

## Reviewer #2

### General comments

This manuscript included interesting topic and monitored not only gaseous organic compounds but also other tracers such as CO<sub>2</sub>, CO, and aerosols which can assess different six fuel types. The authors used several tools using monitoring data to identify characteristics of each fuel type. In the aspects of reduction and verification for their emissions on climate and air quality issue, this study would be valuable reference for research community in the future. Even though its value, this manuscript was not well written. First, this manuscript would be more suitable to ‘measurement report’ type rather than ‘research article’ ([https://www.atmospheric-chemistry-andphysics.net/about/manuscript\\_types.html](https://www.atmospheric-chemistry-andphysics.net/about/manuscript_types.html)).

**Response:** We would like to thank the reviewer for the comments and suggestions to improve the current work. We will have the reviewer comments in black, address the comments in blue, and modified sentences in red.

We have restructured the manuscript. Our measurements not only provide key parameters of combustion (e.g., emission factors) but also offer the ability to identify and quantify rarely measured and previously unidentified organic vapor emissions, particularly those in the chemically complex low-volatility fraction. These insights are crucial for advancing the current understanding of the impact of solid fuel combustion on air quality and climate. Moreover, we applied the Mann-Whitney U test to biofuels and coal, allowing us to identify specific potential new markers for these fuels based on Vocus measurements. These markers serve as valuable references for field campaign studies focused on source apportionment.

Given the depth and significance of our findings, we believe that this work is more appropriate as a “research article” rather than a “measurement report”. It makes a substantial contribution to the scientific community's understanding of emissions and their implications.

Next, the title of this manuscript doesn't cover and represent of whole manuscript. The title makes readers misunderstand that this paper focus on Vocus PTR-TOF instrument. However, the manuscript includes more than that. Therefore, it would be good to find appropriate title.

**Response:** The main instruments we used for organic vapor measurement in this study is Vocus-PTR. However, the study also included the data from other instruments (e.g., CO<sub>2</sub>, BC) for emission factor calculation and interpretation of different combustion phase. Thus, we deleted “Vocus PTR-TOF” in the tile and change it to “Chemical characterization of organic vapors from wood, straw, cow dung, and coal burning”.

The terms were used too complicated to understand. There are several terms which seems like same or subtly different but used together without any clear categorizations (e.g., burning, burning type, fuels, fuel type, biomass burning, wood, and solid fuels). Authors might assume many researchers can distinguish and realize

those terms by themselves without clear explanation. However, in the scientific manuscript, clear definitions and unified term through a whole manuscript are very important tools for making readers understand what authors would like to say. Similarly, many of undefined (or unclear) terms were used in whole manuscript such as ‘common compounds’ and ‘characteristic compounds’ including ‘selected characteristic compounds’. It was not clear what common compounds (and also characteristic compounds) mean and represent of.

**Response:** We agree that clear definitions and consistent usage of terms are essential for ensuring that our readers can easily understand the distinctions we are making.

- 1) **Terminology Consistency:** We have revised the manuscript to ensure that the term “solid fuel type” is used consistently throughout. To prevent any confusion, we have avoided using terms like “fuel sources” or any other variations interchangeably.
- 2) **Clarification of Solid Fuels:** In this study, solid fuels are defined to include both biofuels (such as beech logs, spruce/pine logs, spruce/pine branches and needles, straw, cow dung) and coal. We have added this classification to Section 2.1 to ensure that readers clearly understand what we mean by solid fuels in the context of this research.
- 3) **Removal of Ambiguous Terms:** To avoid ambiguity, we have removed all instances of the term “characteristic compounds” from the manuscript. Instead, we now refer to the selected substances as “potential markers” based on the statistical methods used, similar to those employed by Zhang et al. (2023). This change helps in providing a more precise scientific explanation.
- 4) **Clarification of "Common" Markers:** We have clarified that the term “common” refers to potential markers that are applicable across all biomass fuels, rather than a specific type of biomass fuel. The Mann-Whitney U test was performed to identify potential markers among different types of solid fuels. However, in this study, biomass fuels (such as logs, branches, needles, straw, and cow dung) were analyzed separately from coal due to their distinct characteristics. To address this distinction, we characterized the dominant compounds across various biomass fuels by setting a threshold (relative mixing ratio contribution  $\geq 0.1\%$ ) for compounds that are not potential markers of one specific biomass fuels. This approach allowed us to identify compounds that are more readily detectable in complex environments. Recognizing that "common" is not a precise scientific term, we have removed it from the manuscript entirely to avoid any misunderstanding.

These revisions should help ensure that the terminology used in our manuscript is both clear and scientifically accurate, allowing readers to fully grasp the distinctions we are making in our study.

For the results and discussion Sect., authors handled with lots of information and data all together. This absolutely contributes to research community as a good reference. However, every section displayed very

independently and did not link together without any explanation of purpose for each section. This might be very confusing for readers. Therefore, I would like authors to re-organize structures in the manuscript and make it appealing.

**Response:** We have restructured the manuscript. We first discuss about “The characteristics of EF and MCE from combustion” and we found the average emission factors for organic vapors from different solid fuels have significant difference, depending on the combustion phases and solid fuel types. And then based on above results, we deeper analysis the relationship between MCE and combustion phase (smoldering and flaming) and the chemical composition of organic vapors from different solid fuels (Section 3.2 and Section 3.3). We observed that the chemical composition between different solid fuels is significant (O/C, H/C and relative contribution of different functional group, volatility distribution). Thus, in the Section 4, by using Mann-Whitney U test to biofuels and coal, we would like to find the dominant compounds for all biofuels and identify specific potential new markers for solid fuels based on Vocus measurements.

