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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.

Anonymous Referee #1

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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. 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

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Discussion paper

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.

SPECIFIC COMMENTS:

L26: It isn't realistic to present EFs to 5 significant figures (3121.3 $\pm$ 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.

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.

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.

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.

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?

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.

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?

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.

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.

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.

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.

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?

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?

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.

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 $\pm$ 99.2) is more than the lowest SOAP for biomass fuels (242.6 $\pm$ 32.2). These calculations are still based on very few data.

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?

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%?

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?

MINOR COMMENTS:

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

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

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.

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

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

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

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

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

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

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

L193: Are any of the small variations statistically significant?

L219: Can VM really be known to 0.01%?

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

L249-250 and 262: "k" not "K".

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

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

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

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

Table 1: Please state the units for EF.

Table S2: Typo: bitumate.

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

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.

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.

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