

Comment & response

Overview

1. The manuscript requires extensive English language editing. It is not currently suitable for publication as there are too many places in which weaknesses in English make it unclear, ambiguous or difficult to understand the point the authors are making;

2. The abbreviation “PAMS” stands for Photochemical Assessment Monitoring Stations, referring to the locations at which VOC sampling and monitoring are conducted. It does not refer to the compounds sampled, for which “VOC” (or VOCp to distinguish those isolated here from the generic term) should be used.

3. My chief concern with this work is the limited (and potentially biased) nature of the data collected, as outlined above. This precludes the authors from reaching robust conclusions regarding meteorological drivers of ozone, inter-annual variability, and the precise cause of the observed increase in ozone in Zhengzhou City, as well as preventing them from being able to offer clear policy advice regarding emissions controls.

4. The authors need to give far more detail of the prevailing meteorology in Zhengzhou, at the very least the typical intra- and inter-annual variability to put the sampling time period into context

5. Some of the analysis techniques used have associated limitations which the authors do not discuss. For example, PMF analysis requires that the inputs (here the concentrations) are independent when that will not be the case here. PMF is a well-established and accepted method for source apportionment analysis but the authors should be clear about its limitations. Likewise, the actual rate of ozone formation from any specific VOC is strongly dependent on both chemical and meteorological conditions and can only be reliably estimated using detailed atmospheric chemistry models. While ozone formation potentials based on constant maximal incremental reactivity (MIR) ratios are a useful indicator of which VOCs may be most important to control they are only an indicator. Again the authors need to be clear about this. The following reference may be of use: “Photochemical ozone creation potentials for organic compounds in northwest Europe calculated with a master chemical mechanism”. Derwent, R.G. et al., 1998; doi: 10.1016/S1352-2310(98)00053-3

6. I cannot over-emphasize the importance of wind direction to the analyses presented here. It is critical in terms of both the transport of longer-lived and secondary pollutants and the local production. Local production is affected by meteorology which is often synoptic in scale and therefore correlated with windspeed and direction. Meteorological conditions will affect both photochemical efficiency and pollutant source strengths. This importance should be reflected in both the text and the figures. The back-trajectories tucked away right at the end as Fig. 10 should be incorporated into Fig. 1 and presented in the text ahead of the analysis of possible local production sources.

7. Following on from this, there is also a real need for a windrose plot showing met

conditions of importance (from the text = T, RH, although radiation would also be useful) and concentrations of the various pollutants for each of the 4 locations and possibly also split out by month. While I appreciate the authors have attempted to highlight the contribution of wind direction (and speed) in Fig. 4 and through various color-coding in Fig. 7 I don't think these give the clear oversight required given the key role winds play. See e.g.

<http://www.openair-project.org/examples/BivariatePolarPlots.aspx> or

<http://www.openair-project.org/examples/windpollutionroses.aspx> for open source visualization tools

8. As the manuscript stands, the SI seems rather unnecessary as it consists of a single plot. I have however made several recommendations below regarding moving material out of main text

Response:

Thanks for the reviewer's suggestion and comments on our submitted manuscript. We have made point-to-point responses to each comment as below in detail.

Introduction:

Comment 1

This section is particularly difficult to follow. It is hard to work out which parts refer to previous work and how relevant these are. The authors do not explain how the different regimes reported from e.g. Los Angeles and SE USA relate to Zhengzhou.

Response:

The statement has been revised. The original aim of the statement was to support the importance of VOCs studies in different regions. This has been deleted according to suggestion.

Comment 2

Many of the references are not the most appropriate to the point the authors appear to be making. For example, Capps et al applied a methodology that was developed previously by e.g. Carter et al., Derwent et al., etc.

Response:

We have made more clarification and checked the citations. The reference of Capps et al was replaced by Carter et al.(1994) as suggested.

“Carter, W. Development of Ozone Reactivity Scales for Volatile Organic Compounds; J.

Air & Waste Manage. Assoc. 1994, 44, 881-899. “

Comment 3

L81: “one of the most polluted cities” where? In China? Asia? Globally?

Response:

The statement has been revised as:

“With the rapid growth of industrial activities, as well as increased vehicle emissions and fuel combustion, air quality in Zhengzhou has notoriously deteriorated, exceeding the allowable limits by 65% of days (air quality index, AQI>100) in a year set by Air Quality Guideline in 2013 ”.

Comment 4

L81-2: “its air quality exceeds the allowable limits set by Air Quality Guidelines” - specifically which pollutants exceeded the limits and what are the limits

Response:

The limits are referred to the guideline of air quality index (AQI) in China. In 2012, the Chinese Ministry of Environmental Protection (CMEP) issued the Ambient Air Quality Index (AQI) Technical Provisions (Trial) (HJ 633 – 2012) on the basis of the United States Environmental Protection Agency (U.S.EPA) AQI objective, and it was implemented as the Chinese Ambient Air Quality Standard (CAAQS) (GB 3095-2012). The AQI level is determined by the concentrations of six criteria pollutants including SO₂, NO₂, CO, O₃, PM_{2.5} and PM₁₀. Basically, the AQI acts as a guideline for local government to inform the public and to take proper health protection measures.

Experimental:

Sampling site:

Comment 5

What was the sampling duration and flow rate?

Response:

The description was revised as

“The whole air samples were collected in one minute using 3.2 L stainless-steel canisters (Entech Instrument, Inc., Simi Valley, CA, USA), which were pre-cleaned with high purity nitrogen and pressurized to 20 psi”.

