

Atmos. Meas. Tech. Discuss., referee comment RC1
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Comment on amt-2022-38

Anonymous Referee #1

Referee comment on "Combined organic and inorganic aerosol source apportionment on yearlong ToF-ACSM dataset at a suburban station in Athens" by Olga Zografou et al., Atmos. Meas. Tech. Discuss., <https://doi.org/10.5194/amt-2022-38-RC1>, 2022

This paper describes the analysis of a year long dataset acquired with a TOF-ACSM in Athens, Greece. The paper is well-written and the comparison of two different PMF approaches is interesting and worth publishing. There were a few sections that were confusing. Addressing the following comments should clear those up.

Specific comments:

Comment 1. Lines 119-123: This is not how CDCE is applied. It's not the ammonium to nitrate fraction that matters, but the fraction of ammonium nitrate to total mass loading. Fraction of ammonium nitrate is clearly below 0.4 (based on the time-series in Figure S1 and Table S2), but the particles are also not neutralized, so you need to consider the second part of CDCE for acidic aerosols. Based on Table S2, CE=0.5 is fine for winter and spring, but a bit low for summer (CE=0.55) and fall (CE=0.56). It will only be a 10% error or so to apply CE=0.5 for all seasons, but you should explain why you are doing that.

Comment 2. Lines 250-256: Interquartile to median is a measure of spread or variability, not error so I would not call it an error. Isn't the variability in the mass concentration a reflection of atmospheric variability? It's not clear to me that lower variability necessarily means the PMF factors are a better representation of atmospheric sources. It also seems like a disconnect with the previous paragraph where the combined matrix had higher residuals (worse PMF) but lower spread (better PMF?).

Comment 3. Lines 436-451. This paragraph is very confusing. What is a subtracted f44-f43 plot? You are plotting f44 vs f43 for OOA factors so why do you need to subtract HOA? Or do you mean you subtracted an HOA factor for the raw data points in the background of the Figure S8? In Figure S8, the markers are color coded by month, not

concentration. There is a shading scale for concentration in the legend, but it is impossible to see in the figure. Maybe use size for concentration? The triangle plot does not evaluate the goodness of the PMF fit. It shows if the PMF profiles obtained here are consistent with typical ambient measurements. The description of the trends as vertical or horizontal is not very helpful. Please describe in terms of chemistry, i.e., as more or less oxygenated.

Specific minor comments:

Line 30: This methodology has been published before, so rather than saying this “work presents a new methodology,” say “this work applies a new methodology to a year-long dataset.”

Line 49-50: Do you mean IVOCs? VOCs are typically emitted already in the gas-phase.

Figure 1: I don't find pictures of instrument containers particularly useful and the contrast in the upper left panel is poor. I would suggest making this a 2-panel figure, one panel with a map that shows a scale between the upper left and upper right panels and one panel with the lower right photo. You could put the prevailing winds on the map, at least for winter and spring. In the lower right photo, label the yellow dot and indicate which direction is north.

Line 107: What was the detection limit for your instrument? Which species is this detection limit for?

Line 127: Please describe the approach rather than requiring someone to read the Supplement. Maybe replace “as described in the Supplement (Sect. S1).” with “from the wavelength dependence of the absorption (Sect. S1).”

Line 168-171: This is a very long sentence. Replace “technique application a technique that” with “technique that” and put a period after “size.” Then start a new sentence with “Calculations are repeated...”

Line 174: Since the previous sentence mentions both wind and air mass analysis, indicate which one you are applying CPF to. Maybe start this sentence with “The wind analysis used the” and delete “was used”

Line 182: 1000 m seems really high for back trajectory analysis of ground level measurements. Can you explain why you chose this height?

Line 191: The terminology in PMF can be confusing and I would define the terms in this introductory paragraph. Maybe something like "In the following, profile refers to the mass spectrum of a given factor and variable refers to an individual m/z."

