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Organic vapors from wood, straw, cow dung, and coal burning using Vocus PTR-TOF

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Abstract

Solid fuel (SF) combustions, including coal and biomass, are important sources of pollutants in the particle and gas phase and therefore have significant implications for air quality, climate, and human health. In this study, we systematically examined real-time gas-phase emissions using the Vocus proton-transfer-reaction time-of-flight mass spectrometer, from a variety of solid fuels, including beech logs, spruce and pine logs, spruce and pine branches and needles, straw, cow dung, and coal. The average emission factors (EFs) for organic gases ranged from 6.7 to 74.2 g kg^{-1}, depending on the combustion phases and fuel types. Despite slight differences in modified combustion efficiency (MCE) for some experiments, increasing EFs for primary organic gases were observed with lower MCE. The $C_xH_yO_z$ family is the most abundant group, but a greater contribution of nitrogen-containing species and C_xH_y families (related to polycyclic aromatic hydrocarbons) could be found in cow dung burning and coal burning, respectively. Intermediate volatility organic compounds (IVOCs) also constituted a considerable fraction in solid-fuel combustions (from 12.6% to 39.3%), especially for spruce and pine branches and needles (39.3%), and coal (31.1%). Despite the large variability of EFs in the organic gas emissions, the relative contribution of different classes showed large similarities between the combustion phases in beech stove burning. The product from pyrolysis of coniferyl-type lignin and the extract of cedar pine needle were identified as characteristic compounds in the spruce and pine branches and needles open burning (e.g., $C_{10}H_{14}O_2$, $C_{11}H_{14}O_2$, $C_{10}H_{10}O_2$). The characteristic product ($C_9H_{12}O$) from the pyrolysis of beech lignin was identified as the characteristic compound for beech log stove burning. Many series of nitrogen-containing homologues (e.g., $C_{10}H_{11\text{-}21}NO$, $C_{12}H_{11\text{-}21}N$, $C_{11}H_{11\text{-}23}NO$ and $C_{15}H_{15\text{-}31}N$) and nitrogen-containing species (e.g., acetonitrile, acrylonitrile, propanenitrile, methylpentanenitrile) were specifically identified in cow dung burning emissions. Polycyclic aromatic

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hydrocarbons (PAHs) with 9-12 carbons were identified with significantly higher abundance from coal burning compared to emissions from other studied fuels. The composition of these characteristic organic vapors reflects the burned fuel types and can help constrain emissions of solid fuel burning in regional models.

Keywords: Vocus, solid fuel, primary emission, characteristic compounds, combustion phase

1 Introduction

Solid fuels (SFs), including coal and biomass, are a primary source of domestic heating worldwide (Tao et al., 2018; Oberschelp et al., 2019; Wu et al., 2022). In developing regions, such as India, more than 80% of rural households use biomass as cooking fuel (Balakrishnan et al., 2011). Firewood is mainly used for rural households, followed by crop residues and cow dung 'cakes', which are made of a mixture of dried cow dung and crop residues (Loebel Roson et al., 2021; Chandramouli and General, 2011). In Europe, fireplaces and woodstoves are used for domestic heating in winter, which have considerable impacts on air quality, resulting in intense 'smog' events (Kalogridis et al., 2018; Fourtziou et al., 2017; Bailey et al., 2019; Font et al., 2022). China is the largest producer and consumer of coal in the world. In China and some Eastern European countries like Poland, coal is widely used for domestic purposes, such as heating and cooking of households, due to its cost-effectiveness and easy accessibility (Guo et al., 2021; Stala-Szlugaj, 2018). The combustion of these solid fuels has been recognized as the main source of anthropogenic emission of atmospheric pollutants that elicit adverse effects on air quality and human health (Wu et al., 2022; Zhang and Smith, 2007).

Wildfires or bushfires have become more frequent in many regions due to heatwaves and drought (Weber and Yadav, 2020; Williams et al., 2012). SF combustion, including wildfires, is a major source of non-methane organic gases (NMOGs) to the atmosphere, emitting hundreds to thousands of different organic gas-phase species (Hatch et al., 2019; Koss et al., 2018; Permar et al., 2021). Once emitted, evaporated vapors or freshly emitted burning NMOGs will oxidize to produce oxygenated organic gases with a broad volatility range. These organic gases with sufficiently low volatility will nucleate or condense onto pre-existing aerosols to form secondary organic aerosols (SOA) (Kumar et al., 2023).

The identification of characteristic compounds for each emission source will be highly valuable in evaluating SOA formation potential and ambient source contributions. Liu et al. (2008) identified potential characteristic volatile organic compounds for different emission sources (e.g., biomass burning, mobile sources and painting). Nevertheless, these commonly used characteristic compounds are well-established, yet due to their presence in more than one type of biomass fuel, distinguishing between different biomass-burning sources presents challenges. Since 2009, there have been many advancements in the gas-phase measurements of SF, which include lab studies (Bruns et al., 2017; Bruns et al., 2016; Bhattu et al., 2019) and large field campaigns (e.g., WE-CAN Aircraft Measurements, FIREX-AQ campaign) (Permar et al., 2021; Jin et al., 2023; Majluf et al., 2022). However, efforts toward understanding SOA formation in burning plumes have been hindered by limited identification and quantification of NMOGs emitted by fires, especially intermediate volatility organic compounds (IVOCs) (Akagi et al., 2011). Laboratory and field campaigns suggest that intermediate volatility organic compounds are important precursors of SOA. Grieshop et al. (2009) demonstrated that traditional SOA precursors account for less than 20% of the observed SOA formed from residential

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wood combustion emissions, while IVOCs can contribute approximately 70% of the formed SOA (Li et al., 2024), which highlights the urgent need for more research on IVOCs from biomass burning emissions.

Adding an IVOC emission inventory to an air quality model can significantly narrow the gap between the estimated and measured SOA concentrations(Li et al., 2024; Hodzic et al., 2010; Zhao et al., 2016; Robinson et al., 2007).

