

## Authors' Responses to reviewers

We appreciate the helpful comments made by reviewers.

Following the two reviewer's comments, we have removed Figure 7. Figure numbers were reorganized.

We have changed the configuration of section 3 (Results).

Below, we indicate our point-to-point responses in blue.

### Reviewer 2:

*The authors of this paper report the concentrations of low molecular weight monocarboxylic acids in the gas and particle phase samples collected at a deciduous broad leaf forest site near Sapporo, northern Japan. They claim that acetic acid, isopentanoic acid, and lactic acid are the major particle phase monoacids at the site and they claim that isopentanoic acid and lactic acid originate from soil bacteria activities because they correlate well with gas phase isobutyric acid that may be produced by soil bacteria. While some of the data presented here may be interesting for people working in this field, the paper has major flaws as there are too many speculative statements that do not stand up to scrutiny. I recommend major revision before this paper could be published in ACP.*

*Comment 1: Page 5 Line 12: It is not clear to me why the authors chose sulfate ion as an anthropogenic tracer for local urban emissions. Is the total sulfate ion or is it nss-sulfate? How does the authors exclude the influence of long range transport from China or Russia? This should be clarified.*

### Response:

Thanks for the comment.

We calculated non-sea-salt  $\text{SO}_4^{2-}$  and added the following sentences/phrases in the revised manuscript.

“Concentrations of non-sea-salt  $\text{SO}_4^{2-}$  [nss- $\text{SO}_4^{2-}$ ] is calculated by the following equation:

$$[\text{nss-SO}_4^{2-}] = [\text{SO}_4^{2-}] - 0.25 \times [\text{Na}^+],$$

where  $[\text{SO}_4^{2-}]$  and  $[\text{Na}^+]$  are concentrations of total  $\text{SO}_4^{2-}$  and  $\text{Na}^+$ , respectively (Duce et al., 1983, Berg and Winchester, 1978).” Please see page 3, lines 26-30.

“Nss- $\text{SO}_4^{2-}$  (mean: 2240 ng m<sup>-3</sup>) is major anion ...” Please see page 4, line 16.

“nss- $\text{SO}_4^{2-}$ ”. Please see page 4, line 20.

“nss- $\text{SO}_4^{2-}$  ( $r^2 < 0.14$ )”. Please see page 4, line 22.

We also checked backward air mass trajectories at a level of 300 m and added the following sentences in experimental and result sections.

“We calculated seven-day air mass back trajectories at a height of 300 m above sea level using the Meteorological Data Explorer (METEX) provided by the National Institute for Environmental Studies (<http://db.cger.nies.go.jp/metex/index.html>).” Please see page 3, lines 31-33.

“This forest site is located a few kilometers south of Sapporo city. The dominant wind direction was from east and south throughout the sampling period. We compared the concentrations of individual monoacids together with  $\text{nss-SO}_4^{2-}$ : an anthropogenic aerosol tracer to evaluate the influence of anthropogenic air mass transport from urban area. We confirmed that individual monoacids in both gas and particle phases did not show a correlation with  $\text{nss-SO}_4^{2-}$  ( $r^2 < 0.14$ ). The majority of sampling air was not affected by urban area. In addition, Figure 5 shows seven-day air mass back trajectories (300 m a.s.l.) for the study period from 28 June to 8 July at the sampling site. Most of the air masses passed through the Pacific Ocean during the measurement period, except for 28 June. This result may suggest that the air masses arriving at the forest site are not affected by the outflows from East Asia and far East of Russia.” Please see page 4, lines 19-26.

