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Emission estimates and inventories of non-methane volatile organic compounds from anthropogenic burning sources in India

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ABSTRACT

Comprehensive, spatially disaggregated emission inventories are required for many developing regions to evaluate the relative impacts of different sources and to develop mitigation strategies which can lead to effective emission controls. This study developed a 1 km^2 non-methane volatile organic compound (NMVOC) emission model for the combustion of fuel wood, cow dung cake, municipal solid waste (MSW), charcoal, coal and liquefied petroleum gas (LPG) in India from 1993 to 2016. Inputs were selected from a range of detailed fuel consumption surveys and recent emission factors measured during comprehensive studies of local burning sources. For the census year of 2011, we estimated around 13 (5–47) Tg of NMVOCs were emitted from biomass and MSW combustion in India. Around 54% of these emissions were from residential solid biofuel combustion, 23% from open burning of MSW, 23% from crop residue burning on fields and <1% from LPG for cooking. NMVOC emissions from residential combustion were shown to be highly sensitive to the amount of cow dung cake combusted and this acted as a key pollution source across the Indo-Gangetic Plain. The results of this study indicate that multiple mitigation strategies are required across several different categories of burning source to achieve effective NMVOC emission reduction.

1. Introduction

Biomass burning is the second largest global source of trace gases to the troposphere after biogenic emissions (Yokelson et al., 2008; Andreae, 2019). Major sources include wildfires, agricultural crop residue burning on fields and residential solid fuel combustion. Trace gases are released in varying amounts dependent on the combustion conditions and the material burned (Yokelson et al., 1996). Emission factors have been shown to vary significantly for different energy sources such as fuel wood, straw, grass, peat, and cow dung cake (Andreae, 2019). NMVOCs have the potential to significantly reduce local, regional and global air quality though the formation of tropospheric ozone (Pfister et al., 2008; Jaffe and Wigder, 2012) and secondary organic aerosol (SOA) (Alvarado et al., 2015; Kroll and Seinfeld, 2008).

Emissions from domestic biofuel combustion pose significant health risks as approximately 3 billion people cook with solid fuels globally

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(World Health Organization, 2018; World Bank, 2020). Emissions from burning have been linked to eye disease (Pokhrel et al., 2005), chronic bronchitis (Akhtar et al., 2007; Moran-Mendoza et al., 2008), chronic obstructive pulmonary disease (Dennis et al., 1996; Orozco-Levi et al., 2006; Rinne et al., 2006; Ramirez-Venegas et al., 2006; Liu et al., 2007; PerezPadilla et al., 1996), lung cancer (Liu et al., 1993; Ko et al., 1997), childhood pneumonia (Smith et al., 2011), acute lower respiratory infections (Bautista et al., 2009; Mishra, 2003) and low birth weight of children (Boy et al., 2002; Yucra et al., 2011). The detrimental impact of domestic biofuel combustion on indoor air pollution was estimated to result in 3.9 million premature deaths in 2010 (Smith et al., 2014), 2.8 (2.5–3.3) million premature deaths in 2015 (Kodros et al., 2018) and 3.8 million premature deaths in 2016 (World Health Organization, 2018).

Rapid growth has resulted in India being the second largest contributor to NMVOC emissions in Asia (Kurokawa et al., 2013; Kurokawa and Ohara, 2020). NMVOC emissions from India have been estimated in studies both focussed on Asia (Streets et al., 2003; Ohara et al., 2007; Zhang et al., 2009; Kurokawa et al., 2013; Crippa et al., 2019; Kurokawa and Ohara, 2020) or specifically on India (Varshney and Padhy, 1998; Pandey et al., 2014; Sharma et al., 2015). Lack of data and uncertainties in existing data complicate emission estimates and mean that considerable uncertainty exists over the size of NMVOC emissions from India, as shown in Table 1. Predicting emissions is complicated by a diverse range of sources such as older vehicle fleets, a high reliance on compressed natural gas (CNG), open crop burning on fields, MSW burning and solid biofuel combustion.

Traditional cook stoves represent a large pollution source in India due to their extensive use. Fig. 1 shows an estimation of residential fuel use in India from fuel wood, cow dung cake, LPG, coal, charcoal, biogas, crop residues, kerosene and electricity (see the Supplementary Information S1 for details of calculation). Fuel wood and cow dung cake usage have been relatively constant over the last 25 years, with approximately three quarters of a billion users (Pandey et al., 2014; World Health Organization, 2018; World Bank, 2020). It has been forecast that solid fuel combustion sources will remain an important energy source to India in coming decades. Projections by the International Energy Agency show that with current policies, the proportion of the Indian population using biomass for cooking will reduce to a third of the population in 2030 and represent a quarter of the population by 2040 (IEA, 2020).

Biofuels such as fuel wood and cow dung cake are cheaper than modern cooking fuels, such as LPG and electricity. Traditional methods are also important to many local recipes, with the meals cooked using them considered to be tastier (Mukhopadhyay et al., 2012). Cow dung cakes are commonly used in the north of India because they are sustainable, reduce the demand on local fuel wood resources and are widely available. Despite this, the impact cow dung cake combustion has on air quality is poorly understood. This is because consumption estimates of dried cow dung cakes in India have been shown to vary by around a factor of 3, in the range 35–128 Tg yr^{-1} for the years 2000–2001 (Habib et al., 2004). Emission estimates from cow dung cake combustion are also complicated by the varying moisture content of samples, which has a large influence on burn efficiency and in turn increases uncertainties in inventories.

Table 1
Estimates of NMVOC emissions from India, with the value in brackets showing the estimated contribution from burning sources.

Year	NMVOC/Tg yr^{-1}	Reference
1996	8.0 (6.6)	(Pandey et al., 2014; Sadavarte and Venkataraman, 2014)
1998	8.1 (4.7)	Varshney and Padhy (1998)
2000	8.0 (6.1)	(Pandey et al., 2014; Sadavarte and Venkataraman, 2014)
2000	10.8	Streets et al. (2003)
2003	9.7	Ohara et al. (2007)
2005	9.0 (6.5)	(Pandey et al., 2014; Sadavarte and Venkataraman, 2014)
2006	10.8	Zhang et al. (2009)
2008	16.0	Kurokawa et al. (2013)
2010	9.8 (6.9)	(Pandey et al., 2014; Sadavarte and Venkataraman, 2014)
2010	9.8 (6.5)	Sharma et al. (2015)
2010	11.5	Ohara et al. (2007)
2011	12.1 (6.0)	REAS 3.2 (Kurokawa and Ohara, 2020)
2015	12.0 (7.0)	(Pandey et al., 2014; Sadavarte and Venkataraman, 2014)
2015	13.5 (5.1)	EDGAR 5.0 (Crippa et al., 2019)

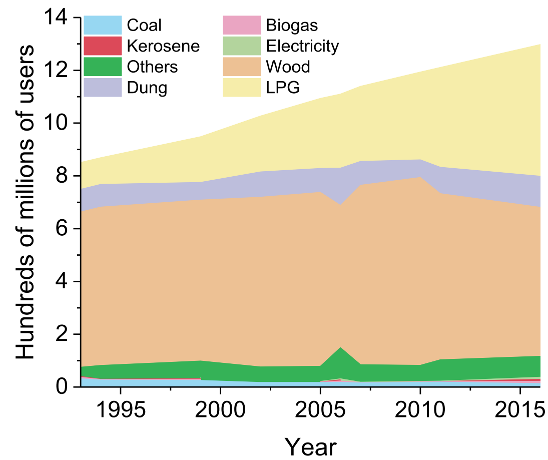

Fig. 1. Approximate fuel use in India by number of users. See the Supplementary Information S1 for details of calculation. The peak in dung and other fuels in 2006 underlines one of the difficulties in accurately establishing fuel usage from surveys scaled up for India.

The open burning of municipal solid waste (MSW) and agricultural crop residues on fields are also likely large emitters of NMVOCs. MSW is burnt for disposal, as a result of fugitive methane emissions in landfill sites and as a source of heating in cold seasons in low income areas (Nagpure et al., 2015). Several recent studies have indicated that MSW burning may result in emissions of around 1.7–1.8 Tg of NMVOCs annually across India (Wiedinmyer et al., 2014; Sharma et al., 2019) with the contribution of on field agricultural crop residue burning estimated at approximately 1.5 Tg from 2008 to 2009 (Jain et al., 2014).

Several recent studies have examined NMVOC emissions from sources relevant to India. Stockwell et al. (2016) examined VOC emissions from burning sources in Nepal, a neighbouring country, from a wide range of region-specific sources which included municipal solid waste, cooking fires and crop residues. 93 VOC species were quantified by collecting emissions into whole air sample canisters with analysis by gas chromatography coupled to a flame ionisation detector, an electron capture detector and a mass spectrometer. Fleming et al. (2018) quantified 76 VOCs emitted from the combustion of fuel wood and cow dung cake fires from Haryana, India. Whole air samples were collected and separated by gas chromatography followed by analysis by two electron capture detectors, two flame ionisation detectors and a mass spectrometer. One major limitation of these gas chromatography-based studies is that multiple separations with different column configurations and detectors are required to analyse different species of emission. They are also not well suited to measurements of intermediate-volatility and semi volatile organic compounds (I/SVOCs).