Thank you for your valuable feedback regarding the organization of the Results and Discussion sections. We understand that presenting a large amount of data and information can be overwhelming for readers, and it’s crucial that each section is cohesively linked to ensure clarity and purpose. We have reorganized the manuscript to create a more logical flow and to clearly connect each section. We now begin with a discussion on “The characteristics of EF and MCE from combustion” (Section 3.1). In this section, we highlight the significant differences in average emission factors for organic vapors from various solid fuels, which depend on both combustion phases and fuel types. Building on these findings, we delve deeper into the relationship between EFs, combustion phase (smoldering and flaming), and the chemical composition of organic vapors from different solid fuels in Sections 3.2 and 3.3. Here, we show that the chemical compositions (O/C, H/C ratios, relative contributions of different functional groups, and volatility distribution) vary significantly among the different solid fuels. Finally, in Section 4, we apply the Mann-Whitney U test for all solid fuel types. This analysis aims to identify specific potential new markers for these fuels based on Vocus measurements. This section builds directly on the findings from the earlier sections and serves to tie the entire discussion together.

These revisions ensure that each section of the manuscript is interconnected, making the study more coherent and accessible. We believe this restructuring enhances the manuscript's appeal and clarity, allowing readers to better understand the progression of our analysis and the conclusions drawn.

Unfortunately, the authors did not seem to give their attention meticulously. For example, some figures should be swapped (not be matched with each figure explanations), there are many abbreviations without full names, reference and explanation were not matched, etc.

**Response:** Thank you for your meticulous review and for pointing out these critical issues. We have addressed

each concern: Figures in Supplement: We have thoroughly reviewed and corrected the placement of figures in the manuscript. The figures are now correctly matched with their corresponding explanations, ensuring that each visual element accurately supports the text.

Abbreviations: We have systematically checked for all abbreviations used throughout the manuscript. We have introduced the full names upon first use.

References and Explanations: We have carefully reviewed all references and their corresponding explanations. Any mismatches have been corrected to ensure that each reference accurately supports the related content.

### Specific comments

1. P1 L19: Through whole manuscript, real-time gas-phase emissions analysis was not occupied large part. There is no timeseries data set without flaming/smouldering stage explanation.

**Response:** We conducted real-time combustion experiments for all solid fuels, and as part of our analysis, we focused on the spruce/pine logs-burning experiment to illustrate the time series data between the flaming and smoldering phases. This example was chosen because the emission factors and other particulate data, such as black carbon (BC) and organic carbon (OC), show significant differences between these two phases. In response to your feedback, we have removed the term “real-time” from the abstract to reflect the content more accurately. However, we have retained the mention of “real-time” in the methods section to clearly describe the experimental setup and procedures.

2. P1 L25-27: Please explain explicitly with a clear sentence. Authors use this type of sentence in the whole manuscript. It would be good to make short sentences with clear sentence.

- The  $C_xH_yO_z$  family is the most abundant group (of what?),

- but a greater contribution of nitrogen-containing species (than what?)

and  $C_xH_y$  families (related to polycyclic aromatic hydrocarbons) could be found in cow dung burning and coal burning, respectively. This sentence is quite vague to read because authors use conjunctions couple of times such as ‘but’ and ‘and’ in one sentence.

**Response:** We have rephrased this sentence and separated it into two sentences (Line 24-29).

The  $C_xH_yO_z$  family is the most abundant group of the organic vapor emitted from all SF combustion. However, among these SF combustions, a greater contribution of nitrogen-containing species and  $C_xH_y$  families (related to polycyclic aromatic hydrocarbons) is observed in the organic vapors from cow dung burning and coal burning, respectively.

3. P1 L28: Please explain explicitly with a clear sentence.

- especially for spruce and pine branches and needles (39.3%), and coal (31.1%)

- To: especially for spruce/pine branches and needles (39.3%) and coal (31.1%)

**Response:** We have rephrased this sentence and separated it into two sentences (line 30-32).

Intermediate volatility organic compounds (IVOCs) constituted a significant fraction of emissions in solid fuel combustion, ranging from 12.6% to 39.3%. This was particularly notable in the combustion of spruce/pine branches and needles (39.3%) and coal (31.1%).

4. P1 L33: What does the characteristic product mean? It is only for C<sub>9</sub>H<sub>12</sub>O? Is it scientific term people use in this field? Is it different from ‘characteristic compounds’ on line 32? What is ‘characteristic compounds’?

5. P2 L64: Author used the term of ‘characteristic compounds’. But never define what it means. This is totally different from the term such as ‘organic compounds’. Because readers can know what ‘organic’ is, it is not necessary to explain. However, ‘characteristic’ means a lot. Therefore if it is scientific term, authors should explain what it is or what kind of gases are belonging to this category. If it is not a scientific term, it is wrong expression.

**Response:** To avoid ambiguous expressions caused by non-scientific terms, we have removed all instances of the term "characteristic compounds" from the manuscript. Instead, we have used the same statistical methods as Zhang et al. (2023) to select substances that are statistical outliers relative to other emission sources and, following this definition, we refer to the selected substances that are statistical outliers as "potential markers." Due to the similarities among different types of wood burning (open and stove burning), and in comparison to spruce burning, we identified only one new potential marker compound, C<sub>9</sub>H<sub>12</sub>O, which originates from the pyrolysis of beech lignin. However, other compounds resulting from the pyrolysis of coniferyl-type lignin could also be considered potential markers for all types of wood burning.

6. P3 L93 and L97: Author mentioned that ‘Vocus PTR time-of-flight mass spectrometer (hereafter Vocus)’ but in L97, mentioned again Vocus PTR-TOF. If it is same word, please add TOF in L93 and change from ‘Vocus PTR-TOF’ to ‘Vocus’.

