

# 1 Emissions of non-methane volatile organic compounds from combustion of domestic fuels 2 in Delhi, India

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## 18 Abstract

19 29 different fuel types used in residential dwellings in northern India were collected from  
20 across Delhi (76 samples in total). Emission factors of a wide range of non-methane volatile  
21 organic compounds (NMVOCs) (192 compounds in total) were measured during controlled  
22 burning experiments using dual-channel gas chromatography with flame ionisation detection  
23 (DC-GD-FID), two-dimensional gas chromatography (GC×GC-FID), proton-transfer-reaction  
24 time-of-flight mass spectrometry (PTR-ToF-MS) and solid-phase extraction two-dimensional  
25 gas chromatography with time-of-flight mass spectrometry (SPE-GC×GC-ToF-MS). 94%  
26 speciation of total measured NMVOC emissions was achieved on average across all fuel types.  
27 The largest contributors to emissions from most fuel types were small non-aromatic  
28 oxygenated species, phenolics and furanics. The emission factors (in g kg<sup>-1</sup>) for total gas-phase  
29 NMVOCs were fuel wood (18.7, 4.3-96.7), cow dung cake (62.0, 35.3-83.0), crop residue  
30 (37.9, 8.9-73.8), charcoal (5.4, 2.4-7.9), sawdust (72.4, 28.6-115.5), municipal solid waste  
31 (87.3, 56.6-119.1) and liquefied petroleum gas (5.7, 1.9-9.8).

32 The emission factors measured in this study allow for better characterisation, evaluation and  
33 understanding of the air quality impacts of residential solid fuel combustion in India.

## 34 **1. Introduction**

35 Biomass burning is the second largest source of trace gases to the troposphere, releasing around  
36 a half of global CO, ~ 20% of NO and ~ 8% of CO<sub>2</sub> emissions (Olivier et al., 2005; Wiedinmyer  
37 et al., 2011; Andreae, 2019). Biomass burning releases an estimated 62 Tg yr<sup>-1</sup> of non-methane  
38 volatile organic compounds (NMVOCs) (Andreae, 2019) and is the dominant source of both  
39 black carbon (BC) and primary organic aerosol (POA), representing 59% and 85% of global  
40 emissions respectively (Bond et al., 2013). Biomass burning includes open vegetation fires in  
41 forests, savannahs, agricultural burning and peatlands (Chen et al., 2017) as well as the biofuels  
42 used by approximately 3 billion people to meet their daily cooking and heating energy  
43 requirements worldwide (World Bank, 2017). A wide range of trace gases are released from  
44 biomass burning, in different amounts depending on the fuel type and the combustion  
45 conditions, meaning that detailed studies at the point of emission are required to accurately  
46 characterise emissions. The gases released lead to soil-nutrient redistribution (Ponette-  
47 Gonzalez et al., 2016; N'Dri et al., 2019), can themselves be toxic (Naeher et al., 2007) and can  
48 significantly degrade local, regional and global air quality through the photochemical  
49 formation of secondary pollutants such as ozone (O<sub>3</sub>) (Pfister et al., 2008; Jaffe and Wigder,  
50 2012) and secondary organic aerosol (SOA) (Alvarado et al., 2015; Kroll and Seinfeld, 2008).  
51 They can also lead to indoor air quality issues (Fullerton et al., 2008).

52 Emissions from biomass burning and their spatial distribution remain uncertain and estimates  
53 by satellite retrieval vary by over a factor of three (Andreae, 2019). Bottom-up approaches use  
54 information about emission factors and fuel usage. However, information for many developing  
55 countries, where solid fuel is a primary energy source, is particularly sparse. Toxic pollution  
56 from burning has been linked to chronic bronchitis (Akhtar et al., 2007; Moran-Mendoza et al.,  
57 2008), chronic obstructive pulmonary disease (Dennis et al., 1996; Orozco-Levi et al., 2006;  
58 Rinne et al., 2006; Ramirez-Venegas et al., 2006; Liu et al., 2007; PerezPadilla et al., 1996),  
59 lung cancer (Liu et al., 1993; Ko et al., 1997), childhood pneumonia (Smith et al., 2011), acute  
60 lower respiratory infections (Bautista et al., 2009; Mishra, 2003) and low birth weight of  
61 children (Boy et al., 2002; Yucra et al., 2011). Smoke from inefficient combustion of domestic  
62 solid fuels is the leading cause of conjunctivitis in developing countries (West et al., 2013).  
63 The harmful emissions from burning also resulted in an estimated 2.8-3.9 million premature  
64 deaths due to household air pollution (Kodros et al., 2018; World Health Organisation, 2018;  
65 Smith et al., 2014), of which 27% originated from pneumonia, 18% from strokes, 27% from  
66 ischaemic heart disease, 20% from chronic obstructive pulmonary disease and 8% from lung

67 cancer, with hazardous indoor air pollution responsible for 45% of pneumonia deaths in  
68 children less than 5 years old (World Health Organisation, 2018). For this reason, hazardous  
69 indoor air pollution from the combustion of solid fuels has been calculated to be the most  
70 important risk factor for the burden of disease in South Asia from a range of 67 environmental  
71 and lifestyle risks (Lim et al., 2012; Smith et al., 2014).

72 The emissions from biomass burning fires are complex and can contain many hundreds to  
73 thousands of chemical species (Crutzen et al., 1979; McDonald et al., 2000; Hays et al., 2002;  
74 Hatch et al., 2018; Stewart et al., 2020a). Measurements of emissions by gas chromatography  
75 (GC) have been made (EPA, 2000; Wang et al., 2014; Gilman et al., 2015; Stockwell et al.,  
76 2016; Fleming et al., 2018), as it has the potential to provide isomeric speciation of emissions.  
77 However, it is of limited use in untargeted measurements from burning due to the complexity  
78 of emissions, leading to large amounts of NMVOCs released not being observed. Some of the  
79 main issues are that GC does not provide high time resolution measurements and several  
80 instruments with different column configurations and detectors are required to provide  
81 information on different chemical classes. Samples can also be collected into canisters or  
82 sample bags and then analysed off-line (Wang et al., 2014; Sirithian et al., 2018; Barabad et  
83 al., 2018), which can increase time resolution, but can also lead to artefacts (Lerner et al., 2017).

84 Recent developments have allowed the application of proton-transfer-reaction mass  
85 spectrometry (PTR-MS) to study the emissions from biomass burning (Warneke et al., 2011;  
86 Yokelson et al., 2013; Brilli et al., 2014; Stockwell et al., 2015; Bruns et al., 2016; Koss et al.,  
87 2018). PTR-MS uses proton transfer from the hydronium ion ( $\text{H}_3\text{O}^+$ ) to ionise and  
88 simultaneously detect most polar and unsaturated NMVOCs including aromatics, oxygenated  
89 aromatics, alkenes, furanics and nitrogen containing volatile organic compounds in gas  
90 samples. PTR-MS can measure at fast acquisition rates of up to 10 Hz over a mass range of 10  
91 – 500 Th with very low detection limits of tens to hundreds of pptv (Yuan et al., 2016). The  
92 more recently-developed technique of proton-transfer-reaction time-of-flight mass  
93 spectrometry (PTR-ToF-MS) has allowed around 90% of total measured NMVOC emissions  
94 in terms of mixing ratio from burning experiments to be speciated (Koss et al., 2018) and has  
95 also been used to study the formation of SOA (Bruns et al., 2016). The main disadvantages of  
96 the PTR-ToF-MS technique are its inability to speciate isomers, significant fragmentation of  
97 parent ions, only being able to detect species with a proton affinity greater than water and the  
98 formation of water clusters needing to be considered (Stockwell et al., 2015; Yuan et al., 2017).  
99 More recently, measurements have also been made using iodide chemical ionization time-of-

100 flight mass spectrometry ( $\Gamma$ -CIMS), which is well suited to measuring acids and  
101 multifunctional oxygenates (Lee et al., 2014) as well as isocyanates, amides and organo-nitrate  
102 species released from biomass burning (Priestley et al., 2018). Multiple measurement  
103 techniques used in concert are therefore complementary, with the use of PTR-ToF-MS and  
104 simultaneous gas chromatography often alleviating some of the difficulties highlighted above.

105 Since the start of the century, rapid growth has resulted in India becoming the second largest  
106 contributor to NMVOC emissions in Asia (Kurokawa et al., 2013; Kurokawa and Ohara, 2019).  
107 However, effective understanding of the relative strength of different sources and subsequent  
108 mitigation has been limited by a deficiency of suitably detailed, spatially disaggregated  
109 emission inventories (Garaga et al., 2018). A current receptor-model study has shown elevated  
110 NMVOC concentrations at an urban site in Delhi to be predominantly due to vehicular  
111 emissions, with a smaller contribution from solid fuel combustion (Stewart et al., 2020b).  
112 However, approximately 60% of total NMVOC emissions from India in 2010 were estimated  
113 to be due to solid fuel combustion (Sharma et al., 2015). Other studies have also suggested that  
114 burning may lead to enhanced concentrations of pollutants such as polycyclic aromatic  
115 hydrocarbons in Delhi (Elzein et al., 2020). A need has therefore been identified to measure  
116 local source profiles to allow evaluation with activity data to better understand the impact of  
117 unaccounted and unregulated local sources (Pant and Harrison, 2012).

118 Approximately 25% of worldwide residential solid fuel use takes place in India (World Bank,  
119 2020), with approximately 25% of ambient particulate matter in South Asia attributed to  
120 cooking emissions (Chafe et al., 2014). Despite large government schemes, traditional solid  
121 fuel cookstoves remain popular in India because they are cheaper than ones that use liquefied  
122 petroleum gas (LPG) and the meals cooked on them are perceived to be tastier (Mukhopadhyay  
123 et al., 2012). The total number of biofuel users has been sustained by an increasing population,  
124 despite the percentage use of biofuels decreasing as a proportion of overall fuel use due to  
125 increased LPG uptake (Pandey et al., 2014). Cow dung cakes remain prevalent as a fuel because  
126 they are cheap, readily available, sustainable and ease pressure on local fuel wood resources.  
127 Few studies have reported emissions data from cow dung cake (Venkataraman et al., 2010;  
128 Stockwell et al., 2016; Koss et al., 2018; Fleming et al., 2018), leaving considerable uncertainty  
129 over the impact that cow dung cake combustion has on air quality. LPG usage has increased  
130 from around 100 to 500 million users over the same period, but only reflects around 10% of  
131 current rural fuel consumption (Gould and Urpelainen, 2018).

132 India-specific inventories which include residential burning indicate a considerable emission  
133 source of total NMVOCs of around 6000-7000 kt yr<sup>-1</sup> (Pandey et al., 2014; Sharma et al., 2015).  
134 Burning is likely to have a large impact on air quality in India, but considerable uncertainties  
135 exist over the total amount of NMVOCs released owing to a lack of India specific emission  
136 factors and information related to the spatial distribution of emissions.

137 Few studies exist measuring highly speciated NMVOC emission factors from fuels specific to  
138 India. Recent studies using PTR-ToF-MS to develop emission factors, which are more  
139 reflective of the range of species emitted from burning, have focussed largely on grasses, crop  
140 residues and peat (Stockwell et al., 2015) as well as fuels characteristic of the western U.S.  
141 (Koss et al., 2018). A previous study measured emission factors of NMVOCs from cow dung  
142 cake using gas chromatography with flame ionisation detection (GC-FID) of 8-32 g kg<sup>-1</sup> (EPA,  
143 2000). Fleming et al. (2018) quantified 76 NMVOCs from fuel wood and cow dung cake  
144 combustion using *chulha* and *angithi* stoves by collecting samples into Kynar bags, transferring  
145 their contents into canisters and off-line analysis using GC-FID, GC-ECD (electron capture  
146 detector) and GC-MS. The emission factors measured from these 76 NMVOCs were 14 g kg<sup>-1</sup>  
147 for cow dung cake burnt in *chulha* stoves, 27 g kg<sup>-1</sup> for cow dung cake burnt in *angithi* stoves  
148 and 6 g kg<sup>-1</sup> for fuel wood burnt in *angithi* stoves. An emission factor from one single dung  
149 burn measured using PTR-ToF-MS was considerably larger at around 66 g kg<sup>-1</sup> (Koss et al.,  
150 2018). Emissions from dung in Nepal have also been measured (Stockwell et al., 2016) by  
151 sampling into whole air sample canisters followed by off-line analysis with GC-FID/ECD/MS  
152 and Fourier-transform infrared spectroscopy (FTIR). However, very few speciated NMVOC  
153 measurements were made and the emission factors were similar to those measured using just  
154 GC (Fleming et al., 2018). Studies have also focussed on making detailed measurements, using  
155 a range of techniques, from the burning of municipal solid waste (Christian et al., 2010;  
156 Yokelson et al., 2011; Yokelson et al., 2013; Stockwell et al., 2015; Stockwell et al., 2016;  
157 Sharma et al., 2019) and crop residues (Stockwell et al., 2015; Koss et al., 2018; Kumar et al.,  
158 2018).

