Itemized Response to Reviewer's Comments

Ms. Ref. No.: ACP-2021-1016

Title: Measurement report: Intra/interannual variability and source apportionment of VOCs during 2018–2020 in Zhengzhou, Central China

1.Some factors have been renamed in the text but not in the corresponding figures. In some figures the factor names are updated and in others not.

Figure 5 has both old and new factor names in the same figure. In addition it has two version of supposedly the same? conditional probability analysis (one with a line underneath the corresponding BW plot and one in filled at the bottom). Unfortunately the overall shape of the curve appears to be very different between the two versions for the same factor. E.g. on the top the biogenic factor has a 0.35 conditional probability to the north (0 to 30 degree) and also towards 150 degree. Yet in the bottom version the conditional probability towards both these directions is < 0.25. Similar problems exist in all panels. Which version is the reader supposed to believe? Are these for different percentiles? For different model runs?

Response: Sorry for the mistake. Figure 1 was drawn using the updated PMF results. Combined with the related sources around the site, the latest version is credible.

The inconsistency in Fig.5 is given by a different PMF run. To better identify each pollution source, the factors are constrained by using tool of toggle constraint in the PMF model based on your constructive comments. After adjustment, the dQ value is still within a reasonable range, which proves that the output result is reasonable. The constrained results with new factor profile are more relevant to the local source profiles and emission inventory, so they can better reflect the actual situation of the local atmosphere.

Meanwhile, we have done the following work, which may affect the results of CPF and PMF.

1. Reject 0 value: Due to instrument failure, some values of wind speed and direction were recorded as 0, which were not excluded in previous studies. In addition, some values in the time series of the PMF results are negative. The above abnormal values were eliminated in the latest study.

2. Unit of PMF: The reviewer mentioned that the unit of PMF should be microgram per m^3 instead of ppbv. Thus, we are running the PMF with mixing ratios in $\mu g/cm^3$.

3. PMF model settings: The technical guide for source analysis of ozone pollution in ambient air requires the error fraction (EF) of VOCs to be

within 30%. The EF value used in previous studies was 30%, while it was set to 10% of the VOC concentration in the latest studies. Meanwhile, information on which species were weak and strong is missing. We have corrected it. VOC species were grouped into strong, weak, and bad according to their signal/noise ratio (S/N), and there were 22 and 4 species grouped into strong and weak, respectively.

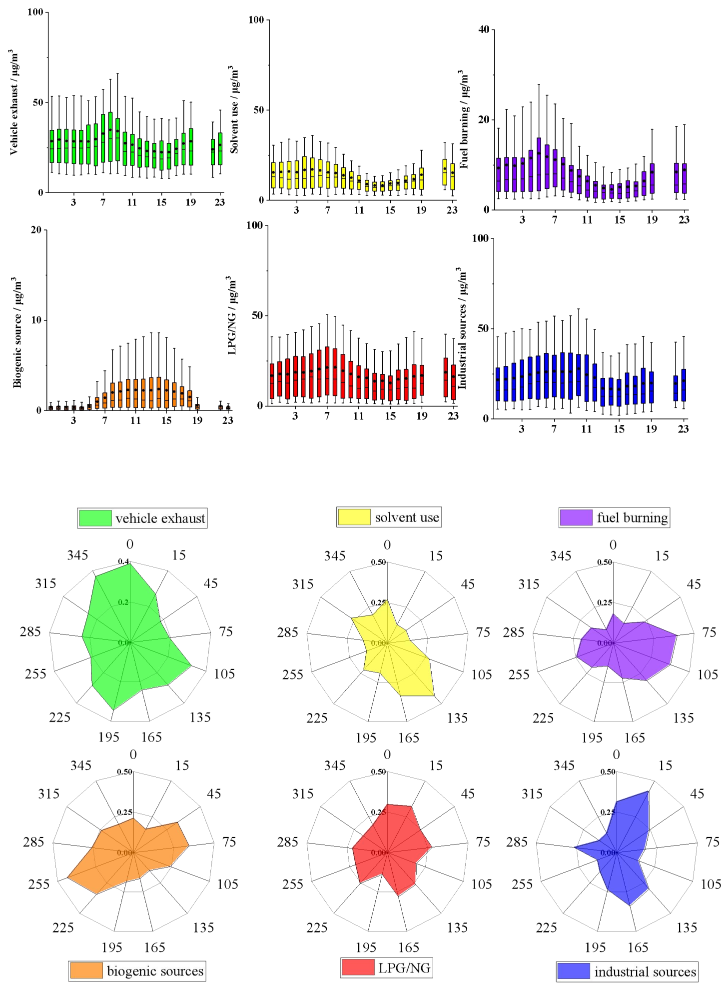
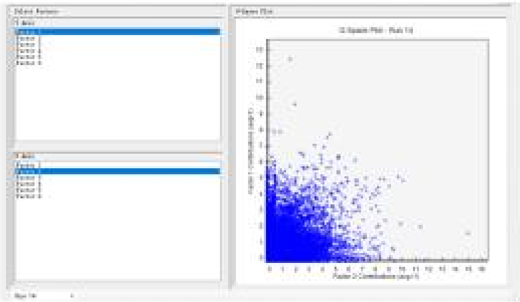


