

Interactive comment on “Inter-comparison of source apportionment models for the estimation of wood burning aerosols during wintertime in an Alpine city (Grenoble, France)” by O. Favez et al.

Anonymous Referee #2

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General:

The paper presents a study comparing three different quantitative methods for assessing ambient concentrations of PM originating from wood burning. The interest of residential wood burning as a source of ambient PM has grown substantially over the last 10 years, as it is a CO₂ neutral energy source likely to be taken more and more into service in the coming years. The emission factor and the chemical composition of PM emitted from residential wood burning are likely to vary substantially with respect to combustion unit, conditions, etc., thus when assessing such emissions based on the PM chemical, physical and optical properties, deviating results can be expected.

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Hence, a study comparing various methods is timely and worth publishing in a well renown journal as ACP. The comparison of the three methods is based on a period of 14 days, which I find somewhat short for a thorough investigation. I'm quite sure that the winter in Grenoble extends more than the actual number of days.

I share some of the concern uttered by referee 1 concerning the Aethalometer approach and its accuracy. I will not go into details on that as it is covered by another referee.

I find that the paper lacks a thorough final quality check. There are quite a few examples of bad language and there are sentences which lack accuracy, impairing the quality of the paper. I will list some of these issues under specific comments, but have no room for addressing them all.

Specific comments:

Page 1, line 13: suggest rephrasing the sentence to "... indicate a major contribution of wood burning organic aerosols (OMwb) to the ambient aerosol organic fraction, with..."

Page 1, line 14: Please make sure that the numbers for OMwb to OM presented in line 14 are correct (compare with line 15 page 21 and Table 4 page 32).

Page 2, line 11: rephrase last sentence including "wealthiest countries".

Page 2, line 21: Szidat, not Svidat

Page 2, line 23: rephrase to "∼ 40%, respectively for winter time ..."

Page 2, line 29 - 30: rephrase to "High concentrations of water soluble potassium, organic markers and humic-like substances (HULIS),"

Page 3, line 3: remove "often"

Page 3, line 9: change to "These source apportionment models are typically composed by two groups:

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Page 3, line 13: recently made change to “made recently”

Page 3, line 19: change from model to “approach” Page 3, line 24: Use the phrase “In the current study” or “In the present study” instead of “In this study” (se also Page 7, line 17, ...)

Page 3, line 25: I guess the winter season in Grenoble exceeds 14 days. ...

Page 4, line 1: change to “. . . aerosol (Donahue et al., 2009 and references therein). It should be mentioned that the AMS-PMF and Aethelometer models consider the entire ambient OA, while. . .”

Page 4, line 8: change to “for the AMS-PMF”

Page 4, line 14: match with the dates in the abstract. What is correct? 29th of the 30th?

Page 4, line 18: change from lying at to “situated”.

Page 4, line 11: For how long were the filter preheated and at which temperature? Provide this information in the text.

Page 4, line 23 - 26: Is the LPI designed for quartz fibre filters? The jet to plate distance is likely to be shifted towards higher size fraction when using a QFF, this could also be a result of the porous surface of the QFF. I don't think it will affect the results too much, but the authors could consider mentioning it. How did you account for the non uniform deposit of particles on the LPI impaction plates when performing the EC/OC analysis, which is important for the EC/OC split?

Page 5, line 1: remove “basis”.

Page 5, line 2: remove “furthermore”.

Page 5, line 3: what is the size range of the SMPS?

Page 5, line 9: change from precipitations to “precipitation”

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Page 5, line 16: change from newly to “recently”

Page 5, line 19: The second mode of EUSAAR2 ends at 850 °C and not 700 °C.

Page 5, line 19 - 21: If you used both EUSAAR2 and NIOSH 5040, it might be useful to say something about how well they compared to each other.

Page 5, line 19 - 21: If you used both EUSAAR2 and NIOSH 5040, it might be useful to say something about how well they compared to each other.

Page 5, line 26: change from polyaromatic hydrocarbons to “polycyclic aromatic hydrocarbons”

Page 5, line 29: change from insaturated to “unsaturated”.

