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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Particulate matter, air quality and climate: lessons learned and future needs" *by* S. Fuzzi et al.

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Fuzzi et al. reviews impacts of particulate matter on climate, air quality and public health. This paper is a comprehensive review written by 21 experts in the field and it is a great contribution to the community. I would be very happy to recommend publication in ACP. I do not have much to criticize, but I found some sections carry only a few references (there are several paragraphs even without references). Moreover, some sections contain many of old references published before 2010 and only a few of recent papers, and thus, I feel some sections are not fully up-to-date. The authors may check very recent papers and see whether there are any of important aspects discussed particularly in the past few years. I have some specific comments that the authors

should consider as below. Obviously I am not an expert for all the topics discussed in this paper, and I hope the other referee can point out aspects that I did not cover.

We thank the reviewer for the appreciation of our work and also for the criticisms which we address specifically below.

Abstract: I found that the current abstract is kept rather general and does not fully represents what are all reviewed. I would suggest extending abstract; please try to make it clear and specify key features of lessons learned and future needs.

As the Reviewer noted, the present review is rather comprehensive and to put all points and conclusions of the paper in the abstract is impossible. Nevertheless, we believe that the current abstract provides a good summary of the paper content. No changes are therefore made.

P530, L10: There are some recent works regarding chemical transformation of allergenic proteins that I would suggest including here: Gruijthuijsen, et al. Int. Arch. Allergy Immunol. 2006, 141, 265; Shiraiwa et al. Nature Chem. 2011, 3, 291; Reinmuth-Selzle, K J. Proteome Res. 2014, 13, 1570.

References added.

P534, L6: I would suggest including Crounse et al. 2013 (J. Phys. Chem. Lett. 2013, 4, 3513), which proposed autoxidation, a key reaction for ELVOC formation.

Reference added.

P534, L20: Amazon, particularly in rainy season, is also close to pristine conditions. I would suggest including description of some measurement results conducted in Amazon. The importance of preindustrial aerosols can be also emphasized in this section (e.g., Carslaw et al., Nature, 2013).

The paragraph has been modified to take this suggestion into account. The reference Carslaw et al., 2013 has been added.

P540: HOA may not be fully equivalent to POA. HOA connects to chemical composition, whereas POA refers to an emission process. For example, dimers have low oxidation state and may be categorized as HOA, but dimers are certainly generated in secondary processes. It is often assumed in the AMS-PMF community that HOA corresponds to POA and OOA to SOA, but I would say this is still not fully elucidated. Please include some discussion or at least mention this issue.

We agree with the reviewer. The equivalence between HOA and POA is merely empirical and relies on the fact that PMF-derived HOA factors typically exhibit time trends that correlate well with trends in CO or black carbon and other tracers of fossil fuel combustion. Most studies on HOA were based in fact on data from polluted environments. By contrast, at background stations where SOA - including dimers - can dominate OM composition, PMF often fails to extract HOA. Even in countries where biogenic SOA are known to form in high quantities, like Finland, HOA could be extracted from AMS records only at urban background sites (Timonen et al., J. Aerosol Sci. 2013). In conclusion, even if both primary and secondary species with low oxygen content can fit the spectral profile of HOA, most published studies reporting HOA concentrations refer to environments where POA were likely the most important contributors. We have modified the text clarifying this point better.

P545, L13: Please spell out PN (particle number?).

Done.

P558, L13: This paragraph carries no references. There are many works done and please include several of those here.

References added.

P560, L26: For importance of particle-phase pathways for formation of low volatility compounds, please include some more recent references on dimer (e.g., Ziemann & Atkinson, Chem. Soc. Rev. 2012, 41, 6582; Shiraiwa et al. PNAS 2013, 110, 11746.)

and organic salt formation (e.g., Yli-Juuti, T. Atmos. Chem. Phys. 2013, 13, 12507.).

References added.

P562, L14: Why suddenly health effects here? You could omit this sentence, or you need to add more descriptions of health effects.

Sentence omitted.

Figure 12: This figure is not easy to understand. What is y-axis? Does it imply only dust, sea salt and PBAP undergo coagulation and removed via deposition? Coagulation should be relevant for ultrafine particles. This figure should be revised for easier understanding and better presentation.

The figure and the relative caption have been modified for easier understanding (new figure 12 attached).

P563, L10: For vapor wall loss, please also include Matsunaga & Ziemann (Aerosol Sci. Technol., 44, 881, 2010) and Loza et al. (Environ. Sci. Technol., 44, 5074, 2010). Particle wall loss is also an important issue for deriving SOA yields in chamber experiments. Please include a sentence with proper references.

We have rephrased this sentence and added the two suggested references. P563, L15: This whole paragraph is again without any references. Please include any appropriate particularly for values for O:C ratio.

References concerning O:C ratio have been added.

P565: Recently it has been found that organic aerosols can adopt an amorphous semisolid state depending on temperature and relative humidity (e.g., Virtanen et al., Nature, 2010; Koop et al., PCCP, 2011). It challenges traditional view of gas-particle interactions including gas uptake, chemical aging, SOA formation, aging and partitioning and CCN/IN activation. This is a hot topic in the aerosol chemistry community and many studies have been conducted in the past few years. I would suggest including

more discussion on this aspect.

We agree with the reviewer. Two new paragraphs have been added to discuss this subject.

P566, L7: For organosulfate, I suggest including Iinuma, Y, Environ. Sci. Technol. 2007, 41, 6678.

Reference added.

