Authors Responses to Reviewers

We appreciate the helpful comments made by reviewers.

Below, we indicate in detail the revisions made to the manuscript.

Text form the original is shown in blue.

Changes in the revised manuscript are shown in red.

Referee #2

Comments to the Author

This study correlates high levels of low molecular weight, monocarboxylic acids (LMWMCA) with Asian dust in the Japanese snow pack. Since the primary sources of LMWMCA are not associated with Asian dust events, the conclusion is the organic acids adsorb onto the dust particles during transport. This changes the surface chemistry of the dust particle and therefore its efficiency as ice nuclei in clouds. The study indicates that while organic acids adsorb on dust particles, coating of dust particles by sulfate or nitrate is not as efficient. The study further shows that the uptake of formic acid and acetic acid largely depended on the amount of Ca in the dust and therefore does not apply to all dust types. This is a good study and I recommend publication with minor corrections and revisions.

I have concerns regarding the conclusion that coated dust particles make better ice nuclei and can therefore cause heavier snowfall. Many studies have shown that coating bare dust particles with organics can reduce the ice nucleating properties of bare mineral dust depending on the chemical composition the dust (Kulkarni et al 2014). Based on the complexity of ice nucleation properties of dust, the authors have not provided enough evidence to claim that these organic acid coated dust particles can alter the snowfall. This does not seem to be an important component of the study, but is an attempt for the authors to provide relevance context. It can be left out.

Response:

Based on the comment 3), we have modified the following descriptions in the manuscript. Please see our responses to comment 3).

Minor Issues:

1) Abstract: Remove last line of the abstract as there is no evidence presented to support this statement.

Response:

As the referee suggested, we have rephrased the sentence "Our study demonstrates that Asian alkaline dusts can uptake volatile monocarboxylic acids during long-range transport, and the dusts coated with organic acids may act as effective ice nuclei to cause a heavy snowfall." to "Our study suggests that Asian alkaline dusts may be a carrier of volatile monocarboxylic acids." (Page 1, Lines 25-26).

2) Page 2, line 1 When referring to urban, forest, marine, and Arctic samples are these air samples or water/snow samples?

Response:

We are sorry for the unclear description. We have modified the following sentence.

The sentence "They have been reported in urban (Kawamura et al., 2000) forest (Andreae et al., 1988), high mountain (Preunkert et al., 2007), marine (Miyazaki et al., 2014), and Arctic samples (Talbot et al., 1992; Legrand et al., 2004)." has been revised to "Gaseous and particulate formic and acetic acids have been reported in urban (Kawamura et al., 2000), forest (Andreae et al., 1988), high mountain (Preunkert et al., 2007), marine (Miyazaki et al., 2014), and Arctic samples (Legrand et al., 2004)." (Page 1, Line 30 - Page 2, Line 2).

We have deleted the following reference to the references section.

Talbot, R. W., Vijgen, A. S., and Harris, R. C.: Soluble species in the Arctic summer troposphere: acidic gases, aerosols, and precipitation, J. Geophys. Res., 97, 16531-16543, 1992.

3) Page 2 line 20 This paragraph focuses on the importance of snow to the region and then discusses how changes in surface chemistry and hygroscopicity may improve ice nucleation of dust. See above comment. Previous studies have shown bare mineral dust to be very efficient IN and that coating may decrease ice nucleation properties, but increase water nucleation. This study provides no evidence either way and this section distracts from the point of the study.

Response:

Kulkarni et al. (2014) and references therein reported that ice nucleation potential of dust particles coated with organics are lower than that of bare dust particles. These are very important. However, our study do not clarify the relationship between ice nucleation potential and Asian dust particles at Murodo-Daira near the summit of Mt.

Tateyama. Based on the suggestion, we have modified the following descriptions in the manuscript.

The following sentences were deleted; "Because these snow precipitations are the sources for agricultural and drinking waters in those areas, snow precipitations in the high mountains are important for the water cycles in Japan. Ice nuclei (IN) supplied from the Asian Continent through long-range atmospheric transport and the water vapours supplied from the Sea of Japan during the winter Asian Monsoon are two important components to promote the heavy snow over the western part of Japan. Asian dust particles may act as effective IN to result in a heavy snow over the Japanese Alps. During the formation of snow crystals, chemical compositions of dust surface may be critical to control the hygroscopic properties of dust particles (Creamean et al., 2013)."

We have deleted the following reference in the references section.