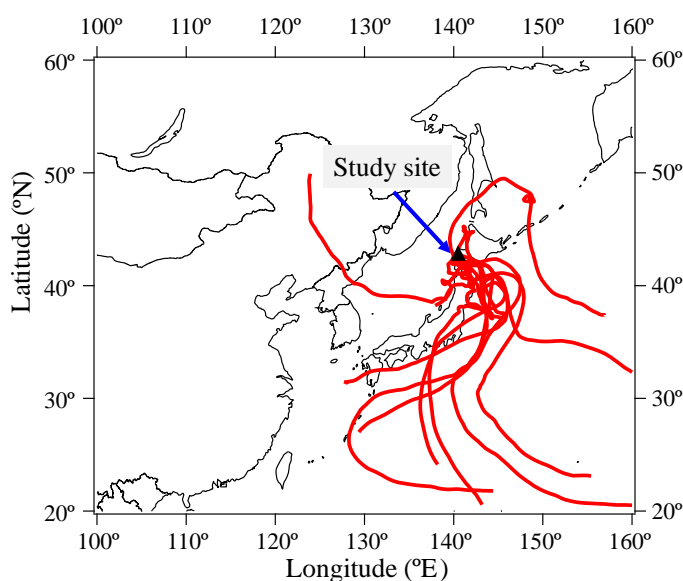


Figure 5. Seven-day air mass back trajectories at a height of 300 m a.s.l. during the sampling period.

*Comment 2: Page 5 Lines 19-20: What are the precursors that microorganisms process to form these compounds?*

**Response:**

Following the comment, we added the following sentence with reference; “Branched chain monoacids including isobutylic acid are known as common metabolites of bacteria (e.g., *Bacteroides distasonis*) and fungi in soils (Uta et al., 2012 and references therein).” Please see

page 4, line 42 - page 5, line 2.

*Comment 3: Page 5 Lines 20-26: This paragraph raises more questions than the answers. How do the authors explain a better correlation of lactic acid with C3-C6 monoacids for nighttime samples? Do the lactic acid producing lactobacillus species more active at night? Even they are produced in soil, how is lactic acid emitted from soil to atmosphere? Couldn't lactic acid be a product of atmospheric oxidation processes? The assumption that C1-C6 monoacids and lactic acid are emitted from soil is not valid without providing concrete evidence for direct soil emission data.*

**Response:**

We deleted the corresponding sentences. Instead, we have added the following sentences in the revised manuscript.

“LMW monoacids such as acetic and propionic acids can be produced by microbiological processes (Uta et al., 2012). In addition, exudation of organic acids is known to occur in vascular plants, mainly from roots (Curl, 1982). Shen et al. (1996) reported that formic, acetic, and propionic acids are contained in forest soil and rhizosphere soil. However, the possible contribution from the forest floor cannot be evaluated in the present study. Variety of emission sources of C<sub>1</sub>–C<sub>6</sub> monoacids may exist in forest soil. We suggest that gaseous C<sub>1</sub>–C<sub>6</sub> monoacids are emitted from the forest floor where soil microorganisms and plant roots contribute to the emissions of gaseous C<sub>1</sub>–C<sub>6</sub> monoacids.

Gaseous lactic acid (hC<sub>3</sub>) in daytime did not show positive correlations with C<sub>1</sub>–C<sub>6</sub> monoacids ( $r^2 < 0.004$ ), whereas lactic acid in nighttime show positive correlations with C<sub>3</sub>–C<sub>6</sub> ( $r^2 = 0.45–0.65$ ) although they were rather scattered. Bacteria (*lactobacillus*) are known to produce lactic acid (Cabredo et al., 2009). Lactic acid can also be produced by the oxidation of isoprene with ozone (Nguyen et al., 2010). We suggest that major portion of C<sub>1</sub>–C<sub>6</sub> acids were emitted within the forest floor.” Please see page 5 lines, 2-11.

*Comment 4: Page 5 Lines 29-32: This paragraph does not add useful information to the manuscript without leaf and soil emission data. They can also be produced from atmospheric oxidation processes.*

**Response:**

Taking account of the comment, we deleted those sentences.

