

Interactive comment on “The hygroscopicity parameter (κ) of ambient organic aerosol at a field site subject to biogenic and anthropogenic influences: Relationship to degree of aerosol oxidation” by R. Y.-W. Chang et al.

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I find that this is a nice study and I recommend it for publication in Atmospheric Chemistry and Physics. In particular it adds new data that fits into the unifying organic aerosol modeling framework recently proposed by Jimenez et al. (2009). However, I also find myself left with some conceptual difficulties regarding the advocated relationship between O/C and kappa as well as the derivation of organic aerosol kappa. I suggest that the authors provide better theoretical and experimental constraints and add a number of qualifications before this

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manuscript appears in print.

General comments

It seems that over the last few years a dramatic shift occurred regarding aerosol-to-CCN closure studies. Not too long ago, we as a community collectively failed to measure aerosol composition and predict CCN concentrations adequately (see McFiggans et al., 2006, ACP; Kreidenweis et al., 2009, Cloud particle precursors in Clouds in the Perturbed Climate System: Their Relationship to Energy Balance, Atmospheric Dynamics, and Precipitation. Struengmann Forum Report, vol. 2, Ervens et al., 2009, ACPD, and Kammermann et al., 2010, JGR for a list of studies), with a systematic trend to overpredict CCN concentrations from size and composition data. Recently, this trend has been reversed and closure is achieved more often than not. It is unclear to me what aspect of the measurements improved. My personal view on the failed closure story has always been the difficulty to match concentration measurements (CCN and integrated CN), as well as the propagation of sizing errors due to the high sensitivity of the Kohler equation to dry size. I should note that at least in the earlier studies there has been great difficulty to reconcile closure conditions with composition.

The conceptual shift that has taken place is that closure is now a given, and that closure conditions are well enough constrained to infer the partial effects of a single sub-component of the aerosol, here the organic fraction. While the numbers presented here are plausible, I still also consider them highly uncertain. In particular, the issue is that the organic kappa reported here subsumes many of the perhaps compensating measurement errors. An incomplete list would be sizing errors, shape effect errors, mixing state uncertainty, and counting errors. For example, how much does the derived organic kappa change if the CCN instrument counts 10% low? How representative is the organic aerosol kappa, if the ratio of organic/inorganic fraction varies with size,

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if it leads to only 'average' closure? How well can hygroscopic inorganic and insoluble inorganic compounds be subtracted when the AMS measures only the non-refractory composition? How much do assumptions about the chemical state of the inorganic compounds matter? In summary, my criticism is that the derived organic kappa values are at least more uncertain than in studies that measure kappa more directly. To address my concerns the authors should present additional analysis that highlights these uncertainties. For example, the authors might want to do a classical error propagation analysis and report that in a supplement. Alternatively, plots of organic fraction vs. total kappa (e.g. Shinozouka et al. 2009, ACP) could show how noisy the data are, and how well organic kappa can be inferred from subtracting inorganic components. In particular the authors should be sensitive to the problem of overprediction at $k_{org} = 0$, which clearly reveals other experimental issues at play.

The reviewer offers an insightful and thoughtful discussion on the current understanding of attempts to assess ambient organic aerosol hygroscopicity from field CCN studies. The paper has been extensively re-written to address the reviewers's comments.

We agree that there has been a shift in the field whereby full closure is now assumed to be possible, and derivation of more specific aerosol properties is then attempted. This approach is probably now possible with improved CCN and on-line composition measurement capabilities. And, we fully agree with the comment that the uncertainties inherent in this (new) approach must be made clear. We hope the revised version of the paper satisfies the reviewer in this regard. In particular, refer to the "Uncertainties" Section (Sect. 4.3) of the new manuscript. Also, note that the assumptions are now repeated throughout the paper (Abstract, Sect. 3.1, 3.3, and 5).

As a specific example we now state that the uncertainty in the counting accuracy of the CCN counter is 20% (Sect. 2.2). This error estimate arises principally from errors in the counting accuracy of the Aerodynamic Particle Sizer, that counts the number

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of activated droplets, as well as the measurement of the sample flow. However, note that the statistical method used to evaluate k_{org} and a compares the mean ratio of predicted to measured CCN concentrations, and so our results are not sensitive to systematic uncertainties in the total concentrations of either the SMPS or the CCN counter.

