

Referee#1 comment RC2

This study investigates the ice nucleation characteristics of two samples of Asian dust collected from the west coast of Japan. Particles with diameters $> 1.1 \mu\text{m}$ were deposited on a hydrophobic Si waver using an impactor. The ice nucleation ability of the collected particles was investigated in immersion freezing mode in a closed cell mounted onto a cold stage of an optical microscope. The ice nucleating particles (INPs) together with a number of inactive particles were characterized first with AFM and micro-Raman spectroscopy followed by SEM-EDX. For comparison several reference samples were investigated with the same methods. This study presents a new approach to characterize atmospheric INPs on an individual particle basis. It is a welcome complement to other combinations of methods used to characterize atmospheric INPs on a single particle basis. It shows once more how complex the composition of aged aerosol particles in the coarse mode is, rendering a clear classification of particles difficult. Nevertheless, the authors succeeded to identify physical and chemical characteristics that increase or decrease the ice nucleation ability of the particles. I recommend this paper for publication in ACP after revisions. Specifically, the experimental procedure needs to be explained in more detail. For the validation of the method, the reference samples need to be analyzed more quantitatively and compared more thoroughly with available literature. Moreover, more references need to be added to the introduction and the formulation of some sentences needs to be improved (see specific comments below).

The authors would like to thank the reviewer for providing positive comments and constructive suggestions by thoroughly going through the manuscript. As suggested by the reviewer, we added more details on the experimental procedures. We have conducted additional and thorough analysis on the reference samples to validate our method, and quantitatively compared the results with the available literature. Please find our response to each of the comments below (in blue italic).

Page 1, line 19: what is meant by “slower”? At lower temperature?

We changed the corresponding sentence as follows:

‘Meanwhile, most of the IN active atmospheric particles formed ice below $-28\text{ }^{\circ}\text{C}$, i.e. at lower temperatures than the standard mineral dust samples of pure components.’

Page 2, line 9: reference to Murray et al. (2012) only is somewhat arbitrary. Add more references or “e.g.” in front of the reference to the Murray et al.review..

We added Murray et al., (2010) and Rosenfeld and Woodley, (2000) as the references which also reported that the homogeneous nucleation takes place below -37°C .

Page 2, line 21 – 22: This sentence needs to be reformulated.

We changed the corresponding sentence as follows and added few more references:

'Based on these results, while mineral dust and biological particles are generally regarded as efficient ice nuclei (Morris et al., 2004; Connolly et al., 2009; Niemand et al., 2012), ice nucleation within mixed phase clouds involving soot and organic particles is still not as clearly demonstrated due to the diverse chemical composition and different experimental conditions (DeMott, 1990; Kireeva et al., 2009).'

Page 2, line 23: “sea salt and sulfates are often not considered as efficient ice nuclei.”: This is not generally true for sulfates. Ammonium sulfate is found to be ice nucleating at lower temperatures as is also stated later in the manuscript by referring to e.g. Abbatt et al. (2006). The sentence here needs to be corrected, e.g. by adding “under mixed phase conditions”.

Indeed, we should state more clearly that we are referring specifically to the mixed phase conditions here.

We made the following change as suggested.

'On the other hand, sea salt and sulfates are often not considered as efficient ice nuclei under mixed phase conditions.'

Page 2, lines 30 – 31: This sentence needs to be improved.

We changed the corresponding sentence as:

'This enabled direct and detailed investigation on the particles representative of the actual deposition mode ice nuclei in cirrus clouds.'

Page 3, lines 1 – 3: this statement should be supported by more recent references.

We added Prenni et al., (2012) and Ardon-Dryer and Levin, (2014) as the references.

Page 3, lines 3 – 5: this statement should be supported by a reference.

We added Korolev, (2007) as the reference.

Page 3, line 12: which “above mentioned artifacts” are meant here? Please, be more explicit.

We made the following changes to be more explicit:

'However, they also reported discrepancies between the results obtained from the three different sampling techniques, and attributed them to potential bias arising from the artifacts such as the possible generation of particles within the instruments and the inherently scarce number of ice nuclei in the atmosphere.'

Page 3, line 20: more than just one reference should be given here to support this statement.

We added Zhang et al., (2003), Sullivan et al., (2007), and Iwasaka et al., (2009) as the references.

Page 3, lines 21 - 22: again, more than one reference should be given to support this statement.

We added Maring et al., (2003) as the reference.

Page 4, lines 25 – 28: Was the dew point kept constant at -6 to -3°C during cooling? Also, the degree of dilution of the particle should be estimated based on the size of the developing droplet. This is important to judge whether a freezing point depression as discussed later on in the manuscript is relevant at all.

The air supply to the chamber is cut during the cooling phase, so the dew point of the supplied air is not actively controlled or monitored during this time. Nonetheless, as the course of the cooling down to -30°C, the remaining water vapor in the chamber is expected to be maintained slightly above the saturation. This is consistent with the fact that the growth in particle size due to condensation can be visually observed.

The dry and wet particle diameters before and after the condensational growth are shown in Table S-1.

The degree of dilution was estimated based on the growth rate. As a result, the average concentration of the solution droplets of the atmospheric particles were estimated to be approximately 0.074 g/ml.

Table S-1: The diameters and volumes of the particles before (dry) and after (wet) the condensational growth. Corresponding concentrations of the sample and test solute particles in the solution droplets are also shown. Here, D_{dry} and D_{wet} denote the circle equivalent diameters obtained from the 2D silhouette of the particles in the microscopic images taken before and after (at approximately -25 °C) the cooling experiment, respectively. The number of the particles in each sample observed by the microscope is shown as n . V_{dry} is the sphere equivalent volume calculated from the corresponding D_{dry} . V_{wet} was calculated by assuming droplets having contact angle of 110° relative to the substrate. The droplet volumetric growth factor GF was determined by the ratio of V_{wet} relative to V_{dry} . By assuming following densities (sampled particles: 2.00 g/cm³, NaCl: 2.16 g/cm³, Ca(NO₃)₂: 2.36 g/cm³), The calculated mass and molar concentrations of the droplet are shown in terms of m and M , respectively. The test solute particles of NaCl and Ca(NO₃)₂ were aerosolized by atomizing their solutions (0.005g/ml) and collected on the substrate with an impactor.