Offline sampling methods such as canisters and adsorption-thermal desorption (ATD) cartridges, along with gas chromatography (GC) analysis, have limitations related to their low time resolution, susceptibility to sampling artifacts, and a limited range of measurable compounds (Hatch et al., 2018; Hatch et al., 2017). In addition to offline techniques, proton-transfer-reaction mass spectrometry (PTR-MS) has been widely used for the online measurement of volatile organic compounds (VOCs) in the atmosphere (Yuan et al., 2017). However, IVOCs still suffer from high losses in the sampling lines and PTR-MS drift tubes. Furthermore, most studies have focused on either primary or aged emissions, with very few examining the real-time influence of combustion conditions on the composition of emitted organic gases (Bruns et al., 2016; Akherati et al., 2020; Tkacik et al., 2017). The recently developed Vocus PTR time-of-flight mass spectrometer (hereafter Vocus) has greatly enhanced sensitivity due to a newly designed chemical ionization source (Krechmer et al., 2018), and it can detect a broader spectrum of VOCs, IVOCs, and their oxygenated products (up to six to eight oxygen atoms for monoterpene oxidation products) (Li et al., 2020; Wang et al., 2021; Riva et al., 2019). With a novel design and chemical ionization source, the Vocus PTR-TOF allows for real-time characterization of gas-phase emissions during various burning phases (e.g., flaming and non-flaming phases) and identifies the characteristic compounds for a wide range of fuels.

The present study compares real-time emissions from different combustion fuels. We systematically characterize the primary organic gas composition using Vocus from a variety of burning fuels from both residential stoves (beech logs, pine and spruce logs, and coal) and open combustion (spruce and pine branches and needles, straw, cow dung). We investigate the characteristics of compounds and emission factors for different combustibles and explore the dependence of individual NMOGs emission intensity, variability, and chemical composition on combustion conditions (open or stove) and combustion phases. We also discuss characteristic compounds for the burning fuels examined in this study. The differences in EFs and profiles between different combustibles can be considerable, and these results illustrate the importance of considering these emission sources individually. Measurements capable of identifying and quantifying rarely measured and presently unidentified emissions of NMOGs, particularly chemically complex low volatility fraction, are vital for advancing the current understanding of the impact of solid fuel combustion on air quality and climate.

2 Materials and methods

2.1 Fuel and burning setup

The experiments were conducted at the Paul Scherrer Institute (PSI) in Villigen, Switzerland and the burning facility that 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 fuels were studied (beech, spruce, pine, straw, cow dung, and coal briquettes) with three to six

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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). These fuels are classified into four categories, representing different types of burning scenarios in real life: 1) residential wood burning, 2) agricultural burning, 3) forest fires, and 4) traditional heating and cooking in two regions. Residential wood burning included beech and spruce/pine logs burned separately in a stove, consistent with the materials used in two previous articles (Bertrand et al., 2017; Bhattu et al., 2019). 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. Forest fires were simulated by burning a mixture of spruce and pine branches and needles. Traditional cooking and heating practices in regions like India are represented by burning cow dung cakes using half-open stoves (Loebel Roson et al., 2021). Traditional cooking and heating practices in rural regions of developing countries are represented by burning coal in a portable cast iron stove purchased from the local market (Liu et al., 2017).

2.2 Instrumentation and sampling

The experimental design is shown in Figure S1. In summary, it is made up of a burner and a set of diluters with heated lines. Before each burn, a continuous stream of pure air was passed through the gas lines overnight to avoid cross-contamination between burns and to ensure a low background of VOCs. From the various combustibles, once a burn is initiated, emissions are sampled from the chimney through a heated line (473 K) and diluted by two dekati diluters by a factor of ~ 100 (473 K, DI-1000, Dekati Ltd.). Note that wood-burning combustion cycles consist of a first cycle referred to as the 'first load' and subsequent cycles, referred to as 'reloads'. The first load consisted of a cold start, flaming, smoldering, and burn-out phase, and the reloads were comprised of a warm start, flaming, smoldering, and burn-out phase. NMOG emissions of solid fuel combustion are released within 10−30 min after loading. We define the time until full ignition duration for burning encompasses 80% of the entire process, starting from loading the fuels to burnout.

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. 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. Black carbon was measured with an Aethalometer (Magee Scientific Aethalometer model AE33) (Drinovec et al., 2015) with a time resolution of 1 minute. A LI-7000 CO_2 analyzer (LI-COR) provided continuous measurements of carbon dioxide (CO_2). The concentrations of total hydrocarbons (THC) and methane (CH_4) were monitored using a flame ionization detector monitor (THC monitor Horiba APHA-370).

We deployed a Vocus to measure organic vapors with a wider range of volatilities. A detailed description of the Vocus is provided elsewhere (Huang et al., 2021; Krechmer et al., 2018). For this study, the Vocus was operated with H_3O^+ as the reagent ion. The sample air was drawn in through a 1 m long polytetrafluoroethylene (PTFE) tube (6 mm o.d.) using a total sample flow of 4.3 L/min, which

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helped reduce the losses in the inlet wall and the sampling delay. Of the total sample flow, only 100-150 cm^3/min went to Vocus, and the rest was exhausted. The Vocus was calibrated before and after measurements every day using a multi-component standard cylinder (Tofwerk AG). Standard gases were diluted by the injection of zero air, producing mixing ratios of VOCs of around 20 ppbv. The calibration components were methanol, acetaldehyde, acetonitrile, acetone, acrylonitrile, isoprene, methyl ethyl ketone, benzene, toluene, m-xylene, α-pinene and 1,2,4-trimethylbenzene. The background measurements were performed using dry zero air every day. Data were recorded with a time resolution of 1 s. The raw data were processed using Tofware v3.2.3 software (TOFWERK, Aerodyne, Inc.). The standard non-targeted analysis workflow developed by Tofwerk was adopted for mass calibration and peak fitting. The mass transmission function and the ratios between the measured and calculated sensitivities for a series of ions were used to quantify the data and convert the ion counts to ppbv. To calculate the mixing ratio for compounds not present in the calibration mixture, the slope of the linear fit was multiplied by the proton transfer rate constants (k_{ptr}) which have been provided in Supplement Table.

