Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2018-36-AC2, 2018

Atmospheric Chemistry and Physics Discussions

Interactive comment on "VOCs emission profiles from rural cooking and heating in Guanzhong Plain, China and its potential effect on regional O_3 and SOA formation" *by* Jian Sun et al.

Jian Sun et al.

zxshen@mail.xjtu.edu.cn

Received and published: 1 May 2018

We greatly appreciate the reviewer's comments, which have helped us to improve the paper. The paper has been revised carefully according to the comments and suggestions.

Anonymous Referee #1 Received and published: 11 April 2018

This paper presents VOC emission factors (EFs) from 8 types of solid fuels and 3 kinds of stoves used in rural China, based on 27 samples. The study compares emissions from biofuels versus coal, and from semi-gasifier vs Heated Kang vs traditional stoves.

Printer-friendly version

Discussion paper

Semi-gasifier stoves were found to control PM emissions more effectively than VOC emissions. VOCs from solid fuel burning were estimated to contribute _20% to O3 levels in the region, and only weakly to SOA formation. While the study presents novel data on VOC emissions from solid fuel burning in rural China, it over-interprets results from a very small sample size (2 samples per fuel/stove combination) and needs a realistic discussion of uncertainty and the limitations of such a sparse dataset. Calculations are presented to as many as 5 significant figures, which is not credible. Basic information is still needed including the concentration data that the EFs are based on and how background was handled in the EF calculations. Some of the EF rankings and OFP statements don't seem right and should be checked. Unfortunately the English is still getting in the way of clarity and sometimes it's difficult to understand what is meant. Please find a way to correct the manuscript for language.

Response: We thank for the reviewer's comments and suggestions. The limitation and uncertainty level throughout the manuscript were discussed in section 3.5. We have also responded to each specific comment and made changes accordingly in the revised manuscript. The language has been polished by an English-speaking researcher.

SPECIFIC COMMENTS: L26: It isn't realistic to present EFs to 5 significant figures (3121.3 _ 1592.4) if the uncertainty is at the 1-2 significant figure level. Same comment on L144 and elsewhere. On L31 and L254 citing the OFP to 5 significant figures (5914.8) isn't credible because it implies it can be calculated to 0.002% – cite the uncertainty and use an appropriate number of significant figures.

Response: As the uncertainties ranged from 28.8% to 45.3 for concentrations, EFs, OFP and SOAP, up to 3 significant figures were used through the revised manuscript. The uncertainties about the calculations were discussed in the part named "Uncertainties" in revised manuscript.

L26: The abstract uses "biomass straw", L85 uses "maize straw" and "wheat straw", and the conclusions uses "biomass residues" (L371). Please define the terms. Does

residues = straws + corncob? If so then biomass straw > woody fuel isn't correct since corncob has the largest EF, followed by firewood/branches, maize straw, another branch, then wheat straw. So they're all mixed together and the different categories overlap in their uncertainty.

Response: To eliminate misunderstanding, all residue(s) were replaced by straw(s) in the revised manuscript. And the category of straws and/or biomass straws includes maize straw, wheat straw and corncob. The average VOC EFs of biomass straw were higher than those of woody fuels although the uncertainties overlapped. The description of this sequence has been rewritten in the revised manuscript.

Line 26-28: Emission factors (EFs) of targeted VOCs varied from 0.047 $\pm$ 0.019 to 3.12 $\pm$ 1.59 g kg$-$1 with a descending order of biomass straw > woody fuels » coal fuels, although the differences between straw and woody fuels were not significant (p> 0.05).

L85: Of these 8 fuels which are predominantly used in rural China? There are only 2 samples per fuel/stove combination, but studies of VOCs from biomass burning have shown that EFs can vary widely within a fuel type or combustion phase. I suspect this also occurs here within a given fuel/stove combination, but the sample size is too small to test this. I agree this is a pilot study, but this is a serious limitation.