	Sampled particles (n = 144)	NaCl (n = 97)	Ca(NO₃)₂ (n = 102)
D_{dry} (μm)	3.9 ± 0.8	4.8 ± 1.4	4.0 ± 1.0
D_{wet} (μm)	9.2 ± 1.8	14.7 ± 4.7	12.9 ± 2.5
V_{dry} (μm³)	4.4 ± 2.2	9.0 ± 7.6	4.9 ± 2.2
V_{wet} (μm³)	156.9 ± 75.0	750.0 ± 677.0	429.2 ± 191.0
GF	43.9 ± 25.0	94.3 ± 55.7	123.7 ± 101.4
m (g/ml)	0.074 ± 0.112	0.029 ± 0.015	0.024 ± 0.009
M (mol/l)	-	0.49 ± 0.25	0.15 ± 0.05

Page 5, lines 26 – 27: This sentence needs to be improved.

We changed the corresponding sentence as follows:

‘Both the IN active and non-active particles collected in the atmosphere were analyzed on an individual particle basis using a series of microscopic techniques. Firstly, an AFM (CombiScoper™ 1000, AIST-NT, Inc., USA) was used to characterize the three-dimensional morphology, followed by micro-Raman spectroscopy to detect surface chemical compounds.’

Page 7, lines 11 – 16: Here, the freezing onset temperatures of the reference samples are compared with literature (Atkinson et al., 2013) and it is concluded that they are consistent. However, this comparison does not take the surface area present in the samples into account. The comparison needs to be based on the surface area present in the investigated reference sample of this study multiplied with published ice nucleating active sites (INAS) surface densities e.g. from Peckhaus et al. (2016) or Harrison et al. (2016) for feldspars. Moreover, It should be specified what frozen fraction was taken as the onset condition.

We have to admit that our technique is not a perfect method that can resolve all technical issues encountered in the ice nucleation experiments. The biggest advantage of course is that we can keep track and be sure which single particle was actually nucleating ice. In return, we have to sacrifice the accuracy and quantitative evaluation of freezing temperatures based on INAS in relation to the evaporation of droplets in the vicinity of the frozen particles (Bergeron-Findeisen effect), as well as due to the limited number and size ($d > 1.1 \mu\text{m}$) of the particles that can be analyzed.

The number of ice crystals formed are compromised especially at lower temperatures as the number and size of ice crystals increase and more droplets in the vicinity of the growing ice are subjected to

evaporation. When comparing our results with those found in literature, we must also note that strict comparison is difficult because of the uncertainties and differences related to the size distribution and composition of dust particles used in different experiments. Assumptions made to calculate the surface area can also be a source of bias. The K-feldspar used in this study as a reference mineral dust was purchased from the same supplier, but the lot number is different (i.e. different source rock) from the K-feldspar used in Atkinson et al (2013). Therefore, composition of K-feldspar used in the current study and that used in Atkinson et al. (2013) are not exactly the same. Nevertheless, we added the ice nucleation active sites (INAS) of the 4 standard mineral particles measured by the current IDFM (Fig. S-2).

The ice nucleation active sites (INAS) obtained in this study agreed within an order of magnitude difference compared to those found in the literature (despite many different experimental conditions). Further, we would like to emphasize that the relative order of the onset temperatures found for different particles is always consistent (i.e. K-feldspar > Na-feldspar > quartz > kaolinite >> pure water) and even the range of the freezing temperatures are not far off from the reported values.

It was probably not clearly stated in the manuscript, so the following change was made with regard to the onset temperatures:

‘Heterogeneous ice nucleation observed in all standard mineral samples tested in this study (K-feldspar, Na-feldspar, quartz, kaolinite) consistently occurred at higher temperatures than the homogeneous freezing temperature. The reference mineral samples were milled to fine grains before being collected on Si wafer substrate by an impactor. Three set of samples were made for each reference mineral to ensure large enough observation area for the IDFM. The total number of the particles monitored during the ice nucleation experiment by IDFM was 4,509, 2,271, 4,759, and 1,435 particles, respectively. In this study, the freezing onset temperature of the sample was defined as the temperature at which the IN active fraction of the total observed particles reached 0.01. As a result, the freezing onset temperatures for K-feldspar, Na-feldspar, quartz, and kaolinite ranged between -22.2 to -24.2 °C, -24.7 to -25.7°C, -24.8 to -26.8 °C and -27.2 to -29.2 °C, respectively (Fig. S-1). Therefore, the ice nucleation activity of K-feldspar was the highest and that of kaolinite was the lowest. The order and the range of observed onset temperatures for these minerals were consistent with the results found in the literature (Atkinson et al., 2013; Marry et al., 2011).’

