

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.

## General comments

In this paper the effects of aged smoke particles on ice nucleation are discussed. The authors study the case of 27<sup>th</sup> November to 3<sup>rd</sup> November 2020, when a smoke layer was detected at the UTLS region over Cyprus. Based on their calculated backwards trajectories the authors find that this layer originated from wild fires over North America (California). Observations of cirrus formation, virga structures and cirrus originating from gravity waves are carried out by means of active remote sensing via lidar. Additionally, simulations of the gravity waves are carried out.

The topics discussed in this paper are in the scope of ACP and the interest of its readers. The authors tackle an interesting subject and manage to successfully answer the set scientific questions. Apart from the introductory part feeling a bit segmented the manuscript is well written. The reader is introduced to the topic and the subject at hand. The methodology can easily be followed. The results and discussion are clear and well accompanied by references. Nevertheless, a list of mostly minor and technical revisions is presented in the following.

## Specific comments

- Lines 25-27: At what altitudes did the measurements take place? Does this refer to the UTLS?

**We are more precise now and mention: free troposphere up to the tropopause.**

- Lines 41-47: It would be good if some papers are referenced regarding these claims.

**We rearranged the text to keep the introduction as short as possible, and with clear focus of smoke on cirrus impact. By this rearrangement, we avoid to provide references to all these points because we did already an extended review on smoke transport, aging, and resulting changes in the physical and chemical properties with all the necessary references in Ansmann et al. (2021) and do not want to repeat all this here.**

- Lines 48-49: Are there any statistics supporting this assumption? An explanation could be added or a paper could be cited at this point supporting the claim.

**There are no statistics! There are several airborne observations (e.g., Dahlkoetter et al. 2014) that point to a core-shell structure. In addition, lidar observations (depolarization ratio) show that the smoke particles are not far away from having a perfect spherical shape. We rearranged the text to meet your comment better.**

- Line 65: A one-sentence explanation of the activation thresholds would be helpful before this statement.

**We removed the respective discussion (and our confusing hypothesis) to keep the introduction short. This discussion is not needed.**

**We mention that these minerals may cause smoke particles to be activated even at high temperature (-13°C). But we do not want to extend speculation here too much. It is just a hypothesis.**

- Line 67: 'Those INPs' refers to the minerals?

**We change the text... But yes, ... minerals.**

- Lines 139-141: Is there an estimation of this potential bias? Studies have shown high water supersaturations even at cirrus-free conditions. Maybe looking into the available water vapor on the measurement period at UTLS would strengthen or weaken this point especially since you have RH data from the radiosondes.

**We improved this point after checking the radiosondes for cirrus events and cirrus free events in the upper troposphere. The uncertainty (bias) is at all less than 10% as stated in Sect.3.**

- Figure 1: The authors could specify what the uncertainty ranges are.

**Is done! We provide relative uncertainties in the text and figure captions.**

- Figure 1: Do the authors have an explanation to offer about the peak in PLDR at 13.5km altitude?

**This is the result of vertical smoothing of very noisy data! We state that at the end of Sect. 4.1!**

- Line 204: Is that the RH with respect to water or ice?

**We now introduce RH as relative humidity over water and  $RH_{ICE}$  as relative humidity over ice. In the figures we now show exclusively  $RH_{ICE}$ .**

- Line 211 and Fig.3: Since the RH in Fig. 3 does not reach supersaturation I would expect that the authors are using relative humidity with respect to water (RHw). This is not necessarily wrong but would not be advisable for the study of ice crystals/cirrus. If available please use relative humidity over ice (RH<sub>i</sub>).

**Agreed and Improved!**

- Line 217: Strong ice nucleation is rather the explanation of the formation and evolution of the virga rather than an observation. Please rephrase accordingly.

**Done!**

- Line 240: A quantification of the good agreement would be helpful.

**We now explicitly add the AOD values observed by AERONET. The AERONET AODs are 0.05-0.1 larger than the smoke layer AOD values. Thus, the lower troposphere (below 5 km) contributed 0.05-0.1 to the overall AOD, the rest is from the smoke layers.**

- Line 249: Same as Line 211, RH<sub>i</sub> would be preferable.