Response: To our best knowledge, the eight types of fuels were commonly used and thus representative to rural Guanzhong region. The selections are based on the market rate. We admit that there is limitation in this study due to the limited sets of experiments conducted. Considered that rare data available on VOC emissions from biomass burning, this manuscript acts as a pilot study on the field of VOC emissions from residential solid fuel burning in this area. Our major focuses are on the difference of VOCs profiles among the fuel types and their impacts on ozone and SOA formations. In the subsequent studies, the massive sample number will be increased to avoid similar limitations.

L91: Please provide some basic information about the sampling and dilution system

here. How accurate and repeatable is the dilution? On L93 how precise was the before and after fuel weighing process? On L95 “the sampling period covered several circles” doesn’t make sense (maybe “cycles”)? If so what cycle? On L96 what do you mean by “Certain number”? On L100 do you mean you’re using a longer time for lower VOC concentrations? On L105 <5% doesn’t seem insignificant. On L107 what was the time interval between the two samples? Overall please quantify the uncertainty in this methodology and how the methodology has been validated.

Response: The detail description of the dilution system and sampling were provided in the revised manuscript. Accuracy and repeatability of the dilution system were also added.

The “circle” has been corrected to “cycle”. The cycle means the time intervals for adding fuels into the stoves.

On L96, the sentence has been rewritten and the number of channels was specified. On L100, the variation of VOCs sampling time was subject to the practical tests, i.e., 30 min for straws and woody fuels burning, and 60 min for anthracite and honey-comb burning.

On L105, the breakthrough test showed a <5% results and it was acceptable in experimental analysis.

On L107, the cooking work was easily repeated in the same day. While for heating samples, it usually needs another day to repeat the experiment because we need the same starting conditions. The source and level of uncertainty in methodology was discussed in section 3.5.

L95-100: The control of dilution rate was realized by a multi-pump system, and the real-time flow of each pinch point was measured by flowmeters (TSI 4140, TSI, MN, USA). The smoke inlet of the dilution system was set perpendicular to the smoke plume and 0.1 m above the chimney. In our tests, the PM weight differences between different

Printer-friendly version

Discussion paper

sampling channels were <5%, and the repeatability of dilution was >80% for field tests. L102-104: For cooking, fuels were weighed using a balance with a precision of 0.1 g before and after burning to clear the net fuel consumption, and the sampling period covered the whole cooking process. L106-107: Six parallel diluted smoke channels were setâĂŤone for VOCs, three for PM2.5, and two for online monitoring (CO and NOx).

L129: More analytical detail is needed. The reported MDLs for the 57 VOCs show a tremendous range (0.003-0.808 ppbv), and 0.808 ppbv is a strangely precise MDL. Which MDL goes with which compound? How many measurements fell below the MDL? The precision at 2 ppbv is <5% but we don't know what typical concentrations were. The reference to Ho et al. seems to be just for C2-C5 aliphatic compounds. Why does the text cite 57 target analytes but Table S2 lists 98 compounds? Which 57 were used?

Response: Table S2 showed the MDL of quantified VOCs in this study and their concentrations in each sample sets were shown in Table S4. To clarify, we can measure 108 VOCs with the absorbent tube method which include 57 ozone precursors and 51 toxic VOCs (i.e., halogenated compounds). And among these target VOCs, 98 of them were detectable and their background concentrations were shown in Table S3. The comparison of concentrations and MDL was discussed in uncertainties part. The total number of VOC quantified has been corrected in the revised manuscript. And the more detailed information on the targeted ozone precursors were referred to Ho et al., 2017 and Ho et al., 2018.

L138-140: The minimum detection limits (MDLs) for 98 target analytes were in the range of 0.001–0.159 ppbv with a sampling volume of 3 L; the MDLs for all VOC species are listed in Table S2. L144-145: Additional details on sampling and analytical methods have been published by Ho et al. (2017) and (2018).

L134: Please discuss the main sources of uncertainty in Eq. 1 and how the error

bars in the EF were calculated (L144). Do they include all sources of EF uncertainty? How are you accounting for background concentrations in the EF calculations? Did you measure them? Since EF calculations require the background to be subtracted off you need to describe how/where the background concentrations were measured and provide them in a statistics table.