*Comment 5: Page 6 Line 15: Is it important for the formation of organic aerosols or is it important for the formation of biogenic SOA marker compounds? It should also be clarified if the acidity is important for enhancing the partitioning of gaseous organic compounds into the particle phase or the acidity is important for the formation of biogenic tracer compounds in the gas or particle phase.*

**Response:**

Following the comments, we deleted the following sentences; “Particulate formic and acetic acids in daytime negatively correlated with pH of the water extracts from aerosol samples ( $C_1$ :  $r^2 = 0.35$ ,  $C_2$ :  $r^2 = 0.43$ ) (Figure 7). Acidity of aerosol particles can enhance the formation of SOA via organic precursors and subsequent heterogeneous reactions in aerosol liquid phase (e.g., Jang et al., 2002; Pathak et al., 2011). Secondary formation of formic and acetic acids is associated with photo-oxidation of biogenic volatile organic compounds (BVOCs) such as isoprene and monoterpene (Ervens et al., 2008; Kawamura et al., unpublished data). Based on the field measurement, aerosol acidity is important for the formation of organic aerosols such as isoprene SOA tracers,  $\alpha$ -pinene SOA tracers and dicarboxylic acids (oxalic acid) from BVOCs in the forest atmosphere (Mochizuki et al., 2015; 2017b). Formic and acetic acids are intermediate products in the complex heterogeneous oxidation of BVOCs. Lower pH may promote the secondary formation of formic and acetic acids from BVOCs in the forest atmosphere (Kawamura et al., unpublished data).”

*Comment 6: Page 6 Line 17: Aren't formic acid and acetic acid end products rather than intermediate products of oxidation? They may be very stable intermediate products if you think CO<sub>2</sub> as the end product though.*

**Response:**

Thanks for your comment. The lifetimes of formic and acetic acids in gas phase are estimated to be 25 and 10 days, respectively (Paulot et al., 2011). Formic and acetic acids are stable in the atmosphere. By taking the comment, we modified the sentence in the revised MS. Please see page 5 lines 24-25.

*Comment 7: Page 6 Lines 18-19: This sentence is confusing. Do the authors mean lower pH values of aerosol than pKa values of formic acid (3.7) and acetic acid (4.7)? If this is the case, wouldn't formic acid and acetic acid go out of the particle phase, leading to lower particle phase concentrations of both species? If the acidity enhances the formation of small organic acids from BVOCs, what are the processes involved in the formation? Do BVOCs get oxidized directly by particle phase acids on the particle surface?*

**Response:**

Based on the reviewer's comment 5, we deleted the relevant sentences.

*Comment 8: Page 6 Lines 19-24: What about the boundary layer height and temperature? It is often observed that the concentrations of particle phase organic compounds are higher in nighttime samples than daytime samples due to shallower boundary layer height and lower temperature.*

**Response:**

Although we do not have the information on boundary layer height, one sentence was added on the boundary layer height following the reviewer's comment.

“The higher concentrations in nighttime may be associated with shallower planetary boundary layer, which can accumulate organic acids near the ground surface.” Please see page 5, lines 20-21.

Ambient temperature and relative humidity are important factors to control the gas/particle partitioning of formic and acetic acids. Following the reviewer’s comment, we have added the following sentences.

“Although  $F_p$  of LMW monoacids did not show a correlation with ambient temperature in nighttime ( $r^2 < 0.16$ ), except for propionic acid ( $r^2 = 0.31$ ), we found average  $F_p$  of LMW monoacids in nighttime were higher than those in daytime (Table 1).” Please see page 5, line 42 - page 6, line 2.

*Comment 9: Page 6 Line 25: Which bacterium is it (species, strain, etc)? What conditions does it need to produce isopentanoic acid? Is it atmospherically relevant condition? I am sure that not all bacteria are isopentanoic acid producers.*

**Response:**

Based on the comment, we rephrased the sentence as follows.

The sentence “Isopentanoic acid can be produced by bacteria (Allison, 1978).” has been changed to “Isopentanoic acid can be produced by bacteria such as *Clostridium* spp. and *Bacteroides* spp. (Uta et al., 2012 and references therein). These microorganisms live in soil.” Please see page 5, lines 27-28.