The more recently developed technique of proton-transfer-reaction time-of-flight mass spectrometry (PTR-ToF-MS) is well suited to measuring emissions from solid fuel combustion sources and has allowed around 90% of total measured NMVOC emissions in terms of mixing ratio from burning experiments to be speciated (Stockwell et al., 2015; Koss et al., 2018). Several recent studies have applied this state-of-the-art technique, alongside gas chromatography, two-dimensional gas chromatography and two-dimensional gas chromatography coupled to time-of-flight mass spectrometry to quantify 192 NMVOCs from burning sources collected from Delhi, India (Stewart et al., 2021a, 2021b). These highlighted large differences in NMVOC emissions between different sources, with emission factors for cow dung cake and municipal solid waste combustion (MSW) measured by Stewart et al. (2021a) ~ 300% and 400% larger, respectively, than for conventional fuel wood combustion. A further study showed that emissions from fuel wood, crop residue for domestic combustion, cow dung cake and MSW were ~ 20, 60, 130 and 220 times more toxic with respect to the types and quantity of polycyclic aromatic hydrocarbons (PAHs) released and 30, 90, 120 and 230 times more reactive with the OH radical than liquefied petroleum gas (LPG) (Stewart et al., 2021c). However, strategic improvement in Indian air quality with effective mitigation policies has been hindered by the lack of adequate, spatially disaggregated emission inventories created using these local source profiles (Garaga et al., 2018).

Uncertainty over data sources for Indian fuel consumption, the base year, emission factors and the spatial distribution of sources leads to large uncertainties in estimates of total emissions. In this study, we have developed comprehensive, spatially disaggregated emission inventories for NMVOCs released from burning sources in India. Inventories are produced for 10 different years from 1993 to 2016 and use recently published emission factors which far better reflect the full range of species released. This study then evaluates the relative contributions of individual sources to emissions to allow an assessment of the overall impact of emissions from burning sources to air quality in India. This is because recent studies have shown that NMVOC emission reduction is needed to accompany NO_x emission reduction to avoid increases in O_3 concentrations in cities like Delhi (Nelson et al., 2021; Stewart et al., 2021d).

2. Methods

2.1. Emission factors

The emission factors used in this study come from a variety of recently published sources. All emission factors applied in this study included measurement by PTR-ToF-MS, a technique well suited to species released in significant quantities from solid fuel combustion such as small oxygenated species, phenolics and furanics. These species are often missed by GC measurement alone. Preference has been given to emission factors from studies which: (1) have many measurements (n), (2) use samples collected from India or (3) use samples collected from similar countries. Fully speciated emission factors are available from the references given. For residential fuel combustion, the emission factors measured by Stewart et al. (2021a) were used and were developed from 76 combustion experiments of fuel wood, cow dung cake, LPG and MSW samples collected from around Delhi. This study was extremely detailed and measured online, gas-phase, speciated NMVOC emission factors for up to 192 chemical species using dual-channel gas chromatography with flame ionisation detection (DC-GC-FID, $n = 51$), two-dimensional gas chromatography (GC × GC-FID, $n = 74$), proton-transfer-reaction time-of-flight mass spectrometry (PTR-ToF-MS, $n = 75$) and solid-phase extraction two-dimensional gas chromatography with time-of-flight mass spectrometry (SPE-GC × GC-ToF-MS, $n = 28$). Comparison of these emission factors to those obtained in similar studies is provided in Stewart et al. (2021a). The emission factors used as part of this study are larger than those measured by Stockwell et al. (2016), Fleming et al. (2018) and several other studies which were based on gas chromatography techniques alone. The emission factors here measure many more NMVOC species, use techniques which target a range of species which more traditional GC analyses do not detect and make online measurements which minimise loss of intermediate-volatility and semi-volatile organic species, which may be lost through the collection of whole air samples, but have been shown to represent a large proportion of total emissions from biomass burning (Stockwell et al., 2015). Table 2 shows the mean emission factors applied in this study.

Emission factors for combustion of crop residues on fields were taken from measurements by Stockwell et al. (2015) made using PTR-ToF-MS of 115 NMVOCs (Stockwell et al., 2015) for wheat straw ($n = 6$), sugarcane ($n = 2$), rice straw ($n = 7$) and millet ($n = 2$). This study also included the mean crop residue emission factor for 19 food crops, for use when no current emission factor had been comprehensively measured using PTR-ToF-MS. The emission factor applied (38.8 g kg^{-1}, see the Supplementary Information S2 for details of calculation) was evaluated against that for crop residues used for domestic combustion in Delhi (37.9 g kg^{-1}). Whilst the values measured by Stockwell et al. (2015) and Stewart et al. (2021a) were comparable, the value from Stockwell et al. (2015) was used as the crop types were more reflective of the crop residues burnt on fields after harvest, compared to those burnt to meet residential energy requirements. The mean emission factor for crop residue combustion on fields was used for specific crop types with smaller levels of cultivation.

Emissions from coal burning were estimated using a mean emission factor from the combustion of bituminous coal from China ($n = 14$), a neighbouring Asian country, made using PTR-ToF-MS. Whilst the chemical composition of the coal may be more important than the development status of the country, there was overall a low level of reported residential coal use and this estimate was included for completeness. A total of 89 NMVOCs were identified, which represented 90–96% of the total mass spectra (Cai et al., 2019).

Indian specific PAH emission factors were recently measured in gas- and particle-phases using PTR-ToF-MS and GC × GC-ToF-MS (Stewart et al., 2021b). This dataset provided PAH emission factors collected from combustion of fuel wood ($n = 16$), cow dung cake ($n = 3$), crop residue from domestic combustion ($n = 3$), MSW ($n = 3$), LPG ($n = 1$) and charcoal ($n = 1$) samples.

2.2. Spatial activity data

High resolution, gridded population data for India (WorldPop, 2017) was used at a resolution of 1 km^2. Officially, urban populations in India are defined as having (Chandramouli, 2011):

- population density > 400 people km^{-2}
- 75% of men employed in non-agricultural industries
- population of town > 5000 people.

Rural populations in India cannot be identified simply by having a population density of < 400 people km^{-2}, as some states such as Uttar Pradesh have an average population density of around 800 people km^{-2}. Rural grid squares were therefore identified by calculating the population density threshold in each state in which the sum of the 1 km^2 grid squares below this threshold correctly reproduced the rural populations in these states from the 2001 and 2011 censuses (Government of India, 2014). Supplementary Information S3 shows that this resulted in good reproduction of rural and urban populations. A small uncertainty existed over the exact population of India and we used population statics indicated by the 2011 census.

NMVOC and PAH emissions from domestic solid fuel combustion were plotted against this high-resolution population data in the R statistical programming language at 1 km^2 for 2001 and 2011, with the population datasets scaled to the percentage changes in Indian population indicated by the World Bank for additional years of interest.

2.3. Fuel wood, LPG, charcoal and coal consumption

Preference was given to large fuel usage surveys which included tens to hundreds of thousands of respondents. The Household Consumption of Goods and Services in India survey by the National Sample Survey Office (NSSO, 2007a; 2012a, 2014) gave state-wise kg $capita^{-1}$ fuel

Table 2
Mean NMVOC and PAH emission factors (g kg^{-1}) from combustion of different fuels, with σ = standard deviation of burns of same fuel type and n = number of measurements.

NMVOC emission factors/g kg^{-1}	Wood	Dung	MSW	LPG	Charcoal	Rice	Wheat	Sugarcane	Millet	Crop	Coal
VOC	18.7	62.0	87.3	5.7	5.4	23.8	15.9	53.6	5.4	38.8	3.7
Σ	17.9	18.4	31.4	5.5	3.9	20.0	8.1	23.7	1.6	19.4	0.9
N	51	8	3	3	2	7	6	2	2	19	14
Ref	[a]	[a]	[a]	[a]	[a]	[b]	[b]	[b]	[b]	[b]	[d]

PAH emission factor/g kg^{-1}	**Wood**	**Dung**	**MSW**	**Crop**	**LPG**	**Charcoal**
PAH	0.25	0.61	1.02	0.75	0.06	0.15
σ	0.21	0.11	0.34	0.52	–	–
n	16	3	3	3	1	1
Ref	[c]	[c]	[c]	[c e]	[c]	[c]

References.
[a] Stewart et al. (2021a).

[b] Stockwell et al. (2015).

[c] Stewart et al. (2021b).

[d] Cai et al. (2019) and

[e] Crop types used for residential solid fuel combustion.

wood, LPG, charcoal and coal burning statistics for rural and urban environments and was used for the years 2004–2005, 2009–2010 and 2011–2012. NMVOC emissions for these years were calculated though equation (E1):

$$NMVOC\ emission_{1km2,\ fuel} = EF_{fuel} \times fuel\ consumption \times population_{1km2} \times \left(\frac{365}{30}\right) \quad (1)$$

where NMVOC $emission_{1km2,fuel}$ = total NMVOC emission from respective fuel combustion per 1 km^2 grid (kg yr^{-1}), EF_{fuel} = mean emission factor for fuel used, fuel consumption = per capita fuel consumption (kg 30 $days^{-1}$) converted from per 30 days to per year by multiplying by (365/30) and $population_{1km2}$ = population in 1 km^2 grid. This calculation was performed separately for rural and urban grid cells to allow accurate incorporation of rural and urban per capita fuel consumption data.

