

For comment 1

General Comments:

Overall, this study applied the observational data and air quality model to study how emission control can improve the air quality in Zhengzhou. I think it can be published after the authors address and fix some issues.

Response: Thank you for your careful reading of our paper and the valuable comments and constructive suggestions. Below are the point-to-point responses to all the comments (The comments are marked in black font and the responses are marked in dark blue font). The major changes that have been made according to these responses were marked in yellow color in the highlighted copy of the revised manuscript. And our own minor changes were marked in red font. Note that the following line numbers are shown in the corrected version.

Please check the detail reply from supplement file.

Specific Comments:

1. "ppbv" and " $\mu\text{g}/\text{m}^3$ " are both used in the manuscript, it is better to use the consistent unit either "ppbv" or " $\mu\text{g}/\text{m}^3$."

Response: Thank you for your suggestions. The units have been homogenized into ug/cm^3 . For the record, some thresholds are obtained by referring to previous references, and their units are fixed. Therefore, when studying the specific ratios (*i/n*-pentane, T/B and VOCs/NO_x), the unit used in this paper is still ppbv rather than ug/cm^3 .

2. Line 249-251: "The highest hourly...respectively." Please confirm that the max hourly NO_x is 357 $\mu\text{g}/\text{m}^3$ and the VOC concentration is 238ppb during the pre-NMG, and explain when they happened because I didn't see the NO_x concentration is higher than 357 $\mu\text{g}/\text{m}^3$, and the VOC concentration is not higher than 120 ppbv in Figure 1.

Response: Sorry for the misunderstanding. The max hourly NO_x has been confirmed, and NO_x concentration is the sum of NO and NO₂. As for VOC concentration in Fig.1, the Y-axis was wrong and has been corrected. It should be noted that the VOC concentration in Figure 1 has been updated from ppbv to $\mu\text{g}/\text{m}^3$.

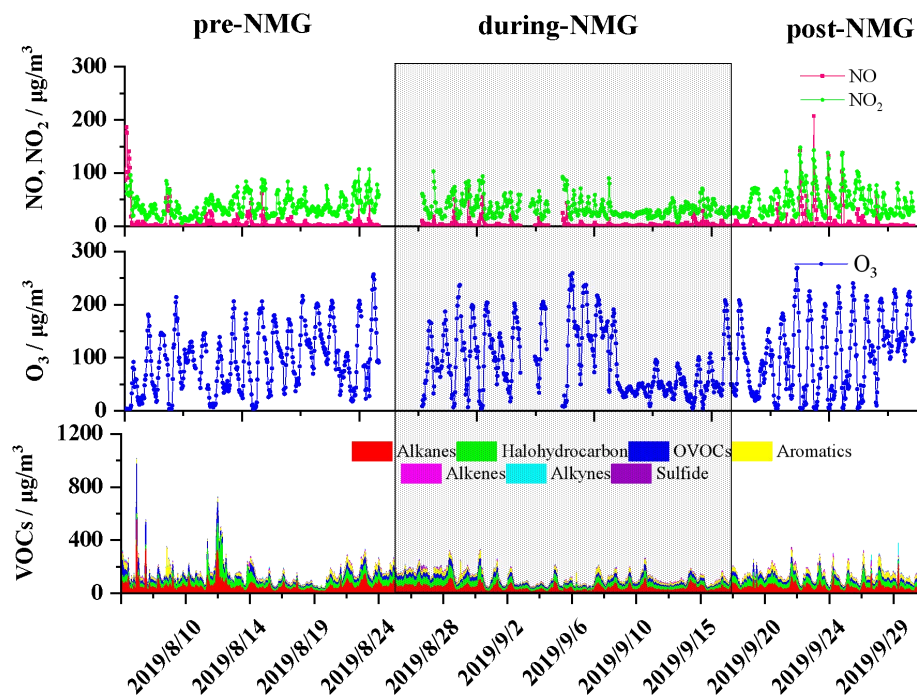


Fig. 1 Time series of VOCs and trace gases during the sampling period in Zhengzhou.

- Line 254-255, “the O₃ precursor concentrations decreased significantly...NO_x”, It is hard to identify the NO_x and VOC time series data change significantly; the T-test or box plots can be used to explain they are different.