**Response:** We have revised the manuscript for consistency. In line with your suggestion, we have added "TOF" to the description to read "Vocus PTR-TOF mass spectrometer (hereafter Vocus)." We have also changed "Vocus PTR-TOF" to "Vocus" to ensure uniformity throughout the text.

7. P3 L113: The title of section ‘Fuel and burning set-up’ can ‘Fuel and burning types set-up’. Without ‘types’, the title sounds like physical set-up (of course, it includes the concept though.)

**Response:** We have updated the section title to "Fuel and burning types" to better reflect the content and avoid any ambiguity regarding the physical set-up.

8. P3 L117: Author mentioned six fuel type but did not explain why those were chosen for experiment.

9. P3 L121 to 130: Through the whole manuscript, it is very difficult to find the linkage of those explanations to the results. More efficient way to deliver how to categorize the burning type is to suggest ‘Six burning type with different fuels’ first. It would be good if authors suggest simple table for it. For example, ‘with those six different fuels, we categorized six burning types for this experiment. 1) beech logs stove, 2) spruce and pine logs stove, 3) spruce and pine branches and needles open, 4) straw open, 5) cow dung open and 6) coal stove. Among the list above, 1) and 2) are representative of residential wood burning..... (Table 1).’

10. P4 L125: There are lots of commas so that it makes readers difficult to understand. On this manuscript, there are many sentences with similar structures to this sentence. ‘Combustion of agricultural waste, straw, and a mixture of fresh spruce and pine branches and needles were in an open stainless-steel cylinder measuring 65 cm in diameter and 35 cm in height.’ Authors can divide into two sentences or make it simple.

**Response:** We have restructured this part and revised and separated the sentence to improve clarity by simplifying its structure.

The revised sentence now reads (Line 113-138):

The experiments were conducted at the Paul Scherrer Institute (PSI) in Villigen, Switzerland. The burning facility is part of the PSI Atmospheric Chemistry Simulation chambers (PACS). Real-time characterization of the primary gas and particle phase emissions was carried out during 28 test burns. Six solid fuels were studied (coal briquettes and biomass fuels: beech logs, spruce/pine logs, fresh spruce/pine branches and needles, dry straw, cow dung) with three to six replicate burns. Material in the beech, spruce, and pine fuels (e.g., logs and needles) was sourced from a local forestry company in Würenlingen, Switzerland. Cow dung cakes (a mixture of cow dung and straw) were collected from Goyla Dairy in Delhi, India. Coal briquettes were purchased from Gansu, China (Ni et al., 2021; Klein et al., 2018).

With those six different fuels, we categorized six burning types for this experiment. 1) beech logs stove, 2) spruce/pine logs stove, 3) spruce/pine branches and needles open, 4) dry straw open, 5) cow dung open and 6) coal stove. We selected these six solid fuels and conducted emissions tests to simulate certain types of burning found in the atmosphere. Among the list above, 1) beech logs stove and 2) spruce/pine logs stove are representative of residential wood burning, which are burned separately in a stove, consistent with the materials used in two previous articles (Bertrand et al., 2017; Bhattu et al., 2019). To represent forest fires or wildfire and agricultural field combustion, 3) a mixture of fresh spruce/pine branches and needles and 4) straw were combusted in an open stainless-steel cylinder (65 cm in diameter and 35 cm in height). Traditional cooking and heating practices in regions like India are represented by 5) cow dung cakes open burning by using half-open stoves (Loebel Roson et al., 2021). Finally, traditional cooking and heating practices in rural regions of developing countries are represented by 6) coal stove burning in a portable cast iron stove purchased from the local market (Liu et al., 2017). Of course, these conditions do not fully accurately represent the conditions found in actual fires, which consist of a variety of burning species (e.g., trees, underbrush, peat soils, etc....), but represent laboratory burning conditions.

11. P4 L132: Section title is not clear. It can be sampling and analysis method.

**Response:** In Section 2.2, we introduce the experimental design for the burning experiment and describe the

instruments used in this study. In Section 2.3, we present the analysis methods. We suggest that the section title could be “Experimental setup and instrumentation”.

12. P4 L134: Pure Air with N<sub>2</sub>, O<sub>2</sub>, Ar? The purity percentage?

**Response:** We have added a sentence to describe the pure air generator and provided two references that used this generator in chamber experiments (Line 141-145).

The zero air was provided by a zero air generator (737-250 series, AADCO Instruments, Inc., USA) for cleaning and dilution (Heringa et al., 2011; Bruns et al., 2015). The zero air generator takes ambient air and scrubs particulates and volatile organic compounds from the air leaving a mixture that is largely made up of N<sub>2</sub>, O<sub>2</sub>, and Ar at ambient concentrations. Other trace gases are scrubbed to lower than atmospheric concentrations including CO<sub>2</sub> (< 80 ppb) and CH<sub>4</sub> (< 40 ppb).

13. P4 L137: diluter ... Diluted by what?

**Response:** We have rephrased this sentence to clarify (Line 147-150).

Once a burn is initiated from the various combustibles, emissions are sampled from the chimney through a heated line (473 K). The emissions (both gas and particle phases) are then diluted by two Dekati diluters (DI-1000, Dekati Ltd.) which dilutes the emissions by a factor of ~ 100 (473 K, DI-1000, Dekati Ltd.).

14. P4 L151: Black Carbon (BC)?

**Response:** We added the “BC”.

15. P4 L152: A LI-7000 CO<sub>2</sub> analyzer (LI-COR) provided continuous measurements of carbon dioxide (CO<sub>2</sub>).

Is it same instrument of CO<sub>2</sub>/CO monitor in Figure S1? In this case, please add carbon monoxide as well.

**Response:** We added the instrument information for CO analyzer (Line 171-173).

A LI-7000 CO<sub>2</sub> analyzer (LI-COR) and APMA-370 CO analyzer (Horiba) provided continuous measurements of carbon dioxide (CO<sub>2</sub>) and carbon monoxide (CO), respectively.

