

Title: Simultaneous observations of peroxyacetyl nitrate and ozone in central China during static management of COVID-19: Regional transport and thermal decomposition.

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Dear Editor

Thank you very much for your time and effort in revising this paper. We apologize for the issues that were not addressed in the revised manuscript. We checked each comment carefully and strive to provide a satisfactory answer. Changes made in response to these comments are marked in yellow in the highlighted copy of the revised version. Our own minor changes are highlighted in red.

The following is a point-by-point response to each reviewer's comments.

Reviewer #1:

your answer begins by implying that meteorology is most important since "emissions from pollutant sources usually change very little over a period of time" while meteorology does change, but later in the same paragraph you state that meteorological differences between the two periods are

"minor" (though the reader does not know what level of difference "minor" entails) and, without any evident justification, say that meteorology is not responsible for any differences between the pollution periods. It's also not clear what the sentence "when comparing the meteorological conditions of the two pollution processes, none of the processes showed a tendency to be more prone to pollution" means; what are the two "processes"? Overall, this section requires a more quantitative approach; by how much should temperature, wind speed, and humidity affect pollution levels (based either on theory or prior research in this area), and therefore, why can we be sure that the differences are not meteorological? It could be that the conclusion remains there's "no discernible trend towards greater susceptibility to pollution" (L280-281) in period 2, but that needs quantitative justification.

Response: Thank you very much for your suggestion. We have revised the presentation of the section:

Previous studies have shown that meteorological factors such as low WS, high RH, and low precipitation are responsible for the increase in PM_{2.5} pollution in Zhengzhou in winter (Duan et al., 2019). Our analysis of the correlation between different pollutants and meteorological conditions during the pollution period showed that PM_{2.5}, TVOCs and NO_x were positively correlated with relative humidity (Fig. S3), which is consistent with the results of some previous studies (Wang et al., 2019). Yu et al.

(2018) identified RH and WS as the most influential meteorological conditions of PM_{2.5} during winter. Their findings revealed a positive correlation between hourly PM_{2.5} concentrations and RH ($r = 0.84$, $p < 0.01$) and a negative correlation between PM_{2.5} concentrations and WS ($r = -0.62$, $p < 0.01$). The WS and RH between the infection and recovery periods were similar in this study which were largely considered to be of the same type of weather (Yu et al., 2018). However, the mean PM_{2.5} concentration during the recovery period was found to be 1.6 times higher than that observed during the infection period. Furthermore, the concentrations of other pollutants (including SO₂, NO₂, CO, and O₃) exhibited analogous trends during the infection and recovery periods. The concentration of TVOCs during the recovery period was 1.2 times higher than that during the infection period, exhibiting a significant upward trend following the resumption of production. It is notable that WS, which is only 0.3 m/s higher in Case 1 than in Case 2, and RH, which is 13% higher in Case 1 than in Case 2, were relatively stable, while the concentration of pollutants is significantly higher in Case 2 than in Case 1. This is presumably attributable to the resumption of production activities in Case 2, which resulted in a notable increase in emissions. Decreased trends of air pollutants were found in other studies before and after the outbreak of the novel coronavirus (COVID-19) in early 2020 (Qi et al., 2021; Wang et al., 2021).

Reviewer #2:

Line 110: This added text is extremely confusing and difficult to read. The tenses switch between present and past, and most of the sentences are run-ons with many connected clauses. It's not clear why details of the city's screening processes are needed for this manuscript. Ideally this paragraph could be clarified, substantially shortened, and boiled down to only the most salient aspects for the present analysis. It would also be helpful if statements about human behavior during the various periods (e.g. "the number of people moving around Zhengzhou has increased", "people basically rested at home", etc.) could be supported by citations to sources that corroborate this.

Response: I'm sorry for the problems that arose. We have simplified the paragraph and corrected the tenses. The revised content is as follows:

In this study, we conducted continuous online observations of VOCs during the polluted winter season at an urban site in Zhengzhou. The study covered the period following the removal of lockdown measures. We focused on pollution events when the daily average PM_{2.5} concentration exceeded 75 µg/m³ (China's Class II standard) for more than three consecutive days. Days with PM_{2.5} concentrations below 35 µg/m³ (China's Class I standard) were classified as clean days. During this period, China lifted zero-COVID strategies, announcing the '10 measures' for optimizing

COVID-19 rules on December 7, 2022 (http://www.news.cn/politics/2022-12/07/c_1129189285.htm, Accessed Jan 2024). Zhengzhou's epidemic prevention and control measures changed with the issuance of Circular No. 163 on December 4, 2022, which allowed the reopening of closed public places. As a result, movement within Zhengzhou increased and social production resumed. Our research specifically examines the period dominated by the COVID-19 Omicron variant, where they demonstrate notable differences from the early virus strains (i.e., original SARS-CoV-2 virus and Delta) in terms of geographical transmission, the scale of the infected population, and symptom manifestation (Petersen et al., 2022; Merino et al., 2023).