Response: Thanks for your suggestions. The box plots have been used to explain the difference in the O₃ precursor concentrations. During the control period, the mean concentrations of VOCs and NO_x were reduced by about 19% and 11%, therefore, the description of Line 254-255 has been corrected to “the O₃ precursor concentrations showed a decreasing trend”.

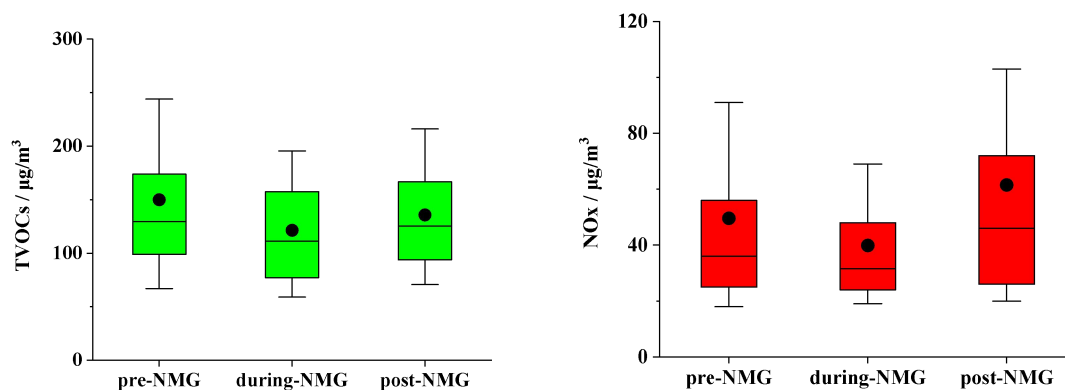


Fig. S4 The box plots for NOx and TVOCs in Zhengzhou during the three periods.

- Line 257 “levelsaccounted,” typo.

Response: Sorry for the mistake. We have corrected it. (L 262)

- Why 2019/08/26-2019/09/08 have high ozone and NOx, but in the same NMG period 2019/09/10-2019/09/15, ozone and NOx level decrease a lot??

Response: During the pre-NMG period, NOx emission intensity is high due to the lack of control. Combined with appropriate meteorological conditions, O₃ concentration is high.

During the NMG period, emission reduction leads to lower NOx concentration, and more rainfall leads to lower mean concentrations of NOx and O₃.

- Line 308, “As illustrated in Fig. 2”, I think it is Fig. 1. Typo.

Response: Sorry for the mistake. We have corrected it. (L 314)

- In Table1, the total VOC concentration or sum of the top 20 VOCs can be presented in the bottom row.

Response: Thanks for your suggestions. The sum of the top 20 VOCs has been presented in the bottom row. And the units (µg/cm³) have been homogenized.

- Line 331, why discuss isoprene here? The isoprene is not in the top 20 VOCs.

Response: Considering that isoprene is a typical biogenic tracer, its characteristics

need to be investigated. However, the isoprene is not in the top 20 VOCs. Therefore, the description of isoprene has been removed.

9. Line 367, why the chloromethane continually increases a lot?

Response: Fig. S11 shows the hotspots diagram of Zhengzhou and its surrounding areas during the observation period, and the number of fire spots in September was significantly higher than that in August. And chloromethane is a tracer of biomass burning. Therefore, the chloromethane continually increases a lot during the control period.

10. Figure 4. In the x-axis “m,” typo.

Response: Sorry for the mistake. We have corrected it. (Figure 4)

11. Figure 5, the time series plot is hard to compare the data pattern in those three periods; the box plot or range plot for each period may be a better way to present.

Response: Thanks for your suggestions. The box plots have been supplemented.

