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Ice crystal formation on soot particles

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Soot particles are primary particles produced by incomplete combustion of both biomass and/or fossil fuels and thus constitute a major anthropogenic pollutant. They are generally complex internal mixtures of black carbon (BC) and organic matter (OM) (Bond et al., 2013; Petzold et al., 2013). While these properties strongly depend on the emission sources, they can be altered during atmospheric cycling, including interactions of the primary particles with other atmospheric matter and/or gases. Physically, soot particles usually form fractal-like aggregates composed of the primary carbonaceous spherules. Such aggregates include a variety of cavities and pores that are formed between primary carbonaceous spherules or as a result of random packing of the fractal chains.

Overall, estimates of the net radiative forcing associated with BC are still associated with large uncertainties, even though being one of the most important climate forcing agents (Bond et al., 2013).

While BC generally absorbs solar radiation, thus contributing to a warming effect on global climate it can also act as a heterogeneous ice nucleating particle (INP) and impact cloud-radiation interactions, potentially cooling the climate (Lohmann, 2002). Prediction of the ice nucleation behavior of soot particles requires consideration of both, the chemical and physical properties of the particles.

Here, we present a systematic laboratory-based investigation of the ice formation behavior of a set of soot particles. Different commercial soot samples are used, along with a soot produced by a propane diffusion burner, using a Combustion Aerosol Standard Generator (miniCAST, JING AG). miniCAST burners are frequently used as proxy for atmospheric soot particles. Ice nucleation ability of these soot types is tested on size-selected particles covering the size range between 100 and 400 nm and a temperature range from 253 K to 218 K, using the Horizontal Ice Nucleation Chamber (HINC, Lacher et al. 2017), a Continuous Flow Diffusion Chamber (CFDC).

We complement our ice nucleation experiments with auxiliary measurements including BET-surface determination and thermogravimetric analysis, in order to characterize the physico-chemical properties of the tested aerosol particles. In addition, analysis of Transmission Electron Microscope (TEM) images allows for a detailed characterization of the morphology associated with our soot samples. This supports investigating the influence of any cavities on our ice nucleation results, considering a Pore Condensation Freezing (PCF) mechanism for ice nucleation onto soot particles. During PCF liquid water is taken up within pores below water saturation due to the inverse Kelvin effect, which can subsequently freeze heterogeneously in presence of an active site (for $T > 235$ K) or homogeneously (for $T < 235$ K) causing ice to nucleate on the soot aggregate.

Results show different activation behavior of the soot over the temperature range investigated. While the commercial soot samples show freezing well below water saturation in the cirrus regime, CAST-brown soot needs conditions above water saturation to show any activation. For the mixed-phase cloud conditions all soot types show droplet activation for high supersaturations w.r.t water.

Significance statement

We present a systematic laboratory study on the ice nucleation behaviour of different soot particles along with detailed measurements on the particle physico-chemical properties. This allows us to shed light on the ice nucleation mechanism of soot, which is poorly understood so far.

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