Data were collected from additional large previously conducted surveys. These surveys collected data in terms of the number of households using specific fuels per 1000 households in different Indian states in rural and urban environments. The Fifth Quinquennial Survey on Consumer Expenditure provided data for 1993–1994 (NSSO, 1997), the Energy Sources of Indian Households for Cooking and Lighting provided data for years 2004–2005, 2009–2010 and 2010–2011 (NSSO, 2007b; 2012b, 2015) and the Household Consumer Expenditure and Employment-Unemployment Situation in India for 2002 and 2006–2007 (NSSO, 2003, 2008). The National Family Health Survey presented India-wide fuel use as a percentage of the population. To reflect spatial variation in fuel use, the raw data from these surveys were accessed (from the DHS Programme, U.S. Agency for International Development), extracted through the SPSS statistics software package and processed in the R programming language. This increased fuel usage data availability as the number of households per 1000 households using specific fuels in Indian states and covered the years 1992–1993, 1998–1999, 2005–2006 and 2015–2016 (International Institute for Population Sciences, 1995, 2000, 2007, 2017). These were extensive datasets with 1992–1993, 1998–1999 and 2005–2006 surveying just under 100,000 households and 2015–2016 around 600,000 households. The fuel use data collected as part of this study are provided in the Supplementary Information S4.

To allow the incorporation of data from years which were based on the number of households using a particular fuel per 1000 households (1993, 1994, 1999, 2002, 2006, 2007 and 2016), a scaling factor was developed. The scaling factor was based on the ratio of fuel use in the state from years where per capita data was available. It was possible to link the Household Consumption of Goods and Services in India and the Energy Sources of Indian Households for Cooking and Lighting surveys for the years 2005, 2010 and 2011. This was done using years where the number of households per 1000 households and kg $capita^{-1}$ fuel usage statistics were available, as it was possible to calculate the amount of fuel a primary user would use. The fuel use of a primary user here was defined as the amount of fuel a person would burn who was recorded to use a specific fuel type. For example, if the per capita consumption in the Household Consumption of Goods and Services survey in India for fuel wood was 10 kg per capita per 30 days, and the Energy Sources of Indian Households for Cooking and Lighting survey showed 250 households per 1000 households used fuel wood, then the fuel use was estimated to be 40 kg per primary user per 30 days. This was achieved by multiplying the per capita usage for a particular fuel type by the inverse of the ratio of fuel usage in that state in rural or urban environments, and is given in E2:

$$Fuel\ use\ primary\ user = Fuel\ use_{capita} \times \frac{1000}{NHH} \quad (2)$$

where Fuel use primary user = amount of a specific fuel type that a person who just burns that fuel type uses (kg 30 $days^{-1}$), Fuel use_{capita} = per capita fuel use per 30 days (kg $capita^{-1}$ 30 $days^{-1}$) and NHH = number of households per 1000 households using a particular fuel type. This was calculated for urban and rural scenarios in Indian states in years where it was possible (2005, 2010, 2011).

The amount of fuel a primary user would use was then used to estimate the amount of fuel consumed per capita in years where only usage per 1000 household statistics were available (1993, 1994, 1999, 2002, 2006, 2007 and 2016) by rearranging E2 to give E3.

$$\frac{Fuel\ use\ primary\ user}{1000} \times NHH = Fuel\ use_{capita} \quad (3)$$

The amount of fuel per primary user was taken from the closest survey where data was available. In some earlier surveys, data were not collected for smaller states and these were either estimated by averages of neighbouring states, or from the nearest available usage values for other years for these states. NMVOC emissions for the years 1993, 1994, 1999, 2002, 2006, 2007 and 2016 were then determined using E1 with the calculated per capita fuel consumption values from E3. This is explained in more detail in the Supplementary Information S5. Biomass fuels are sometimes not used exclusively. This is accounted for as per capita data sampled a range of fuels. For years where data was only available for the number of households per 1000 household, conversions were based on users using a principal fuel as their energy source.

2.4. Cow dung cake consumption

Cow dung cake consumption was only reported as number of households per 1000 in these surveys and the amount of cow dung cake burnt per primary user was determined based on the energy density compared to fuel wood. This was done using calorimetry data which showed that cow dung cake was 1.3–1.9 times less efficient than fuel wood (EPA, 2000; Gadi et al., 2012). For this reason, the amount of fuel wood per primary user for fuel wood in a state has been multiplied by 1.6 to give the equivalent amount of cow dung cake a user would need to burn for their cooking needs. Upper and lower estimates for cow dung cake consumption were based on the range 1.3–1.9. This was then converted to fuel use per capita in kg per user per 30 days by rearranging E2. This has been evaluated to validate this approach, which estimated Indian cow dung cake consumption to be in the range 25.7–79.7 Tg yr^{-1} from 1993 to 2016. This was generally towards the lower end of consumption values previously reported of 35–128 Tg yr^{-1} (Habib et al., 2004). For this reason, emission inventory estimates were also compared to those produced using cow dung cake consumption based on the TERI Energy Data Directory and Yearbook (TEDDY) 2012/2013 data and a study from the Petroleum Planning & Analysis Cell (PPAC) from 2016 with population indicated at the 2011 level (TEDDY, 2012; PPAC, 2016).

2.5. Activity data for MSW combustion

The input for MSW was one of the most difficult inputs to calculate due to lack of reliable data and was consequently one of the most uncertain. An estimation of NMVOCs released from open MSW burning was attempted as there was little information available for India, where MSW burning is potentially a very large pollution source. The amount of MSW burnt was estimated using an established approach (IPCC, 2006; Wiedinmyer et al., 2014) with revised inputs for India based on per capita MSW generation from over 300 Indian cities (Annepu et al., 2012), state wise MSW collection figures (CPCB, 2013) as well as estimates of the amount of urban (NEERI, 2010) and rural MSW burnt (World Bank, 2012). This estimate does not include incineration for electrical power generation.

Wiedinmyer et al. (2014) assessed worldwide emissions from MSW burning based on IPCC guidelines (IPCC, 2006). The approach used here was similar, with modifications to the input data which made them more specific to India. The approach split the amount of MSW burnt into the MSW burnt by rural and urban populations in the country. For rural populations this was given by:

$$W_{Bres} = MSW_{pr} \times P_{rural} \times B_{frac,res} \tag{4}$$

where W_{Bres} = MSW burnt residentially, MSW_{pr} = per capita rural MSW generation, P_{rural} = population of rural grid cell and $B_{frac,res}$ = the fraction of MSW burnt residentially.

Per capita rural MSW generation was set at the lower limit indicated by the World Bank for South Asia of 0.12 kg capita^{-1} day^{-1} and evaluated in the range 0.08 kg capita^{-1} day^{-1} (Parmar and Pamnani, 2018) to 0.12 kg capita^{-1} day^{-1} (World Bank, 2012). The fraction of MSW burnt rurally was set to 0.6 which was the IPCC estimate (IPCC, 2006) and was further supported by a recent study which showed that only around 40% of rural MSW was collected in South Asia (Kaza et al., 2018).

The fraction of MSW burnt for an urban population was estimated by the sum of two calculations. The first was for street MSW burning:

$$W_{Bres} = MSW_{pu} \times P_{urban} \times f_{uncollected} \times B_{frac} \tag{5}$$

where MSW_{pu} = per capita urban MSW generation, P_{urban} = population of urban grid cell and $f_{uncollected}$ = fraction of MSW which was not collected. The weighted per capita urban MSW generation was calculated by averaging per capita MSW generation statistics from 366 Indian cities by state (Annepu et al., 2012), with calculated values given in the Supplementary Information S6. The fraction of MSW which was uncollected was calculated from the Central Pollution Control Board (CPCB), as the difference in the amount of MSW generated and collected (CPCB, 2013). Urban per capita MSW generation was scaled to its estimated change for different years of interest (see the Supplementary Information S6).