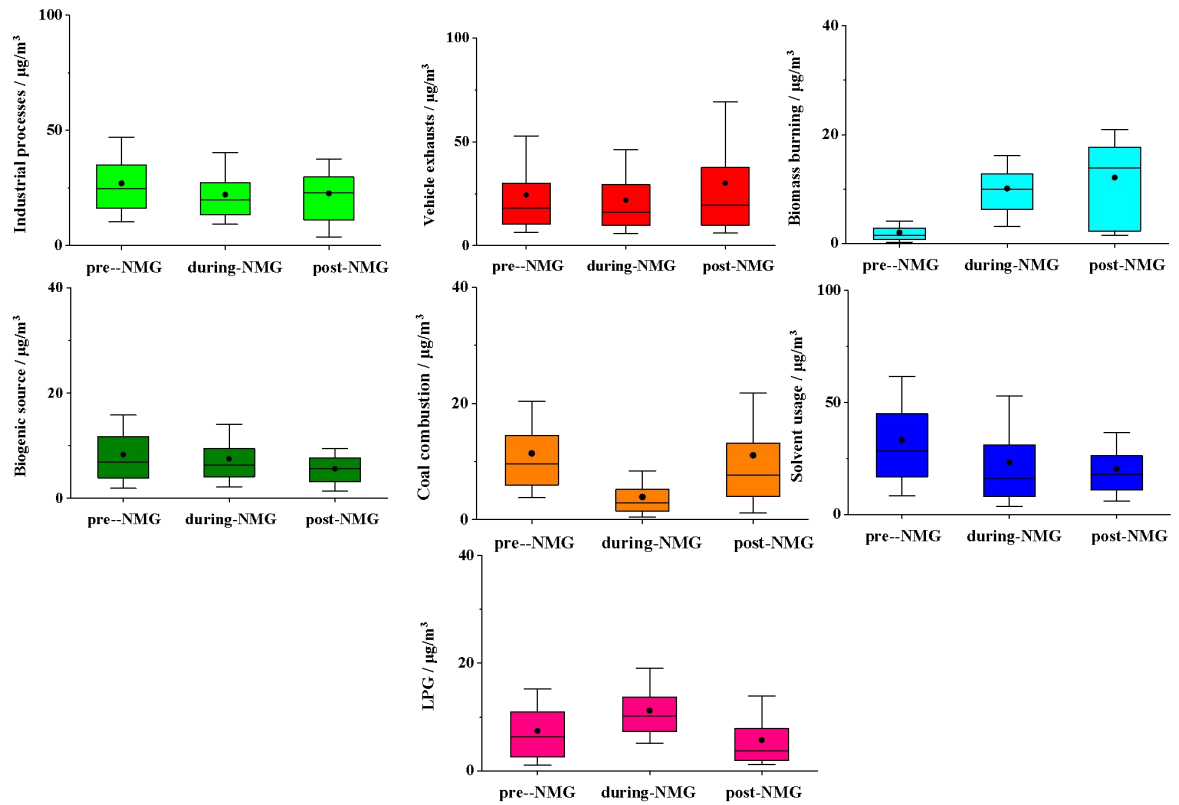


Fig. S10 The box plots for each identified source during the three periods.

12. Figure 8, the three panels are not the same size.

Response: Thanks for your suggestions. We have corrected it. (Figure 8)

13. Table list, Table 2 OFP contributions (“ $\mu\text{g}/\text{cm}^3$ ”), please confirm it is correct.

Please confirm all units are consistent.

Response: Table 2 OFP contributions (“ $\mu\text{g}/\text{cm}^3$ ”) has been confirmed, and all units have been consistent.

14. Figure S10 should provide the data in the support document.

Response: As shown in Table S6, the data has been provided in the support document.

Table S6 The concentrations of VOCs groups to the total OFP

	Alkanes	Alkenes	Aromatics	Halohydrocarbons	OVOCs	Sulfide	Alkyne
pre-NMG	48.5	30.5	98.7	2.8	56.2	0.1	2.9
during-NMG	36	21.9	91.2	2.8	30	0.2	1.3
post-NMG	41.5	21.4	124.6	3.5	27.7	0.2	2.7

15. Line 510 “However.....ozone”, following this sentence, the following section should connect with ozone sensitivity study, but the next section is “3.4.2 Risk assessment of individual VOC species”. This may confuse the readers.

Response:Sorry for the misunderstanding. In subsection 3.4, the atmospheric environmental implications of VOCs were discussed by calculating the values of OFP and risk assessment. We have interchanged the order of 3.4.1 and 3.4.2 to ensure logical consistency.

16. Line 565-568, should be in the method section.

Response:Thanks for your suggestions. We have corrected it.