16. P5 L160: The Vocus was calibrated before and after..... For the reliable data for other species, it would be good to suggest calibration strategy to other instrument as well.

**Response:** The Vocus is the primary instrument used in this study, and we have not included extensive details about its calibration for the chamber experiments.

However, other instruments (e.g., AMS, AE, SMPS) are widely used in atmospheric studies. Therefore, we have not provided detailed descriptions of the calibration processes for these additional instruments. Instead, we have briefly added introduction of the maintenance and calibration in the section 2.2 and cited more references related to the calibration methods applied to these instruments (Line 157-175).

Numerous instruments were connected after the second dekati diluter for the characterization of both the particulate and gaseous phases. A Scanning Mobility Particle Sizer (SMPS, CPC 3022, TSI, and custom-built DMA) provided particle number size distribution information and calibrated by using polystyrene latex (PSL) particle size standards (Wiedensohler et al., 2018; Sarangi et al., 2017). The non-refractory particle composition was monitored by a high-resolution time-of-flight aerosol mass spectrometer (HR-ToF-AMS, Aerodyne Research Inc.). AMS data were processed using SQUIRREL (SeQUential Igor data RetRiEvaL v. 1.63; D. Sueper, University of Colorado, Boulder, CO, USA) and PIKA (Peak Integration and Key Analysis v. 1.23) to obtain mass spectra of identified ions in the  $m/z$  range of 12 to 120. OC (organic carbon) is derived from the ratio of organic mass (OM) to OC (OM/OC) determined with high-resolution AMS analysis (Canagaratna et al., 2015). In the AMS mass spectra, the fraction of  $m/z$  60 ( $f_{60}$ ) represents the ratio of levoglucosan-like species (Schneider et al., 2006; Alfarra et al., 2007). AMS was calibrated for ionization efficiency (IE) by a mass-based method using  $\text{NH}_4\text{NO}_3$  particles (Tong et al., 2021). Black carbon (BC) was measured with an Aethalometer (Magee Scientific Aethalometer model AE33) (Drinovec et al., 2015) with a time resolution of 1 minute. The maintenance and calibration are given in the AE33 user manual – version 1.57. A LI-7000  $\text{CO}_2$  analyzer (LI-COR) and APMA-370 CO analyzer (Horiba) provided continuous measurements of carbon dioxide ( $\text{CO}_2$ ) and carbon monoxide (CO), respectively. The concentrations of total hydrocarbons (THC) and methane ( $\text{CH}_4$ ) were monitored using a flame ionization detector monitor (THC monitor Horiba APHA-370).

17. P5 L162 and L165: Please clarify pure air and zero air difference. If it is same, please unify the term. And also make it clear that only dried air used for background measurement.

**Response:** We have reviewed the terms and confirm that “pure air” and “zero air” refer to the same concept in this context. We have unified the terminology to use “zero air” throughout the document. Additionally, we have clarified that only dried zero air was used for Vocus background measurements.

18. P5 L173: In section 2.4, the title is very clear what will be discussed in the section. However, for section 2.3, the title is very vague.

**Response:** We have combined Sections 2.3 and 2.4, as both sections discuss the calculation of key parameters: MCE, EFs, and volatility. This merger provides a clearer and more cohesive structure for the content.

19. P5 L176: excess mixing ratio above what? If authors use the excess mixing ratio of CO and  $\text{CO}_2$  above background air, please add information of background air (where those data download or how measure it.) And also this methods in Equation 1. seems like little different from the reference (Ward and Radke, 1993).

**Response:** We have corrected the reference and added the information of the background air (Line 197-198). Where  $\Delta\text{CO}$ ,  $\Delta\text{CO}_2$  are the mixing ratios of CO or  $\text{CO}_2$  in excess of background (measured before the combustion), respectively (Christian et al., 2003).

20. P5 L179: The sentence seems very complicated. If authors would like to add instrument information inside



bracket, please add all of instrument information. If authors would like to describe species, please unify the species all. And clarify that meaning of conventional gases.

**Response:** We have rephrased this sentence to improve clarity and make it easier to understand (Line 201-204).

The emission factors (EFs,  $\text{g kg}^{-1}$ ) of species  $i$  was calculated, following a carbon-mass balance approach (Andreae, 2019; Boubel et al., 1969; Nelson, 1982):

$$EF_i = \frac{m_i}{\Delta mCO + \Delta mCO_2 + \Delta mCH_4 + \Delta mNMOGs + \Delta mOC + \Delta mBC} \times W_c$$

Here  $m_i$  refers to the mass concentration of species  $i$ .  $\Delta mCO$ ,  $\Delta mCO_2$ ,  $\Delta mCH_4$ ,  $\Delta mNMOGs$ ,  $\Delta mOC$ , and  $\Delta mBC$  are the background-corrected carbon mass concentrations of carbon-containing species in the flue gas.

21. P5 L182: mass concentration.

Author used units as combination of mixing ratio and mass concentration. However, it is different unit. (mass) mixing ratio: Number of the mass of the target gas (species) per mass of air (possible units are ppmm (also ppmw) = parts per million of air molecules by mass (weight), etc.). A specification whether it refers to dry or moist air is required. mass concentration is mass/volume. Please clarify all units.

**Response:** The data used for MCE calculation are the CO and CO<sub>2</sub> mixing ratios. However, the emission factor calculation uses mass concentration data. The misunderstanding arose from using the same symbol for these two equations. We have corrected the symbol in the emission factor equation to clarify this distinction (Line 201-204).

