

The authors thank the referees to review our manuscript and particularly for the valuable comments and suggestions that have significantly improved the manuscript. We provide below point-by-point responses (in blue) to the referees' comments and have made changes accordingly in the revised manuscript.

Referee #1

The authors have addressed all my questions. Thanks.

Response: Thank you for reviewing our manuscript and making valuable comments and suggestions, which have greatly improved the manuscript. Thank you very much.

Referee #4

The manuscript has some values as a measurement report because it adds a dataset for the characterization of oxidative potential in conditions of high concentrations with a good chemical analysis including also insoluble and soluble trace elements that may be useful to the scientific community. The post processing with correlations and application of receptor model is quite standard with limited novelty. Nevertheless the publication of the manuscript may be valuable. I understand that is already a revised version, nonetheless, there are some aspects not clear, see my specific comments, that should be addressed in a further revision step before considering the paper for publication.

Specific comments

I found a little confusing that the same symbols are used for total and soluble trace elements. Authors should clearly mention in caption and in text to what fraction they refer when discussing correlation, concentrations and so on.

Response: Thanks. We have re-checked the manuscript and made corresponding modifications to any confusion. In line 245 of the revised manuscript, we changed "...the concentration trends of trace elements" to "...the concentration

trends of total trace elements” In line 807 of the revised manuscript, we changed “...(e) elements.” to “...(e) total elements.” The elements in table S1 have also been specified as total elements.

Line 43. I would not say one of the main but one possible mechanism.

Response: Thanks. Change made.

Lines 43-46. I would suggest to mention the recent work of Guascito et al (Journal of Hazardous Materials 448, 130872, 2023) that correlates oxidative potential with biological effects at different sites.

Response: Thank you for your suggestion and this work is worth mentioning. This work has been added as a reference. In line 45 of the revised manuscript, it now reads “... Guascito et al., 2023).”

Lines 83-98. In this part discussing the literature findings, I suggest to mention the efforts in studying size segregated oxidative potential, for example Basis et al (Toxics 11 (1), 59, 2023).

Response: Thank you for your suggestion. In lines 90-98 of the revised manuscript, it now reads, “...Some studies have also measured the OP of particles with different particle sizes, and reported that smaller size fractions typically have higher ROS activity compared to large PM size fractions (Saffari et al., 2014; Shafer et al., 2016; Basis et al., 2023). For example, Basis et al. (2023) measured the OP of water-soluble fraction of size segregated PM (< 0.49, 0.49-0.95, 0.95-1.5, 1.5-3.0, 3.0-7.2 and > 7.2 μm) collected during the cold and warm periods at an urban site in Thessaloniki, northern Greece, and the results showed that the total DTT activity of the PM < 3 μm size fraction were higher (2-5 times) than that of PM > 3 μm size fraction in both warm and cold periods.”

Line 167. Better small rather than little.

Response: Thanks. Change made.

It is mentioned the analysis of levoglucosan, mannosan and others. These are used in PMF but not reported in the tables, why? I strongly suggest to have a table with all measured concentrations.

Response: Thank you for pointing this out. The concentrations of measured organic markers (including levoglucosan, mannosan, galactosan and hopanes) have now been added to Table S2. In lines 346-347 of the revised manuscript, it now reads, “...The average concentrations of these organic markers are shown in Table S2.”

PMF application. It is not clear if two separate runs have been done of only one. Because in some parts they speak about a limited number of samples like if two runs were done. However, results only include one series of profiles like if all samples were pooled together in a single run. This must be explained. I believe that, considering the limited number of samples, the option to use a single run with all samples may give more stable results from a statistical point of view. In any case, it is necessary to give more details on the PMF results and approach. For example, weak or bad chemical species; why it has not been used the WSOC? Stability of the solution in terms of rotational ambiguity, bootstrap and so on. I suggest to have a look at the work of Belis et al, (Atmospheric environment: X 5, 100053, 2020) for receptor models performances.

Response: Thank you for your careful reading and professional comments. In this study, all samples were input into one single run, so there was only one series of profiles. In lines 214-215 of the revised manuscript, it now reads “...For each site, 31 samples (a total of 62 samples) and 23 species were input into PMF model.”