For comment 2

The study by Shijie Yu et al. is a comprehensive assessment of the air quality over Zhengzhou before, during and after the 2019 National Minority Games, during which policy making decisions reduced the emission sources to investigate the implications on the local air quality. This is an excellent study for identifying and quantifying the impact of reduced anthropogenic sources. The paper is very written and it is particularly interesting that the authors have thoroughly analysed the data, providing PMF analysis, modelling, PSCF, O₃ formation potential and eventually health risk assessment. That said, there are a couple of critical points in the study that should be better clarified before publication.

Response: We appreciated it very much for the reviewer's positive comments and valuable suggestions. Below are the point-to-point responses to all of the comments (The comments are marked in black color and the responses are marked in dark blue color). The major changes that have been made according to these responses were marked in yellow color in the highlighted copy of the revised manuscript. And our own minor changes were marked in red font. Note that the following line numbers are shown in the corrected version.

General comments

1. It seems that the ozone formation potential remained unchanged compared to the control and post-event periods. The factors of 0.23 and 0.17 that were derived shall include the uncertainties before reaching a conclusion on whether the reduction of regional emissions had indeed an impact. Generally, the study fails to provide qualitative evidence on the significance of the emission reductions but this is explained by the complexity of regional (not only local) sources and meteorology. The study succeeds in reaching a clear conclusion (VOC:NO_x ratio has to be reduced by a factor of 2) but on the

other hand, ozone production is not an instantaneous process and the assessment does not include O₃ transport from other regions.

Response: Thanks for the suggestion. During the sampling periods, the values of OFP are 238 ± 166 (pre-NMG), 183 ± 115 (during-NMG) and 220 ± 149 $\mu\text{g}/\text{m}^3$ (post-NMG), respectively. And the values of control stage was reduced by 55 and 37 $\mu\text{g}/\text{m}^3$ than those before and after control period, respectively. In other words, OFP decreased during the control period. OFP is calculated from observed data, and the uncertainties is greatly affected by the instrument. The uncertainty of the instrument has been discussed in Section 2.2, and the coefficients of determination (R²) of calibration curves were mostly above 0.99 and the MDL ranged from 0.004 to 0.36 $\mu\text{g}/\text{m}^3$ for each species.

This article mainly studies effect of sporting events and the emission reductions from four aspects (L110-116). The results showed that the concentration of VOC, OFP and health risk were reduced during the control period. Unreasonable emission reduction may aggravate ozone pollution during control periods. It is suggested that emission reduction ratios of the precursors (VOC:NO_x) should be more than 2. The above research proved the significance of the emission reductions at a certain extent.

For the model, the influence of transmission is considered. Before the model was run, the local anthropogenic emissions were individually labeled for O₃-sensitive research, and the results were shown in Figure 9, then the control ratio of VOC/NO_x were got. In other words, when the chemical transport model run, the sensitivity of O₃ also considers local emissions, regional emissions, O₃ generated by natural sources, and transmitted O₃ influence from other regions, and Figure 9 shows the possible effects of controlling local anthropogenic emissions.

2. From Figure 1, the most interesting period seems to be the week of 8-15/9/2019. However, the authors provide little no discussion on this period. In the same lines, the authors conclude that the city of Zhengzhou is within the

VOC-limited but this period indicates the opposite: O₃ is strongly decreased together with the NO_x concentrations while VOCs remain unchanged. A whole new sub-chapter discussing this week and investigating the respective processes is essential.

Response: Thanks for the suggestion. From September 9th to 15th 2019, Judging from the results of observation, concentration changes of O₃ and NO_x seem to have a certain degree of synergy. In fact, the precipitation days were observed from September 10th to 15th, and the precipitation was 0.6 23.5 4.8 0.4 1.7 and 4.0 mm per day, respectively. Therefore, the main reasons for O₃ concentrations decrease were temperature decrease and weaken photochemical reactions of O₃ production caused by precipitation during research time. Considering that there is a lot of precipitation during this period, this part is mainly explained by the model results. That is, although O₃ concentrations and NO_x concentrations have been observed to decrease at the same time, the main contribution of the decrease in O₃ concentration comes from the decrease in light and temperature caused by precipitation, and the decrease in O₃ production, rather than the decrease in NO_x concentration. The precipitation data and related research can be found in our previous research (Su 2021 et al).

