

Response to Reviewer

Abstract Page 3630, Line 1 – There should be a hyphen between water and soluble

Response: It is corrected

Page 3630, Line 11 – Suggest adding (1H-NMR) after resonance since after this the abbreviation is used throughout the abstract

Response: It is corrected

Page 3630, Line 17 – Should methanosulfonate be methanesulfonate?

Response: It is corrected

Page 3631, Line 21 - There should be a hyphen between water and soluble

Response: It is corrected

Page 3631, Line 25 – To be in chronological order the Timonen et al. reference should come first

Response: It is corrected

Page 3632, Line 19 – Should methylsulfonate be methanesulfonate?

Response: It is corrected

Page 3632, Lines 20-21 – I am not sure I understand the part of the sentence from polluted sector on. Is it that a factor for the polluted sector and additional contribution from the clean sector were found? This needs to be clarified.

Response: We revised the statement as follows: “methanesulfonate (MSA), amines, clean marine samples, polluted air mass and clean air masses.”

Page 3632, Line 24 – Suggest changing originated to originating

Response: It is corrected

Page 3632, Line 29 – I am not sure I understand what statistical area means in reference to the Little Rock metropolitan area

Response: The term statistical refers to the definition of metropolitan area by the US Census that has been adapted by the US EPA. Because the term is only used in the US, we eliminate it.

Page 3634, Line 18 – Suggest adding a the before University

Response: It is corrected

Page 3634, Line 19 – The abbreviation for the chemical formula used is not defined

Response: It is corrected

Page 3634, Line 21 – The abbreviation CN is not defined. Also the abbreviations for the chemical formulas used are not defined.

Response: It is corrected

Page 3634, Line 22 – The abbreviation VPDB is not defined

Response: It is corrected

Page 3635, Line 1 – Believe sample and standard should be subscripted

Response: It is corrected

Page 3635, Lines 4-5 – The second thermal optical can be removed

Response: It is corrected

Page 3635, Line 9 – Suggest changing remaining filters were to remaining portion of each filter was. Also, should ultrasound be ultrasonic? Lastly, the abbreviation for the chemical formula used is not defined.

Response: It is corrected

Page 3635, Line 10 – Suggest adding an a before 0.45

Response: It is corrected

Page 3635, Line 11 – Suggest changing transferred in to transferred into

Response: It is corrected

Page 3635, Lines 14-15 - The abbreviations for the chemical formula used are not defined

Response: It is corrected

Page 3635, Line 15 – Suggest changing added in to added into

Response: It is corrected

Page 3635, Line 17 – The abbreviation BBFO is not defined

Response: It is corrected

Page 3635, Line 22 – The abbreviation ACD is not defined

Response: It is corrected

Page 3637, Line 8 – In equation 4 the second α_4 should be α_5

Response: It is corrected

Page 3638, Lines 10-11 – OC/EC ratios are site and source specific. Therefore, I am unsure how the authors know that their ratio indicates biomass and coal combustion since I believe they were just sampling regional emissions of both of these.

Response: The conclusions relies on the analysis of ten years of PM_{2.5} chemical speciation data from the same site and the source apportionment using positive matrix factorization. This analysis has been published last year at Atmospheric Chemistry and Physics and it is cited in the manuscript (Chalbot et al., 2013a). To clarify this point, we revised the statement as follows: “The OC/EC ratio (4.58 ± 1.06) was comparable to those observed in the same region for the 2000-2010 period (Chalbot et al., 2013a) that identified biomass burning and traffic as the most important sources of carbonaceous aerosol in the region.”

Page 3638, Lines 11-14 – The ratios of K^+/K , K/Fe , and mineral elements Al, Si, and Ca the authors mention are not ratios routinely examined in aerosol measurements. Since no literature values or references are provided, it is hard to put these into context. How do the authors know their ratios are similar to those previously observed in the region?

Response: In the revised manuscript, we provide additional information on soluble potassium being a tracer of biomass burning and the range of the K/Fe values. New references are also cited.

Page 3638, Lines 14-15 – The authors mention that the high molar ratio of ammonium to sulfate suggests complete neutralization of sulfate and nitrate. The ratio is over 2, which suggests a non-acidic aerosol. But nitrate is not even included in this. Would it be better to determine and discuss the molar ratio of ammonium/(nitrate + sulfate)?

Response: This is true. We eliminated the discussion about nitrate.

