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Original Research

Global brown carbon emissions from combustion sources

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ABSTRACT

Light-absorbing organic carbon (OC), sometimes known as Brown Carbon (BrC), has been recognized as an important fraction of carbonaceous aerosols substantially affecting radiative forcing. This study firstly developed a bottom-up estimate of global primary BrC, and discussed its spatiotemporal distribution and source contributions from 1960 to 2010. The global total primary BrC emission from both natural and anthropogenic sources in 2010 was 7.26 (5.98–8.93 as an interquartile range) Tg, with 43.5% from anthropogenic sources. High primary BrC emissions were in regions such as Africa, South America, South and East Asia with natural sources (wild fires and deforestation) contributing over 70% in the former two regions, while in East Asia, anthropogenic sources, especially residential solid fuel combustion, accounted for over 80% of the regional total BrC emissions. Globally, the historical trend was mainly driven by anthropogenic sources, which increased from 1960 to 1990 and then started to decline. Residential emissions significantly impacted on emissions and temporal trends that varied by region. In South and Southeast Asia, the emissions increased obviously due to population growth and a slow transition from solid fuels to clean modern energies in the residential sector. It is estimated that in primary OC, the global average was about 20% BrC, but this ratio varied from 13% to 47%, depending on sector and region. In areas with high residential solid fuel combustion emissions, the ratio was generally twice the value in other areas. Uncertainties in the work are associated with the concept of BrC and measurement technologies, pointing to the need for more studies on BrC analysis and quantification in both emissions and the air.

1. Introduction

Brown carbon (BrC) is a part of organic carbon (OC) with strongly wavelength-dependent light absorption properties [1–3]. The light absorptivity of BrC increases as the wavelength shortens from the visible to ultraviolet ranges [4,5]. It was estimated that the absorption of BrC at 440 nm was about 40% of the absorption induced by the black carbon (BC), whereas at 675 nm, the proportion was less than 10% [6]. The radiative forcing raised by BrC absorbance was estimated at about +0.25 W m^{-2} (~19% of the absorption by anthropogenic aerosols). One study showed that by including BrC absorption in the model, the global mean radiative forcing at the top of the troposphere would increase from −0.08 W m^{-2} to +0.025 W m^{-2} [7]. Besides the significant impacts on the climate, BrC is sometimes a concern due to its adverse impacts on human health [8–11].

BrC can be formed in both primary fuel combustion processes and secondarily in the atmosphere [12–15]. High BrC loadings in the air were often reported in areas or periods with strong biomass burning influences [7,16]. For example, in areas with major biomass burning and biofuel combustion, the ambient BrC loading generally exceeds 2 mg m^{-2}, with the fraction of BrC in OC around 40%–50%. In southern Africa, the loading was as high as 15–20 mg m^{-2} [7].

Fractions of light-absorption OC in OC are often discussed, however, there are limited estimates currently available. A few studies are from OC (or BC) inventories with an assumed ratio of BrC [17–20], or using measured emission factors (EFs) along with energy activity data [21–24]. Following the former approach, Lin et al. (2014) assumed the primary organic aerosols from biomass

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burning and biofuel combustion was all light absorption in evaluating BrC contribution to radiative forcing. Feng et al. (2013) adopted a fraction of 92% BrC in OC generated from biomass burning and biofuel combustion, and 0% in primary emissions from fossil fuel combustion, resulting in an estimate of about 30.3 Tg in 2000 and a global mean burden of 0.65 mg m^{-2} in the atmosphere [7]. Following the approach using activity data and corresponding EFs, Jo et al. (2016) [20] estimated that globally the primary BrC emissions from biomass burning and biofuel combustion in 2000 were 3.9 ± 1.7 Tg and 3.0 ± 1.3 Tg, respectively, with relatively high emissions in South America, South and East Asia. Sun et al. (2017, 2021) estimated that the primary BrC emissions from residential coal and biomass burning in China were 592 Gg and 712 Gg in 2013, respectively [21,22]. For natural sources, Evangeliou et al. (2019) estimated that the open fire in Greenland between July 31 and August 21, 2017, yielded about 141 tons of BrC [23]. Available studies focused only on one sector like residential burning emissions, while spatiotemporal differences in emissions are rarely evaluated.

Giving the lack of spatiotemporally resolved results from all combustion sources, the current limited estimation of BrC needs to be improved, including the missing sources and more reliable datasets adopted. In this study, based on the fuel-consumption data product (PKU-FUEL), EF databases of BrC, and the BrC/OC ratios for those without directly measured BrC Efs, we first developed a bottom-up country-level global emission inventory of primary BrC. A total of 76 detailed sources in six sectors (Table S1) from 1960 to 2010 were compiled. Temporal and spatial variations of the BrC emissions and the fractions of primary BrC in OC are discussed. Uncertainties in the estimates are addressed. The inventory would be valuable for studies on global or regional radiative forcing modelling and countermeasures on co-controls of both health and climate-relevant compounds.

2. Method

2.1. Emission inventory development

Although there are debates on the concept of BrC [25–27], we think it is reasonable that BrC consists of special compounds whose mass can be estimated or calculated. Several methods are available in the literature on measuring BrC or specific chemical compositions [12,16,22,28–31]. Results from these methods, such as the mass of water- or methanol-soluble carbon, the mass estimated from the light absorption coefficient and mass absorption efficiency, and the mass of several specific chromophores, like *N*-containing compounds and humic-like substance, can vary largely and may not be comparable. The present study adopted a bottom-up approach to estimate BrC, which is from the activity level (i.e., quantities of energy consumed or amounts of materials produced) and the corresponding emission factors (mass of targets per fuel mass or per production). Natural emissions from forest fires and five anthropogenic sectors, including power plants, industry, agriculture, transportation, and residential sources, are considered.

Energy consumption data were mainly comprised of national statistics and the International Energy Agency database [32–36]. For BrC EFs (EF_{BrC}), several studies reported values for combustion sources [5,21,22,37–41]. As noted above, results for these studies, although limited, are sometimes not comparable due to different measurement methods. Measurement of specific chromophores only assesses a small fraction of BrC, thus these Efs are not included in the present estimate. Besides directly measured BrC mass, some studies reported the ratio of BrC to OC in emission exhausts. These ratios were also collected and compiled into a database to estimate EF_{BrC} from OC EFs (Table 1 and details in Table S1). As there are limited data on EF_{BrC}, uncertainties in the EFs would consequently contribute largely to the uncertainty of the emissions inventory.

Historical changes in the BrC emissions from 1960 to 2010 were estimated based on the sector- and fuel-specific energy consumption over time, EF_{BrC}, and impacts of technology changes such as installation of end-of-pipe treatment facilities and upgrading of stoves on the EFs of BrC and/or OC. For the power plants and industry sectors, the EF_{BrC} of coal depends on the combination of boilers (e.g., pulverized coal or stoker) and dust removal equipment (e.g., uncontrolled cyclones, wet scrubber, electrostatic precipitator, or fabric filter). These ratios vary spatiotemporally, as described in Huang et al. (2014) [42,43]. In the residential sector, bituminous coals in China were divided into chunk and honeycomb briquettes with differentiated EFs. While in areas other than China, because of unavailable data, we assumed that the use of honeycomb briquettes was negligible [43]. The ratios of woodstoves, improved woodstoves, and fireplaces burning biomass fuels are from Bond et al. (2004) and Shen et al. (2013a) [44,45].