*Comment 10: Page 6 Lines 25-29: This paragraph does not add up. Do the authors suggest that lactic acid and isopentanoic acid may be produced by bacteria in soil and emitted to atmosphere because they did not correlate well with particle phase monoacids that might be also produced by soil bacteria? Where are the evidence for particle surface formic acid and acetic acid formation? The data provided in the manuscript is hardly conclusive to suggest the sources or processes involved in their formation.*

**Response:**

Thanks for the critical comment. We have modified the sentence in the revised manuscript as: “We suggest that lactic and isopentanoic acids are linked to the biosynthetic processes, whereas formation processes of  $C_1$ - $C_6$  acids in particle phase may be different from isopentanoic and lactic acids.” Please see page 5, lines 30-31.

*Comment 11: Page 7 Lines 7-8: I am not sure how the comparison of the vapor pressures and  $F_p$  values for these compounds is related to the source of lactic acid and isopentanoic acid. Please clarify this.*

**Response:**

Following the suggestion, we deleted the following sentence; “These results suggest that

particulate lactic and isopentanoic acids may be derived from primary sources in forest ecosystem via microbial activity.”

*Comment 12: Page 7 Line 10: with increasing ambient temperature*

**Response:**

Corrected as suggested. Please see page 7, line 14.

*Comment 13: Page 7 Line 14: What about the nighttime?*

**Response:**

Please see our response to comment 8.

*Comment 14: Page 7 Lines 15-18: Please provide LWC data. What were the state of particles? Were particles deliquesced? How does  $F_p$  correlate with the total particle phase acid concentration? It may well be that particle bound organic mass may be more important for the partitioning of monoacids than LWC.*

**Response:**

We added the aerosol liquid water content in the revised MS with additional discussion.

“We found that  $F_p$  of formic and acetic acids increase with increasing RH ( $C_1$ :  $r^2 = 0.30$ ,  $C_2$ :  $r^2 = 0.55$ ) (Figure 9), whereas other LMW monoacids did not show a correlation with RH ( $r^2 < 0.20$ ), except for butyric acid ( $r^2 = 0.55$ ) in daytime. Although  $F_p$  of LMW monoacids did not show correlation with RH in nighttime ( $r^2 < 0.15$ ), we found that average  $F_p$  of LMW monoacids in nighttime were higher than those in daytime (Table 1). Al-Hosney et al. (2005) and Prince et al. (2008) reported that the uptake of formic and acetic acids by  $\text{CaCO}_3$  can be enhanced by higher RH ( $C_1$ : RH > 62%,  $C_2$ : RH > 53%). In addition, liquid water contents (LWC) of aerosols were estimated using ISORROPIA-II model (Fountoukis and Nenes, 2007) with the data of inorganic ions and meteorological parameters. The aerosol LWC ranged from 1.4 to 14.6  $\mu\text{g m}^{-3}$  (av. 6.4  $\mu\text{g m}^{-3}$ ). Although  $F_p$  of LMW monoacids did not show strong correlations with LWC ( $r^2 < 0.24$ ), strong positive correlations were found between RH and aerosol LWC in daytime ( $r^2 = 0.47$ ) and in nighttime ( $r^2 = 0.74$ ). Higher RH may enhance the partitioning of gaseous formic and acetic acids to aerosol phase as a result of the condensation of water vapour on aerosol particles. Our results suggest that higher temperature depresses a transfer of gaseous formic and acetic acids to aerosol phase and higher RH enhances the partitioning of gaseous formic and acetic acids to aerosol phase in the forest atmosphere.” Please see page 6, lines 5-17.

We have added new Figure 9 as below.