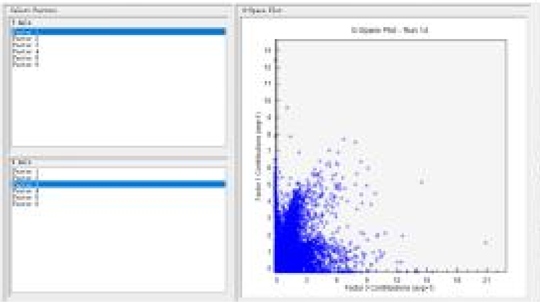
Fig. 1 Directional dependence and hourly record of each source in Zhengzhou.

2.The Q ratios which the authors use to argue the validity of their solution are not a reliable quality indicator when used in isolation. Specifically, they cannot identify a solution with too much rotational ambiguity. In general, the better the Q ratio the higher the risk of having a solution with too much rotational ambiguity. Such solutions also often cause problems in the bootstrap runs (unmapped factors or poor mapping of bootstrap factors on the original factor). Rotational ambiguity can only be assessed with the help of G-space plots or a cross correlation analysis of the factor contribution time series of all factors. Either the G-space plot or the R of the factor pairs needs to be provided. If any factor pair has an R >0.6 then this is a strong indication that there is too much rotational ambiguity and that the same source is getting split into two factors.

Response: Thank you for your suggestions. Choosing the optimal number of factors (P-value) is a critical question in PMF analysis. According to previous studies (Baudic, et al. 2016; Hui, et al. 2020; Liu, et al. 2020; Song, et al. 2019; Wang, et al. 2021; Zheng, et al. 2018), the ratios of Q (ture)/Q (robust) and Q/Qexpected (Qexp) were tested to determine optimum solution.

The G-Space Plot screen (Figure 2) shows scatter plots of one factor versus another factor, which can be used to assess the relationship between source contributions. The results show that there is no correlation between different factors. The G-space scatter diagram is evenly distributed and the edge is parallel to the X and Y axes, and the result is reliable.


