

Effect of processing on emission characteristics of coal briquettes in cookstoves



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ARTICLE INFO

Article history:

Received 6 November 2020

Revised 21 May 2022

Accepted 7 June 2022

Available online 18 June 2022

Keywords:

Bio-coal

Briquettes

Fuel processing technology

Clean coal technology

Health risk

ABSTRACT

Solid fuel usage for domestic cooking energy needs is an integral activity in several developing countries. Emissions from the daily usage of solid fuel lead to release of various pollutants which have adverse health effects. In the present study, an attempt was made to determine the influence of processing of fuel on emission characteristics from cookstoves. Processed fuel included carbonised and briquetted forms of raw coal which were tested in traditional and improved Indian cookstoves. Emission factors (EFs) were developed for fine particulate matter (PM_{2.5}), Carbon Monoxide (CO), Elemental Carbon (EC), Organic Carbon (OC), Polycyclic Aromatic Hydrocarbons (PAHs) and trace metals. Overall, mass-based PM_{2.5} reductions of ~40 % by use of carbonised coal and ~47 % by use of coal briquettes were achieved. Carbonaceous aerosols were found to be the most dominant form of PM_{2.5} mass fraction (50–62 %) for all stove fuel combinations. PAHs and trace-metals represent ~1–5 % of the overall mass but can be critical from a health perspective. EFs of trace metals increased when shifting from raw to processed fuel pointing towards the need for standardization for processed fuel recipes.

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Introduction

Household air pollution (HAP) is one of the leading risk factors for global disease burden (Lim et al., 2012). 60 % of the households in South-east Asia use solid fuel for cooking, and almost 2.8 billion people worldwide are exposed to HAP due to solid fuel usage for domestic energy requirements. In 2014, 3.04 billion people did not have access to clean fuels and technologies for cooking (Framework, 2015). In India, around 3.6 million households use coal as domestic cooking fuel. A previous study in our group found the usage of coal as primary domestic fuel in all the coal mining regions of India (Bojjagani, 2016). The use of coal for cooking produces pollutants like PM_{2.5} (particulate matter with an aerodynamic diameter ≤ 2.5 μm), Carbon monoxide (CO), Oxides of nitrogen (NO_x), Oxides of sulphur (SO_x), Polycyclic-Aromatic Hydrocarbons (PAHs) and toxic metals (Chen et al., 2015; Li et al., 2016b; Oanh et al., 1999) which cause various health and environmental problems.

There is a crucial need for promotion of schemes to shift to clean and energy efficient cooking technologies in rural settings. Only 53 % of households in India have access to Liquefied Petroleum Gas (LPG), of which only

28 % use it as a primary source for cooking (Census, 2011). A recently launched initiative by the Indian government to provide access to clean cooking energy - Pradhan Mantri Ujjawala Yojana (PMUY) offered free LPG connections to various households across India (Patnaik & Tripathi, 2017). However, recent reports reveal that PMUY has failed to meet its deliverables (Patnaik & Tripathi, 2017; Pillarisetti et al., 2019).

There are primarily two approaches to reduce emissions from cookstoves in kitchens: (a) deployment of Improved Cook Stoves (ICs) (Grieshop et al., 2011; Pande et al., 2019; Reece et al., 2017; Wathore et al., 2017) and; (b) use of processed low emission fuels (Ge et al., 2004; Nwabue et al., 2017; Qi et al., 2017; Zhi et al., 2009). There are several coal/ coal-blend cookstoves reported in the literature, such as the TJ4.0 (Altanzul, 2017) and KG5 (World Bank, 2019) that are effective in reducing emissions. ICs have several limitation which makes them very difficult to deploy (Khandelwal et al., 2017; Lewis et al., 2015).

Emissions from cookstoves result from a combination of several factors that include fuel properties, combustion type and operator methods (Roden et al., 2006). In several studies, ICs have been found to produce more emissions than traditional stoves (Kar et al., 2012; Patel et al., 2016; Tryner et al., 2016). Certain stoves are designed specific to a particular fuel composition (Current Science, 2010) and fail to perform efficiently with alternate fuels (Patel et al., 2016; Tryner et al., 2016). Altanzul, 2017 found that unprocessed coal burns cleanest

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in a stove optimized for that fuel. However, several studies report the processed form of briquettes have high efficiency and low emissions compared to raw coal (Ge et al., 2004; Juan Qi et al., 2017; Zhi et al., 2009). This is mainly because of the use of sub-optimal stoves that were not designed to burn the raw fuel properly. Altanzul, 2017 demonstrated that a properly designed raw coal combustor burns cleaner than the same stove burning several processed fuels.

Due to low adoption of IC and un-affordability of LPG, processing of fuel can be an advantageous strategy to reduce emissions from cookstoves. Several policy-based analyses suggest fuel processing as a possible strategy to reduce emissions from cookstoves (Anenberg et al., 2013; Zhi et al., 2009). Recent clean cooking policies developed by the Indian government recommend focus on clean fuels (Aayog, 2017). Use of processed fuel in optimized stoves for reducing emissions, increasing efficiency and reducing fuel consumption has been recommended in several publications, including – so called ‘smokeless fuel’ (Atiku et al., 2016; Habib et al., 2014; Lasek et al., 2019; Mitchell et al., 2016; Nwabue et al., 2017), ‘carbonised fuel’ (Li et al., 2016a; Li et al., 2019), ‘torrefied fuel’ (Atiku et al., 2016; Jones et al., 2017; Qi et al., 2018), ‘briquettes’ (Qi et al., 2017; Zhi et al., 2009) and ‘pellets’ (Champion & Grieshop, 2019; Pradhan et al., 2018).

IC or processed fuel cannot be considered ‘clean’ individually and need to be addressed in combination. In the present study, an attempt was made to understand the role of processing of fuel on emission characteristics. Raw and processed coal in combination with several cookstove types available in India were assessed in the laboratory for emissions of PM_{2.5}, CO, EC-OC, PAHs and trace metals. Processed coal included carbonised and briquetted forms, and the stoves used for this study represented traditional and ICs available in the region. This is perhaps the first study from the Indian sub-continent to quantify source profiles from coal and coal-blend cookstoves for a wide array of pollutants. The emission factors developed can be used for different modeling and inventorization efforts.