2.2. Uncertainty analysis

A Monte Carlo simulation was run 10,000 times to characterize the overall uncertainty. The fuel consumption was assumed to be uniformly distributed, with coefficients of variation of 10%, 20%, 30%, 20%, and 30% for power plants, industry, residential, transportation, and open biomass burning, respectively [36]. The log-normally distributed EF_{BrC} can be taken directly from the database (Table S1), and a variation coefficient of 50% was assumed for the penetration rates of dust removal facilities in power plants and industry sectors.

3. Results and discussion

3.1. Global BrC emission in 2010

The global total BrC emission from both natural and anthropogenic sources in 2010 was estimated at 7.26 (5.55–10.2 as an interquartile range) Tg, with approximately 43% from anthropogenic sources (Fig. 1). Among various anthropogenic sources, emissions from the residential sector were predominant at 2.34 (1.76–3.31) Tg, comprising up to ~74% of the total anthropogenic emissions. The residential sector has been recognized as an important global primary OC and BC source [42,46–48]. A high contribution from residential emissions was associated with abundant biomass fuels burned in low-efficiency stoves, and lacking abatement facilities [5,48–50]. BrC from the residential sector was largely burning bituminous coal, firewood, and crop residue, contributing to 26%, 24%, and 32% of residential BrC emissions, respectively. Following the residential sector, the second-largest anthropogenic source of primary BrC was from the agricultural sector. This sector contributed 23% of the anthropogenic BrC, mostly from the open burning of agriculture wastes. Emissions from other anthropogenic sources were minor, accounting for <5% of total anthropogenic BrC emissions on the global average. There were significant spatial differences in BrC emissions and sector contributions.

The spatial distribution of the annual BrC emissions in 2010 is illustrated in Fig. S1. High BrC emissions existed in Africa, South and Southeast Asia, East Asia, and South America, at 2.99 (2.28–4.23) Tg, 1.33 (1.02–1.88) Tg, 1.09 (0.83–1.55) Tg, and 1.01 (0.77–1.43) Tg, respectively. BrC emissions in Africa and South America were dominated by natural emissions. In the Republic of the Congo, Angola, and Brazil, the three largest emission countries, natural emissions accounted for nearly 80%–90% of the total. In Asia, anthropogenic emissions were dominant, and high emissions were

Table 1
Summarized mean ratio of BrC/OC for different fuels from different sectors. Details can be found in Table S1.

Fuel	Sector Power Plants	Industry	Residential	Transportation	Agriculture	Natural
Coal	0.16	0.30	0.50	–	–	–
Oil	0.05	0.05	0.05	0.05	0.05	–
Gas	0.02	0.02	0.02	–	–	–
Waste	0.30	0.30	0.50	–	0.50	–
Biomass	0.30	0.30	0.2/0.3[a]	0.50	–	0.10/0.25[a]

[a] The ratio differed on fuel types.

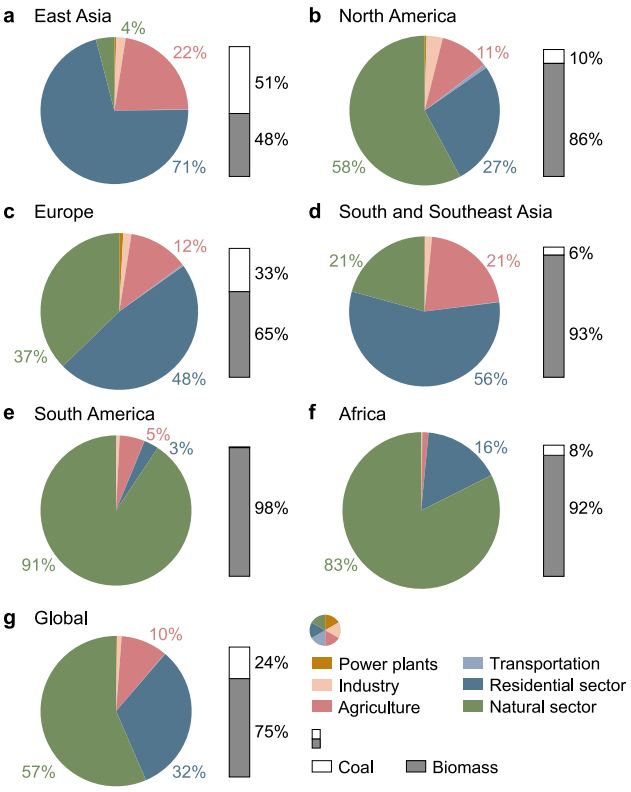

Fig. 1. Relative contributions of different sectors (pie chart) and fuels (bar chart) to the primary BrC emissions globally and in several regions in 2010: **a**, East Asia; **b**, North America; **c**, Europe; **d**, South and Southeast Asia; **e**, South America; **f**, Africa; **g**, Global. (Note: the bar chart only included the percentage of coal and biomass, the other three fuel types—gas, oil, and industry process—are not shown in this figure.)

from the residential sector. In South and East Asia, the residential BrC accounted for over 70% of the total anthropogenic emissions, followed by agricultural emissions (about 25%). But the fuel shares in the residential sector varied across the countries. In China, coal combustions contributed the most to the residential BrC, while in India, indoor biomass burning was the largest contributor to the residential BrC. In Europe and North America, the BrC emissions were 0.34 (0.26–0.48) Tg and 0.21 (0.16–0.30) Tg, respectively, with 37% and 58% from natural sources, while the residential sector was still the largest anthropogenic source.

Fig. 1 also shows the relative shares of different fuels in anthropogenic emissions. Globally, most BrC was from biomass fuels (75%), followed by coals (24%). On the global scale, biomass fuels were consumed in residential (64%), industry (19%), and agricultural (8%) sectors, but BrC emissions from the biomass burning in these three sectors contributed to 68%, 1%, and 31% of the total biomass BrC emissions. This trend is due to the distinct emission factors among these sectors. Coals were largely consumed in the power plants, however, BrC from coal combustion was mostly from the residential sector. This is due to the more efficient burning conditions and complete end-treatment measures in power plants. The other energies contributed less than 2% to the BrC globally. For each region, biomass fuels were the largest contributor, apart from East Asia, where the BrC emissions were largely from coal combustion.

