

REFeree #1

General Comments

This paper focuses on long-term daily measurements of chemical speciation data at 2 coarser size cuts at both an urban site with traffic and a more regional background site in Cyprus in order to distinguish between local and regional emissions and sources. A key finding is a statistically significant drop in particulate matter concentrations which still exceed EU regulations. This paper also focuses on long term trends of individual sources using PMF and found a decrease in traffic-related emissions due to a shift towards EURO-standard vehicles that was largely negated by increases in road dust and biomass burning. They conclude that this region remains a pollutant hotspot due to contributions of desert dust and anthropogenic pollution as well as increases in biomass burning.

The introduction focuses on the effects of PM on human health, glances over climate, and establishes the importance of researching it for public policy. They identify gaps in the literature by describing how scarce long-term data is in these regions and how focus has typically instead been on Greece. For example, the authors describe how the most comprehensive long-term PM trend study in Cyprus lacks chemical data to distinguish factors driving downward trends in local urban and regional PM. The annual trends observed are consistent with the literature and the PMF factors resolved at both sites are well supported by tracer analysis and volume of data. The meat of this study is in the trends of identified PMF sources. Overall, I believe that this body of work is good quality, comprehensive, and novel.

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**.

Specific Comments:

The agreement in section 2.2 is very high between the gravimetric PM mass determination and TEOM-FDMS and does not raise concern, but can you please be more specific on the number of samples substituted this way?

Indeed, very strong agreement is observed between TEOM-FDMS and the gravimetric methods ($R^2 = 0.96$; slope = 1.02). Therefore, the gravimetric $PM_{2.5}$ data was substituted with daily averaged online measurements from a co-located TEOM-FDMS. This substitution was applied to $PM_{2.5}$ daily samples collected between 2011 and 2014, corresponding to a total of ca. 1460 samples, or ca. 20% of the dataset.

In section 2.3, is it known what causes the differences in site-specific conversion factors?

The selection of different OC-to-OM conversion factors for NICTRA and AMX was motivated by the contrasting nature of these sites. NICTRA being an urban traffic site, is expected to be dominated by primary organic aerosol from direct vehicular emissions, which are typically less oxidized and therefore associated with lower OM/OC. On the other hand, AMX being a regional background site is likely to be significantly influenced by aged secondary organic aerosols during long-range transport, leading to higher OM/OC ratios. Based on the work of Turpin and Lim, (2001) proposing an OC-to-OM conversion factor of 1.6 ± 0.2 for urban environments and 2.1 ± 0.2 for remote sites, we have applied a value of 1.8 for NICTRA and 2 for AMX to account for their respective characteristics.

This paper would benefit from a map used to describe the geographic origin of the different source regions as well as a pie chart of their percent frequency.

As suggested, a map used to describe the geographic location of the different source regions is added in Supplementary material (Fig. S11), along with the frequency (in %) of air masses from the various sectors.

This paper would also benefit from a short discussion of the differences in de-seasonalized monthly means and monthly means in section 3.1. I understand it is explained in a previous paper, but it seems odd then to present both results in this paper.

The non-parametric Mann-Kendall trend test is typically applied to de-seasonalized monthly averages rather than monthly means to prevent recurring seasonal fluctuations from masking long-term trends thereby improving the robustness of trend detection. As suggested, a short sentence was added in the revised manuscript.

This paper's flow would benefit with a short description on what drives the decrease in regional dust emissions at AMX earlier in section 3.1.

The decrease in regional dust emissions observed before 2015 is likely driven by changes in regional weather patterns that influence dust activity over the region. Shaheen et al. (2023) reported a negative association between dust-AOD and meteorological parameters such as winter sea-level pressure (SLP) and wind regime between 2010 and 2017, reflecting the suppressing effect of these conditions on regional dust. A short sentence was added to the revised manuscript.

It would be important to describe the methodology for how the optimal number of PMF factors was chosen for both sites. Has separate work been done on temporal correlation of the factors to tracers and of factor concentration to each other? There is often debate about if PMF is distinguishing individual sources or the same source at multiple stages of aging. As an example, an anti-correlation of fresh to aged sea salt could help distinguish these. What method do you use to describe "significant" differences in chemical composition of your factors, specifically in line 285? Given that PMF factor profiles are fixed I wasn't sure how this was determined.

The methodology for selecting the optimal PMF solution is provided in Section S1 (Supplementary material). This section provides additional information on various quality control checks including correlations of factors with external tracers, Pearson distance ($PD = 1 - R^2$), Standardized identity distance (SID), as well as bootstrap results. PD and SID give an indication on the (dis)similarity between PM source profiles. Based on the values of the two metrics, we can conclude that all our PMF factors are significantly different both in terms of temporal correlation ($PD \geq 0.4$) and chemical composition ($SID \geq 0.8$) (see Tables S3 and S4).

