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THREE YEARS OF EXPERIENCE WITH MEASUREMENT OF CLOUD CONDENSATION NUCLEI CONCENTRATIONS USING CLOUD CONDENSATION NUCLEI COUNTER CCN-200

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INTRODUCTION

Aerosol particles in the atmosphere that allow water vapor to condense and form cloud droplets are called Cloud Condensation Nuclei (CCN). Elevated concentrations of CCN tend to increase the concentration and decrease the size of droplets. This can lead to suppression of precipitation in shallow and short-lived clouds and to greater convective overturning and more precipitation in deep convective clouds, Rose et al. (2010). The response of cloud properties and precipitation processes to increasing anthropogenic aerosol concentrations represents one of the largest uncertainties in the current understanding of climate change. One of the fundamental challenges is to determine the ability of aerosol particles to act as CCN under relevant atmospheric conditions. Knowledge of the spatial and temporal distribution in the atmosphere is essential to incorporate the effects of CCN into meteorological models of all scales, Huang et al. (2007). Long-term CCN measurements are performed at aerosol monitoring sites such as those forming ACTRIS (Aerosols, Clouds and Trace Gases Research Infrastructure) network. In this paper, we present the three-year experience of measuring CCN concentrations over the National Atmospheric Observatory Košetice (NAOK), a rural background site in the Czech Republic. The first results of these measurements were presented by Mishra et al. (2022).

EXPERIMENTAL SETUP

The instrument used for the CCN concentration measurements was a Dual Column Cloud Condensation Nuclei Counter (CCN-200) purchased from Droplet Measurements Technologies, USA. The DMT CCNC operates on the principle that heat conduction in air is slower than diffusion of water vapor (Roberts and Nenes, 2005). The CCNC operates by maintaining a positive temperature difference between the top and bottom of the column. Inside the column, the supersaturated water vapor condition is caused by diffusion of water vapor from the warm, moist column wall toward the centerline, at faster rate than heat. In order to obtain homogeneous data sets with high quality CCN measurements, a standard operating procedure (SOP) has been defined within the ACTRIS-2 project (WP3-NA3). The SOPs are defined for both polydisperse and monodisperse measurements. The CCN dual column counter we use allows us to make two simultaneous measurements of CCN concentrations. Different combinations of supersaturations (SSs) can be used in each column, so that data for multiple SSs can be collected during one measurement cycle. Another option is to perform the CCN measurement in one column in a polydisperse manner and in the other in a monodisperse manner. The advantage of monodisperse CCN

concentration measurement is that fractions of activated aerosol particles can be obtained on each SS for several particle sizes, so that two-dimensional CCN concentration spectra can be obtained during one measurement cycle. In our measurements, we mostly used the same SS in both columns during the measurement cycle, but in one measurement period (No. 3 in Table 1) we used different SS in columns A and B. An overview of the measurement campaigns can be found in Table 1.

Tab 1: SS settings of measurements in different time periods. No 1 - period from 24. 6. 2019 to 26. 2. 2020, No. 2 - period from 24. 6. 2020 to 21. 9. 2020, No. 3 - period from 21. 9. 2020 to 14. 10. 2020, No. 4 - period after WiIN10 System Upgrade from 14. 4. 2021 until now.

No. 1	SS [%]	0.1	0.15	0.2	0.3	0.5	1
	Time [min]	10	6	6	7	8	8
No. 2	SS [%]	0.1	0.2	0.3	0.5	1	
	Time [min]	20	10	10	10	10	
No. 3	SS_A [%]	0.15	0.2	0.25	0.5	0.7	
	SS_B [%]	0.1	0.2	0.3	0.5	1	
	Time [min]	20	10	10	10	10	
No. 4	SS [%]	0.1	0.2	0.3	0.5	1	
	Time [min]	10	7	7	8	8	

Apart from the above-mentioned advantages, the CCN-200 also has some disadvantages compared to the single-column CCN-100. They arise from the fact that the same size box can accommodate two instruments instead of one. As a result, the replacement of some spare parts (sheath flow filters, Nafion, etc.) is more complicated due to the poor access to these parts. Another problem occurs during the transition period from the highest to the lowest SS. The amount of heat that must be removed from the cover is twice as large and it takes longer to stabilize the temperatures for new SS than for the CCN-100 single-column instrument. This must be taken into account when setting the time for the measurement cycle overlay. Unfortunately, there is no reference to this phenomenon in the CCN-200 instruction manual. The water consumption is also twice that of the CCN-100, which is not insignificant when measuring in remote locations. Therefore, we have replaced the original 0.5-liter bottles with 2-liter bottles, which allows 8 days of continuous operation of the CCN-200.