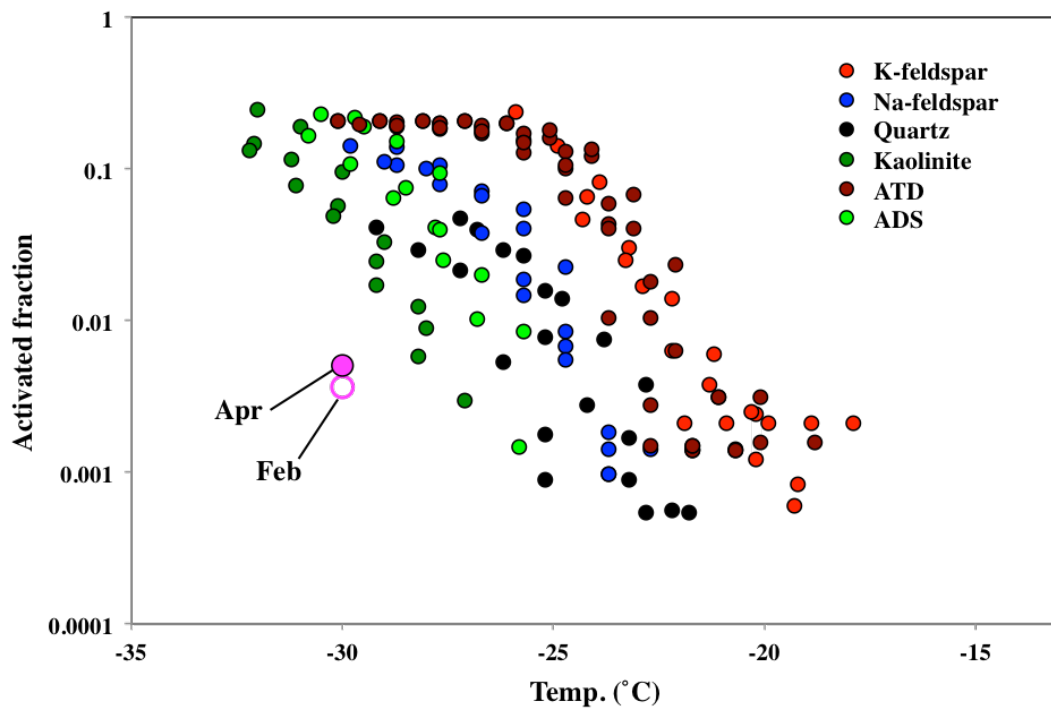


Figure S-1: Activated fractions of the reference mineral dust particles. Results of the atmospheric samples collected in February and April are also shown for comparison.

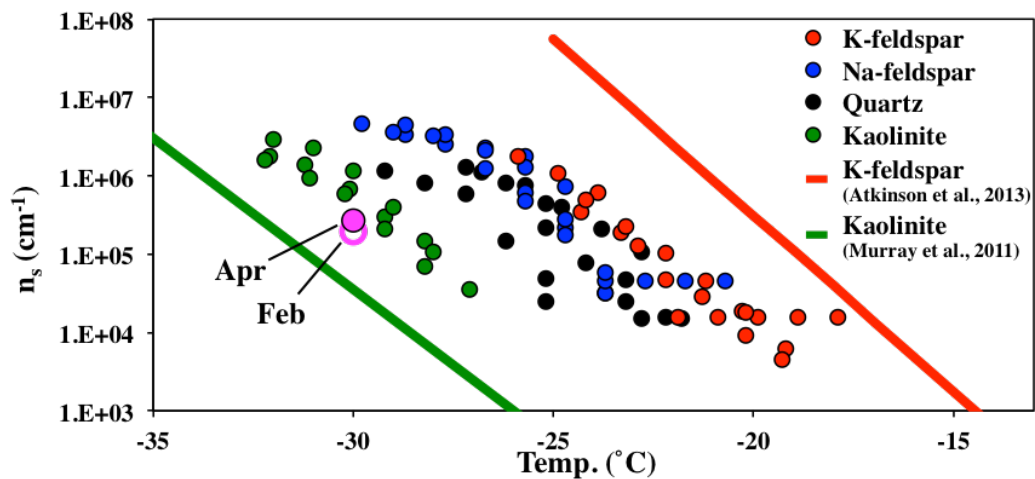


Figure S-2: the ice nucleation active site (INAS) densities for the reference single component mineral dust samples. These INAS densities were calculated from the activated fractions (Fig. S-1) and the averaged sphere equivalent surface areas obtained from the 2D silhouette of individual particles in the microscopic image.

Page 7, line 18: The ATD and ADS samples should be discussed in more detail. How many particles were analyzed? Comparison of freezing temperatures with relevant literature is needed.

In response to the above comment, we added the ice nucleation active sites measured for ATD and ADS, and compared with the natural dust sample found in literature (Niemand et al., 2012)

We also made the following change in the manuscript:

'For comparison, the freezing onset temperatures of the ATD and ADS were -22.1 to -23.7 °C and -25.2 to -27.2 °C, for 2,019 and 1,354 monitored particles, respectively.'

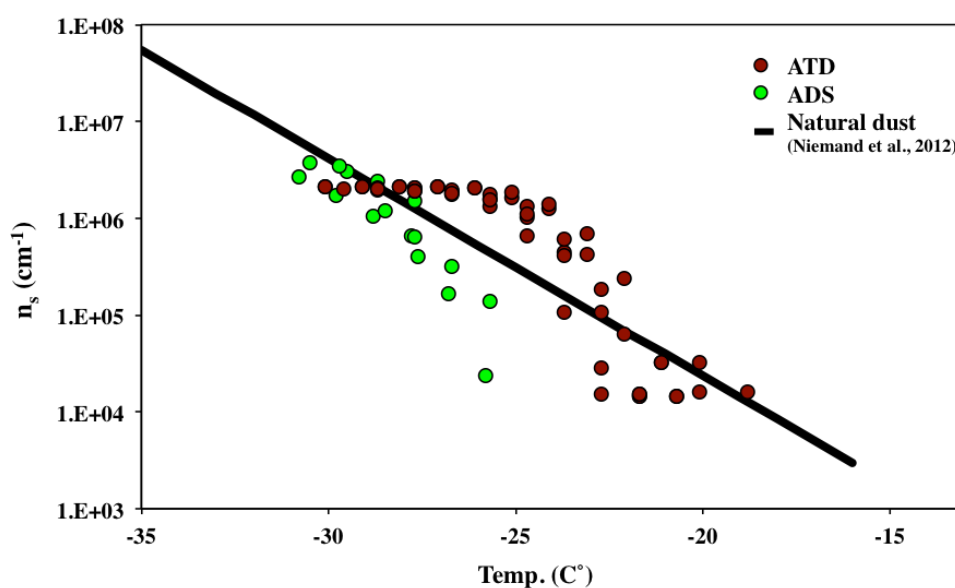


Figure S-3: the ice nucleation active site densities for ATD and ADS.