3.2. Historic change trend in BrC emission

Fig. 2 shows historical changes in the global total BrC emissions, which were significantly driven by the anthropogenic emission changes, while natural emissions contributed presumably to random peaks in the total emissions. Generally, anthropogenic emissions increased from 1960 to 1990, reaching an emissions peak of 3.73 Tg year^{-1}, then showing a slowly decreasing trend until 2010. The anthropogenic emissions in 2010 (3.16 Tg year^{-1}) were about 30% higher than that in 1960 (2.40 Tg year^{-1}). The temporal changes were expectedly distinct across different regions (Fig. 3). The change in the emissions trend in East Asia, which contributed to about 30% of the global total, was similar to the global total emissions. Reduced emissions from 1990 were largely associated with the switch to clean energy and stove upgrades in the residential sector, a major source of BrC in East Asia. In South and Southeast Asia, the BrC emissions were found to continuously increase during the study period, and this trend is expected to continue over the next several years. The fast population growth and relatively slow switching to modern household energies lead to continuously high biomass consumption and consequently increased BrC emissions [22,51,52]. Emissions from North America and Europe were declining, with the emissions in 2010 accounting for only one-third of those in 1960. The decreasing trend was largely attributed to less coal usage after more utilization of modern energies in the residential sector.

Historical changes in the total BrC were dominated by the changes in the residential sector, which was the largest anthropogenic source of BrC. Residential BrC emissions in East Asia and European regions have declined obviously since the peak emission in 1990 (Fig. 3). The decline in East Asia was due to less biomass consumption and improved residential stove efficiencies. The residential biomass consumption in 2010 was only half of that in 1990, and residential EFs also lowered by 8% due to stove upgrades. While in Europe, the residential emissions decreased due to the replacement of coal with clean modern energies [53,54]. The residential coal used in Europe in 2010 was only 12% of that in 1960, but residential gas consumption increased by more than 40 times [32,33]. Residential emissions from the South, Southeast Asia, and Africa were continuously increasing with the growth of population and high reliance on biomass fuels. For example, in India, the population in 2010 increased by about 2.6 times compared to that in 1960 [55].

Agricultural emissions, as the second-largest source of BrC,

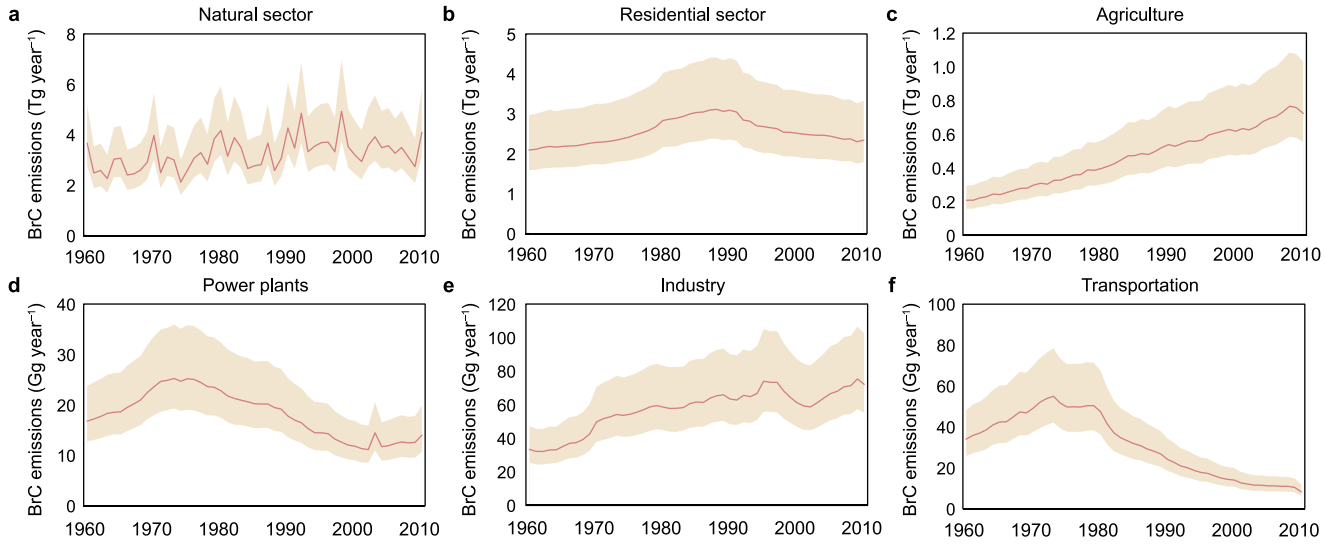

Fig. 2. Temporal changes of global primary BrC emissions in different sectors: **a**, Natural sector; **b**, Residential sector; **c**, Agriculture; **d**, Power plants; **e**, Industry; **f**, Transportation.

generally increased during the study period. Agricultural emissions were mostly concentrated in South Asia and East Asia. Although some countries had policies and controls on open agricultural waste burning in fields [56–58], more yields and consequently high agricultural wastes consumption [59] along with the fast growth of the global population led to a net increase of global BrC emissions from the agricultural sector. Industrial BrC emissions declined since the middle 1990s, mainly due to reduced coal use in coking and brick production, and then the industrial BrC emissions were nearly stable since early 2000. BrC emissions from the power plants and transportation sector both showed declining trends, especially after the 1970s, although their contribution to the global total emissions was minimal. The amount of energy consumed in these two sectors increased by 7.3 and 5.1 times, respectively. However, benefiting from the improved combustion efficiencies, strict emission control standards, and end-of-pipe control facilities [60–64], showed a net decline in BrC emissions from these two sectors by assuming these factors affect BrC similarly to OC.

As mentioned above, biomass burning was the largest emitter of BrC. On the global scale, biomass BrC emissions had a constantly increasing tendency, yet the growth rate reduced since 1990 and maintained relatively stable emissions at about 2.35 Tg year^{-1} (Fig. s2). Before 1990, the increase in biomass BrC emissions was mostly due to increased emissions from East Asia, South Asia, and Southeast Asia. For example, in China and India, biomass BrC emissions nearly doubled from 1960 to 1990. After 1990, as biomass emissions from East Asia declined, increased emissions from South Asia, Southeast Asia, and Africa offset the decline in East Asia,

