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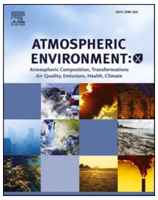

Non-methane volatile organic compounds emitted from domestic fuels in Delhi: Emission factors and total city-wide emissions

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ABSTRACT

In controlled laboratory conditions, 62 samples of domestic fuels collected from 56 grids of Delhi were burnt to quantify the emissions of 23 non-methane volatile organic compounds (NMVOCs), i.e., alkanes (11), alkenes (6), alkynes (1) and aromatic compounds (5). The domestic fuels used for residential activities were comprised of 20 unique types of fuel woods, 3 species of crop residue, dung cakes and coal. These fuels are primarily used for cooking and water/space heating during winters. The current study reports the total emission budget of NMVOCs from domestic burning over Delhi. Furthermore, this study also compares the differences in EFs of NMVOCs which are calculated for different burning cycles and sample collection methods. The EFs of NMVOCs calculated from the samples collected during the flaming stage using canisters were analysed for 23 NMVOCs and then compared with same species emitted from complete burning cycle. In addition to this, 10 consumption and emission hotspot grids were also identified in Delhi; based on the ground survey and laboratory simulated results. The total annual usage of domestic fuels for the year 2019 was found to be 0.415 Mt/yr (million tonnes) in Delhi. 12.01 Gg/yr of annual NMVOC emissions was calculated from domestic fuel burning in which the emissions from dung cake and fuel wood dominated with 6.6 Gg/yr and 5.4 Gg/yr, respectively. The EFs of NMVOCs calculated using canister and online collection method differ significantly from each other. The flaming stage presented enhanced emissions compared to the complete burning cycle by ~7 times which suggests that the method of data analysis and the period of sample collection play a pivotal role in the preparation of an emission inventory and estimating the budget.

1. Introduction

Around 2.8 billion people across the globe use solid biomass fuels (dung cake, crop residue, wood, charcoal and coal etc.) to meet their energy requirement for residential activities (Smith et al., 2013). It is a general practice among the less affluent population in developing/third-world nations of India, Bangladesh, Nepal, Bhutan, China, Brazil, and other African countries (Ewing and Msangi, 2009; Gheewala et al., 2013). Around 25–75% of primary energy requirement for cooking are met by the usage of domestic fuels (Smith et al., 2013; Stockwell et al., 2015, 2016). The national census (2011) assessed that 76% of rural households of India during 2009–2010 are primarily dependent on biomass fuels as a key source of energy for residential activities. A report by TERI Energy & Environment Data Diary and Yearbook (TEDDY), 2009, documented the usage of 252.1 Mt/yr of wood, 99.2 Mt/yr of crop residue, and 106.9 Mt/yr of dung cakes in

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India annually. Delhi alone consumed 0.7 Mt/yr of biomass fuels (0.3 Mt/yr dung cake, 0.2 Mt/yr wood and 0.2 Mt/yr crop residue) for cooking activities (Saud et al., 2012; D. P. Singh et al., 2013). The burning of domestic fuels for residential activities area primary source of particulate matter (PM) and gaseous pollutants such as CO, CO_2, NO_x, SO_x and VOCs (Park et al., 2013), which have significant impact on the atmospheric chemistry, climate change and human health (Estillore et al., 2016; Swart et al., 2004). The estimation of NMVOC emissions from domestic fuels is vital due to their significant role in the photochemistry and ozone formation in the lower troposphere (Atkinson and Arey, 2003). The alkene species of ethene, isoprene and propene are highly reactive and have a high photochemical ozone forming potential (Derwent et al., 1996; Fleming et al., 2018). Ground-level ozone is a strong oxidative agent and responsible for oxidative stress in living beings (Iriti and Faoro, 2007). Secondary organic aerosols (SOA) plays a pivotal role in the alteration of radiative balance of the earth due to constant absorption and scattering of solar radiation (Lin et al., 2016). The NMVOCs emitted from the combustion of domestic biofuels is known to aid in the formation of secondary organic aerosols (SOA). Gilman et al. (2015) reported that the polyunsaturated oxygenated NMVOCs to be major contributors in the formation of SOA while Bruns et al. (2016) estimated that ~80% of the total SOA produced from the combustion of fuelwood is due to phenol, naphthalene and benzene. The usage of biofuels for residential activities plays a vital role in enhancing the concentration of indoor particulate matter and pollutant gas by 10–20 times as compared to the ambient concentrations (Gordon et al., 2014). In a report published by the World Health Organisation (WHO), 2016, around 2.8 billion people were found to be exposed to the harmful emissions of solid biomass fuels from cooking and about 3.8 million premature deaths were attributed due to the household air pollution globally. Pandey and Venkataraman (2014) estimated that India emitted 4900 Gg of NMVOCs annually from residential cooking. Guttikunda and Calori (2013) estimated the emission of 17 Gg of VOCs from domestic activities over Delhi region for the year 2010.

The variation in the emission of NMVOCs from domestic fuels are dependent on multiple factors such as type of biofuels (Stewart and Stewart, 2008), the moisture content (Ni et al., 2015), dominance of different burning phases (Akagi et al., 2011; Andreae, 2019; Aurell and Gullett, 2013; Park et al., 2013), age of solid fuels (Nord-Larsen et al., 2011), method of burning (Fleming et al., 2018; Park et al., 2013), regional climatic condition/soil composition (Sirithian et al., 2018), the elemental composition of fuels (C, H, N and O), part of the plant burnt with cellulose, lignin, pentosane content (K. Singh et al., 2014) and the modified combustion efficiency (MCE) (Andreae, 2019; Aurell and Gullett, 2013). Hence, the estimation of EFs of NMVOCs using local fuels can critically help in reducing the uncertainties which may arise due to the above-mentioned conditions (Zhou et al., 2017).

The earlier reported inventories were generated using the consumption data of domestic fuels taken from a national sample survey and TEDDY reports which are generally produced with a coarse (spatial) resolution. The difference in the usage pattern varies with the socio-economic background of the slum/village dwellers and hence it can be identified by segregating the study area into smaller segments to lower the uncertainty. Further supporting details can be found in supplementary section S1. The present study attempts to reduce the above-mentioned uncertainties for the estimation of complete emission budget of NMVOCs.

The objectives of this study are to compare the differences between the EFs of 23 NMVOC species calculated using different burning cycles and sample collection methods for domestic fuels. The domestic fuels are used as cooking and water/space heating fuel. It is also used as mosquito repellent during the monsoon season. The EFs were calculated for two different periods analysed using two different methods. EF was calculated for the flaming stage, where the flue gas samples were collected using canisters and the analysis was done using GC-FID, whereas, EF was calculated for the complete burning cycle (ignition, flaming and smoldering) where the samples were collected directly and analysed using dual channel GC with FID (DC-GC-FID), comprehensive two-dimensional gas chromatography coupled to a FID (GC × GC-FID; online-method) and PTR-ToF-MS simultaneously (Stewart et al., 2020a). Using the values of EFs of NMVOCs from complete burning cycle. We calculated the total NMVOC emission budget from domestic fuels used in National Capital Territory (Delhi), India.

2. Methodology

2.1. Study sites and descriptions of survey

Delhi [28°12′–28°63′ N, 75°50′–77°23′ E] is the national capital territory (NCT) of India. As per the Census 2011 (https://www.census2011.co.in/census/state/districtlist/delhi.html, *last accessed March 27, 2020*) the total population of Delhi was 16,753,235 and the land area was 1484 km^2, which is distributed in 9 districts (Fig. 1a), namely north-west Delhi, west Delhi, south-west Delhi, north Delhi, north-east Delhi, east Delhi, south Delhi central & New Delhi. As per the Delhi Urban Shelter Improvement Board (DUSIB) 2015 report (http://delhishelterboard.in/main/wp-content/uploads/2015/12/675_JJ_Cluster_List.pdf, *last accessed March 27, 2020*), 636 slums areas were reported in the NCT. For this study, the whole area of NCT was divided into 66 grids of 25 km^2 area. A ground truthing survey revealed that the usage of domestic fuels is primarily dominated among lower-income group residents (slums and villages) as compared to the high income-group residents (colonies and societies). To classify the consumption pattern of domestic fuels, 695 locations (636 slums and 59 villages) were identified for a questionnaire-based field survey. The survey was conducted on 2–3% of residents (~6500 households) of the total population in each identified location and 148 domestic fuel samples were collected (see Fig. 1a). Based on their spatial distribution and fuel type, 62 samples (49 FW, 8 DC, 4 CR and 1 coal) were burnt under laboratory conditions (see Fig. 1b). Based on the survey result, it was found that 56 grids reported the use of domestic fuels for residential activities while 10 grids did not report any such consumption.

