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TOPICAL REVIEW

Review of black carbon emission factors from different anthropogenic sources

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Abstract

Particulate black carbon (BC) affects global warming by absorbing the solar radiation, by affecting cloud formation, and by decreasing ground albedo when deposited to snow or ice. BC has also a wide variety of adverse effects on human population health. In this article we reviewed the BC emission factors (EFs) of major anthropogenic sources, i.e. traffic (incl. marine and aviation), residential combustion, and energy production. We included BC EFs measured directly from individual sources and EFs derived from ambient measurements. Each source category was divided into sub-categories to find and demonstrate systematical trends, such as the potential influence of fuel, combustion technologies, and exhaust/flue gas cleaning systems on BC EFs. Our review highlights the importance of society level emission regulation in BC emission mitigation; a clear BC emission reduction was observed in ambient studies for road traffic as well as in direct emission measurements of diesel-powered individual vehicles. However, the BC emissions of gasoline vehicles were observed to be higher for vehicles with direct fuel injection techniques (gasoline direct injection) than for vehicles with port-fueled injection, indicating potentially negative trend in gasoline vehicle fleet BC EFs. In the case of shipping, a relatively clear correlation was seen between the engine size and BC EFs so that the fuel specific BC EFs of the largest engines were the lowest. Regarding the BC EFs from residential combustion, we observed large variation in EFs, indicating that fuel type and quality as well as combustion appliances significantly influence BC EFs. The largest data gaps were in EFs of large-scale energy production which can be seen crucial for estimating global radiative forcing potential of anthropogenic BC emissions. In addition, much more research is needed to improve global coverage of BC EFs. Furthermore, the use of existing data is complicated by different EF calculation methods, different units used in reporting and by variation of results due to different experimental setups and BC measurement methods. In general, the conducted review of BC EFs is seen to significantly improve the accuracy of future emission inventories and the evaluations of the climate, air quality, and health impacts of anthropogenic BC emissions.

1. Introduction

Black carbon (BC) formed in in-complete combustion is the most important light absorbing particulate component in the atmosphere in respect of global warming (Bond *et al* 2013, IPCC 2014). In addition to absorption of solar radiation and influencing cloud formation, BC can deposit on snow and ice surfaces leading to reduction of ground's albedo, especially in Arctic areas (e.g. Arctic Council 2013, Bond *et al* 2013, IPCC 2014). Especially in urban areas, BC affects public health (Janssen *et al* 2011, 2012, Steiner et al 2016, Chowdhury et al 2022), due to the small size of BC particles and the compounds condensed on their surfaces (e.g. Hakkarainen et al 2022). Prevailing BC sources are typically anthropogenic combustion processes, such as transportation, industry, and residential combustion (Bond et al 2013, Helin et al 2018, Mylläri et al 2019). Due to the importance of BC in research regarding global warming and the potential to affect climate by BC emission mitigation, it has been taken into the policy arenas (Arctic Council 2013). BC emissions are indirectly regulated on some level; for instance, in Europe, the BC emissions of modern diesel and gasoline vehicles have been regulated by tight limits set for the number of solid exhaust particles. These kinds of regulations exist and will come into use also in other applications and geographical areas. To increase knowledge related to BC concentrations in urban areas and to address concerns about the health and environmental effects of BC, WHO has recommended in their recently updated global air quality guidelines to start systematical BC (or elemental carbon (EC)) measurements, create BC inventories and start BC mitigation action where relevant (WHO 2021). In addition, currently the International Maritime Organization (IMO) considers setting direct BC emission limits for marine traffic. These BC emission mitigation actions are expected to decrease the BC emission factors (EFs) and, as a long-term influence, atmospheric concentrations of BC (Ahmed et al 2014, Kanaya et al 2020, Luoma et al 2021).