Material & methods

Stove and fuel types

The stoves used in the present study include, Traditional - Chandrapur Stove (CS) and IC - Neerdhur Stove (NS) and Varanasi Stove (VS). CS

Table 1
Characterisation of coal and coal briquettes used for the present study.

Proximate (%)	Raw coal (RC)	Handmade coal briquettes (HCB)
Moisture Content (as received)	7.40	5.34
Volatile Matter (as received)	24.79	33.34
Fixed Carbon (as received)	40.64	37.50
Ash (as received)	27.17	23.88
Calorific Value (MJ/kg) (as received LHV)	23.7	20.1

and VS were procured locally from Chandrapur, Maharashtra, a coal mining region of India, and Varanasi, Uttar Pradesh – a city in northern India. CS and VS are used locally for cooking and water heating purposes. A multi-fuel natural draft Stove -NS certified by the Ministry of New and Renewable Energy, India, was used as an IC. Detailed description of CS and VS can be found in Das, Bhandarkar, et al. (2019) and Das, Dutta, et al. (2019), while specifications concerning NS are available in Gupta et al., 2020; M. Sharma & Dasappa, 2017.

Fuel used for the present study include raw coal, carbonised coal and coal briquettes. Raw Coal (RC) was procured from Chandrapur (a mining region of India) representative of the coal being used by local people. Coal is broken to a size that is typical of user practice, and a size analysis is carried out using an image analyzer. The volume average diameter is determined to be 45 mm. Handmade coal briquettes (HCB) were made using locally available powdered coal, wheat flour and cow-dung, keeping in mind the experimental formulation adhering to the traditional practices of the region. Coal, wheat flour and cow-dung were powdered and sieved through 18 mesh (< 0.825 mm), separately. The mixture of coal: wheat flour: cow dung in a ratio of 8:1:1 by mass was then well-mixed and water (approximately 1.5 times) by weight was added for making the hand-made briquettes with a volume average diameter of ~50 mm. The briquettes were sun dried (local average temperature: 25–30 °C) for a minimum of 5 days before carrying out experiments. Fig. SI-1 gives the flowchart of the briquetting process used in the study. The details of the fuel samples used for the present study is shown in Table 1. The energy content of the fuel reported here is the lower heating value (LHV) of the fuel. In addition to the raw coal and HCB mentioned above, raw coal samples - CL1 and CL2; their carbonised forms CL1CK and CL2CK, and a commercially available coal briquette - CB were also burnt in different stoves, and their emissions were analysed as indicated in the experimental matrix (Table 2). Details of the characterisation of CL1, CL2, CB are provided in Das, Bhandarkar, et al., 2019 and of CL1CK and CL2CK are provided in Das, Dutta, et al., 2019.

Sampling setup

Gas phase and particulate emissions measurements were taken using a Portable Emission Measurement System (PEMS) developed by Aprovecho Research Center (Coffey et al., 2017; MacCarty et al., 2010). The setup was modified by (a) placing the blower after the sampling port and (b) fabrication of a portable stand which could be easily assembled and deployed for field sampling conditions (Fig. 1). Fig. SI 2 shows the schematic of PEMS and modified PEMS.

The distance between the cookstove and the hood was adjusted such that the lower end of the hood was around 100 cm from the top of the cookstove. Isokinetic sampling was used to collect emissions from the sample port in the sample duct through a polytetrafluoroethylene (PTFE) tube. The sample line was connected to three lines for the PEMS sensor box and two gravimetric samplers. The sample entering the PEMS sensor box passed through a filter, followed by a suction pump (≈4.6 lpm) and CO/CO₂ sensor. PM_{2.5} measurements were

Table 2
Experimental matrix used in the present study to measure emissions released from several stove fuel combinations. ■ = Das, Bhandarkar, et al. (2019); ■ = Das, Dutta, et al. (2019) and ■ = Present Study.

	Coal						Carbonised Coal				Briquettes			
	Chandrapur Stove (CS)			Varanasi Stove (VS)			Chandrapur Stove (CS)		Varanasi Stove (VS)		Chandrapur Stove (CS)		Varanasi Stove (VS)	
	CL1	CL2	RC	CL1	CL2	RC	CL1CK	CL2CK	CL1CK	CL2CK	CB	HCB	CB	HCB
PM _{2.5}	■	■	■	■	■	■	■	■	■	■	■	■	■	■
CO	■	■	■	■	■	■	■	■	■	■	■	■	■	■
OC	■	■	■	■	■	■	■	■	■	■	■	■	■	■
EC	■	■	■	■	■	■	■	■	■	■	■	■	■	■
PAHs	■	■	■	■	■	■	■	■	■	■	■	■	■	■
Elements	■	■	■	■	■	■	■	■	■	■	■	■	■	■