The second calculation was for the MSW burnt on landfill sites:

$$W_{Bdump} = MSW_{pu} \times P_{urban} \times f_{collected} \times B_{frac,dump} \tag{6}$$

where W_{Bdump} = landfill MSW burnt and $f_{collected}$ = fraction of MSW collected. The fraction of MSW collected came from CPCB statistics, but was reduced by 17–50% due to the informal recycling sector, based on very limited data from studies focussed on MSW recovery by the informal sector which showed 17% recovery in Delhi (Talyan et al., 2008), 20% recovery at a landfill site in Pune (Annepu et al., 2012), 4% in Pondicherry (Rajamanikam et al., 2014) and up to 40–50% in Mohali (Nandy et al., 2015). This was due to the large contribution of the informal recycling sector to recycling in India, where waste was collected by waste merchants, garbage collectors and waste pickers from highways, waste depots and landfill sites. This was an important consideration in India as studies have shown recovery of between 8.5 and 80 kg of material per picker per day and large cities such as Delhi having 80,000–100,000 pickers (Nandy et al., 2015). $B_{frac,\ dump}$ was given by NEERI who estimated that 10% of landfill MSW in Mumbai was burnt (NEERI, 2010). This was reinforced by a further study which examined the amount of waste burnt based on satellite studies of a landfill site in India which showed that approximately 10% of the waste that entered the site each day ended up being burnt (Sharma et al., 2019). $B_{frac,\ dump}$ was notably lower here (0.1) than in Wiedinmyer et al. (2014) (0.6) which was based on the 2006 IPCC Guidelines for National GHG Inventories. The estimate used in this study represented a conservative estimate of NMVOC emissions from landfill fires. Due to lack of reliable data in establishing $B_{frac,\ dump}$, and the associated uncertainty, the sensitivity of urban landfill burning emissions over the range 0.1–0.6 was evaluated as part of the uncertainty range given in this study. This provided the upper limit to the uncertainty range of the potential amount of landfill waste burnt. This depicts scenarios before the new MSW management rules in 2016.

2.6. Input to agricultural crop residue burning on fields

NMVOC emissions from crop residue burning on fields in India were estimated to evaluate the relative importance of different burning sources using the most up-to-date input data currently available (see Table 2). A calculation was carried out for 2011, as NMVOC emissions from crop-residue burning on fields showed little year-on-year variation from 1995 to 2009 (Jain et al., 2014). The residue generated from the cultivation of four main categories of crops was estimated. The amount of crop types produced in each state (Ministry of Agriculture, 2012) was collated for cereals (rice, wheat, coarse cereals, maize, jowar, bajra), oilseeds (groundnut, rapeseed, mustard, sunflower and 9 oilseeds), fibres (cotton, jute and mesta) and sugarcane. The amount burnt was calculated using India specific estimates of the residue to crop ratio, dry matter fraction and fraction burnt (Jain et al., 2014). Emissions were estimated using factors from recent studies of crop residues routinely burnt on fields using PTR-ToF-MS (Stockwell et al., 2015). When the exact residue was measured (e.g., rice straw, wheat straw, sugarcane and millet) the correct emission factor was used. For cases where the exact residue was not measured, the mean reported crop residue emission factor was used (see the Supplementary Information S7 for inputs for the crop residue estimate). The spatial distribution of croplands was then either indicated using agricultural land identified by the high-resolution 500 m NASA MODIS land use product reduced to 1 km^2 resolution or through croplands identified at 10 km^2 through evaluation of the distribution of agricultural lands (Ramankutty et al., 2008).

The total amount of crop residue burnt in a state was calculated by:

$$Crop_{emission} = \frac{\sum_0^n CWG \times RTCR \times DMF \times FB \times EF_{crop,i}}{area\ cultivated} \quad (7)$$

where $Crop_{emission}$ = NMVOC emitted in a state from crop residue burning on fields (kg km^{-2}) (Ministry of Agriculture, 2012), CWG = mass of crop produced in the state, RTCR = residue to crop ratio (Jain et al., 2014), DMF = dry matter fraction (Jain et al., 2014), FB = fraction of crop residue burnt (Jain et al., 2014), $EF_{crop,i}$ = emission factor for crop species i (g kg^{-1}), area cultivated = total agricultural area identified in a state from either MODIS (1 km^2) or Ramankutty et al. (2008) (10 km^2) and n = number of different crops produced in the state. An overview of all emission model inputs is given in the Supplementary Information S8.

3. Results

3.1. Emission model

Fig. 2 shows the calculated NMVOC emissions from the burning of fuel wood, cow dung cake, MSW, LPG and charcoal alongside that for crop residue burnt on fields for the year 2011. This year was chosen as the focus for this study, as this was a national census year and had some of the best available fuel consumption data. Additional inventories of all years studied are available to download from the Centre for Environmental Data Analysis (https://doi.org/10.5285/fdb8960260a64c5faf652f8f47c4df81). In general, NMVOC emissions on an area basis were lowest in the very north and north-east region of India around the Himalayas and in the north-west region of the Thar Desert, both areas of low population density. Detailed NMVOC emission estimates by source and state for 2011 are given in the Supplementary Information S9.

3.2. Fuel wood

National NMVOC emissions from fuel wood burning were estimated as 4.3 (1.0–22.3) Tg and were the largest due to the high number of users (600 million) across India (see Fig. 2A). Emissions were significant in many cities which appeared as red dots in Fig. 2A, as well as across the Indo-Gangetic Plain. The greatest emissions were in West Bengal and Kerala, due to high population densities (1028 and 860 people km^{-2} respectively) and high per capita fuel usage. For example, rural and urban fuel wood consumptions in Kerala were reported to be ~32 and 21 kg capita^{-1} (30 days)$^{-1}$, respectively.

3.3. Cow dung cake

Cow dung cake burning represented a significant NMVOC source, with emissions of 2.8 (1.3–4.4) Tg localised to the Indo-Gangetic Plain (see Fig. 2B). This considers cow dung cake consumption based on calorimetry data which likely represents a lower limit emission scenario and is discussed in more detail in section 4.2. Cow dung cakes are often considered a co-product of cattle production (Gupta et al., 2016) and are used as a sustainable fuel in several regions, partly to alleviate demand on local fuel wood supplies. Cow dung cakes remained an important fuel source in northern states, with high per capita usage along the Indo-Gangetic Plain in 2011.33.4% of rural households were reported to use cow dung cakes as a primary fuel source in Uttar Pradesh, 30.3% in Punjab, 24.4% in Haryana, 20.8% in Bihar and 10.6% in Madhya Pradesh (NSSO, 2015).

3.4. Municipal solid waste

Fig. 2C shows NMVOC emissions from the open burning of MSW, which were high from both rural and urban areas. In total, MSW burning in India was estimated to release 3.0 (1.6–6.9) Tg of NMVOCs in 2011. Emissions from the combustion of MSW were significant, particularly to urban areas due to these being regions of high population density.

3.5. Charcoal/coal

NMVOC emissions from charcoal (0.9, 0.4–1.3 Gg) and coal (4.8, 1.7–5.9 Gg) remained low due to low usage and a low emission factor. Fig. 2D shows emissions from charcoal. Coal burning was only noticeable to West Bengal (see the Supplementary Information S10).

3.6. LPG

NMVOC emissions from LPG were low at 71 (24–123) Gg due to a low emission factor, high energy density and low per capita fuel usage (see Fig. 2E). Emissions were principally in urban areas which had higher per capita LPG usage. This source mainly released propane and butanes, which were shown to be significantly less toxic in terms of PAHs and less reactive with the OH radical than the other solid fuel sources studied here (Stewart et al., 2021c).

3.7. Agricultural crop residue on fields

Crop residue burning on fields was estimated to emit 3.0 (1.4–4.5) Tg of NMVOCs in 2011. Fig. 2F shows emissions from crop residue burning on fields visualised using the distribution of geographic lands (Ramankutty et al., 2008). Emissions from agricultural crop residue burning on fields were significant in the north of India and were driven by cereal production in Punjab and Haryana, as well as sugarcane and cereal production in Uttar Pradesh and Bihar. The most significant emissions from Madhya Pradesh and Rajasthan were from the burning of oilseeds crops. Emissions from Maharashtra, Karnataka, Andhra Pradesh and Tamil Nadu were principally from the burning of sugarcane residue.

3.8. PAHs

To better understand the scale and sources of PAH emissions in India, the emissions model was used to evaluate PAH emissions from burning sources in 2011. This is because PAH concentrations have been reported to be high in cities like Delhi (Elzein et al., 2020). The spatial distribution of emissions by source type was like that displayed in Fig. 2 for NMVOCs. Detailed PAH emission estimates by source and state are given in the Supplementary Information S12.

