

We thank Anonymous Referee #1 for his/her thoughtful comments and useful discussion. Below are our point-by-point responses.

Reviewer's comment [1]:

How long were samples boiled in H₂O₂? How was it determined that all the organic material was removed? Why were these two treatments (heat and H₂O₂) chosen?

Authors' response [1]:

We repeated the H₂O₂ treatment until no visible reaction could be detected by addition of more H₂O₂, and used the lack of visible bubbling as an indicator that almost all soil organic matter had been removed. For clarity, we have rewritten the following sentence:

Page 9708, Lines 10-12: "The former samples were prepared by boiling gently in a 30% H₂O₂ solution until almost all organic matter was expected to be digested (i.e., until no visible reaction could be detected by addition of more H₂O₂), followed by rinsing with deionized water and drying."

H₂O₂ digestion was used to remove organic matter almost completely, and dry heating was initially pursued as an alternative way of removing/deactivating organic matter without liquid immersion. As a result, we could confirm that dry heating to 300°C has a similar impact to H₂O₂ digestion [*Page 9710, Line 1-2*], although wet heating to 90-100°C is less effective [*Page 9712, Line 12-14*]. We have written the last part of the first paragraph in Section 3.1 as follows:

Page 9710, Line 1-2: "Dry heating is also known as a technique to remove and/or deactivate organic matter in soils. For example, Fernández et al. (1997) reported that soils heated at 150°C exhibit no significant loss of organic matter, whereas those heated at 490°C lose almost all the organic matter. As for the ice nucleation properties, we confirmed that dry heating to 300°C has a similar impact to H₂O₂ digestion (Fig. 3)."

Reviewer's comment [2]:

Please point out early in the paper that China Loess is used for comparison purposes as a desert dust rather than an agricultural soil.

Authors' response [2]:

We have rewritten the following sentence:

Page 9708, Lines 5-7: "As a reference for natural desert soil dusts, we used China loess soils (CJ-1) (Nishikawa et al., 2000), which were collected in an arid area in Gansu Province, China."

Reviewer's comment [3]:

Are the images in Fig. 4 examples of 600 nm particles even though the scale bar is 300 nm?

Authors' response [3]:

Yes. As described in the Materials and methods section, all data are the results from the experiments with particles having a mobility diameter of 600 nm.

Reviewer's comment [4]:

Do you expect all organic compounds of interest to contain both N and S? Aren't there lots of organic compounds that just contain C, H, O? Were all the CHO particles just soot? Is there a reason why organic compounds in agricultural soils would contain N and S rather than N or S or just CHO? You may be obtaining an underestimate of the number of organic compounds.

Authors' response [4]:

It is well known that SOM serves as a reservoir of nutrients, such as N, P and S (Paul, 2007). We did not necessarily expect all organic particles to contain both N and S, but our results showed that all the particles categorized as "organics" contained both N and S, but not P. We have added the following sentences in Section 3.2:

Page 9710, Line 20: "It has long been known that SOM serves as a reservoir of nutrients, such as N, P and S (Paul, 2007). As for the samples analyzed here, all the particles categorized as organics contained both N and S, but P was not found."

We classify carbonaceous particles lacking N and S as "others" and not "organics", because most of them have a morphology like soot (see Fig. 4e). As seen in Fig. 5a, such particles categorized as "others" are very minor components and their exclusion does not result in a serious underestimate of the number of organic compounds.

Reviewer's comment [5]:

Pg 9710 line 25: My understanding is that a sample of the soil was aerosolized and imaged. Other samples of soil were run through the CFDC or treated and run through the CFDC. The particles that nucleated ice were then collected after the CFDC for imaging. This should be stated in the Materials and Methods Section.

Authors' response [5]:

We have added more detailed explanations in the Materials and methods section as follows:

Page 9709, Line 8: "The dust aerosol particles were collected using a two-stage jet impactor (Matsuki et al., 2010a, b; Tobo et al., 2010). We used only the second stage of the impactor, because the 50% cutoff aerodynamic diameters of the first and second stage at a sample flow of 1 L min⁻¹ are estimated to be 1.6 and 0.2 μm, respectively, and the mobility diameter of the particles is 600 nm (= 0.6 μm). For the IN sampling, the particles that nucleated ice and were grown to ice crystal sizes were collected using a single jet impactor (Prenni et al., 2013; Tobo et al., 2013) installed at the outlet of the CFDC. The 50% cutoff aerodynamic diameter at a sample flow of 1.5 L min⁻¹ is estimated to be 2.9 μm."

