

Interactive comment on “Long-range transport of biomass burning smoke to Finland in 2006” by L. Riuttanen et al.

Anonymous Referee #4

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Overview

This paper is concerned with presenting methods to study transport and lifetime of both gases and particles originating from biomass burning. It is beyond doubt that biomass burning has a detrimental impact on air quality on both the smaller and larger scale, and studying the fate of biomass burning plumes can provide useful insights within this area of research. As pointed out by the authors, biomass burning constitutes a substantial source of both trace gases and aerosol particles to the atmosphere.

In this study, the authors present an extensive data set comprising observations of trace gases and aerosols collected at three different locations in Finland and combine this data with both satellite products and air mass trajectory analysis to study the transport and fate of trace substances and particles in biomass burning plumes. While the

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data set certainly by itself contain information useful for a broader audience with in the field of atmospheric research, the authors fail to present the methods used and results derived in a convincing way. Instead, the methodology applied in many aspects appears murky and it is my belief that several of the conclusions made by the authors in fact must be invalidated after scrutinizing the analysis and methods presented in the study.

Nevertheless, I do think the data and parts of the analysis still deserves publication, but only after the paper has undergone major revisions and the methodology used has been re-evaluated by the authors.

It is thus my recommendation that the authors make changes necessary to either validate the methodology used or removing/replacing the currently excessively qualitative and quantitative parts of the analysis.

Final recommendation: Major Revision

Major comments:

First, the definition, derivation and formulation of “fire sum” is obscure and needs to be clarified.

A large part of this study is concerned with presenting and applying a method to estimate lifetime and concentration of different gases as well as aerosols when air is leaving biomass fire zones in Eastern Europe. This method in turn relies on defining or estimating the contribution to aerosol and trace gas burden caused by biomass burning episodes influencing the air during transport. The authors assume that substances emitted from fire zones will decay by a first order process, and the challenge is to define both the elevation of concentration resulting from biomass burning emissions immediately after leaving the last fire as is the determination of the lifetime of the different trace components.

My prime concern relates to the definition of $\Delta[C](t)$. According to my interpretation

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of the method description, $[C]$ and $[C_{ref}]$ is representative for completely different air masses. Still, the authors use these values to calculate a resulting difference in concentration, $\Delta[C]$, that should be representative for the addition caused by the fire emissions plus the ageing (or decay) during transport from the last fire. During transport, the biomass burning plume will be subjected to chemical decay, various sink processes, dynamical ageing and dilution (mechanisms which also are acknowledged by the authors). The authors further assume in the definition of $\Delta[C]$ that $[C_{ref}]$ represent the regional signal of aerosols and gases minus the fire emissions in the region of transport, although $[C_{ref}]$ in fact corresponds to air arriving from a completely different region which is defined by different sources, sinks and meteorological conditions. Thus, as in the case of the studied spring fire episodes, assuming that the air transported from South-Westerly to Northerly sector is representative of air masses arriving from South-East [minus fire emissions] will inevitable introduce large errors in the subsequent analysis. These errors will mainly be caused by the large differences in regional background of the two air-mass types, which in turn will impact calculations/estimates of both $\Delta[CO]$ and lifetime. For example, it cannot even be remotely suggested that the marine type size distribution presented in figure 5d for the reference period also is representative for transport from the fire sector (but minus the fire emissions). If the authors fail to justify the chosen approach it is my recommendation that this and related discussions are removed from the revised manuscript and instead replaced by a more general discussions of ageing in biomass burning plumes, perhaps adopting a Lagrangian type approach making use of the fact that data from three stations are available downstream of the biomass burning plume.

Thus, with less than showing that $[C_{ref}]$ is independent of transport direction (if no fires are present of course) the derived values of lifetime and $\Delta[CO]$ values lacks relevance as they are based on unjustified assumptions. A language edit is further more recommended as the text currently contains numerous and unnecessary typos and awkward sentences.

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Specific comments:

P4290, l, 12: "Signs...was..." to "Signs...were..."

P4291, l1: "...the properties of and...formation conditions for..."

P 4292, l1: "Studies...gives..." to "Studies...give..."

p4292, l. 5: "in large" to "over large"

p4293, l14-16: Rewrite sentence.

p4294, l8: "eastern Lapland" to "Eastern Lapland"

p4294, l13: "...measurements are made from different heights..."? Perhaps better to reword to observations are performed at different heights or similar.

