Review of egusphere-2026-4493

Title: Environmental impacts of polarity-resolved water-soluble organic matter from animal dung burning on the Qinghai-Tibet Plateau

Authors: Huang et al.

Summary

The authors burn yak and sheep dung in a household stove in one county of the Qinghai-Tibet Plateau, separate the water-soluble organic matter of the $PM_{2.5}$ into three fractions, and measure emission factors, light absorption, functional groups and oxidative potential. These are scaled up to county-level inventories of emissions, radiative forcing and OP for the whole plateau, from which recommendations on fuel policy are drawn.

The measurements may be of some value, and I should have liked to be more encouraging. They cannot, however, carry the regional conclusions that are the manuscript's principal claims. Three difficulties seem to me decisive. First, the regional "radiative forcing" is obtained by applying a forcing efficiency to annual emissions, with no account of atmospheric burden, lifetime or transport; it is not radiative forcing in any accepted sense. Second, the forcing efficiency of total WSOM, and with it the comparison with black carbon, is obtained by adding mass-specific efficiencies that should have been weighted by mass. Third, emission factors from a single stove are applied to the entire plateau without evidence that they are representative, and the very large uncertainties reported are then set aside. The recommendation to replace sheep dung with yak dung rests on these estimates, and on fuel differences whose significance has not been established.

Putting this right would mean rebuilding the quantitative analysis rather than revising the text. I therefore cannot recommend publication in ACP.

Major comments

1. The regional "radiative forcing" (Figure 3a, Figure S2) appears to be the SFE of each fraction multiplied by its annual emission (L144–148). Forcing depends on the mass present in the atmosphere, and hence on lifetime, transport and deposition, none of which enters the calculation; if I have misread the procedure, the authors should set it out. The reported magnitudes raise an additional concern. Since 1 MW km^{-2} is equivalent to 1 W m^{-2}, the highest class in Figure 3a (approximately 2600 MW km^{-2}) corresponds to an exceptionally large radiative flux perturbation, exceeding even the solar constant. This requires a clear explanation of the physical quantity and reference conditions. Nor are the reductions claimed for the substitution scenario (L364–365) readily reconcilable with the mapped values; were densities summed as though they were extensive quantities? Until the quantity, its time basis and its assumptions are defined, the statements on regional warming, the cryosphere and fuel substitution cannot stand.
2. The WSOM SFE of 61.0–193.4 W g^{-1} (L334–336) is the arithmetic sum of the three fractions' per-gram values in Table 1 (e.g. 21.8 + 7.8 + 31.3 = 60.9). Mass-specific efficiencies must be mass-weighted, $\Sigma\, m_i\, SFE_i / \Sigma\, m_i$; the authors should give the fraction

masses and recalculate. Even a correct SFE would not support the comparison with BC (L28–29, L336, L402–403), for a ratio of per-gram efficiencies says nothing about forcing without the atmospheric burdens; that claim should be withdrawn. The SFE calculation retains only absorption. While this may provide an absorption-only estimate, it cannot establish the magnitude or sign of the net radiative effect without accounting for scattering. Furthermore, the Mie calculations lack reported particle size distributions and refractive indices, and the use of solution-phase MAE in particle radiative calculations is not adequately justified.

3. Emission factors from one stove in one county, with single values of r and φ from Hu et al. (2016), are applied to the whole plateau without evidence of representativeness. This introduces an unquantified uncertainty in extrapolating the measured emission factors to the entire plateau, which cannot be captured by Monte Carlo simulations unless the relevant spatial variability is characterized and included. Even the propagated variability gives about −55% to +320% (Table S1), yet the inventory is called "reasonably robust" (L311–313) on the strength of the activity data alone. Please separate measurement uncertainty from representativeness, identify the dominant parameters and compare with independent estimates, or withdraw the word.

4. The recommendation to replace sheep dung with yak dung (L361–368, L406–411) rests on the estimates above and on fuel differences that Section 3.1 never compares. The number of burns is not stated, standard deviations often equal the means (e.g. 0.35 ± 0.36 g kg^{-1}; 21.8 ± 21.5 W g^{-1} in Table 1), and no significance test is offered. Please report all quantities by fraction, fuel and scenario with n, and test the differences. Moreover, DCFH-based OP is an index of redox activity, not of exposure or health outcome; a map of OP is not a map of health risk.