Response: For L144, the error bars of EFs referred to the standard deviations. Uncertainties were discussed individually in the separate section. We have measured the background concentrations of VOCs. In EFs calculation, the sample concentrations were subtracted by the background. The related descriptions have been added in the revised manuscript. The background concentrations of target VOCs were shown in Table S3. The main sources of uncertainty in Eq. 1 were discussed in the revised manuscript.

L142-144: Background samples of VOCs were also collected and analyzed using the same protocol as that of source sample collection. The background concentrations of VOCs were subtracted off when calculating EFs and the data are listed in Table S3.

L142: The paper still needs a basic statistics table showing the concentration data (ppbv) for the 57 target analytes that the EF calculations are based on, as well as the background concentration and the MDL for each VOC. What was the variability between the two samples for each stove/fuel combination?

Response: The concentrations of VOCs for EFs calculation were shown in Table S4. As the calculation and presentation of data in this study were both weight-basis but not concentration basis. Therefore, the unit of VOCs concentrations in Table S4 was μg m-3 instead of ppbv. The background and MDL data were shown in Table S3 and S2, respectively. The average viabilities of VOC concentrations measured for each fuel/stove combination ranged from 24.2 to 69.4% (calculated as standard deviation / average value).

L145: Discuss the representativeness of the samples before different fuel types are

compared. How does variability within each fuel/stove combination compare to that between each fuel/stove combination? While recognizing the value of a pilot study, two samples per fuel/stove is not enough to meaningfully compare one fuel type to another. These limitations need to be discussed in order for the results to be meaningful.

Response: The representativeness of the combinations of fuels and stoves were discussed in Section 2.1. The combinations selected in this study could represent the main fuel consumption styles for cooking and space heating in the rural Guanzhong Plain. The variability within each fuel/stove combination was not huge in our study. In detail, the average viabilities of VOC concentrations measured for each fuel/stove combination ranged from 24.2% to 69.4% (calculated as standard deviation / average value). Compared to literatures, the viability of duplication experiments for PM2.5 (with their chemical components) and gaseous pollutants were typically over 100% (Chen et al., 2016; Li et al., 2009; Shen et al., 2015). The repeatability of field experiments depends on not only the accuracy of sampling and analyzing equipment but also the dramatical variations of combustion conditions which easily affected by multiple factors such as wind speed, igniting condition and temperature. Therefore, the variability of VOCs measured in each fuel/stove group were acceptable and representative in comparation with other groups although the sample numbers were limited. We are now preparing for a new sampling campaign in rural area in Shanxi Province to further detect the characteristics of VOCs emission from solid fuel burning and their influence on local atmosphere and even human health. The limitations were also discussed in section 3.5 in the revised manuscript.

Reference: Chen, Y., Shen, G., Liu, W., Du, W., Su, S., Duan, Y., Lin, N., Zhuo, S., Wang, X., Xing, B., Tao, S., 2016. Field measurement and estimate of gaseous and particle pollutant emissions from cooking and space heating processes in rural households, northern China. Atmospheric Environment 125, 265-271. Li, X., Wang, S., Duan, L., Hao, J., 2009. Characterization of non-methane hydrocarbons emitted from open burning of wheat straw and corn stover in China. Environmental Research Letters

4. Shen, G., Chen, Y., Xue, C., Lin, N., Huang, Y., Shen, H., Wang, Y., Li, T., Zhang, Y., Su, S., Huangfu, Y., Zhang, W., Chen, X., Liu, G., Liu, W., Wang, X., Wong, M.-H., Tao, S., 2015. Pollutant Emissions from Improved Coal- and Wood-Fuelled Cookstoves in Rural Households. Environmental Science & Technology 49, 6590-6598.