When I first saw the proposed relationship $k_{org} = 0.3 * O/C$, I was immediately tempted to 'break' this relationship. The reason for this is that we recently published a paper on the relationship between molecular size and kappa, and showed that the Flory-Huggins entropy of mixing well describes kappa over a broad range of molecular sizes. When we selected compounds to study, we did so based on O/C ratio. In particular we ran $O/C \sim 1$ (carbohydrates) for $C = 3..18$, and $O/C \sim 0.5$, for $C = 6..>60$, with the hope to learn something about the relationship of O/C and kappa. For example, maltotriose hydrate has an $O/C \sim 1$ but kappa of ~ 0.055 (Petters et al., 2009, GRL, Table S1), which does not conform to the proposed relationship here. Taking the argument to the limit, one might conclude from our work that the extent of oxidation has nothing to do with kappa. To add to this critique, one can also break the proposed relationship another way, by considering nitroglycerin (1,3-dinitrooxypropan-2-yl nitrate), which has an O/C ratio of 3, but likely not kappa = 0.9. The implication of this example is twofold: 1) How are organic nitrates accounted for using the AMS technique? 2) Should an O bound with N count the same than an O with C? So based on the above, I have great conceptual difficulty accepting the parameterization of O/C vs. kappa. On the other hand, it is obvious that HOA (or pure hydrocarbons without oxygen) do have kappa ~ 0 , and that oxidizing these compounds will eventually increase kappa. The likely mechanism is that compounds transition from being water insoluble, where they do not contribute to kappa, to water soluble, where they express the kappa according to their molecular size. Further, if acids are produced, the oxidized molecule will likely break apart and lead to smaller more oxidized fragment in the condensed phase.

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Such a process would also indirectly result in an increase of kappa for more oxidized compounds. Finally, for ambient aerosol the matrix of possible oxygenated functional groups may be limited by the chemical pathways producing that aerosol, further leading to a streamlining of the empirical O/C vs. kappa relationship. In fact, my personal view is that the observed linear relationship between kappa and O/C provides more of a constraint on chemical oxidation mechanisms active in the atmosphere rather than offering a prognostic equation to predict CCN, but that is more a philosophical question. In light of my comment, I suggest the following specific changes to the manuscript

The thinking described in the this comment is exactly what drives our assumption that there will be a relationship between increasing O/C ratio of the organic component of ambient aerosol and increasing κ value. In particular, we agree that one can conceive of molecules that will not follow this relationship, with the reviewer pointing out some of these examples. However, in the atmosphere, the majority of the oxygenated component of the aerosol is thought to arise from oxidation of not-too-dissimilar compounds that gives rise to secondary organic aerosol. These gas-phase oxidative processes, or those that might occur via heterogeneous gas-particle or homogeneous aqueous phase oxidation, follow a general direction whereby increasing oxidation gives rise to products that are more oxygenated and fragmented to some degree, so increasing their ability to promote water uptake. On the low O/C side of things, it is reasonable to assume that the HOA-like component, made up of hydrocarbon-like molecules, has a κ value approaching zero. This picture of organic aerosol evolution is nicely described in the Jimenez et al. (2009) paper that was published after our paper was submitted. We now justify our assumptions more clearly in Sect. 3.3 of the revised version of the paper.

We do point out that in the original and revised versions of the paper we do not claim to have observed a "linear relationship between kappa and O/C", as the reviewer

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states. Rather, we say that if such a linear relationship existed (i.e. if the kappa value for HOA was zero and the kappa value for OOA is non-zero, or if the relationship between kappa and O/C is linear), then the values reported in the paper would be derived.

1) A couple of paragraphs that describe these issues and at least qualitatively argue why O/C and kappa should be related.