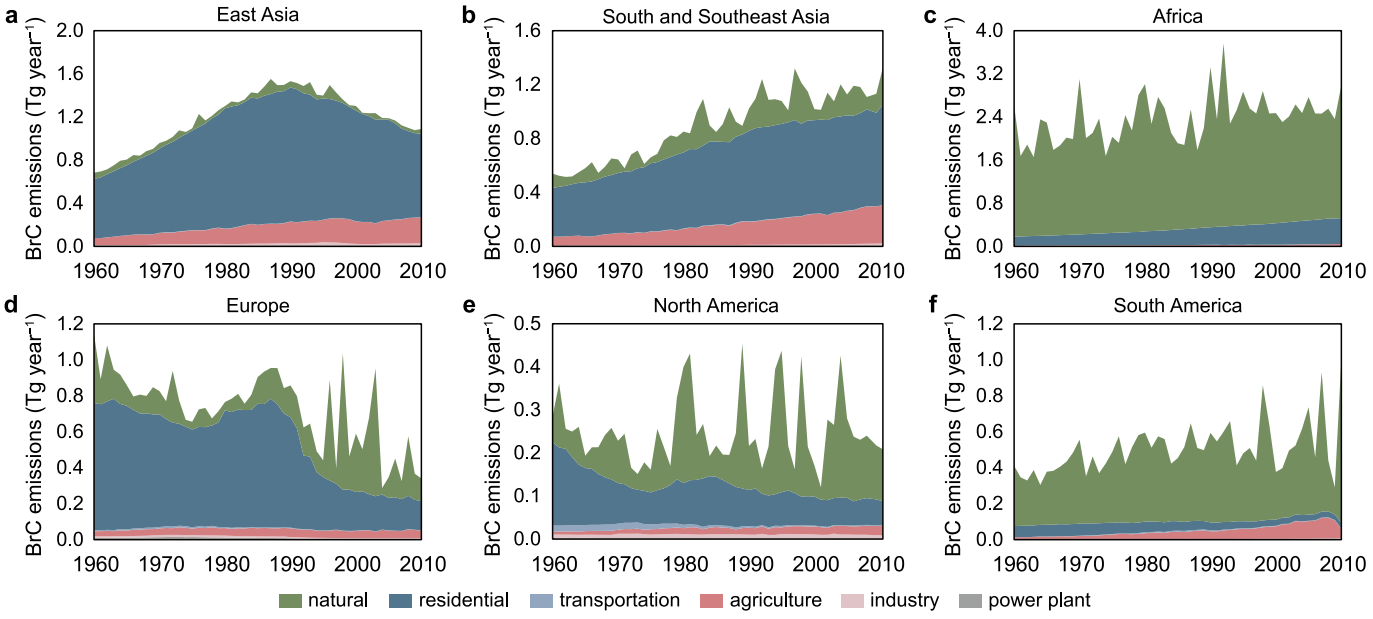

Fig. 3. Temporal changes of continental primary BrC emissions, including natural, residential, transportation, agriculture, industry, and power plants sources in different regions: **a**, East Asia; **b**, South and Southeast Asia; **c**, Africa; **d**, Europe; **e**, North America; **f**, South America.

resulting in nearly steady global biomass BrC emissions. BrC emissions from coal combustion rose slowly until 1990 and then decreased gradually. This tendency was mostly dominated by residential coal combustion emissions. Although coals were largely consumed in power plants, their BrC emissions were smaller compared to residential emissions. Globally in 2010, about 75.3% of the coal was combusted in power plants [32,33], but 95% of the BrC emissions from coal combustion was from the residential sector. Coal BrC emissions from Europe declined significantly since 1990, while in East Asia it also decreased but not as significant as that in Europe. The consumption of other energies (e.g., gas and oil) increased substantially by about five times from 1960 to 2010, while their contribution was rare.

3.3. Spatially resolved fractions of primary BrC in OC

Globally, in primary OC emissions from combustion sources, BrC makes up around 20%, with very slight changes (within −2% to +4%) over the study period from 1960 to 2010. The calculated overall average ratios of BrC in OC (BrC/OC) in the anthropogenic and natural sources were about 29% (±2%) and 16% (±2%), respectively. The BrC/OC expectedly varied significantly among different sectors. The fraction was the highest in the agricultural sector, at ~47% (±1%). This trend was primarily due to higher fractions of BrC emissions in the field burning agriculture wastes. In the residential sector, the fraction of BrC in primary OC was 32% (within ±2% fluctuation). This relatively high ratio was attributed to the dominant emission sources of solid fuels in residential use. The BrC/OC values in emissions from power plants, industry, and transportation sectors were 15% (±2%), 8% (±2%), and 3% (±1%), respectively. These sector differences in the BrC fraction were attributed to distinct fuel shares. In power plants, coal combustion contributed to about 70% BrC leading to higher values of BrC/OC, while in transportation, the BrC/OC ratio was much lower. Resulting from high oil combustion contributions.

The BrC/OC ratio also varied across regions due to different energy structures in each sector (Fig. 4). In areas with higher consumptions of solid coal and biomass fuels in residential and agricultural sectors, the BrC/OC was generally higher. For example, in Asia, where over 97% of the BrC emissions were from the residential sector (e.g., coal and solid biomass) and agricultural burning, the BrC/OC ratio average was ~28% in 2010. This ratio did not significantly change over the study period. In China and India, the BrC/OC ratio values in 2010 were 33% and 27%, respectively, with tiny fluctuations over time. The high proportions were also explained by the high reliance on biomass and coals in the residential sector. It was previously reported that in samples from coal and biomass burning, the BrC/OC ratios could be as high as 53% and 32%, respectively [5,37].

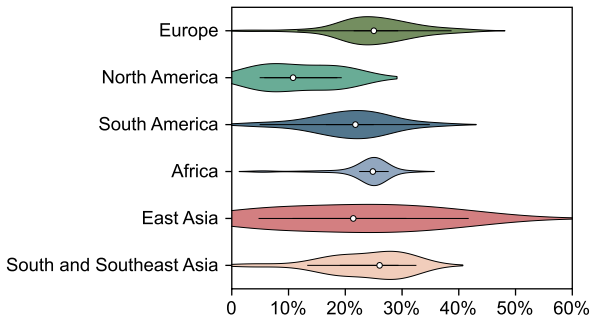

Fig. 4. Distributions of country-level BrC/OC ratios from different regions in 2010.

In Europe, the BrC/OC ratio in emissions from all sources was 18% in 2010, and generally showed a declining trend over time, with fluctuations associated with natural emissions changes. The declining trend was much more obvious if looking into the BrC/OC in emissions from the anthropogenic sources, which was about 36% in 1960 but 28% in 2010. The transition to modern energies from traditional solid fuels in the residential sector led to the obvious decline in the BrC/OC in Europe. For instance, in Germany and Russia, the proportion of gas consumption increased from 3% to 62% and from 1% to 89% during the study period of 1960–2010, whereas the overall average ratio of BrC/OC decreased by 10% and 18%, respectively.

In Africa and South America, the BrC/OC values were around 18% (±1%) and 16% (±2%), respectively, and did not change significantly over time. In these two areas, natural sources comprised largely of the total BrC emissions, and consequently, the ratio of BrC/OC in all source emissions was close to the ratio in the natural source emissions, which did not show significant temporal changes with only about ±1% fluctuation. If looking into emissions from the anthropogenic sources, it contributed much less to the total BrC relative to the natural sources. This observation revealed that the BrC/OC in South America generally increased due to increased emissions from agricultural waste burning.

In North America, the BrC/OC ratio in emissions from all sources was around 13% within −2% to +4% fluctuation over time. Less coal use and higher utilization of clean modern energies resulted in relatively low BrC/OC values in this region. The BrC/OC ratio in the anthropogenic emissions at the beginning was 20% in 1960 and decreased to 13% in 1970. This decrease was mainly due to the increased sharing of clean modern energies in the residential sector. Although there was a continual transition to clean energies in the residential sector, the increased emissions from agricultural waste burning led to a net increase in the BrC/OC from anthropogenic sources in North America that rebounded to 23%. Such temporal trends were observed in both the United States and Canada.

