Public justification (visible to the public if the article is accepted and published):

This is clearly a very substantial and novel piece of work and while the authors have gone to efforts to address the comments of the reviewers, I feel that much of the argument made in the rebuttal is not adequately reflected in the revised document. In general, in spite of the reviewers highlighting the issue, the paper does risk being seen as being 'too technical' or 'too local' to be considered as a research article in ACP. So I would like to extend the opportunity to the reviewers to further revise the article. However because these modifications do not concern the scientific content, I have decided that a further round of peer review is not likely to be necessary.

Response: We thank for the positive feedback from the editor on the significance of this work. Our responses to specific comments are in blue color below:

1. The discussion and conclusions need to more clearly show the wider implications rather than just the technical developments and the observations. In particular, the authors must show that the aerosols observed aren't specific to Singapore and have relevance for the wider atmosphere. While additional articles describing air pollution in Singapore are referred to in the introduction, the wider context needs to come through in the discussion and conclusions, so I would suggest more attention being paid to comparing observations to these previous works. Furthermore, the introduction and conclusions should also describe how typical Singaporean air quality is compared to other locations in the region and following this, a commentary on other areas these observations may be applicable to. These may seem trivial to someone already familiar with the location and the associated literature, but are important bits of contextual information for ACP's general readership.

Response: The abstract, introduction and conclusion have been modified based on editor's suggestions. In particular, the general implications of individual PMF factors identified in this work have been discussed in detail in the conclusion.

The abstract has been revised to reflect the updated main text:

Page 2 lines 4-8: Positive matrix factorization (PMF) analysis of rBC and organic aerosols (OA) (PMF_{base}) identified two traffic factors with differences in rBC content, coating thickness and diurnal pattern, which could potentially help differentiate gasoline and diesel vehicular emissions. Additionally, two secondary OA (SOA) factors influenced by local chemistry and/or regional transport (less-oxidized oxygenated OA (LO-OOA) and more-oxidized OA (MO-OOA)) were identified. Including metals in the PMF (PMF_{metal}) improved the quality of source apportionment significantly.

Page 2 lines 11-13: Although the aged BBOA component was highly oxidized, its strong association with $K_3SO_4^+$ distinguished it from other background MO-OOA, which generally lacked distinctive OA signatures.

The manuscript has been revised as shown below:

Page 4 lines 15-29: Singapore is a highly developed coastal urban environment located at the southern tip of the Malay Peninsula in Southeast Asia. In additional to typical local emissions from vehicular traffic and cooking , Singapore hosts the world's second busiest shipping port (World Shipping Council, 2024) and fifth largest oil refining capacity (Lau et al., 2021), both of which are potentially significant anthropogenic sources of combustion-related PM and toxic pollutants to impact the local air quality. Moreover, air quality and concentrations of atmospheric PM in Singapore can be significantly influenced by SOA and transboundary haze caused by biomass burning emissions transported from surrounding regions in Southeast Asia (Budisulistiorini et al., 2018; Heil and Goldammer, 2001; Kuwata, 2024). Given the unique capability of the SP-AMS to simultaneously detect rBC particles and their associated coatings, this study explores multiple PMF scenarios based on SP-AMS measurements, iteratively incorporating rBC, OA, inorganic aerosols (IA), and metals into the input. This step-by-step PMF interpretation approach underscores the value of including all measured chemical species in the analysis to better resolve local primary emissions and the secondary transformation processes of rBC-containing particles in Singapore. The improved characterization presented in this study provides important insights into how industrial emissions may introduce toxic heavy metals and influence local SOA formation in a complex urban environment. It also sheds light on how regional biomass burning may affect background levels of rBC-containing particles in Southeast Asia's urban atmosphere.

Pages 33-34, lines 20-31: … and subsequently predicting their climatic and health impacts. This work demonstrates that including rBC, OA, IA, and metals fragments improve source apportionment and coating thickness quantification (i.e., Org/rBC ratio) of rBC-containing particles in urban environments under strong influences of multiple anthropogenic emissions, and provide insight into our understanding of secondary processing of rBC-containing particles in Singapore, a complex tropical urban environment with large scale shipping ports and petrochemical industries.