Page 7, line 19 – 21: the measurement procedure needs to be explained better. Was the same sample cooled repeatedly to investigate the ice nucleation activity of the 10,188 and 24,145 particles? If yes, what were the heating/drying conditions between the cooling cycles to remove the ice crystals?

No we did not repeat the freezing and heating/drying cycles though it would be an interesting experiment to show the robustness and reproducibility of the results. To minimize the impact of repeated activation on the particle properties, we limited it to be just one freezing and heating cycle. Besides, it is very labor intensive to go through that many particles for a repeated number of cycles.

The heating/drying condition is described in section 2.1 in the manuscript. We added following explanation in the manuscript:

'For ambient samples, we first determined the total number of target particles by analyzing the optical images recorded with x50 magnification. As a result, ice nucleation activity of 10,188 and 24,145

particles were monitored by the IDFM for the February and April samples, respectively. To avoid too many particles in contact or in proximity to each other in the same field of view, multiple samples were collected during the sampling period on separate silicon wafer substrates to gain a good total number of particles. ‘

Page 7, lines 22 – 23: Ice nucleation active fractions should also be given for the reference samples.

We added those results in Fig. S-1.

Page 7, line 24: specify “onset temperature” in terms of a frozen fraction.

We added the following description in section 3.1 as the definition of the onset temperature.

‘In this study, the freezing onset temperature of the sample was defined as the temperature at which the ice nucleation active fraction of the total observed particles reached 0.01.’

Pages 9 – 10, Section 3.4: The elements C, N, O and Si could not be identified by SEM-EDX. The influence of this restriction should be discussed more explicitly. It should be analyzed whether based on the Raman spectra, some particles might be dominated by organics. In general, it might be helpful to combine the results of the Raman spectra and the SEM-EDX analysis already in the results section and not only in the discussion section. Also, do the compositions derived from the two techniques support each other or are they in some cases contradictory? At the end of this section, it is stated that the Raman peaks are “not necessarily representative of a major component of the particle”. This is certainly true for large samples. However, in case of particles with diameters < 5 μm , the laser spot penetrates the whole sample. This is supported by the presence of Raman peaks from the substrate present in the spectra. Moreover, in the method section, it is stated that Raman spectra were taken “each 750 nm step”. This should lead to a full coverage of the particles in the x- and y-directions.

The restriction of not being able to take into account C, N, O and Si peaks are that we cannot detect organic containing particles by SEM-EDX analysis alone. As a result, if an organic dominant particle is present, it should be identified by the presence of organic related peaks in Raman and absence of other particular elements in the EDX spectra. This restriction however, does not lead to miscounting of mineral dust particles since even a quartz enriched particle can be detected with inclusions of small amount of Al. We added the following description in section 3.4.

‘Note however, that elements C, N, O, and Si were not taken into account in the EDX semi-quantitative analysis. Therefore, we cannot rule out the possibility that organic dominant particles may be overlooked by EDX analysis alone.’

In relation to the results obtained by Raman spectra and EDX analysis, we found that the compositions derived from the two techniques are generally complementary to each other rather than being contradictory. But it is difficult to obtain a 100% match because the Raman peaks depend on the amount, Raman active cross section and peak overlaps of the compounds. And we also cannot determine if the organic matter is the dominant component in a particle or not solely from the obtained Raman peaks. As pointed out, the following statement “not necessarily representative of a major component of the particle” and the related paragraph (Page 10, 1-9) is omitted from the text since it is true that we cannot prove it. We made following change in the manuscript:

‘Particle classification with SEM-EDX relies on characteristic X-ray signals, which are used to estimate the major elemental composition of a particle. This information can be considered to reflect the bulk elemental distribution within a particle and has been commonly used for particle classification in many previous studies. In contrast, micro-Raman spectroscopy detects slight shifts of wavelength in the scattered light that reflect the vibrations of molecular bonds specific to the compounds contained in a sample. Therefore the Raman spectrum was used rather to complement the bulk particle type classification by the EDX analysis, and to detect the internally mixed compounds.’

Page 10 – 12, Section 4.1: This section needs to be improved. The assignment of clay minerals is solely based on a fluorescence signal in the Raman spectra. However, fluorescence is also often taken as an indication for the presence of biological aerosol particles (e.g. Twohy et al., 2016). How can a biological origin of the fluorescence signal be excluded?

Actually, the assignment of clay minerals is not solely based on a fluorescence signal but also taking into account the subsequent SEM-EDX analysis on the same particles. Our conclusion is based on the fact that 87% of the fluorescent particles contained dust related elements such as Mg, Al, Fe.

Indeed, we are aware of the biological fluorescence and it is also stated in Page 8, 29-31 ‘Fluorescence from a particle is typically attributed to a certain group of organics of biological origin, or the intercalated impurities of humic or humic-like substances in clay minerals and amorphous alumino-silicates (Sobanska et al., 2012; Jung et al., 2014).’