2.2. Experimental set-up

The dilution sampler provided the real conditions for burning as it allows sufficient dilution, cooling, and residence time during the experiment. The design of the dilution sampler is presented in supplementary Fig. S1 (Venkataraman and Rao, 2001). More information about the experimental setup can be found in Saud et al. (2012). Before weighing, each sample was dried outside under ambient conditions for 4 h to remove moisture naturally which was practiced by the residential fuel users as well. The samples were weighed in three replicates to reduce uncertainty in the weight of the residential fuels burnt. The laboratory combustion experiment was performed using 200g of each residential fuel sample. Gas samples from the flaming stage were collected in 1L stainless steel containers from the stack at the height of ~2.5m. The internal surface and valve of the canister were coated with inert silica to prevent the absorption of NMVOCs. The sample collection period was 10 min at the flow rate of 0.1 L/min during the flaming phase and analysis was carried out according to US-EPA Compendium method TO-15 (EPA, 1999). The NMVOC analysis was performed using a GC-FID instrument (make PerkinElmer, model Clarus 580). The calibration gas (Linde SPECTRA Environmental Gases, New Jersey, USA) contained 25 species: ethane, ethene, acetylene, propane, propene, isobutane, *1*-butene, *n*-butane, isopentane, *1*-pentene, *n*-pentane, isoprene, *1*-hexene, *n*-hexane, benzene, *n*-heptane, toluene, *n*-octane, ethylbenzene, *o*-xylene, nonane, isopropylbenzene, *n*-decane, *n*-undecane and *n*-dodecane at 100 ppb (±10%). More information about GC is attached in the supplementary section S2. To calculate the modified combustion efficiency (MCE), emissions of CO and CO_2 were analysed using flue gas analyser (make TESTO, model 350).

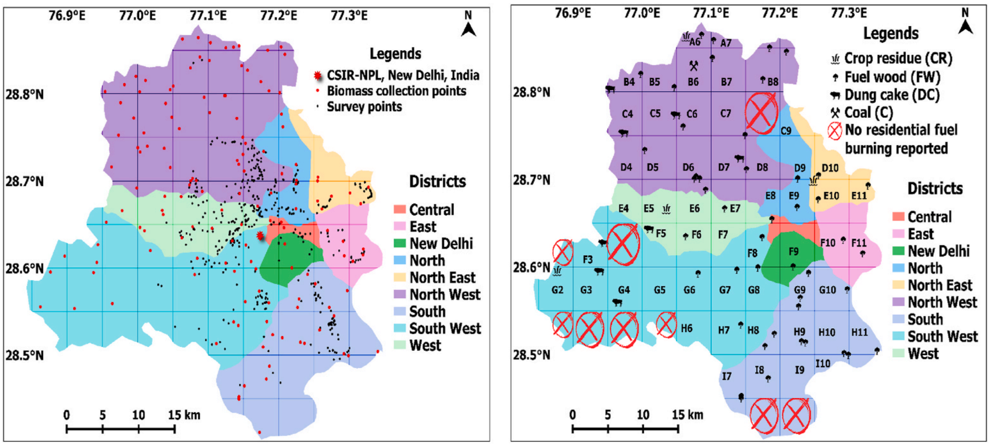


Fig. 1. The spatial distribution of (a) survey points and biomass collection points; (b) types of samples which were burnt as laboratory experiment.

2.3. Data analysis methods

The emission factor (EF) of a compound is defined as the amount of pollutant emitted from a unit weight of fuel burnt in g/kg (Andreae and Merlet, 2001). The EFs of the NMVOCs for domestic fuel combustion were determined based on flue gas volume and the mass concentration of pollutants using equation (1) (Park et al., 2013). Detailed information about each factor is given in the supplementary section S3.

$$EF_{ij}\left(\frac{g}{kg}\right) = \sum\left(\frac{C_i * V_j * A * T_j}{W_j}\right) \quad (1)$$

where.

EF_{ij} = Emission factor of *i*th NMVOC species of *j*th residential fuel sample
C_i = Concentration of *i*th NMVOC species (g/m^3)
V_j = flow rate of gas in stack (mid-point) for *j*th residential fuel sample (m/s)
A = cross-sectional area of stack (m^2)
T_j = sampling time for *j*th residential fuel sample (s)
W_j = weight of *j*th residential fuel sample burnt (kg)

The concentration of NMVOCs were performed using GC-FID and the flow rate, total sampling time, and weight of burned samples were recorded during the combustion (Park et al., 2013; Sen et al., 2014). The activity patterns of domestic fuel usage were calculated using questionnaires. Total emission from specific residential fuel was estimated using equation (2) (Shrestha et al., 2013).

$$TE_{ij} = \sum(F_j \times EF_{ij}) \quad (2)$$

where.

TE_{ij} = Total emission of *i*th *NMVOC* species from *j*th residential fuel sample (g)
F_j = Total annual consumption of *j*th residential fuel type (kg)
EF_{ij} = Emission factor specific to *i*th NMVOC species from *j*th residential fuel sample (g/kg)

The correlation matrix was generated to determine the co-emitter from domestic fuels (Supplementary Fig. S2). Detailed information about the samples can be found in supplementary Table ST1.

The EF of NMVOCs from the flaming stage and complete burning cycle (Stewart et al., 2020a) for the same residential fuels are compared in the present study. Stewart et al. (2020a) used GC × GC-FID and DC-GC-FID for the analysis of gas samples which were collected online throughout the burning period using a thermal desorption system. In addition, they also used a PTR-ToF-MS to expand the range of NMVOCs that could be measured. In contrast, canisters were used to collect gas samples during the initiation of burning (spanned roughly for 10 min; 100 mL per min; total 1 L) to cover the flaming phase in the current study. The uncertainty of the GC-FID instrument was ±10% while the lowest detection limit of the instrument was determined as 2SD (2 × standard deviation of background). In this paper, the EFs of 23 NMVOCs measured in the GC-FID are directly compared between the flaming and complete burn cycle. Stewart et al. (2020a, 2020b) reported a much wider range of EFs of NMVOCs (alkanes, alkenes, alkynes, aromatics, NVOCs, OVOCs, phenolics, furans and furanones), while this study reported only a fraction of the total NMVOCs measured (alkanes, alkenes, alkynes and aromatics).

3. Results and discussion

3.1. Comparison of emission factor between different stages of burning

Fig. 2 presents the comparison of EFs of NMVOCs from different domestic fuels both for the flaming stage and the complete burning cycle. The EFs of NMVOCs have been reported for each domestic fuel following which the average value was calculated. A wide range (2–41 g/kg) of EFs of NMVOCs was reported from the flaming stage of domestic fuel. Further details are summarized in Supplementary Table ST2.

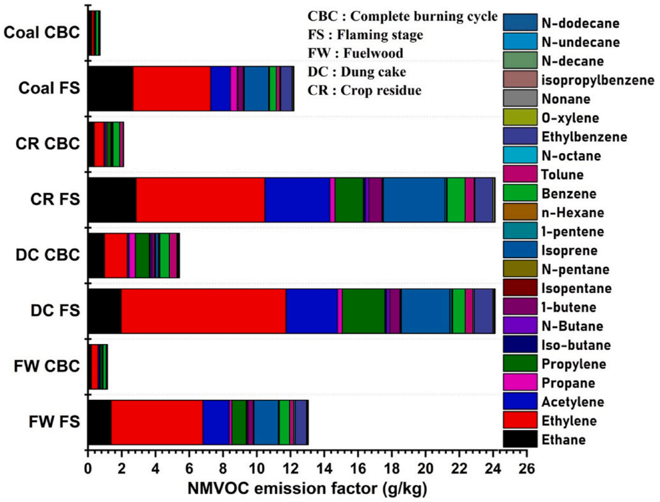


Fig. 2. EFs of measured NMVOCs emitted from domestic fuel burning during flaming stage and complete burning cycle.

3.2. Fuel wood (FW)

Fig. 3 shows the comparison of the measured EFs of NMVOCs from FW during the flaming stage and complete burning cycle. On average 88% increase was recorded in the EFs of NMVOCs during flaming stage as compared to complete burning cycle. The contribution of 10 most abundant NMVOCs (ethene, acetylene, isoprene, ethane, propene, ethylbenzene, benzene, 1-butene, toluene and propane) to the total EF was 97% during the flaming stage (Fig. 4a) and 96% during the complete burning cycle (Fig. 4b). Among all the FW species, *Azadirachta indica* was the highest emitter of ethane, propane, propene, 1-butene, 1-pentene during both the phases. Among different NMVOCs, ethene reported highest EF from all domestic fuels during both the burning phases.

The crop residues used as domestic fuels for residential burning activities were *Brassica nigra* (mustard), *Cocus nucifera* (coconut) and *Solanum melongena* (brinjal) in Delhi. Fig. 5 reports the comparison of EFs of NMVOCs from CR during flaming stage and complete burning cycle. The EFs of NMVOCs during flaming stage were on an average 87% higher than the complete burning cycle. The contribution of 10 most abundant NMVOCs (ethene, acetylene, isoprene, ethane, propene, benzene, ethylbenzene, 1-butene, propane and toluene) to total EF was 98% during the flaming stage (Fig. 6a) and 96% during the complete burning cycle (Fig. 6b).

