

Dear reviewer,

thank you for careful reading of the manuscript and for providing many valuable comments and ideas how to improve the paper.

A brief overview of main changes:

(1) Section 1 (Introduction) has an improved structure, is more straight forward now. Section 2 covers the instrumental part only: Sect. 2.1: CARO, Sect. 2.2: Polly, Sect. 2.3: Nicosia radiosonde. Section 3 describes the lidar data analysis, including the INP parameterizations in Sect. 3.1. We improved the DIN parameterization a bit, introduced the contact angle concept.

(2) RH (over water) is no longer shown. In all figures, we switched to RH_{ICE} .

(3) We show a new simulation figure (Fig.8) to explicitly support the gravity wave observations on 1 November 2020. Afterwards, we show only one simulation figure (Fig.10, for 28 October) in the revised version instead of three (for 28, 30 October, 1 November) as presented in the submitted version.

(4) We went through the entire manuscript and improved the text as a whole along the comments of the reviewers.

Now the step-by-step response to all comments with our response in blue.

The essential changes in the manuscript are indicated in BOLD.

Review of manuscript egosphere-2023-988:

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.."

Considered!

Line 17: Delete "The" ..start sentence with "Smoke.."

Done!

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.

We discuss all this in more detail, including dust and ash aspects (in the introduction and, later on, in the result section).

Line 42: Delete “up to”

Done!

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

We rearranged the text in the introduction to meet these points. However, we try to keep the discussion short. An extended discussion (some kind of a review with references) regarding smoke transport, aging effects ,and smoke chemical and optical properties was already given in Ansmann et al. (2021).

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.

We changed wording (to avoid DIN INP) throughout the manuscript.

Line 57: “th” should be “the”

Improved!

Line 59: “serve as *an* INP”

Improved!

Line 60: suggest replace “take place” with “occur”

Improved!

Line 66: replace “efficacy” with “activity” unless a time component to nucleation is being implied here

Improved!

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.

We changed the text and leave out confusing statements to keep the introduction short.

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 ..”

We rearranged the text here (introduction section) to be more clear and to have a better context of all the mentioned and justified points.

Lines 80-83: is this needed?

A detailed overview of the paper content (at the end of the introduction) is no longer given.

Line 85: inset comma after “..Raman lidar, Polly..”

We changed phrasing a bit.... here ...

Lines 90-91: How about marine aerosol, surely this is also part of the mix in the Eastern Mediterranean aerosol

Yes, considered now!

Line 99: suggest replace “.Meanwhile also smoke is a topic of research (Nisantzi...” with “.. and smoke research more recently (Nisantzi..”

We changed the text to keep all this short.

Line 104: “reflection by” should be “reflection of”

We changed phrasing...!

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.

We extended this part considerably (Section 2.2).

Line 115: replace “by” with “be”

Done!

Line 118: “signal-to-noise”?

Yes!

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 parameterisations 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.

We rearranged the text a bit, but try to avoid a lengthy discussion here. We stick to our INP parameterizations (ABIFM, DIN) as given in Sect. 3.1. The primary goal of the paper is to show that smoke can lead to strong ice nucleation. In this stage of research, the ice nucleation mode is of secondary order of importance. There will be several follow-up papers (by us and by others), and then an extended discussion and the application (and testing) of a variety of INP parameterizations make sense.

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.

The radiosondes show at all RH values below 60% in cloud free air, in the absence of cirrus formations over many hours. And for an RH of <60% RH there is practically no bias (at least always below 10%). We discuss that now at the end of Section 3, before the start of Sect. 3.1.

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 at 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 3, 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.

We improved this section (4.1..... in the revised version 3.1) a bit by keeping the arguments of the reviewer in mind, but did not change much in Section 3.1. We introduced a statement of Berkemeier *et al.* (2014) as an argument that we present both INP parameterizations (ABIFM, DIN). The entire

research field is very new, and so many aspects concerning aged smoke particles and their role in ice nucleation are simply not known. Why should we already now reduce the information and application space?