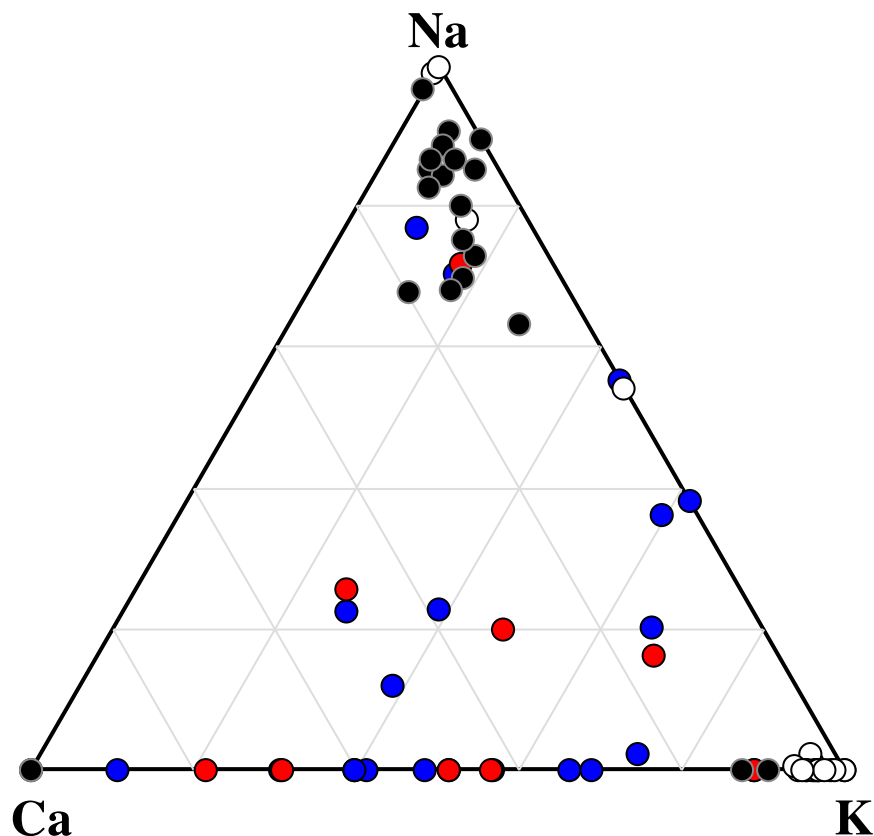
Possibility remains that part of the fluorescence results from internal mixing of biological matter attached to the mineral dust. To make it more clear, the following explanation is now added in the manuscript:

‘Subsequent SEM-EDX analysis showed that most of the fluorescent particles (87 %) contained elements that indicate a mineral composition. The fluorescence is also often taken as an indication for the presence of biological aerosols (Twohy et al., 2016), but considering the fact that all the ADS also showed similar

fluorescence, and the relatively small abundance of biological particles as compared to mineral dust in general (Huffman et al., 2012), these fluorescence signals are mostly associated with mineral dust (especially those enriched in clay minerals) rather than pure biological particles. Possibility remains however, that a fraction of the fluorescence signal results from biological matters attached to the clay minerals. In any case, both the SEM-EDX and micro-Raman analyses indicated that mineral dust particles act as efficient ice nuclei under conditions relevant for mixed phase cloud formation.'

Page 11. lines 5 – 6: An Al-K-(Ca+Na) ternary plot is not a typical way to discriminate K-feldspars from Na/Ca-feldspars. For this purpose a K-Na-Ca plot is usually used.

The ternary plot in Figure 7 is replaced with the K-Na-Ca plot. In any case, atmospheric particles are not gathering at a certain corner but they are rather scattered and show quite mixed characteristics.



Page 11, lines 6 – 7: Why should clay minerals and mica appear in the middle of an Al-K-(Ca+Na) ternary plot? The clay mineral kaolinite contains only Al but no K nor Ca/Na. It should appear therefore at the Al corner. The clay mineral illite contains Al and K but no Ca/Na. It should therefore appear on the Al-K line. The same with the common mica muscovite: it contains Al and K but no Ca/Na.

From our years of experience of SEM-EDX analysis on natural Asian dust aerosols, it is very rare to see a single component dust particle. It is commonly the case that several mineral components are internally mixed within a particle so that plots do not usually stick to Al corner or Al-K line.

Page 11, line 8: How was the presence of mica inferred?

According to Yabuki et al., (2003), mica appears in the center rather than along the Al-(Ca+Na+K) line of the ternary Al-(Ca+Na+K)-(Fe+Mg) plot. Most of our dust particles (both IN and non-IN active) also fall in the area where plots of mica, feldspars, clay minerals appear. Most particles do not show characteristic elemental composition of specific mineral types, which is another indication that particles are forming complex mineral mixtures.

Page 11, lines 17 – 19: Only one study is cited although the sentence starts with "Previous studies". Either give reference to at least one more study or change to "A previous study".

We changed to "A previous study".

Page 11, line 23: improve formulation, e.g. reformulate: “most IN particles active above -30°C.”

Reformulated accordingly to the suggestion as follows.

Furthermore, it was demonstrated that most IN particles active above -30 °C were mineral dust particles composed mainly of clay minerals, with or without minor mixing of other mineral components, that involve fluorescence, most likely as a result of defects and/or impurities (e.g. humic organics) in their crystal structure (Gaft et al., 2005; Jung et al., 2014; Sovanska et al., 2014).

Page 11, lines 23 – 26: The discussion of the origin of the fluorescence signal needs to be improved.

The fluorescence signal may come naturally from the crystal structural defect and/or the organic materials contained in the clay mineral particles. As discussed in section 4.4.(page 16, lines 12-16 in the original manuscript), we even found that all the fresh ADS (Asian dust particles believed to be dominated largely by clay minerals) were fluorescent and contained organic matter to some extent. Therefore the fluorescent particles associated with elements of crustal origin (by EDX analysis) were suggested as clay minerals. We made the following changes to be more explicit:

‘Furthermore, we even found that all the fresh ADS particles (believed to be dominated largely by clay minerals) showed the fluorescence. This fluorescence is likely derived from the defects and/or impurities (e.g. humic organics) in their crystal structure (Gaft et al., 2005; Jung et al., 2014; Sovanska et al., 2014). Therefore, the fluorescent particles associated with elements of crustal origin (by EDX analysis) were

regarded as clay minerals. It was also demonstrated that the most IN particles active above -30 °C were dominated by such fluorescence mineral dust particles.'

Page 12, lines 7 – 8: It should be stated here, that this number has a low bias because contributions from organics are missed in SEM-EDX. An estimate of the contributions of organics based on the Raman spectra might be added.

Following sentence is added to the corresponding section:

Note however, that this elemental fraction of sea salt components is based on SEM-EDX and does not reflect potential contribution from organics and therefore must be regarded as the upper limit.

Page 12, line 23: “at any event” seems a too strong conclusion considering that aged sea salt particles + inclusions were also found as a small fraction in the IN active particle fraction.

We changed the corresponding part as follows:

Based on the results of this study, we suggest that large and aged sea salt particles internally mixed with sulfates, nitrates, or organics are less likely to nucleate ice, although the possibility remains for ice nucleation by pure sea salt and sea spray organic particles in the atmosphere.