INTERCOMPARISON AND CALIBRATION WOKSHOP

In March 2020, we participated in an intercomparison and calibration workshop at TROPOS in Leipzig, Germany, where calibration of flow rates and SSs as well as intercomparison measurements with our CCN-200 and four other CCN-100 instruments were performed. On this occasion we also found out the problem with the cooling of the optical detector. The results are shown in Fig. 1a. We can see that the temperature T_1 is almost constant and the temperatures T_2 and T_3 change up and down quite rapidly when SS changes. T_{Opc} also increases rapidly but decreases very slowly. This is due to the fact

that the optical cell is heated but not cooled. In fact, at the end of the 10-minute measurement interval for SS 0.1%, T_{Opc} is still about 6 °C above T_3 (the correct value for $T_{Opc\,Set}$ in the CCN-200 software is $T_3 + 2$ °C). Of course, T_{Opc} should be higher than T_3 to prevent fogging of the optical cell, but too high T_{Opc} leads to evaporation of condensed water and loss of droplets. Studies with DMT have shown (Operator Manual, 2018) that T_{Opc} can be up to 5 °C above T_3 without decreasing concentration. At a T_{Opc} of 7 °C above T_3, there is a 20% loss of droplets, which is similar to the case of our measurements with 10 min interval for SS 0.1%. After adjusting the measurement interval at SS 0.1% to 20 minutes, see period No. 2 in Table 1, the difference between T_{Opc} and T_3 at the end of the
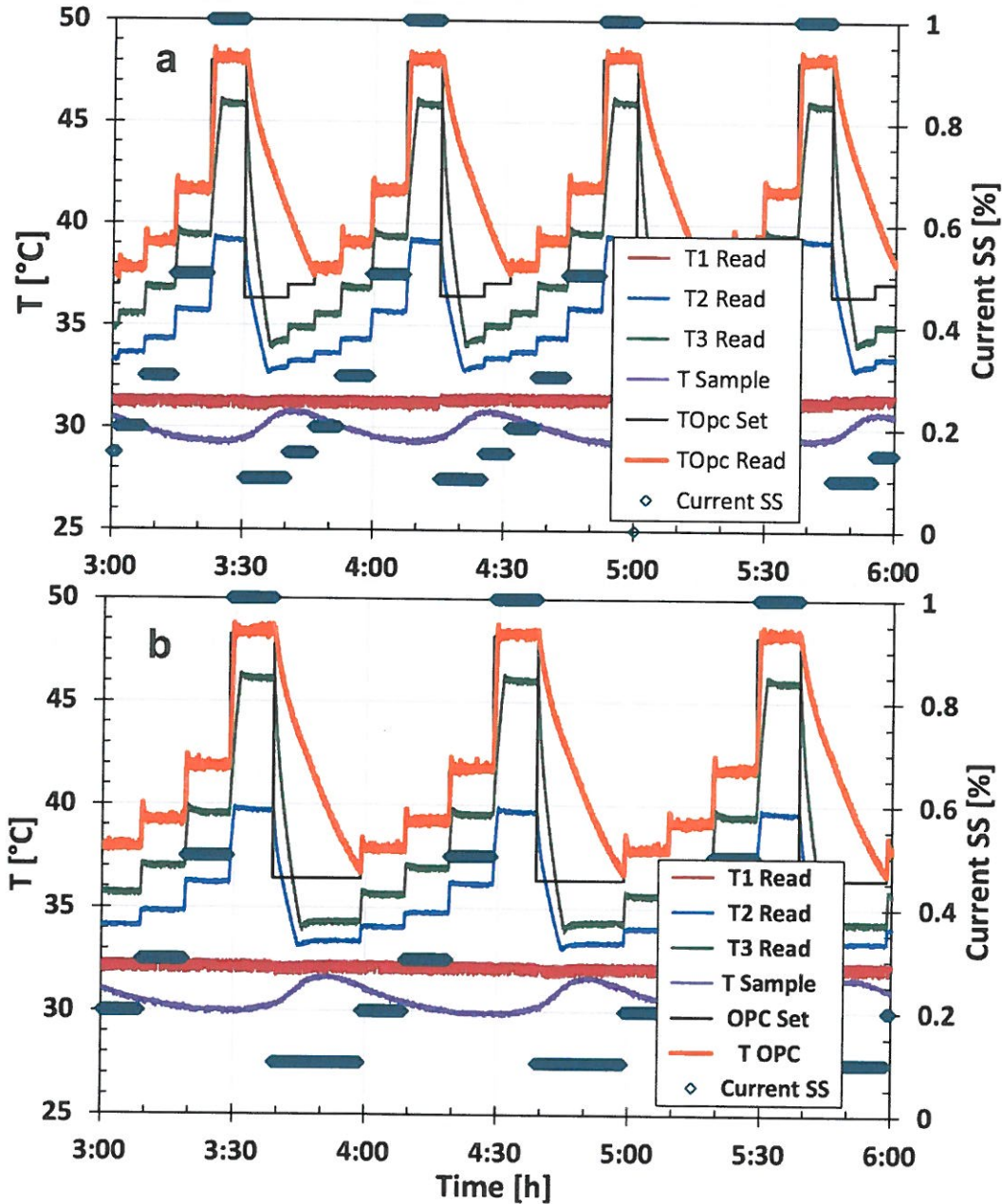

