

Replies to reviewer #2

We thank the reviewer for the careful reading of our manuscript and the positive comments. In the revised version, we have addressed all the comments of the reviewer, as detailed. below:

The reviewer's comments are provided in black and our replies in blue.

This study by Chatziparaschos et al. used a 3D chemical transport model to predict global concentrations of ice nucleating particles based on an emission model accounting for quantities of quartz and feldspar emitted and parameterisations of the ice nucleating effectiveness of quartz and feldspar. The results of this process are shown to agree reasonably well with field measurements of ice nucleating particle concentrations. The study concludes that the high abundance of quartz particles means that their contribution to INP concentrations is significant, despite the lower ice nucleation ability of quartz compared to 'K-feldspar'. Conclusions are also drawn about the contributions of the different minerals to low and mid-level clouds. The study is interesting and appears to be well-conducted. I have a few suggestions below but support publication once these are considered. While mostly well-written, the study would benefit from thorough proofreading. I have highlighted various issues below but there are likely more.

General comments:

I think it is worth noting that a great deal is still not known about why quartz and feldspar nucleate ice. In the absence of proper physico-chemical understanding of how these minerals nucleate ice the parameterisations of Harrison et al. remain entirely empirical. It may still turn out that they are not representative for unforeseen reasons. This doesn't detract from the study but is probably worth mentioning briefly. Relatedly, I think a few words clarifying why it is alkali feldspar (I think use of K-feldspar throughout is fine, although worth noting that some Na-rich feldspar also nucleate ice well), rather than plagioclase feldspars, that are important may be helpful e.g (Kiselev et al., 2021;Whale et al., 2017;Harrison et al., 2016).

Discussion has been added, and the reference by Harrison et al. (2016) has been introduced:

"More recently, it was shown that among dust minerals, alkali feldspars, in particular the potassium feldspars, nucleate ice more efficiently than feldspars in the plagioclase series, which contain calcium (Harrison et al., 2016). Also, sodium-rich feldspars are very ice-active but were found to lose their ice-activity with time compared to K-feldspars (Harrison et al., 2016), thus, K-feldspar is the most efficient INP. Note, however, that little is known about the processes governing the IN capability of these minerals, so more future work is needed to unravel their effect and contribution for all possible ice formation conditions (Burrows et al., 2022)".

The abbreviation INP, is used in various ways throughout. While what is meant is mostly clear it would probably be best to pick a definition and stick with it.

We now use INP for particles having the potential to activate and form ice at a specific temperature, and once activated, we talk about ice crystals.

Suggest checking the format of references throughout the text, there is some variation.

We have checked and corrected the format of the references.

Specific comments

Line 23 – INP usually abbreviates Ice Nucleating Particle, with an 's' added for 'Ice Nucleating Particles'

Done.

Line 23 is confusing. Ice nucleation is the first step of ice formation, remove reference to 'homogeneous formation'

This sentence has been rephrased as follows: "Ice formation is enabled by Ice Nucleating Particles (INPs) and can profoundly affect the microphysical and radiative properties, lifetimes, and precipitation rates of clouds."

Lines 25 – The word 'efficient' implies a ratio. 'Effective' or similar would be better here.

"Effective" is now used.

Line 26 – It isn't clear what is meant by 'ice nuclei' (Vali et al., 2015). 'Ice nucleation activity' would be more typical I think.

Done.

Line 34 – I wonder if it might be possible to briefly explain why differences are seen between cloud regimes? This isn't really discussed in the text either. What causes the differences between quartz and k-feldspar as regards the cloud types they influence?

The relevant sentence in the abstract has been modified as follows:

"Our results show that although K-feldspar remains the most important contributor to INP concentrations globally, affecting mid-level clouds, the contribution of quartz can also be significant. It dominates at the lowest and the highest altitudes of dust-derived INP, affecting mainly low-level and high-level mixed-phase clouds."

And the relevant discussion in section 4: "However, at lower altitudes and below -12°C , low $[\text{INP}]_{\text{total}}$ concentrations ($<10^{-2}\text{L}^{-1}$) are calculated and are mainly derived from quartz dust particles (60%) (Figure 8b). This outcome is attributed to quartz's high number concentration between $30\text{--}40^{\circ}\text{N}$ at and below 700 hPa, partially associated with higher local quartz than feldspar emissions from Asian dust sources (Claquin et al., 1999; Nickovic et al., 2012). At temperatures below -25°C , the quartz contribution becomes increasingly important with increases in INP concentration when the temperature decreases, reaching up to 50 % at -35°C (Figure 8b). These findings agree well with Ilić et al. (2022) and Boose et al. (2016), who showed that quartz could significantly contribute to $[\text{INP}]$ at temperatures between the homogeneous freezing limit and -33°C . Overall, $[\text{INP}]_{\text{quartz}}$ dominates at the lowest and the highest altitudes of dust-derived INP (Figure 8b; reddish colors). This is clearly demonstrated in Supplementary Figure S6 for Eurasia where at the range of 700-900hPa and around 450 hPa model pressure levels the contribution of $[\text{INP}]_{\text{quartz}}$ to total ice crystals from immersion freezing on dust particles exceeds 60%. Figure S7 shows that in the South Hemisphere quartz

contribution to total INP is about 40%. Consequently, $[INP]_{\text{feldspar}}$ is expected to affect mid-altitude clouds, while $[INP]_{\text{quartz}}$ is expected to affect both the low-altitude clouds and the high-altitude cold clouds..'

See also the discussion of Figure 1 provided above, that explains how the combined effect of mineral concentration and its active sites at different pressure and temperature levels changes with height.

Line 71 – '....concentration of INP' maybe?

Modified as suggested.

Line 86 – Doesn't read well if INP means 'Ice nucleating particles'

This sentence was rephrased to address the comments of the other reviewer.

Line 89-90 – Kiselev et al. was in 2017 I think?

The reference was corrected.

Line 105 – The singular approximation assumes that each droplet contains a single ice nucleating particle active in a given temperature interval. I don't think the statement regarding site density is necessarily true.

This sentence was rephrased to include the reviewer's comment: "This approach adopts the so-called singular hypothesis that assumes that each droplet contains a single ice nucleating particle active in a given temperature interval and the time dependence (stochasticity) of nucleation is of secondary importance (Vali et al., 2015)."

Line 125 – may be worth briefly noting that solution environment may well substantially impact the ice nucleation effectiveness of both quartz and feldspar e.g. (Kumar et al., 2019;Whale et al., 2018;Klumpp et al., 2022;Whale, 2022).

We added further explanation following the above suggestion: "In addition, the solution environment may substantially impact the ice nucleation efficiency of both quartz and feldspar (Whale et al., 2018; Kumar et al., 2019), but the study of the impact of this process on INP is beyond the scope of the present study."

Line 187 – I wouldn't call the samples used 'soil'. Mostly they are mineral samples that have been selected for purity. The point that the samples may not be representative of atmospheric conditions is very true however.

We have replaced "soil" with "mineral" as suggested by the reviewer.

Line 339 – I don't think Spracklen and Heald looked at marine organic aerosol?

Spracklen and Heald's ref was misplaced. The sentence now reads: "These differences between model results and observations could be attributed to several factors, such as the omission of the contribution of marine organic (Wilson et al., 2015) and terrestrial biogenic aerosols (Spracklen and Heald, 2014; Myriokefalitakis et al., 2021)."