2.3 Data analysis

Modified combustion efficiency (MCE, Equation 1) is an estimate of the relative amount of flaming and smoldering and is equal to

$$MCE = \frac{\Delta CO_2}{\Delta CO + \Delta CO_2} \qquad \text{Equation (1)}$$

Where ΔCO, ΔCO_2 are the excess mixing ratios of CO or CO_2, respectively (Ward and Radke, 1993). Generally, a higher MCE (> 0.9) suggests dominated flaming combustion, whereas a lower MCE (< 0.9) is mostly associated with smoldering combustion (Zhao et al., 2021; Zhang et al., 2022).

Primary emission factors (EFs, g kg^{-1}) of conventional gases (CO_2, CO, and CH_4), NMOGs (Vocus) and particle phase species were calculated, following a carbon-mass balance approach (Andreae and Merlet, 2001)

$$EF_i = \frac{m_i}{\Delta CO + \Delta CO_2 + \Delta CH_4 + \Delta NMOGs + \Delta OC + \Delta BC} \times \cdot W_C \qquad \text{Equation (2)}$$

Here m_i refers to the mass concentration of species i. ΔCO, ΔCO_2, ΔCH_4, $\Delta NMOGs$, ΔOC, and ΔBC are the background-corrected carbon mass concentrations of carbon-containing species in the flue gas. OC is derived from the OM / OC ratio determined with high-resolution AMS analysis (Canagaratna et al., 2015). W_C is the carbon mass fraction of the burning fuel. The W_C in the fuel a constant average value of 0.46 for wood (Bertrand et al., 2017), 0.45 for straw (Li et al., 2007), 0.45 for cow dung (Font-Palma, 2019), and 0.49 for coal (Zhang et al., 2000) was assumed. Changes of W_C over the burning cycle are expected to be small compared to the variability of pollutant emissions.

2.4 Volatility calculation of gaseous organic compounds

The volatility (i.e. the saturation mass concentration, C^*) for individual organic compounds was calculated based on the number of oxygen, carbon, and nitrogen atoms in the compound using the approach by Li et al. (2016):

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$$log_{10}{}_{C}^{*} = (n_C^0 - n_C^i)b_C - n_O^i b_O - 2\frac{n_C^i n_O^i}{n_C^i + n_O^i} b_{CO} - n_N^i b_N \quad \text{Equation (3)}$$

where n_C^0 is the reference carbon number; n_C^i, n_O^i and n_N^i denote the numbers of carbon, oxygen, and nitrogen, respectively, in the compound. b_C, b_O and b_N are the contributions of each atom to $log_{10}{}_{C}^{*}$, respectively; and b_{CO} is the carbon-oxygen nonideality. The parameters used in this analysis are presented in Table S1. Most notably, the empirical approach used by Li et al. (2016) was derived with only a limited number of organonitrates, which could potentially introduce bias in estimating vapor pressure (Isaacman-Vanwertz and Aumont, 2021). To mitigate this bias, we modified the nitrogen coefficient for CHON formulas that can be forced to equal twice the negative of the oxygen atom (b_N = -2b_0).

2.5 Identification of characteristic compounds

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. To identify the characteristic compounds of emissions from different fuels, we implemented the Mann-Whitney U test (Mann and Whitney, 1947; Wilcoxon, 1945) in MATLAB®, which has been applied in the selection of aerosol markers (Zhang et al., 2023). It is a nonparametric test and is used for between-group comparisons when the dependent variable is ordinal or continuous and not assumed to follow a normal distribution.

This test takes two data samples as parameters, uses the ranks as a measure of central tendency, and then returns the test results with a *p*-value to indicate the statistical significance. When the *p*-value is lower than the significance level of 0.1 (a commonly used *p*-value to study statistical significance in atmospheric research), the median of the tested sample is significantly high or low in the two-tailed test. 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. 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.

The molecules from a specific class of fuel that satisfy the pairwise comparison test between one fuel, referred to as fuel *j*, and other types of fuel, were determined to be significantly high- or low-fraction ions in fuel *j*. These ions have the potential to serve as characteristic compounds for fuel *j*. We have calculated in addition the fold change (*FC*) of ion *i* in fuel *j* was calculated using Equation 4,

$$FC_{i,j} = \frac{f_{i,j}}{f_{i,other}} \quad \text{Equation (4)}$$

Where $f_{i,j}$ represents the fraction of ion *i* in the mass spectra profiles of fuel *j*, and $f_{i,other}$ represents the average fraction of ion *i* in the mass spectra from the other fuels.

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3 Results and discussion

3.1 Emission factors from solid-fuel combustion

The average EFs of CO, CO_2, NMOGs and PM in g/kg as well as the MCE values calculated for the 6 types of fuels, are shown in Table 1. Detailed EFs and MCE values for each experiment can be found in Table S2.

The average MCE values depend on the fuel types and combustion phases (flaming and smoldering). The lowest MCE values, 0.90, were observed during the smoldering phase of the stove-burning of beech logs, while the highest values, at 0.99 ± 0.02, were recorded during the flaming phase of the spruce and pine branches and needles open burning. In all experiments, the highest EFs for a single gas-phase species correspond to CO_2 (1136.2-1711.7 g/kg). Coal burning has the highest average CO EFs (40.6 ± 12.6 g kg^{-1}) and CO_2 EFs (1680.2 ± 32.7g kg^{-1})

Total NMOG EFs reported in Table 1 refer to species quantified using the Vocus. The average EFs of primary organic gases (in the range of 6.7 to 74.2 g kg^{-1}) and the standard deviation are calculated based on the average EFs for the repeatable experiments, which depend on the combustion phases and fuel types. Generally, lower MCE values correspond to higher NMOGs EFs within a given class of burning fuel (Figure S2a). For instance, smoldering beech logs resulted in significantly higher average NMOGs EFs (74.2 ± 42.9) compared to burning pine and spruce logs. Pine and spruce stove and open burning, dominated by the flaming phase (average MCE > 0.95), exhibited average NMOGs EFs of 44.9 ± 17.5 and 39.8 ± 11.4, respectively. 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 g/kg) (Hatch et al., 2017). Despite the slight difference in MCE for some experiments, the increasing EFs for NMOGs with at least six carbon atoms per molecule (⩾ C6) as proxy SOA precursors were observed with lower MCE (Figure S2b) (Bruns et al., 2016). Moreover, the EFs of these SOA precursors are much higher than the primary biomass-burning organic aerosol (BBOA), which suggests a higher potential for SOA formation. Notably, the emission of NMOGs from cow dung and coal was relatively low, at 6.7 ± 2.1 and 11.5 ± 2.6 g kg^{-1}, respectively. Our emission factors align well with previously reported volatile organic compound emission factors from bituminous coal combustion under similar conditions (range of 1.5 to 14.1 g/kg) reported by (Klein et al., 2018).

