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Calibration and measurement uncertainties of a continuous-flow cloud condensation nuclei counter (DMT-CCNC): CCN activation of ammonium sulfate and sodium chloride aerosol particles in theory and experiment

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Experimental and theoretical uncertainties in the measurement of cloud condensation nuclei (CCN) with a continuous-flow thermal-gradient CCN counter from Droplet Measurement Technologies (DMT-CCNC, Roberts and Nenes, 2005) have been assessed by model calculations and calibration experiments with ammonium sulfate and sodium chloride aerosol particles in the diameter range of 20-220 nm. Experiments have been performed in the laboratory and during field measurement campaigns, covering a wide range of instrument operating conditions (650-1020 hPa pressure, 293-303 K inlet temperature, 4-34 K m^{-1} temperature gradient, 0.5-1.0 L min^{-1} flow rate). For each set of conditions, the effective water vapor supersaturation S_{eff} (0.05-1.4%) was determined from the measured CCN activation spectra (dry particle activation diameters) and Köhler model calculations. High measurement precision was achieved under stable laboratory conditions, where the relative standard deviations of S_{eff} were as low as $\pm$1%. During field measurements, however, the relative variability increased to about $\pm$5%, which can be mostly attributed to variations of the CCNC column top temperature with ambient temperature. The observed dependence of S_{eff} on temper-

ature, pressure, and flow rate was compared to the CCNC flow model of Lance et al. (2006). At high S_{eff} the relative deviations between flow model and experimental results were mostly less than 10%, but at $S_{eff} \leq$0.1% they exceeded 40%. Thus, careful experimental calibration is required for high-accuracy CCN measurements - especially at low S_{eff}. A comprehensive comparison and uncertainty analysis of the various Köhler models and thermodynamic parameterizations commonly used in CCN studies showed that the relative deviations between different approaches are as high as 25% for $(NH_4)_2SO_4$ and 16% for NaCl. The deviations were mostly caused by the different parameterizations for the activity of water in aqueous solutions of the two salts. To ensure comparability of results, we suggest that CCN studies should always report exactly which Köhler model equations and parameters were used. Provided that the Aerosol Inorganics Model (AIM) can be regarded as an accurate source of water activity data for highly dilute solutions of $(NH_4)_2SO_4$ and NaCl, only Köhler models that are based on the AIM or yield similar results should be used in CCN studies involving these salts and aiming at high accuracy. Experiments with $(NH_4)_2SO_4$ and NaCl aerosols showed that the conditions of particle generation and the shape and microstructure of NaCl particles are critical for their application in CCN activation experiments (relative deviations up to 18%).

References:

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