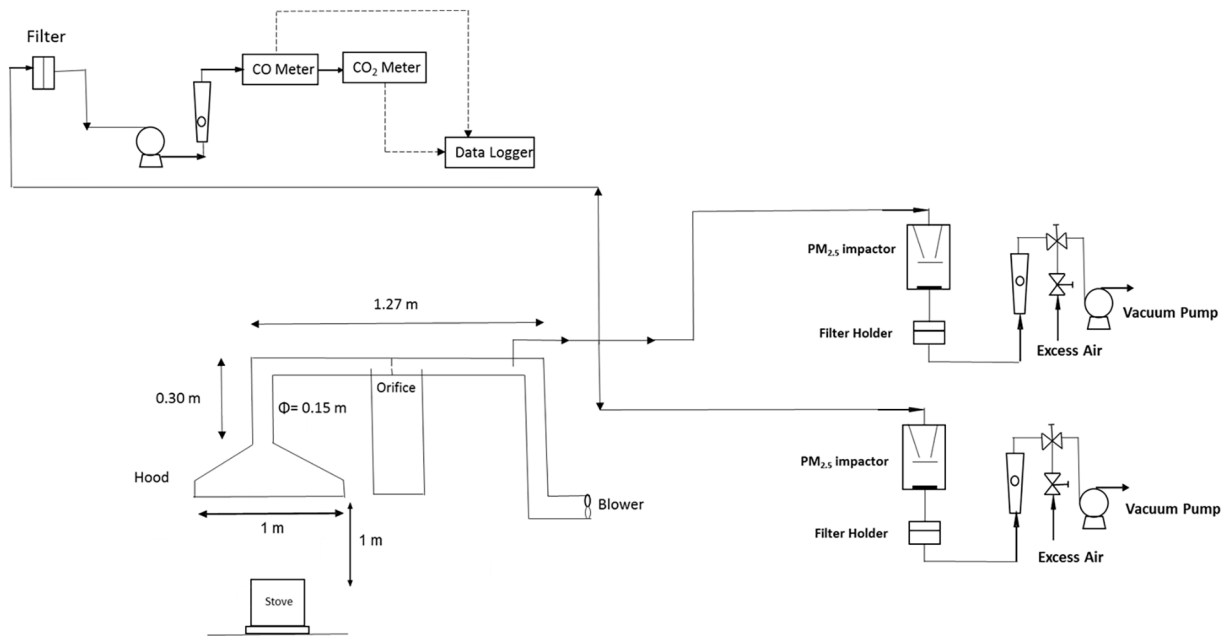


Fig. 1. Schematic diagram of the measurement setup for sampling of PM_{2.5}, CO and CO₂ emissions from cookstoves used for the present study.

carried out gravimetrically using a Airmetrics™ MiniVol™ PM_{2.5} single stage impactor with quartz fiber and PTFE filter substrates connected to a pump at the end of the sampling train operating at 5 lpm. Samples collected on the quartz and PTFE filters were used for EC-OC and trace metal analyses, respectively. The filters were desiccated for 24 h and weighed in triplicates before carrying out the experiments. The filter samples were desiccated after the experiments for 24 h and weighed. The difference in weights before and after sampling gave the mass of PM_{2.5} deposited on the filter.

Sampling protocol

In the absence of cookstove testing protocols available for performance evaluation of coal or coal-blend fuels, a new protocol was designed based on Indian BIS protocol (BIS, 1991) and WBT 4.2.3 (WBT 4.2.3, 2013). The emission sampling cycle resembled the local cooking practice of igniting the coal using wood and kerosene and a cooking period of one hour. Once the fuel was ignited and a stable flame was visible, the pot was placed on the cookstove. Emission characteristics from cookstoves may be affected by the size and temperature of the pots used for cooking (Christian L'Orange, Volckens, et al., 2012). In the present study, cooking pots available in the local region were used to replicate field condition. The pot used was made from stainless steel with a height of 9.75 cm, diameter of 21.30 cm and volume of 3 l. Empty pot mass was approximately 1.486 kg. One hour of cooking time occurred after the pot containing two litres of water was placed on the stove. After every 10 min, 150 g of coal or 110 g of briquettes were added to the cookstove for maintaining consistency and repeatability in different experimental runs. The fuelling method and burn rate was kept consistent with the local cooking practice for each stove fuel combination. Water was heated from ambient temperature to the boiling point, 100 °C, and water continued boiling until the end of the cooking period. The ignition time, time to boil, mass of water evaporated, fuel consumed, mass of PM_{2.5}, mass of CO and mass of CO₂ were recorded for each experiment. Experiments were performed in triplicate for each stove-fuel combination.

After one hour from the starting point of the pot being placed, the fuel was quenched with water replicating field-based practices. Ash attached to the surface of coal or coal briquettes was removed by this

process. The leftover fuel was oven dried for 24 h at 104 °C to remove any moisture from the fuel. The final mass of the fuel burnt was calculated on dry-basis by subtracting the fuel left in the combustion chamber from the initial mass of the fuel.

The emission factors were calculated using Water Boiling Test 4.2.3 (WBT 4.2.3, 2013) exhaust flow method as shown in Eqs. (1) and (2) below:

$$\text{Emission Factor of Pollutant} \left(\frac{\text{g}}{\text{kg}} \text{ or } \frac{\text{g}}{\text{MJ}} \right) = \frac{\text{Total emission of Pollutant (g)}}{\text{Total fuel used (in kg) or Energy delivered to the pot (MJ)}} \quad (1)$$

where,

$$\text{total emission of pollutant(g)} = \text{Flow rate through the hood, } Q (\text{m}^3/\text{s}) \times \text{average pollutant concentration in the exhaust gas} (\text{g}/\text{m}^3) \times \text{time taken for testing(s)} \quad (2)$$

Different studies report different metrics to evaluate the thermal efficiency of a stove-fuel combination. Jetter et al., 2012 defines overall thermal efficiency as a product of combustion efficiency and heat transfer efficiency. WBT 4.2.3 is divided into three phases: cold-start high power phase, hot-start high power phase and simmer phase. In the present study, a modified water boiling test inclusive of only the cold-start high power phase was designed to measure the thermal efficiency from different coal fuelled cookstoves representative of field-based practices. Thermal efficiency of the stove-fuel combination is calculated as the ratio of amount of the energy delivered to the pot to the energy released when fuel is burnt, as shown in Eq. (3) (WBT 4.2.3, 2013). The energy transferred to the water is the sum of the latent heat, sensible heat, and the heat transferred away from the pot via convection, conduction, and radiation. Heat transferred away from the pot, while dominant during the simmering phase of the WBT 4.2.3 test, is negligible during the high-power test phase used in this study.

$$\text{Thermal Efficiency} = \frac{\text{Energy delivered to the pot}}{\text{Energy of the equivalent dry fuel}} \quad (3)$$

Energy delivered to the pot is calculated as the sum of energy required to raise the temperature of water in the pot from room temperature to 100 °C and the amount of energy required to evaporate the water during the cooking cycle (Eq. (4)). The energy consumed is the equivalent mass of dry fuel consumed multiplied by the heating value (Eq. (5)). Calorific value of the remnant fuel was assumed to be same as that of the raw fuel, and this assumption can be identified as a possible limitation of the study. C. L'Orange, DeFoort, et al. (2012) identified that uncertainty in cookstove test results could be reduced by using fuels with consistent moisture content. Thus, in the present study fuel was oven dried for 24 h at 104 °C to remove any moisture from the fuel. However, this is a limitation of the present study which does not reflect the actual use condition (Van Zyl et al., 2019; Yuntunwi et al., 2008).

$$\text{Energy delivered to the pot} = MC_p\Delta T + mL \quad (4)$$

M = Initial mass of water in the pot;

C_p = Specific heat of water;

ΔT = Rise in Temperature of water;

m = mass of water evaporated;

L = Latent heat of water;

$$\text{Energy of the equivalent dry fuel} = (m_f - m_i) \cdot C_v \quad (5)$$

m_f = final mass of the fuel

m_i = Initial mass of the fuel

C_v = calorific value of fuel

Characterisation of particulate matter

The PM_{2.5} filter samples were analysed for EC-OC, PAHs and trace metals. In addition to the filter samples from the stove fuel combinations discussed in the present study, the samples from Das, Bhandarkar, et al. (2019) and Das, Dutta, et al. (2019) were also used to determine the PAHs and trace metal composition within the particulate matter. A detailed description of the experimental matrix is shown in Table 2.

