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Parameterization of the cloud condensation nuclei (CCN) activity of ambient black carbon at different aging levels: Comparison between theoretical and experimental results

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The influence of black carbon (BC) particles on the climate system has recently gained interest because of their very specific properties and the significant fraction of anthropogenic sources they originate from. These particles, emitted by combustion processes, are generally hydrophobic upon emission, but, with aging, they can acquire a coating by the condensation of, or coagulation with, organic and inorganic material. Accurately predicting the variations of CCN-ability from bare to thickly coated BC is a major challenge for current climate models, as it has direct implications on the life-cycle of BC and thus its climate impacts. A recent study from Lund et al. (2017) highlighted the significance of this challenge: when varying the amount of coating required to reach CCN-activation, they reported changes by up to 25-50 % of the radiative forcing of BC via aerosol-radiation interactions compared to a baseline simulation. We performed two field campaigns in order to investigate the cloud droplet activation behavior of BC-containing particles as a function of their size and mixing state. The first campaign took place in the city of Zurich, allowing us to study the activation behaviour of freshly emitted BC, mostly from traffic, in fog. The second one was performed at the high altitude research station Jungfraujoch (3580 m asl), with a focus on activation of heavily aged BC. In order to compare our experimental results with the theoretical activation behaviour of BC, we used a theoretical approach based on a core-shell model and the κ-Köhler theory (Petters and Kreidenweis, 2007) to retrieve the critical supersaturations of BC-containing particles. We also estimated the effective peak supersaturation in cloud/fog events, using a method from Hammer et al. (2014). We could then test the agreement by checking whether the critical supersaturation at 50 % activated fraction matched the range of retrieved effective peak supersaturations. We found very good agreement between experimental and theoretical results in fog, where low supersaturations (typically around 0.05 %) restrict activation to large particles which are detected by our instruments. Although the much lower supersaturations found in fogs compared to clouds make this kind of investigation more difficult in terms of detection limits, we found clear links between the fractions of activated BC-containing particles and the supersaturation in clouds.