5. The fractions share the same activity data and differ only in their emission factors, so their maps and Gini coefficients (0.603–0.615) are nearly identical; what does the fractionation reveal that bulk WSOM would not? HP-WSOM, collected as the HLB effluent, may contain poorly retained inorganic ions, including constituents introduced during pH adjustment (Text S1). Unless its organic content was quantified separately, inorganic material may affect the reported fraction mass and derived mass-specific properties. Please report the quantification, recoveries and mass closure, and define "WSOM-C". Nor can FT-IR "identify" nitroaromatics or quinones (L392–394). The attribution of its elevated OP to transition metals (L195–203) is also unsupported by metal measurements.

6. The WSOM share of $PM_{2.5}$ or organic carbon is never given, and water-insoluble organic matter, reported elsewhere to dominate BrC absorption (Huang et al., 2020, listed but apparently uncited), is ignored. The authors should quantify the WSOM share and discuss WIOM, or confine their conclusions to WSOM.

7. Much cannot be checked. Table S2 contains no Fen-Wei data (L325–327, L347–349), and the totals, percentages and county rankings in Sections 3.2–3.4 cannot be recovered from Figures 2 and 3. The values should be tabulated.

Specific comments

Introduction

- **General:** Qualitative terms stand where numbers are wanted, e.g. "large quantities" and "a major source" (L47–48), "a major fraction" (L62), "a substantial fraction" (L65). Many statements also lack references, e.g. L54–58, L59–60, L71–72, L78–84. These are examples from the Introduction only; the same applies throughout.
- **L74–76:** The HULIS-n / HULIS-a / HP-WSOM scheme is attributed to Mukherjee et al. (2020); it originates, as far as I am aware, with Chen et al. (2016a, b), and credit ought to go there.
- **L77–78:** Hygroscopicity is offered as a distinguishing property but never mentioned again.
- **L92–93:** Polarity-resolved WSOM from biomass and coal combustion has already been reported by the same group (Huang et al., 2022; Zhang et al., 2022c). What is new here beyond the choice of fuel?

Methods and sampling

- **Study area:** The Results single out counties (Gonghe, Huangyuan, Huangzhong, Medog, Mangya, Minhe, Tongde) that few readers will know. Please describe the study area: the number of counties, their location and relevant characteristics, and the position of the sampling site (Gangcha).
- **L108:** Is this a field observation, or a controlled combustion experiment that happens to have been carried out in the field? The two are rather different things.
- **L111–115:** The heating and cooking scenarios are the central variable, yet the reader is referred elsewhere for what distinguishes them. A brief description (air supply, burn rate, temperature, duration) belongs here.
- **L114:** "Three fuels": yak dung, sheep dung and …? I eventually deduced bituminous coal, whose results appear only in Table S2 and are not discussed. Either discuss them or remove them.
- **L115–117:** How many independent burns and samples for each fuel and condition, when, and with what PM mass?
- **L118–119, L133–134:** Brief accounts of the extraction and FT-IR procedures belong in the main text, not solely in Text S1 and earlier papers.
- **L137:** "A certain volume" will not do. Please also state how OP was normalised: per gram of what, and measured how?
- **L159–160:** Why 31.0% and 51.2%? How does Hu et al. (2016) relate to this study, and why may single values be applied to every county? Were their uncertainties included in the Monte Carlo simulation?
- **L161–167:** The inputs are said to be normally distributed, yet the ranges in Table S1 are strongly asymmetric, and with standard deviations as large as the means a normal distribution yields negative emission factors. Please state the distributions used, how

negative draws were treated, and whether the parameters were independent. A reference for Crystal Ball is also needed.

Notation and definitions

- **L24, L93 and passim:** The text drifts between "WSOM" and the undefined "WSOM-C" (major comment 5c). I infer that "-C" means carbon; but in Section 2.1 and Eqs. (1)–(2) "C" means cooking and, for good measure, the non-heating season.
- **L144:** "EF" is never defined.
- **L145, L250, L333:** SFE200-700 in the text, but 280–700 nm in Table 1, Text S2 and Figure 3. Which is it?
- **L150–160:** In Eq. (2) the capital "C" (non-heating season) is easily confused with the lower-case "c" (cooking); by L157 "C" serves for both. In Eq. (1) the subscript "l" is presumably "k". Please list every symbol with its units, and state how many counties and pollutant types are included.
- **L163:** What, precisely, are the "activity data", and from which yearbooks and years?