3.4. Discussion, implications, and limitation

With growing concerns about climate impacts, some studies estimated BrC emissions for specific sources (e.g., residential sector and natural sources) in one year. It is believed that anthropogenic BrC is largely produced from burning traditional solid fuels, especially biomass fuels. Based on OC emission inventories in the literature and the calculated mass ratio of BrC to OC, Jo et al. (2016) simulated that globally primary BrC emissions from biomass burning and biofuels were about 3.9 ± 1.7 Tg and 3.0 ± 1.3 Tg in 2000, respectively [20]. Our estimates of BrC from natural sources and agricultural open biomass burning in 2000 was 3.8 (2.88–5.41) Tg, which was close to 3.9 ± 1.7 Tg by considering uncertainties. The BrC emission from biofuels, including firewood and crop residues used as residential energies, was about 1.4 (1.1–2.0) Tg in 2000 in the present study, which was about half of the estimate by Jo et al. (2016). We noted that the BrC/OC ratio in their estimates (0.442 for firewood and 0.652 for crop residue) was nearly twice of ours. To obtain ratio of BrC to OC in estimating BrC, Jo et al. (2016) calculated the ratio of BrC/BC from the modified combustion efficiency and absorption Ångström exponent and then multiplied it by the BC/OC in the literature. In the present study, we first used the directly measured BrC EF, and secondly, the ratio of BrC/OC by fuel type and sector in the literature for those without measured BrC EFs.

There were a few estimates of BrC emissions from natural sources. From typical burn depths assumed and BrC EFs from peat fires, Evangeliou et al. (2019) [23] estimated that the open fire in Greenland from July 31 to August 21 in 2017 yielded ~141 tons of

BrC in the air. Our present country-level estimates cannot be directly compared with this number. Our estimates of BrC from natural sources was 4.1 Tg globally in 2010, of which over 82% was discharged from Africa and South America, which was about 1.3 times than anthropogenic BrC in the same year.

For country-specific emissions, there were several estimates of BrC from the residential sector in China. Cai et al. (2014) estimated that the BrC emissions from residential coal and open agricultural biomass burning were 95.2 ± 73.7 and 175.4 ± 27.8 Gg, respectively, in 2000, based on the BrC/BC ratios calculated from typical source samples and the previously reported BC emissions in that year. The latter was close to the 195.4 (148.8–277.7) Gg in our present study, however, the former was much less than our estimate of 522.5 (397.8–741.0) Gg for residential coal combustion. This difference was mainly due to different BrC EFs, of which ours was about five times higher than those used in this reference. The BrC/BC ratio was the light-absorption ratio in the visible range that ignores the strong absorption of BrC in the ultraviolet range. In addition, the emission of BC in this reference was probably underestimated by assuming a higher ratio of honeycomb briquettes (40% in 2000) used in China. In another estimate of BrC from residential coal combustion by Sun et al. (2017) [21], the result of 592 Gg was close to our estimate of 485 (369–690) Gg, but Sun et al. (2021) [22] estimated a high emission of 712 Gg BrC from indoor biomass burning in 2013. Recently, Zhu et al. (2021) [19] estimated BrC emissions in China based on the BC inventory [65] and reported that in 2018, the national total of BrC was 3.42 Tg, with 71%, 14% and 15% from the residential sector, vehicle emission, and open biomass burning, respectively. Our estimates showed that in 2010, residential emissions contributed to 74% of the national total BrC emissions, which was also close to the proportion estimated by Zhu et al. (2021). However, from the declining trend observed in the present study, we expected that the national total BrC emissions in 2018 would be less than the 1.04 (0.79–1.48) Tg in 2010 and would be much less than the estimate of 3.42 Tg by Zhu et al. (2021). Besides the differences in the EF database, residential fuel consumption data, especially those for non-commercial biomass fuels, affected the emission discrepancies significantly. These differences are primarily due to the transition to clean modern energies, like gas and electricity for cooking over the past several decades. It has been revealed that household biomass consumption declined significantly, which was not accurately captured in some national statistical data [66–68]. Therefore, emission estimates based on the residential energy from the national statistics or database from IEA failed to capture the declining trends and consequently overestimated residential biomass emissions of most products of incomplete combustion.

In our estimates, over 60% of the total global BrC emissions were from Africa and Asia. In Africa, the BrC was largely discharged by natural sources, while in Asia, anthropogenic emissions (i.e., residential solid fuel use) contributed largely to the primary BrC. The recently updated BC emissions inventory showed that Asia also had high BC emissions [46]. The overlap of high BrC and BC emissions potentially suggests high radiative forcing in these regions associated with carbonaceous aerosols. Moreover, in these high emissions regions, the main emission sources of BrC are similar to BC, indicating that there is potential for co-reductions of BC and BrC under effective controls of the common sources. Transitions to clean energy, advanced fuel processing technologies, and upgrading of stoves in the residential sector may effectively reduce the emissions of these light-absorbing forcers. There will be significant co-benefits in response to climate change and protecting human health.

Primary BrC emissions from different areas had distinct temporal trends suggesting that the BrC spatial distribution would be rather different in the future, which consequently affects the radiative forcing and ecosystem changes under climate change. For countries with relatively high BrC emissions, like China, the total BrC emissions already showed an obvious decreasing trend at an average rate of ~1.4% year^{-1} after 1990. This trend is mostly due to the decreased coal consumption in the residential sector. On the other hand, in India, it is noted that the total BrC emissions still had a continuously increasing tendency with a growth rate of ~1.7% year^{-1}, especially from the biomass burning in the residential and agricultural sectors. Thus, banning open agricultural biomass burning and lowering emissions from residential biofuels are needed in India. Most countries in North America and Europe had an overall downward trend, by about 50% from 1960 to 2010, and maintained relatively low emission levels (<60 Gg year^{-1}). Regional or country-specific well-directed countermeasures should be taken to reduce BrC emissions, and other climate forcers (e.g., BC).

The present study, for the first time, estimates country-level primary BrC emissions from 1960 to 2010 and discusses spatio-temporal characteristics and sectorial changes in driving the temporal and spatial trends. Although uncertainty in the estimates was quantitatively assessed using the Monte Carlo simulation, there are relatively high uncertainties in the estimates. Limited data are available for EF_{BrC} that vary by sources and burning conditions. For some sources with little to no direct measurements, EF_{BrC} was calculated from the OC EFs and BrC/OC ratio, but this ratio was kept the same for different regions. This assumption can hardly hold in real life. Secondly, impacts on technology changes on the EFs of BrC were assumed to be the same as OC. These cannot be captured by the quantitative uncertainty ranges for the estimate reported. Last but not least, it is important to note that there are different opinions and debates on the concept of BrC [25–27], and the associated definitions and measurement methods. Some propose that BrC is part of OC formed from primary emissions but also secondarily from atmospheric reactions, and there are interests in characterizing its chemical compositions and molecular-level characteristics. But there are different voices that BrC is not a compound where mass can be estimated, and it is the intrinsic absorptivity of OC whose properties are not fixed by time, space, or sources. This study did not further assess light absorption attributed to BrC. The scientific significance and implication of BrC studies would be much clearer and useful when its contributions to aerosol light absorption and radiative forcing effects can be evaluated. This highly relies on parameters, such as mass absorption efficiencies and absorption Angström exponent, that usually vary largely in source type and burning conditions. It is important but challenging to assess the radiative forcing of BrC and to reduce uncertainties in calculating aerosol light absorption.