159 Detailed chemical characterisation of NMVOC emissions from fuel types widely used in the  
160 developing world is much needed to resolve uncertainties in emission inventories used in  
161 regional policy models and global chemical transport models. A greater understanding of the  
162 key sources is required to characterise and hence understand air quality issues to allow the  
163 development of effective mitigation strategies. In the present study we measure comprehensive  
164 emission factors of NMVOCs from a range solid fuels characteristic to northern India.

## 165 2. Methods

### 166 2.1 Fuel collection and burning facility

167 A total of 76 fuels, reflecting the range of fuel types used in northern India, were collected from  
168 across Delhi (see Figure 1 and Table 1). Cow dung cake usage was prominent in the north and  
169 west regions, whereas fuel wood use was more evenly spread across the state. Municipal solid  
170 waste was collected from Bhalaswa, Ghazipur and Okhla landfill sites. Collection also included  
171 less used local fuel types which were found being burnt including crop residues, sawdust and  
172 charcoal. A low-cost LPG stove, widely promoted across India as a cleaner fuel through  
173 government initiatives such as the Pradhan Mantri Ujjwala Yojana and Pratyaksh Hanstantrit  
174 Labh schemes, was used for direct emission comparison with other local fuel types.

175 Fuels were burnt at the CSIR-National Physical Laboratory (NPL), New Delhi, under  
176 controlled conditions utilizing a combustion chamber based on the design of Venkataraman  
177 and Rao, (2001). Several previous studies have been based on this chamber design  
178 (Venkataraman and Rao, 2001; Venkataraman et al., 2002; Saud et al., 2011; Saud et al., 2012;  
179 Singh et al., 2013), which was designed to simulate the convection-driven conditions of real-  
180 world combustion and is displayed in the Supplementary Information S1. The burn-cycle used  
181 in this study was adapted from the VITA water-boiling test, which was designed to simulate  
182 emissions from cooking and included emissions from both low- and high-temperature burning  
183 conditions. Fuels were collected and stored in the same manner as local customs using expert  
184 local judgement. This was designed to ensure that the moisture content of fuel wood samples  
185 was like those being burnt locally and that the combustion replicated real-world burning  
186 conditions encountered in local cooking practices, which should consequently give a more  
187 reflective NMVOC emission factor.

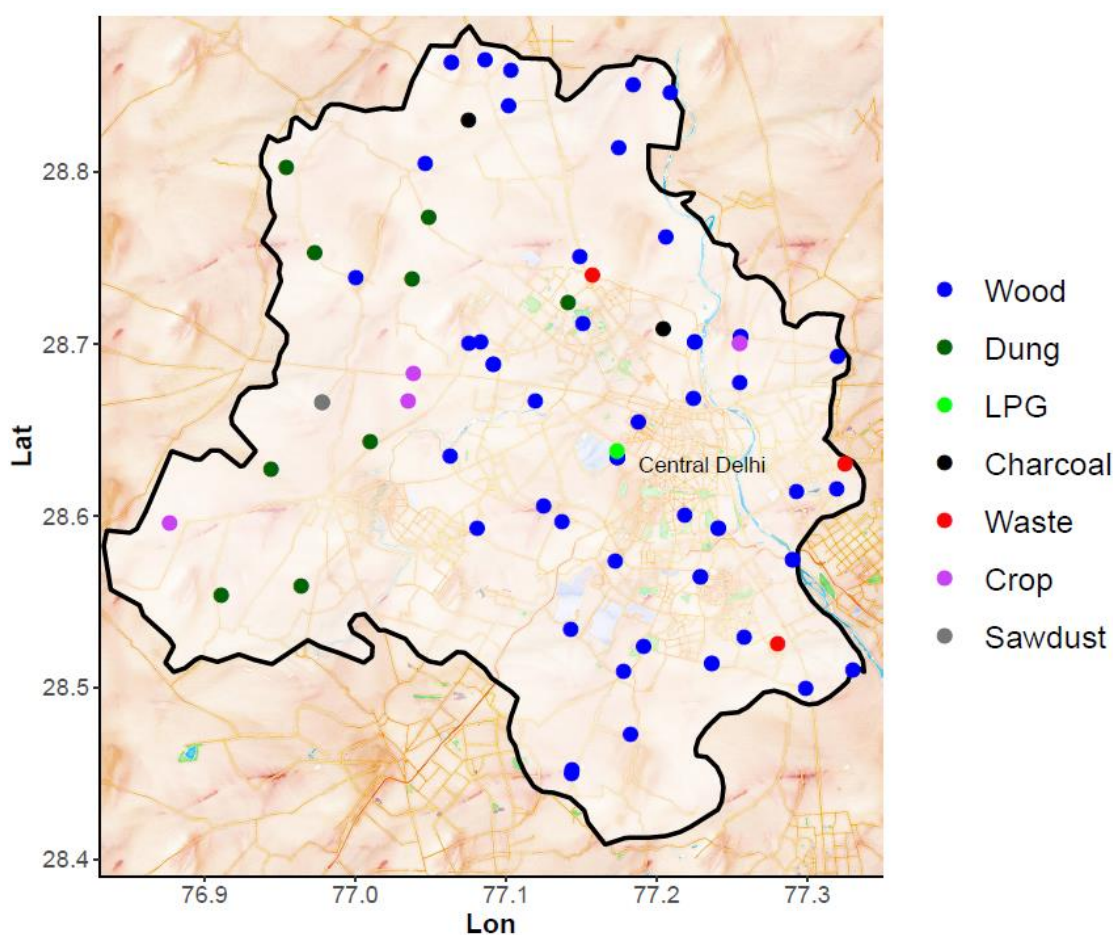
188 Fuel (200 g) was placed 45 cm from the top of the hood and rapidly heated to spontaneous  
189 ignition, with emissions convectively driven into a hood and up a flue to allow enough dilution,  
190 cooling, and residence time to achieve the quenching typically observed in indoor  
191 environments. These conditions have been previously optimised to ensure that emissions  
192 entrainment into the hood did not exert a draft which altered combustion conditions. The mid-  
193 point velocity of gases driven up the flue by convection was measured by a platinum hot-wire  
194 sensor, calibrated for total flow rate using a standard orifice calibrator. Samples were drawn  
195 down a sample line at  $4.4 \text{ L min}^{-1}$  (Swagelok,  $\frac{1}{4}$ " PFA,  $< 2.2 \text{ s}$  residence time) from the top of  
196 the flue, passed through a pre-conditioned quartz filter ( $\phi = 47 \text{ mm}$ , conditioned at  $550 \text{ }^\circ\text{C}$  for  
197 6 hours and changed between samples) held in a filter holder (Cole-Parmer, PFA) which was

198 subsampled for analysis by PTR-ToF-MS, GC×GC-FID and DC-GC-FID instruments at a  
199 distance no greater than 5 m from the top of the flue.

200 Measurements of *n*-alkanes from *n*-tridecane (C<sub>13</sub>) to eicosane (C<sub>20</sub>) were also made from a  
201 subset of 29 burns using solid phase extraction disks (SPE, Resprep, C<sub>18</sub>). Samples were passed  
202 through a cooling and dilution chamber designed to replicate the immediate condensational  
203 processes that occur in smoke particles approximately 5-20 mins after emission, yet prior to  
204 photochemistry which may change composition (Akagi et al., 2011). Further details of SPE  
205 sample collection are given in Stewart et al. (2020a).

206

207



209 Figure 1. Locations across Delhi used for the local surveys into fuel use and collection of representative  
210 biomass fuels. Map tiles by Stamen Design. Data by © OpenStreetMap contributors 2020. Distributed  
211 under a Creative Commons BY-SA License.

212

213 Table 1. Types and numbers of fuels burnt, the mean emission factor of total measured NMVOCs  
 214 (TVOC) in g kg<sup>-1</sup> and standard deviation (SD) from all available burns.

<b>Fuel woods</b>	<b>n</b>	<b>TVOC</b>	<b>SD</b>	<b>Other</b>	<b>n</b>	<b>TVOC</b>	<b>SD</b>
<i>Azadirachta indica</i>	3	18.6	7.9	Cow dung cake	8	61.9	18.4
<i>Morus spp</i>	4	27.4	21.1	<i>Cocos nucifera</i>	2	57.4	23.3
<i>Melia azedarach</i>	2	23.7	13.1	Charcoal	2	5.1	3.9
<i>Shorea spp</i>	2	9.8	2.2	Sawdust	2	71.3	60.8
<i>Ficus religiosa</i>	2	51.9	63.4	Waste	3	87.3	31.4
<i>Syzygium spp</i>	2	8.9	4.9	LPG	3	5.8	5.6
<i>Ficus spp</i>	2	7.1	1.2	Cow dung cake mix	1	34.7	-
<i>Vachellia spp</i>	2	13.5	9.7	<i>Solanum melongena</i>	2	13.6	6.5
<i>Dalbergia sissoo</i>	2	17.9	8.8	<i>Brassica spp</i>	2	41.0	45.5
<i>Ricinus spp</i>	2	8.5	2.5				
<i>Holoptelea spp</i>	2	6.0	0.8				
Mixed woods	1	6.1	-				
<i>Saraca indica</i>	2	12.9	5.2				
<i>Populus spp</i>	1	8.5	-				
<i>Pithecellobium spp</i>	2	19.5	5.4				
<i>Eucalyptus spp</i>	2	6.9	1.9				
<i>Prosopis spp</i>	6	14.5	10.4				
<i>Mangifera indica</i>	2	12.4	3.4				
Plywood	8	26.6	24.3				
Processed wood	2	33.7	17.2				

215

## 216 2.2 PTR-ToF-MS

217 The PTR-ToF-MS (PTR 8000; Ionicon Analytik, Innsbruck) instrument from Physical  
 218 Research Laboratory (PRL), Ahmedabad was used to quantify 107 masses and subsampled the  
 219 common inlet line using ¼ inch PFA. Additional details of the PTR-ToF-MS system used in  
 220 this study are given in previous papers (Sahu and Saxena, 2015; Sahu et al., 2016). The sample  
 221 air was diluted into zero air, generated by passing ambient air (1 L min<sup>-1</sup>) through a heated  
 222 platinum filament at 550 °C, before entering the instrument with an inlet flow of 250 ml min<sup>-1</sup>.  
 223 Samples were diluted by either 5 or 6.25 times (50 ml min<sup>-1</sup> in 200 ml min<sup>-1</sup> zero air or 40 ml  
 224 min<sup>-1</sup> in 210 ml min<sup>-1</sup> zero air). The instrument was operated with an electric field strength  
 225 ( $E/N$ , where  $N$  is the buffer gas density and  $E$  is the electric field strength) of 120 Td. The drift  
 226 tube temperature was 60 °C with a pressure of 2.3 mbar and 560 V applied across it.

227 Calibrations were performed twice a week using a gas calibration unit (Ionicon Analytik,  
 228 Innsbruck). The calibration gas (Apel-Riemer Environmental Inc., Miami) contained 18  
 229 compounds: methanol, acetonitrile, acetaldehyde, acetone, dimethyl sulphide, isoprene,  
 230 methacrolein, methyl vinyl ketone, 2-butanol, benzene, toluene, 2-hexanone, *m*-xylene,  
 231 heptanal,  $\alpha$ -pinene, 3-octanone and 3-octanol at 1000 ppbv ( $\pm$  5%) and  $\beta$ -caryophyllene at 500  
 232 ppbv ( $\pm$  5%). This standard was dynamically diluted into zero air to provide a 6-point



233 calibration. The normalised sensitivity (ncps/ppbv) was then determined for each mass using a  
234 transmission curve (Taipale et al., 2008). The maximum error in this calibration approach has  
235 been shown to be 21% (Taipale et al., 2008). Peak assignment was assisted with results reported  
236 by previous burning studies and references therein (Brilli et al., 2014; Stockwell et al., 2015;  
237 Koss et al., 2018). The results may also contain other indistinguishable structural isomers not  
238 mentioned here.