It confuses me that there is both a decrease in traffic emissions and decrease in heavy oil combustion, but an increase in regional fossil fuel combustion. How well or poorly temporally correlated are the factors to each other? PMF does not provide perfect separation so I worry this conclusion may be driven by regional fossil fuel combustion correlating with dust as it would seem difficult to me to temporally separate local road dust emissions and vehicle emissions. My other concern is that PMF uses a fixed source profile and if there are changes in European car emissions, could other factors be increasing in contribution to compensate? How does the traffic emission factor change in say the first half and second half of the research campaign?

First of all, we would like to recall that the regional fossil fuel combustion factor was only resolved at AMX. This factor presents no correlation neither with traffic ($R^2 = 0.06$) nor mineral dust ($R^2 = 0.04$) identified at NICTRA. Therefore, its trend is unlikely to be associated with local traffic or road dust emissions from Nicosia city.

PMF source apportionment was also conducted for the beginning (2015) and the end (2023) of the study period to evaluate potential changes in chemical factor profile for the traffic source. No substantial differences are observed between 2015 and 2023, either in terms of concentrations of major species or relative contributions (%) of various tracers. Specifically, the OC/EC ratios are nearly identical for 2015 and 2023 (1.78 and 1.77, respectively). The two profiles were further compared using the standardized

identity distance (SID), one of the similarity metrics proposed by the Joint Research Centre. The obtained SID value (0.7) indicates that the two source profiles are similar.

Technical Corrections:

This paper overall reads smoothly with minimal typos. I've added some comments suggesting ways sentences can be restructured to improve readability and clarity.

Line 85

“Following rigorously” to “rigorously following”

Amended, as suggested.

Line 87

“Then, they were” to “They were then”

This was taken into account in the revised manuscript.

Line 147

“Allows building” to “builds”

This has been corrected accordingly.

Line 147

Remove “herewith”

As suggested, we have removed “herewith”.

Line 162

“details” to “detail”

“details” was replaced by “detail” as suggested by the reviewer.

Line 340

“offsetting completely” to “completely offsetting”.

Changes were made in the revised manuscript, accordingly.

Line 417

“minimal variations (statistically insignificant)” to “minimal and statistically significant variations”

Here we meant no statistically significant variations. Changes have been made in the revised manuscript.

Line 424

“As illustrated in Figure 5b, besides having the highest sulfate levels, the West Turkey sector also shows an increasing trend” to “As illustrated in Figure 5b, the West Turkey sector has the highest sulfate levels and also shows an increasing trend”.

Given that this paragraph focuses on sulfate trends rather than concentration levels, the sentence was revised as follows: “As illustrated in Figure 5b, the West Turkey sector not only has the highest sulfate levels but also exhibits an increasing trend....”

Line 429

“Surprising” to “Surprisingly”

We thank the reviewer for pointing out this typo, which has been corrected in the revised manuscript.

Line 440

“Such approach” to “This approach”

As suggested by the reviewer, “Such approach” was replaced by “This approach”.

Line 447

“being common” to “were common”

We have kept “being common” instead of “were common” in this context because it better fits the flow of the manuscript.

Line 448

“at AMX” to “only at AMX”

The sentence was revised accordingly.

Line 461

“(nearly double that of North Africa) to “that is nearly double that of North Africa)”

The sentence was revised following the reviewer’s suggestion.

Line 466

“uncontrolled emissions from local sources (road dust resuspension and biomass burning),” to “uncontrolled emissions, road dust resuspension and biomass burning, from local sources,”

We thank the reviewer for this suggestion. However, we have decided to keep this sentence in its initial form because it better fits the flow of the manuscript.

REFERENCES

- Shaheen, A., Wu, R., Yousefi, R., Wang, F., Ge, Q., Kaskaoutis, D. G., Wang, J., Alpert, P., and Munawar, I.: Spatio-temporal changes of spring-summer dust AOD over the Eastern Mediterranean and the Middle East : Reversal of dust trends and associated meteorological effects, Atmos. Res., 281, 106509, <https://doi.org/10.1016/j.atmosres.2022.106509>, 2023.
- Turpin, B. J. and Lim, H. J.: Species contributions to pm2.5 mass concentrations: Revisiting common assumptions for estimating organic mass, Aerosol Sci. Technol., 35, 602–610, <https://doi.org/10.1080/02786820119445>, 2001.