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

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References

[1] T.W. Kirchstetter, T. Novakov, P.V. Hobbs, Evidence that the spectral dependence of light absorption by aerosols is affected by organic carbon, J. Geophys. Res. 109 (2004), D21208.

[2] R.K. Chakrabarty, H. Moosmüller, L.-W.A. Chen, K. Lewis, W.P. Arnott, C. Mazzoleni, M.K. Dubey, C.E. Wold, W.M. Hao, S.M. Kreidenweis, Brown carbon in tar balls from smoldering biomass combustion, Atoms. Chem. Phys. 10 (2010) 6363–6370.

[3] D.T.L. Alexander, P.A. Crozier, J.R. Anderson, Brown carbon spheres in East asian outflow and their optical properties, Science 321 (2008) 833–836.

[4] T.W. Kirchstetter, T.L. Thatcher, Contribution of organic carbon to wood smoke particulate matter absorption of solar radiation, Atmos. Chem. Phys. 12 (2012) 6067–6072.

[5] L. Zhang, Z. Luo, Y. Li, Y. Chen, W. Du, G. Li, H. Cheng, G. Shen, S. Tao, Optically measured black and particulate Brown carbon emission factors from real-world residential combustion predominantly affected by fuel differences, Environ. Sci. Technol. 55 (2021) 169–178.

[6] R. Bahadur, P.S. Praveen, Y. Xu, V. Ramanathan, Solar absorption by elemental and brown carbon determined from spectral observations, P. Natl. Acad. Sci. 109 (2012) 17366–17371.

[7] Y. Feng, V. Ramanathan, V.R. Kotamarthi, Brown carbon: a significant atmospheric absorber of solar radiation? Atmos. Chem. Phys. 13 (2013) 8607–8621.

[8] V. Ramanathan, C. Chung, D. Kim, T. Bettge, L. Buja, J.T. Kiehl, W.M. Washington, Q. Fu, D.R. Sikka, M. Wild, Atmospheric brown clouds: impacts on South Asian climate and hydrological cycle, P. Natl. Acad. Sci. 102 (2005) 5326–5333.

[9] J.R. Stinziano, D.A. Way, Autumn photosynthetic decline and growth cessation in seedlings of white spruce are decoupled under warming and photoperiod manipulations, Plant. Cell. Envieon. 40 (2017) 1296–1316.

[10] A. Arola, G. Schuster, G. Myhre, S. Kazadzis, S. Dey, S.N. Tripathi, Inferring absorption organic carbon content from AERONET data, Atmos. Chem. Phys. 11 (2011) 215–225.

[11] T.C. Bond, S.J. Doherty, D.W. Fahey, P.M. Forster, T. Berntsen, B.J. DeAngelo, M.G. Flanner, S. Ghan, B. Kärcher, D. Koch, S. Kinne, Y. Kondo, P.K. Quinn, M.C. Sarofim, M.G. Schultz, M. Schulz, C. Venkataraman, H. Zhang, S. Zhang, N. Bellouin, S.K. Guttikunda, P.K. Hopke, M.Z. Jacobson, J.W. Kaiser, Z. Klimont, U. Lohmann, J.P. Schwarz, D. Shindell, T. Storelvmo, S.G. Warren, C.S. Zender, Bounding the role of black carbon in the climate system: a scientific assessment, J. Geophys. Res. Atmos. 118 (2013) 5380–5552.

[12] K.M. Updyke, T.B. Nguyen, S.A. Nizkorodov, Formation of brown carbon via reactions of ammonia with secondary organic aerosols from biogenic and anthropogenic precursors, Atmos. Environ. 63 (2012) 22–31.

[13] Q. Wang, Y. Han, J. Ye, S. Liu, S. Pongpiachan, N. Zhang, Y. Han, J. Tian, C. Wu, X. Long, Q. Zhang, W. Zhang, Z. Zhao, J. Cao, High contribution of secondary Brown carbon to aerosol light absorption in the southeastern margin of Tibetan plateau, Geophys. Res. Lett. 46 (2019) 4962–4970.

[14] X. Zhang, Y. Lin, J.D. Surratt, P. Zotter, A.S.H. Prevot, R.J. Weber, Light-absorbing soluble organic aerosol in Los Angeles and Atlanta: a contrast in secondary organic aerosol, Geophys. Res. Lett. 38 (2011), L21810.

[15] J.M. Flores, R.A. Washenfelder, G. Adler, H.J. Lee, L. Segev, J. Laskin, A. Laskin, S.A. Nizkorodov, S.S. Brown, Y. Rudich, Complex refractive indices in the near-ultraviolet spectral region of biogenic secondary organic aerosol aged with ammonia, Phys. Chem. Chem. Phys. 16 (2014) 10629–10642.

[16] R. Huang, L. Yang, J. Cao, Y. Chen, Q. Chen, Y. Li, J. Duan, C. Zhu, W. Dai, K. Wang, C. Lin, H. Ni, J.C. Corbin, Y. Wu, R. Zhang, X. Tie, T. Hoffmann, C. O'Dowd, U. Dusek, Brown carbon aerosol in urban Xi'an, Northwest China: the composition and light absorption properties, Environ. Sci. Technol. 52 (2018) 6825–6833.

[17] G. Lin, J.E. Penner, M.G. Flanner, S. Sillman, L. Xu, C. Zhou, Radiative forcing of organic aerosol in the atmosphere and on snow: effects of SOA and brown carbon, J. Geophys. Res. Atmos. 119 (2014) 7453–7476.

[18] R.J. Park, M.J. Kim, J.I. Jeong, D. Youn, S. Kim, A contribution of brown carbon aerosol to the aerosol light absorption and its radiative forcing in East Asia, Atmos. Environ. 44 (2010) 1414–1421.

[19] Y. Zhu, Q. Wang, X. Yang, N. Yang, X. Wang, Modeling investigation of Brown carbon aerosol and its light absorption in China, Atmosphere 12 (2021) 892.

[20] D.S. Jo, R.J. Park, S. Lee, S.W. Kim, X. Zhang, A global simulation of brown carbon: implications for photochemistry and direct radiative effect, Atmos. Chem. Phys. 16 (2016) 3413–3432.

[21] J. Sun, G. Zhi, R. Hitzenberger, Y. Chen, C. Tian, Y. Zhang, Y. Feng, M. Cheng, Y. Zhang, J. Cai, F. Chen, Y. Qiu, Z. Jiang, J. Li, G. Zhang, Y. Mo, Emission factors and light absorption properties of brown carbon from household coal combustion in China, Atmos. Chem. Phys. 17 (2017) 4769–4780.