Fig. 3 shows PAH emissions from the combustion of fuel wood, cow dung cake, MSW, charcoal and LPG in India in 2011. Total annual gas and particle phase PAH emissions were estimated to be 121 (52–387) Gg, from the burning of fuel wood (57 Gg, 12–209 Gg), cow dung cake (27 Gg, 18–98 Gg), LPG (0.7 Gg), charcoal (0.03 Gg) and MSW (36 Gg, 21–79 Gg). This result was similar to a previous estimate of PAH emissions from India in 2004 of 90 Gg (Zhang and Tao, 2009), with ~80 Gg from biofuel burning and slightly larger than a different study for 2007 which estimated emissions of 67 Gg, with 59 Gg from residential combustion (Shen et al., 2013). However, the inefficient combustion of MSW represented a considerable additional PAH source in India, which was likely to have significant impacts on human health. Further comparison of PAH emissions estimated in this study with previous inventories is

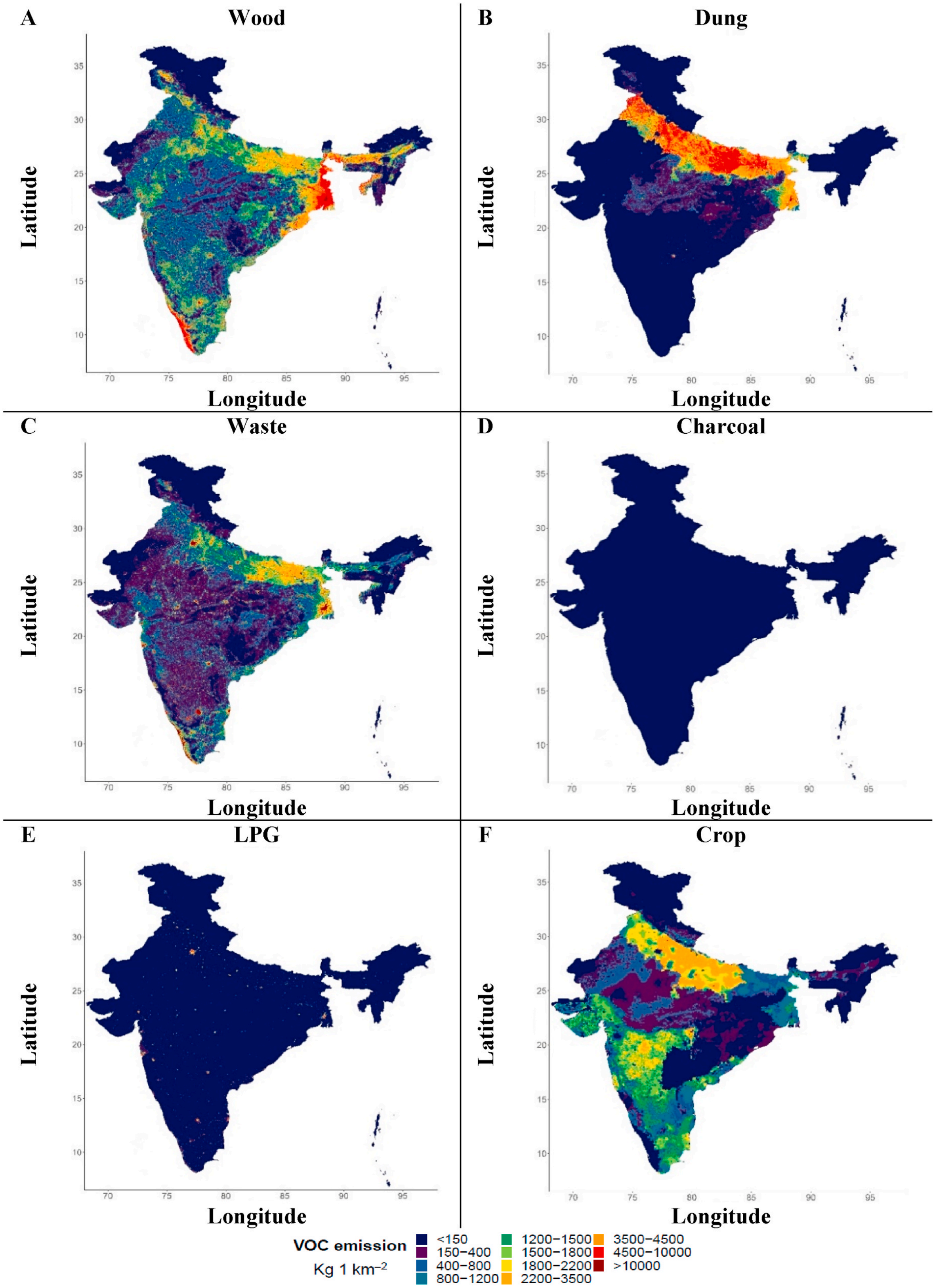

Fig. 2. Spatial distribution and emission of NMVOCs in 2011 from various burning sources in India. Emission maps for charcoal and LPG are provided with a different scale in the Supplementary Information S11. The inventories produced in this study are available to download at the Centre for Environmental Data Analysis (https://doi.org/10.5285/fdb8960260a64c5faf652f8f47c4df81). The declination of international borders on this map are proximate and must not be considered authoritative.

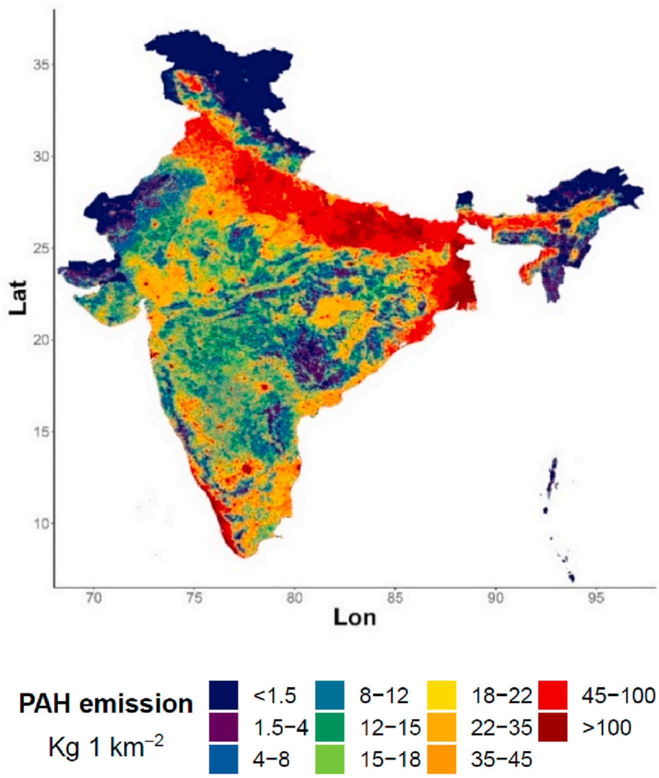

Fig. 3. PAH emissions in India from fuel wood, cow dung cake, MSW, charcoal and LPG burning in 2011. The declination of international borders on this map are proximate and must not be considered authoritative.

given in the Supplementary information S12.

4. Discussion of uncertainties

The generalisation of the laboratory combustion experiments in this study to the burning practices of a country with over 1 billion residents was likely to introduce significant uncertainties in the NMVOC emission estimates. Table 3 shows the fuel consumption values used in this study, the estimated NMVOC emissions and their uncertainties. Table 3 also compares fuel usage values from the limited available literature and previous NMVOC emission estimates from burning sources. Some general uncertainties existed due to the approach used here, as well as uncertainties which were specific to individual combustion sources. This significantly increased the uncertainties in emission estimates of specific combustion sources.

Uncertainties were likely to exist in the fuel consumption data utilised in this study, but these were not reported alongside official data and it was therefore not possible to account for this in the emission model. Furthermore, fuel consumption data was reported at a state-wide level, a much lower resolution than used in this emission model. As a result, sharp distinctions were seen between neighbouring states which had very different reported levels of usage of a particular fuel type. This effect was particularly pronounced for emission estimates from cow dung cake and on-field crop residue combustion. The real distribution of emissions was likely to show a more gradual transition across state boundaries.

The representativeness of this initial laboratory data to real-world conditions potentially lead to large uncertainties in these emission estimates. The modified combustion efficiency was not measured by Stewart et al. (2021a), despite the likely large impact on NMVOC emission. A recent study suggested that emission factors from burning could vary by almost a factor of 2 if fuel was combusted in *chulha* or *angithi* stoves (Fleming et al., 2018). Little information was available about the spatial distribution of different cook stoves across India. Future fuel use statistics should include this, and studies should examine the impact on NMVOC emissions.

The emission factors measured by Stewart et al. (2021a) included speciation that on average represented 94% of the total measured NMVOC emissions. The total measured emission factor reflected the sum of gas-phase organic emissions detected using multiple gas-chromatography instruments and the PTR-ToF-MS. This also included the unspeciated fraction measured on the PTR-ToF-MS. It did not include organic emissions which were not measured by these techniques. For PAH emission estimates, only 21 species were measured. This highlights a more general uncertainty of bottom-up emission estimates as they may underestimate emissions as not all released species may be detected using the measurement techniques deployed. This also complicates comparisons between estimates from different emission inventories as they may not all include the same level of detail.

Varying climates in different regions of India, with different biomass varieties and moisture contents, also increased uncertainties in emission estimates at a countrywide level. This was because small variations, such as seasonal changes to humidity, may have large impacts on burning efficiency and in turn NMVOC emission. Despite this, the methods used in Stewart et al. (2021a) were designed to replicate local practices in Delhi for sample collection, storage and combustion. Furthermore, municipal solid waste samples were collected from landfill sites, stored in sealed bags and combusted within 24 h. These approaches were designed to simulate real-world combustion conditions to ensure that the emission factors were reflective of local residential fuel use.

