REFEREE #4

Summary of Strengths, Weaknesses, and Overall Contribution

This paper presents a valuable long-term dataset on PM concentrations and chemical composition, which enables in-depth analyses of the origins and sources of pollutants. The objectives are clearly defined, and the dataset is of great value to both the local and global scientific community. However, I suggest several improvements to further enhance the quality of the manuscript. Below, I provide general comments followed by more specific ones for consideration.

We would like to thank the reviewer for his/her positive comments. Below a point-by-point response to the comments raised. Reviewer's comments are shown in *ITALIC* and our answers are presented in **BLUE.**

<u>*Major comments:*</u>

-Introduction: While the introduction provides useful background to understand the study's objectives and conclusions, the information is somewhat scattered across paragraphs. I recommend structuring the section so that each paragraph focuses on a specific topic. For example, paragraphs 3 and 4 both address pollutant concentrations in Greece and could be merged and reorganized for clarity.

We thank the reviewer for the comment. The introduction section has been revised accordingly.

-Materials and Methods: Although the manuscript refers to previous publications for details about the sampling sites, I recommend adding a figure showing land use in the study region together with the locations of the sampling sites. This would help readers better understand the rationale for site selection as well as their proximity to specific areas. Known PM sources or prevailing wind trajectories could also be marked to provide additional context for interpreting the results.

A figure describing the study area (for both NICTRA and AMX) have been added to the manuscript.

- Quality control: Although references are provided regarding quality control procedures, it is important to report specific values of detection limits and quantification limits for the analyzed elements, ions, and other compounds. These data are critical when evaluating the reported concentrations. I recommend including a supplementary table with this information.

As suggested by the reviewer, the method detection limits and quantification limits have been reported in Supplementary material (Table S2).

- Results and Discussion: Given the identified origins of PM_{10}, it would strengthen the discussion to include additional references from other authors describing PM sources and variability in the region. This would help contextualize potential sources not only in the study area but also in surrounding regions. Expanding the discussion in this direction would add valuable depth.

Relevant references to other studies in the region have been considered, as suggested by the reviewer

-Line 243: How do the authors conclude that these elements are related to regional biomass burning? I suggest adding a reference that specifically addresses the origin of these compounds in the region.

K^+ (and OM to a lower extent) is usually used as tracer for biomass burning (Fourtziou et al., 2017; Puxbaum et al., 2007; Reche et al., 2012). Given that the influence of local emissions at our regional background site is minimal, this species is likely to originate from regional biomass burning. This is further supported by a peak of K^+ concentrations observed during summer most probably due to agricultural waste and/or forest fires (see Bimenyimana et al., 2025). References were now added, as suggested by the reviewer.

-Urban traffic PM10 (NICTRA): *This section would also benefit from a more detailed discussion, ideally comparing the findings with results from other areas in the region or even from other parts of the world. Such comparisons would make the results more meaningful to the broader scientific community.*

As suggested by the reviewer, discussion on PM trends at other locations of the Mediterranean (Greece and Spain) has been added.

Minor comments:

-Line 119: *While understandable, the technically correct notation is R^2 rather than r^2. Please revise accordingly.*

This has been corrected throughout the manuscript.

-Line 160: *The sentence "These gases, often co-emitted with primary PM pollutants, can further serve as specific markers of combustion-related sources such as traffic and industrial emissions" requires a supporting reference. Please add one.*

A reference has been added, as suggested.

-Line 222: *Ammonium sulfate has been previously referred to by its ionic abbreviation. I suggest maintaining this convention throughout the manuscript for all elements and ions. Alternatively, provide both the abbreviation and the full name at first mention and use only abbreviations thereafter for consistency.*

This suggestion has been considered in the revised manuscript.

-Line 223: *Acronyms should be defined only upon first use. Please review the manuscript carefully to ensure consistency across all acronyms.*

All acronyms are now defined at first use and used consistently, as suggested by the reviewer.

-Line 429: *Please replace "surprising" with "surprisingly".*

This has been corrected in the revised manuscript.

REFERENCES

Bimenyimana, E., Sciare, J., Oikonomou, K., Iakovides, M., Pikridas, M., Vasiliadou, E., Savvides, C., and Mihalopoulos, N.: Cross-validation of methods for quantifying the contribution of local (urban) and regional sources to PM2.5 pollution: application in the Eastern Mediterranean (Cyprus)., Atmos. Environ., 120975, 2025.

Fourtziou, L., Liakakou, E., Stavroulas, I., Theodosi, C., Zarmpas, P., Psiloglou, B., Sciare, J., Maggos, T., Bairachtari, K., Bougiatioti, A., and others: Multi-tracer approach to characterize domestic wood burning in Athens (Greece) during wintertime, Atmos. Environ., 148, 89–101, 2017.

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Reche, C., Viana, M., Amato, F., Alastuey, A., Moreno, T., Hillamo, R., Teinilä, K., Saarnio, K., Seco, R., Peñuelas, J., Mohr, C., Prévôt, A. S. H., and Querol, X.: Biomass burning contributions to urban aerosols in a coastal Mediterranean City, Sci. Total Environ., 427–428, 175–190,

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