All the PMF scenarios analysed in this work can identify two primary traffic-related factors (HOA and rBC-rich OA), which have been previously reported in other urban and near-road environments across different continents (Cui et al., 2022; Farley et al., 2023; Lee et al., 2017; Saarikoski et al., 2019; Wang et al., 2020). As demonstrated in this work, these two factors may serve as useful indicators for distinguishing rBC emissions from gasoline and diesel vehicles in typical urban settings. This can provide quantitative insights to support the development of targeted air pollution control strategies related to traffic emissions in typical urban environments. Furthermore, the observed variability in the mixing degree between rBC core and the HOA coating can result in substantial differences in aerosol optical properties and radiative effects compared to the commonly assumed uniform mixing (Willis et al., 2016). The majority of rBC mass classified into the rBC-rich OA factor could be one of the reasons to explain insignificant lensing effects of BC coating in ambient measurements, but models over-estimated light absorption enhancement of BC coated with OA in urban environment even with high bulk RBC values (Cappa et al., 2012; Fierce et al., 2020; Healy et al., 2015; Liu et al., 2017).

This work provides further separation of an industrial-related factor from primary traffic emissions. Although such industrial and shipping emissions are transported to our sampling occasionally during afternoon sea breeze, the IOA factor accounted for 10-12% of rBC and and 5% of OA coating, suggesting that the negative impacts of industrial emissions on the local air quality should not be overlooked. More importantly, the IOA factor was strongly associated with V^+ and Ni^+, suggesting that this type of rBC-containing particles might pose high potential risks to human health and the environment. This observation is strongly relevant to other coastal cities with major shipping ports and industrial areas with large scale petrochemical facilities. Recently, Tehrani et al. (2023) conducted mobile SP-AMS measurement in the Chester-Trainer-Marcus Hook area of southeastern Pennsylvania, which is home to multiple petrochemical plants, a refinery, and a waste incinerator, showing elevated heavy metal concentrations (e.g., Sb, As, Cd, and V) near the industrial facilities. It is important to note that anthropogenic OA factors related to industrial, shipping, and coal-combustion emissions have been identified in previous standard AMS studies across various locations (e.g., Chen et al., 2022; Daellenbach et al., 2024; Xu et al., 2014). A deeper understanding of the heavy metal constituents in these emissions could offer valuable insights into their toxicity and broader environmental and public health implications.

Biomass burning emissions has been impacting the air quality and regional haze formation in Southeast Asia (Adam et al., 2021; Budisulistiorini et al., 2018; Othman et al., 2022; Van et al., 2022). This work provides additional insight into understanding the fate and transformation of rBC-containing particles in aged biomass burning emissions. Overall, A-BBOA and Night-IA-BBOA derived from PMF_{all} accounted for 12% of OA coatings mass in total, and the mass contribution of MO-OOA to total OA coatings decreased from 31% for PMF_{base} to 24 and 21% for PMF_{metal} and PMF_{all}, respectively. It is important to note that MO-OOA has been consistently reported as a dominant background OA component in total $NR\text{-}PM_1$ across many previous AMS studies (Chen et al., 2022; Jimenez et al., 2009; Zhang et al., 2011; Zhang et al., 2020). However, quantifying its contribution from aged POA, including BBOA, remains a challenge. Recently, Vasilakopoulou et al. (2023) have shown rapid transformation of BBOA into highly oxidized and harmful background MO-OOA in Europe. Although this study focused on OA coatings characterization, our findings highlight the potential to distinguish highly oxidized BBOA-derived materials from the typical background MO-OOA in $NR\text{-}PM_1$ identified by standard AMS techniques. This distinction offers a valuable approach for better evaluating the overall health and environmental impacts of aged or transported BBOA, particularly in Southeast Asia and other regions heavily affected by wildfires and agricultural burning.

2. On a technical note, while the downweighting is described a little better in the text, the justification given in the rebuttal should also be included in the revised text regarding the 'strength' of the variables and what precedent there is in the literature for using the method. This is an essential part of the novel aspect of the methodology, so it must be properly documented.

Response:

We agree with the comment and have added Fig. R1 to SI and included the justification in the main text. SI figure numbers were also updated accordingly due to the new SI figure.

