of that could be explained with multiple charged larger particles selected by DMA. However, activated fraction should still go closer to zero. The reason for high activated fraction should be explained.

Doubly and higher charged particles may have been the reason why the activation curves do not go closer to zero. An indication for this effect is the apparent size dependence of the start of the curves – larger particles will have more multiple charges. According to the study by Rose et al., (2008) the influence of doubly charged particles appears in the activation curves as first plateau in their activation curves plotted as activation ratio vs. particle size at constant supersaturation. Similar curves were obtained in an earlier (unpublished) study with our static thermal diffusion chamber CCNC (Vera Meyer, Kondensationskernaktivierung des wasserlöslichen Anteils in urbanem Aerosol, Diploma Thesis, University of Vienna, 2006; see figure below). In our case, the activation curves are plotted as functions of ΔT (or T_{out}) for constant particle size. The 50% point of these curves, however, which is used to calculate supersaturations, is not influenced by this effect.



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3) Compared to actual differences in activation temperatures, the slope of activation curve is very gentle. Why is that?

The slope depends of course on the spread of the x-axis. We chose a large spread, as the difference in T_{out} is so small for curves obtained for particles of different diameters. In general, the slope of activation curves varies depending on particle size and / or supersaturation. The activation curves obtained e.g. by Giebl et al. (2002) (activation ratio plotted vs. dry diameter) are steeper at higher supersaturations ($SS = 1.08\%$ and $SS = 0.42\%$) than at lower ones.

Do the particles experience highly different supersaturation conditions within the condenser?

In the condenser tube the turbulent flow regime ensures a radially quite homogeneous mixture of water vapour. Supersaturation increases along the tube axis.

It is said that the flow is turbulent, but could this cause different growth behavior for different sized particles?

The growth behaviour of a particle depends on its activation according to Köhler theory, so of course differently sized particles (if present; we used monodisperse particles) will grow differently at specific water vapour supply and temperature. Of course some droplets might be lost to the walls in the turbulent flow, which might be a reason for the difference between theoretical and experimental enrichment factor.

Can the NaCl calibration be used also for particles with greatly lower hygroscopicity or should there some other component also? I see this as a big source of uncertainty and makes me wonder if it is possible at all to use the instrument outside laboratory conditions where the capability for chemical analysis would be really needed?

We used NaCl to calibrate the instrument, i.e. to see which T_{out} gives which supersaturation (at constant T_{s}), so NaCl can be seen here as a “supersaturation sensor”. The new data obtained with PSL show that PSL particles are also activated at the expected supersaturation (obtained from Kelvin theory). The supersaturation achieved in the CCN-VACES depends on the temperature settings, and not on the chemical nature of the particles. Once the instrument’s temperature settings are well established and stabilized, we can determine the supersaturation in the system and enrich other different aerosols particles. Complex aerosols containing volatile or semi-volatile material, however, might experience some changes in the temperature range used in the system. This is true, however, also for other CCN counters that operate at elevated temperatures.

4) Is the data used for figures 3 and 6 the same. Behavior looks highly different and I doubt if observations are as reproducible as claimed by the authors. In Figure 3 the Delta T differs most for smaller particles whereas in Figure 6 T_{out} differs most for larger particles.

The observation is correct, but the figures are correct, too. We clarified this in the revised MS. As the reviewer pointed out, activation curves in Fig. (3) for smaller particles correspond to higher ΔT values while the activation curves for larger particles correspond to lower ΔT values. The activation curve for 30 nm particles is shifted to the right, i.e. to higher ΔT , which is the temperature difference between the saturator temperature and the condenser temperature ($T_{\text{s}} - T_{\text{c}}$). If the saturator temperature is fixed and remains constant, a higher ΔT means lower condenser temperatures T_{c} . As seen in Fig. (3), 200 nm particles are activated at lower ΔT which implies higher T_{c} and consequently a lower critical supersaturation.

A more detailed paragraph has been incorporated in the manuscript (page 8, lines 24-29). In Fig. (6), activation curves are plotted vs. T_{out} . High T_{out} correspond to low supersaturations, so the curves for small particles needing higher supersaturations (and lower T_{out}) to activate are shifted to the left.

An extended explanation has been incorporated in the manuscript (page 9, lines 29-30) and page 10, lines 2-4).

5) There is no variability (deviation from the mean) presented in the observations. Presenting that could give some information how stable the condenser is.

As the temperature difference between the curves is relative small errors bars were omitted in order to show the curves more clearly.

6) Measurement setup involves quite high temperature differences. This might cause problems with particles of semivolatile nature like several organics or even nitrates. How do you account for this?

Previous studies (Sioutas et al., 1998; Saarikoski et al., 2014) performed with the original VACES have demonstrate that the concentration enrichment does not depend on the composition of the measured aerosol. The experimental concentration enrichment factor obtained for (hygroscopic) ammonium sulfate, (semi-volatile) ammonium nitrate, non-hygroscopic PSL and indoor air was practically the same. As in many other instruments, particles containing volatile compounds may be changed, but the VACES not designed to characterize volatile species.

7) In Equation 2 the critical supersaturation is determined by the particles size. The temperature contribution is minimal and thus Table 3 gives no information which could somehow be used to evaluate the performance. Thus it could be removed.

We decided to keep Table 3, as it gives the temperature settings needed to set supersaturations in cases where the chemical composition of the aerosol is unknown.