Fig. 7 shows the comparison of the EFs of NMVOCs from flaming stage which were on an average 31% higher than the complete burning cycle for residential dung cake burning. The burning of dung cake with a lower combustion temperature and consistent burning mostly in smouldering phase may have resulted in similar emission rates during both the flaming stage and complete burning cycle. The contribution of the 10 most abundant NMVOCs (ethene, acetylene, isoprene, propene, ethane, ethylbenzene, benzene, 1-butene, toluene and propane) to the total EF was 97% for flaming stage (see Fig. 8a) and 92% for complete burning cycle (see Fig. 8b).

Fig. 9 presents the comparison of EFs of NMVOCs during the flaming stage and the complete burning cycle, where the EF of NMVOCs during flaming stage was found to be an average 73% higher than complete burning cycle. The contribution of the 10 most abundant NMVOCs (ethene, ethane, isoprene, acetylene, ethylbenzene, benzene, propane, 1-butene, toluene and *o*-xylene) to the total EF was 98% during the flaming stage (see Fig. 10a) and 93% during the complete burning cycle (see Fig. 10b).

The variations in the emission of NMVOCs from different fuel types are primarily due to several factors including the CHNS composition of the residential fuels, chemical nature and anatomical structure of the fuel species(K. Singh et al., 2014). Dung cake and crop residue were found to emit higher amounts of NMVOCs upon burning as compared to the fuelwood species and coal.

3.3. Comparison of flaming stage emission factor with other studies

We have compared the EF from the flaming stage with earlier reported values. Since numerous techniques have been used to burn domestic biomass fuels and different NMVOC species have been reported in various studies, it is very difficult to compile and compare the present values with earlier reported values. However, an attempt has been made to compare the emission of similar NMVOC species from various studies with the present data to understand the variations in EF of NMVOCs from domestic fuels.

Fig. 11 depicts the comparison of EFs of NMVOCs between the present study and earlier reported studies. The EFs of ethane, ethene, acetylene and isoprene from FW in this study are higher than the values for the tropical forest (Andreae, 2019; Andreae and Merlet, 2001), boreal forest (Simpson et al., 2011) and coniferous forest (Stockwell et al., 2015). However, the EFs of ethane and acetylene are comparable with open traditional cooking (Akagi et al., 2011; Bertschi et al., 2003; Fleming et al., 2018; Yokelson et al., 2011). The EFs of benzene, toluene, ethylbenzene propane, propylene, and 1-butene from FW (0.1–0.9 g/kg) almost match with most of the earlier reported values except Verma et al. (2019) and Sun et al. (2019). The EFs of NMVOCs and MCE values (0.93–0.96) from traditional cooking method with single stove (Akagi et al., 2011; Bertschi et al., 2003; Fleming et al., 2018) and laboratory condition with multiple FW species (Stockwell et al., 2015) reported resemblances with this study as compared to the field experiments on a forest fire (Andreae, 2019; Andreae and Merlet, 2001; Simpson et al., 2011; Yokelson et al., 2011). The present study reported highest resemblances with Fleming et al. (2018). In conclusion, the factors

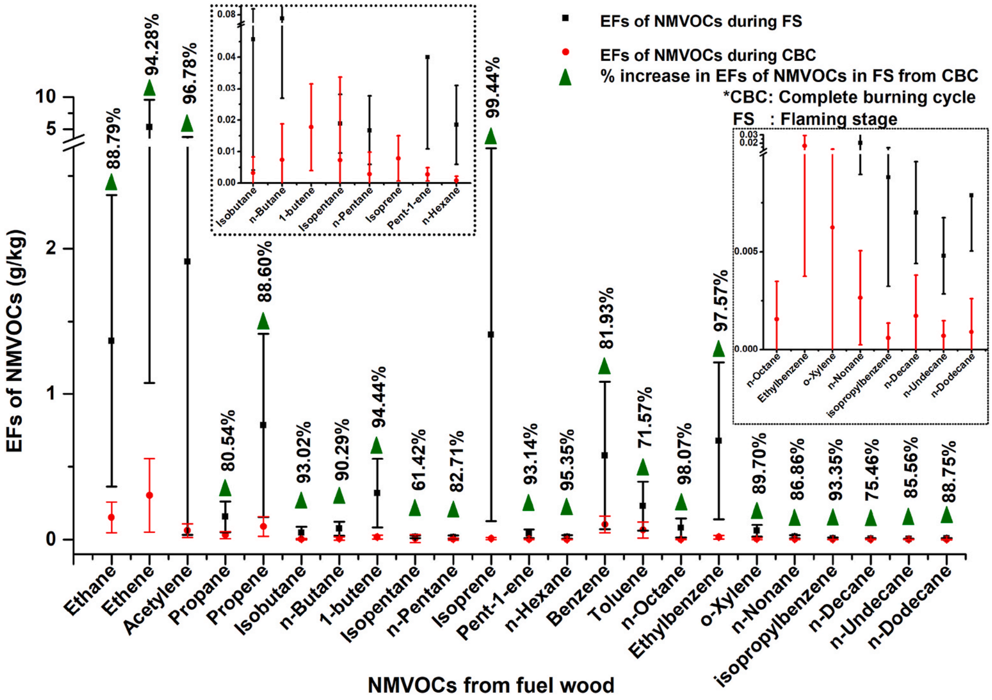
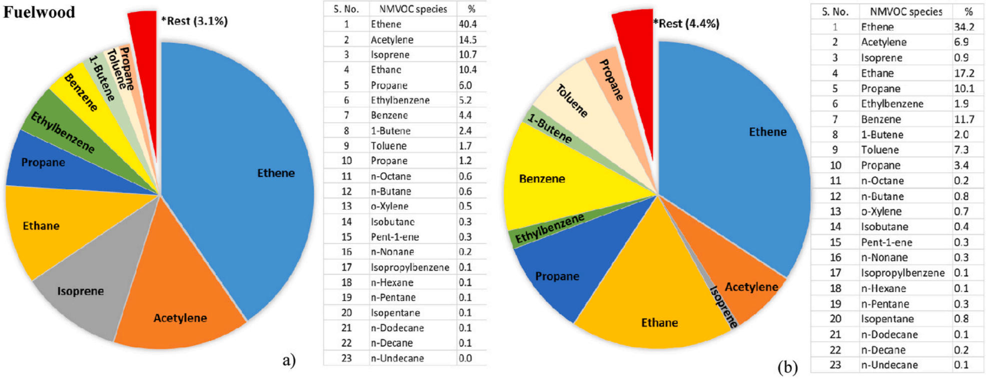


Fig. 3. Comparison between EFs of measured NMVOCs from FW during flaming stage and complete burning cycle.



S. No.	NMVOC species	%
1	Ethene	40.4
2	Acetylene	14.5
3	Isoprene	10.7
4	Ethane	10.4
5	Propane	6.0
6	Ethylbenzene	5.2
7	Benzene	4.4
8	1-Butene	2.4
9	Toluene	1.7
10	Propane	1.2
11	n-Octane	0.6
12	n-Butane	0.6
13	o-Xylene	0.5
14	Isobutane	0.3
15	Pent-1-ene	0.3
16	n-Nonane	0.2
17	Isopropylbenzene	0.1
18	n-Hexane	0.1
19	n-Pentane	0.1
20	Isopentane	0.1
21	n-Dodecane	0.1
22	n-Decane	0.1
23	n-Undecane	0.0

S. No.	NMVOC species	%
1	Ethene	34.2
2	Acetylene	6.9
3	Isoprene	0.9
4	Ethane	17.2
5	Propane	10.1
6	Ethylbenzene	1.9
7	Benzene	11.7
8	1-Butene	2.0
9	Toluene	7.3
10	Propane	3.4
11	n-Octane	0.2
12	n-Butane	0.8
13	o-Xylene	0.7
14	Isobutane	0.4
15	Pent-1-ene	0.3
16	n-Nonane	0.3
17	Isopropylbenzene	0.1
18	n-Hexane	0.1
19	n-Pentane	0.3
20	Isopentane	0.8
21	n-Dodecane	0.1
22	n-Decane	0.2
23	n-Undecane	0.1

Fig. 4. Percentage contribution of EFs of measured NMVOCs from FW during (a) flaming stage; (b) complete burning cycle.