Creamean, J. M., Suski, K. J., Rosenfeld, D., Cazorla, A., DeMott, P. J., Sullivan, R. C., White, A. B., Ralph, F. M., Minnis, P., Comstock, J. M., Tomlinson, J. M., and Prather, K. A.: Dust and biological aerosols from the Sahara and Asia influence precipitation in the Western U.S., Science, 339, 1572-1578, 2013.

4) Page 3 line 10 "several dirty layers were recognized by visual observation due to the occurrence of Asian dusts." This is an awkward statement. How was dust differentiated from a soot layer for example, or other industrial pollutants? How was the occurrence of Asian dust verified?

Response:

We are sorry for the unclear description. In the section 2 stage, we do not decide the dust, soot, or other industrial pollutants. The word "dust" has been replaced with "dirty" in section 2. We have modified the following sentences in response to the above comment.

The following sentences were deleted; "Table 1 provides descriptions of snow samples collected from the snow pit sequence, in which several dirty layers were recognized by visual observation due to the occurrence of Asian dusts. Five snowpack samples including three dust layers were collected from the pit sequence in April 18, 2009. Eleven snowpack samples including four dust layers were collected from the pit sequence in April 17, 2011. In order to evaluate the homogeneity of snow samples within the same snow horizon with dust layer, another snowpack sample (#4') was collected at ca. 1 m away from the location of sample #4 in 2009. Because the thickness

of dust layers in snow pit sequence is ca. 10 cm or more, dusts are deposited together with snowflakes during snow precipitation rather than dry deposition."

We added the following sentenced in the revised MS; "Table 1 provides descriptions of snow samples collected from the snow pit sequence, in which several brown-colored dirty layers were recognized by visual observation. Five snowpack samples including three dirty layers were collected from the pit sequence in April 18, 2009. Eleven snowpack samples including four dirty layers were collected from the pit sequence in April 17, 2011. In order to evaluate the consistent distribution of snow samples within the same snow horizon with dirty layer, another snowpack sample (#4') was collected at ca. 1 m away from the location of sample #4 in 2009. Because the thickness of dirty layers in snow pit sequence is ca. 10 cm or more, brown-colored particles are deposited together with snowflakes during snow precipitation rather than dry deposition." (Please see Page 3, Lines 5-12.)

5) Page 3 line 13 "In order to evaluate the homogeneity of snow samples within the same snow horizon with dust layer, ..." Not sure what is meant by this statement. Need clarification.

Response:

The sentence; "In order to evaluate the homogeneity of snow samples within the same snow horizon with …" has been changed to "In order to evaluate the consistent distribution of snow samples within the same snow horizon with …". (Page 3, Lines 9-10).

6) Page 3 line 25 "The data of inorganic ...." In addition to back trajectory and lidar data, were mineral, or crustal elemental fractions in the snow contaminants compared with reference material for the different Asian dust regions to verify dust from specific regions?

Response:

We apologize for this error. The sentence; "The data of inorganic ions and trace elements in the reference samples are reported elsewhere (Nishikawa et al., 2000, 2013)." has been revised to "The detailed information of reference samples are reported elsewhere (Nishikawa et al., 2000, 2013)." (Page 3, Lines 21-22).

7) Page 3 line 30 Why and how was the pH of the samples adjusted to 8.5 to 9.0? If this is described in Kawamura and Kaplan 1984, then including it here just raises questions and isn't informative. This is again stated on page 4 line 4 without explanation.

Response:

Melt snow samples from the Muroro-Daira were slightly acidic. To avoid the evaporative loss of volatile organic acids from the water samples during analytical procedure, the samples were adjusted to pH = 8.5-9.0 with 0.05 M KOH solution.

We have modified the following sentence in response to the above comment.

The sentence; "150 ml of melted snow samples were transferred to a pear-shape glass flask (300 ml) and the pH was adjusted to 8.5–9.0 with 0.05 M KOH solution." has been revised to "150 ml of melted snow samples were transferred to a pear-shape glass flask (300 ml). To avoid the evaporative loss of volatile monocarboxylic acids from samples during analytical procedure, pH was adjusted to 8.5–9.0 with 0.05 M KOH solution to form organic acid salts (e.g., $CH_3COO^-K^+$)." (Page 3, Lines 25-27).

8) Page 7 line 25 "Although the alkalinity of snow pit samples can be affected…were slightly acidic." Not sure what the relevance of this statement is. I think it is the use of "although" that is throwing me off.

Response:

We are sorry for the unclear description. We have deleted the following sentence from the revised MS.

