Atmos. Chem. Phys. Discuss., 14, C5763–C5765, 2014
www.atmos-chem-phys-discuss.net/14/C5763/2014/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Submicron aerosol source apportionment of wintertime pollution in Paris, France by Double Positive Matrix Factorization (PMF^2) using Aerosol Chemical Speciation Monitor (ACSM) and multi-wavelength Aethalometer" *by* J.-E. Petit et al.

Anonymous Referee #1

Received and published: 11 August 2014

General comments:

Using PMF twice increases uncertainties. The uncertainty derivation for the second step does not appear robust, apparently guided partly by guesswork and partly by how the OA factors should be weighted in PMF Step 2. The final results are less than satisfactory, since the authors are still limited by inherent limitations of PMF; the authors themselves specify the need for further investigation of the uncertainties.

I am hesitant to recommend publication. The science may be better served by using the BC factors directly in the first PMF step; since the authors single out OA fragments for PMF #1 and then add inorganic species in PMF #2, surely they can combine all fragments, ions, and BC/wind data into PMF #1.

Specific comments

This is not real-time characterization of PM sources. Highly-time resolved? Possibly. But not real-time!

Is m/z=60 indicative of wood-burning emissions? Please specify.

Is the assumption that only wood-burning contributes to BrC?

While a slope of 0.99 is good, the somewhat lower correlation coefficient of 0.65 for ACSM+BCaeth (I assume that is what is used for mass closure) against the TEOM even after averaging at 3-h resolution does not lend itself to the strong characterization implied by "validating". Softer language should be used – perhaps couching the comparison in the language of instrument uncertainties (which have not been specified in the measurement section!)

Uncertainties seem large, but not clear what was the justification. For example, how was the 40% uncertainty for the two BC fractions derived – a propagation (e.g. "sum of squares"?) of uncertainties from the two papers mentioned? Why pick 50% for Potassium – how big are the measurement artifacts? Empirically-determining uncertainties for the ACSM data to ensure appropriate weighting for the organic fractions in later PMF analysis seems dubious. Were they set too low compared to the actual values, so as to get "good" PMF results?

"OOA is found to significantly contribute to the traffic organic mass although its exact contribution cannot be determined without the much-needed thorough determination of uncertainties." – This undercuts the novelty of this PMF2 approach, and discounts the next statement that this PMF2 methodology is "especially efficient" in linking OA

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factors and pollution sources.

The EPA's PMF model allows boot-strapping techniques. Yet, authors finish by saying the results need to be refined using boot-strapping of OA factors. There is not a significant discussion of the effects of boot-strapping in the main text, if that was indeed utilized to obtain the results in this manuscript.

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 14159, 2014.