239 Mass calibration and peak fitting of PTR-ToF-MS data were performed using PTRwid software  
240 (Holzinger, 2015). Count rates (cps) of each mass spectral peak were normalised to the primary  
241 ion ( $\text{H}_3\text{O}^+$ ) and water cluster ( $\text{H}_3\text{O} \cdot \text{H}_2\text{O}^+$ ) peaks, and mixing ratios were then determined for  
242 each mass using the normalised sensitivity. Where compounds known to fragment in the PTR-  
243 ToF-MS were identified, the mixing ratio of these species was calculated by summing parent  
244 ion and fragment ion mixing ratios. Before each burn, ambient air was sampled to provide a  
245 background for the measurement.

246 Petrol and diesel fuel samples were collected from an Indian Oil fuel station in Pusa, New  
247 Delhi, and the headspace analysed to allow comparison of benzene to toluene ratios. This was  
248 designed to analyse the ratios in evaporative emissions, as these have been shown to be an  
249 important source of atmospheric NMVOCs (Srivastava et al., 2005; Rubin et al., 2006; Yamada  
250 et al., 2015), which for example represented ~ 15% of anthropogenic UK NMVOC emissions  
251 in 2018 (Lewis et al., 2020). Fuel samples were placed in a small metal container ( $\frac{1}{4}$ " Swagelok  
252 cap) which was connected to a two-way tap ( $\frac{1}{4}$ " Swagelok). The tap was connected to a t-piece  
253 ( $\frac{1}{4}$ " Swagelok) which had a flow of zero air ( $250 \text{ ml min}^{-1}$ ) passed through it and could be  
254 sampled by the PTR-ToF-MS. The tap was then opened to analyse the headspace of fuels.

### 255 **2.3 DC-GC-FID**

256 Gas chromatography was used to analyse entire burns to provide an integrated picture of  
257 emissions from fuel types. The DC-GC-FID sampled 51 burns to measure 19  $\text{C}_2$ - $\text{C}_7$  non-  
258 methane hydrocarbons (NMHCs) and  $\text{C}_2$ - $\text{C}_5$  oxygenated NMVOCs (OVOCs) (Hopkins et al.,  
259 2003). A 500 ml sample (1.5 L pre-purge of  $100 \text{ ml min}^{-1}$  for 15 minutes, sample at  $17 \text{ mL}$   
260  $\text{min}^{-1}$  for 30 minutes) was collected (Markes International CIA Advantage), passed through a  
261 glass finger at  $-30 \text{ }^\circ\text{C}$  to remove water and adsorbed onto a dual-bed sorbent trap (Markes  
262 International ozone precursors trap) at  $-20 \text{ }^\circ\text{C}$  (Markes International Unity 2). The sample was  
263 thermally desorbed ( $250 \text{ }^\circ\text{C}$  for 3 minutes) then split 50:50 and injected into two separate  
264 columns for analysis of NMHCs ( $50 \text{ m} \times 0.53 \text{ mm Al}_2\text{O}_3 \text{ PLOT}$ ) and OVOCs ( $10 \text{ m} \times 0.53$

265 mm LOWOX with 50  $\mu\text{m}$  restrictor to balance flow). The oven was held at 40  $^{\circ}\text{C}$  for 5 minutes,  
266 then heated at 13  $^{\circ}\text{C min}^{-1}$  to 110  $^{\circ}\text{C}$ , then finally at 8  $^{\circ}\text{C min}^{-1}$  to 200  $^{\circ}\text{C}$  with a 30-minute hold.

#### 267 **2.4 GC $\times$ GC-FID**

268 The GC $\times$ GC-FID was used to measure 58 C<sub>7</sub>-C<sub>12</sub> hydrocarbons (C<sub>7</sub>-C<sub>12</sub> alkanes, monoterpenes  
269 and monoaromatics) and collected 3 L samples (100 ml  $\text{min}^{-1}$  for 30 minutes) using an  
270 adsorption-thermal desorption system (Markes International Unity 2). NMVOCs were trapped  
271 onto a sorbent (Markes International U-T15ATA-2S) at -20  $^{\circ}\text{C}$  with water removed in a glass  
272 cold finger at -30  $^{\circ}\text{C}$ , removed and heated to  $\sim$  100  $^{\circ}\text{C}$  after each sample to prevent carryover  
273 of unanalysed, polar interfering compounds. The sample was thermally desorbed (250  $^{\circ}\text{C}$  for 5  
274 minutes) and injected splitless down a transfer line. Analytes were refocussed for 60 s using  
275 liquid CO<sub>2</sub> at the head of a non-polar BPX5 held at 50 psi (SGE Analytical 15m  $\times$  0.15  $\mu\text{m}$   $\times$   
276 0.25 mm), which was connected to a polar BPX50 at 30 psi (SGE Analytical 2 m  $\times$  0.25  $\mu\text{m}$   $\times$   
277 0.25 mm) *via*. a modulator held at 180  $^{\circ}\text{C}$  (5 s modulation, Analytical Flow Products ELDV2-  
278 MT). The oven was held for 2 minutes at 35  $^{\circ}\text{C}$ , then ramped at 2.5  $^{\circ}\text{C min}^{-1}$  to 130  $^{\circ}\text{C}$  and held  
279 for 1 minute with a final ramp of 10  $^{\circ}\text{C min}^{-1}$  to 180  $^{\circ}\text{C}$  and hold of 8 minutes. The GC systems  
280 were tested for breakthrough to ensure trapping of the most volatile components (see the  
281 Supplementary Information S2). Calibration was carried out using 4 ppbv gas standards  
282 containing alkanes, alkenes and aromatics purchased from the British National Physical  
283 Laboratory and through the relative response of liquid standard injections to toluene for  
284 components not in this gas standard, as detailed elsewhere (Dunmore et al., 2015; Stewart et  
285 al., 2020b). Integration of peak areas was performed in Zoex GC image software (Zoex, USA).  
286 Peaks were individually checked and where peaks were split in the software, they were  
287 manually joined. The areas corresponding to alkane isomers were manually joined within the  
288 GC image software and calibration performed by comparing the areas to the corresponding *n*-  
289 alkane. For both GC instruments, blanks of ambient air were made at the beginning, middle  
290 and end of the day and the mean subtracted from samples.

#### 291 **2.5 GC $\times$ GC-ToF-MS**

292 Measurements were made of a subset of 29 burns of C<sub>13</sub>-C<sub>20</sub> alkanes, as well as other gas-phase  
293 species to assist with qualification of masses measured by PTR-ToF-MS, by adsorbing samples  
294 to the surface of SPE disks with analysis by GC $\times$ GC-ToF-MS, as detailed in Stewart et al.  
295 (2020a). Samples of 180 L were adsorbed to the surface of C<sub>18</sub> coated SPE disks (Resprep, C<sub>18</sub>,  
296 47 mm) prewashed with 2  $\times$  5 mL acetone washes and 1  $\times$  5 mL methanol wash. These samples  
297 were collected at 6 L  $\text{min}^{-1}$  over 30 minutes using a low volume sampler (Vayubodhan Pvt.Ltd)

298 which passed samples through a cooling and dilution chamber at 46.7 L min<sup>-1</sup>. Samples were  
299 then wrapped in foil, placed in an airtight bag, and kept frozen until analysis.

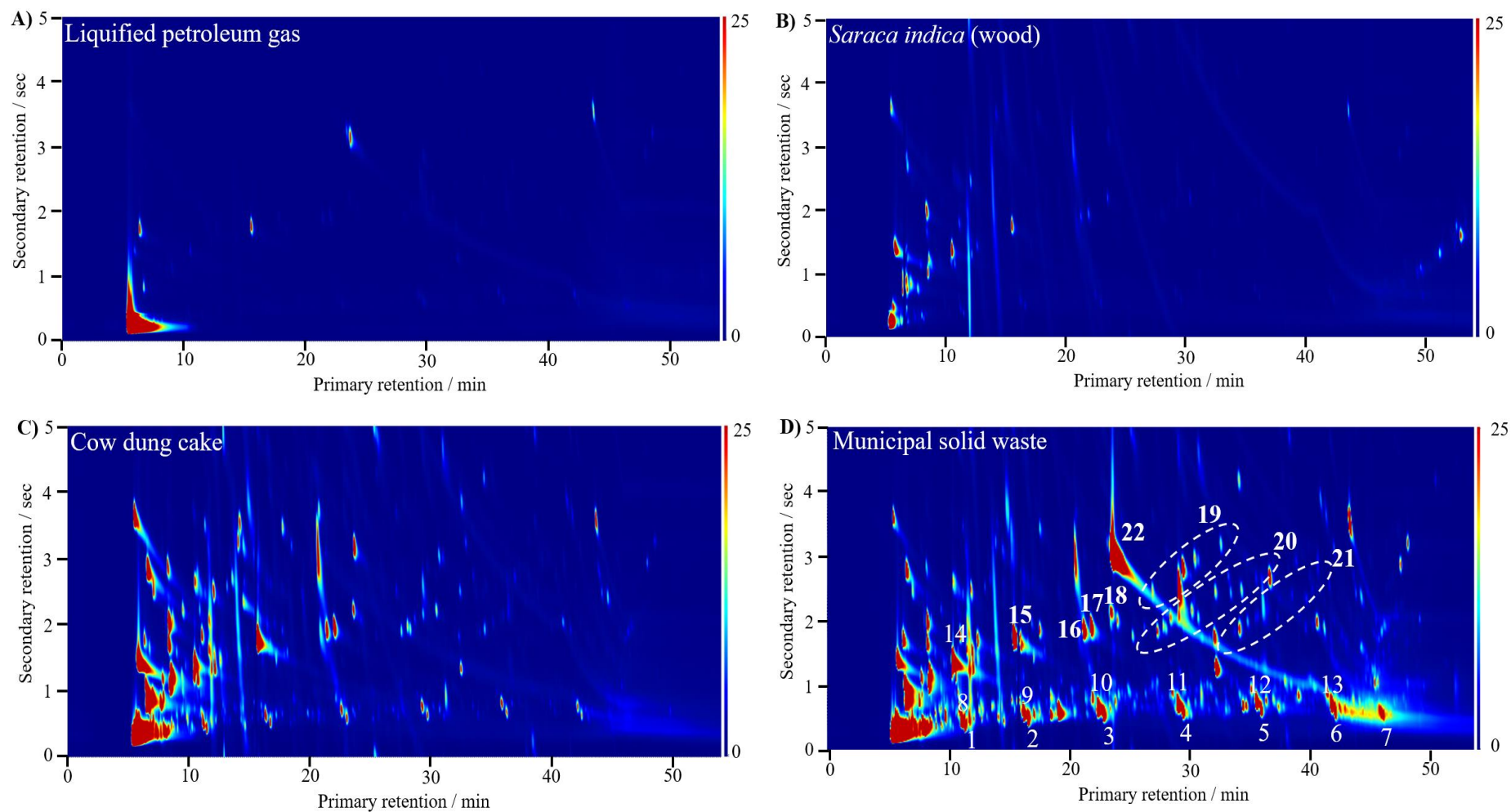
300 SPE extracts were spiked with an internal standard (EPA 8270 Semivolatile Internal Standard  
301 Mix, 2000 µg mL<sup>-1</sup> in DCM) and extracted using accelerated solvent extraction into ethyl  
302 acetate. Extracts were analysed using GC×GC-ToF-MS (Leco Pegasus BT 4D) using a 10:1  
303 split injection (1 µL injection, 4 mm taper focus liner, SHG 560302). The primary dimension  
304 column was a RXI-5SilMS (Restek, 30 m × 0.25 µm × 0.25 mm) connected to a second column  
305 of RXI-17SilMS (Restek, 0.25 µm × 0.25 mm, 0.17m primary GC oven, 0.1 m modulator, 1.42  
306 m secondary oven, 0.31 m transfer line) under a He flow of 1.4 mL min<sup>-1</sup>. The primary oven  
307 was held at 40 °C for 1 min and then ramped at 3 °C min<sup>-1</sup> to 202 °C where it was held for 0.07  
308 mins. The secondary oven was held at 62 °C for 1 min and then ramped at 3.2 °C min<sup>-1</sup> to 235  
309 °C. The inlet was held at 280 °C and the transfer line at 340 °C. A 5 s cryogenic modulation  
310 was used with a 1.5 s hot pulse and 1 s cool time between stages.