Organic Carbon (OC) and Elemental Carbon (EC) concentrations were determined on a DRI Model 2015 Multiwavelength Thermal/Optical Carbon Analyzer using IMPROVE_A protocol. Analytical uncertainty of <7 % was estimated through replicate analyses of the samples (n = 6) for each measured species.

Detection of organic species was carried out using Gas Chromatography/Mass Spectrometry (Agilent GC7890B 5977B MS). Samples were extracted using 50/50 dichloromethane and acetone in a sonicator, followed by evaporation in a rotary evaporator, and blowing down ultrapure nitrogen (Villalobos et al., 2015). The samples were passed through a syringe into a 0.22 μm Polyvinylidene fluoride (PVDF) membrane filter. Two aliquots of the extracts were analysed by GC/MS.

A closed-vessel microwave acid digestion based system (Anton Paar, Austria) was used to extract the filtered samples for metals analysis (Yenisoy-Karakaş et al., 2009). A series of blanks (i.e., both filter and acid blanks) were prepared using the same digestion procedure. All the reagents used in the analysis were analytical or higher grade. The digested samples were analysed for Cr, Mn, Co, Zn, V, Be, Ba, Th, Bi, Ge, Sc, Ni, Cu, As, Pb, Se, Cd and Ag using inductively coupled plasma mass spectrometry (ICP-MS; Thermo fisher Scientific Inc., USA) (USEPA 1999c) (Agency, 1999).

Results and discussion

Improved combustion performance: ignition time, time to boil water and thermal efficiency

Comparisons between the stove-fuel combinations used in the study, were done on the basis of ignition time, time to boil water and

thermal efficiency. Ignition time was defined as the time taken by a stove-fuel combination to produce a stable visible flame. HCB with NS took the least average time to ignite (6 ± 0.01 min) followed by HCB with CS (8 ± 0.58 min), RC with NS (15 ± 0.58 min) stove and RC with CS (19 ± 1.15 min) (Fig. 2). The addition of biomass in the form of binder increased the volatile content of coal briquettes by ~35 % (Table 1) compared with raw coal, which made the briquettes ignite faster (Chen et al., 1996). Further, better ignition rate in NS can be attributed to the enhanced combustion from the secondary air inlets (Caubel et al., 2018; Rapp et al., 2016), which were not available in CS, leading to a high temperature in the combustion zone of the stove and thus leading to faster ignition.

Time taken to boil water in the pot to reach 100 °C was an indication of the rate at which energy was delivered to the pot or power delivery. Fig. 2 shows the comparison of time to boil for the stove-fuel combinations used in the study. RC with NS took the least time to boil followed by HCB with NS, HCB with CS and finally, RC with CS. The time to boil was almost the same for CS-HCB, NS-RC and NS-HCB but almost 2.5 times more in the case of CS-RC. This finding can be attributed to insufficient air available for coal to burn after the pot was placed on CS. The small holes in the grate and sides of CS were almost covered by the coal, and the top opening after putting on the pot was further closed leaving less available air for complete combustion.

Thermal efficiency was measured to be 19.8 % for CS-RC, 23.2 % for CS-CB, 25.5 % for NS-RC and 26.8 % for NS-HCB. Usage of coal briquettes led to 5–17 % increase in the thermal efficiency. The briquette hand making process generated enough pressure to bind the coal-biomass mixture together while creating pores within the solid fuel matrix to ensure entry of oxygen within them, leading to increased combustion efficiency (Qi et al., 2017). Further, the pre-heated secondary air from side holes, primary air from the hole below the grate, uniformity of briquette shape and less resistance for air to flow from the bottom to the top of the combustion bed in coal briquettes cause the NS-HCB combination to generate stable blue flame similar to LPG, leading to higher combustion efficiency.

Reduced emissions: PM_{2.5} and CO

The quantitative values of emission factors of PM_{2.5} and CO developed in the study are shown in Table 3. Use of improved cookstove with coal briquettes has the potential to reduce PM_{2.5} emissions by 65 % (13.85 t/year), use of improved stove with coal by 55 % (18.70 t/year) and use of traditional stove with coal briquettes by 45 % (22.69 t/year). The present results follow similar trend with other studies – 61–67 % (Qi et al., 2017) and 34–44 % (Zhi et al., 2009). The reductions in both PM_{2.5} and CO emissions with the change in stove-fuel combinations point towards the potential advantage of shifting to processed fuel.

The PM_{2.5} EFs for some coal-burning stoves have closely correlated with the volatile matter content (Li et al., 2016a). Unburnt volatile matter can act as a PM precursor and can positively contribute to the formation of particles (Li et al., 2016a,b). Briquettes are usually easily ignitable (Nwabue et al., 2017) and thus reduce the release of initial emissions. Lower ignition time from briquettes combined with IC point towards reduced time weighted average exposure of emission from cooking related activities. Carbonised forms of solid fuels tend to produce more CO emissions than their original forms (Jetter et al., 2012; Mitchell et al., 2016). Gasification of coal can produce syngas that can be more readily combusted with lower emissions of CO and other pollutants. (Sutar et al., 2015; Varunkumar, 2012). A solution to reducing emissions lies in creating staged combustion and complete gasification of the volatile fraction of the fuel. The success of this approach has been reported by World Bank, 2019 and the positive impact on health and air quality has been reported by van Gemert et al., 2019. Patel et al. (2016) found an increase in the air draft of a forced draft cookstove resulted in the decrease in residence time of volatile organic compounds in the flame zone, which in turn increased pollutant concentrations.

