Referee #2

The paper introduces a novel methodology for identifying the sources of both inorganic and organic components of particulate matter. By integrating non-refractory PM compounds with elemental and black carbon data, the authors evaluate model performance using optimal initial parameters compared to results obtained with minimal prior knowledge. This approach is innovative and merits publication in *Egusphere*. The study aligns well with the journal's scope and is generally well presented. Below, I offer several minor suggestions that I believe could enhance the clarity of the paper:

We thank the reviewer for the positive feedback. Please find the responses to the individual comments below.

- **Line 150:** At this point in the manuscript, the definition of "generic conditions" is somewhat unclear. Although the meaning becomes clearer later in the text, providing a brief clarification earlier would greatly improve readability.

The sentence was modified as follows:

Line 162-166: "With respect to the implementation of the RT, the model's performance was evaluated under two conditions: using optimal initial parameters, derived from site-specific information concerning the number and chemical composition of the sources, and under less optimized, generic conditions, which relied on non-site-specific data informed by general knowledge of source composition."

- **Sofi Model:** Line 70 mentions that in the previous publication (Chen, Canonaco, Slowik et al., 2022a), where ACSM data were analyzed, an earlier version of Sofi-RT was used. While section 2.3.1 describes the model's operation in detail, it would be helpful to state early in the manuscript whether the version applied here differs from the earlier one used for evaluating ACSM and Aethalometer data.

We thank the reviewer for the comment. We revised the lines 76 – 80 to:

"Although this study employed state-of-the-art optimized SA approaches, it focused exclusively on the organic fraction of PM rather than the total PM mass, and it did not utilize the capabilities of the latest software version to process data from the Xact instrument."

The SoFi version used in our study was integrated in the manuscript:

Line 187: "SoFi RT v.9 (Source Finder Real-Time) by Datalystica was the tool used…"

- **Line 77:** I believe there is an inconsistency in this paragraph, which states: "In this study, data were collected from an Xact, an Aethalometer, a total carbon analyzer, and low-cost sensors." To my understanding, data from neither a total carbon analyzer nor low-cost sensors were actually used. However, for the Sea Salt/Dust contribution assessment, measurements from a Grimm instrument were included (see Figure S4), yet this device is not mentioned here. Please clarify or correct this.

The authors were referring to the study by Prakash et al. (2021). This was rephrased to be clearer:

Line 83-85: “In another study that took place in Delhi, India, an RT-SA methodology that reports the results online has been set up (Prakash et al., 2021). Data were collected from an Xact, an Aethalometer, a total carbon analyzer, and low-cost sensors. Due to the nature of the input data in Prakash et al. (2021), the source apportionment focused primarily on the speciation of elements, with no information provided about secondary species.”

- **Lines 127-132:** The methodology for handling Aethalometer data lacks clarity. While prior publications by the authors detail their approach to this dataset, the current paper does not fully explain how the multiple scattering effect is accounted for. Please clarify the compensation method for this effect and specify the inlet size used (e.g., PM2.5).

We thank the reviewer for the comment. The methodology for processing Aethalometer data we follow is based on the procedures established by the EBAS database. The AE33 instrument, utilizing its dual-spot technology, automatically corrects for the filter loading effect. The aerosol absorption coefficients at all wavelengths are calculated using the equivalent black carbon (EBC) concentrations, the instrument's specific sigma values, and the H-correction factor provided by EBAS. These absorption coefficients are then adjusted to standard conditions (0 °C, 1 atm).

To obtain an EBC value that more closely corresponds to Elemental Carbon (EC) as measured by EC/OC instruments, a site-appropriate Mass Absorption Cross-section (MAC) value is applied. This is the standard procedure we follow when processing such data.

However, in this study, the AE33 was feeding the data into the RT software in near real-time mode. As a result, the raw EBC data provided by the instrument were directly used by the analysis software without post-processing. To avoid confusion, we have updated the manuscript to reflect this approach, as follows:

Line 137-140: “In this study, the AE33 operated with a $PM_{2.5}$ inlet, and minute-resolved EBC mass concentrations at the relevant wavelengths were used as raw data from the instrument under near real-time conditions.”

- **Line 235:** The authors assert that high a-values are unsuitable for ACSM data. Include a reference or provide justification for this claim.

We agree with the reviewer that this statement deserves further justification. On previous studies conducted worldwide it has been found that for the ACSM data if we constraint with a high a-value the chemical composition of the factor can change significantly to the point that their identification is no longer clear. For that reason it is suggested that the upper threshold is 0.5 for more variable sources such as BBOA, while for the factors that refers to primary organic aerosols the suggested value is usually even lower ~0.2. These suggestion are described both in earlier publications as well as in the publication that refers to the harmonized source apportionment analysis protocol in Europe (Crippa et al., 2014) (Chen et al., 2022).

A reference that suggests tight constraints on the primary factors of the ACSM was included in the text:

Line 252: “Higher a-values are not suitable for ACSM data, as the identity of the factors can change (Crippa et al., 2014; Chen et al., 2022)., in contrast to Xact data.”