Comment 6

Why did the authors select 07:00 and 14:00 as the two sampling times? How do these relate with rush hour? Or mealtimes? Or the working day?

Response 6

The statement has been revised as

“Two samples, one collected at 07:00 with increasing of human activities and another one collected at 14:00 with well-mixed of ambient air, were obtained on each sampling day”.

Chemical analysis:**Comment 7**

What was the specific compound mix in each of the three standard gases? There are issues regarding extrapolating area: concentration scaling factors from 1 compound to another even for structurally similar compounds and those with similar retention times (see e.g. Ruiz-Hernandez et al., 2018; doi:10.1186/s13007-018-0335-2))

Response:

The 57 VOCs (defined as the most critical contributors for ozone) discussed in this paper were quantified with the PAMS standard gas (1 ppm; Spectra Gases Inc, NJ, USA) containing 28 alkanes, 11 alkenes, acetylene and 17 aromatics. The other two standard gases were used to quantify halocarbons and oxygenated VOCs, but they have not been discussed in this manuscript. In order to avoid confusion, the information for the two standard gases has been removed. The extrapolating method has not been used. The co-eluted compounds of m-xylene and p-xylene were reported as m,p-xylene.

QA/QC:**Comment 8**

This text could be moved to the SI if it is retained

Response:

Suggestion taken. The texts have been moved to SI.

PMF:**Comment 9**

The authors go into far more detail of the specific equations (which are a standard technique) than is necessary in the main text. I suggest that the mathematical details of PMF are moved

to the SI and the authors give more information regarding precisely how it was applied to their data.

The authors also need to include a statement regarding the limitations of applying PMF here.

Response:

Thanks for the suggestion. The content in Section 2.3 has been rewritten.

Results and discussions

Mixing ratios and meteorological variations:

Comment 10

“VOCs” not “PAMS” Are the reported meteorological data over the entire month or just the 10 days each month when the sampling was conducted?

Response:

The term of “PAMS” has been replaced with “VOCs” in the text. The meteorological data were covered the 10 sampling days in each month.

Comment 11

Are the average VOC concentrations for all samples taken in that month, i.e. combining the 07:00 and 14:00 sampling times? This would be misleading as it would be expected that values and sources differ markedly between those two times of day.

Response:

Yes, the average concentrations represent the values from two sections (i.e., 07:00 and 14:00) for each sampling day.

We do understand the concern about the variations of concentrations and contributions of the sources from time to time. There were many factors impacted the VOCs levels and composition indeed, while in this study we focus on the impacts from meteorological conditions in different month. Therefore, we have depicted the case by the statement as

“This can be attributed to numerous factors that will be explored later in the paper. Besides the emission sources (to be discussed in Section 3.2), the impacts controlled by meteorological conditions should not be ignored as well”.

And, the diurnal variations have been discussed in other sections.

Comment 12

The large variability (reported standard deviations and month-to-month differences) indicate the clear need for sampling to continue over a much longer time period, taking in different

times of day and for a number of years.

Response:

The objective of this study is to illustrate the characteristics of VOCs during the high ozone level period in a year. The sampling has been conducted for five months in a year, when were sufficiently representative.

Comment 13

L199: “more accumulated at GS”? I don’t understand what point the authors are making here. Do they refer to higher concentrations? Greater influence of transported pollution?

Response:

The sentence was revised as

“The average wind speed at GS ($0.74\pm0.33\text{ m s}^{-1}$) was lower than that at MEM ($1.84\pm0.94\text{ m s}^{-1}$) and YH ($0.97\pm0.36\text{ m s}^{-1}$) (Table 3), indicating poor dispersion conditions at GS. The air pollutants emitted from MEM and YH were more liable accumulated at GS, resulting in a higher level of ΣVOCs at GS in June.”

Comment 14

L205: Likewise I don’t understand what the authors mean by “topographical effect

Response:

Topographical effect is referred to a factor impacted the distribution of VOCs. In this study, the sites of GS and MEM were located at the western part of Zhengzhou, while JK and YH were belong to the eastern part. Since the VOCs levels gradually increased from east to west, we have thus suspected that the concentrations of VOCs could be correlated with the topography.

Comment 15

The authors have used PMF to identify source sectors and present the findings of this analysis in section 3.6. what is presented here is speculation which is entirely superfluous given they have used PMF later.

Response:

Suggestion taken. We have removed Section 3.6 and incorporated it into the whole discussion section.

Comment 16

It would seem to me to make more sense for the authors to discuss likely influences from

long-range transport of pollutants (i.e. HYSPLIT back-trajectory analyses here shown in Section 3.7) BEFORE considering local sources.

Response:

Thanks for the suggestion. This has been moved to Section 3.1.

Comment 17

L208: The authors describe the results as showing “the general consistency of pollution sources in the region” but the large variability in the reported averages do not appear to suggest that.

Response:

Even though the concentrations were variable, the compositions of major compounds were similar at every site. Therefore, we do conclude that there were consistent emission sources for each site.

Comment 18

L221-222: The authors have not convincingly demonstrated this in their presented results.

