This study presents and analyzes carefully designed laboratory measurements describing the ice nucleation behavior of soot particles internally mixed to various degrees with sulfuric acid. Data are discussed at cirrus temperatures ranging from 218 K to 243 K, for relative humidities from ice saturation to above water saturation and for two selected soot particle sizes (200 nm and 400 nm). Importantly, soot particle coatings are varied from thin to thick, corresponding to about 3% to 30% weight fractions of soluble material, respectively. The study finds that H2SO4/H2O coatings suppress the ice nucleation ability of soot particles at cirrus levels to a degree that depends on the coating thickness.

The in-depth discussion of the coating process confirms how crucial it is to take disparate soot particle morphologies (i.e. sources and atmospheric ageing processes) into account when assessing their cirrus-forming potential. The characterization of particle morphologies with the help of high-resolution imagery in Appendix C is particularly valuable supporting data interpretation. The study fills an important gap in the literature by presenting a systematic investigation of how soot particle coatings affect heterogeneous ice nucleation and offering a physical explanation (based on pore condensation and freezing (PCF) and immersion freezing processes) for the observed behavior. Existing data sets are valuable in their own right, but have not led to a consistent picture regarding the role of soot particles in cirrus cloud formation.

In my opinion, this is an excellent contribution to the literature on this topic, and I recommend publication after the authors considered my following comments. Line numbers refer to the initially submitted manuscript (made available for technical review).

<u>Major</u>

l81-83: To me, it is unclear if contrail processing had actually happened in the measurements of Petzold et al. (1998) or whether the peculiar signature of the probed soot particles (bimodality) was a feature of the fresh emissions. Please check.

l354: Please define what a mesopore is. This is especially relevant as pore properties feature prominently in the data discussion (mostly in section 3.1). In l402, the term ‘micropore’ is used and it is unclear what sets it apart from a mesopore.

l668ff: The authors may want to more clearly state in this paragraph that current data suggest that only soot particles with sizes 200 nm and 400 nm actually contribute to cirrus formation, hence are ‘atmospherically relevant’. In the case of aviation soot (l683f), most particles emitted at altitude belong to the Aitken mode, i.e., they are much smaller.

Even poor INPs can be efficient in altering cirrus if present in sufficiently high concentrations. BC particles are often present in high number concentrations close to their sources. It may be good to add that besides size-dependent ice nucleation ability the soot particle number concentration must be known in order to judge their potential to affect cirrus formation notably.

I was also wondering whether the authors like to add the importance of further measurements probing smaller soot particles (if feasible, down to 50 nm) to confirm the trend of ice nucleation ability diminishing with decreasing size. This would be especially important for the case of aviation soot, but possibly for other high-temperature combustion sources, too.

Minor

l11: I believe you mean ‘net warming’. Same in l39.

l38: The plural of aircraft is still aircraft.

l39: As written, it is unclear whether the climate warming effect relates to cirrus or to aviation soot.

l48: Please clarify that for T<235 K, homogeneous ice formation occurs in liquid aerosol droplets only, setting cirrus apart from mixed-phase clouds.

l63: Sentence ending with ‘where aeroengines exhaust sulphur emissions’ appears to be incomplete.

l73: Sentence ends with ‘but remaining unconstrained’ sounds awkward.

l118: What is a ‘potential’ pore?

l292+293: Sentence sounds awkward.

l331: Define AF.

l418: Please clarify that water uptake is reduced to the inverse Kelvin effect, which, what I believe, is what the authors mean to say.

l686ff: A recent study* concluded that even a small number of ice-active aviation soot particles is capable of modifying the total number and mean size of cirrus ice crystals when nucleating ice alongside homogeneous freezing, albeit with a minor impact on cirrus optical depth. It was found that, based on soot-PCF, only uncoated (barely coated) soot particles with sizes >100 nm contributed to enhanced ice nucleation activity (after contrail-processing). This information may be used in this paragraph relating to mCAST black soot to better explain what is meant by ‘may not inhibit or compete with (aerosol) droplet homogeneous freezing’ (l689f).

In sum, while not making ‘a significant contribution to cirrus cloud formation via PCF’ (l693), soot particles may still perturb cirrus microphysical properties. This cautionary note also applies to the subsequent paragraph (l697ff) discussing FW200 soot samples.

*https://www.nature.com/articles/s43247-021-00175-x

l706: consider replacing ‘nucleated’ with ‘water-activated’ to avoid confusion with ice nucleation

l727: ‘require RH $>RH_{hom}$ conditions’ — for what?

l738: ‘systemically’ -> systematically

l739: ‘soot role’ -> role of soot

Bernd Kärcher, DLR-IPA