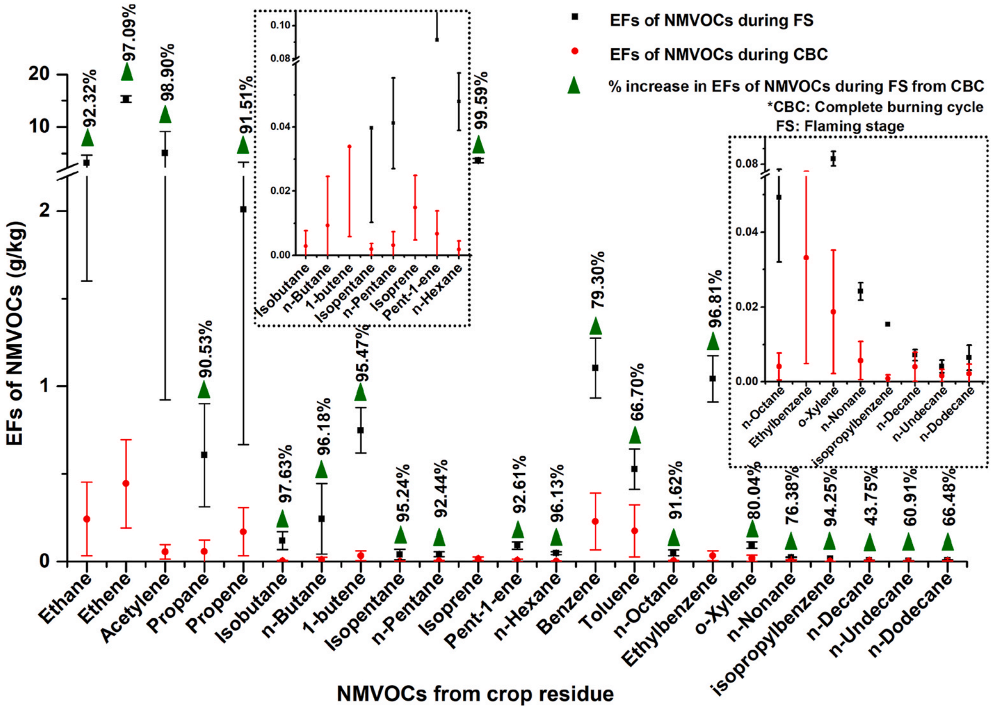
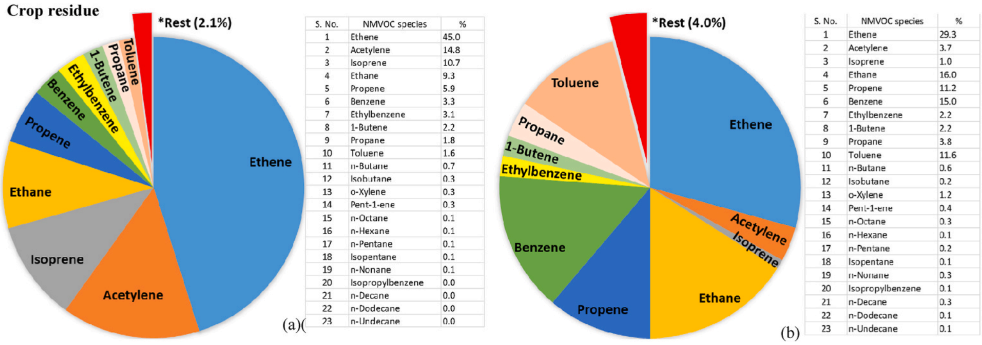


Fig. 5. Comparison between EFs of measured NMVOCs from CR during flaming stage and complete burning cycle.



S. No.	NMVOC species	%
1	Ethene	45.0
2	Acetylene	14.8
3	Isoprene	10.7
4	Ethane	9.3
5	Propene	5.9
6	Benzene	3.3
7	Ethylbenzene	3.1
8	1-Butene	2.2
9	Propane	1.8
10	Toluene	1.6
11	n-Butane	0.7
12	Isobutane	0.3
13	o-Xylene	0.3
14	Pent-1-ene	0.3
15	n-Octane	0.1
16	n-Hexane	0.1
17	n-Pentane	0.1
18	Isopentane	0.1
19	n-Nonane	0.1
20	Isopropylbenzene	0.0
21	n-Decane	0.0
22	n-Dodecane	0.0
23	n-Undecane	0.0

S. No.	NMVOC species	%
1	Ethene	29.3
2	Acetylene	3.7
3	Isoprene	1.0
4	Ethane	16.0
5	Propene	11.2
6	Benzene	15.0
7	Ethylbenzene	2.2
8	1-Butene	2.2
9	Propane	3.8
10	Toluene	11.6
11	n-Butane	0.6
12	Isobutane	0.2
13	o-Xylene	1.2
14	Pent-1-ene	0.4
15	n-Octane	0.3
16	n-Hexane	0.1
17	n-Pentane	0.2
18	Isopentane	0.1
19	n-Nonane	0.3
20	Isopropylbenzene	0.1
21	n-Decane	0.3
22	n-Dodecane	0.1
23	n-Undecane	0.1

Fig. 6. Percentage contribution of EFs of measured NMVOCs from CR during (a) flaming stage; (b) complete burning cycle.

determining the EFs of NMVOCs from FW were dependent on type of stove, MCE and sample collection method. Further details are summarized in supplementary Table ST3.

ANOVA analysis was performed within 20 FW species for 11 abundant NMVOCs (benzene, toluene, ethylbenzene, ethane, ethene, propane, propylene, isoprene, isopropylbenzene, acetylene and *o*-xylene) out of which only 2 NMVOCs (acetylene and *o*-xylene) reported a significant difference in mean EF within fuelwood species while the

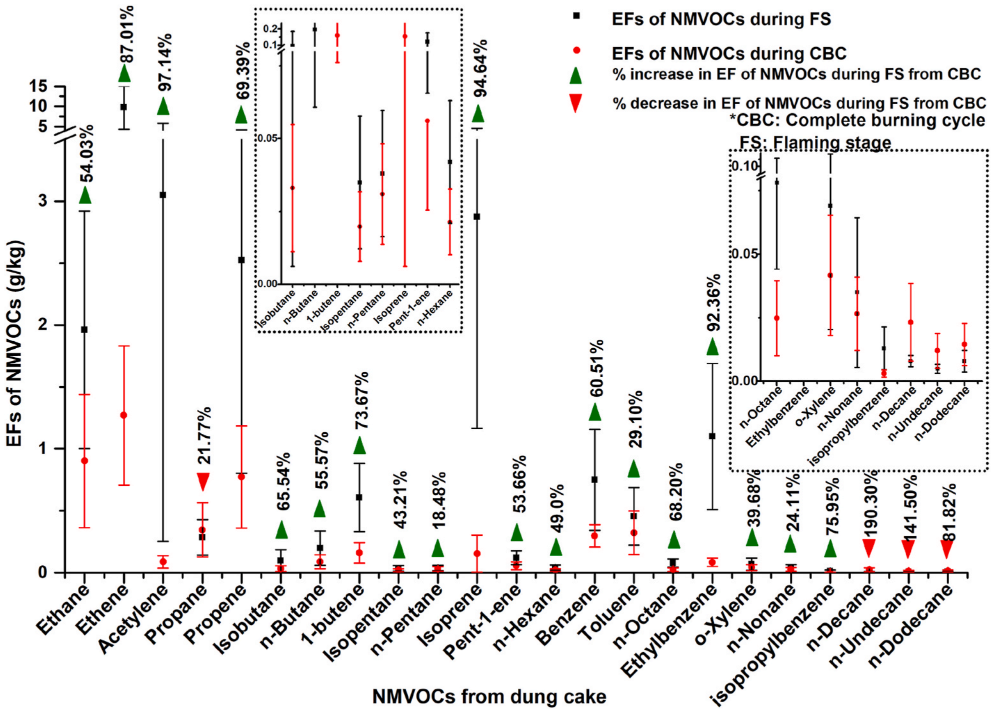
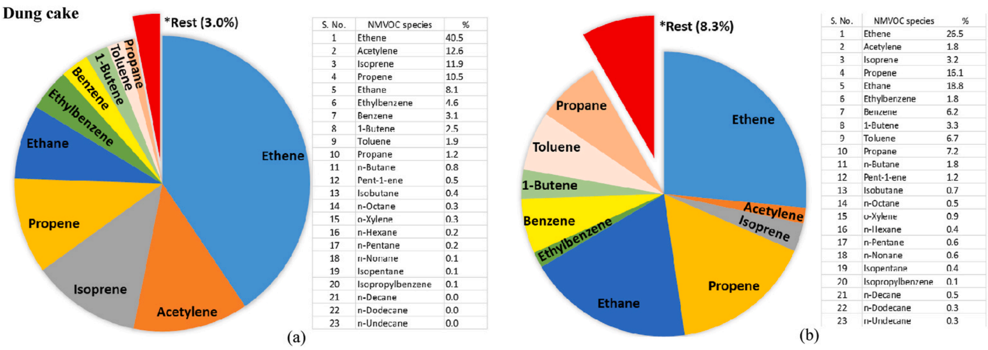


Fig. 7. Comparison between EFs of measured NMVOCs from DC during flaming stage and complete burning cycle.