Fig. 1a, b: Time dependences of temperatures at CCN-200 for 45 and 60 minutes measuring cycles. T_1, T_2 and T_3 - temperatures at column top, middle and bottom, T_{Sample} - temperature in the inlet manifold, T_{Opc} and Opc Set - measured and set temperature in the optical cell.

measurement interval did not exceed 5 °C, see Fig. 1b, so we can consider the measurements in period No. 2 to be correct.

CCN-200 – WIN10 SYSTEM UPGRADE

At the end of 2020, we accepted a quote from DMT for CCN-200 - WIN10 System Upgrade. The upgrade included: WIN10 Electronic System Hardware Upgrade, WIN10 Operating Software & Driver Installation, Win10 CCN-200 Software Installation and regular maintenance, factory calibration and door-to-door logistics. We were told that the functionality of the unit after the upgrade was the same as before, but that was not the case. Water consumption is now about 15% higher, so the 2-liter supply bottle is used up in 7 days instead of 8. The main difference, however, is in the setting of the SSs. In the Win7 CCN-200 software, the length of the measurement time for each SS was exactly the same as specified by the user in the SS settings table, regardless of whether the temperatures were stabilized or not, so the times for each SS and for the entire measurement cycle were precisely defined. In the WIN10 CCN-200 software, the measurement time for each SS consists of two parts: the unknown time required to stabilize all temperatures, including T_{opc}, and the time defined by the user for the measurement itself in the SS settings table. Temperatures are considered stabilized when the difference between the set temperature and the measured temperature is ≤ 0.4 °C. The sum of unknown times for each SS during the measurement cycle is almost 20 minutes, mainly for the transition from the highest SS to the lowest. Originally, this time was even longer, but it was significantly shortened by using a new insulation of the optical cell. From the above, it is obvious that we cannot specify exact times for individual SS or for the entire measurement cycle. Another novelty of the upgrade is that the user can choose between two measurement modes: System Stability and Individual Stability. In system stability, the SSs and time intervals in both columns must be the same. The measurements in both columns are then synchronized, i.e. the measurement cycles in both columns start and end at the same time. In Individual Stability mode, the user can set different SSs and different time intervals in each column, but the measurement cycles in each column are different in length and the measurements are not synchronized.

Even with Win7 system, CCNC would occasionally reboot spontaneously, with a frequency of reboots every month or two. This was strange, but acceptable. However, after the system upgrade, the frequency of reboots gradually increased, eventually ending in a cyclic reboot and eventual system collapse. As a result, we sent CCNC back to DMT for warranty repair and were surprised to find that the problem was resolved very quickly. We were told to use UPS for the CCNC power supply to avoid voltage fluctuations. However, this did not work. After about two months, the problem repeated itself with spontaneous reboots and the system breaking down. The diagnosis made by our employee from IT together with technicians from DMT revealed that the problem was most likely a damaged PC of the CCNC. Initially, we thought we would have to buy a new PC and replace it ourselves. However, we were pleasantly surprised. DMT staff (sales manager and technicians) visited our institute and replaced the damaged PC, helped calibrate the flow rates, and accompanied us to NAOK to see the installation of the CCNC at a remote site. Since then (November 16, 2021), the CCN-200 has been working properly with no spontaneous reboots.

RESULTS AND CONCLUSIONS

We have been collecting CCN concentration data through NAOK since June 2019. Until October 2020, we used the Dual CCN.exe program running on WIN7 operating system. This data is almost ready to be submitted to the EBAS database. However, the data set from June 2019 to February 2020 contains CCN concentrations at SS 0.1%, which were underestimated by 10 - 20% due to the short measurement interval and T_{Opc} being too high. Data collected after CCN-200 - WIN10 SYSTEM UPGRADE have an undefined measurement cycle length. For these data, the procedure for submission to the EBAS database still needs to be worked out and approved by NILU.

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