311 Peaks assignment was conducted through comparison of retention times to known standards  
312 and comparison to the National Institute of Standards and Technology (NIST) mass spectral  
313 library. Integration was carried out within the ChromaTOF 5.0 software package (Leco, 2019).  
314 Eight blank measurements were made at the beginning and end of the day by passing air from  
315 the chamber (6 L min<sup>-1</sup> for 30 mins) through the filter holder containing a PTFE filter and an  
316 SPE disk. Blank corrections have been applied by subtracting the mean of blank values closest  
317 to measurement of the sample. An 8-point calibration was performed for *n*-alkanes using a  
318 commercial standard (C<sub>7</sub>-C<sub>40</sub> saturated alkane standard, certified 1000 µg mL<sup>-1</sup> in hexane,  
319 Sigma Aldrich 49452-U) diluted in the range 0.25 – 10 µg ml<sup>-1</sup>.

### 320 **3. Results**

#### 321 **3.1 Comparison of chromatograms from combustion of different fuel types**

322 Figure 2 shows GC×GC-FID chromatograms obtained from collecting the emissions during  
323 the combustion of LPG (Figure 2A), *Saraca indica* fuel wood (Figure 2B), cow dung cake  
324 (Figure 2C) and municipal solid waste (Figure 2D). Figure 2D is labelled to show the position  
325 of NMVOCs measured and displays a homologous series of *n*-alkanes from *n*-heptane (C<sub>7</sub>) to  
326 *n*-tetradecane (C<sub>14</sub>) along the bottom, with the 1-alkenes positioned to the left. Above are more  
327 polar species such as monoterpenes, aromatics from benzene to substituted monoaromatics  
328 with up to 5 carbon substituents, and at a higher second dimension retention time even more  
329 polar species, such as styrene.



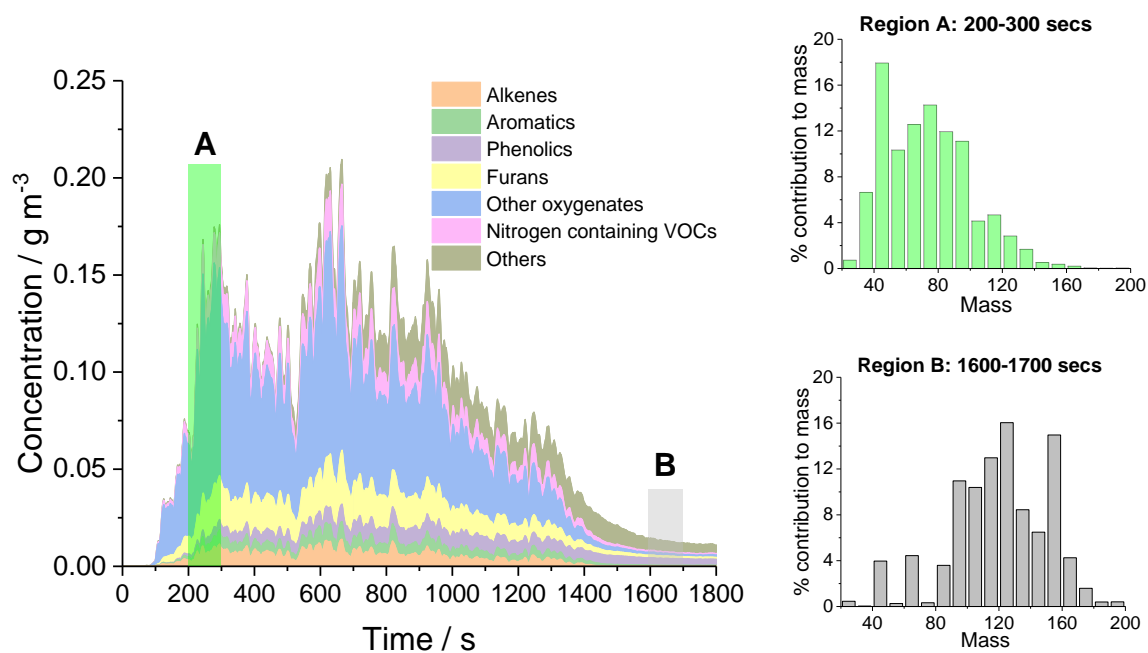
330 Figure 2. GC×GC-FID chromatograms from burning (A) = LPG, (B) = *Saraca indica* (fuel wood), (C) = cow dung cake and (D) = municipal solid waste  
 331 samples where 1-7 = *n*-octane – *n*-tetradecane, 8-13 1-octadecene – 1-tridecene, 14 = benzene, 15 = toluene, 16 = ethylbenzene, 17 = *m/p*-xylene, 18 = *o*-xylene,  
 332 19 = C<sub>3</sub> substituted monoaromatics, 20 = C<sub>4</sub> substituted monoaromatics, 21 = C<sub>5</sub> substituted monoaromatics and 22 = styrene. Samples A-D were collected with  
 333 the same sample collection parameters and the chromatograms are set at the same contrast level to allow direct comparison between different fuel types.

334 Many peaks were present in the chromatograms for cow dung cake and municipal solid waste,  
335 and these fuels released significantly more NMVOCs per unit mass than fuel wood and LPG  
336 (see Table 1). Cow dung cake and municipal solid waste released a range of NMVOCs  
337 including *n*-alkanes, alkenes, and aromatics. The municipal solid waste (Figure 2D) showed a  
338 particularly large and tailing peak 22 owing to large emissions of styrene. Several unidentified  
339 peaks were observed in these complex samples which were broad in the second dimension.  
340 These were assumed to be from polar, oxygenated species formed during burning such as  
341 phenol. These species could not be identified and were not analysed using the GC×GC-FID.  
342 Peaks have been omitted if these species were found to interfere significantly. Analysis has  
343 only been carried out using the DC-GC-FID from ethane (C<sub>2</sub>) to *n*-hexane (C<sub>6</sub>) owing to the  
344 significant presence of coeluting peaks. The large peak in the LPG chromatogram (Figure 2, 1°  
345 ~6 min, 2° ~ 0.5 s) was from unresolved propane and butane because of the high concentrations  
346 from this fuel source.

### 347 **3.2 PTR-ToF-MS concentration time series analysis**

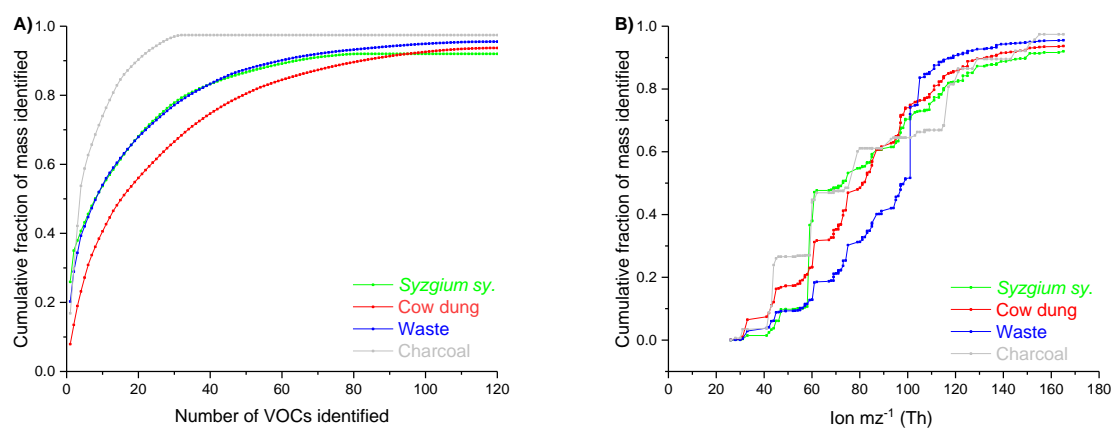
348 Figure 3 shows an example concentration-time series measured by the PTR-ToF-MS for a cow  
349 dung cake burn. A sharp rise in NMVOC emissions was seen from the start of the burn which  
350 decreased as the fuel was combusted. Emissions of small oxygenated species as well as  
351 phenolics and furanics were dominant throughout most of the burn. At the beginning, a greater  
352 proportion of lower mass species were released, as shown in the binned mass spectrum of  
353 region A in Figure 3. At the end in the smouldering phase, emissions were dominated by  
354 heavier and lower volatility species (Figure 3, Region B). A previous study indicated larger  
355 molecular weight phenolics were from low temperature pyrolysis (Sekimoto et al., 2018).

356 Figure 4 shows the cumulative mass of species measured from burns of fuel wood, cow dung  
357 cake, municipal solid waste, and charcoal as a proportion of the total mass of NMVOCs  
358 quantified using PTR-ToF-MS. The results were like those reported by Brillì et al. (2014) and  
359 Koss et al. (2018): 65-90% of the mass of NMVOCs at emission originated from around 40  
360 NMVOCs, with around 70-90% identification by mass when quantifying around 100  
361 NMVOCs. The largest contributors to the NMVOC mass from burning of fuel wood and cow  
362 dung cake were methanol (*m/z* 33.034), acetic acid (*m/z* 61.028) and a peak that reflected the  
363 sum of hydroxyacetone, methyl acetate and ethyl formate (*m/z* 75.043). For municipal solid  
364 waste samples around 28% of total mass was from methyl methacrylate (*m/z* 101.059) and  
365 styrene (*m/z* 105.068), and two of the three municipal solid waste samples released significant  
366 quantities of styrene, most likely the result of degradation of polystyrene in the samples.



367 Figure 3. PTR-ToF-MS concentration-time series during the first 30 minutes of a cow dung cake burn  
 368 coloured by functionality with regions A and B displaying mass spectra placed into  $m/z$  bins of 10 Th.  
 369 Fuel collected from Pitam Pura, Delhi.

370

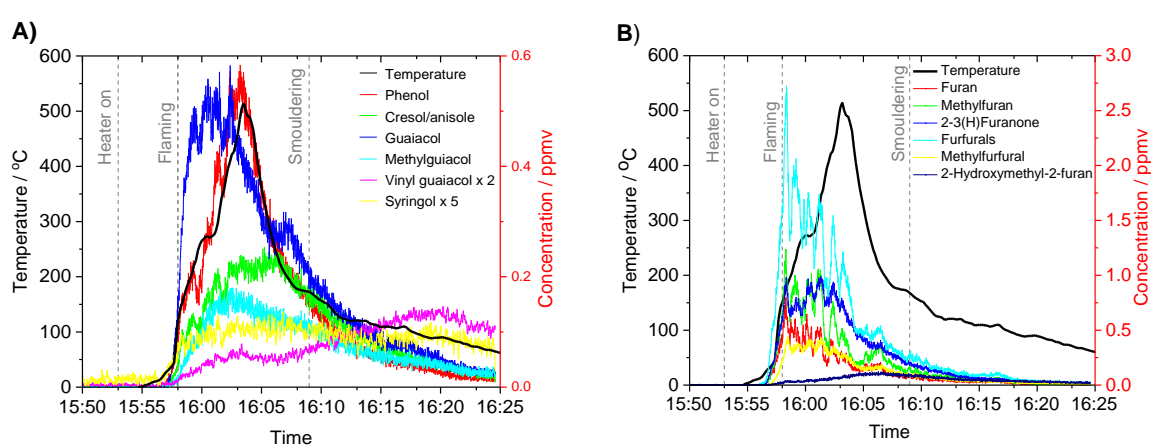


371 Figure 4. Cumulative NMVOC mass identified from PTR-ToF-MS compared with total NMVOC signal  
 372 measured by PTR-ToF-MS with (A) ordered by decreasing NMVOC mass contribution and (B) ordered  
 373 by ion mass. High quantification of emissions from charcoal was due to a low emission factor ( $2.4 \text{ g}$   
 374  $\text{kg}^{-1}$ ).