"Although the alkalinity snow pit samples can be affected by titration of alkaline dust particles, melt snow samples from the Murodo-Daira were slightly acidic."

Some, but not all grammatical clean-up:

9) Abstract Line 17 remove "being" before consistent.

Response:

The term remain the same.

10) Page 1 line 30, comma after Kawamura citation.

Response:

Based on the suggestion, we have added "," after (Kawamura et al., 2000). (Page 2, Line 1).

11) Page 2, line 8 have a variety of sources (insert of)

Response:

Based on the suggestion, the phrase; "Formic and acetic acids have variety sources such as primary emission from motor exhausts (Kawamura et al., 2000)…" has been revised to "Formic and acetic acids have a variety of sources such as primary emission from motor exhausts (Kawamura et al., 2000)…". (Page 2, Line8).

12) Page 5 line 7 insert "the" before laser.

Response:

Based on the suggestion, the sentence "The observation wavelength of laser is 532 nm." has been revised to "The observation wavelength of the laser is 532 nm." (Page 5, Lines 6-7).

13) Page 7 line 9 change has to was before "involved"

Response:

Based on the suggestion, the sentence; "… Asian Continent has involved with a heavy snow precipitation." has been revised to "… Asian Continent was involved with a heavy snow precipitation." (Page 7, Line 9).

14) Check uses of "although" and "however", the authors use these two conjunctions are used a lot and not always appropriately.

Response:

Based on the suggestion, we modified following sentences.

The sentence; "However, concentrations of lactic and glycolic acids are 1 and 2 orders of magnitude lower than those of major monocarboxylic acids (C_1 and C_2), respectively." has been revised to "Concentrations of lactic and glycolic acids are 1 and 2 orders of magnitude lower than those of major monocarboxylic acids (C_1 and C_2), respectively." (Page 5, Lines 30-31).

The sentence; "Although the pathways of microbial production of branched chain monocarboxylic acids and lactic acid may be different, this strong correlation indicates

that these organic acids are closely linked in the biosynthetic processes associated with bacterial activity in soils." has been revised to "This strong correlation suggests that these organic acids are closely linked in the biosynthetic processes associated with bacterial activity in soils." (Page 9, Lines 20-21).

The sentences "Although bacteria species responsible to branched monocarboxylic and lactic acids have not been reported in the Tateyama snow samples (Maki et al., 2014), our results suggest that branched chain monocarboxylic acids are produced by bacterial process in soils of the Asian Continent and transported over the Japanese Islands with Asian dust. However, contribution of biogenic monocarboxylic acids is much lower than anthropogenic monocarboxylic acids." have been revised to "Bacteria species responsible to branched monocarboxylic and lactic acids have not been reported in the Tateyama snow samples at this time. Our results suggest that branched chain monocarboxylic acids may be produced by bacterial process in soils of the Asian Continent and transported over the Japanese Islands with Asian dust. Contribution of biogenic monocarboxylic acids is much lower than anthropogenic monocarboxylic acids." (Page 9, Lines 25-28).

We have deleted following sentence from the revised MS; "However, it was detected in our snow pit samples."

In addition, we have modified the following sentences.

The sentence "Details of analytical procedure were described previously (Kawamura et al., 2012)." has been revised to "Details of analytical procedure were described previously except for the pH adjustment with KOH solution (Kawamura et al., 2012)." (Page 4, Lines 16-17).

The sentence "They are adsorbed on the pre-existing particles via atmospheric titration with alkaline Kosa particles during the long-range atmospheric transport over the Japanese Islands." has been revised to "They are adsorbed on the pre-existing alkaline Kosa particles via atmospheric titration during the long-range atmospheric transport over the Japanese Islands." (Page 9, Lines 4-5).

The phrase "Total MCA-C/DOC ratio (av. 21%) in 2009 is significantly higher than

those reported in rainwater from Los Angeles …" has been revised to "Total MCA-C/DOC ratio (av. 21%) in 2009 is significantly higher than those reported in rainwater samples from Los Angeles …" (Page 9, Lines 8-9).

The phrase "… suggesting that entrainment of organic acids in snow flakes is significant during the atmospheric transport from China to Japan." has been revised to "… suggesting that entrainment of organic acids in alkaline dusts and snow flakes is significant during the atmospheric transport from China to Japan." (Page 9, Lines 12-13).

The phrase "… secondary photochemical oxidation of anthropogenic toluene, indicating that … " has been revised to "… secondary photochemical oxidation of anthropogenic toluene and other aromatic hydrocarbons, indicating that …" (Page 10, Lines 5-6).