In this study, the source apportionment of DTT_v was performed using PMF as implemented by the multilinear engine (ME-2; Paatero, 1997) via the source-finder (SoFi) interface written in Igor WaveMetrics (Canonaco et al., 2013). Compared to EPA PMF, which does not require any prior information but may have a substantial degree of rotational ambiguity (Paatero, 1997), ME-2 can partly constrain the factors based on a priori information (such as factor profiles)

to reduce rotational ambiguity and direct the solution towards environmentally-meaningful rotations (Huang et al., 2014; Lin et al., 2018). In PMF&ME-2 model, species cannot be set as “Strong”, “Weak” or “Bad”. The species input into the PMF model all have concentrations much higher than the corresponding uncertainty values, with a signal-to-noise ratio (S/N) > 2. In line 79 of the Supporting Information, it now reads “All data input into PMF model has a signal-to-noise ratio (S/N) greater than 2.”

In order to obtain more accurate and refined source apportionment results, only tracer species were input into PMF model, and thus WSOC and NACs were not used.

In PMF&ME-2 model, there are no bootstrap (BS), displacement (DISP), and bootstrap combined with displacement (BS-DISP) results. To reduce rotational ambiguity and obtain an environmentally reasonable solution, the factor profiles were partly constrained in PMF&ME-2 model according to previous studies (Huang et al., 2014; Wang et al., 2017), as shown in Table R1 (added as Table S1). The uncertainties for PMF analysis of these sources were 2-14%. In lines 219-220 of the revised manuscript, it now reads, “...For a clear separation of sources profiles, the contribution of corresponding markers was set to 0 in the sources unrelated to the markers (see Table S1).” In lines 368-369 of the revised manuscript, it now reads, “...The uncertainties of PMF analysis for these sources were 2-14%.”

Table R1. F matrix elements constrained in the ME-2/chemical species 6 factors solution. The 0 value denote the $f_{h,j}$ values constrained in ME-2, while hyphens denote unconstrained elements.

Species	Biomass burning	Coal Burning	Traffic-related	Secondary Formation	Dust	Oil combustion
DTTv	-	-	-	-	-	-
Ti	-	-	-	0	-	-
V	-	-	-	0	-	-
Cr	-	-	-	0	-	-
Mn	-	-	-	0	-	-
Fe	-	-	-	0	-	-
Co	-	-	-	0	-	-
Ni	-	-	-	0	-	-
Cu	-	-	-	0	-	-
Zn	-	-	-	0	-	-
As	-	-	-	0	-	-
Sr	-	-	-	0	-	-
Cd	-	-	-	0	-	-
Ba	-	-	-	0	-	-
Pb	-	-	-	0	-	-
Picene	-	-	-	0	0	-
Hopanes	0	-	-	0	0	0
Galactosan	-	0	0	0	0	0
Mannosan	-	0	0	0	0	0
Levoglucosan	-	0	0	0	0	0
<i>o</i> -ph	0	0	0	-	0	0
<i>m</i> -ph	0	0	0	-	0	0
<i>p</i> -ph	0	0	0	-	0	0

The discussion of the difference between results in North and South is not very conclusive, probably further studies with longer datasets are needed and this should be reported in the conclusions (updating lines 421-424). In addition, in lines 240-255, I would suggest to mention that there are other studies showing that samples with similar concentrations may have a strong differences in DTT activity or the other way around interpreted by difference in chemical composition as well as possible antagonistic and synergistic effects, see for example, Lionetto et al (Atmosphere 12 (4), 464, 2021).

Response: Thank you for your professional comments and suggestions. In lines

436-439 of the revised manuscript, it now reads “...Besides, in order to gain a more comprehensive understanding of the regional differences in PM_{2.5} OP, sources and its relationship with chemical composition, longer periods and different seasonal datasets are also need to be studied in the future.” In lines 253-257 of the revised manuscript, it now reads “... Ahmad et al. (2021) also reported that the concentrations of PM_{2.5}, WSOC, and most elements in Lahore, Pakistan, were higher than those in Peshawar, Pakistan, while the DTT_v values of the two sites were similar, and the DTT_m value in Peshawar was higher than that in Lahore.” In lines 263-266 of the revised manuscript, it now reads “...Due to the complex chemical composition of PM_{2.5}, there may also be antagonistic and synergistic effects, contributing to the inconsistent relationship between DTT activity and compounds content(Xiong et al., 2017; Lionetto et al., 2021).”

Line 281. Not toxicity here just oxidative potential.

Response: Thanks. Change made.

Figure 1. Are these total or soluble data for trace elements? The same for Table S1.

Response: Figure 1 and Table S1 both show total trace elements. Corresponding clarifications have been made.

Figure 3. Just use a single threshold for statistical significance. It is not necessary to use two. The same for fig. S6.

Response: Change made.

References

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