3.2 Chemical composition of primary organic vapors

3.2.1 Overview of the measurements

To assess the feasibility of distinguishing differences between combustion fuel sources based on the measured species, we evaluated the similarity of the mass spectra obtained from each experiment using the correlation coefficient (*r*), as shown in Figure 1. Primary NMOGs from the same burning fuel are strongly correlated (0.82-0.99), indicating the general repeatability of the experiments. Furthermore, we observed a weak intra-fuel correlation between coal and other biomass sources (0.44-0.78), suggesting significant differences in chemical composition. By contrast, the separation between different biomass samples is not stark and all possess a correlation between 0.6-0.98. We also note the slight difference in composition between smoldering and flaming beech wood.

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Figure 1 also shows the average mixing ratio contribution of full ignition duration from *m/z* 40 to 300 for each experiment, and is categorized into C_xH_y, $C_xH_yO_z$, C_xH_yN and $C_xH_yO_zN$ families based on their elemental composition. In all primary NMOGs, the $C_xH_yO_z$ family is the most abundant group, making the largest contribution to beech logs (88.6%), pine and spruces logs (82.1%) and straw (81.7%). These percentages are higher than those for coal (63.1%) and cow dung (68.9%). Coal burning results in considerably higher contributions in the C_xH_y families (33.7%) than beech logs (9.3%), consistent with the bulk chemical composition of the fuels.

Figure 2 separates emitted compounds based on their carbon (x-axis) numbers. The dominant signals in primary organic gases for all fuels are attributed to C3-6 compounds, while more species with higher carbon numbers (#C > 10) are observed in pine and spruce branches and needles open burning. The bin containing H/C ratios between 1.2 and 1.5 has the largest contribution in all fuels except the straw, ranging from 27% to 31.2%. O/C ratios less than 0.15 contribute significantly to coal burning (42%), which corresponds to the high proportion of C_xH_y families (Figure 1). Wood and straw burning emitted more oxygenated organic vapors than coal and cow dung burning with more contribution of higher O/C species (O/C> 0.5). The results show similarities to the comparison between burning wood and cow dung in the particle phase. Specifically, cow dung exhibits a slightly lower fraction of high O/C compared to other fuels studied. (Zhang et al., 2023).

Generally, the total fraction of nitrogen-containing species (C_xH_yN and $C_xH_yO_zN$) is significantly higher in the primary NMOGs emitted from open burning of cow dung (18.8%) compared to the other fuels (2.1% to 7.3%). This trend is consistent with both our results from aerosol composition measurement and previous literature (Stewart et al., 2021b; Zhang et al., 2023; Loebel Roson et al., 2021). Generally, nitrogen containing compounds in cow dung consist mainly of one nitrogen atom and have a wide range of carbon numbers between 2 and 7 (Figure 2). Stewart et al. (2021a) also reported that cow dung was the largest emitter of nitrogen-containing NMOGs than other fuelwood and crops in India, releasing large amounts of acetonitrile and nitriles. These nitrogen-containing organic gases are likely formed from the volatilization and decomposition of nitrogen-containing compounds within the cow dung cake, such as free amino acids, pyrroline, pyridine, and chlorophyll (Ren and Zhao, 2015; Burling et al., 2010).

We categorized NMOGs by functional groups into 10 classes, including alcohols, carbonyls (including acid), hydrocarbons, furans, N-containing compounds, O-containing < 6 (where the number of carbon atoms is less than 6), O-containing ≥ 6 (where the number of carbon atoms is equal or greater than 6), oxygenated aromatics, polycyclic aromatic hydrocarbons (PAHs), single-ring aromatics (SRA) (Bhattu et al., 2019). Figure S3 and Figure S4 show a comparison of the organic gas composition observed from different fuel types. The measured emissions exhibit significantly different compositions, reflecting the variability of organic components produced from different fuel types. The emissions of all solid fuels are overwhelmingly dominated by carbonyls in the range of 23.1% (coal) to 45.1% (straw). For all emissions, furans represent the second largest group, which account for more than 14% of the emissions. Comparatively, aromatic compounds are less significant in biomass burning: 5.9% - 12% for oxygenated aromatics, 0.5% - 2.1% for PAHs, and 2.1% - 5.8% for SRA. In contrast, aromatic emissions are relatively larger in coal burning emissions (13.6%, 8.1%, and 13.8%, respectively).

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3.2.2 Volatility of organic compounds

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, the gaseous organic compounds were grouped into a 14-bin volatility basis set (Donahue et al., 2006) (Figure 3). We choose to classify the volatility in four main classes with units of μg 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 $\log_{10}(C^*) < -0.5$).

Comparison and compilation of gaseous biomass burning emissions sorted by volatility and functional group are shown in Figure 3, and the distribution of average EFs as a function of binned saturation vapor concentration is shown. The VOC class was found to be the most abundant, ranging from 58.7% to 87% (Figure S5). For all burns, carbonyls, furans, and SRA families are overwhelmingly dominant in VOCs, accounting for more than 60% of the VOC emissions. The high fraction of oxygenated VOCs like carbonyls in biomass burning emissions is in stark contrast to VOCs emitted from coal combustion, which is dominated by aromatic hydrocarbon emissions, particularly PAHs. This difference may be attributed to the condensed structure of coal. PAHs are a group of organic matter compounds containing multiple aromatic rings that mainly result from incomplete combustion (Mastral and Callen, 2000).