As recommended by the reviewer, a new section (Sect. 3.3) has been included that discusses how κ and the degree of oxygenation of the aerosol may be related. To quote that section:

"It should be noted that our goal in this work is to extend previous studies by working from an assumption that has been used in the past, i.e. that some part of the organic aerosol component leads to CCN-activity and another does not. Whereas past aerosol-CCN closure studies have only assumed that a constant fraction of the organic aerosol component is soluble and that the remainder is insoluble (Broekhuizen et al., 2006; Chang et al., 2007; Medina et al., 2007; Stroud et al., 2007), in this paper we suggest that the oxygenated factors from the PMF analysis represent the soluble organic component while the unoxygenated factor represents the insoluble organic component. Or, using the concept of hygroscopicity, we postulate that the oxygenated factors contribute to the hygroscopicity of the aerosol, and that the unoxygenated factor does not, whether due to its solubility, as suggested by Petters et al. (2009b), or its intrinsic hygroscopicity.

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Although extraction of the κ of both the oxygenated and un-oxygenated components from an unconstrained fit of the data to a model would be the ideal approach, the uncertainties in our measurements were too great to evaluate the difference between sets of κ parameters. In part, this is because of the relatively narrow range of oxygenation observable at one field site during one campaign. And so, we constrained our model by assuming that the un-oxygenated (HOA) component of the organic aerosol contributed minimally to the aerosol hygroscopicity. This is not unreasonable because HOA is thought to be hydrocarbon-like (Zhang et al., 2005) and aliphatic in nature, and in general, these types of compounds have not been found to be CCN-active in laboratory experiments (e.g. Pradeep Kumar et al., 2003; Raymond and Pandis, 2002). In addition, ambient measurements of particles at locations in which HOA is high, especially at the smaller sizes, have shown that the organic component does not appear to contribute to the CCN-activity (Cubison et al., 2008; Quinn et al., 2008; Broekhuizen et al., 2006). Based on these literature results, we feel that the assumption that the un-oxygenated component is non-hygroscopic is reasonable.

In contrast to the HOA factor, the OOA factors, which make up most of the oxygenated component of the organic aerosol, are thought to be characteristic of highly processed organic aerosols (Zhang et al., 2005) formed from secondary processes (Zhang et al., 2007). Again, laboratory experiments of compounds that are formed from these types of processes such as glutamic, glutaric, pinonic and norpinic acids have found that these aerosols are moderately CCN-active (Raymond and Pandis,

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2002) and smog chamber studies of gaseous organic precursors with ozone or OH have also found that the organic aerosols formed are moderately CCN-active (e.g. Duplissy et al., 2008). In addition, this assumption is supported by laboratory studies that have found that oleic acid particles that have undergone ozonolysis become somewhat CCN-active (Broekhuizen et al., 2004; Shilling et al., 2007) and chamber studies that show that ageing organic aerosols can also increase their CCN-activity to varying amounts (Duplissy et al., 2008; Petters et al., 2006; Wex et al., 2009). We do note that there is also some evidence that the hygroscopicity of SOA under supersaturated conditions in chamber studies do not vary with ageing (Prenni et al., 2007; Jurányi et al., 2009).

These findings lend credence to treating the oxygenated component of the organic aerosol as CCN-active and that it is reasonable to assume that this factor dominates the overall organic aerosol hygroscopicity. Thus, the main objective of this work was to attempt to constrain the hygroscopicity of the oxygenated organic component of ambient aerosols.

The hygroscopicity of the BBOA component has uncertainty associated with it. Originally, we viewed the full BBOA factor to be similar to the biomass burning marker levoglucosan (see Lanz et al., 2007), which is quite hygroscopic (Petters and Kreidenweis, 2007) and was therefore grouped with OOA in the oxygenated component. However, more recent work has suggested that there are other components that make up the BBOA fraction (Petters et al., 2009a) that are not as hygroscopic, which is why its solubility is considered in Sect. 4.3.

