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Interactive comment on "A naming convention for atmospheric organic aerosol" *by* B. N. Murphy et al.

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article

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Response to Referee #1

(RC1.0): Firstly, the authors are to be congratulated on attempting to bring an improved system to the complex world of organic aerosol nomenclature. The paper is well written, makes many good arguments in a logical manner, and after some changes deserves to be published as a valuable contribution to an important discussion. My feeling is still that the organic aerosol world is so messy and changeable that it will outlive even this valiant new attempt at organisation, and while reading this paper I couldn't help but think of other ways to name such compounds. Still, the main test of such a contribution is time – whether the community finds it useful or not, and one can hope that in any case this paper will stimulate more careful consideration of OA properties and definitions.

(AC1.0): We thank the reviewer for his thoughtful reading and helpful feedback. We agree that the main test of such an effort will be time, but with the reviewer's suggestions, we feel we have been able to put forth an even more holistic and useful scheme. Below we discuss individual issues and corrections.

Major issues

(RC1.1): 1. The scheme is essentially 100% VBS in character, so most suited for modellers who make use of that framework. However, even the original 1-D VBS framework is giving way to 2 D versions, recognising that volatility alone is insufficient to characterise OA. Why didn't the author's propose a scheme which accounts for this 2-D framework?

(AC1.1): The reviewer's point is appreciated and we also of course recognize the insights that have been gained in the aerosol research community from considering oxidation state. However, the goal of our proposed scheme, and the paper, is not to present an exhaustive, all-encompassing nomenclature that accommodates every attribute of OA that is interesting to the organic aerosol research community. Rather, our intention is to distill those characteristics that we think are most vital to discussion with the broader scientific and public policy communities. These characteristics (primary/secondary pollutant affiliation, phase, volatility, and emission source) are adequate also for a wide breadth of the organic aerosol community. Indeed, some of the individual elements of the framework we propose is not new, but is already in common use, albeit with confusing variations, in ongoing research.

But, more central to our focus, the characteristics we have chosen connect particulate mass and number concentrations, which are important for assessing societal impacts, directly to mitigation strategies, and, in this sense, stream-line a lot of possibly confusing complexity. We realize that the first draft of the manuscript did not clearly communicate this motivation, and so we have rewritten the abstract, added two paragraphs to the introduction, and added some to the conclusions to clarify this extremely important point. These revisions follow:

Abstract: ”While the field of atmospheric organic aerosol scientific research has experienced thorough and insightful progress over the last half century, this progress has been accompanied by the evolution of a communicative and detailed yet, at times, complex and inconsistent language. The menagerie of detailed classification that now exists to describe organic compounds in our atmosphere reflects the wealth of observational techniques now at our disposal as well as the rich information provided by state-of-the-science instrumentation. However, the nomenclature in place to communicate these scientific gains is growing disjointed to the point that effective communication within the scientific community and to the public may be sacrificed. We propose standardizing a naming convention for organic aerosol classification that is relevant to laboratory studies, ambient observations, atmospheric models, and, quite importantly, the various stakeholders for air quality problems. This framework classifies organic material as primary or secondary pollutants and distinguishes among fundamental features important for science and policy questions including emission source, chemical phase and volatility. Also useful is the addition of a suffix describing the volatility of the organic material or its precursor when emission occurred. With this framework, we hope to introduce into the community a consistent connection between common notation for the general public and detailed nomenclature for highly-specialized discussion. In so doing, we try to maintain consistency with historical, familiar naming schemes, unify much of the scattered nomenclature presented in recent literature, reduce the barrier of comprehension to outside audiences, and construct a scaffold into which insights from future scientific discoveries can be incorporated."