IVOCs also constituted a considerable fraction in solid-fuel combustions (from 12.6% to 39.3%), particularly in spruce and pine branches and needles (39.3%), cow dung (24.3%) and coal (31.1%) (Figure S5). Significant differences in the bulk volatility of organic compounds were observed among different types of wood burning. 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. This difference may be attributed to a lower percentage of terpenes in woody tissues compared to needle/leaf tissues (Greenberg et al., 2006). In addition, open burning provides more oxygen than stove burning, which enhances the formation of partially oxidized organic compounds. Within the open burning experiments, the oxygenated molecules (both aromatics and $C \geq 6$) are enhanced relative to the other experiments and result in the largest emission factors of IVOCs. In addition to the burning conditions, the fuel properties are also an important factor affecting the IVOC component. Notably, cow dung comprised a higher fraction of N-containing species within their IVOC emissions compared to other fuels. The emission in volatility bins in this study is important for the modeling of organics with the VBS scheme.

3.3 Comparison between flaming and smoldering of wood burning

Figure 4a shows a typical burning cycle during beech log wood experiments with distinct emission characteristics between flaming and smoldering phases. In the top panel, the MCE is used to indicate the flaming stage with a significant CO_2 enhancement, while the smoldering stage exhibits high levels of CO. The flaming phase shows considerable BC emission, while the smoldering phase is dominated by OA emissions without visible flame. The AAE during the smoldering phase is approximately twice that of the flaming phase, possibly due to the presence of "brown carbon" in organic aerosols. *f*60 represents the prevalence of primary combustion products such as levoglucosan and is used as an indicator for fresh BB emissions (Schneider et al., 2006; Alfarra et al., 2007). During the starting/flaming phase, when the temperature is higher, *f*60 increases, whereas for lower temperatures

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in the smoldering phase, *f*60 decreases (Weimer et al., 2008). The mixing ratio of most of the compounds correlates negatively with the MCE as expected with a significant increase in the smoldering phase (Figure 4a and Figure S6). However, some compounds like benzene have different enhancement rates from flaming to smoldering, which is similar to previous studies (Warneke et al., 2011).

Figure 4b illustrates the measured emission factors for flaming and smoldering wood fire stages. On average, emission factors for organic gases in the flaming stage are approximately four times lower (31.4 g/kg) than those in the smoldering stage fires (121.9 g/kg). Despite significant variability in organic gas emission strengths, the relative contribution of different classes showed large similarities among combustion phases (Figure S7). Hardwood (beech) is a fibrous substance primarily composed of three chemical elements: carbon, hydrogen, and oxygen and these basic elements are incorporated into several organic compounds, i.e. cellulose, hemicellulose, lignin, and extractives formed into a cellular structure (Asif, 2009). The flaming stage is associated with more complete oxidation with a relatively higher contribution of OVOCs (e.g., furan, oxygenated aromatics, O-containing, Figure S8). Conversely, during the smoldering stage, more CO and organic gases are emitted relative to the flaming stage (Figure 4a). OVOCs, such as carbonyl, furan, oxygenated aromatics, and O-containing species, form the major fraction (> 88%) of emissions in both flaming and smoldering fires. They are followed by the sum of C_xH_y, and SRA (5-10%). As shown in Figure S9, the volatility distribution of beech log stove burning between the flaming phase and smoldering phase is also similar, only with a relatively higher contribution of carbonyls and C_xH_y in the smoldering phase than flaming phase. This suggests that the emission factor plays a more important role than the chemical composition in differentiating between flaming and smoldering fires.

3.4 Common compounds emission for biomass-burning combustion and identification of characteristic compounds for different fuels

3.4.1 Common compounds for biomass burning

To conduct a comprehensive analysis aimed at identifying characteristic compounds among emissions, the Mann-Whitney U test (refer to Sect. 2.5) was performed on the relative contribution of primary organic gases derived from various fuels as measured by Vocus. The results of the pairwise Mann-Whitney test are presented in Figure S10, illustrating the average $-\log_{10}$ of the *p*-value as a function of the $\log_2$ of the fold change (*FC*). Species that yield *p*-values lower than 0.1 in the two-tailed test for all pairwise comparisons are deemed significantly more abundant or scarce in a particular fuel type compared to all other fuels. These species are indicated as colored circles in Figure 5. In cases where species do not meet this criterion once or multiple times, they are represented as gray circles, even if their average *p*-value falls below 0.1. A higher $-\log_{10}$ (*p*-value) signifies a reduced likelihood that the fractional medians of two species are equivalent. Simultaneously, a greater *FC* (as per Equation 4) indicates an increased presence of the species' fractional contribution in the tested fuel in comparison to the average contribution across all other fuels. This suggests a higher degree of exclusivity for this species in the given context. The selected characteristic compounds, *p*-values, fold changes, and threshold results are listed in the Supplement Table.

As shown in Figure 1, biomass fuels (wood, straw and cow dung) are different fuels from coal in this study. To address this, we characterized the common compounds from varied biomass fuels by

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establishing a threshold (relative mixing ratio contribution ⩾ 0.1%) for compounds that are not specific characteristic compounds of biomass burning, which allowed for the identification of compounds that are more readily detectable in intricate environments. As shown in Figure S11, the gas-phase analysis revealed several dominant species: $C_5H_4O_2$ (furfural, 2.2-10.1%), $C_2H_4O_2$ (acetic acid, 2.1-5.8%), $C_3H_6O_2$ (methyl acetate, 1.7-4.6%), and C_2H_4O (acetaldehyde, 1.3-3.9%), which were also reported prior studies on biomass burning (Bruns et al., 2017; Stockwell et al., 2015; Christian, 2004; Sarkar et al., 2016). Furthermore, the category of common compounds represents the primary set of compounds associated with biomass burning, contributing from 46% to 69% of the emissions (Figure S12). Carter et al. (2022) expand the representation of fire NMOGs in a global chemical transport model, GEOS-Chem, which contributes substantially to atmospheric reactivity, both locally and globally. Our results could provide more input information for global or regional chemistry transport models.