Page 5, line 22 – Page 6, line 19: There is a pronounced difference in the level of details used when describing the GC and the LC methods. I would appreciate if this section is improved showing more consistency. Also the sentence starting on line 16 and ending on line 17 (Page 6) needs to be improved (which measurements are you actually referring to when using the word “these”. Starting a sentence with “this” or “these” without being followed by an object is typically not a good solution).

Page 7, line 6: change to “R(ATN)⁶ λ, t was determined according to the following equation:”

Page 7, line 26: BCPM1 are estimated from the LPI measurements based on TOA. How was EC converted to BC (Conversion factor?)?

Page 8, line 1: change from time-of-light to “Time-of-flight”.

Page 8, line 14: change from inorganic/organic to “inorganic and organic”

Page 8, line 17: change to “m/z signals”

Page 8, line 26: change from these calculations to “.the abovementioned calculations”.

Page 8, line 17: change to “a satisfactory correlation is obtained between the two

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datasets”.

Page 9, line 8: I suggest the following: “Figure 1 shows the time series of the main submicron aerosol components, i.e., organic matter, ...”

Page 9, line9: change estimated as with “being”

Page 9, line 11: change to “Independently of”

Page 9, line 12 - 13: change from carbonaceous aerosols to “carbonaceous material”, from representing to “constituting”, and from the sum {BC+OM+NO3+SO4²⁻ + NH4⁺} to “of the {BC+OM+NO3+SO4²⁻ + NH4⁺} sum”.

Page 9, line 24: replace furthermore with “also”.

Page 9, line 26: replace here with “in the present study”.

Page 9, line 28: replace latter with “biomass burning” and large with “high”

Page 10, line 1-2: change to “Even when using a low value of 5, wood burning OC (OC_{wb}) accounts for 55% of OC_{total} on average for the period in question ...”

Page 10, line 12: change to “the concentration of a ...”

Page 10, line 16: Should it not be S_{jk} instead of S_{ik}?

Page 10, line 28-29: Could you please be more specific concerning what you mean by delineate the different combustion sources?

Page 11, line 5: replace these compounds with “cholesterol and n-alkanes”

Page 11, line 14 to page 12, line 24: The exercise made by the authors to pick the most “suitable” biomass burning profile very well underlines the great uncertainties associated with biomass burning emissions. There are knowledge gaps such as lack of intercomparison between various analytical approaches for determining monosaccharide anhydrides, non existing data for local tree types (e.g. has the assumed low levoglucosan – mannosan ratio for coniferous trees and correspondingly high ratio for

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deciduous trees been confirmed by other than Schmidl et al. for Europe, and how does the ambient lev/man ratios look like across Europe? Do they correspond to the most commonly used tree types used as fire wood?). The authors exclude two of the profiles because the BBOC to total OC exceeds 1, but the relative standard deviation is in excess of 30%, thus allowing for a BBOC to total OC of 0.7. As far as I can see, the authors have not considered the influence of negative and positive sampling artifact of OC when concluding upon their choice of profile. Hence, one could argue that the profile has been chosen on the basis that it fits the modeling results the best and thus generates fewer problems. However, it could be that it gives the better answer, but not necessarily for the right reason.

Page 13, line 8: remove “of study”

Page 13, line 9: change to “Vehicular emissions contribute on average for 6% of the total OM”

Page 13, line 11-12: Change to “The “other OM” fraction is 26% on average. A diurnal trend was observed with higher contributions of biomass burning to OM during the night (~79%). ...”

Page 13, line 13-15: Remove of (2) and replace are with “were”.

Page 13, line 17-18: Change to: “The CMB analysis clearly suggests a predominance of POA with ~75% of the total OM originating from wood burning, traffic and natural gas combustion.”

Page 14, line 17-18: Change to: “...instrumental error matrix, both typically being ...”

Page 15, line 17-18: Change to: “The best solution following this methodology...”. What are you referring to by this? You need to be more accurate!

Page 15, line 29: the ranges “5-25%”, “25-50%” and “40-55%” is totally decoupled from the HOA, pBBOC and OOA. Please rephrase this.

Page 15, line 30: remove “it has been” and “here”.