4.1. Wood

The NMVOC emission factor used for fuel wood came from a large dataset based on 51 measurements. The large number of measurements should significantly increase the representativeness of the mean emission factor used for fuel wood emission estimates in India during this study. Despite this, the emission factors measured from fuel woods were highly variable, by over a factor of 20 from around 4–97 g kg^{-1}, even under repeatable laboratory conditions. The uncertainty and repeatability of emission factor measurements was discussed in detail in Stewart et al. (2021a). The species of fuel wood and the composition of the sample burnt will vary considerably across India and will include species not measured here from different climatic conditions. This significantly increased the uncertainty in the NMVOC emission estimate, which was calculated for 2011 to be in the range 1.0–22.3 Tg. Further discussion of the uncertainties in emission estimates of fuel wood are given in the Supplementary Information S13.

4.2. Cow dung cake

The uncertainty in NMVOC emissions from cow dung cake combustion included uncertainty in the calorific conversion used to estimate fuel consumption, uncertainty in the emission factor and different reported levels of fuel usage. The uncertainty in the calorific conversion increased the uncertainty range by around 20%. This was reflected in the range of estimated cow dung cake consumption in India, which was 36.3–53.4 Tg in 2011.

Eight measurements were made of NMVOC emissions from cow dung cake combustion, with emission factors varying over a smaller range than for fuel wood from approximately 35-83 g kg^{-1}. The combined uncertainties in the calorific conversion and emission factor resulted in an uncertainty range of NMVOC emission estimates from 1.3 to 4.4 Tg in 2011, which was notably smaller than for fuel wood combustion.

One of the largest uncertainties in the NMVOC emission estimate from cow dung cake combustion was the different levels of fuel consumption reported by different surveys. Different studies report varying levels of cow dung cake usage in India between 5 and 15% of the population (EPA, 2000; International Institute for Population Sciences,

Table 3
Comparison of fuel consumption and NMVOC estimates in this study with literature.

	2011 fuel use this study/Tg	2011 NMVOC estimate this study/Tg	Literature use/Tg	NMVOC estimate literature/Tg	Year	Reference
Fuel wood	230	4.3 (1.0–22.3)	220	–	1985	Yevich and Logan (2003)
			271	–	1990	Streets and Waldhoff (1998)
			169	–	1990	Smith et al. (2000)
			302	–	1996	Reddy and Venkataraman (2002)
			265	–	1996	Bond et al. (2004)
			281 (192–409)	–	2000	Habib et al. (2004)
			316	–	2000	Streets et al. (2003)
			154 [b]	1.1 (0.6–1.7)	2005	Venkataraman et al. (2010)
			256	–	2007	TEDDY 2007 (Singh et al., 2013)
Cow dung cake [a]	45 (36.3–53.4)	2.8 (1.3–4.4)	93	–	1985	Yevich and Logan (2003)
Cow dung cake [b]	106	6.6 (3.7–8.8)	124	–	1990	Streets and Waldhoff (1998)
Cow dung cake [c]	161	10.0 (5.7–13.4)				
			54	–	1990	Smith et al. (2000)
			121	–	1996	Reddy and Venkataraman (2002)
			128	–	1996	Bond et al. (2004)
			62 (35–128)	–	2000	Habib et al. (2004)
			105	–	2000	Streets et al. (2003)
			–	1.8	2005	Venkataraman et al. (2010)
			106	–	2007	TEDDY 2007 (Singh et al., 2013)
MSW	35 (28–56)	3.0 (1.6–6.9)	81.4	1.8	2010	Wiedinmyer et al. (2014)
				0.1	2011	EDGAR 5.0
				0.01	2011	REAS 3.2
			68 (45–105)	1.7 (1.4–2.1)	2015	Sharma et al. (2019)
Agricultural crop residue on fields	83.8	3.0 (1.4–4.5)	107.3	1.5	2008	Jain et al. (2014)
			93	1.7 (0.6–4.0)	2010	Pandey et al. (2014)
				0.7	2010	Sharma et al. (2015)
				1.8 (0.6–4.1)	2015	Pandey et al. (2014)
				0.3	1997–2009	Pandey and Sahu (2014)
				0.6	– 2011	(Ministry of Agriculture, 2014) EDGAR 5.0
LPG	12.5	71 (24–123) × 10^{-3}	–	0.2	2005	Venkataraman et al. (2010)
			–	0.2 (0.1–0.4) [d]	2010	Pandey et al. (2014)
			–	0.3 (0.2–0.5) [e]	2015	Pandey et al. (2014)
Coal	1.3	4.8 (1.7–5.9) × 10^{-3}				
Charcoal	0.2	0.9 (0.4–1.3) × 10^{-3}				
Solid fuel total	276.5 (267.8–284.6)	7.1 (2.3–26.7)	450	4.9 (1.6–11.6)	2010	Pandey et al. (2014)
				4.9 (1.6–11.6)	2015	Pandey et al. (2014)
				5.9	2010	Sharma et al. (2015)
				4.2	2011	EDGAR 5.0
				5.9	2011	REAS 3.2

[a] Fuel use based on calorimetry data.

[b] Fuel use based on TEDDY, 2012/2013 data.

[c] Fuel use based on PPAC 2016 for 2011.

[d] Also includes estimate of kerosene use.

[e] Also includes charcoal use.

2007; NSSO, 2012b). This study estimated cow dung cake fuel consumption from 1993 to 2016 using calorimetry data to be in the range 25.7–79.7 Tg. This was smaller than many previous estimates of Indian dung consumption (see Table 3). The cow dung cake fuel usage inputs used in this study which determined fuel use based on calorimetric data were generally closer to 5–10% of the population and thus represented a more conservative case study for NMVOC emissions from cow dung cake combustion across India. This study may therefore underestimate the potential impact of cow dung cake combustion in India and emphasised the need for better official reporting of cow dung cake fuel usage. Emission estimates using fuel consumption taken from TEDDY, 2012/2013 data indicated that emissions from cow dung cake consumption could be as high as 6.6 Tg in 2011, whilst those from the PPAC showed that it could be even higher at around 10 Tg in 2011 (see the Supplementary Information S14 for inventories and further discussion). The estimated emissions from cow dung cake combustion should be refined in future studies through collection of accurate per capita cow dung cake consumption data.

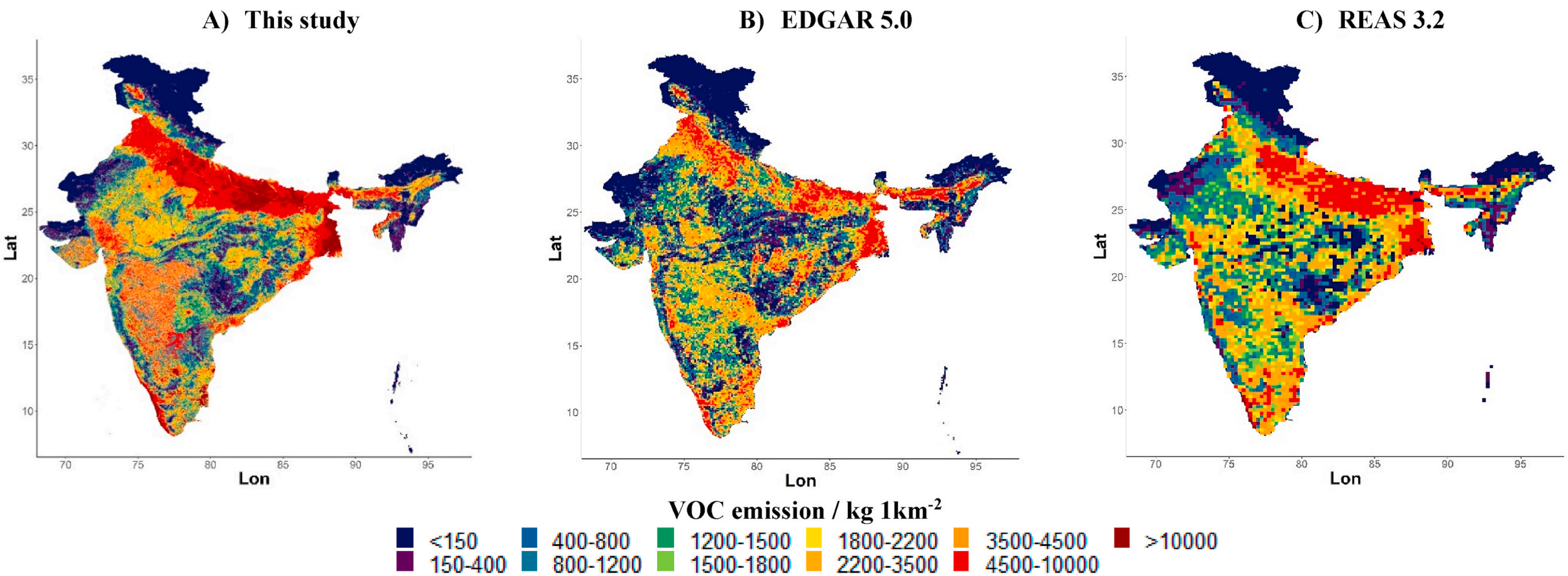

Fig. 4. Comparison of NMVOC emissions from solid fuel combustion sources from 2011 in A = this study, B = EDGAR 5.0 and C = REAS 3.2, with data taken from Crippa et al. (2019) and Kurokawa and Ohara (2020). Plots for EDGAR 3.2 and REAS 3.2 by individual source sector are given in the Supplementary Information S16. The declination of international borders on this map are proximate and must not be considered authoritative.