P566: Dry deposition of particulate matter is reviewed extensively. Wet deposition is also as important or even more important as dry deposition for certain conditions. I would suggest that wet deposition of particulate matter should be also reviewed.

The reviewer is correct, it is an omission. Our wish was to bring the process understanding of dry deposition of particulate matter up to date. This seemed important as there were important disagreements in the literature a decade or so ago, and these have been largely (but not entirely) resolved. We had not intended a similar exercise for wet deposition, in part because there has been much less mechanistic work on wet scavenging processes. However, it is an omission and we have added a short section of text to rectify this omission with additional references. A more extensive review of wet deposition would add much more to the length than the value of the paper. It is notable that the interest in phoretic processes and within and below cloud scavenging, mainly from the days of radioactive fallout, has not really advanced much.

Section 2.4: This big section cites only a few papers published after 2010. Are there not much progress made in this field after 2011?

There are relatively few papers on the process understanding after 2010, though we do cite one in 2011 and one in 2012. Most of the recent papers on fluxes are focussed in urban areas and are on emission, not deposition.

P571, L12: Lelieveld, J et al. (Atmos. Chem. Phys. 2013, 13, 7023) and Giannadaki et al. (Atmos. Chem. Phys., 14, 957, 2014) could be included here for estimation of

premature mortality due to air pollution and dust, respectively.

References added.

P578, L20: Naphthalene SOA exhibits redox activity (McWhinney et al., Atmos. Chem. Phys., 13, 9731, 2013). You could also check some other studies using DTT assay to examine oxidative potential of PM.

Two references have been added.

P581, L12: In addition to formation mechanisms, formation kinetics would be also necessary.

Change made as suggested.

P583: I would suggest mentioning about hazardous air pollutants (HAPs) and toxic air contaminants (TACs) (e.g., Finlayson-Pitts and Pitts, Chemistry of the upper and lower atmosphere, Academic Press, 2000).

We were dealing in this paragraph only with pollutants which are regulated directly in EU legislation and there are many HAPs and TACs which are not. In any case, we have changed the sentence to include also these species as suggested by the reviewer.

P584, L24: You need several references here. Otherwise you cannot write “these studies”.

References added.

Figure 28: Is it known why there are peaks for both nitrate and sulfate around 2003?

Sulphate and nitrate peaks in 2003, arise from the prolonged period of very high temperatures and intense photochemical activity in Europe in that year.

P597, L12: Recent studies have shown that glassy SOA can nucleate ice. It is worth mentioning this aspect (e.g., Wang et al., J. Geophys. Res. 117, D16209, 2012; Schill et al., Environ. Sci. Technol., 48, 1675, 2014; Berkemeier et al. Atmos. Chem. Phys.,

14, 12513, 2014).

Short sentence and references added.

P602, L8: Please include reference here.

See next comment.

P602, L9: Jacobson, 2001 is not a lab study. Please include lab works here (ex. Schnaiter et al., JGR, 2005; Zhang et al., PNAS, 2008).

We have rephrased the paragraph and corrected the references.

P602-603: Refractive index of BC is still uncertain and that of BrC are highly uncertain, which directly affect the assessment of their radiative forcing. This may be worth mentioning.

The reviewer is correct. In fact, the literature provides fairly diverse refractive index values for (uncoated) BC. We have added this point to the discussion. Furthermore, the refractive index of BrC is even more uncertain. Very recently, Moise et al. (Chem. Rev. 2015) presented a comprehensive compilation of laboratory and field data of light absorbing organic particle optical properties. We have added a reference to this review.

P616: Would it be possible that ocean acidification affects biological activity in the sea surface microlayer that eventually affects emission of sea spray aerosols and biogenic precursors?

We are not aware of any studies of ocean acidification and aerosol emissions, therefore we don't want to speculate in the present review.

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 521, 2015.

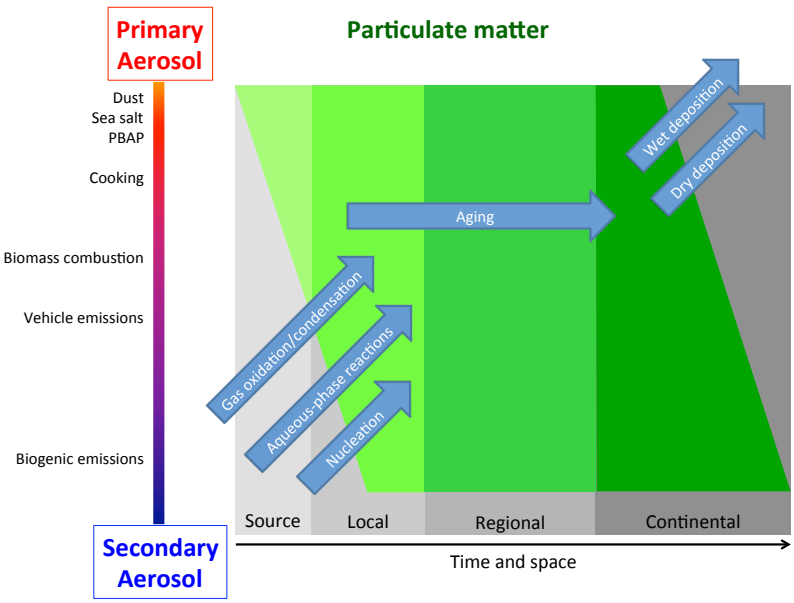

Fig. 1. Fig. 12 - Sources and processes affecting atmospheric aerosol composition from near-source to continental scales. The y-axis indicates the relative importance of primary vs. secondary aerosol for a gi