

# ***Interactive comment on “Distributions and sources of gaseous and particulate low molecular weight monocarboxylic acids in a deciduous broadleaf forest from northern Japan” by Tomoki Mochizuki et al.***

## **Anonymous Referee #2**

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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

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that do not stand up to scrutiny. I recommend major revision before this paper could be published in ACP.

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.

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

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 night-time 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.

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.

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.

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.

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Page 6 Lines 18-19: This sentence is confusing. Do the authors mean lower pH values of aerosol than pKa values of formic acid ( $\sim 3.7$ ) and acetic acid ( $\sim 4.7$ )? 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?

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.

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.

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.

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.

Page 7 Line 10: with increasing ambient temperature

Page 7 Line 14: What about the nighttime?

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

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concentration? It may well be that particle bound organic mass may be more important for the partitioning of monoacids than LWC.

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.

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

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