Page 13, line 30 – 31: “Therefore, the liquefied $\text{Ca}(\text{NO}_3)_2$ coating is expected to show strong molar depression of freezing point, which could explain their weak IN ability”. This argumentation is only valid in the case of concentrated solutions. Its validity in the present case depends on the degree of dilution of the soluble material in the droplet. The dilution should be estimated to test the validity of this argumentation.

As shown in Table S-1, the average mass concentration of the suspension droplets formed by the atmospheric particles was estimated to be 0.074 g / ml. Similarly, the mass concentrations of droplets formed by pure NaCl and $\text{Ca}(\text{NO}_3)_2$ measured for comparison were 0.029 g / ml and 0.024 g / ml, respectively. These concentrations are equivalent to 0.49 mol / l and 0.15 mol / l.

We conducted additional experiments to compare the freezing temperatures of pure water and NaCl solution and found that the droplets formed by NaCl particles froze consistently at lower temperatures than the pure water droplets by the current method (Fig. S-4). this result suggests that the molar depression of freezing point by NaCl or $\text{Ca}(\text{NO}_3)_2$ may occur within the range of concentrations expected in the current ice crystal formation experiment, unless there is an inclusion of extremely active ice nuclei. Furthermore, Pruppacher and Neiburger (1963) suggested that the freezing point depression by inorganic salt may be significant with concentrations down to 10^{-3} mol / l. Therefore, with the degree of

dilution involved in the current experiment, freezing point depression may be effective and act to hinder freezing of droplets activated from NaCl or Ca (NO₃)₂ enriched particles.

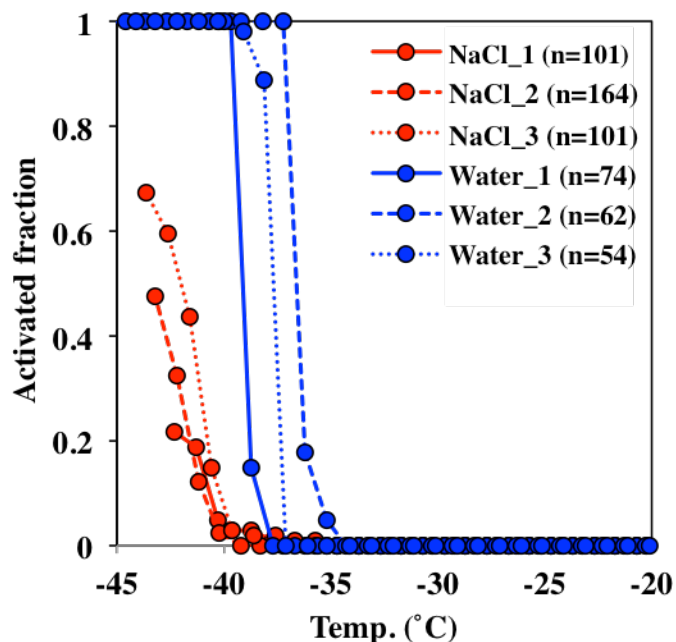


Figure S-4: Activated fractions of NaCl and pure water droplets. Three set of samples were tested for both NaCl and water. The number of particles and droplets observed under the microscope is shown as n. The test NaCl particles were aerosolized by atomizing their solutions (0.005g/ml) and collected on the substrate with an impactor. The pure water droplets were also collected by spraying directly onto the substrate.

Page 14. Line 31: Can the OH peaks be associated with either alcohol or carboxy functionalities?

Detections of both the OH peak (3200 – 3650 cm⁻¹) and the organic peak (2800 – 3100 cm⁻¹) in this study suggest that the detected particles are highly oxidized organic matter containing hydroxyl group (Laskina et al., 2013). Although it is difficult to draw any conclusions, these Raman spectral features are also similar to OH bands observed for glassy oxygenated organic material (Tong et al., 2011; Baustian et al., 2012).

Page 15, lines 25 – 26: improve this sentence. You might split it: “...mainland Japan. Further upstream...”

The sentence was now split accordingly to the suggestion as the follows:

'The study area, Kanazawa City, is located along the west coast of mainland Japan. Further upstream of the westerly continental outflow are the vast arid regions of inland China and Mongolia. Every spring, frequent dust outbreaks are observed, transporting a massive amount of mineral dust aerosols (Asian

dust) across the region and beyond (Iwasaka et al., 2009).'

Page 16, line 2: why exactly 3.27 ± 1.80 particles/cm³? Please explain.

This value is basically what we observed as an average concentration during Asian dust events in February 2016 at our monitoring station. This is explained in the manuscript for better clarity as follows: 'The average concentration observed during Asian dust events in February 2016 was 3.27 ± 1.80 particles/cm³, so the concentrations observed during February sampling period were not as high as that expected during major Asian dust events '

Page 16, line 8: how far away is NOTOGRO from the sampling location?

We added the following information in the manuscript:

(NOTOGRO; 37.45°N, 137.36°E; 116km north east of the sampling location)

Page 16, lines 20 – 27: this paragraph reads like a conclusion and might be deleted here and merged with the conclusion section.

The corresponding paragraph is now merged with the conclusion.

Page 17, line 5: the text in the bracket needs to be formulated better. Is it meant that mineral components such as clay minerals have defects in their crystal structure and contain impurities?

Yes, the corresponding part is now changed accordingly to the suggestion:

These mineral dust particles were suggested to be mixtures of several clay mineral components rather than single mineral species, having defects in their crystal structure and contain impurities.

Page 28, Fig. 3 and page, 35, Fig. 10: the line along which the transect of the particle was scanned should be indicated. The length bar of 0.5 μm given on top of the AFM images does not seem to correspond with the length axis given at the bottom in some images. Please check.