Introduction: "Atmospheric aerosols have documented, although not fully understood, impacts on public and environmental health as well as the climate system (Seinfeld, 2004; Seinfeld and Pandis, 2006; IPCC, 2013). As a result, researchers have worked for decades to quantify these impacts and to report their findings back to the public in hopes of improving societal outcomes through better-informed decision-making. Without a doubt, scientific understanding of atmospheric aerosol in general and organic aerosol in specific has grown considerably over the past two or three decades (Seinfeld and Pankow, 2003; Seinfeld, 2004; Fuzzi et al., 2006; Donahue et al., 2009). The atmospheric aerosol research community has invented and refined a large number of analytical techniques and conceptual models to characterize the observed complexity of the corresponding pollutants and their precursors. As understanding of aerosols has been refined so has the language used to describe it. With the application of each new instrument, our ability to sort and classify pollutants according to previously undetectable properties (e.g., oxidation state) (Zhang et al., 2011) has emerged, or our ability to resolve familiar properties has increased (e.g., at one time separating particulate matter with diameter smaller than 2.5 μm from the larger particles was considered challenging; now the scanning particle size magnifier enables detection of particles smaller than 2 nm) (Kulmala et al., 2013).

"Concurrent with such advances, new labels typically arise to identify new categories or properties in order to facilitate discussion of any differences among them: different societal impacts, different emission sources, etc. As a result, the field has developed several useful schemes for classifying organic aerosol (OA) that are logical, self-consistent, and effective (see for example Seinfeld and Pankow, 2003; Pöschl, 2005; Fuzzi et al., 2006; Donahue et al., 2009; Mohr et al., 2009; Ervens et al., 2011; Zhang et al., 2011). But when viewed as a whole, the landscape of jargon, operationally-defined terms and subtle caveats in OA research nomenclature has become imposing even for scientific experts, and insurmountable to stakeholders."

Conclusion: "By introducing a standardized rule for communicating phase state via subscript, the scheme emphasizes the importance of phase transitions and dynamic interactions observed by the scientific community while maintaining consistency with the terms used to communicate to the policy community."

(RC1.2): 2. I wonder in particular why a stronger link to O:C ratios was not made. This information is readily available from both models and measurements these days, and provides a natural link to both the 2-D VBS approach and observable quantities.

(AC1.2): Please refer to author comment AC1.1 above. We completely agree that O:C ratios are an important property to consider for the OA system. But we do not consider it of vital importance to communicate to the broader community at the expense of clarity. If future analyses reveal a more direct link between O:C ratio and environmental impacts (e.g. through CCN activity or dose-response relationships) such that it becomes compelling to discuss oxygen content of organic pollution with the outside community, then we look forward to revising the proposed, quite modular, scheme to accommodate O:C ratio. We have added the following discussion to the text:

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Section 2, Second to last paragraph: ”One can think of other attributes to accommodate with this scheme: for instance, oxidation state (or O:C ratio) or functional group as detected by Fourier transform infrared spectroscopy (Russell et al., 2011). Although there is a wealth of data and analysis in recent literature showing the importance and usefulness of these properties, volatility is most directly relevant for gas/particle partitioning under typical atmospheric conditions, which is most relevant for predicting condensed-phase concentrations and societal impacts. Oxidation state can be further incorporated into this scheme if future observations warrant (e.g. if a direct connection to negative human health outcomes is shown).”

(RC1.3): 3. If using VBS species, one knows if the C* value is 1 μg m-3, 10 μg m-3. Why lose information by converting to broad classes such as LV, SV etc. As an example, why does a shift in C* from 0.1 to 0.001 μg m-3 not result in a change of nomenclature (both are LV-), whereas one from 1000 to 100 μg m-3 does, from IV to SV? (Why not use the log10(C*) values as an index instead of broad letter codes?)

(AC1.3): This well-taken point also reflects the debate we try to clarify in AC1.1 above. Our intention is to maintain reasonable quantitative precision in the context of informing broader scientific and public policy decision-making while simultaneously standardizing the terms we in the aerosol community use commonly. These are terms we hope will be useful in reporting results but also in submitting proposals, writing blog articles, and commenting through popular media channels. We subsequently would expect the outside community to preferentially latch onto broad classes (e.g. SV-) that readily map to descriptions (e.g. semivolatile) that are useful for broader discussions. We have added the following lines:

Section 2, Third to last paragraph:”Furthermore, the nature of the proposed naming framework, with its reliance on alphabetic rather than numeric identifiers, is well-suited

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for communicating broad concepts quickly, rather than relying on the audience's knowledge of the relationships between log10(C*) and partitioning."