Although the BC measurement technologies have significantly developed lately due to application of novel and improved measurement and data analysis methods (e.g. Schwarz et al 2008, 2012, Carbone et al 2015, Drinovec et al 2015, Caubel et al 2019), uniform metrics do not exist to evaluate BC emissions, concentrations, and effects. BC measurement instruments are based on indirect methods, in atmospheric sciences typically on thermal and optical determination of BC concentration and in emission studies on optical and photo-acoustic methods and size distribution measurements (Birch and Gary 1996, Harris and Maricq 2001, Petzold and Schönlinner 2004, Schindler et al 2004). These issues, together with the challenges in BC instrument calibration methods (Baumgardner et al 2012), significantly complicate BC emission measurements, affect the reliability of existing BC emission information, and decrease the comparability of BC emission data produced by different investigations. Furthermore, these challenges also affect the policy and air quality actions related to BC and estimating effects of BC on global climate and human health.

BC emissions have been studied experimentally both in laboratory and field conditions. While the laboratory studies have typically produced detailed source-specific emission information and EFs (e.g. Giechaskiel et al 2010, Karavalakis et al 2014, Timonen et al 2017, da Silva et al 2018, Yang et al 2019, Aakko-Saksa et al 2021), the field studies have included measurements using portable emission measurement systems (PEMS) (e.g. Zheng et al 2015, 2021), mobile measurements (e.g. Pirjola et al 2016, Wren et al 2018, Järvinen et al 2019, Chambliss et al 2020, Lepistö et al 2022) and next-to-source experiments (e.g. Dallmann et al 2013, Liu et al 2021, Saarikoski et al 2021). Results of many field studies do not represent a single BC source, but e.g. the whole traffic fleet. It is evident that the EFs determined by different methods and in different research environments may not be fully comparable and their utilization e.g. in emission inventories and climate and air quality models requires careful and critical evaluation.

In addition to diversity of experimental methods, the utilization of existing BC emission information is challenged by the reporting practices of the BC EFs; the EFs are presented in variable units in different studies, depending mostly on emission source categories. For mobile sources such as passenger cars, trucks, buses and aircrafts, the EFs are typically reported in mg km^{-1}, mg kg^{-1} fuel or mg kWh^{-1} (Oanh et al 2010, Aakko-Saksa et al 2016, Durdina et al 2017, Kinsey et al 2019), and for stationary sources in mg MJ^{-1} or mg kg^{-1} fuel (e.g. Fachinger et al 2017, Mylläri et al 2019). In addition, some studies have reported the BC emissions relative to CO_2 or NO_x emissions (e.g. Krecl et al 2017, Martinet et al 2019). The conversion of units can significantly increase the uncertainty of results when the reported values are used in larger context, e.g. in air quality models.

BC EFs are widely used in emission inventories describing the temporal variation and trends related to BC emissions from various sources. Emission inventories and scenarios can affect the society level emission mitigation targets (e.g. Savolahti et al 2016, Harmsen et al 2020). BC EFs are needed also in climate models used in investigations to describe the radiative forcing of atmospheric particulate matter (e.g. Wittbom et al 2014, Hienola et al 2016, Sumlin et al 2017). Furthermore, BC is increasingly the part of air quality modeling which is needed to evaluate the local and regional differences in air quality and health effects of air pollution (e.g. Lugon et al 2021). Thus, it is utmost important that the EFs used in emission inventories, models, and other evaluations, are accurate and the experimental data are representative.

The aim of this review article is to collect the newest information from peer-reviewed scientific literature on the BC EFs of major anthropogenic BC sources, i.e. traffic, residential combustion, and energy production. The review was limited to measurements of BC or EC that have been published in peer-reviewed scientific literature. We included EFs measured directly from individual sources and EFs derived from ambient measurements. Each source category was divided into sub-categories to find and demonstrate systematical trends, such as influence of fuel, combustion technologies and exhaust/flue gas cleaning systems on BC EFs. Resulted comprehensive BC EF compilation can be used when new emission regulations are planned, new emission inventories are made and when estimating radiative forcing effects as well as health and air quality impacts of anthropogenic BC emissions.