Response:

The paragraph has been revised as,

“Among the four major organic classes, alkane was the most abundant group as a result of busy traffic in urban city and its longevity (Fig.7), and accounted for 52.85%, 62.52%, 53.38%, 53.39% of the total Σ VOCs values at JK, MEM, GS, and YH, respectively. The highest composition of alkane was observed at MEM due to the stronger contributions of ethane, iso-pentane, cyclohexane, methylcyclohexane, 2-methylhexane and 3-methylhexane (Fig. S3), which were identified as the components heavily impacted by petrochemical industries (Jobson et al., 2004; Mo et al., 2015). In accordance with the higher contribution of petrochemical source (11.56%) at MEM (Fig. 3)”

Comment 19

L231-232: Samples were taken at 07:00 and 14:00 only. The authors cannot make general comments about morning and afternoon as they have not presented any data to suggest that the conditions at 07:00 (14:00) persist throughout the morning (afternoon)

Response:

The paragraph has been revised as,

“Due to the variation of the planet boundary layer (PBL) height, solar radiation and

emission sources etc. day and night, the concentrations of VOCs displayed obvious differences between morning and afternoon time (07:00LT and 14:00LT in this study). Compared with morning time, the aromatic compounds showed lower compositions around 14:00 LT (Fig. 7), because of the increased planet boundary layer and the active photochemical reactions, while alkenes always peaked in afternoon time. According to the dataset, the increases in alkene compositions (about 4.3% uplift) were mainly due to higher contributions of isoprene (about 1.4% at morning and 7.6% in the afternoon), which was mainly emitted from biogenic sources and increased exponentially with temperature (Jiang et al., 2018). In addition, with the heavier impact from l biogenic source (10.03%) in JK (Fig.3), isoprene's contribution to Σ_{VOCs} value at JK at was 12% at 14:00 LT (Fig.7), much higher than those at three other sites (range from 4.02% to 7.92%)”.

Temporal variations:

Comment 20

L261: “wash-out” specifically refers to rain which I don’t think is what the authors mean.

Response:

The sentence has been revised as

“The occurrences of precipitation and raining were also frequent in most areas of China during summer, resulting in decreasing background level of air pollutants”

Comment 21

L276: “sharp changes in local emissions” - such as?

Response:

These could be resulted from the local emissions such as coal combustion and leakage of compressed natural gas (CNG) or LPG. We have explained this case in the following statements:

“At MEM, the distinctive increment was always accompanied by obvious increases of alkanes or aromatics (Fig. 9), combined with the simultaneous increased concentrations of SO₂, CO and NO_x. Illustrating the potential impact come from combustion sources, which may be caused by the nearby thermal power plant. At GS, the increase of Σ_{VOCs} in June was usually with extremely high levels of aromatics, due to the disturbance from solvent use for building renovation during this period, and the abnormal high levels of Σ_{VOCs} in other months were related to the rising concentrations of C3-C4 alkanes, which mainly originated from consumptions of compressed natural gas (CNG) or LPG (Huang et al., 2015b), indicating the possible impact from a gas-fueled power plants located about 1 km southwest of the site (about 18% was western wind at GS during May to

September) ”.

Comment 22

L278-9: It is not clear how changes in T and RH lead the authors to conclude combustion sources were enhanced.

Response:

Since the T and RH were often consistent under normal conditions, the contributions from gas evaporation should be also constant. Therefore, the simultaneous change of SO₂, CO and NO_x should be caused by the enhancement of combustion sources. In order to avoid confusion, we have rewritten the statement as:

“At MEM, the distinctive increment was always accompanied by obvious increases of alkanes or aromatics (Fig. 9). Since the T and RH were often consistent during the sampling period, the direct gas evaporations should be constant. Therefore, the simultaneous increased concentrations of SO₂, CO and NO_x could illustrate the potential impacts from combustion sources, such as emissions from nearby thermal power plant.”

Comment 23

It would be extremely helpful to have a more detailed map of Zhengzhou City showing the 4 sites, key emission sources and prevailing wind.

Response:

Suggestion taken. The newly figure 1 and 6 have been revised as below:

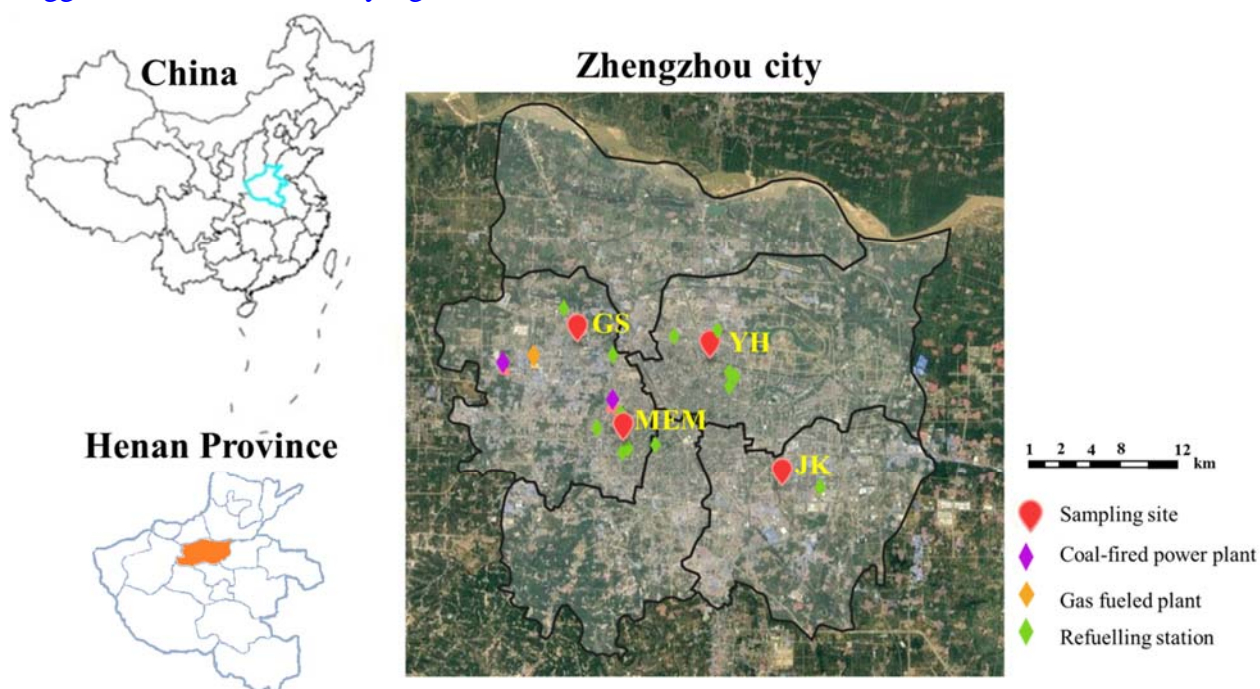


Fig 1. Map of Zhengzhou, China showing the locations of sampling sites.

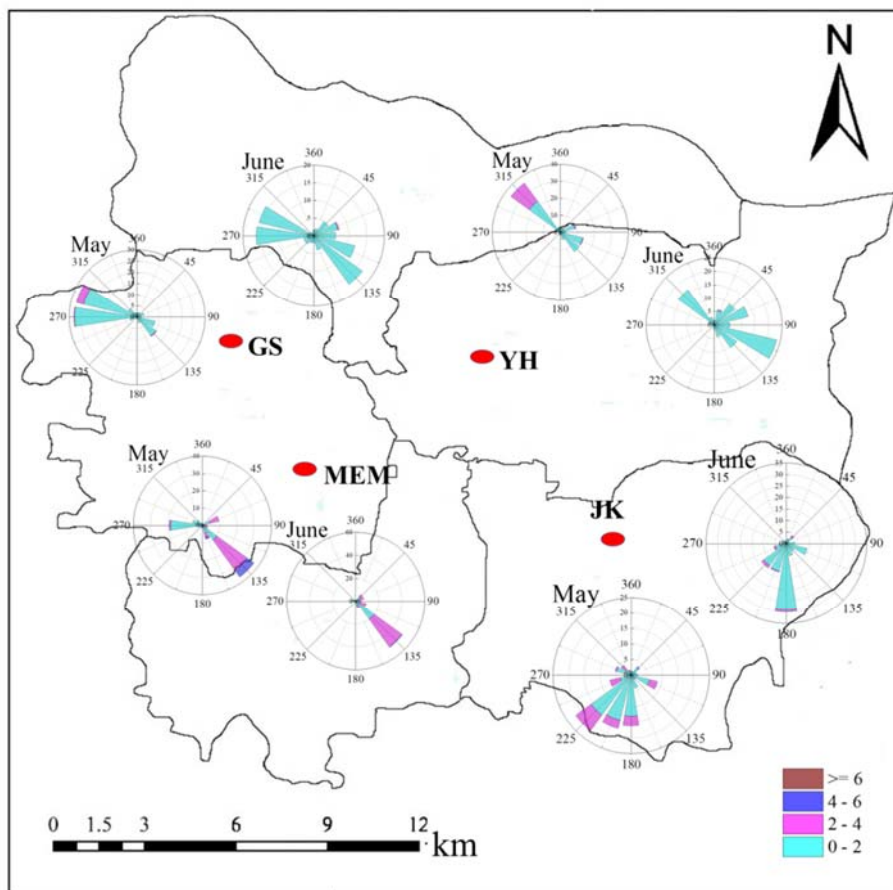


Fig. 6 Wind rose of each site in May and June (the wind distribution in other three months were illustrated in Fig S2)

Spatial variations:

Comment 24

L298-299: O₃-NO_x-VOC interactions and reactions are always highly complex and non-linear, hence the development of ozone isopleths (see e.g. Silman, 1999)

Response:

Thanks for the recommend reference. There is a particular section (i.e, section 3.4) for discussion on the ratio of VOCs/NO_x.

Comment 25

L300: Is this peak (i.e. hourly) O₃ or 8-hour O₃?

Response:

It is hourly average.

Comment 26

L306-307: This sentence appears to contradict the results presented in L301-302.

Response:

There is no contradiction. The original L306-307 just pointed out that a reversible observation on concentrations of O₃ and VOCs/OFP was seen at the two sites of MEM and JH only. There are many different factors controlling the ozone formation. The general trend is unchanged.

Comment 27

L321-323: “when synoptic conditions were favorable” - yet in the abstract and conclusions the authors state categorically that O₃ formation in Zhengzhou is VOC-sensitive. If there is a caveat it should also be made clear in these other sections.

Response:

The discussion on ozone formation regime was presented in the newly Section 3.4.

Comment 28

Please give typical T/B ratios for relevant sources.

Response:

We have provided the information as:

“T/B is an efficient tool to differentiate between pollution sources, both the tunnel studies and roadside researches indicated that T/B ratio varied within the range of 1 - 2 when the atmosphere was heavily impacted by vehicle emissions (Gentner et al., 2013; Tang et al., 2007; Huang et al., 2015b; Wang et al., 2002), and when the ratio was less than 0.6, it may be due to other sources, such as coal combustion and biomass burning (Tsai et al., 2003; Akagi et al., 2011). The industrial activity would become more important when the value of T/B ratio is higher than 3 (Zhang et al., 2015)”.