Lines 192-202: The sentence about the mass concentration should go with the next paragraph that describes the mass loading calculation.

Line 200: You downweight the errors for the variables, not the whole species.

Line 212: I'm not sure what "for each matrix" is referring to here. Maybe end the sentence after "number of factors."

Line 243: Maybe "subjective" or "qualitative" is better than "user-dependent" as in "the interpretation of PMF results is qualitative"

Line 247: Aren't OOA's and oxidized aerosols the same thing?

Lines 273-276: A step is missing in this description. Hysplit does not track species, only air masses. Do you mean that the Hysplit back trajectories are colored by the sulfate concentration at the end point? Also, the red blob looks more diffuse in winter, spring and summer than fall so I'm not sure about your conclusion that fall sulfate is more regional.

Lines 294-296: Since the sulfate is not neutralized with ammonia, that suggests more local than regional sources. It would be more accurate to say that the flat diurnal in winter is consistent with regional sources. The afternoon peak in spring suggests local photochemical activity. The increase at night in summer and fall is due to larger changes in boundary layer height compared to other seasons. Make sure that your conclusions from the diurnals are consistent with your conclusions from the Hysplit back trajectories.

Lines 297-299: The formation of NH_4Cl would be higher at night when the temperature is lower. How does that explain an afternoon peak?

Line 339: Change the section heading to make it clear that this section is about the OA

PMF analysis, not organic aerosols in general.

Line 341: Be more specific, e.g., "the mass spectrum from m/z 100 to 200 is depicted"

Figure 3: It is difficult to see what is going on in the panel with both BBOA and temperature. I would suggest a separate y-axis for temperature. Or, alternatively, you could average the temperature to daily values since it is the seasonal trend that matters.

Lines 377-382: Make it clear that these previous studies are high resolution analysis that gives specific ion fragments at m/z 55 and 57. Also, not clear why you are giving chemical names of gas-phase species for ions from particles. I would delete the chemical names.

Line 387: Why surprising? It is the same site.

Line 394: What does "with a total duration of 8 h" mean? Are you comparing only 8 hours of data? If so, specify which 8 hours.

Line 452: Change the section heading to make it clear that this section is about the combined PMF analysis, not submicron aerosols in general.

Figure 4. Would suggest averaging the temperature data to make the panel with both T and BBOA clearer. It's very hard to see RH and T in the panel with AN. I would make a separate panel. The caption should say "aerosol factors" not "sources."

Lines 472-474: Many of these changed by 0.01. Is that significant? Maybe point out one or two cases where there was a significant improvement.

Line 507: Insert "The CPF polar plot in" before "Figure S10" so the reader does not have to go to the SI to find out what S10 is.

Line 522: You need something to transition between these two sentences. Maybe "As noted above for the OA PMF analysis,"

Line 536: I think you mean the "attribution of organics" rather than the "contribution of

organics." This sentence should end in a comma, not a period, because the next sentence is not a complete sentence.

Figure 5. Use the same y-axis scaling for both panels so that it is easier to interpret the discussion in the text.

Lines 557-568: This whole paragraph seems like it should go earlier in this section since it is more of an overview of the results. Also, some of it repeats information that you have already discussed in more detail. And the phrase "will be discussed below" doesn't make any sense since this is the last paragraph of the results.

Table S1: Why is SFBOA in this table? It is not discussed in the paper.

Figure S1: It would be helpful to indicate the seasons with vertical bars.

Figure S2. I would expand the maps so the detail is clearer. Asia and Africa are not relevant to the discussion. Also, it would be nice to have the season label on each panel.

Figure S4. It would help to have species labels on each panel. Same comment for Figure S7.

Table S4: Since NH₄ is a linear combination of SO₄ and NO₃, and in this data dominated by SO₄, correlations with NH₄ do not add any new information. I would delete the two NH₄ rows.