L168: What would explain the high EF contribution of i-pentane from many fuels? Why would i-pentane be higher than the shorter-chain alkanes? This is the opposite of what is seen in open fires. Does it make sense that dodecane and n-butane dominate for anthracite and honeycomb coals, and benzene for bitumite? On L190 Andreae and Merlet isn't an appropriate reference for a semi-gasifier. If the point is that benzene and toluene have high EFs from biofuels, then why doesn't ethene factor into the top 10 since its EF from biofuel is similar to benzene in Andreae and Merlet. On L212 why would woody fuels emit more propene than ethene? Please provide more in-depth discussion about the top 10 rankings as some of them don't make sense.

Response: We also noticed that the EF for iso-pentane was higher than those of shorter-chain alkanes. We have carefully validated the raw data and calculation. Even though iso-pentane was reported to one of markers derived from vehicle emission (Hwa et al., 2002; Liu et al., 2008), our experimental results demonstrate its large contribution from the examined fuels as well. In fact, there are potential differences due to the combustion conditions against open fires. On line 190, the reference of Andreae and Merlet was used for inter-comparison but it has been removed in the revised manuscript. For anthracite (honey-comb) and bitumite, benzene and higher-chain alkanes (i.e., C11 and C12) had high EFs, while dodecane and n-butane showed the highest contributions in both. Among these three fuels, benzene had a higher contribution in bitumite burning than anthracite (honey-comb). The possible explanation is that the volatile matter combustion phase of bitumite is prone to formation of aromatic hydrocarbons (Chen et al., 2016). Ethylene showed low EFs for all groups in this study and this phenomenon could be attributed to the variations of combustion conditions with other studies. In lines 216-220: Our data showed iso-pentane was listed in the top ten

species for profiles of most testing fuels including woody. Contamination from vehicle emission in such a large scale in combustion emission sampling was hardly possible. The formation mechanism of iso-pentane in fuels is thus valuable to be conducted in subsequent studies. Reference: Chen, Y., Shen, G., Liu, W., Du, W., Su, S., Duan, Y., Lin, N., Zhuo, S., Wang, X., Xing, B., Tao, S., 2016. Field measurement and estimate of gaseous and particle pollutant emissions from cooking and space heating processes in rural households, northern China. Atmospheric Environment 125, 265-271. Hwa, M.-Y., Hsieh, C.-C., Wu, T.-C., Chang, L.-F.W., 2002. Real-world vehicle emissions and VOCs profile in the Taipei tunnel located at Taiwan Taipei area. Atmospheric Environment 36, 1993-2002. Liu, Y., Shao, M., Fu, L., Lu, S., Zeng, L., Tang, D., 2008. Source profiles of volatile organic compounds (VOCs) measured in China: Part I. Atmospheric Environment 42, 6247-6260.

L200: The coefficient of divergence is being used to measure the similarity between profiles, but first need to discuss the uncertainties in the profiles and whether the profiles make sense.

Response: The average viabilities of VOC concentrations measured for each fuel/stove combination ranged from 24.2% to 69.4% which is acceptable for field experiments. The total uncertainty level of VOC profiles was calculated to be 53.1% including the main sources of uncertainties, more discussion about it was supplied in the section "Uncertainties". The results indicated that the profiles were reasonable.

L238: How do you know that "huge variations can be observed for the same fuels and stoves if different burning modes (heating or cooking) were applied"? There just aren't enough samples to quantify and understand variability in a statistical sense.

Response: This sentence has been rewritten in the revised manuscript which represents the results instead of statistical mean. More on statistical calculation have been discussed in the section named "Uncertainties".

L284-285: Notable differences (CD = 0.57) can be observed for the same fuels and

stove types (branch and traditional stove) with different burning modes (heating or cooking).

L252: Please show the VOC concentrations that the OFP calculations are based on and state the uncertainties and limitations for the OFP calculations. For example OFP assumes that O3 production is VOC-limited – does this apply to at your site in China? How well do the Carter MIR values apply at the study sites in China? How does the limited sample size impact the interpretation of OFP values?

