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Interactive comment on "Application of positive matrix factorization in estimating aerosol secondary organic carbon in Hong Kong and insights into the formation mechanisms" *by* Z. B. Yuan et al.

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1. General comments

The present paper by Yuan *et al.* deals with the challenging job of source apportionment for atmospheric aerosols in Hong Kong using the positive matrix factorization method for a dataset of 4.5 years. The challenge therein arises from the multitude of

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different sources in the city and the surrounding area as well as 'long range' sources, transported to Hong Kong by advection. Neither the latter nor the local sources are understood in detail yet, partly not even in the basics such as most of the secondary organic sources of anthropogenic volatile organic compound (VOC) oxidation products participating in the production of the aerosol mass. Additionally, the contribution of different VOCs to the total ambient VOC concentration will vary with measurement site and season, causing different aerosol constituents throughout the year with different characteristic properties.

The structure of the paper is well done and written in an understandable way, but I would have liked more details concerning the exact method. Sometimes the referenced web pages were hard to reach, so the reader has to patient in some way. The presented results are fitting nicely into the scope of ACPD and are certainly of interest. However, the claim to have found indications for cloud-processes acting to form aerosol mass is not valid in this context. Although this might be the case, the conclusion cannot be drawn from the present dataset as outlined below in 'specific comments'. Therefore, the title might be changed somewhat at the end.

2. Specific comments

Several specific comments need to be made to this presentation. One specific point to be made concerns the annual averaging of the source apportionment in Table 1. Although quite useful for long-term aerosol effects, this will smooth out seasonal effects and impacts on human health. Especially the secondary sources vary significantly between the different sites and throughout the year, causing most probably different health effects. Because of this, the authors did monthly analysis for the elemental carbon (EC), organic carbon (OC), secondary carbon (SOC) and sulfates. Doing this, the authors indicated a stronger increase of SOC from summer to winter time than the

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sulfates, but showing a clear dependency of SOC on sulfates or vice versa. This is all reasonable and valid.
However, the conclusion to link this to cloud processes is much more difficult and can not be done in that way because of the multitude of linkages in atmospheric processes and chemistry. For example aromatic compounds like benzene and toluene as well as further anthropogenic VOCs are oxidized nearly exclusively by OH with some small exceptions similar as SO_2. Therefore in the presence of a strong OH source like intensive solar radiation photochemistry is impacting on both similarly, only with a higher velocity for the VOCs because of their faster reaction rate constants. Once the anthropogenic VOCs are released either by transport, industries or heating, they are oxidized rapidly because of their local high concentrations capable to start aerosol mass production quite fast but not hours or days later as stated by the authors. Some of these compounds are emitted in conjunction with SO_2 (heavy fuels etc) even pronouncing this relationship.
Interesting further is the minimum of both secondary aerosol sources during summertime compared to a maximum in the winter. As the authors stated, rainfall is substantial higher during summertime than during wintertime reducing at least the water-soluble compounds by washing-out significantly. Because of the tropical conditions the temperature differences between the two seasons is not that large and due to the mild climate rather small compared to e.g. the mega-cities in the Northern hemisphere. Nevertheless, local pollution sources like heating, changes in transport and changes in weather pattern including transport of pollutants might be the reason for the increase in e.g. VOC (total NMHC) concentrations. This is expressed by the variation in total NMHC during the year clearly marking the transition periods as well as summer and winter. By contrast, SO_2 possesses quite a long lifetime with respect to gas-phase oxidation by OH (about 10 days), which is reduced by cloud-phase reactions to about 3 days, causing a rather well mixing in the troposphere as a source for aerosol sulfates. Therefore, Figure 7 of the present paper is quite understandable. But I would not claim to prove any cloud-phase aerosol production, which nevertheless might be acting. The

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findings can be explained by local sources and photochemistry as well. For me as a reader it seems that the wash-out processes (rainy seasons) and the distribution and strength of the individual sources are determining both the obtained results.
To proceed in this context an analysis with respect to the wind direction or back trajectories would be worth, checking about the results after passing clouds. But most probably the obtained data points are too scarce for this. Further I would be interested in any intercomparison with a chemical transport model as described for the second approach for source apportionment. How much is understood at the moment and how does this fit to the results using the OC/EC technique and the positive matrix factorization. This would give a hint for the cloud issue too. Unfortunately this is not available here, but this would be a challenging and time consuming step too.

3. Technical comments

The authors name the problem of the multitude of different secondary organic aerosol sources and compounds involved giving a few citations. There might be several, but two mightbe included here as well:
i) Seinfeld, J.H. and Pankow, J.F., Organic atmospheric particulate matter: a review. Annual Rev. Phys. Chem. 54, 121-140 (2003).
ii) Kanakidou et al., Organic aerosol and global climate modelling: a review. Atmos. Chem. Phys. 5, 1053-1123 (2005).

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