The emission factors (EFs,  $\text{g kg}^{-1}$ ) of species  $i$  was calculated, following a carbon-mass balance approach (Andreae, 2019; Boubel et al., 1969; Nelson, 1982):

$$EF_i = \frac{m_i}{\Delta mCO + \Delta mCO_2 + \Delta mCH_4 + \Delta mNMOGs + \Delta mOC + \Delta mBC} \times W_c$$

22. P5 L182:  $\Delta OC$ : This was not discussed in section 2.2.

23. P5 L184: OM also was not discussed at all.

**Response:** We have added the discussion and definition of OC and OM in section 2.2 (Line 165-166).

OC (organic carbon) is derived from the ratio of organic mass (OM) to OC (OM/OC) determined with high-resolution AMS analysis (Canagaratna et al., 2015).

24. P6 L203: In this study, the mixing ratio relative contribution for more than 1500 species from six different fuels for all 28 test burns was quantified by using Vocus. It is difficult to understand the meaning of ‘mixing ratio relative contribution’. Relative contribution to what?

**Response:** The relative contribution of species  $i$  was calculated as the mixing ratio of species  $i$  divided by the mixing ratio of total organic vapor, multiplied by 100%. We have rephrased the sentence (Line 221-222).

In this study, the relative contribution of the mixing ratio for over 1,500 species from six different fuels was quantified across all 28 test burns using the Vocus.

25. P6 L204: characteristic compounds. This term never mentioned above in this manuscript.

**Response:** We have changed the term “characteristic compounds” to “potential markers” throughout the manuscript to ensure consistency and clarity.

26. P6 L213-L218: make the sentences clear. However, due to the similarity in fuel types between burning spruce and pine logs, as well as spruce and pine branches and needles, they were categorized as separate fuel sources for this test and not compared with each other but were only compared with the other four types of fuels.

- It would be good to make it simple: for this test, ‘spruce and pine logs’ and ‘spruce/pine branches and needles’ were in the same fuel type category due to similar characteristics.

- And then readers can have questions, if so (they have same characteristics), why did authors use all of them as different fuels at the beginning?

- If author define 6 different fuel type, please keep the term of ‘fuel type’ in the whole manuscript. Please don’t change to fuel sources or other. It makes confusions. Using term should be clear and same. For burning type as well.

- The sentence below is difficult as well. Similarly, due to the composition of cow dung "cakes," which comprise a mixture of dried cow dung and crop residues and a relatively high correlation between cow dung and straw (Figure 1), the Mann-Whitney U test was carried out without accounting for the presence of the other fuels.

**Response:** Spruce/pine logs and spruce/pine branches and needles were not in the same solid fuel type category. The chemical components produced from burning spruce/pine logs differ significantly from those produced by burning spruce/pine branches and needles. However, in the Mann-Whitney U test, to identify potential markers for one type of fuel, such as spruce/pine logs, the spruce/pine branches and needles were included in the comparison with other fuels. This could result in the loss of many common characteristic markers since these two types of fuel actually come from the same type of tree. Therefore, when identifying markers for spruce/pine logs using the Mann-Whitney U test, spruce/pine branches and needles were not included in the comparison group. Similarly, due to the composition of cow dung “cakes”, which are a mixture of dried cow dung and crop residues, the approach used in the Mann-Whitney U test is consistent with the above method.

We have ensured that the term “solid fuel type” is consistently used throughout the manuscript to avoid confusion.

We have made sure not to use “fuel sources” or any other terms interchangeably. Similarly, we have maintained

consistent terminology for “burning type” throughout the text. This should provide clarity and prevent any confusion (Line 240-250).

To identify potential markers the Mann-Whitney U test was used to compare the emissions observed for one type of fuel, (e.g., spruce/pine logs) with the gaseous emissions observed for other fuels. The data used for the comparison was the average composition measured throughout a full burning cycle, excluding the initial ignition period.

However, due to the similarity in solid fuel types between burning spruce/pine logs, as well as spruce/pine branches and needles, they were categorized as separate solid fuel types for this test and not compared with each other but were only compared with the other four types of fuels. This could result in the loss of many same markers since these two types of fuel actually come from the same type of tree. Therefore, when identifying markers for spruce/pine logs using the Mann-Whitney U test, spruce/pine branches and needles were not included in the comparison group. Similarly, due to the composition of cow dung 'cakes,' which are a mixture of dried cow dung and crop residues, the approach used in the Mann-Whitney U test is consistent with the above method.

27. P7 L226: ‘Emission factors from solid-fuel combustion’ to ‘The characteristics of EF and MCE from different fuel types.’ ? make it more clear.

**Response:** We change it to “The characteristics of EF and MCE from different solid fuel types”.

28. P7 L237: did author mention that NMOG is primary organic gases in the above manuscript? It would make easier to readers if authors mention that NMOG is primary organic gases in the introduction. or Just use NMOG in whole manuscript. It is very important that using same term in whole manuscript for readers.

**Response:** We have reviewed the manuscript and confirmed that we use the term “organic vapors” consistently throughout, aligning with the manuscript's title.

29. P7 L243: The slightly higher EFs for pine and spruce wood burning can be attributed to the more extensive analysis of NMOGs in our study compared to previous research (37.3 245 g/kg) (Hatch et al., 2017).

- Can we say this value is higher than pervious study even though the difference is in the uncertainty levels?

**Response:** Based the comments, we have rephrased this sentence (Line 268-270).

This value is higher than pervious study (37.3 g kg<sup>-1</sup>) even though the difference is in the uncertainty levels , which can be attributed to the more extensive analysis of organic vapor in our study (Hatch et al., 2017).

30. P7 L249: emissions. Does it mean EF? Authors know the difference between emissions and EF.

**Response:** Yes, we corrected it to “EFs” in the manuscript.

31. P7 L250: emission factors. If authors started using EF in place of emission factors, please use EF.

**Response:** We have standardized the terminology throughout the manuscript. We ensured that “EFs” is used

consistently in place of “emission factors” throughout the manuscript.