[22] J. Sun, Y. Zhang, G. Zhi, R. Hitzenberger, W. Jin, Y. Chen, L. Wang, C. Tian, Z. Li, R. Chen, W. Xiao, Y. Cheng, W. Yang, L. Yao, Y. Cao, D. Huang, Y. Qiu, J. Xu, X. Xia, X. Yang, X. Zhang, Z. Zong, Y. Song, C. Wu, Brown carbon's emission factors and optical characteristics in household biomass burning: developing a novel algorithm for estimating the contribution of brown carbon, Atmos. Chem. Phys. 21 (2021) 2329–2341.

[23] N. Evangeliou, A. Kylling, S. Eckhardt, V. Myroniuk, K. Stebel, R. Paugam, S. Zibtsev, A. Stohl, Open fire in Greenland in summer 2017: transport, deposition and radiative effects of BC, OC and BrC emissions, Atmos. Chem. Phys. 19 (2019) 1393–1411.

[24] R.K. Chakrabarty, S. Pervez, J.C. Chow, J.G. Watson, S. Dewangan, J. Robles, G. Tian, Funeral pyres in South Asia: Brown carbon aerosol emissions and climate impacts, Environ. Sci. Technol. Lett. 1 (2014) 44–48.

[25] J. Yan, X. Wang, P. Gong, C. Wang, Z. Cong, Review of brown carbon aerosols: recent progress and perspectives, Sci. Total Environ. 634 (2018) 1475–1485.

[26] H. Moosmüller, R.K. Chakrabarty, K.M. Ehlers, W.P. Arnott, Absorption Ångström coefficient, brown carbon, and aerosols: basic concepts, bulk matter, and spherical particles, Atmos. Chem. Phys. 11 (2011) 1217–1225.

[27] M.O. Andreae, A. Gelencsér, Black carbon or brown carbon? The nature of light-absorbing carbonaceous aerosols, Atmos. Chem. Phys. 6 (2006) 3131–3148.

[28] W. Marrero-Ortiz, M. Hu, Z. Du, Y. Ji, Y. Wang, S. Guo, Y. Lin, M. Gomez-Hermandez, J. Peng, Y. Li, J. Secrest, M.L. Zamora, Y. Wang, T. An, R. Zhang, Formation and optical properties of Brown carbon from small α-dicarbonyls and amines, Environ. Sci. Technol. 53 (2019) 117–126.

[29] T.B. Nguyen, A. Laskin, J. Laskin, S.A. Nizkorodov, Brown carbon formation from ketoaldehydes of biogenic monoterpenes, Faraday Discuss 165 (2013) 473–494.

[30] R. Saleh, E.S. Robinson, D.S. Tkacik, A.T. Ahern, S. Liu, A.C. Aiken, R.C. Sullivan, A.A. Presto, M.K. Dubey, R.J. Yokelson, N.M. Donahue, A.L. Robinson, Brownness of organics in aerosols from biomass burning linked to their black carbon content, Nat. Geosci. 7 (2014) 647–650.

[31] L.T. Fleming, P. Lin, J.M. Roberts, V. Selimovic, R. Yokelson, J. Laskin, A. Laskin, S.A. Nizkorodov, Molecular composition and photochemical lifetimes of brown carbon chromophores in biomass burning organic aerosols, Atmos. Chem. Phys. 20 (2020) 1105–1129.

[32] International Energy Agency (IEA), Energy Statistics and Balances of OECD Countries, 2007–2008. Paris, 2010a.

[33] International Energy Agency (IEA), Energy Statistics and Balances of Non-OECD Countries, 2007–2008. Paris, 2010b.

[34] United States Geological Survey (USGS), Aluminum Statistics and Information 2007, 2010 available at: http://minerals.usgs.gov/minerals/pubs/commodity/aluminum/.

[35] UN Industrial Development Organization (UNID), International Yearbook of Industrial Statistics 2008, Edward Elgar Publishing,Inc., Cheltenham, Glos GL50 1UA, UK, 2008.

[36] H. Shen, Y. Huang, R. Wang, D. Zhu, W. Li, G. Shen, B. Wang, Y. Zhang, Y. Chen, Y. Lu, H. Chen, T. Li, K. Sun, B. Li, W. Liu, J. Liu, S. Tao, Global atmospheric emissions of polycyclic aromatic hydrocarbons from 1960 to 2008 and future predictions, Environ. Sci. Technol. 47 (2013) 6415–6424.

[37] Z. Luo, L. Zhang, G. Li, W. Du, Y. Chen, H. Cheng, S. Tao, G. Shen, Evaluating co-emissions into indoor and outdoor air of EC, OC, and BC from in-home biomass burning, Atmos. Res. 248 (2021), 105247.

[38] C.E. Stockwell, T.J. Christian, J.D. Goetz, T. Jayarathne, P.V. Bhave, P.S. Praveen, S. Adhikari, R. Maharjan, P.F. DeCarlo, E.A. Stone, E. Saikawa, D.R. Blake, I.J. Simpson, R.J. Yokelson, A.K. Panday, Nepal Ambient Monitoring and Source Testing Experiment (NAMaSTE): emissions of trace gases and light-absorbinng carbon from wood and dung cooking fires, garbage and crop residue burning, brick kilns, and other sources, Atmos. Chem. Phys. 16 (2016) 11043–11081.

[39] J. Aurell, B.K. Gullett, Emission factors from aerial and ground measurements of field and laboratory forest burns in the southeastern U.S.: PM2.5, black and Brown carbon, VOC, and PCDD/PCDF, Environ. Sci. Technol. 47 (2013) 8443–8452.

[40] J. Aurell, B.K. Gullett, D. Tabor, Emissions from southeastern U.S. Grasslands and pine savannas: comparison of aerial and ground field measurements with laboratory burns, Atmos. Environ. 111 (2015) 170–178.

[41] X. Fan, T. Cao, X. Yu, J. Song, Y. Wang, X. Xiao, Y. Xie, F. Li, Emission characteristics and optical properties of extractable brown carbon from residential wood combustion, China Environ. Sci. 39 (2019) 3215–3224.

[42] Y. Huang, H. Shen, Y. Chen, Q. Zhong, H. Chen, R. Wang, G. Shen, J. Liu, B. Li, S. Tao, Global organic carbon emissions from primary sources from 1960 to 2009, Atmos. Environ. 122 (2015) 505–512.

[43] Y. Huang, H. Shen, H. Chen, R. Wang, Y. Zhang, S. Su, Y. Chen, N. Lin, S. Zhuo, Q. Zhong, X. Wang, J. Liu, B. Li, W. Liu, S. Tao, Quantification of global primary emissions of $PM_{2.5}$, PM_{10}, and TSP from combustion and industrial process sources, Environ. Sci. Technol. 48 (2014) 13834–13843.