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.

We improved all this. We clearly distinguish RH (relative humidity over water) and RH_{ice} (relative humidity over ice) now. All figures show RH_{ice} now.

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.

We follow this suggestion!

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.

We removed this confusing point (ICNC of 100 L⁻¹ vs INP reservoir, n_{250} , of 6000 L⁻¹) from the discussion. Such a discussion is not needed.

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.

We removed the confusion! One of the mentioned velocities was the horizontal wind speed, i.e., the travel speed of the entire gravity wave (and not an updraft speed). This is now explicitly mentioned.

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.

We mention at several places in the revised version that DIN was probably responsible for smoke-related ice nucleation. Only in the last figure, we show simulations for ABIFM (immersion freezing) and DIN (deposition ice nucleation). We show this just for comparison.

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 $T \ll 235K$. 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.

We removed this part of the explanations in the revised version. We will present this kind of discussion (as given in the paper of Berkemeier et al., 2014) in several follow up papers. It is too much here. And we agree that DIN is most likely responsible for ice nucleation. And this message is clearly given in the paper.

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?

As pointed out by Berkemeier, even at very low temperatures (below -50°C) but high humidities (RH above 70-90%), organic particles can become liquid if we wait long enough, at least they can develop a liquid surfacearound the glassy particle..... in that case we have immersion freezing...

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.

OK! No problem to agree with this.

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.

Yes, agree!

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.

Yes, probably! But all this remains hypothetical... and may be clarified in follow-up papers and discussions in papers, workshops, and conferences.

Line 312: replace "the authors" with "we"

The text was removed.

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?

In the revised version we show ABIFM vs DIN only in the last figure. This is now a minor point of the paper. We leave out to explain why we get about 100 INPs per liter in the case of ABIFN and only 50 per liter in the case of DIN. We may do that in follow up papers.

Line 333: The units of ICNC is wrong

This part of the text is removed in the revised version.

Line 352: “Basis” should be “Basin”

Improved!

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 this, 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.

We do not understand the requirement here! You mean INPC, or you mean ICNC? Disregarding this special point, an uncertainty discussion is not easy in this field of observations, parameterizations, and simulations with focus on ICNC and INPC. The uncertainty for each approach is usually large (within 1-2 orders of magnitude), and not just 20-50%, if one would apply a rigorous uncertainty analysis.

However, the comment triggered us to provide more information about uncertainties in the INPC computations (in Sect. 3.1 and in the conclusions, Sect. 5). To our opinion, consistency checks and closure experiments are usually the only ways to obtain trustworthy conclusions for field observations by combining the observations with supporting simulations and estimations/parameterizations. All this is now briefly outlined in Sect. 3.1 and 5.

The goal of the paper is clear. We present observations that show a strong link between smoke layer occurrence and ice nucleation occurrence. That is the main message of the manuscript! All the simulations and estimations are given to provide some numbers in terms of INPC. And these simulated (estimated) INP number concentrations are reasonable, disregarding whether the uncertainties are within 1 order of magnitude. In follow-up papers we plan to combine estimations of INPC (from lidar observations) and estimations of ICNC (from combined lidar-radar observations) in the framework of closure studies as presented in Ansmann et al. 2019b (in the case of dust-cirrus interaction). This is planned in the case of our MOSAiC observations (Arctic smoke-cirrus observations) and Punta Arenas field campaigns (cirrus evolution in Australian wild fire smoke). But a cloud radar was not available at Limassol in October-November 2020.

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.

Yes, we moved from RH (we leave that for relative humidity over water) and RH_{ICE} in Figs. 3, 5, 6, and 7.

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.

Improved!

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.

Improved! Apparent redundancy arises from the fact that a short explanation of shown curves, lines, bars, and areas is given in the first part. In the second part, the specific information and references are given to each set of curves, lines and areas.

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.
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- 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.

As new references we consider Chou et al. (2013), Kilchhofer et al. (2021), and DeMott et al. (2015) in the revised version.