32. P7 L253: title. Authors never mentioned about primary organic vapors in previous sections. It can be ‘The characteristics of NMOG chemical composition from different fuel types’.

**Response:** Based the comments, we revised it to:

3.3 The characteristics of organic vapor from different solid fuel types

33. P7 L254: I don’t think section 3.2.1 can be overview. In this section, general characteristics can be handled.

**Response:** Based the comments, we revised it to:

3.3.1 Chemical composition of organic vapor from combustion

34. P7 L255: combustion fuel source mean burning type? Please keep same term.

**Response:** We have ensured that the term “solid fuel types” is consistently used throughout the manuscript to avoid confusion.

35. P7 L257: what is primary NMOGs? So is it different from just NMOGs?

**Response:** We have ensured that the term “organic vapors” is consistently used throughout the manuscript to avoid confusion.

36. P7 L261: biomass samples. What kind of burning type are belonging to biomass samples. Please defined clearly.

**Response:** We have revised it to “solid fuel types” to avoid confusion.

37. P8 L273: What is H/C ratio?

38. P8 L274: What is O/C ratios?

**Response:** We have clarified the definition of H/C and O/C in the manuscript (Line 335-338).

Hydrogen to carbon ratios (H/C, calculated as the ratio of hydrogen atoms to carbon atoms in a molecules).

Oxygen to carbon ratios (O/C, calculated as the ratio of oxygen atoms to carbon atoms in a molecule).

39. P8 L278: Suggested its value. Science paper should suggest certain number otherwise 'slightly lower' means very subjective.

**Response:** We have rephrased this sentence and added the value (Line 340-343).

The results show similarities to the comparison between burning wood and cow dung in the particle phase (Zhang et al., 2023). Specifically, cow dung exhibits a lower fraction of high O/C (0.22) compared to other fuels studied.

40. P8 L290-L301: I suggest this paragraph can come up the beginning of this section. And then authors can easily explain Figure 1 and Figure 2. On the other hand, I also wondered NMOG will be categorized only primary organic gases and secondary as well.

**Response:** In this study, all emissions from solid fuel combustion are considered “primary”. To avoid confusion, we have consistently used the term “organic vapors” throughout the manuscript without emphasizing “primary”. Additionally, we have moved this explanation to the beginning of the section for better clarity.

41. P9 L303: Hard to understand this sentence. Based on the  $\log_{10}C^*$  values of all organic compounds parameterized with the modified approach of Li et al. (2016) described in Sect. 2.4.

**Response:** We have rephrased this sentence (Line 73-78).

The parameterization described in Sect. 2.4 uses the modified approach of Li et al. (2016) to estimate the volatility of each of the measured compounds by the VOCUS in  $\log_{10}(C^*)$  [ $\mu\text{g m}^{-3}$ ]. The gaseous organic compounds were grouped into a 14-bin volatility basis set (VBS) (Donahue et al., 2006) (Figure 4). Following the suggestions in recent papers (Wang et al., 2024; Li et al., 2023; Donahue et al., 2012; Huang et al., 2021; Schervish and Donahue, 2020), the volatility was aggregated into four main classes with units of  $\mu\text{g m}^{-3}$ : VOCs as  $\log_{10}(C^*) > 6.5$ , IVOCs as  $\log_{10}(C^*)$  between 6.5 to 2.5, semi-VOCs (SVOCs) as  $\log_{10}(C^*)$  between 2.5 to - 0.5 and low-VOCs (LVOCs) as  $\log_{10}(C^*) < - 0.5$ .

42. P9 L308: What kind of burning type are involved in biomass burning emissions?

**Response:** We have rephrased this sentence (Line 380-381).

Comparison and compilation of organic vapors sorted by volatility and functional group classification are shown in Figure 4.

43. P9 L311: For all burns. Did author mean for all experiment or for all burning types?

**Response:** Yes. We have corrected it to “all burning types”.

44. P9 L320-323: I think the reference cannot support author’s finding. In general, spruce and pine branches and needles open burning released a higher proportion of IVOCs (39.3%) into the gas phase compared to stove logs burning (12.6% and 23.9%). Pallozzi et al. (2018) also reported a similar result, showing that needle/leaf combustion released a greater amount of volatile organic compounds into the atmosphere than branch combustion.

**Response:** We have deleted this sentence after carefully reviewing Pallozzi et al. (2018). Upon further examination, we found that the study only mentions specific OVOC compounds being higher in branches and needles, such as the combustion of leaves releasing more benzene than needles, and branches and needle litter

from pine releasing higher amounts than oak. However, this does not sufficiently support our findings, so we have removed the reference Pallozzi et al. (2018).

45. P9 L332: VBS. What does VBS mean?

**Response:** We have added the definition here (Line 73-379).

The parameterization described in Sect. 2.4 uses the modified approach of Li et al. (2016) to estimate the volatility of each of the measured compounds by the VOCUS in  $\log_{10}(C^*)$  [ $\mu\text{g m}^{-3}$ ]. The gaseous organic compounds were grouped into a 14-bin volatility basis set (VBS) (Donahue et al., 2006) (Figure 4). Following the suggestions in recent papers (Wang et al., 2024; Li et al., 2023; Donahue et al., 2012; Huang et al., 2021; Schervish and Donahue, 2020), the volatility was aggregated into four main classes with units of  $\mu\text{g m}^{-3}$ : VOCs as  $\log_{10}(C^*) > 6.5$ , IVOCs as  $\log_{10}(C^*)$  between 6.5 to 2.5, semi-VOCs (SVOCs) as  $\log_{10}(C^*)$  between 2.5 to - 0.5 and low-VOCs (LVOCs) as  $\log_{10}(C^*) < - 0.5$ .

