

***Interactive comment on “Size distributions of dicarboxylic acids, ketocarboxylic acids,  $\alpha$ -dicarbonyls and fatty acids in atmospheric aerosols from Tanzania, East Africa during wet and dry seasons” by S. L. Mkoma and K. Kawamura***

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Reviewer #2 (Comments to Author):

The authors greatly appreciate the reviewer's comments that improved the quality of our manuscript. We made changes and error corrected according to the suggestions. Please find our replies preceded by “Reply:” We also indicate the actions that were taken in the revised version of the manuscript.

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The paper "Size distributions of dicarboxylic acids, ketocarboxylic acids,  $\alpha$ -dicarbonyls and fatty acids in atmospheric aerosols from Tanzania, East Africa during wet and dry seasons " by Mkoma and Kawamura deals with the sources of dicarboxylic acids and related compounds in atmospheric aerosols from Tanzania. The study is based on two different field campaigns conducted during wet and dry seasons in 2011 at the same rural sampling site in Tanzania. Even though there are no scientific findings in this manuscript, the measurement site is quite interesting because very few diacids and related compounds data are available in Africa region. However, interpretations of results are quite speculative and vague. Hence, the reviewer does NOT recommend publication of this manuscript in the Atmospheric Chemistry and Physics in its current form. A major revision is required before publication.

Reply: We believe that we made some new finding based on the campaigns of wet and dry seasons for various organic and inorganic species. For example, we found apparent discrepancy in the relationship to diacids between  $K^+$  and levoglucosan. This point is discussed with possible interpretation on the difference in the biomass burning process (flaming and smoldering) in lines 495-501 and conclusion section in the revised MS. The reviewer's comments on "speculative and vague" have been considered to improve our discussions in the revised MS (for details, please see the responses below).

Major comments -P2, L40: Diacids and related compounds can only explain app 2% of total organic aerosols as you mentioned in the abstract. Thus, the term "organic aerosols" is not suitable. It is better to confine to organic acids which you identified in this study.

Reply: The term "organic aerosols" has been replaced by "organic acids" in lines 98-99, 218, 316, and 382.

-P2, L41-47: Both total diacids carbon fractions in TC and water-soluble OC are higher in PM<sub>10</sub> size mode compared to PM<sub>2.5</sub>. For the reviewer's knowledge, if photochem-

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ical oxidation is important for diacids production, those ratios in PM2.5 mode should be higher than PM10 size mode. Additionally, authors mentioned that heterogeneous reaction on aerosols under high humidity is important for the production of diacids. However, RH during wet season is much higher than dry season.

Reply: The reviewer is correct in terms that RH during wet season is generally much higher than dry season. However, during our sampling period the RH was comparable in wet season (av. 66%) and dry season (av. 64%). The sentence was a bit modified in the revised MS. See lines 177-179.

-P6, section 2.4: Please add S.D. to average value.

Reply: S.D were added to the average value in section 2.4.

-P6, L169-170: Average temperature during the warm wet (avg. 26 °C) and cold dry (24.6 °C) seasons are comparable. The difference is 1.4 °C. What is the threshold value for the separation of warm and cold season?

Reply: The reviewer may be correct in terms that during sampling periods the temperatures are comparable. But the idea was that in Tanzania the dry season (July-September) is generally colder than wet season (March-May). The words “warm and cold seasons” were deleted to avoid any confusion in the revised manuscript.

-P6, L171-172: Please add some evidence of temperature inversion. References or sonde data.

Reply: The reference of Mkoma et al. (2009) was added in the revised manuscript.

-P7, L210-214: Authors mentioned that higher abundance of oxalic acid may suggest enhanced emission from biomass burning, photochemical oxidation, and aging of organic acid during the long-range atmospheric transport. However, the interpretation is not sufficiently underpinned by the data. Authors have to explain more in detail to support your interpretations. Throughout the manuscript, interpretations are too much speculative and vague. Using only one compound data, authors simply characterized

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a certain phenomenon without deep scientific consideration.

Reply: The sentence has been removed in revised MS. Please see lines 215-219.

-P8, L222-223: Interpretation is not sufficiently supported by the data.

Reply: The sentences were reworded in the revised MS. Please see lines 226-228.

-P8, L223-226: Too much speculative. How do authors know C9 is mainly produced from biogenic unsaturated fatty acids emitted from local vegetation? In late part of section 3.1, authors mentioned that fatty acids are emitted from ocean or long-range transported. Which is correct?

Reply: The sentences were reworded.

-P8, L240-243: Interpretation is not sufficiently supported by the data. The reviewer cannot understand how level of ketoacids can explain emission of volatile biogenic precursors from local vegetation. This is certainly not the only possible explanation. There may be several reasons for enhanced concentration of ketoacids during the dry season.