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The reasoning behind the postulated relationship between κ_{org} and O/C is similar to that above. Since PMF finds factors by reconstructing the organic mass spectrum as linear combinations of these factors, the O/C of the organic component (since it is based on the signal at m/z 44) can also be calculated as a linear combination of the O/C of our factors, with the O/C of the unoxygenated component approaching zero and the O/C of the most oxygenated component at 0.81 (see Table 2). Organic aerosols with mixed compositions will lie between these two values. Mathematically, both Eq. 5 and Eq. 6 can be considered for an organic component that is composed entirely of the oxygenated factors, resulting in a κ_{org} of κ_{ox} or $a \times (\text{O/C of the oxygenated component})$, while a completely unoxygenated organic aerosol would have $\kappa_{\text{org}} = 0$ using either equation (since the O/C of the unoxygenated (HOA) component approaches 0). In reality, these two methods are not exactly the same since the factor method uses the volume-weighted average while the O/C method uses mass-weighted averages, which result in a slightly different shape to the curves. However, for the most part, these two methods essentially represent the same relationship between the degree of oxygenation of the aerosol and its hygroscopicity."

2) A big qualification, to be also included in the abstract, that the equation $\kappa_{\text{org}} = 0.3 \times \text{O/C}$ does not apply to arbitrary molecules, but only bulk O/C ratios measured for atmospheric aerosol using the AMS.

This has been included in the Abstract as well as the discussion of the "Calculations

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using O/C" (Sect. 4.1.2). Specifically, the new Abstract states:

"Under these assumptions, the κ of the entire organic component for bulk aerosols measured by the AMS can be parameterized as $\kappa_{\text{org}} = (0.29 \pm 0.05) \times (\text{O/C})$, for the range of O/C observed in this study (0.3 to 0.6). These results represent our five-week study at one location using only the AMS for composition analysis."

3) A qualifier on how AMS O/C ratios may differ from actual O/C ratios, e.g. for organic nitrates.

The effect that organonitrates (and organosulfates) have on the O/C ratio as observed by the AMS, when compared to the true value, is an important qualification to include in the paper. In particular, if substantial organonitrates are present then the "O" related to them is presently somewhat attributed to the inorganic nitrate signal, and does not affect the O/C ratio calculated for the organic component of the aerosol. And so, the O/C ratio of these particles is likely underestimated by the present approach in the AMS. It is difficult to assess quantitatively the size of this effect, but this caveat is now included in Sect. 4.1.2 of the revised manuscript:

"As a general caveat, the slope in Eq. 7 may only be representative of the conditions in this study, which examine the bulk ambient, non-refractory organic aerosols measured specifically by the AMS. Also, it does not account for organonitrate or organosulphate components that may be present. Not only do these compounds contribute to an underestimation

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of the actual O/C (Farmer et al., 2010), but they are also oxygenated and likely hygroscopic, without contributing to the O/C itself, further underestimating the organic component's hygroscopicity."

4) A qualifier that the O/C ratios explored here are only valid for the narrow range of O/C explore here (this should be part of the equation).

This has been added to the Abstract and to the equation itself.

5) A statement that the relationship is likely due to the fact that the observed O/C correlates with solubility and molecular size and not directly controls kappa. If the authors have alternative or additional explanations to the ones I give here they should of course be included as well.

A paragraph has now been added to the discussion in Sect. 4.1.2 that reads:

"It is important to note that a relationship between O/C and κ_{org} would not necessarily indicate that the former controls the latter. Rather, it would suggest that O/C correlates to molecular properties that affect an organic particle's hygroscopicity, such as molecular weight and solubility. Nonetheless, we feel that this relationship may provide useful insight since O/C can be derived directly from mass spectral data."

6) A detailed plot on the observed O/C vs. kappa relationship, including error

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bars in x and y. This is important to show if the observations are indeed linear (which I would not expect from theory), and how it compares with the Jimenez et al. data points.

This figure (new Fig. 4) has been added to the revised manuscript along with additional discussion that in part addresses how it compares with the Jimenez et al. (2009) results (Sect. 4.1.2).

As stated in the original version of the manuscript, the uncertainties in the data preclude us from comfortably concluding on the functional relationship between κ_{org} and O/C. It is for this reason that we stated in the paper that the linear relationship with an intercept at the origin is an assumption. Nevertheless, with that being said and disregarding the high uncertainties, the data points in the plot are quite similar to those from measurements of κ_{org} presented in the Jimenez et al. (2009) paper, as extracted for subsaturated conditions.

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