We checked and changed Fig.3 and Fig. 10 as follows:

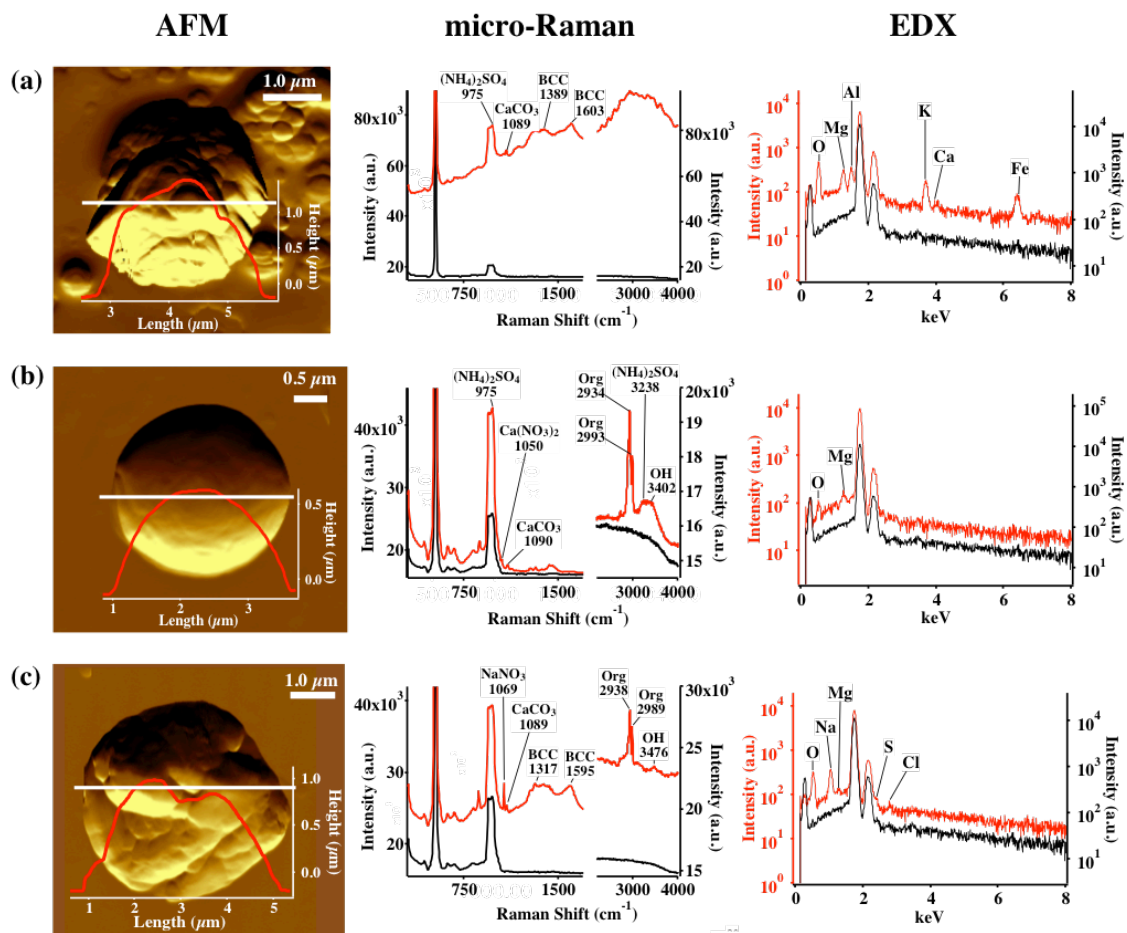


Figure 3: AFM topographic images of representative IN active particle (a) and non-active particle (b, c) groups, and their corresponding Raman and EDX spectra. The AFM images were obtained in probe amplitude mode. The inset in the AFM image shows the scanned height along the white transect of each particle. The red and black curves indicate the spectra of the particles and the substrate background, respectively.

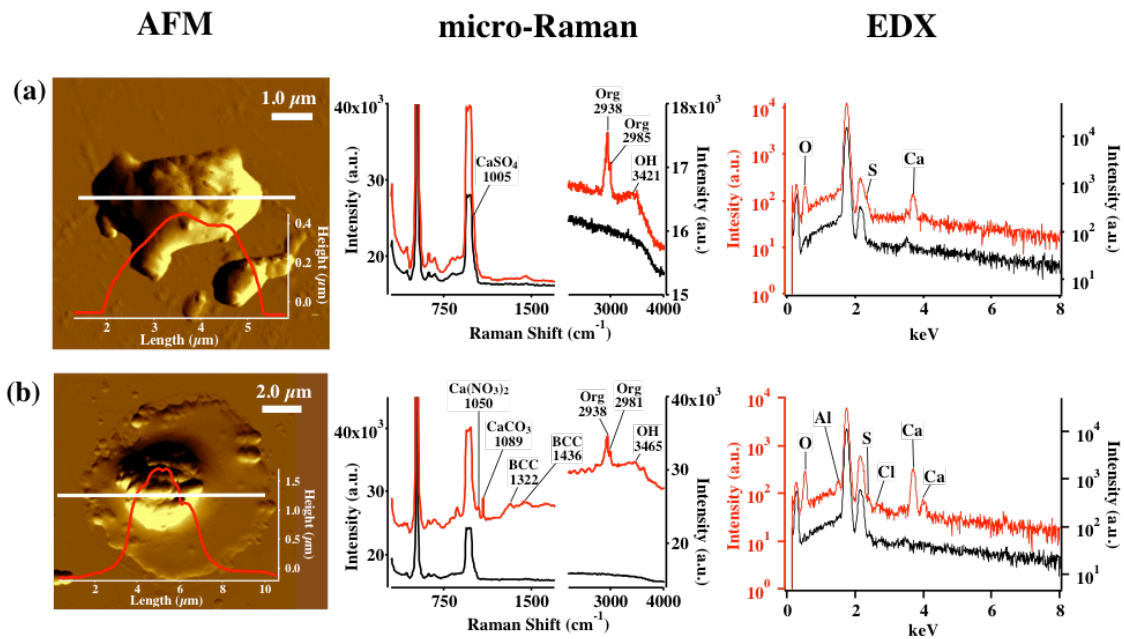


Figure 10: AFM topographic images of representative Ca-rich particles in IN active (a) and non-active (b) groups, and their corresponding Raman and EDX spectra. The AFM images were obtained in probe amplitude mode. The inset in the AFM image shows the scanned height along the white transect of each particle. The red and black curves indicate the spectra of the particle and the substrate background, respectively

Page 30, Fig. 5: what sulfates are meant here if not ammonium sulfate or calcium sulfate?