2. Methods

2.1. Articles

Because the BC emissions are emitted to atmosphere from various sources and they are utilized e.g. in emission inventories separately for several emissions source categories, we conducted the literature search separately for following categories: individual vehicles, marine engines, aircrafts, residential combustion, and power and heat generation done in larger units. In addition, we included in the review the studies utilizing BC measurements of ambient air close to the original BC source and where the EFs were determined from the obtained data. While the emission measurements directly from source provide EFs for a certain selected single source, frequently operated in controlled conditions, the BC EFs determined based on ambient BC measurements are affected by several individual sources and background concentrations, being thus more prone for interpretations. However, in many ambient studies the experiment was conducted near the source, typically close to traffic, and the EFs have been determined by well-defined and liable methods. In general, the separate searches for each category were done by using different keywords, as described in table 1. We decided to limit the review to peer-reviewed scientific articles, and the searches were done from peer-reviewed literature database Scopus. In addition, we included such scientific articles to the material that we were aware of or became aware of during the review.

The resulted articles were included in the further analyses if (a) the articles were written in English, (b) they included experimentally determined EFs for BC or EC, and (c) the articles included sufficient description of methods used in emission factor determination. We excluded articles that evaluated the BC EFs indirectly e.g. from particulate matter (PM) measurements without any direct BC emission information as well as articles which used algorithms-based conversions from smoke number or other parameters such as particle number and geometric mean diameter.

The included studies contained variety of different emission factor units. Regarding ambient studies, the BC EFs were found typically in units mg $\mathrm{kg_{fuel}}^{-1}$ or mg km^{-1}. Regarding the figures of this review, we converted all these values to use units of mg $\mathrm{kg_{fuel}}^{-1}$ by using for fleet and light-duty a fuel consumption of 0.1 l km^{-1} and a fuel density of 0.74 g l^{-1} by making a rough assumption that fleet and light duty consist mostly of gasoline vehicles. For heavy duty (HD) traffic we used a fuel consumption of 0.5 l km^{-1} and a fuel density of 0.84 g l^{-1} by assuming that most of the HD consists of diesel vehicles. All the BC EF values from ambient studies are presented in table S1 in supplemental material in the units given in original publications. Regarding the BC EF studies of individual passenger cars, L-class vehicles and HD vehicles, most of the original BC EFs were in mg km^{-1} but, however, the units of mg mile^{-1} and mg kg^{-1} fuel were also used. The BC EFs from residential combustion were typically given in units mg MJ^{-1} or mg $\mathrm{kg_{fuel}}^{-1}$. Due to the difficulties in estimation of heat values of highly variable fuels, the figures and tables of this review present the residential combustion EFs values in original units. For marine engine BC EF studies, selected articles were limited to ones containing information needed to present BC EFs in mg $\mathrm{MJ_{fuel}}^{-1}$. For aviation, the BC EFs are presented in mg $\mathrm{kg_{fuel}}^{-1}$, both in the original articles and in this review.

3. Results and discussion

BC EFs for each source category as well as related supporting parameters (information on fuel, combustion technology, exhaust cleaning technology, emission standards) are presented in the following chapters. In addition, the measurement and calculation methods of BC EFs are described below. The analysis is divided so that the BC EFs determined from ambient measurements are analyzed first and then the BC EFs determined in source-specific investigations, separately for each source category, i.e. road traffic, marine traffic, aviation, residential combustion and heat and power generation.

3.1. BC EFs measured in ambient air

BC EFs measured in ambient air comprise dominantly EFs measured at stationary sites at roadside, above the road in a bridge or in tunnels. Additionally, BC EFs were measured with mobile equipment while driving within the traffic. This chapter includes only the BC EFs that have been calculated for the total fleet or/and heavy and light duty vehicle traffic separately in the original papers. Coarse assumptions are made; BC EFs for diesel fleet are considered as HD and BC EFs for gasoline fleet are incorporated into the light duty. EFs for individual vehicles are discussed later. In addition to road traffic, some BC EFs for trains and airplanes have been extracted from ambient data. The measurement types, locations and instruments are given in table S1 in supplemental material.

3.1.1. Measurement techniques and calculation methods for BC EFs

BC concentrations for the determination of the BC EFs in ambient air were measured with various instruments and techniques. Widely used instrument was an aethalometer (AE, Drinovec *et al* 2015),

Table 1. Used search terms, number of found articles and corresponding data tables for different BC sources. Additionally, citing information was used to search relevant publications. Scopus search tool (www.scopus.com) was used.

