

Review

Measurement report: Ice nucleating particles active ≥ -15 °C in free tropospheric air over western Europe by Conen et al.

Summary

The study presented by F. Conen et al. reports INP concentrations at -10 °C and -15 °C at the High Altitude Research Station Jungfraujoch, a site which is located in the lower free troposphere. The measurements are based on 24-hour collected filters throughout several years, and focus on sampling intervals when the site was not impacted by boundary layer air. By performing heat treatment tests, it is suggested that the majority of INPs in this temperature regime are microorganisms at Jungfraujoch. While this is an important finding and good motivation for future studies, I have some concerns about the methods used and the conclusions drawn from them.

General remarks

- One of the key points in your study is the distinction between free tropospheric conditions and boundary layer intrusion. However, you use only one method to quantify this, namely the radon concentration. In the study by Herrmann et al. (2015) it was found that “local radon emissions as well as the comparatively long radon lifetime blur the distinction between free tropospheric conditions and boundary layer influence. This further supports the conclusion that the CO/NO_y approach offers the best distinction between free troposphere conditions and boundary layer influence.” Is there a reason why you did not consider the ratio between CO and NO_y to identify free tropospheric conditions, e.g., in addition to the Radon concentration?
- The identification of Saharan dust impact is solely based on “high PM₁₀ loads” (lines 80 – 81). Is there a study showing that such measures can be used to determine Saharan dust events at Jungfraujoch? If not, you need to provide more results on this approach, as for example a comparison to other methods used to determine Saharan dust impact, such as aerosol particle scattering properties (Collaud Coen et al., 2004), occurrence of larger particles (e.g., Kammermann et al., 2010) and back trajectory analysis.
- As discussed by the authors, the 24-hr filter collection are limited in their use to distinguish free tropospheric from disturbed free tropospheric conditions. Did the authors consider performing shorter filter sampling times?
- The study comprises only 28 datapoints of INP concentrations, which is not much to draw general conclusions about seasonal variability.
- You do not present the freezing spectra but only INP concentrations at specific temperatures. It might be interesting to the reader to see those spectra, e.g., in the appendix.

Abstract

Lines 7 – 8: The statement “... typically starting a few km above ground” is not correct. Ice formation in mixed-phase clouds can occur at ground level; e.g., Jungfraujoch is per definition a ground-based station and primary ice formation frequently occurs there (e.g., Mertes et al., 2007), which is also true for other locations in mountains. Thus, ice formation in clouds is rather a question of temperature, supersaturation, and presence of INPs, not of altitude above ground.

Lines 16 - 18: I am struggling with this last statement. Jungfraujoch is a site which can be impacted by boundary layer intrusions, which can occur on short time scales. How certain are you, that you excluded such potential boundary layer intrusions from your 24 hr filters (see also my comment above)? In addition, Jungfraujoch is impacted by touristic activities (e.g., smoking), and your 24-hour filters are impacted by this. Has one filter been sampled without any impact from touristic activities, e.g., during a lockdown in 2020 due to the COVID pandemic? Generally, to make such a strong statement, more of such measurements should be conducted with shorter filter collection times (to

better avoid boundary layer intrusions or emissions from touristic activities), and at other free-tropospheric sites over central/Western Europe to confirm this. Your study is a good motivation for such future research.

I recommend to specify the time period during which you collected your samples (months, years).

Introduction

Lines 22 – 24: Please check the grammatic of this sentence. I am unsure if I understand the sentence correctly. Do you mean that this community of airborne microorganisms have their emission sources within several tens of kilometers (local/regional scale)?

Line 27: The statement regarding altitude for mixed-phase cloud occurrence is not correct, see my comment before.

Line 28: Please add more citations to this very important statement. E.g., many airborne INP measurements are conducted in the free troposphere, and there are more free-tropospheric measurements sites where INP studies were conducted.

Lines 32 - 34: The cited percentages refer to ice particle residuals, not to the INP concentration measurements at -31°C.

Lines 35 – 36: I recommend to also include citations from ice particle residual analysis performed at Jungfraujoch (e.g., Mertes et al., 2007; Kupiszewski et al., 2015; Schmidt et al., 2017; Lacher et al., 2021).

Material and methods

I recommend to specify the time period during which you collected your samples (months, years) and to refer to the respective table in the appendix.

Line 52: What is the error associated to a counting by eye?

Lines 71: I recommend to also cite Herrmann et al. (2015) here.

Lines 80 – 81: Please provide more information about the method used to identify impact from Saharan dust event. Which threshold has been used? Has such a method been established in previous work, resp., has it been validated against other measurements or back trajectory calculations (see also my comment above)?

Line 82: It is not clear to me why you analyzed the median radon concentration here, while before (line 76) the mean radon concentration was used to identify free tropospheric conditions.

Results and discussion

Lines 128 – 135: Your discussion is based on the assumption that during Saharan dust events all aerosol particles are dust particles. With the methods used here you cannot quantify the fraction of dust particles in the overall particle population.

Lines 151 – 153: During Saharan dust events, the total number concentration of aerosol particles does not necessarily increase; more important here would be to quantify the concentration of dust particles and compare it to an increase in INP concentration.

Technical

Line 27: I assume you mean “-5 °C”.

Line 30: It should read “-31 °C”.

Line 45: I assume it should read “At the site,...”?

Line 60: “Sample” instead of “Samples”?

References

Collaud Coen, M., Weingartner, E., Schaub, D., Hueglin, C., Corrigan, C., Henning, S., Schwikowski, M., and Baltensperger, U.: Saharan dust events at the Jungfraujoch: detection by wavelength dependence of the single scattering albedo and first climatology analysis, *Atmos. Chem. Phys.*, 4, 2465–2480, 10.5194/acp-4-2465-2004, 2004.

Kammermann, L., Gysel, M., Weingartner, E., and Baltensperger, U.: 13 month climatology of the aerosol hygroscopicity at the free tropospheric site Jungfraujoch (3580 m a.s.l.), *Atmos. Chem. Phys.*, 10, 10717–10732, <https://doi.org/10.5194/acp-10-10717-2010>, 2010.

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Lacher, L., Clemen, H.-C., Shen, X., Mertes, S., Gysel-Beer, M., Moallemi, A., Steinbacher, M., Henne, S., Saathoff, H., Möhler, O., Höhler, K., Schiebel, T., Weber, D., Schrod, J., Schneider, J., and Kanji, Z. A.: Sources and nature of ice-nucleating particles in the free troposphere at Jungfraujoch in winter 2017, *Atmos. Chem. Phys.*, 21, 16925–16953, <https://doi.org/10.5194/acp-21-16925-2021>, 2021.

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