The manuscript has been revised as shown below:

Pages 8-9, lines 23-2: The signals of K^+, Rb^+, V^+, and Ni^+ were either downweighted or upweighted to produce SNR comparable to those of typical OA and rBC fragments. For instance, strong fragments such as $C_2H_3O^+$ and C_3^+ exhibit SNRs of ~25 and ~21, respectively. To bring K^+ into a comparable range, its signal was downweighted by a factor of 10, resulting in an SNR of ~13. In contrast, the signals of Rb^+, V^+, and Ni^+ were each upweighted by a factor of 3 to achieve SNRs of ~6. Figure S1 demonstrates how adjusting SNR of K^+ improves the performance of PMF as an example. Previous studies of SP-AMS measurements have also used a similar approach to integrate metal-related ions into PMF analysis (Bibi et al., 2021; Carbone et al., 2015; Rivellini et al., 2020). To further investigate the nature and formation of OA, IA-related fragments from sulfate, nitrate, and chloride were included in the PMF analysis. This PMF analysis included all the measured aerosol components (i.e., OA + rBC + metal + IA) (denoted as PMF_{all-n} hereafter). Sulfate ions that exhibited SNRs between 10 and 30 were downweighted by a factor of 3. Nitrate ions and $K_3SO_4^+$ had SNR values in the range of 5 to 15 that do not require adjustment. Noting that downweight and upweight signals of AMS data for PMF analysis has been more systematically investigated by previous studies to improve the separation of factors (Adam Reff et al., 2012; Lee and Hopke, 2006; Paatero and Hopke, 2003).

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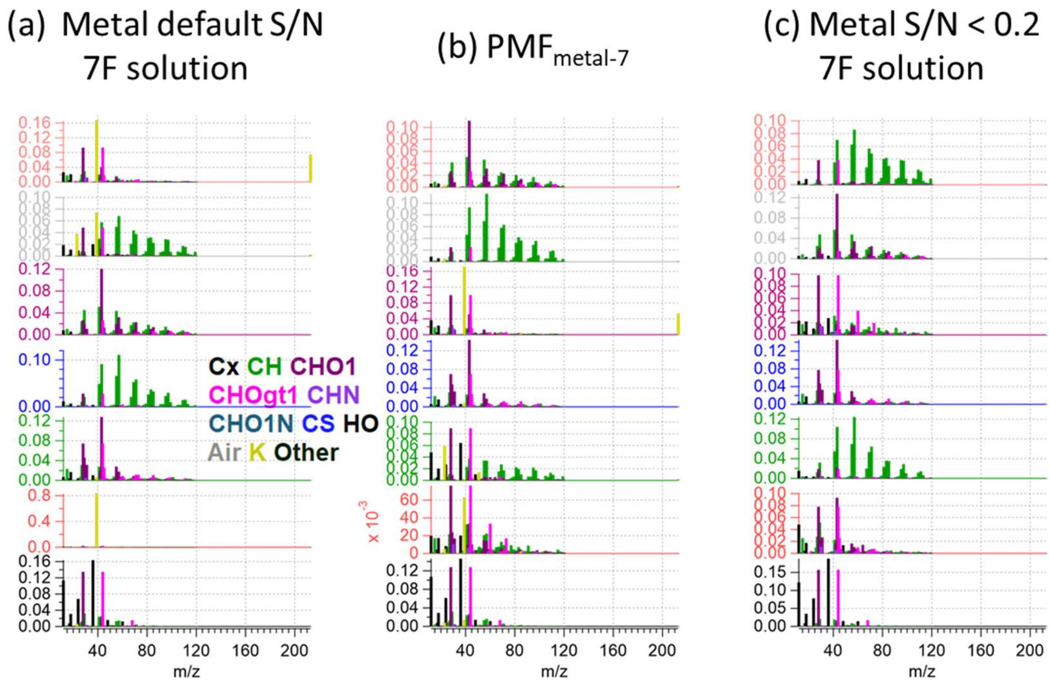

Figure S1. The comparison of 7-factor solutions using default S/N with K^+ dominated factor (i.e., poor separation, a), adjusted S/N in $PMF_{metal-7}$ (b), and over-adjusted S/N showing over-separation (i.e., S/Ns < 0.2, c).