Results: figures, tables and data

- **L214–219, L231–235, L266–269:** I could not obtain the quoted totals, densities and percentages from the figures. Does Figure 2 show the heating season only, and do the values refer to carbon? How were the Gangcha measurements applied to each county?
- **L220, L237, L265, L375–384:** Please mark the counties named on the maps. References for the GDP data are needed.
- **L323, L326:** "1.2 and 4.5 times higher" and "2.1–14.3 and 27.3–169.7 times higher" than what? Please give the original values.
- **L327:** Where is the Fen-Wei Plain, what does "biomass and bituminous coal combustion" refer to, and where are the data and references?
- **Figure 1:** The right-hand axis of (a) reads "Concentration (g/m^3)" with values up to 5; mg or μg is presumably meant. Concentration of what, and where? The EF of babs-365 is in $m^2\ g^{-1}$ at L176 but $m^2\ kg^{-1}$ in (b). Several error bars exceed the bars themselves, and one runs off the panel. The three-dimensional pie charts show nothing that a stacked bar would not show better.
- **Figure 2:** The caption says "concentrations", but the legends are in kg and kg km^{-2}; these are emissions. What distinguishes (a) from (b), and which season is shown? County totals, as in (a), reflect the size of a county as much as anything else. A single colour scale leaves the HP-WSOM panels almost blank.
- **Figures 2 and 3:** The maps do not convey the results: there are no county names, no marker for the sampling site, and neither scale bar nor coordinates. Fewer and larger maps, with the key counties labelled and a table or ranked bar chart of county values, would serve better.
- **Figure 3:** The OP unit, AU $s^{-1}\ g^{-1}\ km^{-2}$, is a per-gram quantity divided by an area; what does it represent? The legend of (a) is barely legible, and the chord diagrams (c1, c2) carry no units. A stacked bar chart would be clearer.

- **Figure 4 and L369–384:** Neither axis is labelled, and "G" and the dashed line are unexplained; "Lorentz" (L387) should read "Lorenz". More substantively, please specify how the counties were ranked, what is accumulated on each axis, and whether any weighting was applied. Without this, the relation between the curves, the Gini coefficients and the conclusion on economic inequality cannot be evaluated; that conclusion appears to rest on two selected counties.

Interpretation and wording

- **L183–191:** The band at 1634 cm^{-1} overlaps aromatic C=C and the O–H bend of water; 1716 cm^{-1} is characteristic of carbonyl and carboxylic C=O rather than quinones; 1053–1220 cm^{-1} and 1400 cm^{-1} overlap sulfate and ammonium.
- **L194–195:** Figure S1c is cited for the 1716 cm^{-1} band, which appears to be labelled in panel (b). Please check.
- **L217–218, L222–223:** Why should HULIS-a rise 6.1-fold in the heating season but HULIS-n only 1.5-fold? How does sheep dung burnt for cooking supply 17.8% of the emissions but 64.7% of the forcing (L268) and 66.6% of the OP (L292)? An explanation is wanted, not merely the numbers.
- **L238–240:** References are needed for the grassland coverage and the share of sheep dung.
- **L249–259:** That the values differ is not in itself a finding. What are we meant to learn from them?
- **L293–294, L299–302:** The health impacts of WSOM are said to be "dominated" by sheep dung burnt for cooking. OP is not a health impact (major comment 4), and WSOM has other sources. Please withdraw or tone down considerably.
- **L306–311:** Please provide data or a reference for the reliability claimed for the activity data.
- **L325–332:** Attributing the difference from the Fen-Wei Plain to altitude conflates differences in fuel, stove, operation and laboratory. Without the same fuel and stove at low altitude, the inference does not follow.

References

- **General:** The list should be brought into line with ACP style. Journal names are given in full in some entries (e.g. L436–447) and abbreviated in others (e.g. L448–450); page or article numbers are missing from several (e.g. L436–447); titles are in title case in some entries and sentence case in others.
- **Errors:** Charrier and Anastasio (2012a, b) are the same paper listed twice (L460–465). Afsana et al. is dated 2022 but carries what appears to be a 2023 DOI (L440–443). Author names are corrupted at L499 and L649.
- **Mismatches:** He et al. (2024) (L226), Jiang et al. (2023) (L330) and Cai et al. (2014) (L356) are cited but not listed; "Zhang et al. (2022)" (L225–226) does not say which of 2022a–c is meant. Several entries appear to be cited nowhere (e.g. Bellouin et al., 2013; Schaap et al., 2004; Zhang et al., 2019).

Supplement

- **Text S1:** At pH 7 acidic compounds are largely deprotonated and poorly retained on HLB, yet the retained "neutral" fraction is eluted with ammoniated methanol, the treatment normally reserved for acids. Please justify the scheme.
- **Text S2:** Eq. (7) is said to simplify Eq. (5); Eq. (4) is presumably meant. The constant K (L74) is never defined or used.
- **Table S1:** Please give the contributions of the individual parameters, not merely the total ranges.
- **Figure S2:** The units (MW) differ from those of Figure 3 (MW km^{-2}).