Reviewer's comment [6]:

Pg. 9710 line 27: The DMA size selection only produces monodisperse particles if the particles are spherical. It is unlikely that you have imaged enough particles to plot a size distribution, though it would be helpful if it could be provided. The distribution of particles studied can be referred to by their 600 nm mobility diameter, but cannot be called monodisperse.

Authors' response [6]:

According to your suggestion, we have avoided using the expression "monodisperse" in the revised manuscript [*Page 9710, Line 27; Legends in Figs. 2 and 5*].

Reviewer's comment [7]:

Pg 9711 line 7: A few more sentences could be added to this paragraph to clarify Fig. 5b.

Authors' response [7]:

We have added/rewritten the following sentences:

Page 9711, Lines 11-12: “~ that all organic compounds can be digested and removed via H₂O₂ treatment, but inorganic components are not altered by H₂O₂ treatment”

Page 9711, Lines 13-17: “The temperature-dependent changes in the fractions of organic and inorganic IN estimated from this calculation (Fig. 5b) are roughly consistent with the results from the SEM/EDX analysis (Fig. 5a). If H₂O₂ treatment can cause a certain reduction in the ice nucleating ability of inorganic components, the possibility remains that the results in Fig. 5b may somewhat overestimate the fractions of organic IN. We have not exhaustively analyzed the influence of H₂O₂ treatment on all known minerals that may be present in the soil dusts. Nevertheless, the results presented here indicate that the reduction of the ice nucleating ability of the agricultural soil dusts after H₂O₂ treatment (Fig. 2) can be explained mainly by the removal of organic matter.”

Reviewer's comment [8]:

Kaolinite is unlikely to be affected by 300°C temperatures or H₂O₂. What are the other common minerals in the soil? How do these differ from the common minerals in China Loess? Could any of these minerals be altered by heat or H₂O₂? If minerals are also deactivated, Fig. 5b may overestimate the amount of organic.

Authors' response [8]:

In the calculation, we assume that inorganic components are not altered by H₂O₂ treatment. As you point out, Fig. 5b may somewhat overestimate the amount of contributions of organics to ice nucleation activity if minerals are deactivated, and we have not exhaustively analyzed the influence of H₂O₂ treatment on all known minerals that may be present in the soil dusts (see Authors' response [7]). Nevertheless, our results demonstrate that the differences in the IN fraction of kaolinite and China loess soil dust between before and after treatments (i.e., H₂O₂ digestion or dry heating to 300°C) are relatively small (at least, within the range of uncertainty of the CFDC measurements; see Figs. 2 and 3).

Reviewer's comment [9]:

Pg 9712 line 5: The structure of this paragraph is confusing. Please start with the main point (the last sentence) and then compare to literature. Is soil from England and Wyoming very different in composition?

Authors' response [9]:

As suggested by the reviewer, we have largely changed the structure of this paragraph as follows:

Page 9712, Lines 5-19: "It is noteworthy that although agricultural soil dusts collected in Wyoming have similar ice nucleating abilities to those collected in England (Fig. 2b), the major component responsible for ice nucleation at temperatures between about -36°C and -15°C has been interpreted in different ways. Our results demonstrate that the presence of organic compounds (i.e., SOM particles) has a significant influence on the ice nucleating ability of the Wyoming soil dusts throughout the entire temperature range down to -36°C . In contrast, O'Sullivan et al. (2014) suggested that mineral components are more important than biogenic components for the ice nucleating ability of the England soil dusts at temperatures colder than about -15°C . The suggestion by O'Sullivan et al. (2014) was based on the results from freezing experiments with the soil dusts before and after wet heating to 90°C . It is expected that wet heating to $90-100^{\circ}\text{C}$ deactivates only certain organic matter (e.g., heat-sensitive proteins or proteinaceous compounds, Christner et al., 2008). In fact, it has been reported that wet heating to $90-100^{\circ}\text{C}$ is less effective than H_2O_2 digestion in reducing the ice nucleating ability of fertile soil dusts (Conen et al., 2011; O'Sullivan et al., 2014). In this study, we applied treatments designed to remove and/or deactivate almost all organic matter (i.e., H_2O_2 digestion or dry heating to 300°C). Thus, although the possibility remains that the soil dusts from Wyoming and England are very different in composition, we speculate that the different interpretations of the major component responsible for ice nucleation are in part attributable to the different experimental approaches."