S. No.	NMVOC species	%
1	Ethene	40.5
2	Acetylene	12.6
3	Isoprene	11.9
4	Propene	10.5
5	Ethane	8.1
6	Ethylbenzene	4.6
7	Benzene	3.1
8	1-Butene	2.5
9	Toluene	1.9
10	Propane	1.2
11	n-Butane	0.8
12	Pent-1-ene	0.5
13	Isobutane	0.4
14	n-Octane	0.3
15	o-Xylene	0.3
16	n-Hexane	0.2
17	n-Pentane	0.2
18	n-Nonane	0.1
19	Isopentane	0.1
20	Isopropylbenzene	0.1
21	n-Decane	0.0
22	n-Dodecane	0.0
23	n-Undecane	0.0

(a)

S. No.	NMVOC species	%
1	Ethene	26.5
2	Acetylene	1.8
3	Isoprene	3.2
4	Propene	16.1
5	Ethane	18.8
6	Ethylbenzene	1.8
7	Benzene	6.2
8	1-Butene	3.3
9	Toluene	6.7
10	Propane	7.2
11	n-Butane	1.8
12	Pent-1-ene	1.2
13	Isobutane	0.7
14	n-Octane	0.5
15	o-Xylene	0.9
16	n-Hexane	0.4
17	n-Pentane	0.6
18	n-Nonane	0.6
19	Isopentane	0.4
20	Isopropylbenzene	0.1
21	n-Decane	0.5
22	n-Dodecane	0.3
23	n-Undecane	0.3

(b)

Fig. 8. Percentage contribution of EFs of measured NMVOCs from DC during (a) flaming stage; (b) complete burning cycle.

rest showed no significant differences (Friedman test, $p > 0.05$). The post hoc test reported a significant difference in mean EFs of acetylene (*Melia azedarach-Saraca asoca, Vachellia nilotica-Saraca asoca, Mangifera indica-Saraca asoca, Terminalia catappa-Saraca asoca, Raw Plywood-Saraca asoca, Morus alba-Saraca asoca, Eucalyptus spp-Saraca asoca and Eucalyptus spp-Ficus religiosa*) and *o*-xylene (*Saraca asoca-Ricinus communis, Acacia nilotica-Saraca asoca, Vachellia nilotica-Saraca asoca, Raw Plywood-Vachellia nilotica and Raw Plywood-Acacia nilotica*). Information about ANOVA and post hoc is summarized in supplementary Table ST7-8.

Fig. 12 shows the comparison of EF of NMVOCs from crop residues with earlier reported values. Among crop residue (CR) species, the *rape* and *Brassica nigra* (mustard) belong to the same family. Wang et al. (2014) reported the emission profile for alkanes(*ethane* > *propane* > *n-butane*) and for the aromatic compounds (*benzene* > *toluene* > *ethylbenzene*) during the burning of *rape*. The profile for *Brassica nigra* in this study resembles with Wang et al. (2014) except for ethylbenzene. This study reported higher EFs of NMVOCs than previously reported values for agriculture/crop residue burning (wheat, rice maze bine, rape, mix CR burning etc.) in U.S., Germany, Mexico, Nepal, and Thailand (Akagi et al., 2011; Andreae, 2019; Andreae and Merlet, 2001; Stockwell et al., 2015, 2016; Yokelson et al., 2011). Verma et al. (2019) reported higher EFs of benzene (17-fold) and toluene (2-fold) from CR than the present study. Ciccioli et al. (2001) have shown the highest similarities with the present study as compared to the field experiments studies. The present study and earlier reported observations reported MCE value in the range of 0.92–0.96 (supplementary Table ST3-5). This study is designed to understand the emission from those CR species that are utilized for residential energy requirements. The correlation matrix shows negative correlation among the EFs of NMVOCs from CR burning (supplementary Fig. S3). The negative correlation indicates that the EF of NMVOCs vary significantly among the CR species which might have occurred due to differences in CHNS composition of the species burnt (K. Singh et al., 2014).

Fig. 13 shows the comparison of EF of NMVOCs from dung cake with earlier reported values. The emission profile for alkenes (*ethene* > *propene*>*1-butene* > *isoprene*>*1-pentene*) reported by other studies (Andreae, 2019; Fleming et al., 2018; Stockwell et al., 2016) almost coincided with the present study except for isoprene. Similarly, the emission profile for aromatics (*benzene* > *toluene* > *ethylbenzene* > *o-xylene*) reported by other studies (Fleming et al., 2018; Stockwell et al., 2016) almost overlapped with this study except for ethylbenzene. The

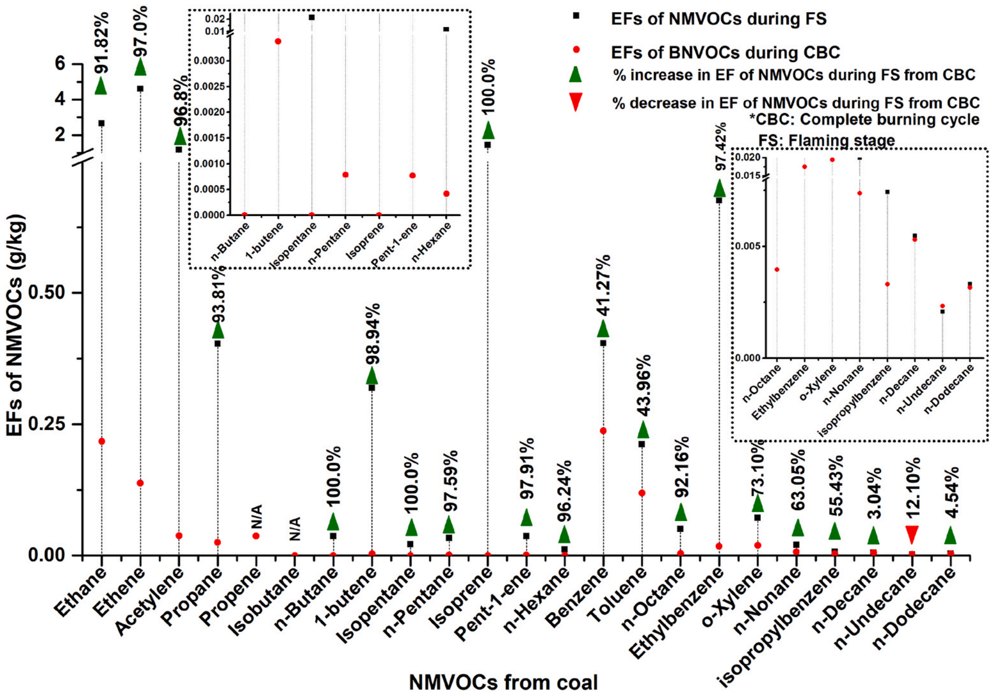
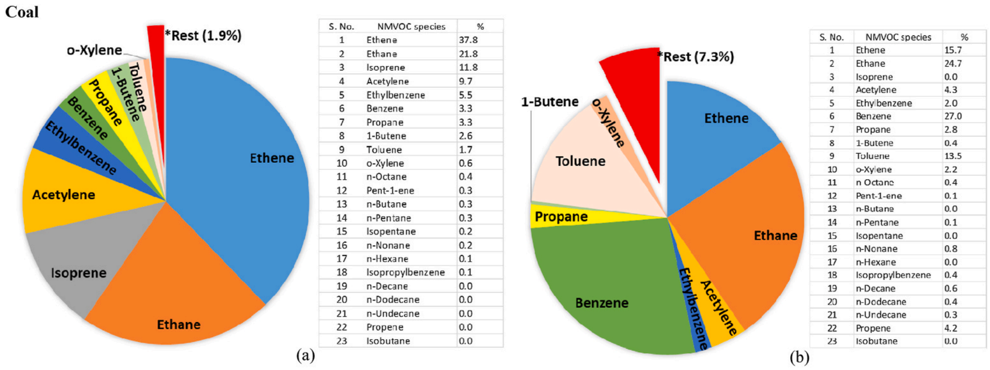


Fig. 9. Comparison between EFs of measured NMVOCs from coal during flaming stage and complete burning cycle.



S. No.	NMVOC species	%
1	Ethene	37.8
2	Ethane	21.8
3	Isoprene	11.8
4	Acetylene	9.7
5	Ethylbenzene	5.5
6	Benzene	3.3
7	Propane	3.3
8	1-Butene	2.6
9	Toluene	1.7
10	o-Xylene	0.6
11	n-Octane	0.4
12	Pent-1-ene	0.3
13	n-Butane	0.3
14	n-Pentane	0.3
15	Isopentane	0.2
16	n-Nonane	0.2
17	n-Hexane	0.1
18	Isopropylbenzene	0.1
19	n-Decane	0.0
20	n-Dodecane	0.0
21	n-Undecane	0.0
22	Propene	0.0
23	Isobutane	0.0

(a)

S. No.	NMVOC species	%
1	Ethene	15.7
2	Ethane	24.7
3	Isoprene	0.0
4	Acetylene	4.3
5	Ethylbenzene	2.0
6	Benzene	27.0
7	Propane	2.8
8	1-Butene	0.4
9	Toluene	13.5
10	o-Xylene	2.2
11	n Octane	0.4
12	Pent-1-ene	0.1
13	n-Butane	0.0
14	n-Pentane	0.1
15	Isopentane	0.0
16	n-Nonane	0.8
17	n-Hexane	0.0
18	Isopropylbenzene	0.4
19	n-Decane	0.6
20	n-Dodecane	0.4
21	n-Undecane	0.3
22	Propene	4.2
23	Isobutane	0.0

(b)

Fig. 10. Percentage contribution of EFs of measured NMVOCs from coal during (a) flaming stage; (b) complete burning cycle.