**We generally switched from RH (relative humidity over water) to  $RH_{ICE}$  (relative humidity over ice) throughout the result section.**

- Line 254: Same as Line 217

**We removed the paragraph with this statement.**

- Figure 4: Similar to above. 'Ice nucleation is expected at the top of the ice virga'.

**Improved!**

- Figures 5, 6 & 7: Consider Using RH<sub>i</sub> instead or RHw.

**Done!**

- Lines 268-269: How do the authors come to this conclusion?

**To avoid a lengthy, speculative discussion (conclusion), we removed this statement (on updraft speed).**

- Line 270: Using the RHi and nucleation thresholds for the available INPs would strengthen this claim. Some INPs activate already at very low supersaturations while others need high values. Having the RHi as a reference would be beneficial.

**We provide a simple gravity wave simulation (in a new Fig.8), directly after this statement! ... with temperatures and  $RH_{ICE}$  or  $S_{ICE}$  values from the radiosonde on 1 November 2020. This keeps the discussion simple and explains reasonably well what supersaturation levels were needed to explain the gravity wave observation on 1 November.**

**We do not like the idea to discuss different ice activity efficiencies of particles of the same aerosol type in this paper with the main goal: lidar observations show that smoke initiated ice nucleation. Such points may be discussed in follow-up papers.**

### Technical corrections

- Line 3: Patterns instead of pattern

**We changed the abstract text and removed pattern.**

- Abstract: Sentence 'Our study... to Cyprus' could be moved one sentence earlier, before 'we found... cirrus layers'. Introducing first the study before referring to results.

**Improved!**

- Line 18: 'was transported' instead of traveled

**Improved!**

- Line 20: 'in the future'

**Improved!**

- Line 22: 'fire storms'

**We removed this paragraph in order to have a more compact introduction.**

- Line 23: 'source of smoke'

**We removed this paragraph...**

- Line 30: 'with in-depth'

**Improved!**

- Lines 30-31: 'has already been shown'

**We removed the respective sentence to keep the introduction short.**

- Line 39: aged smoke particles originating from fires'

**We rephrased this part of the text.**

- Line 55: 'INPs' not necessary

**We rephrased all text parts with ... DIN INP ... accordingly....**

- Line 57: 'the'

**Improved!**

- Line 57: Here it is probably meant 'When the smoke particles take up supercooled water'

**Improved!**

- Line 59: 'completely dissolve and become liquid (and no insoluble material within the particles is left), homogeneous freezing will take place on the resulting aqueous solutions at temperatures below  $-38^{\circ}\text{C}$ '

**Improved!**

- Line 58: warmer instead of higher temperatures to avoid potential confusion with negative values

**Meteorologists do not like 'warmer' temperatures! We removed the sentence!**

- Line 75: Remove 'here'

**The sentence is removed.**

- Line 75: Replace 'were generated' with 'formed'

**The sentence is gone.**

- Lines 80-81: A new sentence for each section description. The first letter after the section number does not need to be capital

**All this is gone.**

- Line 86: 'are presently'

**We changed the text.**

- Line 156: Not clear what is meant. Please rephrase

**Improved, we provide an example.**

- Line 178: Observations & Discussion

**Improved!**

- Line 183: 'Figure 1 contains'

**Improved!**

- Figure 1 legend: It would make it easier to define every panel separately

**Improved!**

- Figure 1 legend: 'line in (f) represents the temperature'

**We changed the text.**

- Line 228: remove 'of'

**Improved!**

- Line 279: 50-100 m/s probably.

**You mean line 269? However, these numbers are gone at all. We removed the statement.**

- Line 310: Does this still refer to RH<sub>ice</sub>?

**We removed the respective paragraph.**

- Figure 8: RH could be denoted as RH<sub>w</sub> for clarity

**We use RH and RH<sub>ICE</sub> throughout the paper now.**

- Figure 8: Consider changing colors of naphthalene and fulvic acid. They are not easily distinguishable

**Improved!**

- Line 321: remove 'now'

**We changed the text.**

- Line 329: Remove 'not'

**Improved!**

- Line 331: Clarify if RH<sub>ice</sub>

**Done!**

- Figures 9-11: Could be unified in one figure for easier intercomparison

**Improved! We now show just one Figure (Fig.10, for 28 October). We leave out to show very similar figures for 30 October and 1 November in the revised version.**

- Line 352: 'Mediterranean basin'

**Improved!**