Results & Discussion question 1: your response does not address the reviewer's point that the NO_x decreased in the recovery period.

Response: I apologize for omitting a response to this question due to my oversight. Thank you very much for raising it. The reason for the reduction of NO_x in Zhengzhou during the recovery period (here the reviewer actually refers to Case 2) may be related to the reduction of motor vehicle travel. A few days before Case 2, the Zhengzhou Municipal People's Government initiated a severe pollution weather level II response (<https://sthjj.zhengzhou.gov.cn/tzgg/7037130.jhtml>) and introduced emission control measures for industrial and mobile sources. We counted

the concentrations of NO_x at other monitoring sites in the urban area of Zhengzhou during Case 1 and Case 2 (Table 1). Compared with Case 1, both found that Case 2 NO_x concentrations decreased somewhat.

Table 1. NO_x value of other urban sites in Zhengzhou (μg/m³)

Monitoring station	Case 1	Case 2
Station 1	111.0 ± 56.4	108.1 ± 52.2
Station 2	117.0 ± 67.6	112.3 ± 70.5
Station 3	113.0 ± 103.1	93.7 ± 55.8
Station 4	115.1 ± 61.8	114.7 ± 66.9

on VOC ratios -- my interpretation is that the reviewer was not implying that the isomer ratio method has limitations, but that its application is incorrect here, and that hasn't been fixed in your revisions. For example, your measured ratio of isobutane to n-butane is squarely within the range for LPG, above the range for vehicle emissions, and below the range for natural gas emissions (L338-340). Possible interpretations of your measured value of 0.5 imply (a) a mix between all three sources, (b) a mix between vehicle and natural gas sources, or (c) emissions dominated by LPG. However, you conclude on L342 that your measured value implies a contribution from natural gas, without explaining how you got there, even

though that doesn't line up with the range in which the measured value falls. The analysis of isopentane/n-pentane ratios seems similarly problematic (L332-337); the measured ratio of 1.4 doesn't necessarily mean that pentane is "mainly" derived from liquid petrol and fuel evaporation, since the same ratio could be achieved by mixing sources from coal and vehicle exhaust in the proper ratio. The analysis in this section needs reworking.

Response: Thank you very much for your suggestions. We didn't understand the reviewer's comments before and have some issues with the use of VOC ratios. We have revised this aspect of the analysis and incorporated additional information into Fig. 3. We believe that this modification offers a more illustrative representation of the distribution of VOC ratios across a given source interval, thereby providing a more robust foundation for the conclusions presented in this section. The modifications are as follows:

The toluene-to-benzene ratio (T/B ratio) was widely used to assess the relative importance of different sources. Specifically, T/B ratio with a value of 1.3-3.0 was observed in vehicle emissions for vehicles with different fuel types (Schauer et al., 2002; Wang et al., 2015). The reported T/B ratio for combustion processes was between 0.13 and 0.7 (Li et al., 2011; Wang et al., 2014). The mean value of T/B ratio for the entire period was 1.0, with the majority of the data (99%) falling between 0.1 and 3.0 and

concentrated within the 0.7-1.3 range (49%). This suggests that both traffic emissions and combustion are significant sources of VOCs.

The isopentane/n-pentane concentration ratios of 0.6-0.8 represent mainly coal combustion emissions, ratios of 0.8-0.9 represent LPG emissions, 2.2-3.8 represent vehicle exhaust emissions, and 1.8-4.6 represent fuel evaporation (Conner et al., 1995; Liu et al., 2008; Li et al., 2019). The sources of isopentane and n-pentane in this study were intricate and multifaceted. The mean isopentane/n-pentane ratio was 1.4, with the majority of data points (99%) falling within the range of 0.1-4.6, with a notable concentration in the 0.8 to 1.8 interval. This indicates that pentane is influenced by a combination of emissions from LPG and fuel evaporation.

Isobutane/n-butane concentration ratios of 0.2-0.3 represent vehicle emissions, 0.4-0.6 represent LPG usage, and 0.6-1.0 represent natural gas emissions (Russo et al., 2010; Zheng et al., 2018). The mean isobutane/n-butane ratio in this study was 0.5, with the majority of data points (99%) falling within the 0.4-0.6 range, indicating that VOCs at the observation sites were significantly influenced by the use of LPG. (Shao et al., 2016; Zeng et al., 2023).