EFs of NMVOCs from DC using *Chulha* reported by Fleming et al. (2018) have shown the highest similarities with the present study. The reported MCE values from earlier literatures were in the range of 0.88–0.95 (supplementary Table ST5).

In a nutshell, the comparison of average EFs of NMVOCs from domestic fuels obtained in the present study were compared with several studies done by different methodologies and different burning cycles are given in Fig. 14. The EFs of NMVOCs at flaming stage from domestic fuels in the present study shows highest similarities with Fleming et al. (2018) (supplementary Table ST6). This could be due to several reasons e.g., the similar CHNS composition of domestic fuels due to the close proximity of sample locations (around 200 km), similar MCE values (0.93–0.94), type of burning (all sample burnt in each study on the single stove either in a laboratory or open *Chulha* in the field) and number of domestic fuel samples (Fleming et al. (2018), n = 45; present study, n = 62). Another study by Stockwell et al. (2016) also showed close resemblances with the present values due to consideration of CR, FW, DC for EFs of NMVOCs by both studies although slight variations might have occurred due to the different study areas (India and Nepal), different MCE values and method of burning. The present study shows large difference in EFs of NMVOCs from domestic fuels from other studies (Akagi et al., 2011; Andreae, 2019; Andreae and Merlet, 2001; Ciccioli et al., 2001; Stockwell et al., 2015; Sun et al., 2019; Yokelson et al., 2011) because of high variation within fuel species, the number of samples burnt, type of fuels, method of burning and sampling, MCE values and difference in sample collection sites. Verma et al. (2019), reported highest values of emission of benzene and toluene as compared to other studies which might be due to the usage of kerosene to initiate the combustion process. Almost all the studies reported the highest EF for ethene. Out of 11 abundant NMVOC species (benzene, toluene, ethylbenzene, *o*-xylene, isopropyl-benzene, ethane, ethene, propane, propylene, isoprene and acetylene), only two NMVOC species i.e., propylene and isoprene had shown a significant difference in mean value of EFs of NMVOCs at $p < 0.05$. However, the post hoc analysis revealed that only propylene reported significant difference at $p < 0.05$ between FW-DC but isoprene had not reported any significant difference at $p < 0.05$ in mean EFs of NMVOCs between fuel types (supplementary Table ST9-10).

3.4. Biomass consumption pattern over Delhi

Biomass usage pattern have been defined on the basis of the survey

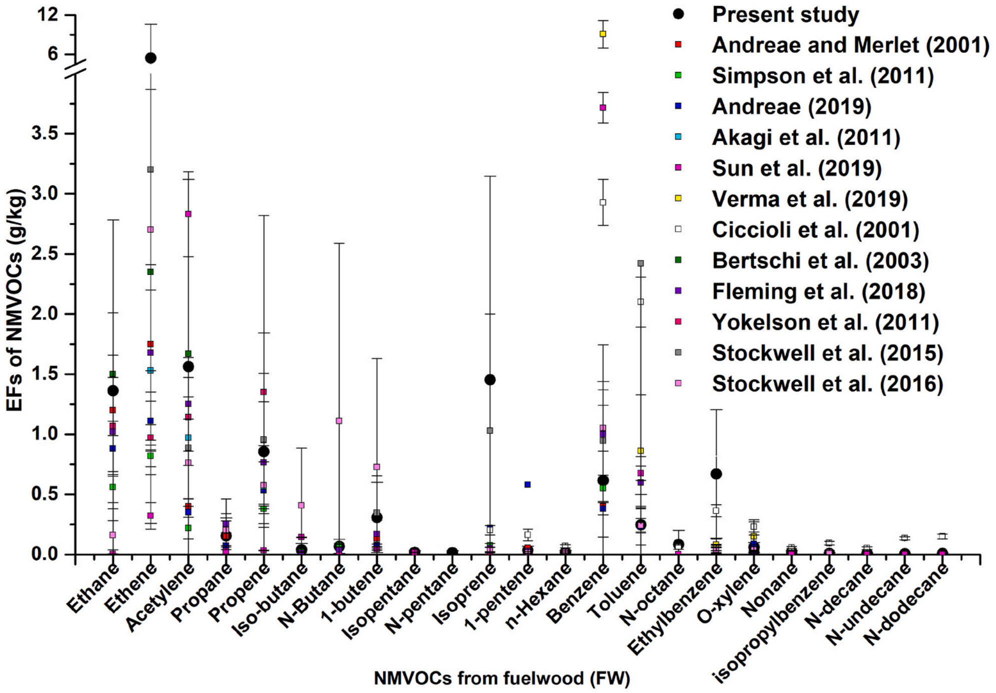


Fig. 11. Comparison of EFs of measured NMVOCs from fuel wood.

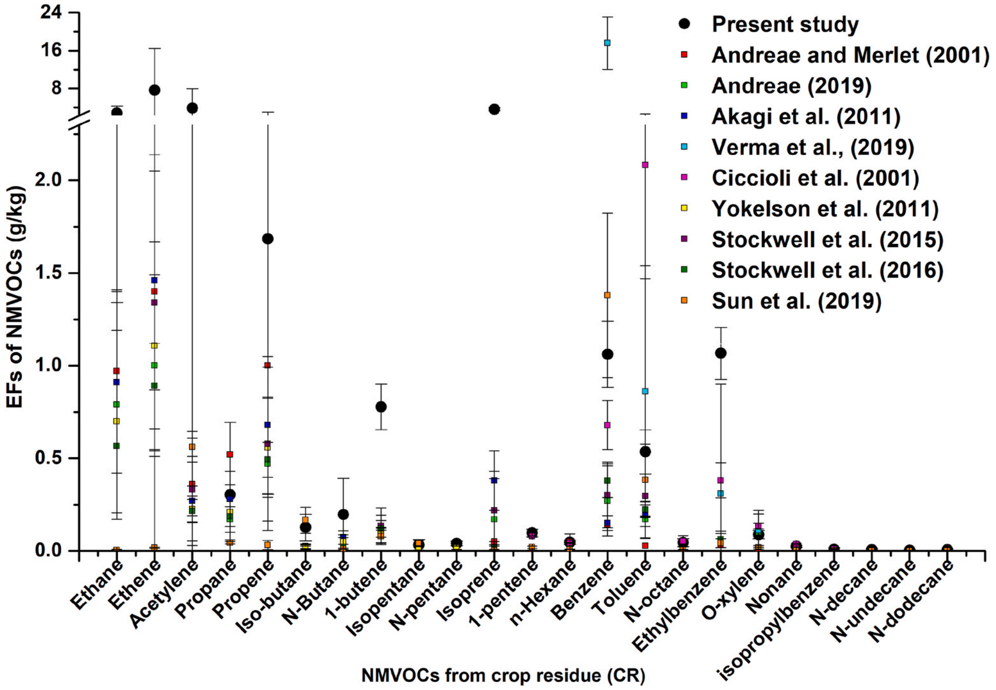


Fig. 12. Comparison of EFs of measured NMVOCs from crop residue.

results. A total of 6498 households were surveyed where 4567 households (~72%) were found to rely on LPG only. The remaining 1931 households (~28%) relied on traditional stoves for household activities out of which 19% of households were completely dependent on domestic fuels and the rest 9% were found to use both traditional stove as well as LPG. The respondents were asked about the amount of domestic fuels used for cooking at one time. The fuels were then weighed using spring balance to estimate the total weight of domestic fuel used per meal. It was then multiplied with total number of meals cooked in a day to estimate the consumption of domestic fuels per day per household. The spatial distribution of annual consumption of different domestic fuels over the 9 districts of NCT is reported in Table 1. The estimated annual consumption of FW, DC, CR, coal in the year 2019 was 0.300 Mt/yr (~72.5%), 0.112 Mt/yr (~26.8%), 0.002 Mt/yr (~0.48%), 0.0008 Mt/yr (~0.2%) respectively which resulted in a total of 0.415 Mt/yr over the NCT. The highest and lowest consumption of domestic fuels was reported in the north-west Delhi and central Delhi districts respectively. Only few rural areas from four districts reported the use of CR. Coal consumption was reported in two districts of north Delhi and north-west Delhi. Previous literatures (Gurjar et al., 2004; Saud et al., 2012; D. P.