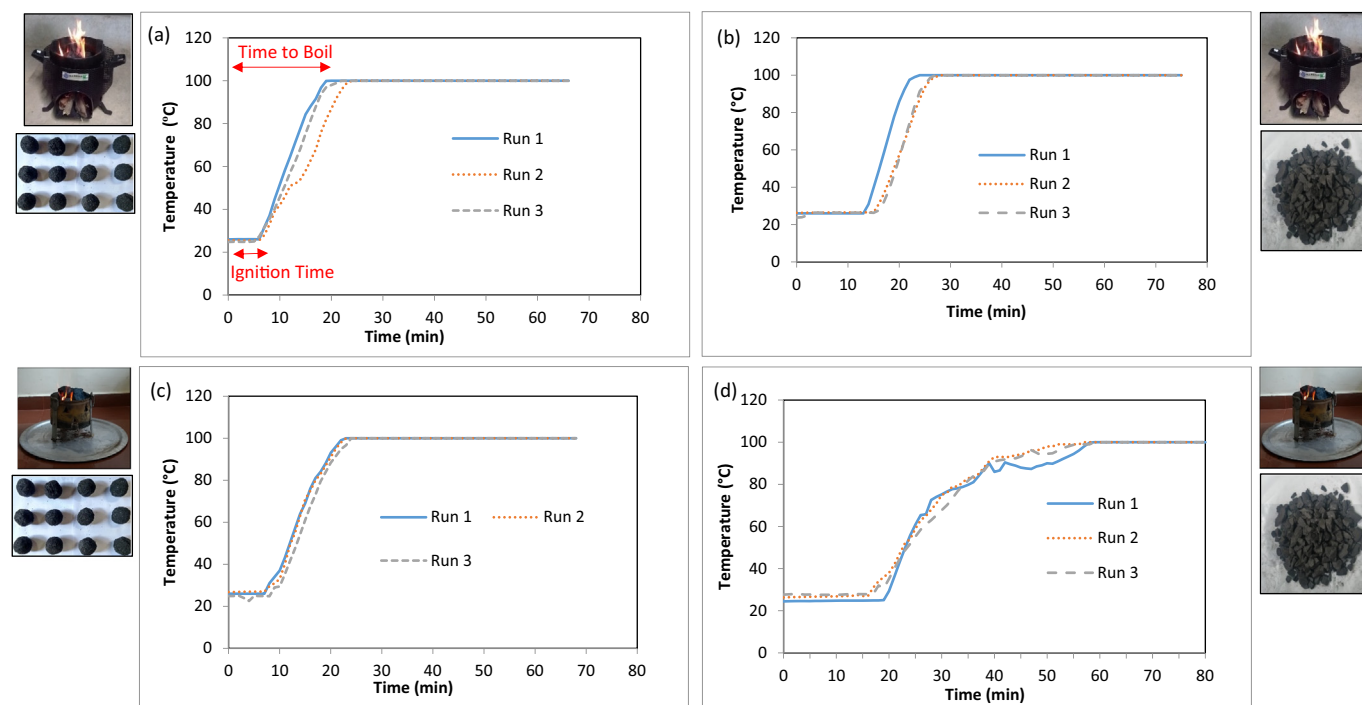


Fig. 2. Temperature plot w.r.t time for water in the pot for complete combustion cycle for four cases: (a) NS + HCB: Neerdhur Stove with Handmade Coal Briquettes; (b) NS + RC: Neerdhur Stove with Raw Coal; (c) CS + HCB: Chandrapur Stove with Handmade Coal Briquettes (d) CS + RC: Chandrapur Stove with Raw Coal.

Findings of the study clearly indicate that HCB performed better than RC in both the traditional and IC. None of the stoves were able to burn the fuel efficiently, and there is a need for more efficient stove designs. IC and processed fuel combination was found to be the most favourable option as it has highest $PM_{2.5}$ and CO emission reduction potential, lowest ignition time and highest thermal efficiency (Table SI 1). In practicality, transition to combination of IC and HCB is difficult to implement as it requires changes in both the existing stove and the fuel. Such transition may require a sudden change in cooking habits of people which may lead to low adoption of the stove-fuel combination. The pathway focused towards the change in the fuel used from coal to processed coal may be more fruitful for regions where adoption rate of IC is poor, powdered coal is available, and people are ready to change the fuel but not the cookstove.

Chemical characterisation: EC-OC, PAHs and trace metals

Carbonaceous aerosols, comprised of elemental and organic carbon, were found to be the dominant contributor (50–62 %) to the $PM_{2.5}$ aerosol mass fraction in all stove fuel combinations measured. Overall emission results indicate that the highest $PM_{2.5}$ emissions were emitted from raw coal samples (7.91 ± 3.49 g/kg), followed by carbonised coal samples (4.75 ± 1.19 g/kg) and subsequently the least emissions were emitted by the briquettes (4.18 ± 1.29 g/kg) (Table SI 2)(Fig. 3). The EFs of OC for raw, carbonised, and briquetted coal were

Table 3

The $PM_{2.5}$ and CO emissions factors in g/kg of fuel and g/MJ delivered for four cases: (a) CS + RC: Chandrapur Stove with Raw Coal; (b) CS + HCB: Chandrapur Stove with Coal Briquettes; (c) NS + RC: Neerdhur Stove with Raw Coal; (d) NS + HCB: Neerdhur Stove with Coal Briquettes. The standard deviations are also shown in brackets for triplicate runs.

Stove-Fuel combinations	$PM_{2.5}$ emission factor		CO emission factor	
	g/kg of fuel	g/MJ _d	g/kg of fuel	g/MJ _d
CS + RC	5.36 (0.57)	1.34 (0.49)	47.75 (1.91)	12.07 (1.50)
CS + HCB	3.49 (1.08)	0.74 (0.20)	48.76 (7.50)	10.44 (1.21)
NS + RC	3.10 (0.05)	0.61 (0.05)	29.69 (7.51)	5.77 (1.44)
NS + HCB	2.44 (0.43)	0.45 (0.10)	31.17 (3.06)	5.77 (0.23)