The ratio of X/E can be used to infer the photochemical age of the air mass. X/E ratios around 2.5-2.9 are typical of urban areas, indicating that VOCs

are mainly from the urban area (fresh air mass) (Kumar et al., 2018). When this ratio is significantly lower than 3.0, it indicates that VOCs are mainly transported from distant sources (aging air masses) (Kumar et al., 2018). The average X/E value in this study was 2.0 (Fig. 3(d)), indicating low photochemical activity and aging of the air mass at the observation site. Potential source analyses also indicate that air masses are affected by long-range transport (Fig. S4).

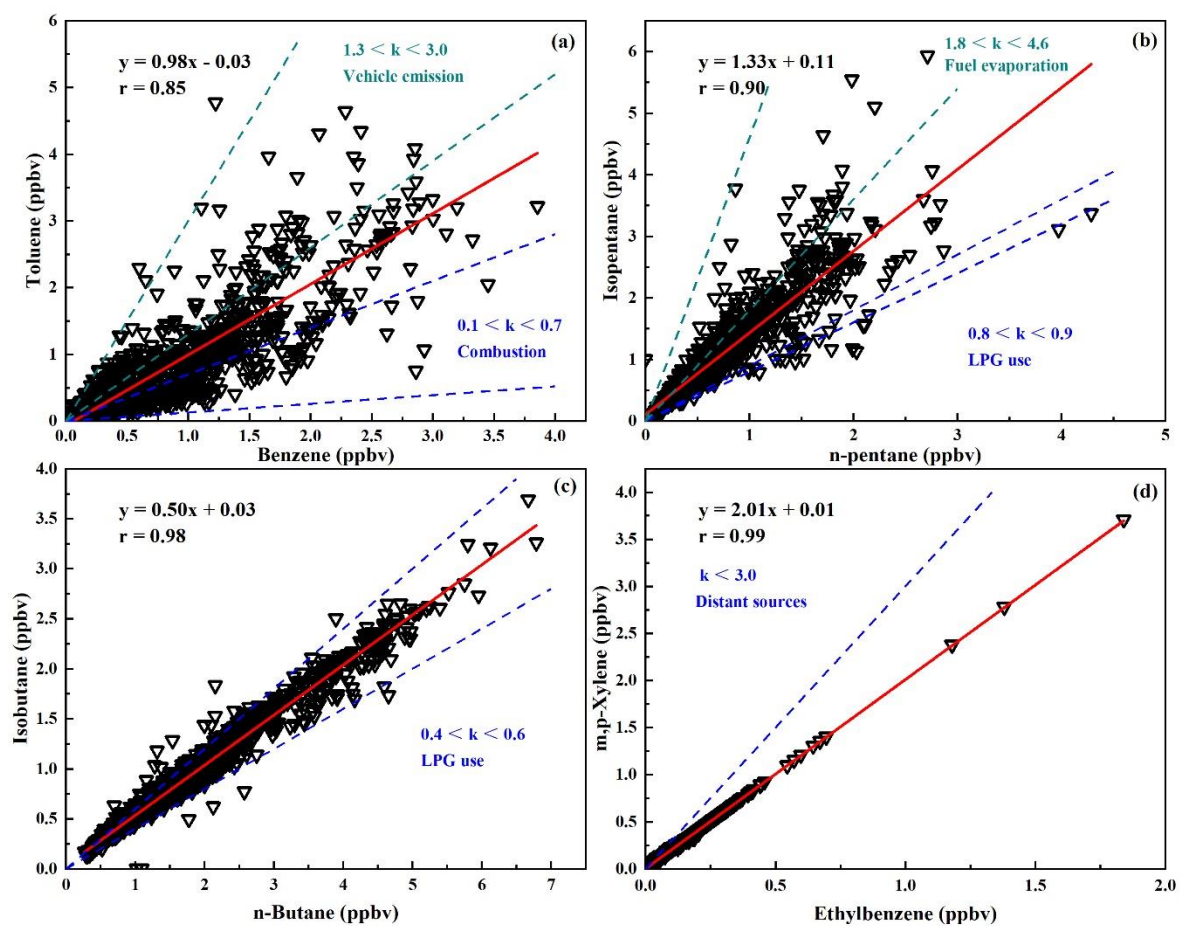


Fig. 3. Correlation analysis between specific VOC species.