Review of manuscript egusphere-2023-988, Wildfire smoke triggers cirrus formation: Lidar observations over the Eastern Mediterranean (Cyprus) for publication in ACP by Rodanthi-Elisavet et al.

The paper describes the detection of a smoke plume from forest fires in North America (California) taking 8 days to arrive in Europe in 2020 and crossing the Mediterranean from Portugal to Cyprus arriving as an aged biomass burning plume while being transported in the mid-troposphere to lower stratosphere (6-14 km). Remote sensing detection methods are used including a polarisation Raman lidar for particle backscatter and extinction coefficients as well as depolarization ratios. Cirrus formation events, virga and alternating cirrus structures from gravity waves.

The paper is of interest to the readers of ACP, in particular the cold cloud, IN and remote sensing and aerosol remote sensing community. However, the following minor revisions and not-so minor points need to be clarified. I have listed the questions/edits in order of appearance and not in order of importance.

Line 2: Suggest "Presently one key aspect of research is whether or not.."

Line 17: Delete "The" ..start sentence with "Smoke.."
Line 27: The authors should elaborate more on the components biomass burning particles. Here they state OA and sulfate are the major contributors but BC cores and ash or mineral particles are also known to be part of the plume. I suggest discussing their contributions here and their emission likely hood as well. Also, because later in the manuscript the authors refer to these very components (BC and minerals) as being important to determine ice nucleation and so it seems appropriate to introduce them here.
Line 42: Delete "up to"
Lines 41- 47: These are all valid claims as far as my knowledge goes, but the authors should certainly include references from the literature to support these claims that chemistry and morphology of particles change with aging and cloud processing
Lines 54-55: The way the sentence is structured here is awkward to me, I suggest change to "can serve as deposition ice nucleating particles (DINPs). DIN INPs is redundant.
Line 57: "th" should be "the"
Line 59: "serve as *an* INP"
Line 60: suggest replace "take place" with "occur"
Line 66: replace "efficacy" with "activity" unless a time component to nucleation is being implied here
Line 67-69: want not able to follow the reasoning here. The sentences above with references are support that biomass burning particles can act as INPs but then this sentence says it remains to be shown if smoke particles can influence MPC and cirrus cloud development. Perhaps the authors wish to state that the former were lab studies, and it remains to be shown in-situ is this is the case. This should be made clear. Also sentence starting with "Those INPs.." which INPs, some specificity would be good to make it more clear to the reader. At the end of this sentence, the authors can link back to the mineral/ash particles that I suggested introducing earlier, since I think the authors are referring to these particles here.

Line 75: Delete “here” and move “ice cloud to earlier … i.e. I suggest “In this article, we will discuss a series *of ice cloud* lidar observations that were ..”
Lines 80-83: is this needed?

Line 85: inset comma after “..Raman lidar, Polly..”
Lines 90-91: How about marine aerosol, surely this is also part of the mix in the Eastern Mediterranean aerosol
Line 99: suggest replace “.Meanwhile also smoke is a topic of research (Nisantzi…” with “.. and smoke research more recently (Nisantzi..”

Line 104: “reflection by” should be “reflection of”
Lines 101-105: For a non-expert in remote sensing, this is a little too brief especially the part where the pointing to an off-zenith angle of 5° to avoid bias. Could this be elaborated a little more as it is important to distinguish the signal from ice in MPCs vs. Cirrus virga.
Line 115: replace “by” with “be”
Line 118: “signal-to-noise”?
Lines 125-129: I agree that *s* is used as an input parameter for the INP parameterisation, but I don’t see why the authors don’t use n_{250} as an input parameter for a parameterisation as well. I understand the commonly used DeMott 10 and 15 parameterisations [*Demott et al.*, 2010; *Demott et al.*, 2015] are for immersion freezing, but there are some cirrus parametrisations available for instance from the AIDA chamber work. Is it a good assumption that all particles larger than 500 nm are available as INPs? Perhaps more explanation or justification is needed here.
Lines 138-141: For the assumption that the aerosol retrievals are for dry conditions, this sounds reasonable, but can the authors also state the range of RH during the cirrus free conditions for when the retrieval was conducted? That would support their assumption to neglect water uptake and depend on the dry aerosol retrievals.