Response: The VOCs concentrations for OFP calculation were listed in Table S4. The VOCs levels in Guanzhong area were satisfied with the condition of MIR calculation, and some previous studies in which the atmospheric concentrations of VOCs in urban and rural Guanzhong also indicated that the MIR method could be properly applied in this area (Li et al., 2017; Xue et al., 2017; Wang et al., 2012). And in Carter's study, the MIR calculation needs a high NOx scenario, that matches the atmospheric conditions in winter (Zhang et al., 2015). The applicability of MIR model was supplied in the revised manuscript. The limited samples would lead to relatively high uncertainties in OFP evaluation and the uncertainty part was discussed in an individual section.

L293-295ïijŽIn Carter's study, the MIR model required a VOC-limited and high NOx condition which applied to the Guanzhong area (Li et al., 2017; Xue et al., 2017; Zhang et al., 2015). Reference: Li, B., Ho, S.S.H., Xue, Y., Huang, Y., Wang, L., Cheng, Y., Dai, W., Zhong, H., Cao, J., Lee, S., 2017. Characterizations of volatile organic compounds (VOCs) from vehicular emissions at roadside environment: The first comprehensive study in Northwestern China. Atmospheric Environment 161, 1-12. Xue, Y., Ho, S.S.H., Huang, Y., Li, B., Wang, L., Dai, W., Cao, J., Lee, S., 2017. Source apportionment of VOCs and their impacts on surface ozone in an industry city of Baoji, Northwestern China. Scientific Reports 7, 9979. Zhang, Q., Shen, Z., Cao, J., Zhang, R., Zhang, L., Huang, R.J., Zheng, C., Wang, L., Liu, S., Xu, H., Zheng, C., Liu, P., 2015. Variations in PM2.5, TSP, BC, and trace gases (NO2, SO2, and O3) between haze and non-haze episodes in winter over Xi'an, China. Atmospheric Environment

112, 64-71. Wang, X., Shen, Z.X., Cao, J.J., Zhang, L.M., Liu, L., Li, J.J., Liu, S.X., Sun, Y.F., 2012. Characteristics of surface ozone at an urban site of Xi'an in Northwest China. Journal of Environmental Monitoring. 14, 3000-3008.

L289: Table S5 needs uncertainties and realistic significant figures. A 516.2 m boundary layer height (20 cm precision) is not realistic. What time of day and season is this for? The uncertainties in the emission rate calculation will be large and need to be quantified, including all uncertainties leading to the 20% contribution (L292). On L304 how do you know the largest source of uncertainty in the box model was fuel consumption? What about other parameters?

Response: The average height of boundary layer was 516.2 m in winter of 2013 (Niu et al., 2017), and the precision was 1 m. The uncertainties in the box model calculation were calculated and discussed, including the ones from fuel consumption, EFs calculation, OFP calculation and other possible uncertainties. ReferenceïijŽ Niu, X.Y., Cao, J.J., Shen, Z.X., Ho, S.S.H, Tie, X.X., Zhao, S.Y., Xu, H.M., Zhang, T., Huang R.J. PM2.5 from the Guanzhong Plain: Chemical composition and implications for emission reductions. Atmospheric Environment, 2016, 147: 458-469.

L323: What are the uncertainties with using the chamber test results in this evaluation? How were the uncertainties in Figure 2 calculated and how well do they capture the overall uncertainty? Throughout the paper, each time an uncertainty is cited please state what it is (standard deviation? standard error?) and how it was calculated.

Response: For all the data presented in this study with the form of "A$\pm$B", the B value were standard deviation (SD) and calculated as below: SD=$\sqrt{}$(1/N*$\sum(xi-\mu)$ˆ2) Where N is the sample number in group, xi is the sample value in the group, μ is the average of x1 - xN. The data processing part has been supplied in the revised manuscript.

And the uncertainties throughout the paper were discussed in section 3.5.

L157-164: 2) Average and standard deviation calculation For EFs, O3 formation poten-

tial (OFP), SOA formation potential (SOAP), and other data are presented with error bars as average and standard deviation (SD) values. The average and SD were calculated as Average (μ) = ()/N, (2) where Xi is the value of sample No. i and N the total sample number; and SD=$\sqrt{}$(1/N*$\sum(xi-\mu)$ˆ2), (3) where Xi is the value of sample No. i, N the total sample number, and μ the average of Xi.