3.4.2 Characteristic compounds for solid-fuel combustion

Mass defect plots of characteristic compounds are visualized in Figure 5, for each burning source, respectively. Many characteristic compounds are identified for each unique type of burning (Supplement Table). As shown in Figure 5, characteristic compounds of all wood burning are mainly composed of compounds from the C_xH_y and $C_xH_yO_z$-family. However, the selected characteristic compounds for spruce and pine branches and needles have higher molecular weights and are more oxidized, which aligns with their characteristics of the mass spectrum. In contrast, compounds from open burning of straw and cow dung contribute considerably more to nitrogen-containing families but less to oxygen-containing species, consistent with their bulk chemical composition characteristics. Additionally, characteristic compounds for coal consist mainly of compounds from C_xH_y-family, which also aligns with its bulk chemical composition and relatively higher H/C ratios (Figure 2).

For all softwood (i.e., spruce and pine logs and spruce and pine branches and needles in this study), monoterpenes ($C_{10}H_{16}$) are a characteristic compound along with the fragment at *m/z* 81.07 (C_6H_8). However, monoterpenes cannot exclusively be related to biomass burning given their abundance in the atmosphere. Monoterpenes are also the biogenic volatile organic compounds (BVOCs) emitted from natural trees and other vegetation (Hellén et al., 2012). However, the emission rates of terpenes vary with season, with a higher rate in spring and summer and a lower rate in autumn and winter (He et al., 2000; Noe et al., 2012). In winter, monoterpenes could be a characteristic compound for softwood burning due to minor natural emissions from spruce, but in summer, terpene emissions from natural trees would dominate the contribution making it a non-characteristic compound. P-cumenol ($C_9H_{12}O$), as one of the characteristic compounds for beech logs, was discovered to be one of the prominent products of beech wood pyrolysis of lignins (Sengpiel et al., 2019; Keller et al., 2020). Homologues of $C_{10}H_{8\text{-}18}O_2$ are determined for spruce and pine branches and needles, with $C_{10}H_{10}O_2$ being β-phenylacrylic acid, which is one of the main chemical compositions of the extract of the cedar pine needle. $C_{10}H_{14}O_2$ could be 1-guaiacylpropane, which is proposed as a characteristic compound for coniferyl-type lignin pyrolysis products from pine (Simoneit et al., 1993; Liu et al., 2021). Homologues of $C_{11}H_{8\text{-}18}O_2$ are also seen, for example, $C_{11}H_{14}O_2$, likely 1-(3,4-dimethoxy-phenyl)-1-propene, which is stated as a representative compound found in lignin (Alves et al., 2003; Hill Bembenic, 2011).

Noticeably, cow dung has a significantly different chemical composition. As a result, many characteristic compounds are identified from the burning of cow dung compared to other fuels. These

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characteristic compounds predominantly contain nitrogen in chemical composition and overlap all characteristic compounds for straw, owing to the mixture of dried cow dung and crop residues in "cow dung cakes." Many nitrogen-containing characteristic compounds are found in straw and cow dung, such as C_4H_5N, C_5H_5N, C_5H_7N, and C_6H_7N, which could likely be assigned to pyrrole, pyridine, methylpyrrole and methyl pyridines respectively. Pyrolysis of the constituents in the crop residue is a probable pathway for these compounds (Ma and Hays, 2008). Acetonitrile (C_2H_3N), acrylonitrile (C_3H_3N), propanenitrile (C_3H_5NO), and 4-methylpentanenitrile ($C_6H_{11}N$) were found as characteristics compounds for cow dung with generally higher FC and higher relative contribution. Additionally, several series of nitrogen-containing homologues are found, such as $C_{10}H_{11\text{-}21}NO$, $C_{12}H_{11\text{-}21}N$, $C_{11}H_{11\text{-}23}NO$ and $C_{15}H_{15\text{-}31}N$. These nitrogen-containing gases have also been detected, especially in emissions from cow dung cake in India compared to fuelwood and are likely formed from the volatilization and decomposition of nitrogen-containing compounds within the cow dung cake. These compounds originate primarily from free amino acids but can also arise from pyrroline, pyridine, and chlorophyll (Stewart et al., 2021a).

Coal is also a distinct solid fuel compared to other biomass fuels in this study, showing a relatively lower correlation coefficient (Figure 1). Consequently, many series of C_xH_y-family homologues are found. Compounds with 9-12 carbon atoms, as shown in Figure 5 for coal burning, could be PAHs-related, such as C_9H_8 (1-Indene), $C_{10}H_8$ (naphthalene), $C_{10}H_{10}$ (1-methylnapthalene), $C_{12}H_{10}$ (acenaphthene), $C_{12}H_{12}$ (2,6-dimethylnaphthalene). The EFs of the characteristic compounds also show an increasing trend with the decrease of MCE (Figure S13), which suggests EFs of the characteristic compounds are not only dependent upon the type of fuel burnt but also on the burning condition.

4 Conclusions

In this study, we first investigated the real-time emissions of primary organic gases using Vocus-PTR-TOF during typical 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). Average emission factors of CO, CO_2, primary organic gases and PM were calculated. This work provides a comprehensive laboratory-based analysis of the chemical composition of organic gases emitted from the different combustibles and different combustion phases. We discuss the prominent net combustion emissions from biomass burning and identify new characteristic compounds using the Mann-Whitney U test.

The results indicate that wood burning has higher primary organic gas emission factors compared to other fuels. The emissions varied significantly, ranging from 6.7 to 74.2 g kg^{-1}, depending on the combustion phases and fuel types. Despite the slight difference in modified combustion efficiency (MCE) for some experiments, the increasing EFs for primary organic gases were observed with lower MCE. Moreover, the EFs of these SOA precursors are much higher than the primary biomass-burning organic aerosol (BBOA), which suggests a higher potential for SOA formation. The $C_xH_yO_z$-family is the most abundant group (63.1% to 88.6%) for all solid fuels, followed by C_xH_y (9.3% to 33.7%). A larger contribution of nitrogen-containing species (C_xH_yN and $C_xH_yO_zN$) is found in cow dung burning, while coal burning has a higher contribution from the C_xH_y families. Moreover, the VOC class was found to be the most abundant (58.7% to 87%), followed by the IVOC class (12.6% to 39.3%). Primary semivolatile/intermediate-volatility organic compounds (S/IVOCs) have been proposed as important

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SOA precursors from biomass burning. Li et al. (2024) found that IVOCs from residential wood burning (~ 13% of total organic gases) can contribute ~70% of the formed SOA. Overall, these data will help update the IVOC emission inventory and improve the estimates of SOA production. Specifically, these results demonstrate that open burning (e.g., wildfire) emissions have enhanced IVOC emission factors, suggesting that the SOA potential from open-burning sources is larger in comparison to their wood stove counterparts.