Reply: The sentence (High concentrations for ketoacids were. ...) was deleted because the reasons for possible enhanced concentrations of ketoacids during dry season are explained in the following sentences. We also add a short sentence. Please see lines 247-248 in the revised MS.

-P9, L257-259: The reviewer thinks that photochemical production of  $\alpha$ -dicarbonyls may be enhanced under strong solar radiation during the dry season compared to the wet season.

Reply: We agree with the reviewer. The sentences have been modified and one sentence was added. See lines 259-262 in the revised MS.

-P9, L276 and L280: How did authors know lipid compounds are long-range transported? How about local emission? Can you separate local and long-range transport?

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Reply: The phrase (, suggesting that secondary formation of C9 is significant during long-range atmospheric transport) has been deleted. See page 9 in the revised MS.

-P9, L284-286: Interpretation is not sufficiently supported by the data. Too much speculative. There may be several reasons of lowest C9/C18:1 ratio during the wet season. For example, depressed photochemical degradation of C18:1 during the wet season compared to the dry season and etc.

Reply: The sentences were reorganized and reworded in revised MS. See lines 286-295.

-P10, L290-291: Photochemical degradation may be enhanced under strong solar radiation. However, sky is mostly covered by clouds during the wet season.

Reply: The sentence is modified in revised MS. See lines 291-295.

-P10, L303-304: The reviewer cannot understand the statement in L303-304.

Reply: The statement and related sentence have been deleted in the revised MS.

-P10, L304-307: Enhanced photochemical production is acceptable. However, increased emissions of their precursors are not scientifically supported by the data.

Reply: The sentence on “increased emission” has been removed in the revised MS.

-P10, section 3.3: Authors mentioned PM2.5 and PM10 as fine and coarse mode particles, respectively in the MS. However, coarse mode particle is defined as a difference between PM10 and PM2.5. Is the data shown in the MS for PM10 calculated from a difference of PM10 sample and PM2.5 sample? If not, authors have to avoid using the term “coarse” in the MS.

Reply: We totally agree with the reviewer’s comment. Use of fine and coarse in the MS has been avoided. We just use terms of PM2.5 and PM10 in the revised MS.

-P10, L319-321: Interpretation is not sufficiently supported by the data. How about a

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correlation between Ca<sup>2+</sup> vs.  $\alpha$ -dicarbonyls.

Reply: The correlation coefficients between Ca<sup>2+</sup> and  $\alpha$ -dicarbonyls were added in the revised MS. See lines 317-320.

-P11, section 3.4: In section 3.3, authors mentioned that total diacids mostly present in fine mode. If it is so, total diacids to PM<sub>2.5</sub> mass (or PM<sub>2.5</sub>-TC, PM<sub>2.5</sub>-WSOC) ratio should be higher than those to PM<sub>10</sub> mass (or PM<sub>10</sub>-TC, PM<sub>10</sub>-WSOC) ratio. However, authors showed opposite results in section 3.4. Authors have to clarify them.

Reply: We think that the ratios for total diacids in PM<sub>2.5</sub> to those in PM<sub>10</sub> could not be necessary similar to those ratios of total diacids to PM mass or PM mass to TC or WSOC. Thus, discussions on the relative abundances of diacids in TC and WSOC are kept in the revised MS, but one sentence was added for clarification. Please see lines 351-354 in section 3.4 in the revised MS.

-P12, L359-365: This kind of intercontinental comparison has a high risk. Because total diacids are minor compounds in total PM mass, relative abundance of diacids to PM mass is governed by other major compositions such as inorganic ions.

Reply: We agree with the reviewer's comments, however, since diacids study is limited in the African region we want to make the compassion to get an idea on what levels have been reported in other regions.

P12, L378-380: Too much speculative. Interpretation is not sufficiently supported by the data. In L241-242, authors mentioned that elevated ketoacids during the dry season is due to additional emission of BVOC from local vegetation.

Reply: The sentences have been reworded for consistency and clarification. Please see lines 378-379 in the revised MS.

P13, L418-420: The statement is vague. The reviewer cannot understand how high C<sub>4</sub>/C<sub>3</sub> ratio suggests both biomass burning activities and oxidation of diacids.

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Reply: This section was completely re-written for the clarification. Please see lines 424-435 in the revised MS. The phrase of “biomass burning activities” has been deleted from the sentence.

P14, L422-426: Interpretation is not sufficiently supported by the data. The reviewer thinks that C3/C4 ratios at this site is relatively lower than other sites over the world. Thus, photochemical degradation of C4 is not important for the production of C3 in this study.

Reply: Same above.

-P14, L439: The reviewer cannot understand the term “primary” in L439. Can correlation analysis among diacids and related compounds give information regarding primary emission of C2?