[44] T.C. Bond, D.G. Streets, K.F. Yarber, S.M. Nelson, J.H. Woo, Z. Klimont, A technology-based global inventory of black and organic carbon emissions from combustion, J. Geophys. Res. Atmos. 109 (2004), D14203.

[45] G. Shen, S. Tao, S. Wei, Y. Chen, Y. Zhang, H. Shen, Y. Huang, D. Zhu, C. Yuan, H. Wang, Y. Wang, L. Pei, Y. Liao, Y. Duan, B. Wang, R. Wang, Y. Lv, W. Li, X. Wang, X. Zheng, Field measurement of emission factors of PM, EC, OC, parent, nitro-, and oxy- polycyclic aromatic hydrocarbons for residential briquette, coal cake, and wood in rural shanxi, China, Environ. Sci. Technol. 47 (2013) 2998–3005.

[46] H. Xu, Y. Ren, W. Zhang, W. Meng, X. Yun, X. Yu, J. Lin, Y. Zhang, G. Shen, J. Ma, B. Li, H. Cheng, X. Wang, Y. Wan, S. Tao, Updated global black carbon emissions from 1960 to 2017: improvements, trends, and drivers, Environ. Sci. Technol. 55 (2021) 7869–7879.

[47] Q. Lu, Q. Zhang, D.G. Streets, Sulfur dioxide and primary carbonaceous aerosol emissions in China and India, 1996-2010, Atmos. Chem. Phys. 11 (2011) 9839–9864.

[48] E.D. Vicente, C.A. Alves, An overview of particulate emissions from residential

biomass combustion, Atmos. Res. 199 (2018) 159–185.
[49] H. Shen, Z. Luo, R. Xiong, X. Liu, L. Zhang, Y. Li, W. Du, Y. Chen, H. Cheng, G. Shen, S. Tao, A critical review of pollutant emission factors from fuel combustion in home stoves, Environ. Int. 157 (2021), 106841.
[50] C. Yang, M. Zheng, A.P. Sullivan, C. Bosch, Y. Desyaterik, A. Andersson, X. Li, X. Guo, T. Zhou, O. Gustafsson, J. Collett, Chemical characteristics and light-absorbing property of water-soluble organic carbon in Beijing: biomass burning contributions, Atmos. Environ. 121 (2015) 4–12.
[51] S. Ramachandran, M. Rupakheti, M.G. Lawrence, Black carbon dominates the aerosol absorption over the Indo-Gangetic Plain and the Himalayan foothills, Environ. Int. 142 (2020), 105814.
[52] G. Gautam, N.C. Hsu, M. Kafatos, S.C. Tsay, Influences of winter haze on fog/low cloud over the Indo-Gangetic plains, J. Geophys. Res. Atmos. 112 (2007), D05207.
[53] A.N. Menegaki, Growth and renewable energy in Europe: a random effect model with evidence for neutrality hypothesis, Energy Econ. 33 (2011) 257–263.
[54] D. Connolly, H. Lund, B.V. Mathiesen, Smart Energy Europe: the technical and economic impact of one potential 100% renewable energy scenario for the European Union, Renew. Sustain. Energy Rev. 60 (2016) 1634–1653.
[55] Registrar general & census commissioner, India. Census of India. http://censusindia.net/. (Accessed November 2021).
[56] S. Bhuvaneshwari, H. Hettiarachchi, J.N. Meegoda, Crop residue burning in India: policy challenges and potential solutions, J. Environ. Res. Public Health 16 (2019) 832.
[57] The Law of the People's Republic of China on the Prevention and Control of Atmospheric Pollution.
[58] A.M. Duque, U.L.J. Belmonte, G.F.J. Cortes, F.F. Camacho, Agricultural waste: review of the evolution, approaches and perspectives on alternative uses, Glo. Eco. Conser. 22 (2020), E00902.
[59] Food and Agriculture Organization of the United Nations (FAO), FAOSTAT food and agriculture statistics. http://faostat.fao.org/default.aspx. (Accessed 8 May 2012).
[60] S. Wang, B. Zhao, S. Cai, Z. Klimont, C.P. Nielsen, T. Morikawa, J. Woo, Y. Kim, X. Fu, J. Xu, J. Hao, K. He, Emission trends and mitigation options for air pollutants in East Asia, Atmos. Chem. Phys. 14 (2014) 6571–6603.
[61] Q. Wang, B. Su, J. Sun, P. Zhou, D. Zhou, Measurement and decomposition of energy-saving and emissions reduction performance in Chinese cites, Appl. Energy 151 (2015) 85–92.
[62] Y. Wu, S. Zhang, J. Hao, H. Liu, X. Wu, J. Hu, M.P. Walsh, T.J. Wallington, K. Zhang, S. Stevanovic, On-road vehicle emissions and their control in China: a review and outlook, Sci. Total Environ. 574 (2017) 332–349.
[63] I.A. Resitoglu, K. Altinisik, A. Keskin, The pollutant emissions from diesel-engine vehicles and exhaust aftertreatment systems, Clean Technol. Environ. Policy 17 (2015) 15–27.
[64] A.Y. Saber, G.K. Venayagamoorthy, Plug-in vehicles and renewable energy sources for cost and emission reductions, IEEE Trans. Ind. Electron. 58 (2011) 1229–1238.
[65] R. Wang, S. Tao, W. Wang, J. Liu, H. Shen, G. Shen, B. Wang, X. Liu, W. Li, Y. Huang, Y. Zhang, Y. Lu, H. Chen, Y. Chen, C. Wang, D. Zhu, X. Wang, B. Li, W. Liu, J. Ma, Black carbon emissions in China from 1949 to 2050, Environ. Sci. Technol. 46 (2012) 7595–7603.
[66] S. Tao, M. Ru, W. Du, X. Zhu, Q. Zhong, B. Li, G. Shen, X. Pan, W. Meng, Y. Chen, H. Shen, N. Lin, S. Su, S. Zhou, T. Huang, Y. Xu, X. Yun, J. Liu, X. Wang, W. Liu, H. Cheng, D. Zhu, Quantifying the rural residential energy transition in China from 1992 to 2012 through a representative national survey, Nat. Energy 3 (2018) 567–573.
[67] X. Zhu, X. Yun, W. Meng, H. Xu, W. Du, G. Shen, H. Cheng, J. Ma, S. Tao, Stacked use and transition trends of rural household energy in mainland China, Environ. Sci. Technol. 53 (2019) 521–529.
[68] G. Shen, R. Xiong, Y. Tian, Z. Luo, B. Jiangtulu, W. Meng, W. Du, J. Meng, Y. Chen, B. Xue, B. Wang, Y. Duan, J. Duo, F. Fan, L. Huang, T. Ju, F. Liu, S. Li, X. Liu, Y. Li, Wangmo, Y. Nan, B. Pan, Y. Pan, L. Wang, E. Zeng, C. Zhan, Y. Chen, H. Shen, H. Cheng, S. Tao, Substantial transition to clean household energy mix in rural households in China, Natl. Sci. Rev. (2022) nwac050.