- **Line 407:** The construction of the constraint profiles remains unclear, particularly the criteria for identifying variables that can be classified as "irrelevant." Please elaborate on this process.

We thank the reviewer for the comment. We define 'irrelevant variables' as those that are not identified in previous studies as being emitted by specific sources. For example, silicon (Si) is not typically associated with brake wear; therefore, its contribution to this factor was set to zero. The determination of relevant variables for factor identification was based on established knowledge of source compositions, findings from prior studies, and the optimized source apportionment results obtained in the present study. The following statement has been added for clarification:

Line 320-322: “This approach involved setting certain variables to zero (those deemed irrelevant for factor identification, on established knowledge of source compositions, findings from prior studies, and the optimized source apportionment results obtained in the present study)”

- Although the authors clearly state that ACSM analyzes PM1 while Xact measures PM2.5, the source apportionment percentages in Figures 4 and 5 are reported without distinction between these size fractions. Additionally, the abstract refers generically to contributions to "PM mass" without specifying size ranges. Given that each source inherently exhibits its own particle size distribution, we recommend elaborating on this concept and explicitly addressing the limitations of presenting combined results across different size fractions.

We thank the reviewer for bringing up this important point. Based also on the suggestion of Reviewer 1, we have now specified the size fraction measured by each instrument used in this study.

Although we acknowledge that using consistent size fractions across all instruments is preferable, we consider our approach to be justified and methodologically sound for two main reasons:

(a) The data from the different instruments were not combined prior to the source apportionment analysis. Instead, each dataset was treated independently. Consequently, differences in the size distribution of sources do not directly influence the analysis, as each dataset reflects the characteristics of its respective size fraction.

(b) Carbonaceous aerosols are typically concentrated in the fine fraction. BC is primarily associated with fine particulate matter due to its formation from combustion processes (Saarikoski et al., 2021). Supporting this for the organic fraction, studies employing collocated ACSMs equipped with both PM_1 and $PM_{2.5}$ lenses have demonstrated that organic aerosol components in both size fractions are comparable in terms of chemical composition and mass.

For instance, Liu et al. (2024) evaluated the performance of the Time-of-Flight Aerosol Chemical Speciation Monitor with a capture vaporizer (TOF-ACSM-CV) during the RITA-2021 field campaign. Two identically configured TOF-ACSM-CV instruments were deployed to simultaneously measure non-refractory PM_1 and $PM_{2.5}$ at the Cabauw Experimental Site for Atmospheric Research (CESAR) in the Netherlands. The study found that PM_1 accounted for approximately 85% of the $PM_{2.5}$ organic aerosol mass, with both fractions exhibiting similar chemical compositions.

Similarly, in a study conducted in urban Nanjing, China, Zhang et al. (2017) deployed a $PM_{2.5}$-capable ACSM alongside a standard PM_1 ACSM. The measurements revealed that the mass spectra and time series of primary and secondary organic aerosols (POA and SOA) were highly consistent between the PM_1 and $PM_{2.5}$ size fractions, indicating that organic aerosols are similarly distributed in both.

Nevertheless, we recognize that this approach has certain limitations. The degree of similarity between fractions of carbonaceous aerosol content can vary depending on local sources, atmospheric conditions, and site characteristics. Therefore, while the findings presented here support the validity of our approach for this specific campaign, we caution against generalizing this method without site-specific validation. To reflect this, we have included the following statement in the manuscript to acknowledge the potential limitations of using mixed size fractions across instruments:

Line 149-152: " In this study, different PM fractions were used to implement the RT-SA approach. For this reason, we do not attribute the sources collectively to specific size fractions, as doing so would introduce uncertainty. It is important to note that, although the comparison of carbonaceous aerosol fractions appeared consistent in this study leading to good reconstruction of $PM_{2.5}$ mass, this may not be the case in all environments. Variations in local emission sources, atmospheric processing, and particle size distributions can lead to inconsistencies between size fractions. Therefore, where possible, harmonized size cuts should be applied, and caution should be exercised when interpreting what the total mass represents.”

Liu, X., Henzing, B., Hensen, A., Mulder, J., Yao, P., van Dinther, D., van Bronckhorst, J., Huang, R., and Dusek, U.: Measurement report: Evaluation of the TOF-ACSM-CV for PM1.0 and PM2.5 measurements during the RITA-2021 field campaign, Atmos. Chem. Phys., 24, 3405–3424, https://doi.org/10.5194/acp-24-3405-2024, 2024.

Saarikoski, S., Niemi, J. V., Aurela, M., Pirjola, L., Kousa, A., Rönkkö, T., and Timonen, H.: Sources of black carbon at residential and traffic environments obtained by two source apportionment methods, Atmos. Chem. Phys., 21, 14851–14869, https://doi.org/10.5194/acp-21-14851-2021, 2021.

Zhang, Y., Tang, L., Croteau, P., Worsnop, D., et al.: Field characterization of the PM2.5 Aerosol Chemical Speciation Monitor: insights into the composition, sources, and processes of fine particles in Eastern China, Atmos. Chem. Phys., 17, 14501–14517, https://doi.org/10.5194/acp-17-14501-2017, 2017.