G-Space Plot - Run 14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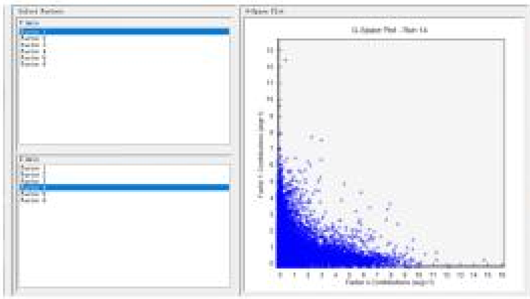
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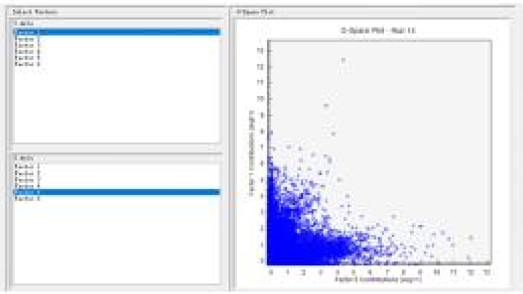
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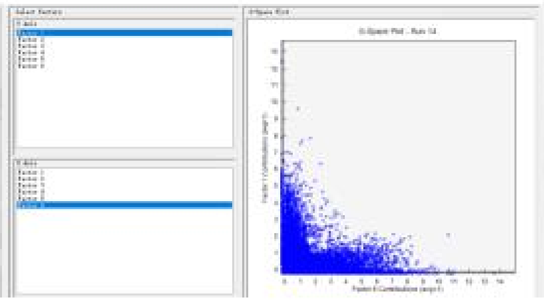
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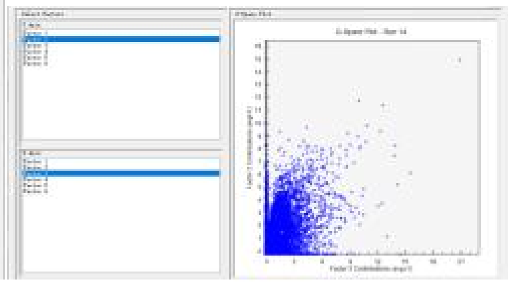
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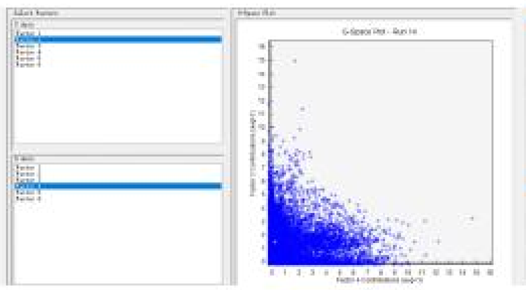
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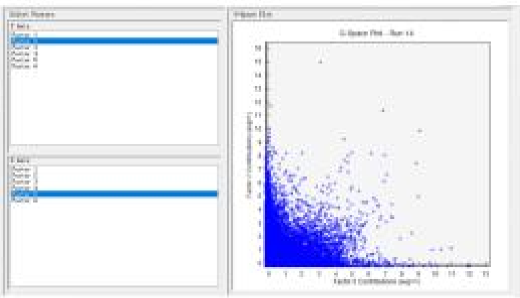
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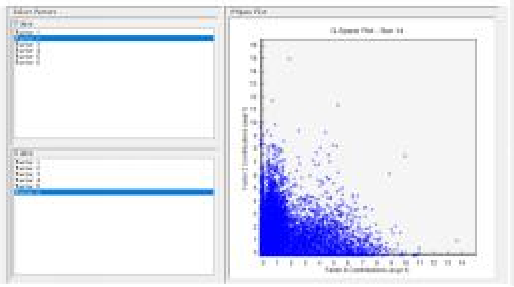
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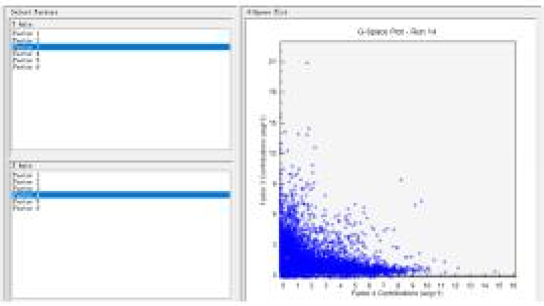
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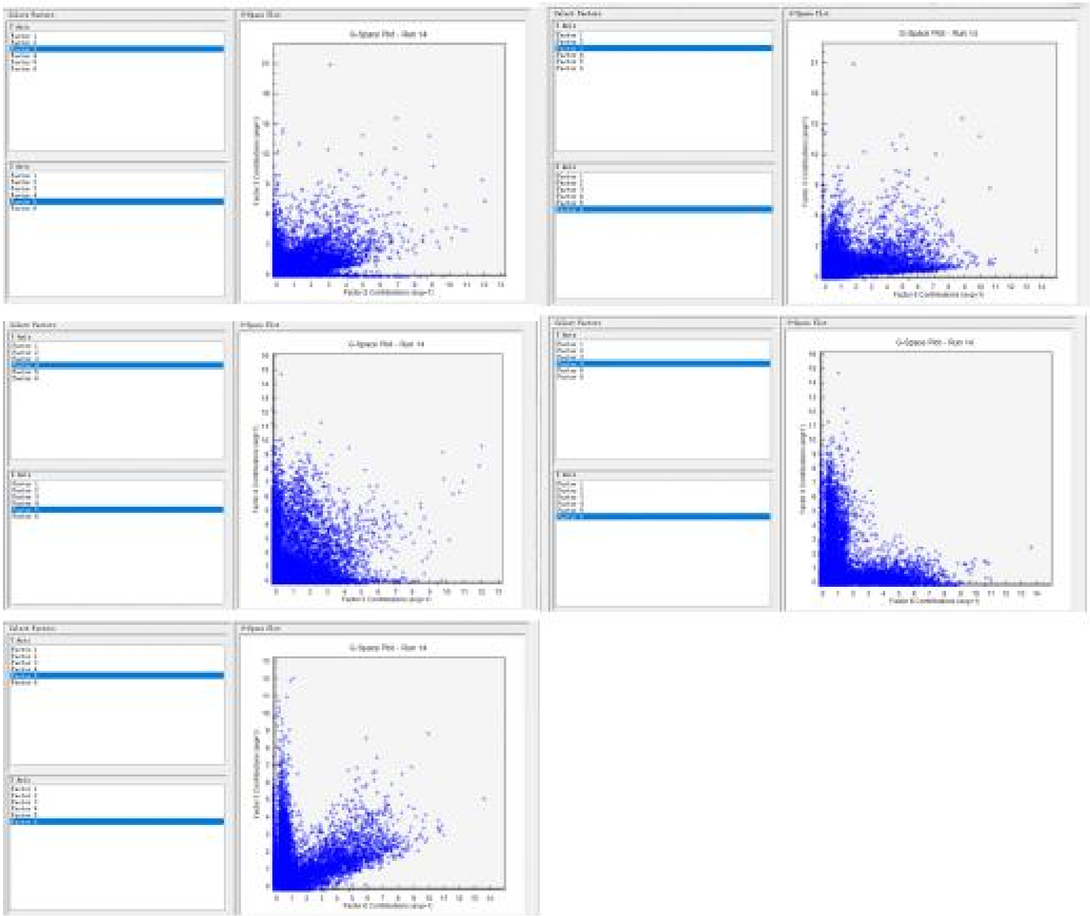