L336: It's not correct to say alkenes are inactive in photo-oxidation reactions, and it doesn't make sense that alkanes are considered to be more reactive than alkenes. L345: Quantify "much lower". Though the average for coal may be lower than for biomass fuels, the range in SOAPs is so large that the highest SOAP for coal (364.6 _ 99.2) is more than the lowest SOAP for biomass fuels (242.6 _ 32.2). These calculations are still based on very few data.

Response: Yes, we corrected these in revised manuscript. On L336, the explanations of the low contribution of SOAP from alkenes and OVOCs were rewritten in the revised manuscript. For L345, the much lower has been quantified and the sentence has been rewritten in the revised manuscript to make it reasonable.

L371-373ïijŽSince alkenes and oxygenated VOCs (grouped as "Others") generally have lower toluene-equivalence index than aromatic hydrocarbons and alkanes (especially long-chain ones), their contributions to SOAP were low.

L348: Clarify "not so significant". Was it significant or not? How was the uncertainty for the overall biomass fuel average calculated? How about for the bitumite average?

Response: The difference was not significant. The statement has been corrected in the revised manuscript. The standard deviations were calculated for biomass and coal fuels separately. For the uncertainties, they were discussed in section 3.5.

L389-391: Specifically, the lowest SOAP of 0.011 $\pm$ 0.004 g kg$-$1 was observed for honeycomb-CS, whereas bitumite-SG had the highest SOAP of 0.365 $\pm$ 0.099 g kg$-$1, even lower than the average value for the biomass burning although the difference was

not significant ($p > 0.05$).

L356: It's unrealistic to present these emission rates to 5 significant figures (to 10 g/day). This calculation is based on extremely few data. Quantify and discuss each uncertainty in this calculation, for example using bitumite as a surrogate for all coal fuels, etc. What is the range of emission rates that can be calculated? What is the uncertainty on the estimate of 0.23%?

Response: The daily SOAP emission was calculated to be two orders of magnitude lower than OFP emission, thus 3 significant figures were used for SOAP in revised appendix and manuscript. The uncertainty was calculated and discussed in the section named "Uncertainties".

L381-383: We don't know when this study occurred. Does your statement that controlling VOCs could decrease O3 levels apply in all seasons? Does the 20% contribution apply in all seasons?

Response: The main sampling work was conducted in winter 2014 and it was supplied in the revised manuscript. And in the conclusion and abstract, we concluded that the 20% contribution from VOCs to O3 and 0.23% to SOA in Guanzhong was in residential heating season in winter. And the emission reduction by semi-gasifier and clean fuel technologies was limited to winter and in Guanzhong area.

L75-77ïijŽVOC samples were collected at three typical agricultural villages in the rural Guanzhong Plain (Fig. S1) in winter 2014. The sampling campaign lasted for 3 weeks; each week was devoted to one sampling site.

MINOR COMMENTS: L35: The reader doesn't know when the study was conducted, so "in 2013" lacks context here.

Response: "In 2013" was deleted. Because in the context, the OFP and SOAP discussed were all referred to this study, and the time of this study conducted was shown in methodology part.

L43: 60% of households in China or globally? What do you consider to be a “traditional” fuel?

Response: The estimation of 60% was in rural China and it was corrected in the revised manuscript. The traditional biomass fuels refer to the raw ones without being processed, such as raw straws of wheat and maize, and forestry waste i.e. branches and wood logs. By contrast, the modern biomass fuels were the ones being deep processed, i.e. straw pellets, straw briquettes, biomass charcoal, and even biogas.

L44-45ïijŽOver 60% of households in rural China adopt traditional biomass resources as major fuels (Hou et al., 2017).