Page 3638, Line 18 - There should be a hyphen between water and soluble

Response: It is corrected

Page 3639, Lines 1, 2, and 7 – The units should be ug C/m³

Response: It is corrected

Page 3641, Line 6 – Suggest adding the word range after 1.8ppm

Response: It is corrected

Page 3641, Line 8 – Suggest changing proton to protons

Response: It is corrected

Page 3641, Line 9 – A period is missing at the end of the sentences

Response: It is corrected

Page 3641, Lines 12-14 – The authors mention that long-chain alkanolic acids, aldehydes, and alkanes were particles with diameters less than 0.96 μm . Is this referring to the samples analyzed by the authors? It is not clear based on the sentence before it and the fact a reference is provided.

Response: It refers to previous studies. We revised the statement as follows: “It was previously observed that long chain ($\text{C}_6\text{-C}_{30}$) *n*-alkanoic acids, *n*-aldehydes and *n*-alkanes accumulated in particles with $d_p < 0.96 \mu\text{m}$ (Kavouras and Stephanou, 2002).”

Page 3641, Lines 22-23 – Should this line indicate that these species were not observed in the coarse particles? The plots referred to in Figure 2 are for the larger size particles and the previous sentence is discussing coarse acetate and formate observed in soil dust.

Response: We revised the text as follows: “These species were observed in the coarse fraction (Fig. 2e-f) but not in fine and ultrafine particles (Figs. 2a-c).”

Page 3642, Line 8 - There should be a hyphen between water and soluble

Response: It is corrected

Page 3642, Line 14 – I believe it should be (Fig. 2a-c)

Response: It is corrected

Page 3642, Lines 16-17 – I believe it should be Fig. 4a-b and c-d

Response: It is corrected

Page 3642, Line 26 – There should be a ppm after 3.77 and 3.91

Response: It is corrected

Page 3643, Line 17 – Suggest changing diameter to diameters

Response: It is corrected

Page 3643, Line 23 – I am not sure I understand the phrase not visible of solvent residue. Is it trying to indicate that the H-3 at 4.64 ppm was not visible due to interference from solvent residue?

Page 3643, Line 24 – Suggest changing of ultrafine and fine ¹H-NMR to of the ultrafine and fine particle ¹H-NMR spectra

Response: It is corrected

Page 3643, Line 28 to Page 3644, Line 3 – I am not sure I am completely following the discussion in these lines. The samples analyzed by the authors were not heavily impacted by biomass burning, it was more background biomass burning that was sampled. So then what does the discussion of H-C-X have to do with the determination of protons from levoglucosan? Is it that the previous assignment was wrong?

Response: The discussion for H-C-X is not associated with the detection of levoglucosan. We separated the test in two paragraphs.

Page 3644, Line 15 – The authors mention that trigonelline was identified in the NMR spectra. Could the authors tell us more about this compound? It is not commonly measured in aerosol samples. So a mention of its prevalence in atmospheric samples could give the reader a better context.

Response: We added additional discussion on the presence of trigonelline on the leaves of oak trees, the prevailing type of trees in the monitoring areas and its detection in pollen particles at the study area. New references are cited.

Page 3645, Line 26 – I believe it should be Table 2 that is referenced

Response: It is corrected

Page 3646, Line 1 – Suggest adding the word different before OC

Response: It is corrected

Page 3646 Line 23 through Page 3647, Line 12 – I believe this section is referring to Table 3, but at no point in the text is Table referenced

Response: It is cited

Page 3646, Line 29 – Suggest changing WSOC that can be interpreted by the condensation to WSOC, that can be interpreted as a result of condensation

Response: It is corrected

Page 3647, Line 6 – Suggest changing an MMAD to a MMAD

Response: It is corrected

Page 3647, Line 23 – The slope mentioned in the text is not the same as indicated in Figure 7a

Response: It is corrected

Page 3647, Line 24 – Suggest changing an CV(RMSE) to a CV(RMSE)

Response: It is corrected

Page 3648, Line 16 – Suggest changing characteristic of water soluble to characteristics of water-soluble

Response: It is corrected

Page 3648, Line 20 – Suggest changing and water soluble to and the total watersoluble

Response: It is corrected

Page 3651, Line 1 – I believe this is an extra line and can be removed

Response: It is corrected

The Decesari et al., 2011, Miyazaki et al., 2009, and Pietrogrande et al., 2013 references are missing from the list.

Response: They are included in the revised manuscript.

Tables Table 1 -The units for elemental carbon should be $\mu\text{g C/m}^3$ -I believe the range for temperature and pressure is being shown in parentheses, but it is not indicated in the caption or column heading

Response: It is corrected and the caption is modified to accurately describe the values for meteorological and air pollution parameters.

Table 2 -In caption, suggest adding the word Mean before Particle -The units for WSOC should be $\mu\text{g C/m}^3$

Response: It is corrected