Distinct particulate/gas emissions at different combustion phases are observed for stove burning of beech logs: initial compositions of flaming or smoldering plumes were dominated by BC or OA, respectively, with much higher primary organic gas emission in smoldering. Despite the large variability in organic gas emission strengths, the relative contribution of different classes showed large similarities among combustion phases. Therefore, the enhanced emission factor under smoldering conditions means there is a greater potential for SOA formation when compared to flaming conditions.

However, each source generally emits a wide spectrum of organic gas species, leading to considerable overlap between organic gas species from different sources. Based on the Mann-Whitney U, we selected species that were unique in certain emissions as possible characteristics compounds for the characteristic compounds and the common compounds for all biomass fuels. Due to minor natural emissions from spruce in summer, monoterpene ($C_{10}H_{16}$) and its fragment could be characteristic compounds for all softwoods (i.e., pine logs and spruce and pine branches and needles in this study) in winter. More products of the pyrolysis of coniferyl-type lignin and the cedar pine needle extract could be found in spruce and pine branches and needles open burning (e.g., $C_{10}H_{14}O_2$, $C_{11}H_{14}O_2$, $C_{10}H_{10}O_2$). The prominent product ($C_9H_{12}O$) resulting from the pyrolysis of beech lignin is identified as the characteristic compound for beech log stove burning. Many series of nitrogen-containing homologues and nitrogen-containing species (e.g., acetonitrile, acrylonitrile, propanenitrile, methylpentanenitrile) are identified (e.g., $C_{10}H_{11\text{-}21}NO$, $C_{12}H_{11\text{-}21}N$, $C_{11}H_{11\text{-}23}NO$ and $C_{15}H_{15\text{-}31}N$), particularly from open burning of cow dung. Coal is a unique solid fuel compared to biomass and more PAHs-related characteristic compounds are identified from coal burning with 9-12 carbon. These characteristic compounds provide important support for future global or regional chemistry transport modeling and source apportionment. Overall, our study provides a comprehensive understanding of the molecular composition and volatility of primary organic compounds, as well as new insights into the identification of characteristic compounds from the burning of solid fuels.

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Tables and figures

Table 1 Average emission factors of CO, CO_2, Organic gases and PM as well as MCE for 6 types of burning.

Burning type	Carbon content	MCE	Emission factors (g kg^{-1} fuel)			
			CO	CO_2	NMOGs	PM
beech logs stove (n=6)	0.46	0.96 ± 0.03	38.9 ± 25.9	1409.4 ± 177.1	74.2 ± 42.9	2.5 ± 1.7
spruce and pine logs stove (n=5)	0.46	0.97 ± 0.01	28.5 ± 14.3	1511.7 ± 68.5	44.9 ± 17.5	1 ± 0.6
spruce and pine branches and needles open (n=3)	0.46	0.99 ± 0.001	2.8 ± 0.8	1579.2 ± 29.7	39.8 ± 11.4	0.9 ± 0.4
straw open (n=4)	0.45	0.97 ± 0.01	24.4 ± 6.6	1488.4 ± 87.2	42.6 ± 33.7	2.8 ± 0.7
cow dung open (n=5)	0.45	0.98 ± 0.01	21.6 ± 14.5	1583.8 ± 37.8	6.7 ± 2.1	1.6 ± 1
coal stove (n=5)	0.49	0.96 ± 0.01	40.6 ± 12.6	1680.2 ± 32.7	11.5 ± 2.6	0.9 ± 0.3

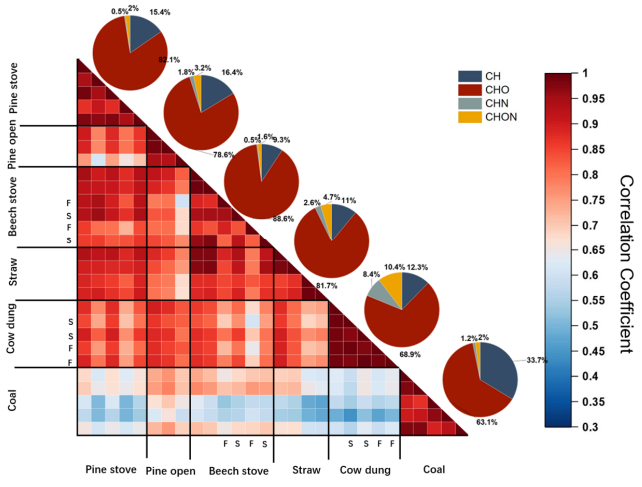

Figure 1. The correlation matrix of primary organic gases measured with Vocus (F represents flaming phase and S represents smoldering phase and unmarked columns and rows represent mixtures of both flaming and smoldering phases). Pie charts showing the contribution of elemental families are on the diagonal.