L50-52: What VOC source does the “relatively rare” statement refer to? The reference is for prescribed burning in Georgia. I thought you meant VOCs from residential burning of solid fuel? If so please cite all existing studies for this, so the reader can see how your study fits into the existing literature. Response: Yes, we mean VOCs from residential burning of solid fuel as the reviewer’s mentioned. Study of Lee et al (2005) cited here was to state the adverse effects of VOCs from combustion sources were comparable to those from PM. The existing studies about VOCs emissions from residential solid fuel burning were cited in the revised manuscript.

L52-54: Studies on trace gases, including VOCs, have been relatively rare (Chagger et al., 1999; Chen et al., 2016; Evtyugina et al., 2014), even though their effects on atmospheric pollution are comparable to that of PM (Lee et al., 2005).

L66: Eight fuel types and three stoves is a good start, but 27 viable samples is not a “comprehensive measurement campaign”.

Response: In fact, not only VOCs samplings but also PM2.5, size-segregated PM and some gaseous pollutants (i.e. CO, NOx and SO2) were sampled and measured during the field sampling. Therefore, it could be a comprehensive one. In this study, as only VOCs data were used, we corrected this sentence in the revised manuscript.

L69-72: In this paper, we particularly focus on the VOC emissions from solid fuel burning in the Guanzhong Plain. A measurement campaign was conducted in its rural environments to determine the EFs of VOCs from rural solid fuel burning activities and their contributions to regional O3 and SOA formation.

L71: Please state when the field study occurred (what year and season, over how many days).

Response: The information was provided in the revised manuscript.

L75-77: VOC samples were collected at three typical agricultural villages in the rural Guanzhong Plain (Fig. S1) in winter 2014. The sampling campaign lasted for 3 weeks; each week was devoted to one sampling site.

L72: "village in Weinan city" doesn't make sense: do you mean a village near Weinan city? Same comment on L74 and L75.

Response: Yes, we agreed with the reviewer's opinion, and we corrected these in revised manuscript.

L81: "simple ion structure" doesn't make sense here. Maybe "iron"?

Response: It has been corrected in the revised manuscript.

Line85-86: Traditional coal heating stoves have simple iron structures and are commonly used heating devices in Northern China.

L143 and elsewhere: Use "to" rather than "-" to show a range so it doesn't look like a minus sign.

Response: Corrected.

L176: What temperatures do you mean by "combustion temperatures"? Please quantify.

Response: The temperature here is not specified. The sentence means that the in-

creasing of combustion temperature could lead to higher EFs of aromatic hydrocarbons. And it has been rewritten in the revised manuscript.

L211-212: The generation of aromatic hydrocarbons demonstrated a positive correlation with increases in combustion temperatures (Shen et al., 2015).

L186 and throughout: It isn't realistic to cite EFs to 0.1% based on two samples.

Response: We admit that there is limitation in data processing due to the limited sample number. While if 1% precision was adopted, the orders of contributor should be the same and hard to discuss. Discussion of uncertainties on VOC profiles were conducted to make it realistic.

L193: Are any of the small variations statistically significant?

Response: Main alkanes contributed 29.4% in anthracite burning emitted VOCs while this proportion was 37.6% in bitumite burning emitted VOCs. Considering the uncertainty level of profiles (28.8%), the variation was not significant.

L219: Can VM really be known to 0.01%?

Response: Proximate analysis was conducted by the Analytical Center of Chinese Academy of Guangzhou Institute of Energy Conversion, CAS, and the results we got were with precision of 0.01%. And this precision for proximate analysis is general in previous studies (Chen et al., 2016; Chen et al., 2015; Ni et al., 2015). ReferenceïijŽ Chen, Y.J., Tian, C.G., Feng, Y.L., Zhi, G.R., Li, J., Zhang, G., 2015. Measurements of emission factors of PM2.5, OC, EC, and BC for household stoves of coal combustion in China. Atmospheric Environment 109, 190-196. Ni, H., Han, Y., Cao, J., Chen, L.-W.A., Tian, J., Wang, X., Chow, J.C., Watson, J.G., Wang, Q., Wang, P., 2015. Emission characteristics of carbonaceous particles and trace gases from open burning of crop residues in China. Atmospheric Environment 123, 399-406. Chen, Y., Shen, G., Liu, W., Du, W., Su, S., Duan, Y., Lin, N., Zhuo, S., Wang, X., Xing, B., Tao, S., 2016. Field measurement and estimate of gaseous and particle pollutant emissions

from cooking and space heating processes in rural households, northern China. Atmospheric Environment 125, 265-271.

