From real-time to long-term source apportionment of PM10 using high-time-resolution measurements of aerosol physical properties: Methodology and example application at an urban background site (Aosta, Italy)

The authors reported in detail the new PM10 source apportionment approach called RASPBERRY that combined OPC-based particle size distribution and spectrally resolved light absorption. Specifically, the manuscript includes inter-comparison between this physical-based PMF and the commonly used chemical PMF in addition to specific case studies across the Po Valley over the period of 2020-2024.

Overall, the paper is well-written with occasional repeated information between figure captions and the main text. The similar inter-comparison between chemical-PMF and this physical-based analysis is very interesting. One of my main concerns is the lack of uncertainty estimates on some of these comparisons. This manuscript would benefit from a major revision that I hope will address my questions and comments below.

Specific comments/questions:

Abstract: The particle size detection limit should be specified when the authors mentioned particle size distribution (L4) (Dp > 0.18 um). The lack of detection of nucleation and Aitken mode particles has implication (e.g., missing fresh emission). For the light absorption, the authors should also include the measurement wavelengths for this analysis (370-950 nm).

L127: 'foehr': Is that a typo?

L142: "all measurements were averaged to a common temporal resolution of 1 hour" – for long term diurnal or seasonal trends I can see that this time averaging is good enough. But for point source emissions like fresh smoke, 1-hr average may be too long. I guess since the particle size measurements do not extend to small particles, the current equipment setup is incapable of capturing these short-term events. Is there any concern about diluting short burst of emissions due to this long time averaging? Or have the authors looked at 30-s or 5-min average of size distributions to ensure they are not missing any point source emissions?

L290-291: "no thorough assessment exists for the measurement uncertainties or detection limits associated with the optical spectrometer used in this work." – There have been other studies on uncertainties associated with optical-based particle sizing instrument (e.g., UHSAS by DMT with uncertainties estimated by Kupc et al., AMT, 2018). Have the authors tried Monte Carlo simulation to estimate the particle size and counting statistics uncertainty?

L306-307: "Achieving an optimal solution required us several tests, as small variations in the uncertainty configuration often produced unpredictable changes in the final outcome." – Does this mean that the reported equations for the uncertainty configuration (Eq 4-5) are based on this tuning process?

L329-330: For the random subset of 4000 samples, the authors picked samples for all seasons. Did the authors also include data representative of diurnal or temporal trend?

L340: "assuming the profiles remain stable over time". If the profiles are different, how does RASPBERRY handle this? Would it generate inaccurate source apportionment?

Figure 3: I would plot particle diameters (right column) on a log scale instead of a linear scale.

L475: The authors mentioned that residential biomass burning agree with previous studies that detected maxima around 100-200 nm. Since the OPC cannot measure below 180 nm, we cannot actually see any peak below this detection limit. I would not claim this in the manuscript.

L447: Reported AAEs on Fig. 3 are rounded to 1 decimal point. I would keep that consistency in the manuscript: 1.1 and 1.8 instead of 1.06 and 1.79.

Figure 7 caption: There is no 'pink' color in panel (a). Did the authors mean 'yellow'?

L626: The elevated baseline of RASPBERRY vs. CAMS ensemble – is this indicative of CAMS ensemble not capturing the background dessert dust level OR is it an issue with RASPBERRY algorithm? The authors should add some explanation for this discrepancy between the 2 datasets.

Figure 8 right column: It would account for uncertainties in both chemical PMF and RASPBERRY algorithm if the authors apply ODR fitting instead of OLS. It would change the slope value drastically depending on what the uncertainty estimates may be.

Figure 13: The color of the maximum values for both AOD and CO color bars are too similar, so I suggest the authors to change colors on one of the color bars.

L918-923 paragraph: Have the authors considered comparing the constraining profiles from Po Valley against those collected during the AEROMMA 2023 field mission over North America? The relatively complete aerosol in-situ measurements (3 nm to ~50 um) would provide additional examples of constraining profiles. There were sampling in a lot of megacities (NY, Toronto, Chicago, LA) that captured biomass-burning-influenced aerosols from Canadian wildfires as well as urban pollutions. Despite the geographic differences, I highly suggest the authors to compare the aerosols profiles to provide additional constraining profiles for RASPBERRY.

L939-944 paragraph: The authors could add the recommendation of additional in-situ aerosol measurements with more comprehensive instruments (e.g., extend particle size detection to nucleation mode) to provide additional training datasets for RASPBERRY algorithm.