Comment 29

L334-336: Without knowing that the emission sources remained constant throughout the time it is not possible to state definitively that the differences were due to photochemistry.

Response:

Considering the variations of compositions of major VOCs groups (Fig. S6), the increases of alkyne and aromatics in September from June and July represent that the changes of emission sources had impacts on higher T/B ratios at September.

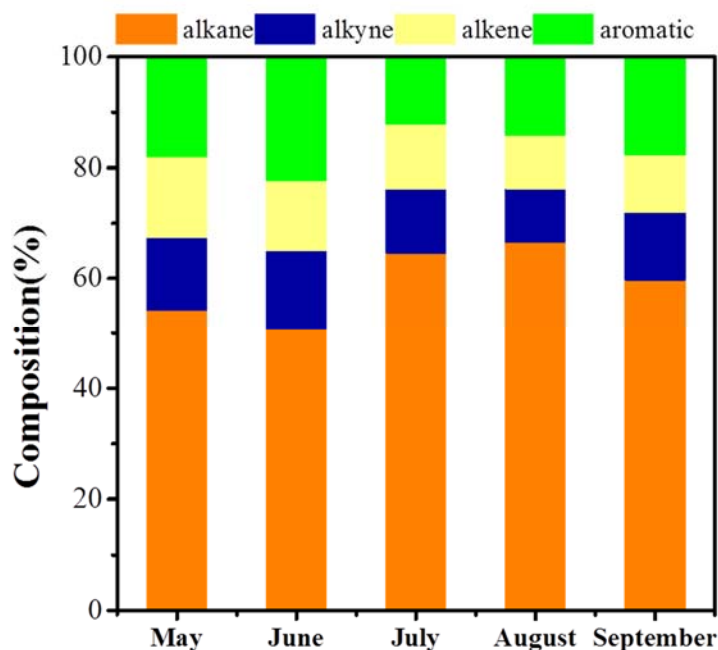


Fig S6. Temporal variation of compositions of major chemical groups

Therefore, the relative statements have been revised as:

“In addition, the atmospheric lifetimes of toluene and benzene are different, it was reported that when the $\bullet\text{OH}$ concentration was assumed to be 10^6 rad cm^{-3} (Monod et al., 2001), the lifetime for toluene and benzene was 1.9 days and 9.4 days, respectively, which maybe one of factors for the lower T/B ratios at 14:00 LT than those at 07:00 LT, and higher T/B ratios in September, due to more rapid consumption of toluene at 14:00 and the weaken photochemical reaction in Sept.”

Comment 30

L336-337: The authors make many statements such as this without attempting to explain why the observed difference may have occurred.

Response:

The incoherent statements have been deleted.

Comment 31

L340-342: The absolute values should be reported before the R2 value.

Response:

Thanks for the reminder. The absolute values have been reported.

Comment 32

L349: It would be possible to achieve these values with zero vehicle emissions but a mixture

of industrial and biomass burning emissions. Hence my previous comment regarding the superfluity of this speculation given the authors have conducted PMF for source-apportionment. But perhaps other pollutants monitored at the site also provide insight into most likely sources?

Response:

The statement has been revised as

“Except for September, the average T/B ratio at JK lied within the range of 1.47 - 2.72, testified the heavily impact form vehicle emissions sorted by PMF, where traffic related source accounted for more than 29% emissions (Fig.3).”

Comment 33

L355: How are the outliers (“abnormal values”) identified and removed?

Response:

The abnormal values defined as the data points did not distribute in the range of 5-95%, and thus were not taken into consideration in Section 3.5.

Comment 35

L360-361: As previously noted, a windrose plot would be extremely helpful.

Response:

They have been now presented in newly Fig. 6 and Fig. S2.

Comment 36

L360-361: This might be the prevailing wind, but what about the specific days sampled?

Response:

The prevailing wind directions were often consistent with the wind directions on our 10 sampling days each month.

Comment 37

i-pentane and n-pentane: Is this the ratio of i/n that the authors are reporting from these previous studies?

Response:

In newly L370-374, the ratios from other studies have been added, and the values observed in this study were presented in Fig.15.

Comment 38

I would suggest that the authors reverse the order and discuss i/n ratios first as they are NOT influenced by different reaction rates, and then discuss T/B ratios which are.

Response:

The relative statement has been moved to newly L368-389.

Reactive chemicals:**Comment 39**

L389-390: As noted previously, there are caveats associated with OFPs. It is not just the “reactivity” that matters when assessing the contribution of each individual species to overall O₃ formation. Different mixtures of VOCs result in competition between different species, leading to different relative yields, different reaction paths, etc.

Response:

The statements have been revised as:

“The reactivity of individual species was different, and the various mixtures of VOCs would result in competition between different species, leading to different reaction paths and relative ozone formation yields. Ozone formation potential (OFP) is a useful tool to estimate maximum O₃ productions of each compound under optimum conditions, from which the vital species in ozone formation could be identified.”

Comment 40

L392-395: The authors need to make it abundantly clear that this is an entirely hypothetical potential (or maximum) possible O₃ formation for each compound in isolation.

Response:

The statement has been revised as:

“Ozone formation potential (OFP) is a useful tool to estimate maximum O₃ productions of each compound under optimum conditions, from which the vital species in ozone formation could be identified. The calculation of OFP is based on mixing ratios and maximum incremental reactivity (MIR) of individual compound, which are expressed in Eq. (4).”