46. P9 L333: wood burning. Did author define wood burning and biomass burning somewhere? What kind of burning type are in this category? And why do authors want to compare two stages? why do authors only analyze wood burning for this comparison?

**Response:** In this study, solid fuels include the biofuels (beech logs, spruce/pine logs, spruce/pine branches and needles, straw, cow dung) and coal. We have revised the sentence: We have added the classification of biomass fuels and coal in the Section 2.1 (Line 118-119). We observed that straw burning typically occurs very rapidly, often within just 10 minutes. While we also compared the flaming and smoldering phases of cow dung burning, the differences between these phases were not significant. Therefore, we focused on wood burning as a representative example to analyze the variations in organic vapor and other particle-phase concentrations, chemical compositions, and other parameters across different combustion stages.

Previous research have demonstrated distinct emission profiles of organic aerosol between flaming and smoldering phases (Heringa et al., 2011; Li et al., 2021). This prompted us to explore the changes in chemical composition and emission factors (EFs) of organic vapors during these different phases. Understanding these variations can provide valuable insights for future research on secondary organic aerosol (SOA) formation and model simulations (Stefenelli et al., 2019).

Six solid fuels were studied (coal briquettes and biomass fuels: beech logs, spruce/pine logs, spruce/pine branches and needles, straw, cow dung) with three to six replicate burns.

47. P9 L335-337: I don't agree with authors point of view. CO increased when the stage was changed from flaming to smoldering. In the top panel, the MCE is used to indicate the flaming stage with a significant CO<sub>2</sub> enhancement, while the smoldering stage exhibits high levels of CO.

**Response:** I'm not entirely certain I understood your comment fully, but to avoid any potential misunderstanding,

I have rephrased the sentence for clarity (Line 281-283).

In the top panel, the MCE, CO, and CO<sub>2</sub> concentrations, along with our experimental records, are used to indicate the flaming and smoldering stages.

48. P9 L338: What AAE means?

**Response:** “AAE” is an abbreviation for “absorption Ångström exponent” We have added this definition in the manuscript for clarity.

49. P9 L339: f60. Have ever author described what it is in the manuscript or table?

**Response:** “f60” is an abbreviation for “the fraction of  $m/z$  60”. We have added this definition in the Section 2.2 in the manuscript for clarity (Line 166-168).

In the AMS mass spectra, the fraction of  $m/z$  60 ( $f_{60}$ ) represents the ratio of levoglucosan-like species (Schneider et al., 2006; Alfarra et al., 2007).

50. P9 L341: BB. What does it mean?

**Response:** “BB” is an abbreviation for “biomass burning”. We have added this definition earlier in the manuscript for clarity.

51. P10 L350: (31.4 g/kg) and (121.9 g/kg). Please add standard deviation or uncertainty  $\pm$  value.

**Response:** We have added the standard deviation in the manuscript (Line 294-296).

On average, EFs for organic vapors in the flaming stage are approximately four times lower ( $31.4 \pm 7.1$  g kg<sup>-1</sup>) than those in the smoldering stage fires ( $121.9 \pm 24$  g kg<sup>-1</sup>).

52. P10 L351: Large similarity. I cannot understand large similarities. Did authors mean similar trend?

**Response:** We acknowledge that the term "large similarity" might be unclear. To clarify, we mean that while there are significant differences in emission strength (such as emission factors or concentrations) between the flaming and smoldering phases, the average carbon and oxygen distribution of organic vapors remains relatively consistent across these phases. In other words, although the overall emission levels vary, the average carbon and oxygen distribution of organic vapors in the emissions does not change significantly (Line 296-298).

Despite significant variability in the strength of organic vapors emissions (EFs), the average carbon and oxygen distribution of organic vapors remained largely consistent across the combustion phases (Figure S4).

53. P10 L352: Figure S7. doesn't have information that what each of #c and #o represents for and also its explanation does not match to the Figure.

**Response:** We apologize for the mistake. The legends for Figure S7 and Figure S8 (now Figure S4 and Figure S5) were mistakenly swapped. We have now corrected this issue so that the figures and their explanations match accurately.

54. P10 L356: What does OVOC stand for?

**Response:** “OVOC” is an abbreviation for “oxygenated VOCs”. We have added this definition in the manuscript for clarity.

55. P10 L356: Figure S8. I assumed Figure S8 was not the figure author would like to show.

**Response:** We apologize for the mistake. The legends for Figure S7 and Figure S8 were mistakenly swapped. We have now corrected this issue so that the figures and their explanations match accurately.

56. P10 L365: What does 'common' mean?

**Response:** We have changed the title of this section to “**Chemical characteristics of dominant compounds from all biomass fuels**”. In this section, we aim to distinguish dominant compounds that apply to all biomass fuels (not just a specific type of biomass fuel) and those that are specific to particular solid fuels. The “common” means potential markers that apply to all biomass fuels. However, it is not a scientific term. We have deleted this term in the whole manuscript.

57. P10 L382: Unify the term. there is no 'wood' in Figure 1.

**Response:** In this study, solid fuels include the biofuels (beech logs, spruce/pine logs, spruce/pine branches and needles, straw, cow dung) and coal. We have revised the sentence (Line 422-423):

**As shown in Figure 2, biomass fuels (logs, branches and needles, straw and cow dung) are different fuels from coal in this study.**

58. P11 L395: solid-fuel combustion. This is very confusing that solid fuel, biomass burning, fuel type, wood... all things are very tangled so that hard to understand. Please keep same term and define each term clearly.