4294, l16: remove "...measurement..."

P4295, l4: perhaps "sampled from" is better choice than "measured in" in current context.

P4295, l 13: suggest reword to: "The 31 m measurement tower is located on a hill 26m a.s.l. "

p4296, l2: "measured for" to "measured (or used?) in"

p4296, l 6-7: rephrase p4296, l25-26: I doubt that eq1 in fact describes the fire emissions. The authors must do a better job in explaining the physical meaning of the definition of "fire sum"

P4297, l10-11: a description of the other "fire sum" methods and why they were not suitable would clearly be beneficial for the integrity of the MS. As I read it now it seems that the authors just tried different formulation until suitable results was achieved.

P4298, section 2.2.1: The method used to define the spring reference period makes use of properties of a completely different air-mass (marine-mixed air from South-West

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to North) as compared to air likely to be associated with continental air-mass properties from South-East during the actual fire episode. What about the differences in trace gas concentration, aerosols, meteorological history and associated sink processes etc differentiating the two transport conditions even if now fires would be present? It appears a little bit like comparing “apples with pears”. It is for me hard to understand how Cref and Co can be used in same equation to derive lifetime, and the results are very likely to be erroneous. The authors do touch upon this issue on page 4308, where it is concluded that other emissions are of importance as well and that appr. one fourth of observed concentration increase can be explained by fire emissions. Thus, 75% percent is explained by other sources. This statement alone invalidates all calculations of lifetime of trace substances in biomass burning plumes using the method presented in this study. It would be interesting to have seen a plot of some parameters versus the calculated “firesum”.

With less than scientifically justifying the assumptions made I doubt that method is suitable for publishing. The same reasoning can also be applied for the summer period.

P4298, I13: this approach will not provide atmospheric lifetime (see general comment.)

P4299, I25-26: A can agree that a qualitative relation between fires and air quality can be made to some extent using the data presented in this study. I do however strongly doubt that the method (i.e fire sum) is suitable for a quantitative assessment of the impact of fires.

P4299, I23: do the authors mean the summer reference period?

P4299, I25-26: Sentence seems out of place. (“Therefore, it is. . .”)

P4299, I13: “None of the. . .” Is this based on trajectory analysis or due to no signal in measurement data? Somehow this is unclear. . .

P4300, I2: from to in P4300, I.11-12: “. . .reduced to 16km on minimum..” to “. . .was reduced to a minimum of 16km..”

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P4300, I23: "...measurements are available..."

p.4300, I25: What about the other months? On page 4299 it is said that none of the smoke plumes was transported to Värriö. Clarification needed.

P4302, I18: oxdies to oxides

P4302, line5-7: I believe that a strong candidate to explain the difference between Mebust 2011 and the current study is due to the fact that properties of non-comparable air masses have been utilized to derive the lifetimes. The authors seem neglect the fact that air masses from East/South-East will be associated with higher NO_x concentration also in the absence of fire emissions. These arguments are applicable to all lifetime calculations presented in this study. As the selection of reference concentration directly influence calculated values of lifetime and concentration enhancement from biomass burning, my ultimate conclusion is thus that the applied method cannot be used to derive lifetimes in a way that is presented here.

P4305, I7-8: Provide reference for this statement.

P4307, I6: "all BC"?

P.4307, I24: "clear sky conditions"? Please clarify.

P4308, I15-20 and associated table three: following the discussion, it can be concluded, using the data presented in table three, that the fire sum can explain 9% of the variation in accumulation mode concentration ($r^2=0.09$). How does this fact agree with previous statement that the key for distinguish between smoke and clean air episodes lies in the accumulation mode concentration? (see page 4307, I25-27)

Page 4310, line 14-15: Emissions? Change also table caption of table 4 to something more suitable, e.g. net bio-mass burning contribution or similar.

P4310, line 24: Speculate on the reasons for this.

Page 4311, I4-7: Don't follow.

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P4311, I20: "...smoke originated..." to "...smoke originating..."

Figure 5: Why not present distributions on same number scale or even as normalized distributions in same figure (could without loss of information replace volume concentration in figure 5b)

P4311, I23-24: Please explain what is meant with the statement that coagulation widens the size distribution. Sounds counterintuitive...

P4313, I5: expand on what is meant with "...fire sum succeeded to detect fire periods..." Also, again, what does the following statement indicate WRT to applied method "...at least one fourth of variations ... originated from fires..."

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 4289, 2013.

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