measured to be 3.14 ± 1.24 , 1.78 ± 0.52 and 1.36 ± 0.22 g/kg respectively. The EFs of EC for raw, carbonised, and briquetted coal were estimated to 1.49 ± 1.70 , 0.92 ± 0.35 and 0.56 ± 0.09 g/kg, respectively. OC was found to be the largest contributor to the $PM_{2.5}$ mass. EC abundances were 1.5–2.5 times higher for raw coal as compared to the processed coal, suggesting less complete combustion in raw coal (Chen, 2009; Ge et al., 2004; Zhi et al., 2008). The Carbonaceous aerosol fraction was measured to be 62 ± 16 % for coal, 56 ± 5 % for carbonised coal and 50 ± 13 % for coal briquettes. Average EC/ $PM_{2.5}$ ratio was found to be 0.20 for raw coal, 0.19 for carbonised coal and 0.15 for briquettes. Coal had the highest fraction of OC and EC in its particulate emission composition (42 %), followed by carbonised coal (37 %). EC to total carbon (TC = EC + OC) ratio ranged from 0.19 to 0.43 for all stove fuel combination, with an average of 0.32 for raw coal, 0.34 for carbonised coal and 0.29 for coal briquettes. The EC/PM ratio ranged from 0.08 to 0.34 for all stove fuel combination, with an average of 0.20 for raw coal, 0.19 for carbonised coal and 0.15 for coal briquettes. Findings of the study are well within the range of several other literature studies (Atiku et al., 2016; Roden et al., 2006). Kar et al. (2012) found the EC proportion to be highest for IC. While carbonaceous aerosols are associated with radiative forcing and climate change (Roden et al., 2006), there are some studies which report health effects associated with the organic fraction of carbonaceous aerosols and with black carbon (Harrison & Yin, 2000; Urch et al., 2004).

The emission factors of 20 PAHs are presented in Fig. 4. It was estimated that $\Sigma PAHs$ EF is 29.47 ± 7.91 mg/kg for raw coal; 23.36 ± 3.09 mg/kg for carbonised coal and 12.69 ± 0.31 mg/kg for coal briquettes. $\Sigma gen-PAHs$ (sum of six carcinogenic PAHs, Benzo(a)anthracene (BaA), Chrysene, benzo(b,k)fluoranthene (BbKF), benzo[a]pyrene (BaP), Indeno(1,2,3)cd-pyrene and Dibenz[a,h]anthracene (DbAhA) which are classified as GroupB2, BaPE (BaPE, benzo[a]pyrene equivalent carcinogenic factor = $BaA * 0.06 + BbKF * 0.07 + BaP + DbAhA * 0.06$) calculated for the different stove fuel combinations was estimated to be 4.45 ± 1.35 for raw coal, 4.26 ± 0.57 for carbonised coal and 2.14 ± 0.17 mg/kg for coal briquettes.

EFs of 18 trace metals were measured. Trace elements represented 0.27–5.11 % of $PM_{2.5}$ for all stove fuel combination. In the present

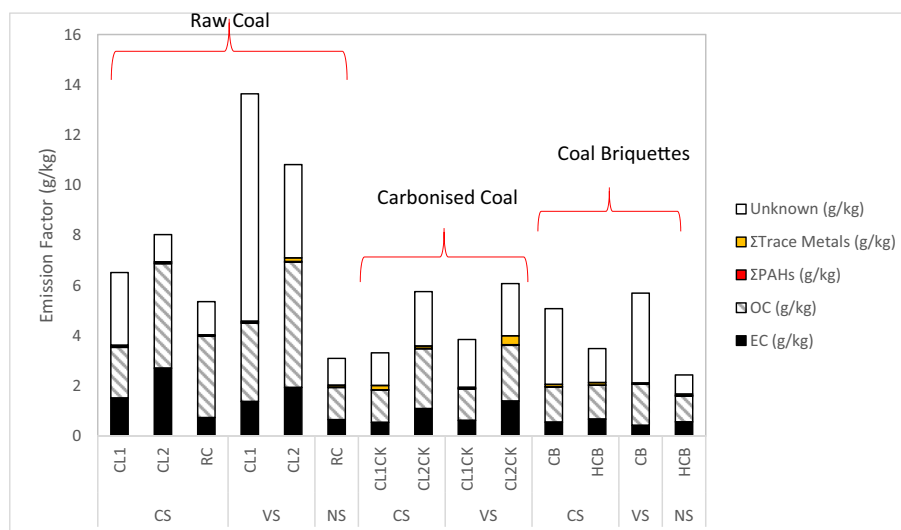


Fig. 3. Comparison of Emission Factors from different coal and coal-blend fuel. The proportions of EC, OC, PAHs and trace metals in the PM_{2.5} fraction are indicated to highlight the relative contributions.

study, the overall elemental emissions were highest for carbonised coal (163.11 + 118.92 mg/kg) followed by briquetted coals (70.95 + 22.96 mg/kg) and the least for raw coal (61.40 + 38.88 mg/kg) (Fig. 5) (Fig. SI 3). Li et al., 2019 also found carbonised form coal and biomass fuels having high mass-based emissions of trace elements. Sun et al., 2019 also found several elements had higher EFs in the processed form of coal as compared to raw fuels. Lead, cadmium, mercury and iron are some of the elements which have been shown to have toxic effects (Harrison & Yin, 2000). In the present study, the main contributors were Co, Zn and Ba (in descending order) – totally contributing to almost 90 % of elements. Se, As, Cr, Co, Cu, and Al are characteristic elements for emissions from coal combustion (Ge et al., 2004; Morawska & Zhang, 2002). Cobalt, one of the characteristic elements for emissions

from coal combustion (Ge et al., 2004; Morawska & Zhang, 2002), was the most abundant element with an order of magnitude higher than other elements. Ge et al., 2004 identified lead to be the most abundant element from coal cookstove emissions. In the present study, EF of Ni ranged from 0.29 mg/kg for NS-RC to 0.38 mg/kg for NS-HCB. The increase in Ni content in the PM mass fraction can be attributed to the biomass-based binding material used for the study (Sun et al., 2019). Briquetting of coal led to reduction in EFs of toxic elements As, Se and Pb. Similar findings were also reported by Qi et al., 2017. Decrease in Ba and Pb content due to carbonization and briquetting were also reported by Sun et al., 2019. Ge et al., 2004 also reported briquettes with higher emissions of Cd and Ni in PM_{2.5}. The phenomenon is closely linked to the ‘enlarged oxygen inlet through the porous structure in