Comment 41

L396: The authors should present their own results first and then put them into context against previous studies. It's not clear why they would expect relative abundances and relative contributions to O₃ formation to be the same across different regions with different sources and different meteorological conditions.

Response:

The paragraph has been rewritten as,

“In Zhengzhou city, it was alkenes contribute most ($55.91 \pm 14.17\%$) to the sum of OFP, of which ethylene taken a large portion. This is different from the results estimated based on emission inventories by Wu and Xie (2017), where the largest contributor of total OFP in North China Plain (NCP), YRD and PRD was aromatics, hinting at the relatively lower surface coating industry in Zhengzhou”.

The estimation was not considered meteorological conditions, and the provincial emissions were calculated using equations:

$$E_t = \sum_{p=1}^N \left(\sum P_{m,n} \times VMT_{m,n} \times EF_{m,n} + \sum (1 - R_k) \times EF_{s,k} \times A_{s,k} \right) \times 10^{-12}$$

where E_t is the total VOC emission (Tg), $P_{m,n}$ is the vehicular population of category m with emission standard n in province p ($N = 31$, including all of the provinces, municipalities, and autonomous regions in mainland China), $VMT_{m,n}$ and $EF_{m,n}$ are the corresponding annual average mileage (km) and emission factor (g/km), respectively, R_k is the removal efficiency with technology k , and $EF_{s,k}$ and $A_{s,k}$ are the corresponding emission factors and activity data for source s (except on-road vehicles), respectively.

Comment 42

L401: “fraction” rather than “composition”?

Response:

Yes, it should be “fraction”. The statement has been revised as:

“the percentage of acetylene ($4.51 \pm 0.34\%$) weighted in OFP was higher than many other areas in China”

Comment 43

L410-411: Demonstrating the caution required in using and interpreting OFPs

Response:

Thanks for the reminder. The statement has been revised as,

“With the lower RH, higher T and OFP (88.13 ± 30.32 ppbv) values, the O_3 level at YH was unexpectedly lower than that at MEM on sunny days, keeping in mind that the OFP

was estimated with the assumption that reactions were proceed under optimum conditions, the above phenomenon reflected the unsatisfied ozone formation conditions at YH.”

Comment 44

L414: And/or increased the importance of local versus long-distance sources.

Response:

The statement has been revised as

“The total OFP was highest at JK in June, while the highest O_3 levels was observed at GS, during this period GS was downwind of other sites with lowest WS ($0.74 \pm 0.33 \text{ m s}^{-1}$), and the concentration level of O_3 usually increased with wind speed (Fig.S7), particularly when the eastern wind was dominant, illustrating the disturbance from long-distance sources in urban center.”.

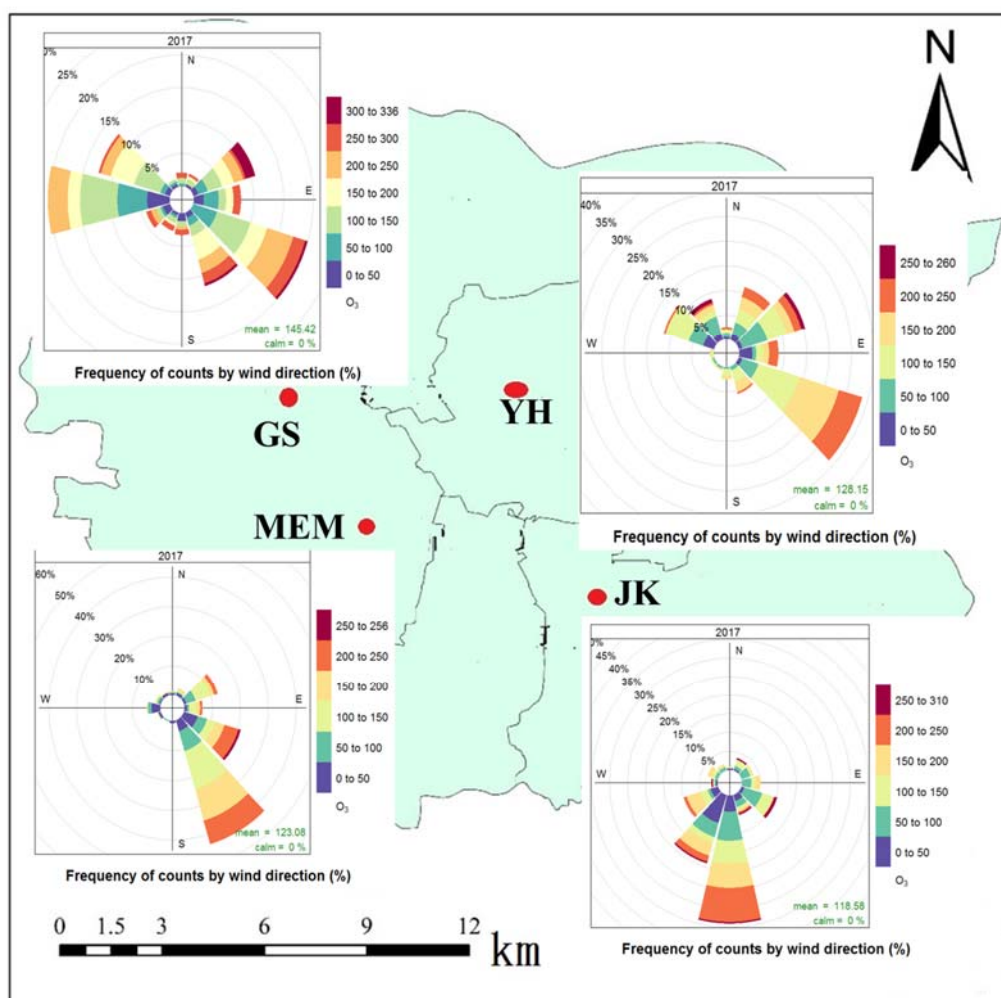


Fig.S7 Relationship among O_3 ($\mu\text{g m}^{-3}$), wind direction and wind speed (m s^{-1}) during sampling period in June, 2017