Page 16, line 5-21: I’m not quite sure I understand the argument put forward in this section. You find that pBBOA have the same spectral features as those of levoglucosan (m/z 29, 60 and 73), which predominantly appears to be associated with flaming conditions in the study of Weimer et al. (2008) and not with smoldering conditions characterized by “oxygenated fragments” (m/z 18 and 44). On the basis of this you predict that the OOA fraction may contain some mass fragments related to primary oxygenated wood burning OA. Is it then the lack of correlation between levoglucosan and (m/z 18 and 44) which make you conclude that the PMF fail to apportion m/z 18 and 44 to wood burning OA? Secondly, some studies, not AMS/PMF studies, claim that the formation of levoglucosan is governed by smoldering combustion. This is contradictory to what you and Weimer et al. (2008) find. Do you have any explanation for why AMF seem to deviate from this?

Page 16, line 24-25: remove “against” and the “s” in nighttime; replace are with “were” and be maximum with “peak”

Page 16, line 27: change to “However, concentrations of OOA increased on. . .”

Page 17, line 27: change to “Equation [5] can be solved when combined with equations 6 – 9:”

Page 19, line 4: There are three chapters (4.2; 5.2 and 6.2) named results; A more specific text ought to be stated.

Page 19, line 5: Replace within the with “for”.

Page 19, line 8: How was EC converted to BC (Conversion factor?)?

Page 19, line 9: Replace are with “were”.

Page 19, line 16 - 17: with a 4 – 50% range for EC from wood burning I doubt whether you could make this statement.

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Page 19, line 20: replace good with “high”.

Page 19, line 22: change to “. . . the WSOC. . .”

Page 19, line 23: to what extent did WSOC correlate with OOA?

Page 19, line 24: Please provide OC/EC ratios or EC/TC ratios. The wide range of OC to OM conversion factors used makes the OM/EC ratio useless for comparison. Please do also rephrase the sentence.

Page 19, line 26-29: Please provide OC/levoglucosan ratios. The arguments are similar as those raised for Page 19, line 24. Lines 26 – 29 also have a rather poor language and lack accuracy. Please make an effort to improve this. Finally, I disagree with your statement that the aethalometer model is accurate. If you additionally judge its performance with respect to EC, I would say it is rather poor. Please also straighten out your EC/BC terminology.

Page 20, line 1 – 5: Fairly heavy sentence I would say. Some suggestions which might improve it readability: At least replace short time scale with “a high time resolution”; remove “here”; change PMF analyses to “the AMS/PMF approach”; add “. . . ratio from the size distribution measurements using the LPI”.

Page 20, line 7: If there is such a high correlation you might think of adding the correlation coefficient?

Page 20, line 9-10: change to: Such high correlation between OM_{wb} and the m/z 60 signal has been reported previously by Sandradewi et al. (2008c).

Page 20, line 11: change to “aerosol source”

Page 20, line 12: replace well-marked with “pronounced”.

Page 20, line 15: replace against with “as compared to” and drop the s in nighttime!

Page 20, line 15-16: Change to “Maximum contributions of wood burning aerosols were

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observed between 20:00 and 02:00, when OMwb accounted for approximately 70% of the aerosol total organic matter content, whereas a second maximum was obtained after 7:00.

Page 20, line 19: change ranges to “ranged” (please quality assure your paper with respect to the tenses of the verb, your are notoriously mixing past and present)

Page 20, line 20: ?????????

Page 20, line 26: include “the” in front of CMB

Page 20, line 29: “The only source that is apportioned in the three models is wood burning”. What do you mean by that???

Page 21, line 2: replace globally with “generally”.

Page 21, line 5-6: remove “now”; change to “. . .the aethalometer model. . .”

Page 21, line 17-29: I think this section could be condensed. Replace than by “as”. And please state where is “here” in line 23!

Page 22, line 6: change in this to “for by the”

Page 22, line 10: change in this to “accounted for by the”

Page 22, line 16: replace accounted with “interpreted”.

Page 22, line 16 - 17: The content of the sentence is not easily understandable. Please make an attempt to improve this.

Page 22, line 22 - 25: Rephrase and make the content of this section more readily available to the reader. Its content is currently rather vague.

Page 23, Conclusion: Improve language along guidelines provided previously in the review.

Table 4: Check the consistency of C1 in Table caption (360 000) compared to Page 14

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line 15 (C1 = 320 000).

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 559, 2010.