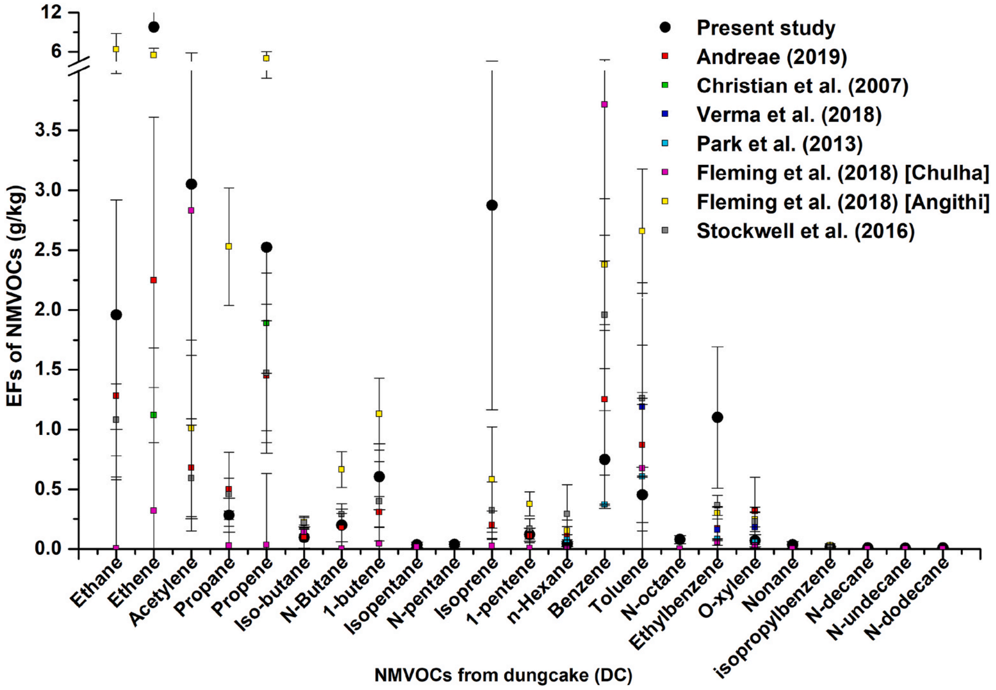


Fig. 13. Comparison of EFs of measured NMVOCs from dung cake.

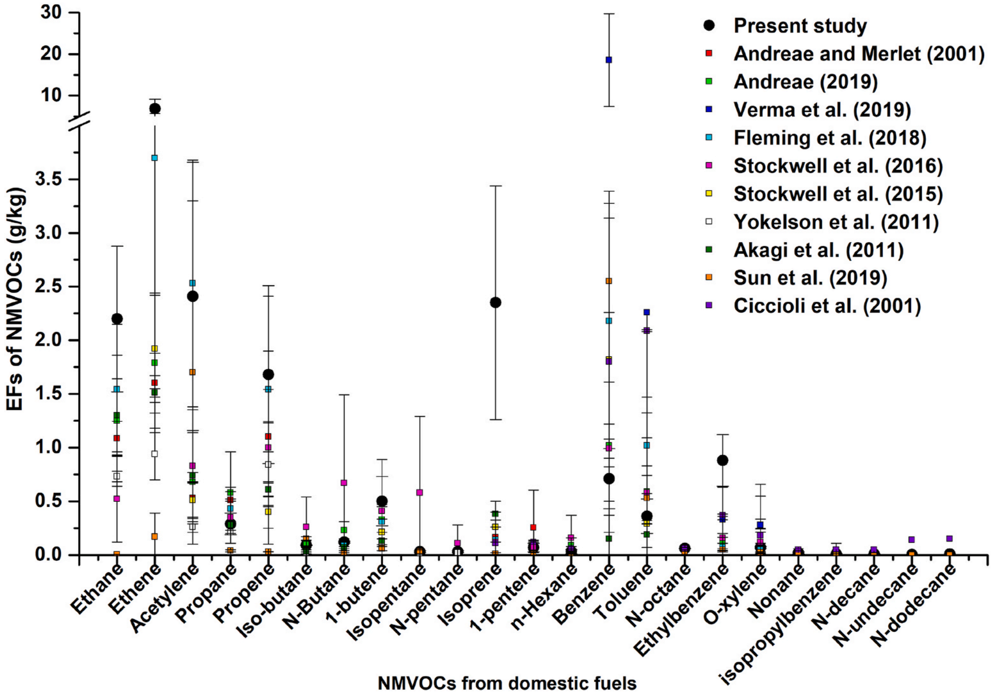


Fig. 14. Comparison of EFs of NMVOCs from domestic fuel.

Singh et al., 2013) reported higher consumption of DC and CR as compared to the present study but the consumption of FW was comparatively lower than the present study (supplementary Table ST10). The domestic fuel usage might have lowered due to some government policies for below poverty line (BPL) individuals under which an LPG connection is provided at a subsidized rate to improve the socio-economic status of the BPL people of NCT.

Fig. 15 presents the domestic fuel consumption hot spot grids (top 10 grids) for NCT of Delhi and supplementary Table ST11 shows the spatial distribution of hot spots for different types of domestic fuels. Around 50% of the total consumption of domestic fuels was reported by these 10 grids, hence the grids can be considered as consumption hot spots (CHS). The CHS were distributed in 6 districts (north-east, north-west, west, north, south, south-west) with134 locations (~19%). E10 was the highest consumption grid, located in north-east Delhi. The spatial distribution for top CHS were located in west Delhi (3 grids) at 40 locations followed by north-west Delhi (3 grids) with 35 locations.

This study also evaluated the consumption pattern for clean fuel (LPG) and domestic fuels over CHS regions (supplementary Table ST12). 83% of the households from the grid E10 (highest consumption grid)

Table 1
Annual consumption of different types of domestic fuels for residential purposes in Delhi.

S. No.	District	FW usage (Mt)	DC usage (Mt)	CR usage (Mt)	Coal usage (Mt)	Total usage residential fuels (Mt)
1	North-west	0.0742	0.0580	0.0001	0.0008	0.1331
2	West	0.0571	0.0307	0.0017		0.0895
3	South-west	0.0354	0.0141	0.0002		0.0498
4	South	0.0345	0.0007			0.0352
5	North	0.0398	0.0023		0.0000004	0.0421
6	North-east	0.0397	0.0056	0.000003		0.0453
7	East	0.0086				0.0086
8	Central	0.0028				0.0028
9	New-Delhi	0.0080	0.00004			0.0080
	Total	0.30007	0.11154	0.00207	0.00084	0.4145

FW: fuel wood; DC: dung cake; CR: crop residue; Mt: Million tonnes

reported to use clean energy (LPG) while 16.3% of the residents used domestic fuels. Less than 1% of the households in grid E10 used mixed fuel for the residential activities. This study also revealed that many households use traditional stoves in spite of having an LPG connection which might be due to cheaper access to domestic fuels, difficulties in refilling the cylinder due to economic constraints and lack of awareness about the health threats due to the use of domestic fuels.

3.5. *Budget estimate*

Using the values of EFs of NMVOCs from Stewart et al. (2020a) the total emission of NMVOCs from domestic fuel burning over Delhi for the year 2019 was found to be 12.01 Gg/yr out of which 5.4 Gg/yr NMVOC were being emitted from fuelwood while 6.6 Gg/yr of NMVOC were found to be the resultant of dung cake burning. Crop residue and charcoal burning accounted for 0.04 Gg/yr and 0.005 Gg/yr of NMVOCs respectively. Estimates of the annual emissions of NMVOCs from domestic fuels over Delhi have been reported by quite a few studies (Gurjar et al., 2004; Guttikunda and Calori, 2013; Jain et al., 2014) often with an undefined number of NMVOC species included. Hence, it was difficult to directly compare the variations in total annual NMVOC emissions for different years. Gurjar et al. (2004) reported annual emission of NMVOC to be 12.20 Gg/yr from domestic fuels (fuel wood 2.7 Gg/yr, crop residue 6.56 Gg/yr, dung cake 2.94 Gg/yr) for the year 2000. Jain et al. (2014) reported total NMVOC emission from crop residues which include open burning as well as residential usage to be 0.26 Gg/yr for 2009. Guttikunda and Calori (2013) reported total NMVOC emission in the order of 17.3 Gg/yr from residential activities using domestic fuel (kerosene, LPG, solid biofuel) for the year 2010 using a modelling approach.

The grids E4, D4, E10, B4, E5, D7, C6, B8, E11 and F3 are the highest NMVOC emitting grids from domestic fuel usage over Delhi for the year 2019 (Fig. 16). These 10 grids contributed to the emission of 6.24 Gg/yr which makes up the ~52% of the total NMVOCs emitted over whole Delhi.

The total NMVOC emissions are primarily dependent on two factors- (i) amount of consumption of residential fuels and (ii) EF of NMVOCs of the residential fuels used. Most of the hotspot grids reported high NMVOC emissions due to higher consumption of residential fuels. Certain grids with high usage of CR and DC as residential fuels also reported higher NMVOC emissions as the values of EF of NMVOCs from CR and DC are relatively higher than that of the FW species.

4. Conclusion

A large variation in EF of NMVOCs was observed while comparing the values derived from the complete burning cycle and the flaming stage. EFs of NMVOC during flaming stage are 88% higher than complete burning cycle in case of FW and CR, whereas, 33% for DC and 73% for coal. Among all FWs, *Azadirachta indica* was the highest emitter of ethane, propane, propene, 1-butene, 1-pentene during both phases. DC and CR were higher emitters of NMVOCs as compared to FW and coal.