Su, F., Xu, Q., Wang, K., Yin, S., Wang, S., Zhang, R., Tang, X., Ying, Q., 2021. On the effectiveness of short-term intensive emission controls on ozone and particulate matter in a heavily polluted megacity in central China. *Atmos. Environ.* 246, 118111.

3. The references are ca 80% from Asian authors. While indeed the study has been conducted in an Asian city, the chemical processes and source composition signatures are universal. Highlight papers from atmospheric chemistry have been ignored and replaced with regional references and therefore, the authors should try to increase their literature, including international studies. Some examples are mentioned in the specific comments below but this comment applies to the complete text.

Response: Thank you for your suggestions. According to the suggestions, the references of this paper are improved. And highlight papers from atmospheric chemistry and international studies have been supplemented.

4. The units (ppb and ug/cm³) should be homogenized.

Response: Thank you for your careful reading of our paper and the valuable comments and constructive suggestions. The units have been homogenized into ug/cm³. For the record, some thresholds are obtained by referring to previous references, and their units are fixed. Therefore, when studying the specific ratios (*i/n*-pentane, T/B and VOCs/NO_x), the unit used in this paper is still ppbv rather than ug/cm³.

Specific comments

1. L41. Please include the uncertainties on the values

Response: Thanks for your suggestions. We have corrected it.

2. L59. 'Some' VOCs

Response: Sorry for the mistake. We have corrected it.

3. L104. Remove ‘,’

Response: Sorry for the mistake. We have corrected it.

4. L269. Please elaborate further. If ozone precursors decreased significantly (I would argue that this significance is not illustrated by the results presented) then apparently, the ozone has been transported from other regions.

Response: O₃ formation is a highly nonlinear process, thus varying NO_x and VOCs emissions may lead to fluctuations in ozone sensitivity to precursors. Therefore, identification in ozone sensitivity to precursors is essential to elucidating the underlying reasons of ozone increase.

The paper assess the O₃-NO_x-VOCs sensitivity and propose control strategies for ozone episodes. The results showed that Zhengzhou is under VOC-sensitive regimes. Cutting VOCs can alleviate ozone pollution, while excessively reducing NO_x concentration might lead to the increase of ozone concentration. Unreasonable emission reduction may aggravate ozone pollution during control periods. Therefore, the concentration of ozone precursors decreased during the control period, and the ozone pollution had not improved.

5. L312-314. It is not evident that this reduction is significant (Figures S5 and S6). Please include errorbars in Figure S5

Response: Thanks for your suggestions. The box plots have been used to explain the difference in VOCs concentrations.

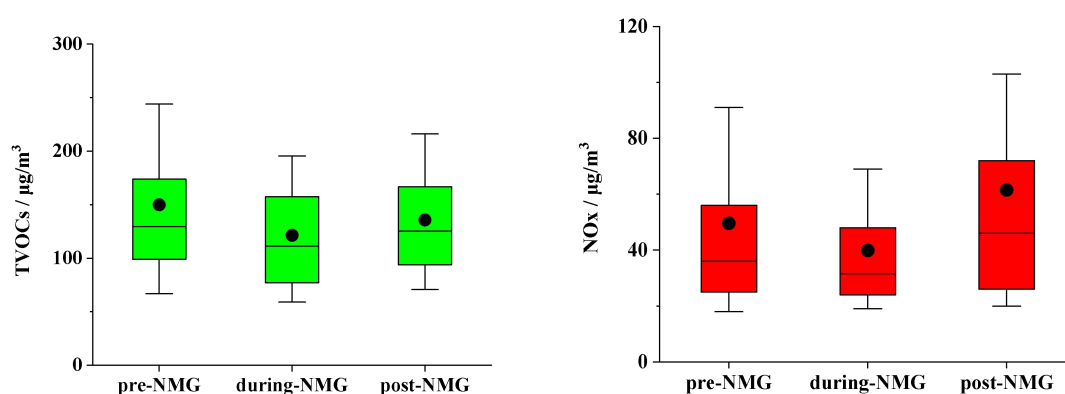


Fig. S4 The box plots for NO_x and TVOCs in Zhengzhou during the three periods.