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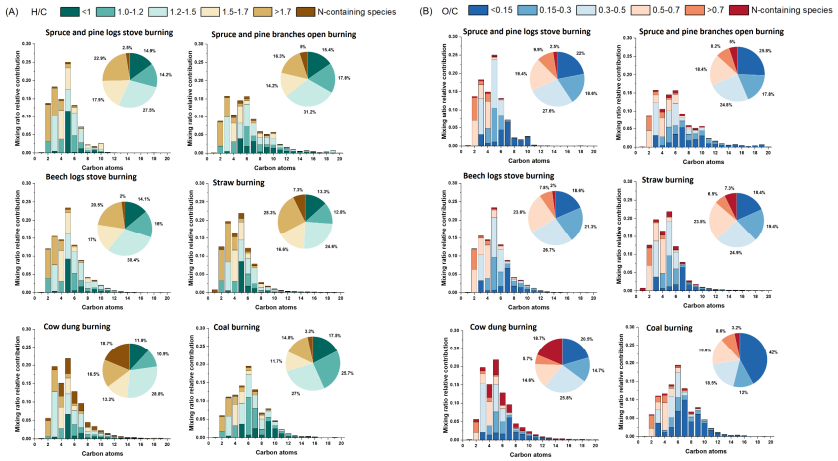

Figure 2. The average carbon distribution is colored by the H/C (**A**) and O/C (**B**) for non-N-containing species. The pie charts are the corresponding contribution of a range of H/C or O/C ratios.

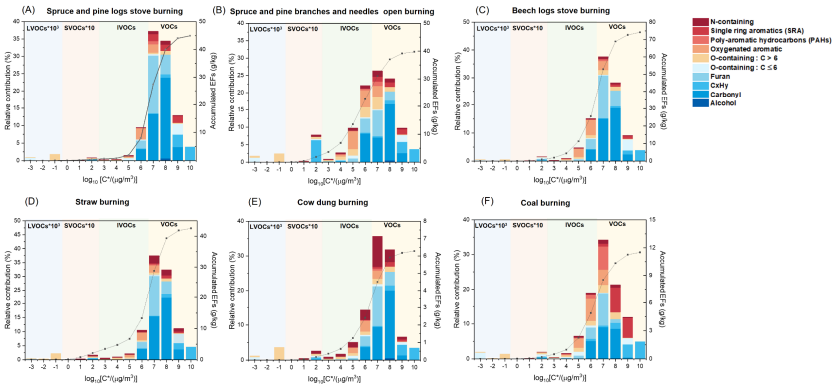

Figure 3. Volatility and average accumulated emission factors (assume the average molecular weight of each bin are same) the distribution of primary emissions as a function of binned saturation vapor concentration. Shaded areas indicate the volatility ranges with units of μg m^{-3}: VOCs (yellow) as $\log_{10}(C^*) > 6.5$, IVOCs (blue) as $\log_{10}(C^*)$ between 6.5 to 2.5, semi-VOCs (SVOCs, green) as $\log_{10}(C^*)$ between 2.5 to - 0.5 and low-VOCs (LVOCs, orange) as $\log_{10}(C^*) < -0.5$). The relative contribution of LVOCs and SVOCs are multiplied by a factor of 1000 and 10, respectively.

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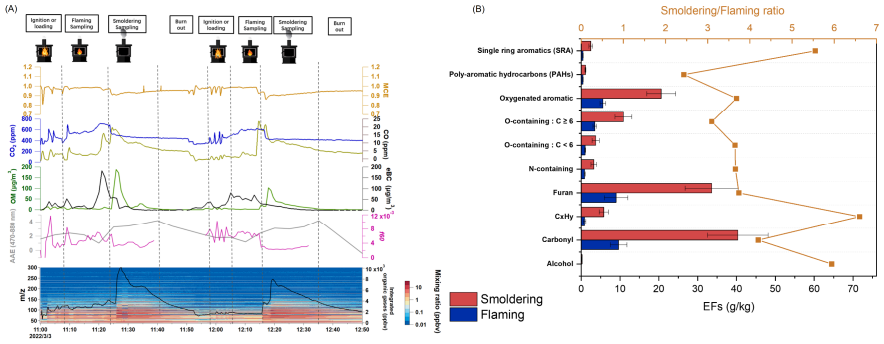

Figure 4. (A) Temporal profiles of mixing ratios measured by Vocus and evolution of CO, CO_2, AAE, *f*60, MCE and key aerosol compositions during burning cycles of beech logs stove burning (B) Geometric mean of the primary emission factors for gas-phase species of different functional groups during flaming 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_2 was observed from the flaming phase to the smoldering phase.

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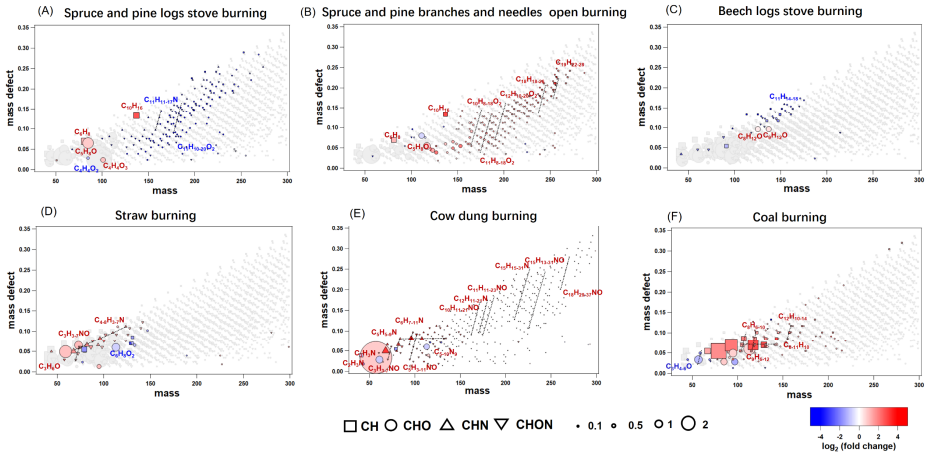

Figure 5. Mass defect plots identifying characteristic compounds sized by the square root of fractional contribution (%) and colored by $\log_2$ (the fold change). The dashed line represents the series of homologues.

Data availability

The data presented in the text and figures are available in the Zenodo online repository (https://doi.org/10.5281/zenodo.10804011).

Author contributions

TTW, JZ, HL, KL, RKYC, EG, LK, DMB, and RLM conducted the burning experiments. TTW analyzed the data and wrote the paper. MB, ZCJD, LK, DMB, KL, RLM, IEH, HL, JGS, and ASHP participated in the interpretation of data.

Competing interests

The authors declare that they have no conflict of interest.

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