This sulfates in Fig. 5 means the detection frequency of either ammonium sulfate or calcium sulfate. We also changed Fig. 5 for better clarity as the follows:

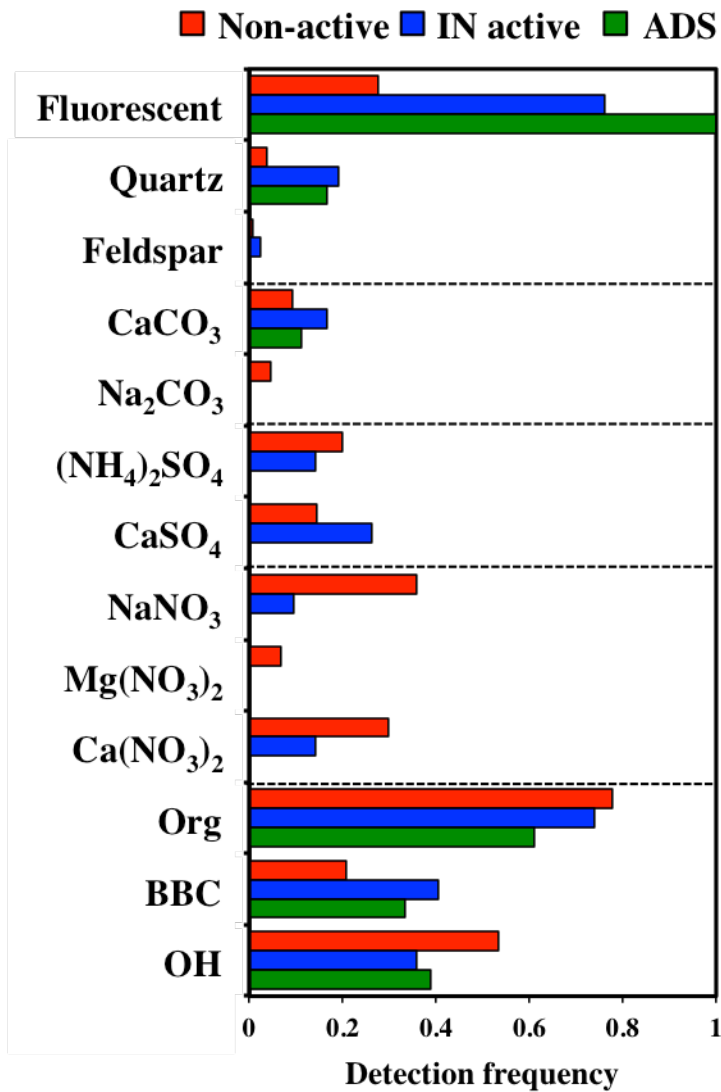


Figure 5: Summary of the detection frequencies of the assigned components in non-active and IN active particles by micro-Raman analysis. Data from Asian dust source (ADS) particles are shown for comparison.

Page 3, line 9: “residues” instead of “residue”.

Corrected accordingly.

Page 8, line 31: Sobanska et al. (2012) is missing from the reference list.

Corrected accordingly.

Page 14, line 4: “nucleus” instead of “nuclei”

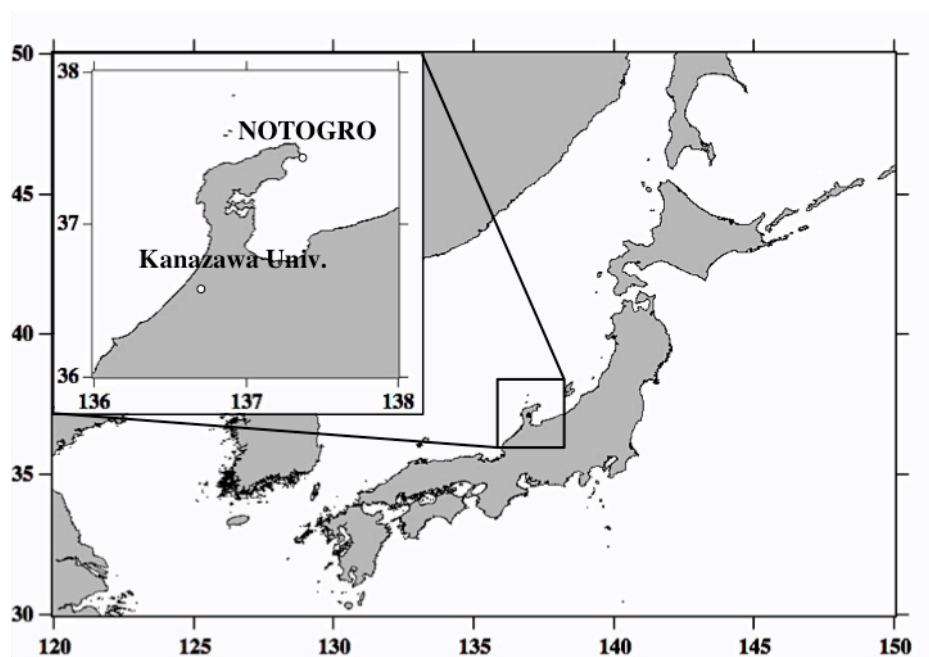
Corrected accordingly.

Page 26: figure caption: “supersaturation” instead of “super saturation”.

Corrected accordingly.

Page 27, Figure 2: the axis numbers are too small to be readable.

Corrected accordingly as follow.



Page 28, Fig. 3 and page, 35, Fig. 10: The label “BCC” in the Raman spectra should read “BBC”.

Corrected accordingly.

References:

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