375

376 Figure 5 shows a concentration time series for phenolics and furanics from the burning of an  
 377 example fuel wood. Most species of similar functionality tracked each other. Stockwell et al.  
 378 (2015) demonstrated that benzene, phenol and furan could act as tracers for aromatic, phenolic  
 379 and furanic species released from biomass burning. Figure 5A shows that heavier, more

380 substituted phenolics appeared to be released at cooler temperatures. Guaiacol (dark blue) was  
 381 released at the start of the flaming phase before the temperature increased and more phenol  
 382 (red) was released at higher burn temperatures. Later in the burn, a larger proportion of vinyl  
 383 guaiacol (pink) and syringol (yellow) were emitted. This agreed well with previous results  
 384 which showed that species emitted from lower temperature depolymerisation had a larger  
 385 proportion of low-volatility compounds compared to higher temperature processes during  
 386 flaming (Sekimoto et al., 2018; Koss et al., 2018). Figure 5B shows concentration time series  
 387 of furanic species, with most species showing similar characteristics throughout the burn. The  
 388 only species to peak later in the burn was 2-hydroxymethyl-2-furan.



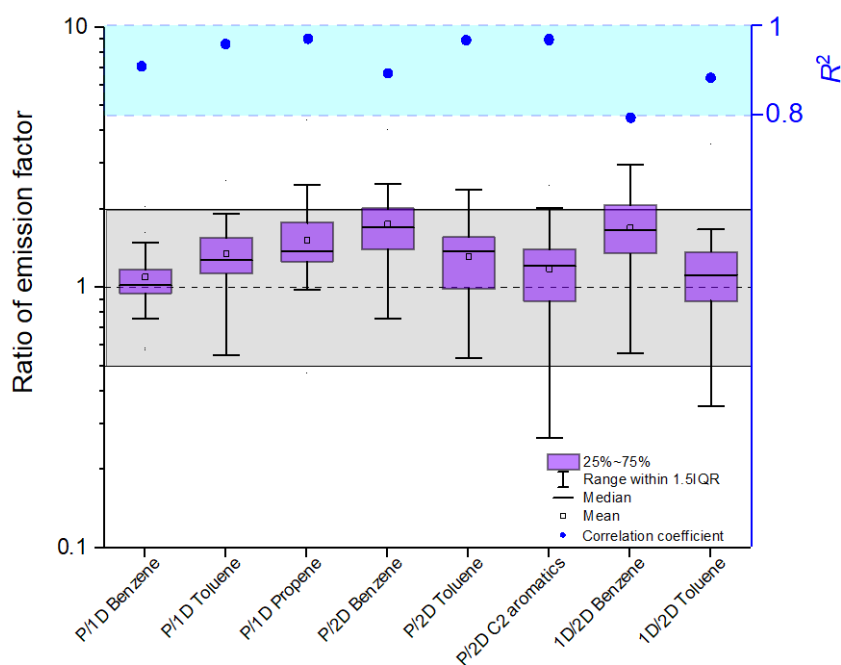
389 Figure 5. Concentration time series analysis of phenolic and furanic compounds released from burning  
 390 of *Azadirachta indica* which released  $27.0 \text{ g kg}^{-1}$  of NMVOCs. Temperature corresponds to the increase  
 391 in temperature above ambient measured in the flame directly above the combustion experiment.

392

### 393 3.3 Comparison of emissions data obtained with different instruments

394 Previous instrument intercomparisons from biomass burning samples were between PTR-MS,  
 395 GC-MS and open path FTIR (Gilman et al., 2015) and between PTR-ToF-MS, FTIR, airborne  
 396 cavity-enhanced spectroscopy (ACES) and I-CIMS (Koss et al., 2018). Gilman et al. (2015)  
 397 showed generally good agreement of slopes of measured emission factors between benzene,  
 398 ethyne, furan, ethene, propene, methanol, toluene, isoprene and acetonitrile using different  
 399 instruments/techniques with slopes of  $\sim 1 \pm 0.3\%$  and correlation coefficients  $> 0.9$ . Koss et al.  
 400 (2018) showed mean measured values of most NMVOCs from all burns with other instruments  
 401 compared to the PTR-ToF-MS which agreed within a factor of two and had correlation  
 402 coefficients  $> 0.8$  for most species except butadienes, furan, hydroxyacetone, furfural, phenol  
 403 and glyoxal. These previous comparisons indicate the level of consistency expected with  
 404 instrument comparisons of quantitative NMVOC measurements from burning experiments.

405 Figure 6 shows a comparison of measurements made using the DC-GC-FID, GC×GC-FID and  
 406 PTR-ToF-MS techniques. Bar plots show that the mean and lower/upper quartiles of all  
 407 measurements agreed within a factor of two. The correlation coefficient between different  
 408 instruments is given by blue circles, with all > 0.8. Generally, the mean values measured for  
 409 the PTR-ToF-MS were slightly larger than using the GC instruments, which was attributed to  
 410 the presence of other undistinguishable structural isomers measured by the PTR-ToF-MS.  
 411 Comparison between DC-GC-FID and GC×GC-FID measurements were also complicated by  
 412 high levels of coelution of additional NMVOC species released from combustion with similar  
 413 retention times ( $R_t$ ) to benzene/toluene ( $R_t = 21/25$  mins) on the DC-GC-FID instrument.  
 414 Generally, the smallest values were measured with the GC×GC-FID instrument, consistent  
 415 with the greatest ability to speciate isomers and limit the impacts of coelution. Significant  
 416 efforts were made to synchronise the sample periods for the three instruments as best as  
 417 possible; however, slight uncertainty existed over the exact time each instrument started  
 418 measuring when calculating mean sample windows ( $\pm 30$  s). These factors combined, may help  
 419 to explain the slight differences observed between different instruments during this study.  
 420 When multiple instruments have measured the same NMVOC in this study, preference was  
 421 given to the data from the GC×GC-FID due to the ability of this instrument to resolve coeluting  
 422 peaks, followed by the DC-GC-FID and then the PTR-ToF-MS.



423

424 Figure 6. Comparison of PTR-ToF-MS to DC-GC-FID and GC×GC-FID with the black dashed line  
 425 representing slopes equal to one, grey shaded region = slopes agreeing within a factor of two, shaded  
 426 blue region indicating correlation coefficients > 0.8 and P = PTR-ToF-MS, 1D = DC-GC-FID and 2D  
 427 = GC×GC-FID.



### 428 3.4 NMVOC emission factors from biomass fuels

429 Figure 7 shows a detailed breakdown of the mean NMVOC emission factors by fuel type  
430 measured for all 76 burns (see the Supplementary Information S3 for values). Emission factors  
431 have been determined by calculating the mean NMVOC concentrations up the flue over a 30-  
432 minute period, in line with the GC sample time, with any small emissions after this sample  
433 window not included. This has been related to the total volume of air convectively drawn up  
434 the flue and the mass of fuel burnt (see the Supplementary Information S4 for details). The data  
435 is split by functionality to show trends for different chemical types. This shows that burning  
436 released a large amount of different NMVOCs, across a wide range of functionalities,  
437 molecular weights and volatilities. The large variety of NMVOCs are likely to have different  
438 influences on O<sub>3</sub> formation, SOA production and the toxicity of emissions.

439 Figure 7A shows very large emissions of smaller oxygenated species which were driven by  
440 methanol, acetic acid and the unresolved combined peak for hydroxy acetone, methyl acetate  
441 and ethyl formate. For the fuel wood samples, acetic acid/glycolaldehyde (2.6 g kg<sup>-1</sup>), methanol  
442 (1.8 g kg<sup>-1</sup>) and acetaldehyde (0.6 g kg<sup>-1</sup>) compared well with mean values reported by Koss et  
443 al. (2018) for pines, firs and spruces (2.7/1.3/1.2 g kg<sup>-1</sup>) and the mean values measured by  
444 Stockwell et al. (2015) mainly from crop residues, grasses and spruces (1.6/1.3/0.9 g kg<sup>-1</sup>). The  
445 emission factor from this study for the unresolved peak of hydroxy acetone, methyl acetate and  
446 ethyl formate (1.4 g kg<sup>-1</sup>) was larger than those previously reported by Koss et al. (2018) and  
447 Stockwell et al. (2015) of 0.55 and 0.25 g kg<sup>-1</sup>, respectively.

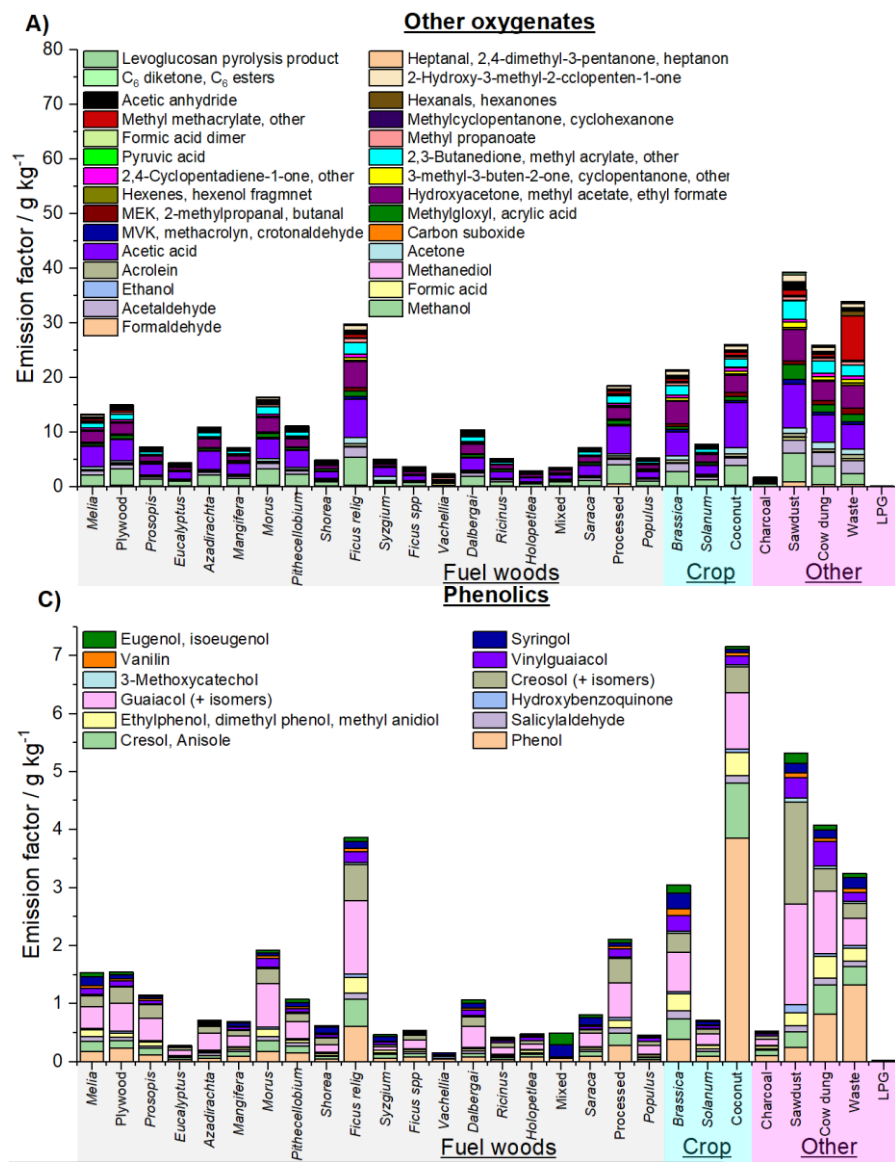
448 Figure 7B shows that there were large emissions of furans and furanones from combustion,  
449 mainly from methyl furans, furfurals, 2-(3H)-furanone, methyl furfurals and 2-methanol  
450 furanone. The World Health Organisation consider furan a carcinogenic species of high-  
451 priority (WHO, 2016) with furan and substituted furans suspected to be toxic and mutagenic  
452 (Ravindranath et al., 1984; Peterson, 2006; Monien et al., 2011). Furan emissions originate  
453 from the low temperature depolymerisation of hemi-cellulose (Sekimoto et al., 2018) and from  
454 large alcohols and enols in high-temperature regions of hydrocarbon flames (Johansson et al.,  
455 2016). The OH chemistry of furans has been the subject of several studies (Bierbach et al.,  
456 1994; Bierbach et al., 1995; Tapia et al., 2011; Liljegren and Stevens, 2013; Strollo and  
457 Ziemann, 2013; Zhao and Wang, 2017; Coggon et al., 2019) and often produces more reactive  
458 products such as butenedial, 4-oxo-2-pentenal and 2-methylbutenedial (Bierbach et al., 1994;  
459 Gómez Alvarez et al., 2009; Aschmann et al., 2011, 2014). Photo-oxidation of furans may also  
460 be a potentially important source of small organic acids such as formic acid (Wang et al., 2020).