Section 4.1: In this section I think more justification for this method is needed or more clarification. If the authors treat the aerosol at DINPs, then why do they need to compute the INP from immersion mode at cold cirrus temperatures. The latter would only be relevant if the organic shell takes up water and dissolves, in which case if the core is BC, these would not be immersion freezing active since BC does not have active sites [*Kanji et al.*, 2020], but rather only freezes by deposition mode or PCF for temperatures below 235 K [*Chou et al.*, 2013]. And if a bulk droplet exists as these temperatures, then the freezing mechanism is homogeneous nucleation. Only when the $RH_i < 140\%$ is when PCF or DIN is considered relevant.

Also, the assumption that the particles are in equilibrium with the environment is not a good one for these conditions because the viscosity of the organic coatings really limits diffusion of water in the organics, so the very assumption of having glassy state or organic coatings, is contrary to assuming equilibrium conditions. The only relevance of immersion freezing at such cold temperatures would be if the organic coating is dissolved or diluted and the core is a mineral ash or dust compound.

In this regard, I would simplify and only consider DIN as the process and use that to retrieve INPs from the data and not immersion freezing since OA has been shown to nucleate ice via

DIN or PCF/DIN [*Kilchhofer et al.*, 2021; *Knopf et al.*, 2018; *Knopf et al.*, 2010]. And this suggestion is in line with what the authors write in section 5.1, (lines 192-196) that the fast lofting into the dry upper troposphere would limit core-shell structure formation and thus DIN would be supported over immersion freezing by water uptake.
Lines 203-209: The discussions here refer to supersaturated air and subsaturated air, but with respect to ice, but in Fig. 3a and c, the RH is plotted presumably with respect to water, because no SS RH regions are observable in Fig. 3a and c. Also, it is not clarified in the the caption of Fig. 3 that the RH is wrt water.

Lines 217: here I would reword to saying that an intensification of ice/virga was observed emerging from the smoke layer implying that strong ice nucleation by the smoke particles occurred. The way it is phrased now, is incorrect, as the process of nucleation was not observed by the remote sensing, but the ice virga evolution is observed.

Lines 250-255: Is there a reason why the highest number of calculated ICNC is 100 L-1 but the reservoir of INPs calculated was up to 6000 L-1, is this because not all particles are DIN in active, or the competition for water vapour? This would be better if the scale were RH_i rather than RH_w, so the reader can tell how close to ice saturation these values are.

Line 263 and 269, the units provided for updraft velocity seems different here. Is that intended, if so it should be stated that the GW observed here in this work had updraft speeds much lower than typical velocities mentioned on line 263.

Line 297: Here the authors should add that the heterogeneous IN was likely via DIN. It does not seem plausible to me that immersion freezing is the mechanism, see comments below.

Lines 300-315: The assumption of immersion freezing here is flawed in my opinion or not sufficiently justified. The authors nicely explain that the shell of the aerosol or the organic phase will likely not be liquified because of the diffusion limitations of water uptake therefore the aerosol might still be highly viscous or in the glassy state, as shown in Fig 8. What then should the water uptake mechanism be, if the OA is still glassy? If water condenses onto an OA shell that is not miscible with the condensed water, then this aerosol coated with water should freeze homogeneously since the T << 235K. If the water mixes with the OA coating and freezes at these low humidities, then it can be postulated that immersion freezing is taking place with the core promoting it because the RH is below that required for homogeneous freezing of solution drops at this temperature. But it can't be that the OA is in the glassy state, and acts as a core for the water to condense and the core of the OA is initiating immersion freezing in the droplet. IF bulk water is present at these conditions, it would freeze homogeneously.

What would be the active site on the OA core promoting immersion freezing and how can this be validated given the low T where the homogeneous freezing rate of the condensed water onto the glass OA shell would be very high as well?

I agree, the data in Fig. 8 show nicely that the ice occurrence is below the glassy transition lines, so it is likely that the OA is in the glassy state, as such with the above explanations DIN

is the only likely mechanism. For immersion freezing to take place, the OA shell should become miscible with part of the water taken up.