Fig. 2 The G-Space Plot by using PMF model.

Baudic, Alexia, et al.2016 Seasonal variability and source apportionment of volatile organic compounds (VOCs) in the Paris megacity (France). Atmospheric Chemistry and Physics 16(18):11961-11989.

Hui, Lirong, et al.2020 VOC characteristics, chemical reactivity and sources in urban Wuhan, central China. Atmospheric Environment 224.

Liu, Y., et al.2020 Characterization and sources of volatile organic compounds (VOCs) and their related changes during ozone pollution days in 2016 in Beijing, China. Environ Pollut 257:113599.

Song, Mengdi, et al.2019 Sources and abatement mechanisms of VOCs in southern China. Atmospheric Environment 201:28-40.

Wang, M., et al.2021 Impact of COVID-19 lockdown on ambient levels and sources of volatile organic compounds (VOCs) in Nanjing, China. Sci Total Environ 757:143823.

Zheng, Huang, et al.2018 Monitoring of volatile organic compounds (VOCs) from an oil and gas station in northwest China for 1 year. Atmospheric Chemistry and Physics 18(7):4567-4595

3.Furthermore, the authors should upload the dataset used for their study in a public repository (e.g. Zenodo), and cite the corresponding doi.

Response: The dataset has been uploaded to the public repository (Zenodo), and the link is https://zenodo.org/record/6815259#.Ysq_lnZBy3A .