(RC1.4): 4. The suggested nomenclature seems very logical in many ways, but then exceptions occur. Notably, on page 29987 the grouping aqSOA is introduced, with no distinction between anthropogenic, biogenic, biomass-burning or any other source. Why are aerosols produced by aqueous pathways not treated in similar ways to aerosols produced via the gas-phase? As another example, how should one denote an organic nitrate formed from anthropogenic NOx and biogenic VOC? Or oligomers of both ASOA and BSOA compounds? These types of problems suggest to me that many papers will have to re-invent terminology anyway.

(AC1.4): We agree that the inconsistency between aqSOA and the other source terms is indeed problematic. It is also illustrative of a fundamental issue in atmospheric pollution science: how does one propagate classifications from precursors to products? Incidentally, this is why we try to focus the scheme on communicating attributes of organic compounds at definitive states (i.e. emission and observation). The debate about the meaning secondary OA for instance is an example of similar confusion, since it centers not around what phase the OA started or ended as (by definition it begins in the gas phase and ends in the particle phase) but on how it was transformed (i.e. through condensation alone or through condensation before or after some reaction). We have solved this confusion by introducing explicitly the volatility suffix, thereby freeing the "S" in SOA to identify material that has actually reacted in the atmosphere.

In principle, one could replace or preface the "S" with other designations for different formation pathways such as high-NOx, aqueous-phase, etc. However, as the reviewer points out, if they mix and form new products, it is completely unclear what label should survive. To avoid this confusion, we have eliminated aqSOA from the proposed scheme. We have added the following discussion to the text:

Section 2, Fourth paragraph: ”We have chosen to omit the common modifier ”aq” for identifying organic aerosol formed during reaction in an aqueous phase. While this formation pathway is an area of intensive and exciting study, our scheme focuses on attributes of OA relevant to its state at emission or current state, not on the formation process. Hypothetically, confusion arises when OA compounds formed via gas-phase reaction react in a dry particle with compounds formed via aqueous-phase reaction to form new products. Is this product material also aqSOA? More information is needed regarding the importance of this formation pathway, and the fate of its products in order for it to be accurately incorporated into the proposed scheme. Meanwhile, the phase subscript offers a method to identify hydrophilic organic material currently in an aqueous phase.”

The concept of mixing source terms is also problematic, as the reviewer notes. This discrepancy is manifested in other atmospheric science applications as well: for instance, when applied to particle number concentrations and coagulating particles from distinguishable sources. However, it is vital that an effective naming scheme be capable of communicating source information to the outside community; this is from a certain perspective, its most important task. For that reason, we argue that the modifiers for source type as currently proposed are critical to the usefulness of this scheme.

(RC1.5): 5. OM, OC, OCA? Here I am not sure. The traditional use of OM and OC has been for the particulate phase, synonymous with OA. The authors are both logical and consistent here in defining OM = OG + OA, but there is significant potential for confusion with respect to other papers. Why not simply add ’T’ as prefix, thus TOM or similar to TOOC as used in Heald et al. (ACP, 2008)? Personally, I would have preferred suffixes, similar to chemical practice, e.g. OM(g), OM(p), OM(t) could have represented the gas, particle and total concentrations. This could be easily extended to OM(aq), OM(glassy), etc., and would be consistent with today's use of such phase identifiers in the standard chemistry literature.

(AC1.5): We thank the reviewer for his useful insight on this aspect of the framework. We have modified the scheme to incorporate phase explicitly with a subscript following the two-letter token, OA. Thus to refer to the vapor phase concentration, one would use OAvapor (or OAv) while total OA would become OAtotal (or OAt). To refer to organic carbon, the OA is straightforwardly replaced with OC. We have also introduced an important caveat. When a subscript is omitted, the acronym refers to just the condensed-phase material (i.e. OA, OAp, and OAparticle are all synonymous). This keeps consistency with the outside community, which likely does not have much use for quantities like OAvapor or OAtotal anyway. We have modified discussion in the text to read:

Section 2, Second paragraph: "The root name (Table 1) along with the phase subscript concisely describes several aspects of the organic species of interest. This core of the framework is the most familiar to the atmospheric aerosol community. It begins with designation of the species as a primary ("P") or secondary ("S") constituent followed by a token identifying all of the organic material ("OA") or just the material from carbon ("OC"). Here, we define pollutant material as primary if it has not undergone a chemical change in the atmosphere (i.e. it could have changed phase). Pollutant material is secondary if it has been chemically changed in the atmosphere. The specific phase of the material is then identified with a subscripted word (or letter for shorthand): particle ("p"), vapor ("v"), or total ("t"). One can easily identify other phases as well (see Table 2). We finally propose one critical addendum to this scheme. If no phase subscript is provided (e.g. OA, POC, SOA), the acronym is assumed to refer only to condensed-phase material (i.e. no subscript “p” is needed). This addition effectively clarifies discussions with the non-aerosol and non-scientific community when the distinction of phase causes more confusion than clarity and is anyway subordinate to broader issues.”

(RC1.6): The definitions and discussion of POA assume that emission factors are largely derived from dilution sampling. Such emission factors are often derived from ambient sampling though, e.g. tunnel experiments or regressions compared to other pollutants. How should such POA emissions be defined and indicated? (The discussion leading to the 320 μg m-3 criteria for POA in section 4 is difficult to apply when dilution sampling is not the source of the emission factors.)

(AC1.6): To apply emissions data to the volatility-focused framework we outline, an experiment only needs to note the temperature and OA ambient concentrations for which these data are derived. These two pieces of information are enough to characterize at least broadly the volatility of the species (ignoring mixing effects which are outside the scope of what is currently feasible).

(RC1.7): Table 2 POA, POG - what about compounds emitted as gases (POG) at high temp, but which condense on cooling? These are excluded from the POA and SOA definitions.

(AC1.7): Compounds emitted at high temperature are not excluded from the current definitions. All of the volatility classifications are specified at 298 K. If a species condenses upon cooling in the atmosphere, it is still POA because it has not reacted (please refer to AC1.4). Clearing up this confusion is also discussed in detail throughout section 4. Going forward, it is important for emissions measurements to likewise report the temperature of the measurement so that emission factors can be corrected to ambient temperatures.

Smaller issues

(RC1.8): p. 29985 Add some references to support the statements on the 1st lines here
(AC1.8): We have added appropriate references to texts and literature surveys.

(RC1.9): p. 29985, last line. I think the sentence that ”This view was at odds with what a traditional....” reflects only certain model setups. I think the reason for many of the low SOA/POA ratios found in some of the early modelling studies was the use of very low yields, and other model-specific assumptions. Even in the earliest EMEP OA models (Andersson-Sköld and Simpson, JGR, 2001, Simpson et al., 2007), we found very large SOA/POA ratios.
(AC1.9): We have added the following lines to the text:

”Following the common assumption that HOA generally aligned with POA and OOA with SOA, some traditional models were predicting SOA/POA ratios that were extremely low compared to observations (Volkamer et al., 2006; Shrivastava et al., 2008). Results from the European Monitoring and Evaluation Programme (EMEP) Model, using higher SOA yields, showed higher SOA/POA ratios in suburban/rural areas of Europe (70-80%) (Andersson-Sköld and Simpson, 2001; Simpson et al., 2007). Still, those results were consistent with the lower bound estimates of the very large OOA/HOA ratios measured by the AMS at urban, suburban, and remote sites during later campaigns (Kulmala et al., 2011; Crippa et al., 2013).”

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(RC1.10): p. 29988 Line 8. Lanz et al. 2007 is not a good reference for ^{14}C work (they used AMS + PMF). One of the Szidat et al. papers would be better for the Swiss studies on this issue. It would be good with some non-European studies also.
(AC1.10): We have updated the text.

(RC1.11): p. 29988 Explain what assessment methods 5, 201A and 202 are.
(AC1.11): We have updated the text with the following descriptions:

Section 1, Third to last paragraph: "Assessment method 5 dictated that particulate matter be withdrawn from the source collected on a glass fiber filter maintained at a temperature of 120±14 °C. The particulate matter mass was defined as any material that condensed at or above the filtration temperature. By encouraging methods 201A and 202 (FR, 2010), the agency acknowledged the importance of capturing both filterable (particulate mass at stack temperature without dilution) and condensable particulate matter (particulate mass at 30°C without dilution), respectively. Method 201A built upon method 5 by introducing a PM2.5 cyclone before the collection train and specified a cutoff temperature (30 °C) for stack operation. If this cutoff temperature was exceeded, the "stack tester" was required to complete method 202: capture condensable particulate matter in a dry impinger, extract the material with hexane and water, dry the sample and weigh it."