The present study resembles the values reported by Fleming et al. (2018) and Stockwell et al. (2016). It might be due to similar sample collection methods (samples collected using canisters) which was limited to a certain time period and specific burning phase. Closer proximity ensured that the domestic fuels used for residential activities

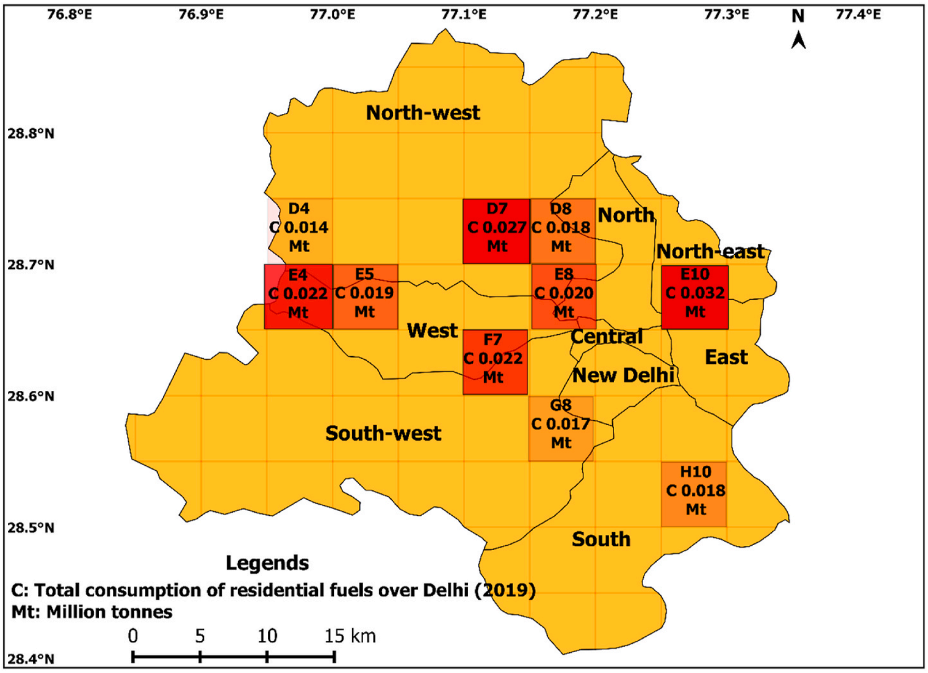


Fig. 15. Gridded distribution over NCT-Delhi for consumption hotspots.

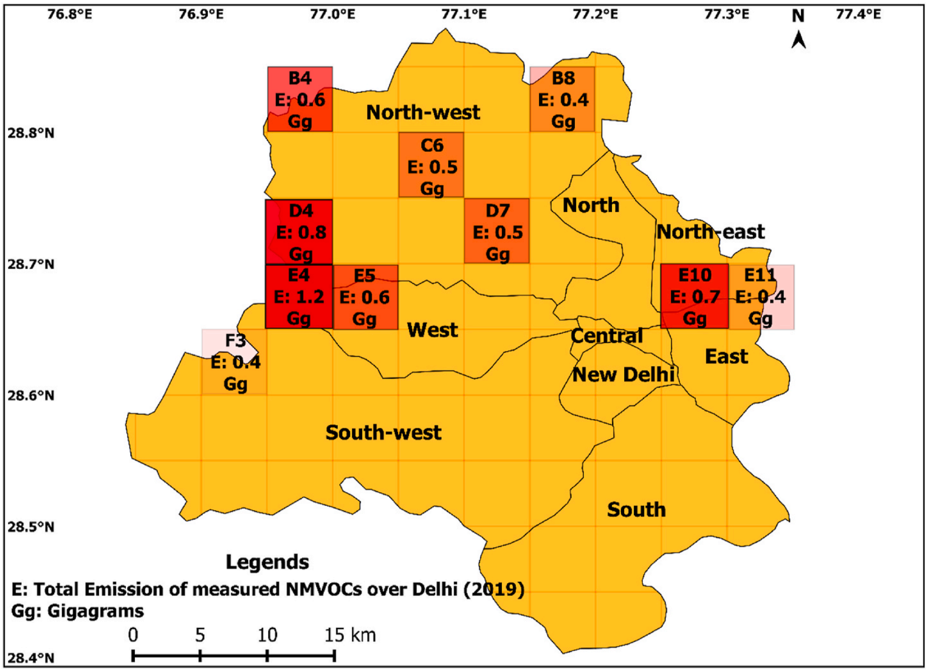


Fig. 16. Gridded distribution over NCT-Delhi for emission hotspots.

have been grown in similar climatic conditions.

ANOVA analysis, which was performed within 20 FW species for 11 abundant NMVOC species suggests that only 2 NMVOCs (acetylene and *o*-xylene) reported significant difference in mean EF within FW species. The negative correlation among EFs of NMVOCs from CR burning indicates that EF of NMVOCs vary significantly among CR species.

The annual consumption of domestic fuels in the national capital region of Delhi is about 0.415 Mt/yr of which 0.3 Mt/yr (~72%) fuel wood, 0.11 Mt/yr (~27%) dung cake, 0.002 Mt/yr (~0.5%) crop residue and 0.001 Mt/yr (~0.2%) coal are used. North-west district of Delhi reported the highest consumption of domestic fuel annually, which is about 32% of the total annual usage of Delhi. 92% of domestic fuels are being used for cooking purposes, followed by 7% for heating of water during winter season, 1% for space heating during winters and 0.5% of the total domestic fuels are used as mosquito repellent over Delhi.

Using EFs of NMVOCs from Stewart et al. (2020a), total emission of NMVOCs from domestic fuel burning over Delhi for the year 2019 was 12.01 Gg/yr. FW (5.4 Gg/yr) and DC (6.6 Gg/yr) are major emitters of NMVOC as compared to CR (0.04 Gg/yr) and charcoal (0.005 Gg/yr). The total emission of 23 reported NMVOCs calculated using the EF from the flaming stage was found to be ~7 times higher than that of the complete burning cycle. If we consider only one stage of burning cycle e.g., flaming stage, it could mislead the budget estimation representing the upper limit of the emission. Hence the EFs of NMVOCs from complete burning cycle should be used to calculate the total emission budget.

CRediT authorship contribution statement

Arnab Mondal: collected samples, carried out the experiment, analysed the samples using GC-FID and led the manuscript preparation. **Ummed Singh Saharan:** carried out the statistical analysis and contributed to the manuscript. **Rahul Arya:** participated in the survey, collected samples, participated in the experiment and contributed to the manuscript. **Lokesh Yadav:** participated in the survey, collected samples, participated in the experiment and contributed to the manuscript. **Sakshi Ahlawat:** participated in the survey, collected samples, participated in the experiment and contributed to the manuscript. **Ritu Jangirh:** participated in the survey, collected samples, participated in the experiment and contributed to the manuscript. **Garima Kotnala:** participated in the survey and collection of the samples. **Nikki Choudhary:** participated in the survey and collection of the samples. **Rubiya Banoo:** participated in the survey and collection of the samples. **Akansha Rai:** participated in the survey and collection of the samples. **Pooja Yadav:** participated in the survey and collection of the samples. **Martina Rani:** participated in the survey and collection of the samples. **Shyam Lal:** contributed in shaping manuscript into the final format. **Gareth J. Stewart:** participated in the experiment and analysis of NMVOC data using PTR-ToF-MS, GC × ×GC-FID and DC-GC-FID and contributed in the preparation of the manuscript. **Beth S. Nelson:** participated in the experiment and analysis of NMVOC data using PTR-ToF-MS, GC × ×GC-FID and DC-GC-FID and contributed in the preparation of the manuscript. **W. Joe F. Acton:** participated in the experiment and analysis of NMVOC data using PTR-ToF-MS, GC × ×GC-FID and DC-GC-FID and contributed in the preparation of the manuscript. **Adam R. Vaughan:** participated in the experiment and analysis of NMVOC data using PTR-ToF-MS, GC × ×GC-FID and DC-GC-FID and contributed in the preparation of the manuscript. **Jacqueline F. Hamilton:** participated in the experiment and analysis of NMVOC data using PTR-ToF-MS, GC × ×GC-FID and DC-GC-FID and contributed in the preparation of the manuscript. **James R. Hopkins:** participated in the experiment and analysis of NMVOC data using PTR-ToF-MS, GC × ×GC-FID and DC-GC-FID and contributed in the preparation of the manuscript. **C. Nicholas Hewitt:** participated in the experiment and analysis of NMVOC data using PTR-ToF-MS, GC × ×GC-FID and DC-GC-FID and contributed in the preparation of the manuscript. **Lokesh K. Sahu:** assisted in the analysis of PTR-MS data and contributed in the manuscript. **Nidhi Tripathi:** assisted in the analysis of PTR-MS data and contributed in the manuscript. **S.K. Sharma:** contributed in shaping manuscript into the final format. **T.K. Mandal:** provided overall guidance in planning, conducting the whole study and contributed in shaping the manuscript into the final format.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.aeaoa.2021.100127.

Ethics approval and consent to participate

Not applicable.

Consent for publication

Not applicable

Availability of data and materials

All data generated or analysed during this study are included in this published article and its supplementary information files.

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