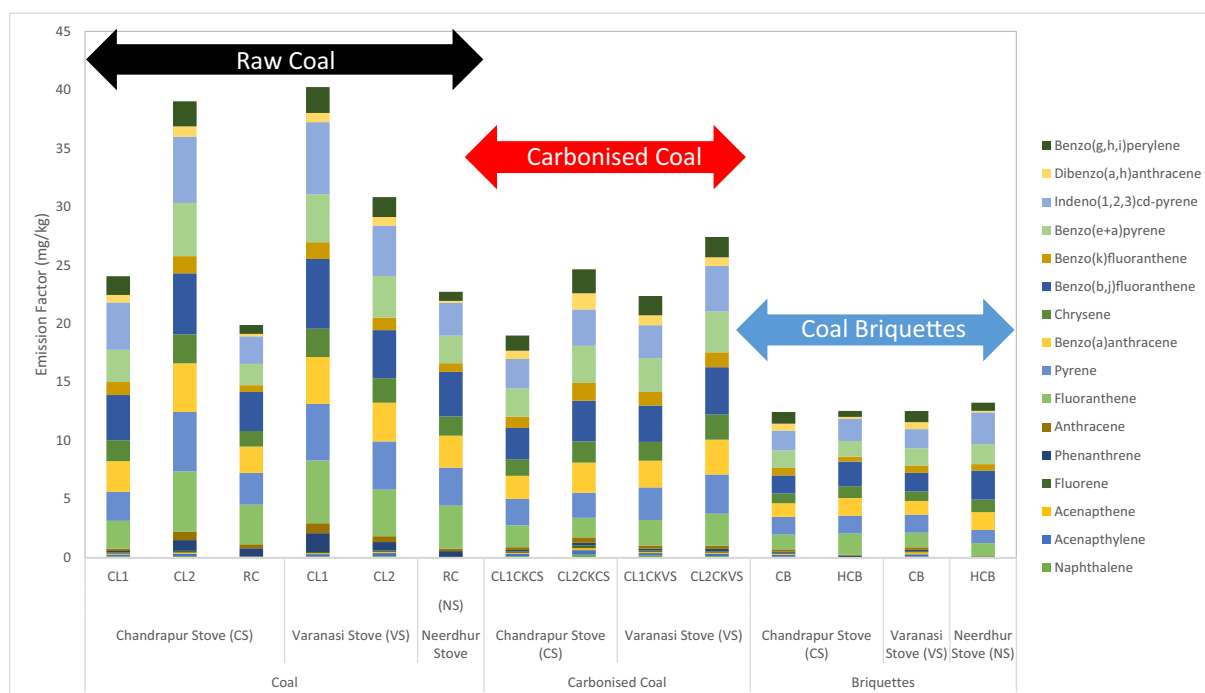


Fig. 4. Emission Factors (mg/kg) of individual PAHs from Domestic Combustion of Coal, Carbonised Coal, and Coal Briquettes.

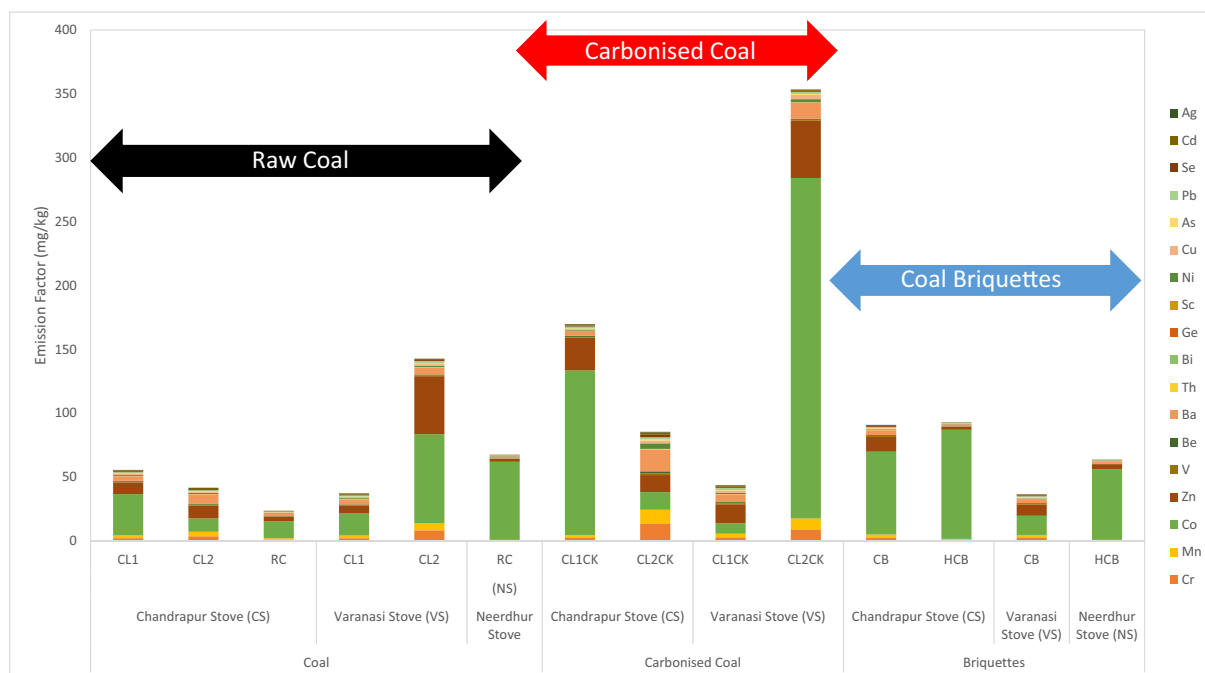


Fig. 5. Emission Factors of 18 trace metals for all tested samples of different stove fuel combinations. These trace metals were extracted from the corresponding PM_{2.5} samples.

the briquettes, possibly leaving more toxic elements in the form of oxides in the ash instead of releasing them into the air via flue smoke' (Qi et al., 2017; Sun et al., 2019; Liu et al., 2007; Li et al., 2016b). Any fuel processing that devolatilizes (semi-cokes) the fuel will concentrate the metal and ash content. This can be expected to increase the EF for those metals.