The DIN can be readily explained, here water vapour can adsorb onto the organic shell/coating of the aerosol and eventually the adsorbed water nucleates ice, or water vapour deposited on the surface nucleates ice. One can even imagine that small cracks or pores in the organic aerosol (due to ageing while being transported) can condense small pockets of liquid water which freeze homogeneously because the temp is low enough thus inducing PCF/DIN.

What should be the reason water condenses onto a glass aerosol at sub saturated conditions, if the glassy aerosol is not absorbing water due to the high viscosity and low diffusion rates? I think these two explanations do not go hand in hand.

Line 312: replace “the authors” with “we”

Line 325: The DIN ICNC are also higher in line with this mechanism. But also what is causing the differences between the DIN assessed ICNC and the immersion freezing one?

Line 333: The units of ICNC is wrong
Line 352: “Basis” should be “Basin”

In the conclusions or elsewhere in the discussion, the authors should address the differences between the ICNC derived from the simulations vs. the remote sensing methods. The max for instance was 75/L vs. 100/L which uncertainties can account for his, or at least use the remote sensing derived uncertainties to say that perhaps this difference is negligible given the uncertainty in the measurement. Some acknowledgement that this are not completely similar needs to be made.

Figures 3, 5 and 6: I would consider changing the RH scale to RH_i instead of RH_w. This allows evaluation of the cases of cirrus clouds based on supersaturation and the relevant phase is ice here, not liquid.

Figure 7. Please switch order so that the caption refers to panel a before panel b. Also the caption is disorganised, the authors refer first to panel b then to panel a and then back to b. This can be better consolidated.

Figure 8: The light blue area (last line caption) and the bluish area (caption line 3) are mentioned twice, but I think they refer to the same region in the plot. Please consolidate or correct. I only see one light blue/bluish area.

References

Chou, C., Z. A. Kanji, O. Stetzer, T. Tritscher, R. Chirico, M. F. Heringa, E. Weingartner, A. S. H. Prevot, U. Baltensperger, and U. Lohmann (2013), Effect of photochemical ageing on the ice nucleation properties of diesel and wood burning particles, *Atmospheric Chemistry and Physics*, *13*(2), 761-772, doi:10.5194/acp-13-761-2013.

DeMott, P. J., A. J. Prenni, X. Liu, S. M. Kreidenweis, M. D. Petters, C. H. Twohy, M. S. Richardson, T. Eidhammer, and D. C. Rogers (2010), Predicting global atmospheric ice nuclei distributions and their impacts on climate, *PNAS*, *107*(25), 11217-11222, doi:10.1073/pnas.0910818107.

DeMott, P. J., A. J. Prenni, G. R. McMeeking, R. C. Sullivan, M. D. Petters, Y. Tobo, M. Niemand, O. Moehler, J. R. Snider, Z. Wang, and S. M. Kreidenweis (2015), Integrating laboratory and field data to quantify the immersion freezing ice nucleation activity of mineral dust particles, *Atmospheric Chemistry and Physics*, *15*(1), 393-409.

Kanji, Z. A., A. Welti, J. C. Corbin, and A. A. Mensah (2020), Black Carbon Particles Do Not Matter for Immersion Mode Ice Nucleation, *Geophys. Res. Lett.*, *47*(11), 9, doi:10.1029/2019gl086764.

Kilchhofer, K., F. Mahrt, and Z. A. Kanji (2021), The Role of Cloud Processing for the Ice Nucleating Ability of Organic Aerosol and Coal Fly Ash Particles, *J. Geophys. Res.-Atmos.*, *126*(10), 21, doi:10.1029/2020jd033338.

Knopf, D. A., P. A. Alpert, and B. Wang (2018), The Role of Organic Aerosol in Atmospheric Ice Nucleation: A Review, *ACS Earth and Space Chemistry*, *2*(3), 168-202, doi:10.1021/acsearthspacechem.7b00120.

Knopf, D. A., B. Wang, A. Laskin, R. C. Moffet, and M. K. Gilles (2010), Heterogeneous nucleation of ice on anthropogenic organic particles collected in Mexico City, *Geophys. Res. Lett.*, *37*, L11803, doi:10.1029/2010gl043362.