Reply: The sentence has been reworded by removing “primary”. Please see the first paragraph of section 3.6 in the revised manuscript.

-P14, L446: Can correlation analysis among diacids and related compounds give information regarding formation of C2 from BVOC via aqueous phase reactions? Too much speculative.

Reply: The sentence has been reworded in the revised MS. See lines 453-455.

-P15, L458-459, L466-467, L480-482: Three sentences are conflict each other. Authors mentioned that biomass burning is important for the production of C2 and related compounds in L458-459 but not important in L466-467 and again important in L480-482.

Reply: The sentence in L466-467 has been deleted.

-P15, L470-472: The reviewer cannot understand how ratios of dicarboxylic acids to biomass burning precursor between two observation sites can give the information regarding local and regional transport of biomass burning aerosols.

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Reply: The sentence structures were modified and the sentences were reworded in revised MS. See lines 468-474.

-Section 3.7: The interpretations of PCA analysis are quite vague. In the reviewer's opinion, 10 and 11 data set are not enough for PCA analysis. Thus, the reviewer suggests deleting whole section 3.7 and corresponding tables and figures.

Reply: Following the reviewer's comment, we decided to delete the section 3.7 and Table 7. -Section 4, Conclusion: It is better to summarize major findings of the study and their interpretations. For example) -Mass concentrations of C2, C9, wC9, and fatty acids in coarse mode (PM10-PM2.5) are highly enhanced during the wet season than the dry season. -Two biomass burning tracers showed different regression patterns with total diacids in PM2.5 during the wet and dry seasons.

Reply: Conclusion section revised. Please see lines 503-525.

-Figure 6: C6 concentration in PM2.5 is much higher than those in PM10 during the dry season. Because PM10 means sum of PM2.5 and coarse mode (PM10-PM2.5) particles, mass concentrations of compounds in PM10 should be higher than those in PM2.5. There might be errors on data processing. Similarly, Ph concentrations in PM2.5 are much higher than those in PM10 during the wet season.

Reply: Because of the possible contamination of C6 and Ph during sampling/storage/transport, the relevant data may be unreliable. Thus, we decided to delete Fig. 6d,f from the revised MS. -Figure 10: nss-K<sup>+</sup> showed different regression pattern with total diacids in PM2.5 during the wet season. No correlation was observed. However, levoglucosan correlate well with total diacids in PM2.5 during the wet season. Can authors explain reason of this result in the text?

Reply: nss-K<sup>+</sup> may be emitted only by flaming process, but not emitted by smoldering process in contrast to levoglucosan. These points are briefly added in the revised MS. See lines 494-500. Conclusion section was also modified. See lines 516-521.

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Minor comments -Throughout the MS, authors have to check subscript and superscript.

Reply: The authors double checked the subscript and superscript in the revised MS.

-Please check abbreviation. AVG, S.D., MIN, MAX, etc in text and tables.

Reply: The authors revised the abbreviations in MS

-P2, L26: “gas chromatography (GC)” ) “gas chromatography (GC)/ flame ionization detector (FID)”

Reply: The sentence reworded in revised MS.

-P2, L31: The term “C2” have to be defined first.

Reply: The term “C2” was defined first

-P2, L37: “higher to” => “higher than”

Reply: Correction is done

-P3, L57: Remove a comma after dot.

Reply: Comma is removed

-P3, L68: Add “and” after a comma.

Reply: “and” is added in revised MS.

-P4, L87: “reading” => “leading”

Reply: The error is corrected.

-P4, L116: Remove a parenthesis in the phrase (PM2.5 and PM10).

Reply: A parenthesis is removed in revised MS.

-P5, L147: Add “/Flame Ionization Detector (FID)” after “HP 6890”

Reply: “/Flame Ionization Detector (FID)” is added in revised MS

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-P6, L158-160: It is better to delete lines starting “Lower field blanks : : : Yasui (2005).”

Reply: The line is deleted in revised MS.

-P6, L183: It is better to remove the web address of HYSPLIT.

Reply: the web address of HYSPLIT is deleted in revised MS

-P7, L205: Add “and” before “29.2.4”. The term “29.2.4 164.8 ng m<sup>-3</sup>” should be corrected.

Reply: Error is corrected and the word “and” is added before the value.

-P14-16, L447-461: The authors mentioned that OC and EC can be used as a tracer of biomass burning. However, the reviewer cannot understand how EC can be used as a tracer of biomass burning. EC alone cannot be used as a tracer of biomass burning whereas elevated OC/EC ratio can be used as a tracer of biomass burning.

Reply: We agree with the reviewer. The sentences have been reworded for clarity.

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Interactive comment on Atmos. Chem. Phys. Discuss., 12, 25657, 2012.

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