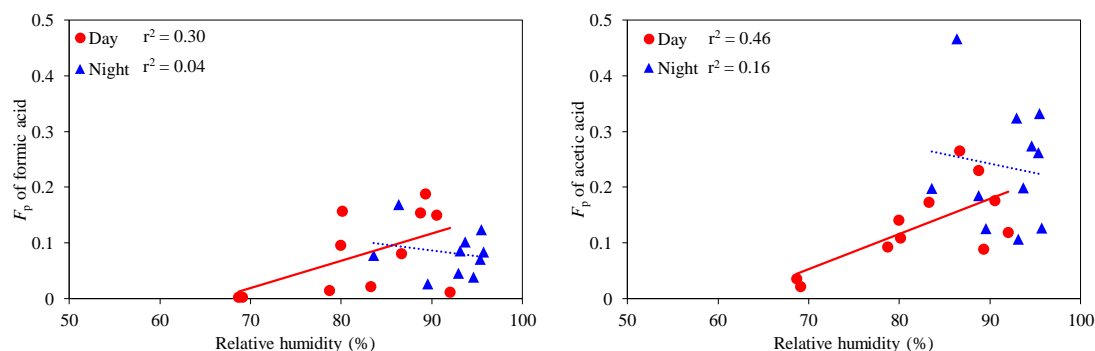


Figure 9. Particle-phase fractions ( $F_p$ ) of formic and acetic acids against relative humidity.

We also added the following sentences in the revised manuscript.

“ $F_p$  of LMW monoacids showed positive correlations with mass concentrations of total LMW monoacids in particle phase ( $r^2 = 0.24$ – $0.46$ ), except for  $C_3$ ,  $C_9$ , and  $C_{10}$ . Although we did not measure the aerosol mass, gaseous LMW monoacids in the forest atmosphere may be adsorbed on the existing particles.” Please see page 6, lines 29-31.

*Comment 15: The section 4.3 does not add useful information to the manuscript as the data compared here are only snapshots of monoacid concentrations at each sampling site, and do not take seasonal or temporal variations of monoacid concentrations into account. I suggest the authors removing this section completely.*

**Response:**

Deleted as suggested.

*Comment 16: The summary and conclusion section need to be revised. There are too many speculative statements here rather than evidence based statements.*

**Response:**

According to the comment, we have revised the summary and conclusions.

“Particle-phase fractions ( $F_p = P/(P+G)$ ) of major LMW monoacids were low ( $F_p$ : 0.04-0.50), except for isopentanoic ( $F_p = 0.68$ ) and lactic ( $F_p = 0.66$ ) acids. Concentrations of long-lived monoacids (formic and acetic acids) in gas phase showed positive correlation with short-lived monoacids ( $C_3$ – $C_6$ ). Concentrations of LMW monoacids in gas and particle phases did not correlate with  $\text{nss-SO}_4^{2-}$  (anthropogenic tracer). These results suggest that anthropogenic contribution is not important, but local contribution, that is, photochemical oxidation of biogenic VOCs, is more important as a source of LMW monoacids. Concentrations of  $C_1$ – $C_6$  monoacids in gas phase showed positive correlations with isobutyric acid ( $iC_4$ ) ( $r^2 = 0.21$ – $0.91$ ). Branched chain monoacids can be used as an indicator of soil microorganism processes (Uta et al., 2012 and references therein). Many kinds of LMW monoacids are exuded by plant roots (Curl, 1982) and derived from soil microorganism (Uta et al., 2012). We suggest that forest floor is another important source of gaseous LMW monoacids. Isopentanoic acid in particle

phase showed a positive correlation with lactic acid ( $r^2 = 0.98$ ), which is produced by soil microbes. Acetic acid in particle phase showed a positive correlation with nonanoic acid ( $C_9$ ), which is produced by the oxidation of unsaturated fatty acids such as oleic acid.  $F_p$  of formic and acetic acids showed good correlations with ambient temperature ( $C_1$ :  $r^2 = 0.49$ ,  $C_2$ :  $r^2 = 0.60$ ) and RH ( $C_1$ :  $r^2 = 0.30$ ,  $C_2$ :  $r^2 = 0.55$ ) in daytime, suggesting that these parameters are important for the gas to particle conversion in the forest atmosphere. The present study demonstrates that deciduous broadleaf forest is an important source of LMW monoacids in gas and particle phases in the atmosphere of northern Japan.” Please see page 6, line 37 - page 7, line 10.

#### References added.

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