4.3. Municipal solid waste

The NMVOC emission estimate from MSW burning was one of the most uncertain, with large and potentially unquantifiable uncertainties in parts of the calculation. These included the low number of emission factor measurements, the high emission factor applied, uncertainty in the total mass of MSW generated in India, uncertainty in the amount of MSW recycled and uncertainty in the amount of MSW burnt in rural and urban environments. This emission estimate was presented as a discussion point, which should be treated with caution and could clearly be refined and improved as newer and better data becomes available. The uncertainties associated with MSW burning are discussed in considerable detail in the Supplementary Information S15.

4.4. Agricultural crop residue on field

Uncertainty in the estimate of NMVOC emissions from crop residue burning on fields was related to the timing as well as spatial distribution of emissions, uncertainties in emission factors and the measurements not being of crop residue samples collected from fields in India.

The spatial distribution of emissions from crop residue burning on fields was like Jain et al. (2014) with emissions from cereals impacting the northern states, oilseeds to Rajasthan and Madhya Pradesh, fibre to Maharashtra, Gujarat and Andhra Pradesh and sugarcane to Uttar Pradesh, Karnataka and Tamil Nadu. Despite this, uncertainty existed in the timing and spatial distribution of emissions. Emissions from crop residue burning on fields will show large seasonality, which was not accounted for here and could potentially be inferred in future studies using satellite data (e.g., NASA VIIRS fire counts) to provide information on the timing of data. Emissions will be predominantly during the pre-monsoon season for rabi crops (Apr–May) and during the post-monsoon season for kharif crops (Oct–Nov) (Gopal, 2014). Agricultural land was identified using both MODIS land use data and through previously published data which evaluated the distribution of agricultural lands (Ramankutty et al., 2008). A better understanding of the true impact of emissions from crop residue burning on fields would require data about the relative distribution of fires on agricultural lands.

Jain et al. (2014) used the emission factors from a review (Andreae and Merlet, 2001). This study used recently measured emission factors using PTR-ToF-MS, a technique which has been shown to measure a far greater amount of emissions from biomass burning than conventional techniques such as GC, due to measurement of additional species such as small oxygenates, phenolics and furanics. The emission factors used came from a dataset of 19 experiments and ranged from 4 to 69 g kg^{-1}. When the exact residue was measured (e.g., rice straw, wheat straw, sugarcane and millet) the emission factor was used, but for crops which were less widely produced, emission factors were not measured and the average crop value calculated by Stockwell et al. (2015) was used. This generalisation of emission factors measured by PTR-ToF-MS, and lack of measurements of some residues (e.g., sugarcane), led to uncertainty in the overall estimation. Notably these samples were not from India, with rice straw samples from China and Taiwan and millet from Ghana. Uncertainty was largest for generalised emission factors applied to crops with lower yields as well as millet and sugarcane, as these were only measured from two burns. However, high emissions from sugarcane were recorded previously using FTIR (Stockwell et al., 2014), which helped to validate the higher emission factor used in this study. Measurement of emission factors from combustion of crop residues collected from fields in India, as well as improved understanding of the quantity of crop residues burnt on fields, is required to better evaluate this source.

4.5. PAHs

The estimate of PAH emissions from cow dung cake and MSW combustion remained the most uncertain due to the low number of samples. These emission factors were based on only three samples and a

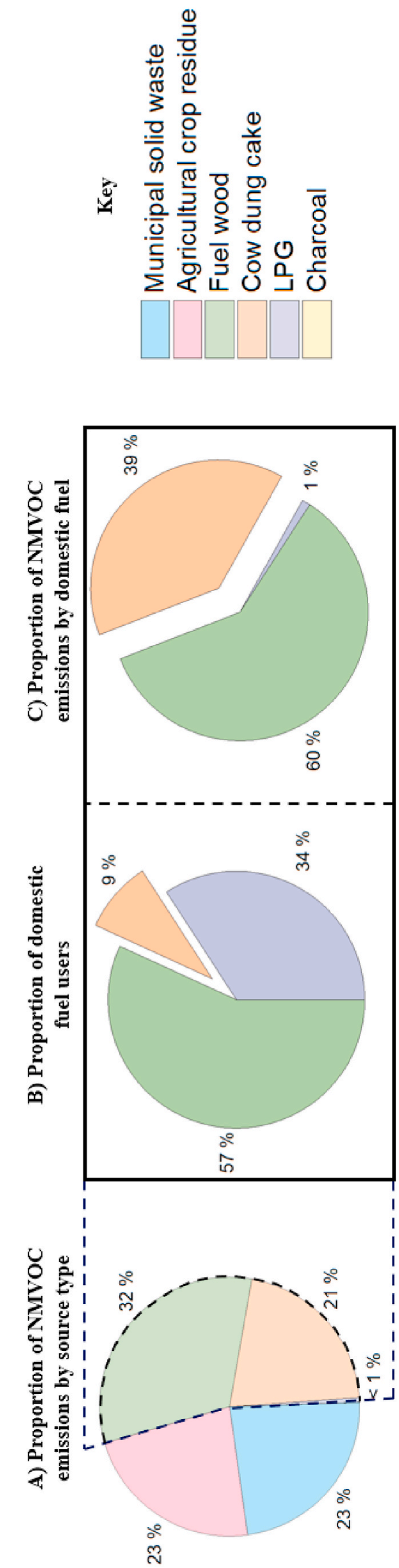

Fig. 5. Breakdown of contributions of different burning source sectors to emissions in 2011 where A = relative contributions of different burning sources to total burning related NMVOC emissions, B = relative proportion of number of residential fuel users for fuel wood, cow dung cake and LPG and C = relative proportion of fuel wood, cow dung cake and LPG to residential combustion related NMVOC emissions.

Table 4
NMVOC pollution (Tg yr^{-1}) from various fuel types in India. NMVOC emissions from charcoal were omitted and are in the range 2–6 × 10^{-3} Tg yr^{-1}. NMVOC emissions from coal were omitted and decreased from 11 × 10^{-3} Tg in 1993 to 4 × 10^{-3} Tg in 2016.

Year	Wood	Dung	LPG	MSW	Crop	Total
1993	3.8 (0.9–19.9)	2.5 (0.9–3.2)	0.02 (0.006–0.03)	2.1 (1.0–4.6)	–	8.4 (2.8–27.7)
1994	3.9 (0.9–20.0)	2.5 (0.9–3.2)	0.02 (0.006–0.03)	2.1 (1.1–4.7)	–	8.5 (2.9–27.9)
1999	3.9 (0.9–20.4)	1.9 (0.7–2.4)	0.03 (0.01–0.06)	2.3 (1.2–5.3)	–	8.1 (2.8–28.2)
2002	4.0 (0.9–21.3)	2.6 (1.0–3.4)	0.04 (0.01–0.07)	2.4 (1.3–5.6)	–	9.0 (3.2–30.4)
2005	4.2 (1.0–21.6)	2.6 (1.2–4.1)	0.05 (0.02–0.09)	2.7 (1.4–6.2)	–	9.6 (3.6–32.0)
2006	3.6 (0.8–18.6)	4.1 (1.9–6.5)	0.05 (0.02–0.09)	2.8 (1.4–6.3)	–	10.6 (4.1–31.5)
2007	4.3 (1.0–22.3)	2.4 (1.1–3.8)	0.06 (0.02–0.09)	2.8 (1.4–6.4)	–	9.6 (3.5–32.6)
2010	4.4 (1.0–22.7)	1.9 (0.9–3.1)	0.07 (0.02–0.11)	3.0 (1.5–6.7)	–	9.4 (3.4–32.6)
2011	4.3 (0.9–22.3)	2.8 (1.3–4.4)	0.07 (0.02–0.12)	3.0 (1.6–6.9)	3.0 (1.4–4.5)	13.2 (3.8–33.7) [a]
2016	4.0 (0.9–20.5)	3.1 (1.4–4.9)	0.09 (0.03–0.16)	3.3 (1.7–7.5)	–	10.5 (4.0–33.1)

[a] Includes estimate from crop residue burning on fields in India in 2011.

better assessment is needed, as the effect of composition and moisture content of fuels on PAH emission was not accounted for in this study. MSW and cow dung cake samples in Stewart et al. (2021b) had high emission factors, likely due to the low modified combustion efficiencies of the burns. In addition, this study quantified 21 major PAHs; however, the total number of PAHs released from solid fuel combustion was likely to be larger than predicted in this study (Stewart et al., 2021b).

5. Inventory comparison

Fig. 4A shows the spatial distribution of the total NMVOC emissions estimated as part of this study from burning sources in India during 2011 of 13 (5–47) Tg. Residential combustion represented ~53% of total emissions with fuel wood and cow dung cake respectively contributing ~32% and ~21% of total NMVOC emissions (see Fig. 5A). MSW and crop residue burning on fields each contributed ~23% to total NMVOC emissions.