6. L320-321. It seems that meteorological conditions and transport is more significant than the local emission reductions.

Response: Thanks. Meteorological conditions and transport is more significant that can influence pollutant compositions. And we have corrected it.

In fact some research have reported that control measures sometimes do not work well (Cheng et al., 2018; Xu et al., 2019). The implementation of short-term control

measures may change ozone sensitivity, and even increasing the O₃ concentration. But considering the long-term environmental improvement goal, these measures are necessary. At the same time, our research can also support managers to spend this intermediate stage reasonably without causing many environmental problems.

Xu, W., Liu, X., Liu, L., Dore, A.J., Tang, A., Lu, L., Wu, Q., Zhang, Y., Hao, T., Pan, Y., Chen, J., Zhang, F., 2019. Impact of emission controls on air quality in Beijing during APEC 2014: Implications from water-soluble ions and carbonaceous aerosol in PM_{2.5} and their precursors. *Atmos. Environ.* 210, 241-252.

Cheng, N., Chen, Z., Sun, F., Sun, R., Dong, X., Xie, X., 2018. Ground ozone concentrations over Beijing from 2004 to 2015: Variation patterns, indicative precursors and effects of emission-reduction. *Environ. Pollut.* 237:262-274.

7. L330-332. Isoprene is indeed considered as an excellent biogenic tracer. However, anthropogenic isoprene sources may be also considered.

Response: Thank you for providing useful information. However, the isoprene is not in the top 20 VOCs. And the description of isoprene has been removed.

8. L335-336. In line with my general comment, TVOCs seem to remain unchanged and therefore, the conclusion is driven mainly by model studies.

Response: Thanks for the suggestion. During the sampling periods, the values of TVOC are 150±93 (pre-NMG), 121±55 (during-NMG) and 136±60 µg/m³ (post-NMG), respectively. And the values of control stage was reduced by 29 and 15 µg/m³ than those before and after control period, respectively. In other words, TVOC decreased during the control period. As shown in Fig. S7, the diurnal variations of TVOCs during the three periods are similar. During the control period, the daytime TVOC concentration was significantly lower than last period, which proved that the emission reduction was effective.

9. L339. Biased reference selection on VOC oxidation.

Response:Sorry for the misunderstanding. And we have corrected it. (Carter, 2010)

Carter, W., 2010. Development of the SAPRC-07 chemical mechanism. *Atmos. Environ.* 44, 5324 – 5335.

10. L347-348. Halogenated compounds have increased lifetimes so claims on night-time chemistry are somehow unsuitable.

Response:Thank you for proving useful information. Therefore, we have deleted the description of halogenated compounds at night.

11. L350-353. I-n pentane is used for decades as an excellent tracer for source identification and there are dozens of widely cited studies to illustrate this. Please add more references.

Response:Thanks for your suggestions.The references have been supplemented.

(Gentner et al., 2009; Zheng, et al. 2018)

Zheng, H., Kong, S., Xing, X., Mao, Y., Hu, T., Ding, Y., Li, G., Liu, D., Li, S., Qi, S., 2018. Monitoring of volatile organic compounds (VOCs) from an oil and gas station in northwest China for 1 year. *Atmos. Chem. Phys.* 18, 4567-4595.

Gentner, D., Harley, R., Miller, A., Goldstein, A., 2009. Diurnal and seasonal variability of gasoline-related volatile organic compound emissions in Riverside, California. *Environ. Sci. Technol.* 43, 4247 – 4252.

12. L357-358. Please add more references.

Response:Thanks for your suggestions.The references have been supplemented.

(Brito et al., 2015; Dorter, et al. 2020; Yenisoy-Karakas, et al. 2020)

Brito, J., Wurm, F., Serrano, A., Assuno, J., Artaxo, P., Godoy, J., Artaxo, P., 2015. Vehicular Emission Ratios of VOCs in a Megacity Impacted by Extensive Ethanol

Use: Results of Ambient Measurements in Sao Paulo, Brazil. *Environ. Sci. Technol.* 49:11381-7.