**Response:** The title of this section has been changed to “Identification of potential markers for specific solid fuels”. In this study, solid fuels include the biofuels (beech logs, spruce/pine logs, spruce/pine branches and needles, straw, cow dung) and coal. Biomass burning is the burning of biomass biofuels. In the section of 3.4.1, we discuss about the chemical characteristics of dominant compounds from all biomass fuels.

59. P11 L398: add number of table.



**Response:** We have uploaded a separate Excel table as the Supplementary Table.

60. P11 L399: Very confusing between characteristic compounds and selected characteristic compounds. What are the differences between two of them?

61. P11 L401: In contrast, compounds from open burning of straw and cow dung Does it different form characteristic compounds and selected characteristic compounds?

**Response:** We have removed the “selected characteristic compounds”

62. P12 L448: solid fuel combustion, including residential burning (beech logs, a mixture of spruce and pine logs, and coal briquettes) and open combustion (spruce and pine branches and needles, straw, and cow dung).

- Author also investigated biomass burning and woods? did authors use biomass burning same to solid fuel?

- The burning type category is very important here. Please use it clearly.

**Response:** In this study, solid fuels include the biofuels (beech logs, spruce/pine logs, spruce/pine branches and needles, straw, cow dung) and coal. We have added the classification of biomass fuels and coal in the Section 2.1 (Line 118-119). We also rephrased the sentence in the conclusion (Line 48-489).

Six solid fuels were studied (coal briquettes and biomass fuels: beech logs, spruce/pine logs, spruce/pine branches and needles, straw, cow dung) with three to six replicate burns.

In this study, we investigated emissions of organic vapors using Vocus during typical solid fuel combustion, including burning of beech logs, spruce/pine logs, spruce/pine branches and needles, straw, and cow dung and coal briquettes.

63. P13 L481: Still have questions of characteristic compounds and the common compounds.

**Response:** We have addressed some similar comments before.

To avoid ambiguity, we have removed all instances of the term “characteristic compounds” from the manuscript. Instead, we now refer to the selected substances as “potential markers” based on the statistical methods used, similar to those employed by Zhang et al. (2023). This change helps in providing a more precise scientific explanation.

Clarification of "Common" Markers: We have clarified that the term “common” refers to potential markers that are applicable across all biomass fuels, rather than a specific type of biomass fuel. However, recognizing that "common" is not a precise scientific term, we have removed it from the manuscript entirely to avoid any misunderstanding.

64. Figures and table.

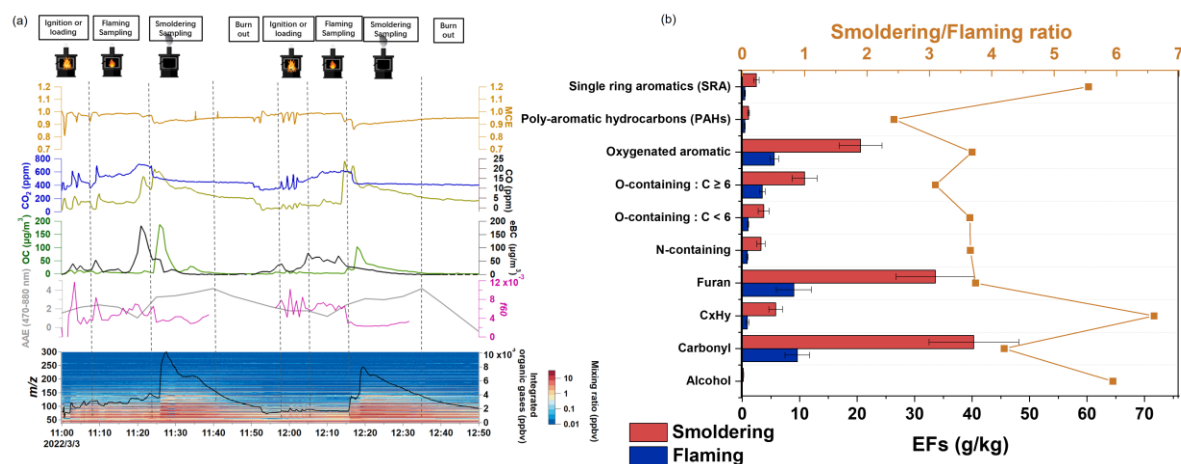
- Figures label fonts were too small to read.

- Figure labels and explanations in the captions should be same to the explanation in the manuscript.

- Figure 4. What is key aerosols? Figure 4(b) the colour bar of Smouldering and Flaming can be moved to right side of the panel.

**Response:**

- We have increased the label font size for Figures 1-4 to improve readability. Since the manuscript currently only allows uploading images in PDF format, which may result in compression and reduced image resolution, we will upload the figures separately after the review process is completed.
- We have reviewed and ensured that the labels and explanations in the figure captions match the explanations provided in the manuscript.
- In Figure 4, the term "key aerosols" refers to organic matter (OM) and black carbon (BC). We have clarified this in the figure caption. Additionally, we have adjusted Figure 4(b) by moving the color bar for the smoldering and flaming phases to the right side of the panel, as suggested.



**Figure 1.** (A) Temporal profiles of mixing ratios measured by Vocus and evolution of CO, CO<sub>2</sub>, AAE, *f*<sub>60</sub>, MCE and key aerosol compositions during burning cycles of beech logs stove burning (B) Geometric mean of the primary EFs for gas-phase species of different functional groups during flaming and smoldering phase, respectively (the flaming and smoldering was separated by the experimental record and calculated MCE). Error bars correspond to the sample geometric standard deviation of the replicates. The square represents the mixing ratio between smoldering and flaming. In this study, the MCE is used to indicate the flaming stage and smoldering and a significant decrease of MAC and CO<sub>2</sub> was observed from the flaming phase to the smoldering phase.

65. Reference.

- Please give the space between the references at least.

**Response:** We will ensure that there is adequate spacing between the references in the revised version of the manuscript.

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