Figure S8: Explain what the black dots are – is this the subtracted data? The shading for size is not visible. Maybe use marker size instead? Not clear what "on the upper size," "on the lower size," and "in the middle" mean. I would delete.

Table S5. Make sure that the tables are in the same order as mentioned in the text.

Technical corrections:

Line 28: No "s" at the end of "aerosols"

Line 37: "causes of" is better than "reasons for"

Line 44: No "s" at the end of "clouds"

Line 45: Start the sentence with "The"

Line 58: No "s" at the end of "vehicles"

Line 64: Start the sentence with "The"

Lines 64-67: This sentence would be easier to understand if you delete "by real-time measurements" at the end and insert "real-time" before "quantification"

Line 66, 68, 93: Define all acronyms: PM1, PMF, GAW, ACTRIS, PANACEA

Line 78: "fingerprint" not "footprint"

Line 96: replace "North east from" with "northeast of"

Line 113: Particles are focused into a "narrow beam" not a "narrow airbeam"

Line 115: Delete "in a tungsten filament" The electrons come from a tungsten filament, but the ionization doesn't occur in the filament.

Line 115: Replace "following" with "according to"

Line 118: Should be a comma before "while" rather than a period before "While"

Lines 146-149: I would put Eq 1 after the sentence that ends "G and F (Eq.1)."

Line 201: Add "s" to "variable"

Line 211: Add "the" before "winter"

Line 214: Delete "s" at the end of "factors"

Line 226: Do you mean "lunchtime" or "mealtime"? It's not "noon" if it's 14:00.

Line 228: Insert "of" between "fractions" and "m/z"

Line 246: Replace "was well-fitted" with "fit the data well"

Line 265: Replace "at" with "in"

Line 278: Replace "portioning" with "partitioning"

Lines 285-287: The end of this sentence is confusing. Do you mean "except for sulfate, for which concentrations were more similar."

Line 287: Replace "rest" with "other"

Lines 292-293: This is a run on sentence. Replace "night, however" with "night. However,"

Line 352: Not clear what "distinctly recognized" means. Do you mean "identified based on its distinctive mass spectrum"

Line 355: Delete the chemical formulas since you have them in parentheses later in the sentence.

Line 357: Replace "most" with "quite"

Line 360: Insert "with the" after "correlated"

Line 363: Insert "The" at the beginning of the sentence.

Line 365: Insert "the" before "HOA factor"

Line 385: Replace "in" with "at"

Line 392: Replace "configuring right" with "separating"

Line 394: Replace "respective" with "loading"

Line 406: Replace "to" with "with"

Line 412: Replace "were very relevant to" with "was highly correlated with"

Line 416: Replace "at" with "in"

Line 417: Insert "the" before "BBOA"

Line 424: Insert "The" before "MO-OOA"

Line 427: Do you mean Figure 3c?

Line 428: Replace "on" with "to"

Lines 455-456: Replace "daily trends...concentration" with "diurnal trends (Fig. 4c) of each factor"

Line 471: Replace "resembles significantly to" with "is similar to"

Line 472: Replace "configuration of this factor with" with "agreement of this factor between"

Line 476: Add "s" to "contribution"

Line 478: Delete "highly" before "agreed" and "s" on "COA"

Line 502: "the ammonium nitrate peak" sounds better than "ammonium nitrate's peak"

Line 505: Replace "leaded" with "led"

Line 506: Delete "s" on "nitrate"

Line 510: "predominantly" not "dominantly"

Lines 530-531: Delete "highly" Delete "s" on "SOAs" Delete "particles" after "sulfate"

Line 534: "and higher" might be better than "while peaked"

Line 573: Replace "sources" with "factors"

Line 577: Open parenthesis missing.

Line 586: Replace "simulations" with "sources"

Line 591: Replace "errors" with "variability" or "spread"

Line 598: Replace "on ambient aerosols...best of information that can be obtained by" with "of ambient...best information from"

Line 605: Replace "finding" with "funding"