(RC1.12): p. 29988 Line 27, "at" low concentrations.
(AC1.12): Fixed.

(RC1.13): p.29990, line 10. Add base-10 to qualify logarithmic average.
(AC1.13): Added.

(RC1.14): p.29991, line 5. Please give some hint as to how this alternative looks, it is frustrating to have to start guessing at this stage.
(AC1.14): This statement no longer appears in the revised text.

(RC1.15): p.29993, line 4. Vegetation (and other biological systems) can emit oxidised VOC, some of which must end up in aerosols even without chemical reactions. This would seem to be outside your SOA definition, but would be measured as OOA?
(AC1.15): We have revised the relevant lines to read:

"There are exceptions because some primary compounds, notably biomass burning emissions and some biological VOCs, may be partially oxidized upon emission. These compounds are technically POA under the proposed framework."

(RC1.16): p.29993, line 23. Here I disagree that one can call the proposed framework quantitative, when the span of volatility can be four orders of magnitude.
(AC1.16): We have replaced the term with "semi-quantitative" to soften the weight of the statement.

(RC1.17): p.29996, items 2 and 3. If SOA mass evaporates and reacts, then the loading must change too. These reactions normally add oxygen to the system.
(AC1.17): We completely agree with the reviewer's point. For each of those statements, we emphasized that the carbon mass itself remains unchanged.

(RC1.18): p.29997, line 5. The statement here that the proposed system is consistent with 'ongoing field and laboratory' studies is misleading I think, except in the sense that the very broad categories OA, SOA, etc. can still be used. As noted on page 29994, field campaigns will have limited or no access tothe information needed.

(AC1.18): We have rephrased this statement to read: ”This system is consistent with general classifications used in many field and laboratory studies, as well as developing model frameworks.” We have also added, in response to the other reviewers, examples of how field and laboratory campaigns can utilize the information contained within this framework.

(RC1.19): p30000 The FR references need more information.
(AC1.19): Added.

(RC1.20): p30007, Table 2. Better to have the Base Terms before the Modifiers, since the latter make use of the former.
(AC1.20): We have reorganized the tables in response to the modifications in the overall scheme.

(RC1.21): I think Table 4 confuses more than it helps. The SV-OOA here includes both 0.01 and 100 μg m-3, which conflicts with Table 1, and the qualifier ‘in the literature’ is too vague to be helpful.
(AC1.21): We have added references to the table and a statement recommending readers to assume SV-OOA map precisely to the definition of semivolatile in the proposed scheme, due to the uncertainty in the measurements and broadness of the classification itself.

(RC1.22): Table 5 is also confusing, Here we can have compounds like SV-SOA-sv which have C* of between 1–100 μg m^{-3}, classified as POA in the ‘traditional framework’. A problem here is whose traditional framework one refers to. Many emissions in Europe have been derived from ambient data with concentrations of order far lower than 100 μg m^{-3}; the definition of POA in this case becomes rather tricky.

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(AC1.22): The reviewer correctly points out a problem here. In line with our focus of presenting to the field a framework that can be understood by the broader community, we have altered the labels here from ”Proposed” and ”Traditional” to ”Detailed” and ”Simplified”. However, a discrepancy for defining a compound like SV-SOA-sv at different OA loadings would not seem to pose a problem currently given that emissions for European models like EMEP specify NMVOCs and particulate organic matter (Simpson et al., ACP, 2012). There are exceedingly few models that explicitly treat the production of SOA from semivolatile emissions, although the US EPA is rapidly probing this frontier (Pye and Pouliot, 2012). Given this attention, we feel it all the more important to standardize the terminology across political and academic borders.

(RC1.23): Finally, and this is personal taste of course, I find these ascii text strings such as SV-bSOA-v a little inelegant, looking more like equations rather than chemical compounds. Again I would have considered some sub and superscript notation.
(AC1.23): We have revised the scheme with this comment in mind and look forward to the reviewer's and community's feedback.

References

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