Source apportionment

Comment 45

It would be extremely useful to have a map showing (roughly) the key emission sources for these 7 or 8 factors near each of the 4 sampling sites.

Response:

They have been incorporated in newly Fig.1.

Comment 46

L420-425: These describe the methodology and should be included in Section 2.

Response:

Done.

Comment 47

L423-424: As suggested previously, I would move Table 6 to SI together with the detailed mathematical description of the method

Response:

Suggestion taken.

Comment 48

L424-425: Explain how the number of factors was determined.

Response:

The description has been revised as:

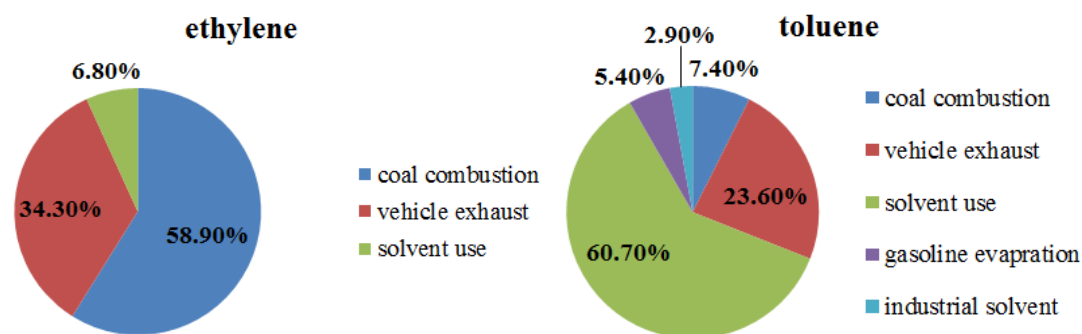
“Three to nine factors were selected to initiate running of PMF, the $Q/Q(\text{exp})$ for every site at fixed factor size were presented in tableS4. With the increase of factor number, the ratios $Q/Q(\text{exp})$ were declined due to additional factors. When the factor size changing from 3 to 4, 4 to 5, and 5 to 6, the decrement of $Q/Q(\text{exp})$ were larger (about 12-23%), while the change was lower than 10% after factors increased to 7, combined with the field conditions, seven factors were defined at MEM, YH, and GS, and eight factors were defined at JK (Fig.2).”

Comment 49

L427-428: Was it possible to identify specific alkanes or alkenes? Were there any clear differences between sites or times of day?

Response:

Yes, sources can be identified with specific compounds using all of samples collected in this study. The PMF results indicate that coal combustion (58.90%) and vehicle exhaust (34.30%) was the most important source of ethylene, while as to toluene, solvent use (60.7%) contributed most.



The differences among the four sites were described in the original L469-482:

The smallest contribution from CNG and gasoline evaporations was found at GS (24.99%), which was lower than the values observed at JK (35.06%) and YH (34.28%), and was probably caused by the heavier traffic at JK and larger consumption of CNG (from the condensed canteens and active household cooking) at YH. The distribution of petrochemical source was uneven on the regional scale, with the largest portion observed at MEM (11.56%) and comparable values found for JK (3.44%), YH (6.21%) and GS (5.56%). These results are in accordance with the fact the distance between the petrochemical plants and the sampling site of MEM was the shortest (ca. 2 km). The weighted percentages attributed to the solvent use were similar among the four sites, with the highest value of 12.41% at MEM, which was consistent with the fact that there was less abundance of aromatics in Zhengzhou city. The emission source related to diesel fuel was more evident at JK (11.76%). The area was rife with logistics companies and frequently suffered from disturbance of heavy-duty cargo vehicles powered by diesel fuel. Coal combustion impacted the ambient air heavily at MEM (17.84%) and GS (13.50%), which were only 2 km and 5 km away from the thermal power plants, respectively.”

Comment 50

I suggest the authors emphasize the differences between the identified factors more than they do. It is the trimethylbenzene that distinguishes the second from the first factor, but it is likely that ratios of e.g. toluene to xylene also differ

Similarly with source 3: it is the ratio of toluene:benzene and other aromatics that makes this distinct from the first factor and leads to the conclusion that one is gasoline and the other diesel.

Does source 4 also correlate with SO₂ which would strengthen the case that this is specifically coal burning rather than another fossil fuel?

Response:

We have attempted to re-calculate as suggestion. However, the correlation between SO₂ and factor 4 was weak.

Again, the fifth factor seems little different from the first three. Emphasise the unique markers of each.

Comment 50

Source 6 seems to exhibit the same compound mix as source 1. How do they differ? Is it that they have very different ratios of some of the compound classes?

Response:

Yes, the mixture was similar between these two factors, but the relative abundance was varied a lot. For factor 1, there was a high portion of C₂-C₄ alkanes, while ethylene and propylene were more dominant in factor 6. According to previous studies, we defined factor 1 as fuel evaporation and factor 6 as vehicle exhaust.