461 Oxidation can also occur by nitrate (Berndt et al., 1997; Colmenar et al., 2012) or chlorine  
462 radicals (Cabañas et al., 2005; Villanueva et al., 2007). As a result, furans have recently been  
463 shown to be some of the species with highest OH reactivity from biomass burning, causing an  
464 estimated 10% of the O<sub>3</sub> produced by the combustion emissions in the first 4 hours after  
465 emission (Hartikainen et al., 2018; Coggon et al., 2019). Oxidation of furans can lead to SOA  
466 production (Gómez Alvarez et al., 2009; Strollo and Ziemann, 2013) with an estimated 8-15%  
467 of SOA caused by furans emitted by burning of black spruce, cut grass, Indonesian peat and  
468 ponderosa pine and 28-50% of SOA from rice straw and wiregrass (Hatch et al., 2015),  
469 although SOA yields are still uncertain for many species (Hatch et al., 2017).

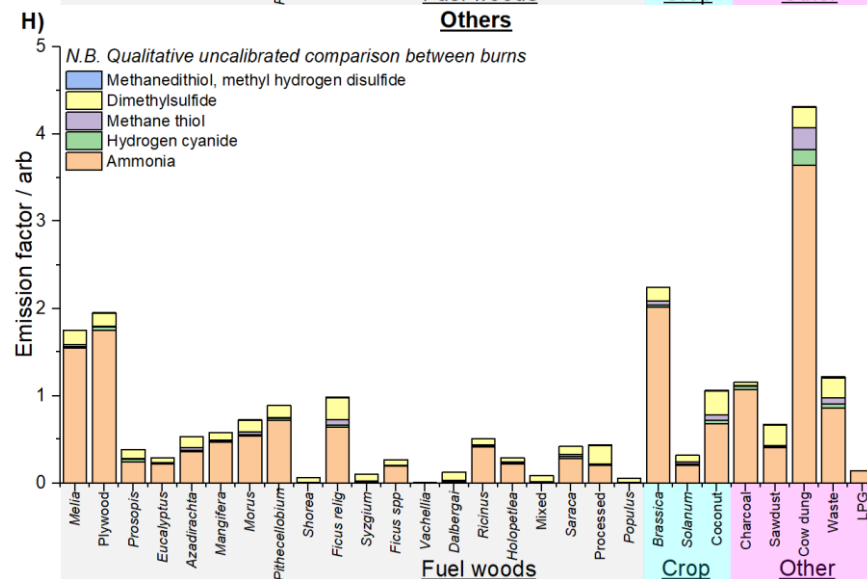
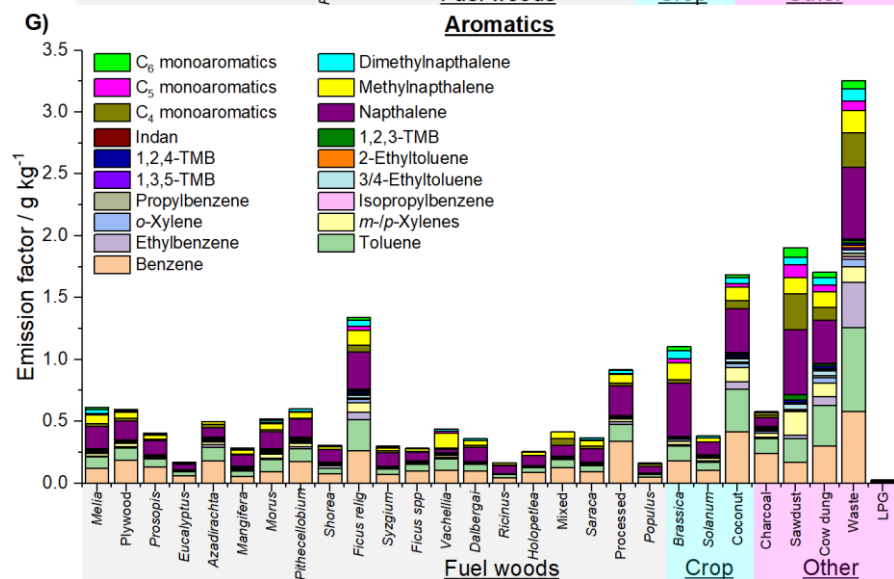
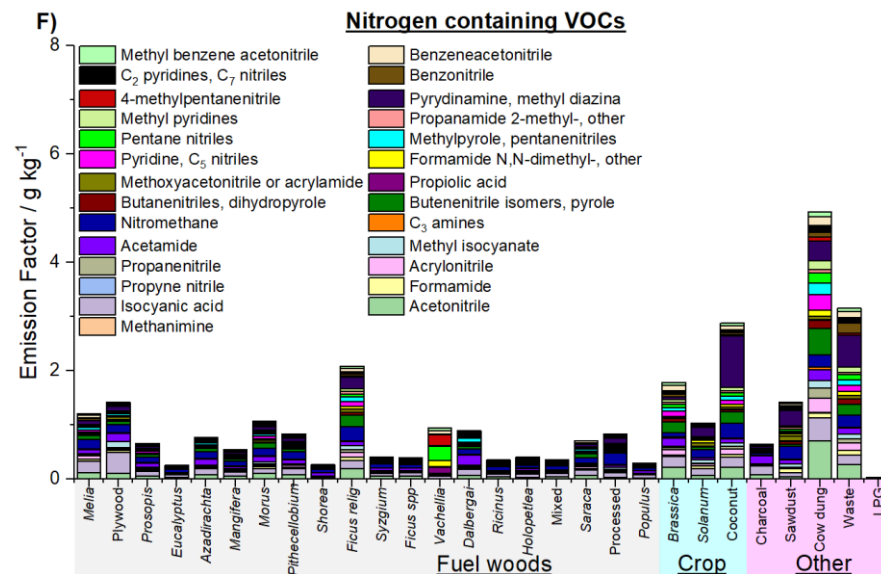
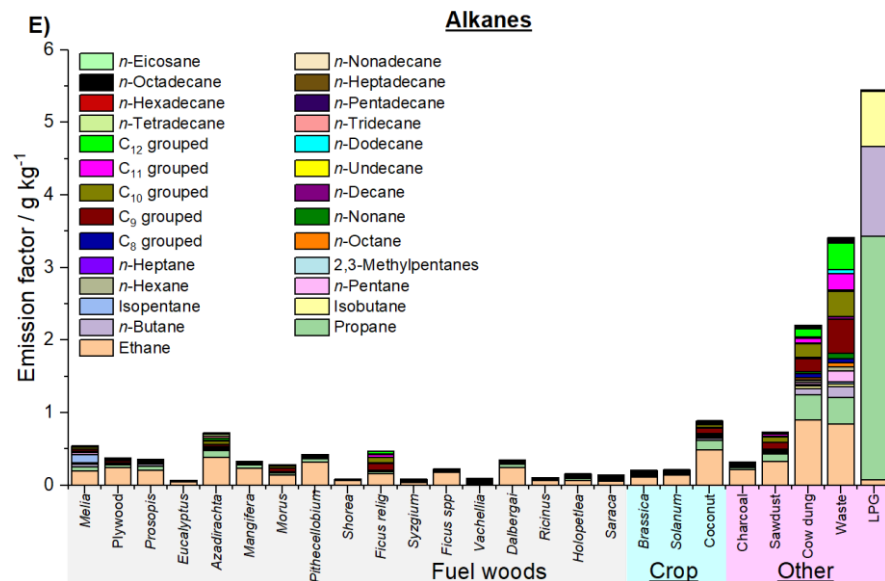
470 Phenols are formed from the low-temperature depolymerisation of lignin (Simoneit et al., 1993;  
471 Sekimoto et al., 2018) which is a polymer of randomly linked, amorphous high-molecular  
472 weight phenolic compounds (Shafizadeh, 1982). Owing to their high emission ratios and SOA  
473 formation potentials, phenolic compounds contribute significantly to SOA production from  
474 biomass-burning emissions (Yee et al., 2013; Lauraguais et al., 2014; Gilman et al., 2015;  
475 Finewax et al., 2018). Figure 7C shows that the largest phenolic emissions from fuel wood in  
476 this study were methoxyphenols, with significant contributions from phenol, guaiacol, cresols  
477 and anisole. Phenolic emissions from sawdust were dominated by guaiacol and creosol.  
478 Phenolic emissions from coconut shell were greatest, most likely as a result of the lignin rich  
479 nature of coconut shell (Pandharipande, 2018). The larger mean emission of furanics (3.2 g kg<sup>-1</sup>)  
480 compared to phenolics (1.1 g kg<sup>-1</sup>) from fuel wood was consistent with wood being composed  
481 of around 75% cellulose/hemicellulose and 25% lignin (Sjöström, 1993).

482 Figure 7D shows that the largest alkene emission was styrene from burning municipal solid  
483 waste, likely caused by the presence of polystyrene in the fuel. Emissions of alkenes from fuel  
484 woods were dominated by ethene and propene, species with high photochemical ozone creation  
485 potentials (Cheng et al., 2010). Monoterpenes, which are extremely reactive with the OH  
486 radical (Atkinson and Arey, 2003), were emitted from combustion of sawdust, cow dung cake  
487 and municipal solid waste samples.

488 Ethane and propane dominated the alkane emissions for fuel wood samples (see Figure 7E). A  
489 wider range of alkanes from C<sub>2</sub>-C<sub>20</sub> were observed from combustion of coconut, cow dung  
490 cake and municipal solid waste. The largest alkane emission by mass was from LPG due to  
491 unburnt propane and butane.



492 Figure 7. Measured emission factors grouped by functionality.



493 Figure 7 continued.

494 Nitrogen containing NMVOCs are formed from the volatilisation and decomposition of  
495 nitrogen-containing compounds within the fuel, mainly from free amino acids but can also be  
496 from pyrroline, pyridine and chlorophyll (Leppalahti and Koljonen, 1995; Burling et al., 2010;  
497 Ren and Zhao, 2015). Nitrogen containing NMVOCs are of interest because nitrogen may be  
498 important in the development of new particles (Smith et al., 2008; Kirkby et al., 2011; Yu and  
499 Luo, 2014) which act as cloud condensation nuclei (Kerminen et al., 2005; Laaksonen et al.,  
500 2005; Sotiropoulou et al., 2006) and alter the hydrological cycle by forming new clouds and  
501 precipitation (Novakov and Penner, 1993). They can also contribute to light-absorbing brown  
502 carbon (BrC) aerosol formation, effecting climate (Laskin et al., 2015). Additionally, nitrogen  
503 containing NMVOCs can be extremely toxic (Ramírez et al., 2012, 2014; Farren et al., 2015).  
504 Cow dung cake was the largest emitter of nitrogen containing NMVOCs ( $4.9 \text{ g kg}^{-1}$ ), releasing  
505 large amounts of acetonitrile and nitriles, likely to have a large impact on the toxicity and  
506 chemistry of emissions (see Figure 7F).

507 Figure 7G shows emissions of aromatics from fuel wood, cow dung cake and municipal solid  
508 waste were principally benzene, toluene and naphthalenes. Large emissions of benzene were  
509 unsurprising as biomass burning is the largest global benzene source (Andreae and Merlet,  
510 2001). Emissions of benzene, toluene, ethylbenzene and xylenes (BTEX) from cow dung cake  
511 ( $0.5\text{-}1.7 \text{ g kg}^{-1}$ ) were in line with previous measurements of  $1.3 \text{ g kg}^{-1}$  (Koss et al., 2018) and  
512  $1.8 \text{ g kg}^{-1}$  (Fleming et al., 2018) but lower than the  $4.5 \text{ g kg}^{-1}$  reported from cow dung cake  
513 combusted from Nepal (Stockwell et al., 2016). Emissions of BTEX from municipal solid  
514 waste burning ( $0.9\text{-}2.6 \text{ g kg}^{-1}$ ) were comparable to that measured previously ( $3.5 \text{ g kg}^{-1}$ )  
515 (Stockwell et al., 2016).

516 Figure 7H shows a qualitative comparison of species such as ammonia, HCN and dimethyl  
517 sulphide which were measured during experiments, but could not be accurately quantified as  
518 their sensitivity was too different from the NMVOCs used to build the transmission curve. Cow  
519 dung cake emitted significantly more of these species than other fuel types.

