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Surface structure, crystallographic and ice-nucleating properties of cellulose

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Increasing evidence of the high diversity and efficient freezing ability of biological ice-nucleating particles is driving a reevaluation of their impact upon climate. Despite their potential importance, little is known about their atmospheric abundance and ice nucleation efficiency, especially non-proteinaceous ones, in comparison to non-biological materials (e.g., mineral dust). Recently, microcrystalline cellulose (MCC; non-proteinaceous plant structural polymer) has been identified as a potential biological ice-nucleating particle. However, it is still uncertain if the ice-nucleating activity is specific to the MCC structure or generally relevant to all cellulose materials, such that the results of MCC can be representatively scaled up to the total cellulose content in the atmosphere to address its role in clouds and the climate system. Here we use the helium ion microscopy (HIM) imaging and the X-ray diffraction (XRD) technique to characterize the nanoscale surface structure and crystalline properties of the two different types of cellulose (MCC and fibrous cellulose extracted from natural wood pulp) as model proxies for atmospheric cellulose particles and to assess their potential accessibility for water molecules. To complement these structural characterizations, we also present the results of immersion freezing experiments using the cold stage-based droplet freezing BINARY (Bielefeld Ice Nucleation ARaY) technique. The HIM results suggest that both cellulose types have a complex porous morphology with capillary spaces between the nanoscale fibrils over the microfiber surface. These surface structures may make cellulose accessible to water. The XRD results suggest that the structural properties of both cellulose materials are in agreement (i.e. P21 space group; a=7.96 Å, b=8.35 Å, c=10.28 Å) and comparable to the crystallographic properties of general monoclinic cellulose (i.e. Cellulose Iβ). The results obtained from the BINARY measurements suggest that there is no significant difference of the immersion ice nucleation activity of MCC and fibrous cellulose in supercooled water. Overall, our findings support the view that MCC may be a good proxy for inferring water uptake, wettability and ice nucleating properties of various cellulose materials. In addition, we discuss the ice-nucleating efficiencies of both cellulose samples and plant debris from the AIDA (Aerosol Interaction and Dynamics in the Atmosphere) chamber experiments in comparison to the BINARY results. The influence of the acid processing of cellulose on its ice nucleation propensity may also be discussed to further demonstrate their atmospheric relevancy.

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