In the revised version, we have described the main point in the first sentence. Unfortunately, it is not clear whether the compositions of the soil dusts from Wyoming and England are similar or not. Although the possibility remains that these soil dusts are very different in composition, we speculate that the different interpretations of the major component responsible for ice nucleation are in part attributable to the different experimental approaches. In the revised version, we have described this point in the last sentence of this paragraph.

Reviewer's comment [10]:

Pg 9712 line 23: It is hard to tell whether the impact on the ice nucleating ability of China Loess is small because of the log scale. Please give numbers rather than referring to changes as “small” or “large”.

Authors' response [10]:

A problem is that it is difficult for the CFDC to obtain the IN fraction data accurately, especially at warmer temperatures. The differences in the IN fraction of China loess soil dust between before and after treatments are “within the range of uncertainty” of the CFDC measurements. For this reason, we consider that although it is hard to discuss the quantitative differences, the impact on the ice nucleating ability of China Loess must be relatively small as compared to the case of the Wyoming soil dusts. Thus, instead of giving numbers, we have added the statement “~ is relatively small (within the range of error).” to a sentence in Section 3.1 [Page 9710, Line 1].

Reviewer's comment [11]:

Fig. 4 legend: Please specify that the particles are also Au-coated.

Authors' response [11]:

We have added an explanation that “*The SEM/EDX images were acquired after Au-coating.*” in the legend of Fig. 4.

Reviewer's comment [12]:

Figs. 4&5: EDX is not generally used as a quantitative technique, and it is challenging to make it quantitative. When imaging the particles, the background is also imaged, and the peaks due to the background should be reduced in intensity. The amount these peaks come through the particles of the sizes given in Fig. 4 could be estimated using a SEM simulator like Casino. How far into the particle does the EDX beam penetrate? Why is there so little variation in the intensities of the carbon peaks? What is being normalized? The minerals have additional carbon; I suspect they are coated with CHO compounds. Why are so many particles pure organic or pure mineral and so few internally mixed? Is this typical of soils? Why does this separation between organic particles and mineral particles occur in soils?

Authors' response [12]:

Thank you for pointing out a need for clarification in the manuscript. The X-ray spectra from the particles were acquired for 20 sec of live time at an acceleration voltage of 20 kV. At this voltage, it is expected that the electron beam penetrates the entire particle (Laskin and Cowin, 2001). Here, we have added the following explanations:

Page 9709, Line 12: “After the SEM images of individual particles were taken, the X-ray spectra from the particles were acquired for 20 sec of live time at an acceleration voltage of 20 kV.”

Page 9710, Line 17: “A major difficulty in the EDX analysis of submicron particles is their high transparency for the primary electron beam (Laskin and Cowin, 2001). Since the electron beam penetrates the entire particle, the EDX spectrum from the particle projection area contains the background signal from the butvar film supported by Ni-grids as well as the signal of the particle coated with Au. For this reason, as illustrated in Fig. 4, we compare the EDX spectrum of the particle projection area with that from the particle-free area.”

As the reviewer points out, the particles categorized as “minerals” have additional carbon. However, since the particles categorized as minerals contained Ca and Mg, we consider that the detection of the C peak in particles categorized as “minerals” is mostly attributable to the presence of carbonates (e.g., CaCO_3 , $\text{CaMg}(\text{CO}_3)_2$). It is also important to note that the major elements of particles categorized as organics are C, N and S. For these reasons, in this study, only particles containing the major elements of both organics and minerals (i.e., C, N, Si and S) are categorized as “organo-mineral mixtures”. It is difficult to explain the reason why so many particles are pure organic or pure mineral and so few internally mixed and why this separation between organic particles and mineral particles occur in soils. It is also difficult to know if this is typical of soils. This is because few studies have reported the composition and mixing state of agricultural soil dusts in the submicron size range (i.e., agricultural soil dusts emitted at this atmospherically-relevant size) using single particle analysis. However, we have to admit that the possibility remains that Si-rich particles containing N- and S-free organics or very small amounts of organics are categorized as “minerals” and not “organo-mineral mixtures”. Therefore, we have added/rewritten the following descriptions:

Page 9710, Lines 23-24: “We consider that the detection of the C peak in particles categorized as minerals may be attributable to the presence of carbonates (e.g., CaCO_3 , $\text{CaMg}(\text{CO}_3)_2$). In this study, only particles containing the major elements of both organics

and minerals (i.e., C, N, Si and S) are categorized as “organo-mineral mixtures”. Therefore, the possibility remains that Si-rich particles containing N- and S-free organics or very small amounts of organics are categorized as minerals and not organo-mineral mixtures.”

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