520 Table 2 shows the total measured emission factors of NMVOCs for different fuel types. The  
521 total measured emission factor has been calculated as the sum of the PTR-ToF-MS signal,  
522 excluding reagent ion peaks ( $< m/z 31 \text{ Th}$ ), water cluster peaks ( $m/z 37 \text{ Th}$ ) and isotope peaks  
523 identified for all masses (SIS, 2016). The emission factors for all alkanes and alkenes measured  
524 by the GC instruments were also included, as alkanes up to *n*-hexane had proton affinities less  
525 than water and larger alkanes had proton affinities similar to water (Ellis and Mayhew, 2014;

526 Wróblewski et al., 2006). This low sensitivity meant that no peaks were present in the PTR-  
 527 ToF-MS spectra for these larger species. Any alkenes measured by the DC-GC-FID were  
 528 excluded from the PTR-ToF-MS data. Further information on the species included in the  
 529 calculation of the total measured emission factor is given in the Supplementary Information  
 530 S5.

531

532 Table 2. Mean total measured NMVOC emission factors ( $\text{g kg}^{-1}$ , including IVOC fraction) where  
 533 high/low EF represent the largest/smallest emission factor measured for a given sample type ( $\text{g kg}^{-1}$ )  
 534 and IVOC is the sum of emission factors of species with a mass greater than benzaldehyde ( $\text{g kg}^{-1}$ )  
 535 where  $n$  = number of measurements made.

	<b>Wood</b>	<b>Dung</b>	<b>Waste</b>	<b>LPG</b>	<b>Charcoal</b>	<b>Sawdust</b>	<b>Crop</b>
NMVOC	18.7	62.0	87.3	5.7	5.4	72.4	37.9
High EF	96.7	83.0	119.1	9.8	7.9	114.0	73.8
Low EF	4.3	35.3	56.3	1.9	2.4	28.3	8.9
IVOC	3.5	12.6	13.2	0.2	1.4	16.9	8.0
$n$	51	8	3	3	2	2	6

536

537 Coconut shell, sawdust, cow dung cake and municipal solid waste released the greatest mass  
 538 of NMVOC per kg of fuel burnt. The mean emission factor for all fuel woods ( $18.7 \text{ g kg}^{-1}$ ) was  
 539 comparable to that for chaparral ( $16.6 \text{ g kg}^{-1}$ ) measured using PTR-ToF-MS by Stockwell et  
 540 al. (2015). This may be due to similarities between north Indian fuel wood types with chaparral,  
 541 which is characterised by hot dry summers, and mild wet winters. The mean fuel wood  
 542 emission factor was smaller than Stockwell et al. (2015) reported for coniferous canopy ( $31.0$   
 543  $\text{g kg}^{-1}$ ). The NMVOC emission measured for cow dung cake ( $62.0 \text{ g kg}^{-1}$ ) was comparable to  
 544 that previously reported ( $66.3 \text{ g kg}^{-1}$ ) in literature using PTR-ToF-MS (Koss et al., 2018), but  
 545 2-3 times larger than that measured by GC-FID/ECD/MS likely due to those techniques  
 546 missing significant amounts of emissions (Fleming et al., 2018). Whilst the total measured  
 547 emissions reported by Fleming et al. (2018) might therefore be an underestimate, it is  
 548 noteworthy that the emission factors measured by Fleming et al. (2018) in *angithi* stoves for  
 549 cow dung cake were ~ factor of 4 greater than fuel wood under the same conditions. This result  
 550 was comparable to this study which showed that cow dung cake emissions were ~ factor of 3  
 551 larger than fuel wood, but the techniques used here targeted a greater proportion of total  
 552 emissions. Moreover, Fleming et al. (2018) reported emission factors from combustion of  
 553 biomass fuels from a neighbouring state, Haryana, and there may be slight heterogeneity  
 554 between the different fuels collected in both studies. Venkataraman et al. (2010) and Koss et

555 al. (2018) also showed NMVOC emissions from dung combustion to be greater than from fuel  
556 wood. NMVOC emissions from municipal solid waste ( $87.3 \text{ g kg}^{-1}$ ) were significantly larger  
557 than the  $\sim 9 \text{ g kg}^{-1}$  (Stockwell et al., 2015) and  $\sim 35 \text{ g kg}^{-1}$  (Stockwell et al., 2016) previously  
558 reported. This was likely due to differences in composition and moisture content of the fuels  
559 collected from Indian landfill sites for the present study, compared with the daily mixed waste  
560 and plastic bags collected at the US fire services laboratory (Stockwell et al., 2015) and a  
561 variety of mixed waste and plastics collected from around Nepal (Stockwell et al., 2016). It  
562 seems noteworthy that combustion experiments of fuels collected from developing countries  
563 in Stockwell et al. (2016) had larger emission factors than those collected from, and burnt at a  
564 laboratory (Stockwell et al., 2015). The mean crop residue combustion emission factor ( $37.9 \text{ g}$   
565  $\text{kg}^{-1}$ ) was comparable to that reported by Stockwell et al. (2015) ( $36.8 \text{ g kg}^{-1}$ ), despite the small  
566 number of samples in this study and compositional differences.

567 Considerable uncertainties exist in consumption estimates for fuels such as cow dung cake and  
568 municipal solid waste in India. A previous study estimated that in 1985 in India fuel wood  
569 consumption was 220 Tg and cow dung cake consumption 93 Tg (Yevich and Logan, 2003).  
570 A different study made an India-wide estimate for 2000 which estimated fuel wood  
571 consumption to be 281 (192-409) Tg and cow dung cake consumption to be 62 (35-128) Tg  
572 (Habib et al., 2004). A more recent study estimated fuel wood usage at 256 Tg and cow dung  
573 cake consumption at 106 Tg for 2007 (Singh et al., 2013). Estimates of the amount of municipal  
574 solid waste burnt in India are even fewer than for cow dung cake consumption. Two previous  
575 studies have estimated that 81.4 Tg of municipal solid waste was burnt in India in 2010  
576 (Wiedinmyer et al., 2014) and that 68 (45-105) Tg was burnt in 2015 (Sharma et al., 2019).  
577 The mean emission factors for cow dung cake and municipal solid waste combustion were  
578 considerably larger than for fuel wood and highlight that at an India-wide level these may  
579 represent significant NMVOC sources.

580 Intermediate-volatility organic compounds (IVOCs) are defined as having effective saturation  
581 concentration,  $C^*$ ,  $=300\text{-}3\times 10^6 \mu\text{g m}^{-3}$  (Donahue et al., 2012). The  $C^*$  of several species was  
582 estimated using a previously established approach (Lu et al., 2018), with the IVOC boundary  
583 defined in this study at benzaldehyde ( $m = 106.12$ ) for which  $C^*$  was  $\sim 7\times 10^6 \mu\text{g m}^{-3}$ . Table 2  
584 also shows an approximation for the mean amount of IVOCs released by fuel type. This  
585 approach was approximate as vapour pressures depend on both mass and functionality. The  
586 fuels tested in this study showed that mean emissions of IVOC species represented  
587 approximately 18 – 27% of total measured emissions from all fuel types other than LPG. This

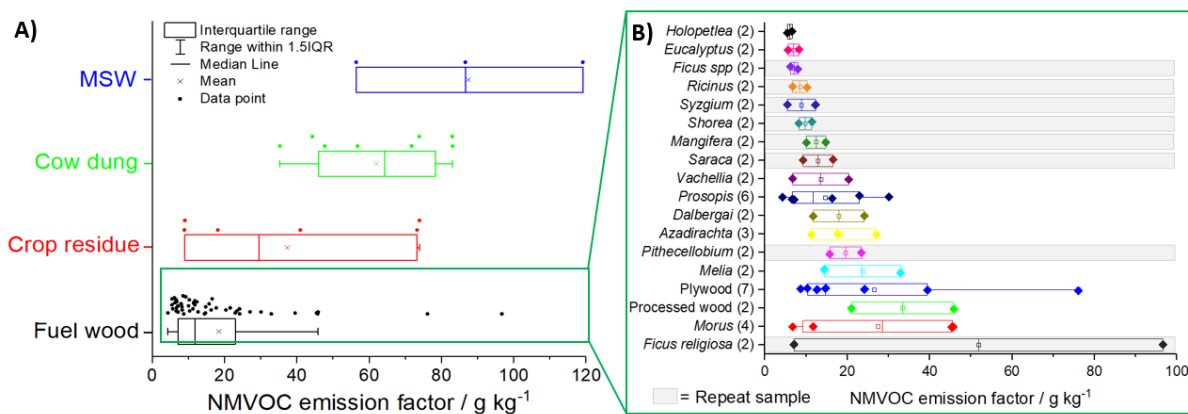
588 demonstrated that domestic solid fuel combustion is potentially a large global source of IVOCs.  
589 In addition, this may represent an underestimate because the quartz filter placed on the sample  
590 line may remove IVOC species which have partitioned to the aerosol phase due to the high  
591 aerosol concentrations present during source testing. Further studies are required to better  
592 understand the contribution of IVOC emissions from biomass burning to SOA formation.

593 Figure 8A shows the distribution of total measured NMVOC emission factors for fuel wood,  
594 cow dung cake, crop residues and MSW. Boxplots show the mean, median, interquartile range  
595 and range within 1.5IQR. The solid circles display the spread of measured emission factors by  
596 fuel type. The zoomed green region given in Figure 8B specifically focuses on the variability  
597 in emission factors of individual species of fuel wood, which has been explored in detail due  
598 to the large number of samples. Repeat samples collected from the same location are shaded in  
599 grey. For fuel wood, measured NMVOC emission factors varied by over a factor of 20 between  
600 4.3-96.7 g kg<sup>-1</sup>. The NMVOC emission factors showed a right skewed distribution with a  
601 median of 11.7 g kg<sup>-1</sup>, mean of 18.7 g kg<sup>-1</sup> and an interquartile range of 15.3 g kg<sup>-1</sup>. For repeat  
602 measurements of identical species of fuel wood collected at the same location, except for *Ficus*  
603 *religiosa*, measured emission factors from repeat experiments varied over a much smaller  
604 range, by up to a factor of 2.3. Variation between emissions from these samples were likely  
605 due to different moisture contents of actual samples measured and the specific combustion  
606 conditions of individual burns. The large variation observed for *Ficus religiosa* was likely due  
607 to the samples being significantly different in terms of composition. Despite the samples for  
608 *Holoptelea spp* and *Eucalyptus spp* coming from different locations, emission factors for these  
609 samples were quite reproducible and only varied by a factor of 1.2-1.5. For remaining identical  
610 species of fuel wood collected from different locations, emission factors varied over a much  
611 larger range by factors of ~ 2-9.

612 For the crop residue species studied here, NMVOC emissions were right skewed with a with a  
613 median of 29.5 g kg<sup>-1</sup> which was less than the mean of 37.9 g kg<sup>-1</sup> and varied from 8.9-73.8 g  
614 kg<sup>-1</sup> with an interquartile range of 53.9 g kg<sup>-1</sup>. *Cocos nucifera* and *Solanum melongena* were  
615 repeat measurements of fuel collected from the same location and varied by factors of 1.8-2.  
616 NMVOC emissions from *Brassica spp* fuel, which was collected from different locations,  
617 varied by a factor of ~ 8. Cow dung cake and MSW samples were all collected from different  
618 locations and varied by up to factors of up to 2.4 and 2.1, respectively.

619





620

621 Figure 8. Variability in NMVOC emission factor by fuel type. A) = Range of emission factors measured  
 622 for fuel wood, cow dung cake, crop residue and municipal solid waste samples with box plots showing  
 623 the mean, median, interquartile range, range within 1.5IQR and solid circles showing the spread of  
 624 measured emission factors by fuel type. B) = Zoomed green region displaying range of NMVOC  
 625 emission factors measured for individual species of fuel wood with grey shaded region indicating repeat  
 626 samples from the same sample collection location and diamonds indicating the measured NMVOC  
 627 emission factors.

628

629 Figure 9A shows the mean total emissions measured in this study for different fuel types split  
 630 by functionality. Large variability in total measured emissions were observed for fuel woods,  
 631 with emission factors from individual burns varying by ~ factor 20. Figure 9B shows the mean  
 632 emissions by functionality as a proportion of total measured emissions averaged by overall fuel  
 633 type. Oxygenates were the largest emission (33-55%), followed by furanic compounds (16-  
 634 21%), phenolics (6-12%) and aromatics (2-9%) for all fuel types except LPG. LPG emissions  
 635 were mainly alkanes, with a small emission of furanic species. These have previously been  
 636 reported to be produced in hydrocarbon flames (Johansson et al., 2016).