Comment 51

L465: Do the authors mean different vehicle types (e.g. hybrid, LPG, etc) or different styles of driving (e.g. more idling, lower speeds with increased braking, etc)?

Response:

It was pointed to different styles of driving.

Comment 52

It seems to me that Factor 8 is simply a sub-set of Factor 6. How are they distinguishable?

Response:

Yes, it is. We have thus combined it with factor 6 in discussion.

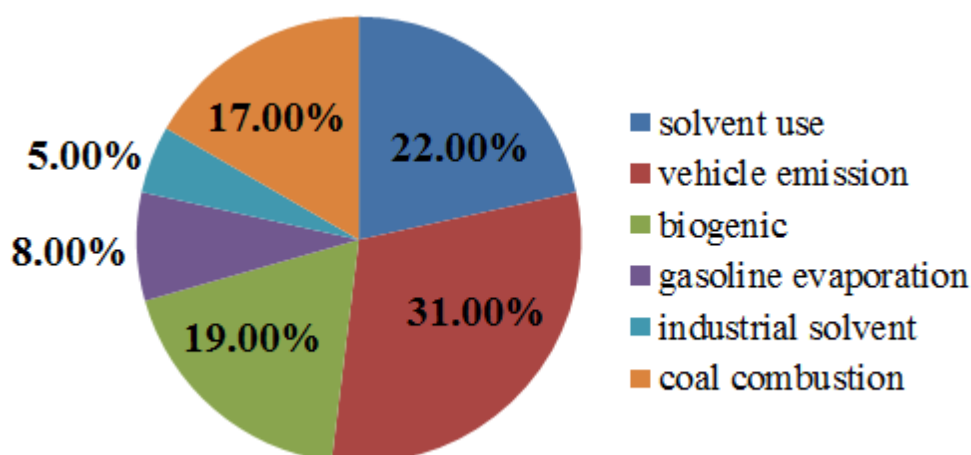
Comment 53

L482-484: It would be really nice if the authors now brought together the two quantified analyses they have conducted: source-apportionment and OFPs to identify the sectors that were most polluting at each site. Presumably here “important” refers to magnitude but would that also be the most important if considering OFP? Or toxicity?

Response:

The below figure presents the source apportionment result using OFPs obtained at the four sites. Vehicle emission was the most influential source (31%) in ozone formation, and solvent use (22%) ranked only second to vehicle emission with lower weighted percentages (9-13%

at the four sites) .



Long-range transport:

Comment 54

This entire section should be moved forward and presented ahead of all the sections describing possible local sources of precursor emissions. As O₃ is a secondary pollutant, regional and long-range transport is typically the greatest source.

Response:

This has been moved to Section 3.1.

Figures and Tables

Comment 55

Table 1: Suggest move to SI; not essential for main text. However, MIR should be included in the current Table 5.

Table 2: See comments regarding wind.

Tables 2-4: Should be combined into a site overview table with all met variables discussed within the text, average concentrations of all pollutants, specific VOC concentrations

Table 5: Would suggest to add % contribution to total VOC concentration as a neat comparison against % contribution to OFP and to put MIR in this table as it is used to calculate OFP

Fig. 1: I suggest the authors combine this with Fig. 10; air mass back trajectories are important for virtually all of the analysis presented in this study so should not be relegated to the final figure (and similarly should be included far earlier in the text than they are.

Fig. 2: It is not possible for any individual class of VOC to account for >100% of the composition of total VOCs. Either the authors should be using a stacked bar chart with each segment of the bar representing the different compound classes or a side-by-side bar chart as in the insert for isoprene. I would also suggest that isoprene should be included as part of the chart and not as an insert.

There is no obvious reason why the right-hand and left-hand panels should use different types of chart given they are showing the same thing.

The caption should be expanded to actually explain the figure; it is not just a title. For example, do the

bars for YH, GS, etc on the left-hand panel include both 07:00 and 14:00 data? And is that comparable with the data from other cities? The panel would be less cluttered if the authors listed the references as footnotes rather than on the chart itself.

Fig. 3: This figure is very poorly presented. The authors are attempting to fit too much data on each panel with too few different axes scales. Using a reverse scale on the secondary y-axis makes it almost impossible to assimilate the information and see correlations between the different variables. Using an axis ranging from 0 to 200 means that the T (in degC) is compressed to the point of masking any hour-to-hour fluctuations; CO (even scaled to ppm instead of ppb) has become a featureless red line. Why is SO₂ coloured in rather than just presented as a line?

The data requires splitting across additional panels, firstly helping to de-clutter and secondly allowing additional axes for clarity.

Again the caption should be expanded to be more descriptive. What is the significance of the two dates that are shaded? Are the tick marks corresponding to the dates shown at the bottom indicating 00:00 on that date? Or 12:00 (i.e. the middle of the day)?

Fig. 4: Why are the colours used for each class here (and in Fig. 3) different from those used in the left-hand panel of Fig.2 ?

Given that the left-hand panel has a continuous temporal scale on the x-axis would it make more sense to present all 07:00 data before all 14:00 data on right-hand panel rather than splitting by site?

Please include more information in the figure caption.

Fig. 5: Again, the authors are attempting to fit too many different variables on each panel, each with different scales, chart types, symbols and symbol colours. This requires splitting out into separate panels or charts.

Response:

Tables and figures have been revised according to the suggestion.