

Response to Referee number 1

27 March 2024

The authors would like to thank Referee no. 1 very much for her/his expert, detailed and valuable comments which allow us to further improve and clarify the MS. We have considered all recommendations and made the appropriate alterations. As a follow up of a comment (of Referee no. 2), we realised that the dispersion correction was based on the seasonal mean VC values, and not on the mean VC averaged for the overall dataset. The latter choice is in accordance with the original idea and further adaptations of the method. Therefore, we repeated the calculations with the appropriate correction factor. Some numbers and figures were slightly modified because of this, while the tendencies remained the same. To completely fulfill another request, we inserted a new section 3.3 Relevance of the dispersion correction, and consequently extended the text by more detailed discussions, performed structural alterations or reorganisations at several places in the MS. We also adopted some additional smaller changes. Furthermore, we prepared in total 10 new compound figures for various purposes and inserted them into the Supplement. They all improved the quality of the revised material and further validated or documented the interpretations. Our specific responses to the comments are as follows, while most textual modifications amended to the MS can be traced in its marked-up version, which is available online.

Specific comments

Line 69. Please define WS the first time that it is used.

Response 1: The definition of the abbreviation WS was shifted from Line 129 to Line 69 of the original MS.

Lines 87-96. Regarding the use an the performances of receptor models, I suggest to mention the work of Belis et al (Atmospheric environment: X 5, 100053, 2020).

Response 2: The reference to this review-type paper was also added.

Line 105. Better sources rather than source.

Response 3: Adopted.

Lines 110-117. If I have well understood the measurements were taken at the second site only during one year and not in parallel and a direct comparison of the two sites was not provided. In the time series analysed, the data have been put all together? This should be mentioned here.

Response 4: This was specified in Lines 141–142 of the original MS. Nevertheless, we inserted two brief sentences already at this place to indicate these details as: “The exact timings of the measurement years are detailed in Table S1. The experimental data from the two measurement sites were merged into one dataset which was evaluated jointly.”

Lines 168-179. What values are used for A? In addition, the C3 value of 0.2 was used for each channel or only for the total concentrations (i.e. the sum of all channels)?

Response 5: The fine tuning of α for particle number concentrations was performed by changing the multiplication factor A from 0.1 to 5. The factor C_3 was used for the total particle number concentration and pollutants. With this realisation, we avoided tuning the parameters on the sample-by-sample basis. Since these two values control the deviation of the variables from zero and from the measured concentrations, respectively, it was necessary to change some other parameters as well to achieve good fits. The final parameters are summarised in Table S3. The missing details of this aspect were adopted in the MS, a new Table S3 containing the final values of the uncertainty parameters, and new composite Figs. S1–S4 on additional uncertainty analyses were inserted to the Supplement.

It is interesting the comparison between the traditional PMF and that corrected with ventilation coefficient. I have not understood if the correction has been done at hourly level in the dataset. If yes, how calm of wind or absence of wind have been treated? In addition, the comments in lines 599-605 seems to be oriented in looking at the corrected PMF as more reliable, however, this is not demonstrated. Is there any reason to think that results with correction are more reliable than the traditional ones? If not, better to modify this sentence.

Response 6: The ventilation correction was performed using the hourly mean datasets. The hourly mean WS values were obtained from the 10-min WS and WD data using vectorial averaging. The occurrence of the zero hourly-mean WS value was very low in the resulted dataset; the share of $WS < 0.1 \text{ m s}^{-1}$ was 0.06 %. An explanatory sentence was added as: “The ventilation coefficient represents the maximum volume into which the particles undergo dilution after their release into or formation within the ambient air per unit time. The main purpose of this data treatment is to correct each concentration data to have the same VC as the mean VC over the whole, 11-year-long dataset. The latter quantity was $1768 \text{ m}^2 \text{ s}^{-1}$ in our case.

It is challenging to directly prove the higher reliability of the DC-PMF results with respect to the uncorrected outcomes. We added the following two arguments: “It was demonstrated earlier (e.g., for Budapest lastly in Salma et al., 2020) that the local meteorological properties can influence the ambient atmospheric concentrations and size distributions in cities in a comparable extent than the changes in the source intensities (Li et al., 2023). The dispersion correction was dedicatedly introduced to remove a large part of the extra covariance between the variables, which is frequently or enduringly caused by the common effect of the meteorology on all concentrations. This basic motivation already implies that the corrected concentrations and concentration contributions are expected to be closer to reality and of higher reliability than their uncorrected counterparts. At the same time, the correction did not considerably alter the source profiles, temporal behaviours and patterns. Furthermore, some previous papers have also demonstrated the value of the dispersion correction in estimating the source contributions (e.g., Dai et al., 2020, 2021; Hopke et al., 2024).”

Discussion of Figure 1. The midday peak seems actually to be present only during the warm seasons, rather than in every season as it seems to be mentioned here. Better to add a legend on Fig. 1.

Response 7: The midday peak in the N_{6-1000} diel variation is visually more obvious in spring and summer than in the other seasons. Its interpretation was based on both our current and previous research work indicated by the cited references. They jointly prove the diel structure. The sentence under consideration was reformulated to indicate the basis of our statement more precisely as: “Conclusively, there are three peaks present with variable relative areas in the diel variations; namely an early-morning peak and an evening peak at the rush hours of 06:00–08:00 and 18:00–21:00, respectively, largely generated by vehicular road traffic, and a midday peak predominantly produced by NPF events driven by photochemistry.” The requested legend was also added to Figure 1.

Line 339. I would not say contributions. The factors are loaded with NO_x meaning that there is an association among particles in these factors and gas but not a contribution. The same for line 396.

Response 8: The sentences were reformulated to: “Both factors were strongly associated with NO, NO₂ and CO as well.” and to “The factor was also considerably associated with SO₂ and PM₁₀ mass.”

Section 3.2.3. Is it possible that this source includes a contribution from resuspended dust, for example road dust resuspended by traffic? This may be possible for particles around 0.5 μm or more, see for example Conte et al. (Environmental Pollution 251, 830-838, 2019).

Response 9: The road and soil dust resuspension by moving vehicles cannot be excluded. Nevertheless, its contribution to N_{6-1000} for the urban diffuse source is expected to be limited in Budapest. The N_{6-1000} for this source showed lower levels in summer and spring than in winter and autumn (Fig. S6c), while the former seasons are generally dryer than the latter periods. Hence, the expected change in the resuspension intensity does not show up in the concentrations. It is mentioned that scattering de-icing mixtures of sand and salts on roads in winter, which could confuse this conclusion, is limited both in its extent and frequency in Budapest. A brief sentence was added: “In principle, resuspension of road and soil dust particles could also add (Conte et al., 2019) as a minor contributor in Budapest.”

Imre Salma
for the coauthors