Drter, M., Odabasi, M., Yenisoy-Karaka, S., 2020. Source apportionment of biogenic and anthropogenic VOCs in Bolu plateau. *Sci. Total Environ.* 731:139201.

Yenisoy-Karaka, S., Drter, M., Odabasi, M., 2020. Intraday and interday variations of 69 volatile organic compounds (BVOCs and AVOCs) and their source profiles at a semi-urban site. *Sci. Total Environ.* 723:138028.

13. L366. Biased reference selection.

Response: Sorry for the misunderstanding. And we have corrected it. (Mcroberts et al., 2015; Schauer et al., 2001)

Mcroberts, W., Keppler, F., Harper, D., Hamilton, J., 2015. Seasonal changes in chlorine and methoxyl content of leaves of deciduous trees and their impact on release of chloromethane and methanol at elevated temperatures. *Environ. Chem.* 12, 426-437.

Schauer, J., Kleeman, M., Cass, G., Simoneit, B., 2001. Measurement of emissions from air pollution sources. 3. C1–C29 organic compounds from fireplace combustion of wood. *Environ. Sci. Technol.* 35, 1716–1728.

14. L378. Figure S8. Seems that the benzene to toluene ratio splits in two clusters, further supporting the impression that the NMG period has two distinct chemical composition regimes that need to be further analysed and discussed.

Response: Thanks for your suggestions. In this study, measured ratios of toluene/benzene were selected as the potential emission sources. The results show that low correlations ($R^2 = 0.3-0.5$) were found during the three periods, suggesting a more complex set of sources for the two species. According to the ratios, the VOCs were

greatly influenced by the mixed source of coal combustion and vehicle emissions. However, this method aims to preliminarily judge the source of VOC and provide support for the source analysis results. Therefore, distinct chemical composition regimes of VOCs were distinguished by the PMF model.

15. L426-427. Isoprene emissions from vegetation should contain a reference from A. Guenther

Response: Thanks for your suggestions. Relevant references have been supplemented. (Guenther et al., 1995 and 1997)

Guenther, A., 1997. Seasonal and spatial variations in natural volatile organic compound emissions. *Ecol. Appl.* 7, 34–45.

Guenther, A., Hewitt, C.N., Erickson, D., Fall, R., Geron, C., Graedel, T., Harley, P., Klinger, L., Lerdau, M., McKay, W.A., Pierce, T., Scholes, B., Steinbrecher, R., Tallamraju, R., Taylor, J., Zimmerman, P., 1995. A global model of natural volatile organic compound emissions. *J. Geophys. Res.* 100, 8873–8892.