L234: Why would the same stove be more likely to show similar emissions despite different fuels? What would explain this?

Response: The hypothesis is that the structure may limit the combustion temperature and oxygen supply, thus the products could be similar to some extent in the same stove. As this hypothesis needs more evidence in our future studies, we did not put it as an explanation in the manuscript.

L249-250 and 262: “k” not “K”.

Response: Corrected.

L257: This sentence doesn’t make sense. It starts with alkanes but finished with alkenes.

Response: This sentence has been rewritten in the revised manuscript.

Line306-308: Alkanes showed relatively low contributions to the total OFP for most fuels except anthracite because of their relatively low MIRs (Duan et al., 2008), even though the concentrations of iso-pentane and dodecane were high.

L262: What do you mean by “relatively high”? Alkane reactivity is low compared to say alkenes.

Response: This sentence was to express that long-chain alkanes had higher reactivity than short-chain ones, thus the VOCs sources with high long-chain alkanes proportions might be over-estimated in OFP calculation by Prop-Equiv method. It has been rewritten in the revised manuscript.

Line 311-314: Even though long-chain alkanes have relatively high kOH values (Carter, 2012), their reactivity toward O3 formation through photochemical reactions is comparatively nonsignificant. As a result, the Prop-Equiv method might reasonably over-

estimate the contributions of alkanes to O3 formation, especially for the VOC sources with high percentages of long-chain alkanes.

L306-307: Not sure what is meant by this sentence.

Response: This paragraph has been deleted and the uncertainties throughout the paper have been discussed in section 3.5.

Please add a distance scale to Figure S1 (lower panel).

Response: Corrected.

Table 1: Please state the units for EF.

Response: Corrected.

Table S2: Typo: bitumate.

Response: Corrected.

Table S3: “Two sampling sites” or “two samples”? I thought it was two samples at the same site.

Response: Corrected.

Figure S3: The colors don’t show up, a lot of it looks like black lines. Having numbers (referenced to Table S1) rather than VOCs on the x-axis makes it very difficult to interpret. Typo: abthracite.

Response: Color of bolder line has been changed and the fill color of columns need to be shown in the proper scale. Figure S3 was aided for Table 2 and Table S5, and if the x-axis was name of VOC species, it could be hard to show them in a limited chart. The typo has been corrected.

TECHNICAL CORRECTIONS: The paper still needs careful proofing for English (too many errors to list). Examples from the abstract: L1: Title: “VOCs” should be “VOC” L21: Change “burning” to “burned” L23, 31 and 37: Change “in Guanzhong” to “in

the Guanzhong" L24: Change "using adsorbent" to "using an adsorbent" L27: Change "that semi-gasifier" to "that semi-gasifier stoves" L27 and 36: Change "VOCs" to "VOC" L29: Change "variations on" to "variations in" L33: Change "SOAP" to "SOAPs" L34: Change "OFP" to "OFP values" L36: Change "impact to" to "impact on" L40: "occurred" should be deleted L54: "towards with" should be "along with" L55: Should be "leads to" and "air pollution" L56: "With discrepancies" isn't the right wording L57: "valuable" or "variable"? L62: "dependency level" is unclear L63: No need to repeat the Hou et al. reference And so forth through the manuscript.

Response: All errors listed and the similar ones throughout the paper have been corrected. And this revised manuscript has been proofed by an English-speaking researcher.

Please also note the supplement to this comment:
https://www.atmos-chem-phys-discuss.net/acp-2018-36/acp-2018-36-AC2-supplement.zip

---

Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-36, 2018.