The inventory developed for this study in Fig. 4A was compared to inventories which were part of the Emission Database for Global Atmospheric Research (EDGAR 5.0, see Fig. 4B) and the Regional Emission inventory in ASia (REAS 3.2, see Fig. 4C). The estimated emissions from these inventories for residential combustion in the year 2011 (EDGAR 5.0 = 4.2 Tg, REAS 3.2 = 5.9 Tg, see Table 3) were of similar magnitude to this study of 7.1 (2.3–26.7) Tg. The larger emissions from residential combustion estimated in this study were likely driven by the larger NMVOC emission factors used as part of this study, which measured a greater number of gas-phase organic species. This study highlighted a potentially larger NMVOC source from the combustion of crop residue on fields of 3.0 (1.4–4.5) Tg when compared to EDGAR 5.0 of 0.6 Tg. It also highlighted that the waste sector (3.0 (1.6–6.9) Tg in 2011) may be responsible for a significantly greater NMVOC emission than estimated by EDGAR 5.0 (0.1 Tg) and REAS 3.2 (0.01 Tg).

One of the most detailed current India specific inventories focussed on the year 2010 and used a 36 km × 36 km grid. This estimated NMVOC emissions of 5.9 Tg yr^{-1} from residential combustion (Sharma et al., 2015). The emission factor for fuel wood (15.9 g kg^{-1}) used by Sharma et al. (2015) was comparable to our study (18.7 g kg^{-1}), however, that for cow dung cake (10.4 g kg^{-1}) was significantly lower compared to the present study (62.0 g kg^{-1}). Sharma et al. (2015) examined the percentage fuel use in urban and rural environments in India and used emission factors from comparable studies. Whilst the estimate was relatively close to that of this study (see Table 4, 6.2 Tg yr^{-1} from fuel wood and cow dung cake combustion in 2010), the scale of NMVOC emissions from cow dung cake and the countrywide spatial distribution of emissions were lost. Table 4 highlights how these NMVOC emission estimates may vary from year to year through the detailed use of different fuel use inputs.

This study also suggested a significant MSW burning source, often omitted from inventories, but which was calculated to represent ~23% of total NMVOC emissions from burning. The estimate of NMVOCs from burning in this study was larger than two previous estimates. Wiedinmyer et al. (2014) estimated NMVOC emissions of 1.8 Tg yr^{-1} from open MSW burning for 2010 and Sharma et al. (2019) estimated emissions of 1.4–2 Tg yr^{-1} for 59 NMVOCs in 2015. The larger NMVOC emission estimate in this study was due to measurement of a larger emission factor, partly driven by the inclusion of many additional NMVOCs. The NMVOC emission factor in this study was notably large and underlines the need for more detailed studies of NMVOC emissions from a greater number of MSW burning samples to truly understand the potential impact of this source.

The estimated total NMVOC emission from crop residue burning on fields for 2011 in this study was 3 Tg, around twice that estimated previously for 2008–2009 by Jain et al. (2014) of ~1.5 Tg. This was principally due to greater sugarcane production in this year and larger emission factors from PTR-ToF-MS studies of crop residue burning capturing a greater amount of NMVOC emissions. However, a need was identified for better characterisation of crop residues specifically burnt in India using these techniques.

The total NMVOC emissions estimated here were larger than other anthropogenic source sectors at a country-wide level (see the Supplementary Information S17).

6. Impact of selective source reduction

Cow dung cake combustion represented only 6–14% of total fuel use in India by number of users when considering fuel wood, cow dung cake, LPG, coal and charcoal, but was responsible for ~27–53% of total NMVOC emissions from these residential combustion sources when considering fuel use based on calorimetry data (see Fig. 5B and C). This significantly increased NMVOC emissions across the Indo-Gangetic Plain. NMVOC emissions from cow dung cake combustion were highly sensitive to small changes in consumption. An interesting case was 2006, which had approximately 540 million fuel wood and 140 million cow dung cake users. Table 4 shows that the NMVOC emissions from cow dung cake (4.1 Tg) exceeded those of fuel wood (3.6 Tg) and demonstrated that a relatively small number of users burning cow dung cakes could have a disproportionately large impact on total NMVOC emissions. Despite this, no factor in isolation could resolve the complex emissions of NMVOCs from burning sources in India, with multiple mitigation strategies required to target each of these different sources.

The emission model was used to evaluate the impact of potential emission reduction strategies. Two case studies were considered which aimed at 50% and 75% reductions in the total mass of NMVOCs released in 2011 (see the Supplementary Information S18 for more details). A 50% reduction in total NMVOC emissions was achieved through the complete conversion of cow dung cake users to LPG and a 65% reduction in emissions from agricultural crop residue burning on fields and MSW waste combustion. This impact was significant, with NMVOC emissions from India in 2011 reduced to 6.5 (2.0–26.4) Tg, with only a small increase in LPG emissions to 90 (30–154) Gg. The second case study required more significant reductions of 80% in agricultural crop residue burnt on fields and MSW burning, complete conversion of cow dung cake users to LPG and 55% conversion of residential fuel wood use to LPG. This resulted in NMVOC emissions of 3.3 (1.1–12.5) Tg in 2011, with LPG combustion emissions that only increased to 135 (45–233) Gg.

7. Evaluation of LPG uptake

Current NMVOC emission reduction policy in India is focussed on the replacement of solid fuels with LPG (Gould and Urpelainen, 2018). Recent government initiatives have included the Pradhan Mantri Ujjwala Yojana and Pratyaksh Hanstantrit Labh schemes (IEA, 2020). Fig. 1 shows that from 1993 to 2016 there were around 400 million new Indian LPG users, whilst levels of other fuel usage remained relatively constant. This policy of increased LPG uptake was calculated to only increase NMVOC emissions from 19 Gg in 1993 to 94 Gg in 2016 (see Table 4).

The effect of this policy was evaluated within the emission model, compared to these 400 million new LPG users burning solid fuels. This was achieved by comparing total NMVOC emissions in 2016 to a scenario where the proportion of LPG usage had not increased from the 1993 level. Whilst total emissions from solid fuel combustion in India remained high due to the large numbers of users, the policy of increased LPG uptake was estimated to have prevented NMVOC emissions of 2.9 (0.7–14.7) Tg by 2016 compared to these new users burning solid fuels.

8. Conclusions

This study compiled recently measured emission factors and fuel consumption data to evaluate the magnitude and spatial distribution of NMVOC emissions from different solid fuel combustion sources across India. This was achieved by producing high-spatial resolution emission inventories, which addressed the yearly magnitude and spatial distribution of emissions. This showed the relative contributions of fuel wood (32%), cow dung cake (21%, based on calorimetry data), municipal solid waste (23%), agricultural crop residue on fields (23%), charcoal (<1%), coal (<1%) and LPG (<1%) to burning related NMVOC emissions of 13 (5–47) Tg in 2011 in India. Small oxygenated, phenolic and furanic species represented half to three quarters of total emissions from the solid fuel combustion sources in this study. Better understanding of the chemistry of phenolic and furanic compounds is essential to further understand the impact of these reactive chemical species on air quality in developing regions, where burning is a large air-pollution source. Certain sources, such as the combustion of fuel wood and cow dung cake for cooking, will remain relatively constant throughout the year. Combustion of fuel wood for heating and lighting will however be higher during winter months, particularly in the north and mountain areas. Other burning sources, such as agricultural crop residue burning, will show large seasonality and occur predominantly during the kharif (Apr–May) and rabi (Oct–Nov) crop burning seasons. This was not accounted for in these emission inventories and means that these sources may have a disproportionally large impact on emissions during these seasons.

This study showed that cow dung cake was a disproportionally high NMVOC emission fuel and was responsible for a high proportion of total residential combustion related NMVOC emissions, particularly across the Indo-Gangetic Plain. This study also evaluated current emission reduction policies from 1993 to 2016, which incentivised LPG uptake, and were predicted to prevent emissions of almost 3 Tg of NMVOCs a year by 2016. Despite this, total NMVOC emissions were here calculated to increase by over 2 Tg over this period, highlighting the limits of this policy in the face of rapid population expansion. For successful future net NMVOC emission reduction, policy should focus on replacement of solid fuels with LPG or other low emission renewable energy sources at a rate faster than the increase in population. Emission reduction from residential combustion can be accelerated by selectively replacing cow dung cake fuel use with LPG. This will lead to a three to four times greater reduction in NMVOC emissions per user compared to each fuel wood user replaced when considering the lower limit scenario based on calorimetry data. In addition, countrywide measures are required to prevent the burning of agricultural crop residues on fields and of MSW to reduce the significant NMVOC emissions from these source categories.

Data availability

The inventories produced as part of this study are available at The Centre for Environmental Data Analysis, UK (https://doi.org/10.5285/fdb8960260a64c5faf652f8f47c4df81).

Author contributions

GJS collected model inputs, developed emission model, evaluated results in context of literature and led paper. SSMY contributed to R code for emission model development and advised on the on-field crop residue burning emission estimates. BSN, JRH, WJFA, ARV, CNH, OW, EN, RG, LKS, TKM, ARR, BRG, JDL and JFH assisted with data interpretation.

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.2021.100115.

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