16. L497-499. Biased reference selection.

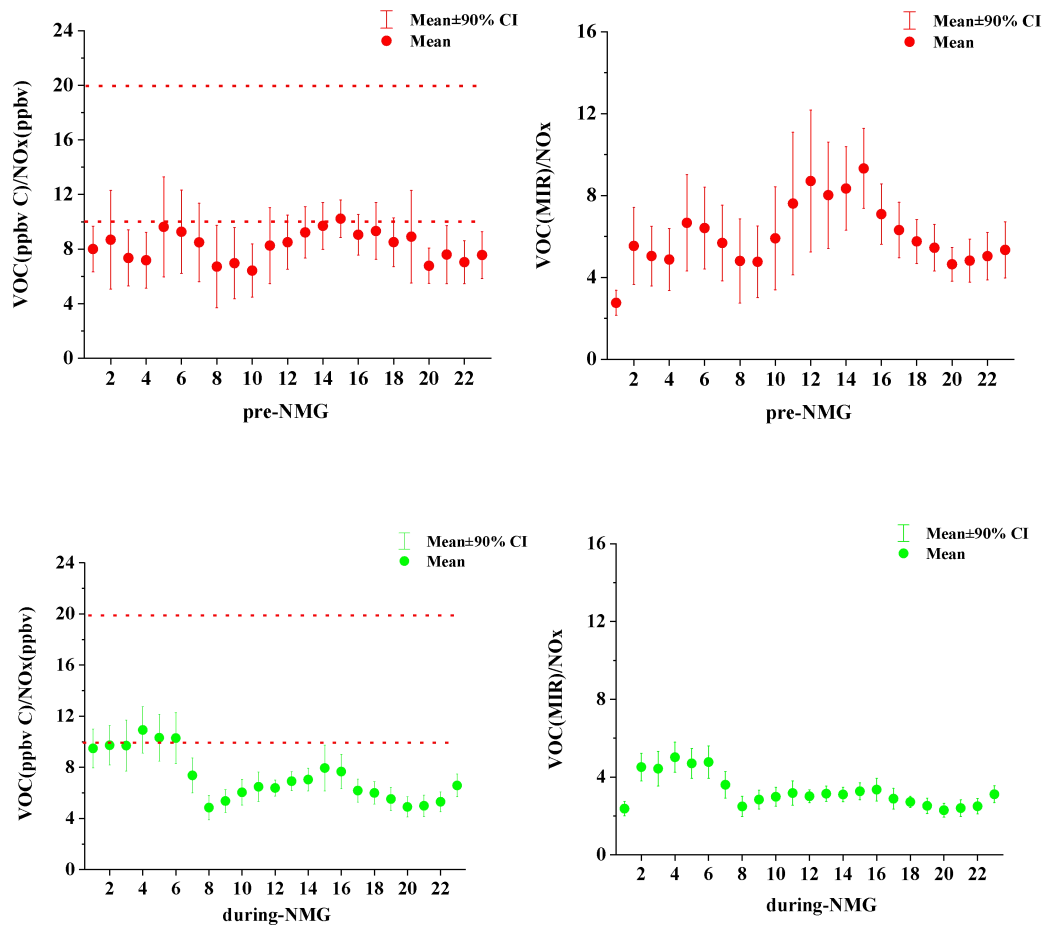
Response: Sorry for the misunderstanding. The reference has been updated. (Wang et al., 2013; Zou et al., 2015)

Wang, H., Chen, C., Wang, Q., Huang, C., Su, L., Huang, H., Lou, S., Zhou, M., Li, L., Qiao, L., Wang, Y., 2013. Chemical loss of volatile organic compounds and its impact on the source analysis through a two-year continuous measurement. *Atmos. Environ.* 80, 488–498.

Zou, Y., Deng, X.J., Zhu, D., Gong, D.C., Wang, H., Li, F., Tan, H.B., Deng, T., Mai, B.R., Liu, X.T., Wang, B.G., 2015. Characteristics of 1 year of observational data of VOCs, NO_x and O₃ at a suburban site in guangzhou, China. *Atmos. Chem. Phys.* 15, 6625–6636.

17. L550. It would be interesting to see the ozone production lines in a second y-axis.

Response: Thanks for your suggestions. The the ozone production lines have been supplemented. And the daily variation of VOC (MIR)/ NO_x is similar to that of VOCs/NO_x (ppbv C/ppbv).



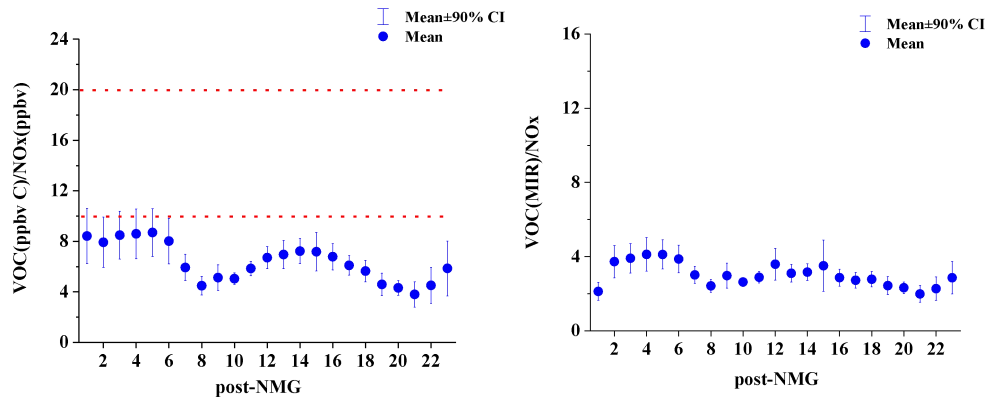


Fig. 8 Daily variations in the VOC/NOx ratio in Zhengzhou before, during, and after NMG periods.