Although PAHs and trace metals occupy a negligible portion of the entire PM_{2.5} mass, they can be an important contributor from a health perspective. Harrison & Yin, 2000, in their review concerning particle properties which affect human health, identified trace elements as toxic in sizeable dose. Chemical characterisation of the particulate matter fraction reveals that while the overall mass-based emissions decrease with the processing of fuel, there can be a substantial increase in the 'toxic' component of the PM mass fraction, mostly attributable to PAHs and trace metals. Pulmonary response and cell injury following exposure to PM are related to soluble material (metal ions and PAHs) rather than to the number or composition of the insoluble particles (Adamson et al., 1999). Higher toxicity influences mechanisms of oxidative stress and inflammation leading to oxidative DNA damage and membrane lipid peroxidation. (Valavanidis et al., 2008). This increase in trace metals in the processed fuel is a major concern and should be considered in policy-based regulation. Further work is also needed towards the development of stoves with good combustion of raw/processed fuel which could lower PAHs and trace metal concentration on a mass basis.

The EFs of PM_{2.5}, EC, OC and PAHs for the stove-fuel combinations used for the present study demonstrated that raw coal > carbonised coal > briquettes. However, EFs of trace metal for the different stoves show that carbonised coal > coal briquettes > raw coal. Different studies report different trends in the order of EF for raw and different processed coal. Sun et al., 2019 found that charcoal (i.e., a carbonised form of biomass fuel) as more efficient in reducing PM_{2.5} and carbonaceous fraction emissions than briquettes. PAHs EFs showed: raw fuel > briquettes > charcoals (Sun et al., 2019), raw fuel > briquettes > charcoals (Oanh et al., 1999) and briquettes > raw fuel (Chen et al., 2015). The process of briquetting usually involves carbonization (Nwabue et al., 2017), which leads to an increase in the pyrolytic

activity of the coal and thereby an increase in the PAHs emission (Chen et al., 2015). Due to inter-dependence of the carbonization and the briquetting process, and due to many other variables, including the combustion process, it is difficult to make conclusions regarding which type of fuel is best for a particular stove.

The nature of emissions from a processed fuel, apart from appropriateness of the match between combustor and the fuel, can be affected by the fuel making process, carbonization temperature (Qi et al., 2018), duration of carbonization of raw materials, (Handayani, Ningsih and Meriansyah, 2019), types and nature of binders (González et al., 2020; Olugbade et al., 2019; Song et al., 2019), compacting pressure and processing temperature, (Olugbade et al., 2019), density, moisture content, binder percentage (Song et al., 2019) and method used for briquetting (Dinesha et al., 2019; Song et al., 2019). These factors strongly affect the combustion properties of the fuel, and depending on the combustion device (stove), influence the emission characteristics. Wide variability of the emissions generated from processed fuel requires the need for assessment of stove-fuel combinations, rather than assessment of fuels or stoves alone.

Conclusions

In the present study, we have attempted to quantify EFs of PM_{2.5}, CO, EC-OC, 20 PAHs and 18 trace metals from 3 types of raw coal, 2 carbonised coal and 2 briquetted coal in 2 traditional and 1 improved cookstove. The performance of any combustion device will depend on suitability of its design to the fuel quality. Some studies assume fuel itself is clean or polluting, without considering the type of stove being used. There is also a need to improve the existing protocols to represent field conditions adequately (Lombardi et al., 2017, 2018). This study highlights the need for stove-fuel-based testing protocols, where the fuel is characterised, and the stove appropriately selected for that fuel. Laboratory testing was done on improved cookstoves to evaluate their performance with different fuel types: coal and briquettes. Emissions measurement included EC-OC, PAHs and elements in addition to the commonly measured pollutants (CO, PM). In the present study, overall PM_{2.5} reductions of ~40 % by use of carbonised coal and ~47 % by use

of coal briquettes were achieved. Briquettes, with simple process and reasonable cost, demonstrated a good ability in decreasing PM_{2.5} emissions with potential of 45–65 % (22.69–13.80 t year⁻¹). However, one-way ANOVA analysis indicated that the EFs of raw/ processed coal in different stoves were not significantly different from each other ($p > 0.05$) suggesting the need for more test replicates and further study. While processed fuels have the potential to reach the performance capability of LPG (Champion & Grieshop, 2019), inability to maintain homogeneity could lead to variable emission characteristics (including increase of the 'toxic' fraction of PM mass) which need to be standardised through protocols. Processing the fuel also has several limitations, one being fuel cost and another being the problem of the disposal of fuel processing residues. Availability of analytical techniques suggest the need for going beyond the PM mass emissions as an indicator of better performance. Fuel specifications, risk assessment of toxic component of emissions (Leavey et al., 2017), fuel processing strategy, and stove-fuel as a combination are recommended as parameters for possible inclusion in cookstove testing protocols.

The findings of the present study can serve as a potential pathway for different stakeholders in formulating adequate solid fuel usage standards for domestic needs. This could be extrapolated to solve several relevant problems - crop burning in India (Sharma et al., 2010) using crop-residue as briquettes/pellets, recycling of plastic (Nwabue et al., 2017) and municipal solid waste management problems. Apart from tackling air quality problems due to inefficient burning, the current solution of processing of powdered raw coal (for fuels to be used in appropriate stoves) for domestic cooking needs can also be a waste management solution for coal mines, thermal power plants, and industries which use coal as their fuel. Coal "waste" briquettes using binder clay and additives can be a useful path to pursue for reducing domestic cooking emissions. Further work may be carried out on different compositions of biomass and coal briquettes to find optimum coal to biomass ratio for Indian coal that may further lead to reductions in emissions. Large scale production of processed fuel for domestic application needs could be achieved through trailer mounted briquetting mobile facility run on a co-operative model.

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.

Acknowledgement

The study is partly supported through project funds from Maharashtra Pollution Control Board, Mumbai; Tata Center for Technology and Design, IIT Bombay; CLEAN New Delhi; and Professor Shireesh Kedare, DESE, IIT Bombay for development of the Cookstove Laboratory. The support is gratefully acknowledged. The authors thank National Environmental Engineering Research Institute, Nagpur for providing IC used for the study. Part of this work was carried out at IIT Bombay as a part of M. Tech (Prerit Tak) and Ph.D. (Darpan Das) projects. The authors also thank Professor Satish Agnihotri, Professor Upendra Bhandarkar and Professor Virendra Sethi, IIT Bombay for their scientific inputs regarding the project. Help with chemical analysis from Ms. Pradhi Rajeev, Mr. Gyanesh Singh, Professor Abhas Singh and Professor Debajyoti Paul at IIT Kanpur is gratefully acknowledged. The authors are also thankful to the anonymous reviewers for the time and expertise they have shared to improve the quality of this manuscript.

Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.esd.2022.06.001>.

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