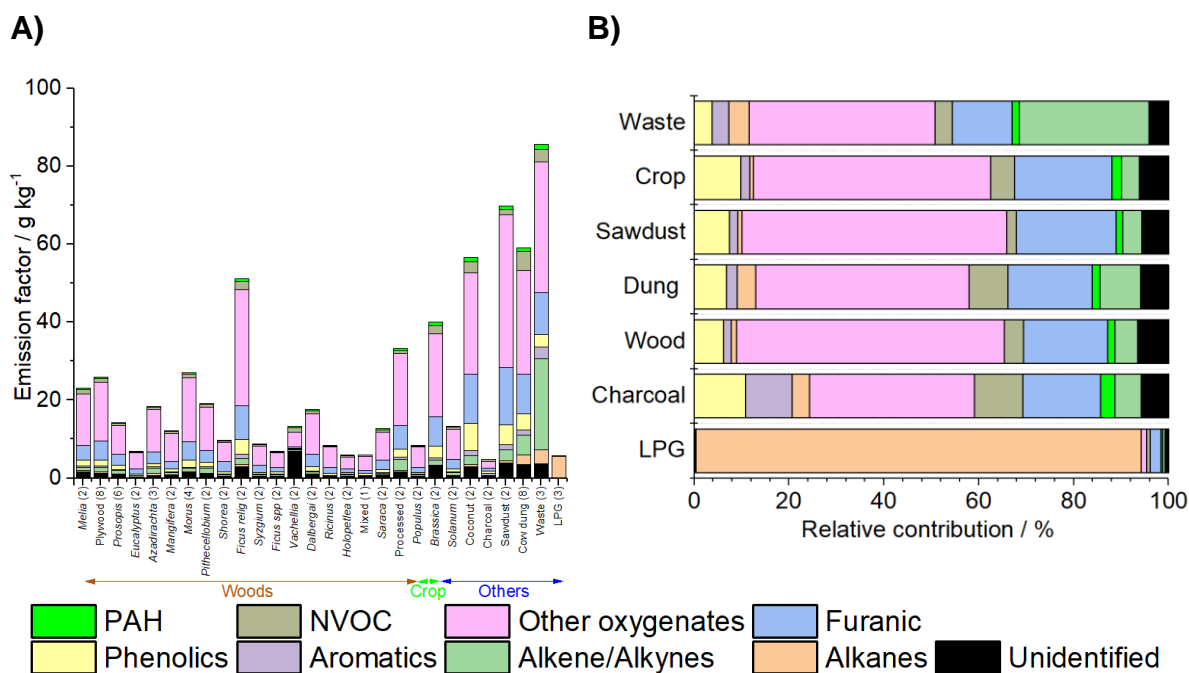
637 Figure 9A-B also show the amount of NMVOC which remained unidentified (black). On  
 638 average 94% of all measured NMVOCs emitted across all burns were speciated. Speciation  
 639 was greater than 90% for all sample types, except *Vachellia spp* due to several large  
 640 unidentified peaks (see the Supplementary Information S6). Mean speciation by fuel type was  
 641 between 93-96% for all other fuels, except LPG where speciation was > 99%.

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645



646 Figure 9. NMVOC emissions from burning sources in Delhi, India grouped by functionality with  
 647 unidentified emissions given by the total NMVOC signal measured by the PTR-ToF-MS minus the  
 648 fraction quantified using DC-GC-FID, GC×GC-FID, GC×GC-ToF-MS and PTR-ToF-MS instruments  
 649 with (A) all fuel types and (B) mean values by type of fuel.

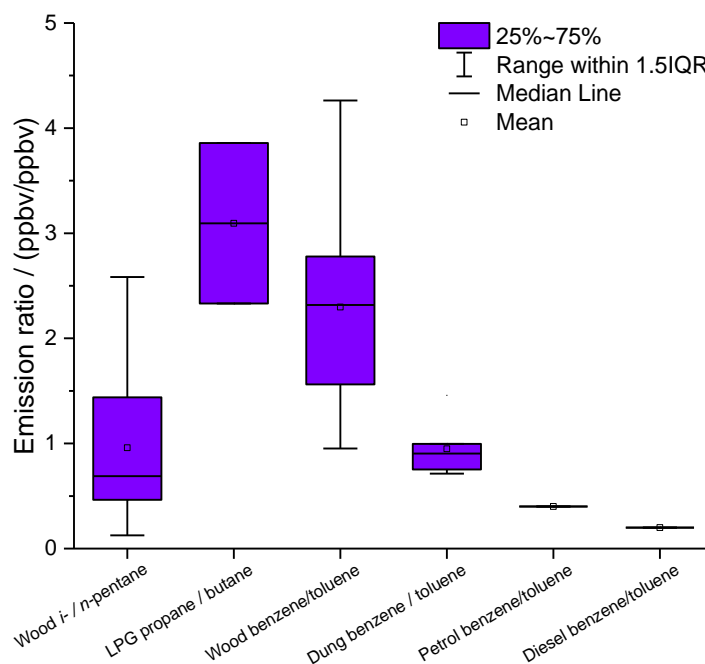
650

### 651 3.5 Emission ratios

652 The ratio of the mixing ratios of NMVOCs in the emitted gas can be a useful indicator of their  
 653 source(s) in ambient air. Ratios can be specific to sources and can allow one source to be  
 654 distinguished from another. The ratio of *i*-*n*-pentane can be a useful indicator of whether  
 655 emissions are anthropogenic or from biomass burning, with a ratio 2.2-3.8 indicative of  
 656 vehicular emissions, 0.8-0.9 for natural gas drilling, 1.8-4.6 for evaporative fuel emissions and  
 657 < 1 from burning (Stewart et al., 2020b). Benzene/toluene ratios can also be useful and have  
 658 been reported from traffic exhaust to be around 0.3 (Hedberg et al., 2002).

659 *i*-*n*-Pentane indicator ratios have been evaluated for fuel wood sources, propane/butane ratios  
 660 for LPG and benzene/toluene ratios for fuel wood and cow dung cake (see Figure 10). The  
 661 range of values for multiple different burns have been evaluated rather than just reporting mean  
 662 and median ratios. The median of *i*-*n*-pentane ratios from biomass samples measured during  
 663 this study was ~ 0.7 (see Figure 10). The mean ratio was ~ 1.0, with an interquartile range  
 664 (IQR) ~ 0.5-1.5, which suggests caution is required when assigning burning sources based on  
 665 emission ratios due to considerable variability. Despite this, the ratio from solid fuel  
 666 combustion sources was often less than expected from petrol emissions. The mean ratio of  
 667 propane/butane from LPG burning was measured to be 3.1. The ratios of benzene/toluene

668 varied considerably between different sources and was measured for fuel wood combustion  
 669 (2.3), cow dung cake combustion (0.94), petrol liquid fuel (0.40) and diesel liquid fuel (0.20).  
 670 The range of benzene/toluene ratios for fuel wood was large, with an IQR of ~ 1.5- 2.8 and the  
 671 range within 1.5 IQR shown by the whiskers in Figure 10 from ~ 0.9-4.2. Despite the variability  
 672 of ratios from specific source types, the considerable range of benzene/toluene ratios could  
 673 potentially be a useful indicator of the origin of unaged (fresh) ambient emissions in Delhi.  
 674 However, further study would be required to assess if these ratios were also true in the exhaust  
 675 of petrol and diesel vehicles in India, or just limited to fugitive emissions. These findings agree  
 676 well with literature which report mean benzene/toluene ratios of 1.4-5.0 from fuel wood and  
 677 0.3 from automotive emissions (Hedberg et al., 2002), indicating that on average biomass  
 678 burning releases a greater molar ratio of benzene than toluene when compared to automotive  
 679 emissions.



680

681 Figure 10. Summary of ratios of NMVOCs measured during this study from the burning of fuel wood,  
 682 LPG and cow dung cake and from the headspace of liquid petrol and diesel fuels collected in India. The  
 683 different mean and median values have been considered to evaluate the ratios at emission of specific  
 684 sources.

#### 685 4. Conclusions

686 This study was based on comprehensive measurements of NMVOC emissions using a range of  
 687 detailed and complementary techniques across a large range of functionalities and volatilities.  
 688 It presented detailed burning emission factors for different NMVOCs from a range of fuels

689 used in the Delhi area of India for residential combustion. This work allowed for a better  
690 understanding of the impact of residential combustion on air quality and showed that fuel wood,  
691 cow dung cake and municipal solid waste burning sources released significantly more  
692 NMVOCs than LPG.

693 A range of areas where future studies are required to better improve and understand emissions  
694 from burning have been highlighted:

695 1. Better understanding of stove burn conditions on emissions

696 The impact of stove conditions on NMVOC emissions remains poorly understood.  
697 Experiments in this study were carried out using expert local judgment to attempt to ensure  
698 that laboratory conditions reflected real-world burning conditions. A range of stoves are used  
699 in India for combustion of local fuels, such as *chulha* and *angithi* stoves, and an evaluation of  
700 the impact of these on emissions and their relative use and spatial distribution requires further  
701 study.

702 2. Better understanding of the effect of moisture content on modified combustion  
703 efficiency

704 Fuels in this study were collected and stored in a manner designed to be reflective of local  
705 practices to ensure that laboratory combustion conditions, and in turn emissions, reflected local  
706 burning practices. Future studies should conduct detailed compositional analysis of fuel types  
707 and moisture content prior to burning. These studies should also measure CO and CO<sub>2</sub> to allow  
708 an evaluation of the impact of modified combustion efficiency on emissions from different fuel  
709 types.

710 3. Limited measurements of some fuel types

711 Few measurements were made from domestic, commercial and industrial waste, and the  
712 emission factors measured in this study were higher than those observed in previous studies.  
713 The effect of moisture content on waste burning has been suggested to impact emissions of  
714 particulate matter by around an order of magnitude (Jayarathne et al., 2018). Furthermore, only  
715 one LPG stove was used to evaluate emissions from this fuel source, with emissions likely to  
716 vary by the type of burner used. Future studies should also make more measurements from  
717 waste burning to better understand the effect of composition on emissions. Comprehensive  
718 measurements should also be made of emissions from combustion of a range of additional crop  
719 residues, as these are an important NMVOC source in India (Jain et al., 2014).

720 4. Evaluation of the impact on O<sub>3</sub> and SOA production as well as the toxicity of emissions  
721 Better understanding of the drivers of photochemical O<sub>3</sub> and SOA production from burning  
722 emissions is required. A large variety of high molecular weight species with likely low  
723 volatilities, such as phenolic and furanic compounds, were released from burning. These  
724 NMVOCs are expected to have a large influence on subsequent atmospheric chemistry, and a  
725 detailed understanding of this chemistry is required to truly assess the impact of biomass  
726 burning on air quality.

727 5. Evaluation of the relative importance of fuel types to air quality in India

728 Detailed evaluation of fuel use across India is required to evaluate the relative impact of  
729 emissions from fuel wood, municipal solid waste, cow dung cakes and LPG. The emission  
730 factors measured for cow dung cake and municipal solid waste in this study were much higher  
731 than for fuel wood and LPG and indicated that these sources are likely to contribute  
732 significantly to poor air quality.

733 The comprehensive characterisation of emissions from fuel types in this study should be used  
734 to produce spatially disaggregated local emission inventories to provide better inputs into  
735 regional policy and global chemical transport models. This should allow a better understanding  
736 of the key drivers of poor air quality in India and could allow meaningful mitigation strategies  
737 to alleviate the poor air quality observed.

738 *Author contributions.* GJS made measurements with GC×GC-FID, combined and analysed  
739 datasets and lead the writing of the manuscript. WJFA made measurements of NMVOCs by  
740 PTR-ToF-MS, supported by CNH, LKS and NT. BSN made measurements with DC-GC,  
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742 LY and SKS collected fuels, carried out burning experiments and measured gas volumes up  
743 the flue. RED worked on GC×GC-FID method development. SSMY assisted with data  
744 interpretation. EN, NM, RG, ARR and JDL worked on logistics and data interpretation. TKM  
745 and JFH provided overall guidance with setup, conducting, running and interpreting  
746 experiments. All authors contributed to the discussion, writing, and editing of the manuscript.

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