Category	Search terms	Number of chosen articles	Data table
Ambient	Emission factor AND black carbon, Emission factor AND EC, emission factor AND on-road	60	Table S1
Passenger cars	Passenger car OR light-duty vehicle OR diesel OR gasoline AND black carbon AND elemental carbon AND exhaust particles	17	Table S2
Heavy-duty vehicles	Heavy AND duty AND black carbon OR pems OR dynamometer OR on-road AND emission factor OR black carbon. Another search was made by substituting heavy with medium.	8	Table S3
L-class vehicles	Moped OR motorbike OR motorcycle OR auto-rickshaw AND black AND carbon AND aerosol	2	Table S4
Marine engines	Ship AND black carbon, ship AND BC, ship AND elemental carbon, marine engine AND black carbon, marine engine AND BC, marine engine AND elemental carbon.	13	Table S5
Aviation	Black carbon AND aviation	19	Table S6
Residential combustion	Residential combustion AND black carbon AND emission factor and wood AND combustion	27	Tables S7 and S8
Power and heat generation	Black carbon OR elemental carbon AND emission factor AND power plant AND measurement	2	Table S9

however, the various models of AEs (AE1, AE2, AE9, AE10, AE16, AE22, AE31, AE33, AE42, AE51) were deployed for the measurements. Another optical, filter-based instrument was multi-angle absorption photometer (MAAP, Petzold and Schönlinner 2004) that was used for example in Mexico City measurements (Thornhill *et al* 2010). In AE, the wavelength used for BC is typically 880 nm whereas in the MAAP it is 660 nm. Krecl *et al* (2017) utilized also custom-built particle soot absorption photometer at 525 nm. A photoacoustic soot spectrometer (PASS-3) was also deployed in Toronto (Wang *et al* 2015) for the particle absorption at 781 nm. Photoacoustic extinctiometer that utilized photoacoustic absorption at 870 nm was utilized in California, US (Haugen and Bishop 2018). Liggio *et al* (2012) used high-sensitivity laser-induced incandescence (LII) instrument and single particle soot photometer (SP2) to determine BC EF in Toronto, Canada. Both instruments are based on LII of BC.

In some studies, BC EFs were based on particulate matter collected to the filter samples. Typically, BC on the quartz filter was determined with the sunset organic carbon (OC)/EC instrument (Birch and Gary 1996) or DRI reflectance method (Chow *et al* 1993) and therefore it was called in these cases EC. Same technique is utilized in the semi-continuous OC/EC instrument that was used in Pittsburgh, US with hourly time-resolution of EC (Li *et al* 2020). Additionally, Sánchez-Ccoyllo *et al* (2009) utilized smoke stain reflectometer for the filter samples to obtain BC EFs in Sao Paulo, Brazil.

The most common method to calculate the BC EF at the roadside is a tracer method that utilizes concurrently measured gas (typically CO_2) concentrations by assuming that BC and the tracer component dilute with the same rate and the dilution is faster than other atmospheric processes affecting the measured concentrations. In the simplest method, the background concentration of BC is subtracted from the ambient BC concentration and the resulted value is divided by the ambient CO_2 concentration subtracted by the background CO_2. This ratio is then multiplied by the carbon mass fraction of the fuel (kg $kg^{-1}{}_{fuel}$). In some investigations, also the concentration of CO has been considered by using the sum of CO_2 and CO instead of solely CO_2.

In addition to CO_2, also NO_x has been used as a tracer for vehicle emissions when calculating BC EFs (e.g. Krecl *et al* 2017, de Miranda *et al* 2019, Martinet *et al* 2019). NO_x has been shown to be a good tracer at street level (Imhof *et al* 2005) as it has strong correlation with other traffic-related species like BC and its emissions are fairly well-known for various traffic situations and available on several traffic emission data bases. We note that NO_x may not be a good and unambiguous tracer method in all the studies e.g. due to the larger utilization of selective catalytic reduction (SCR) or exhaust gas recirculation (EGR) technologies that mitigate NO_x but not BC. BC EFs have also been calculated by considering the measured increment concentration of BC due to the traffic emissions, total traffic rate and the dilution rate (Krecl *et al* 2018). Traffic increment is determined by utilizing the measurement for example at the location not influenced by the local street traffic. Dilution rate can be modeled by the inverse modeling (Madueño *et al* 2019).

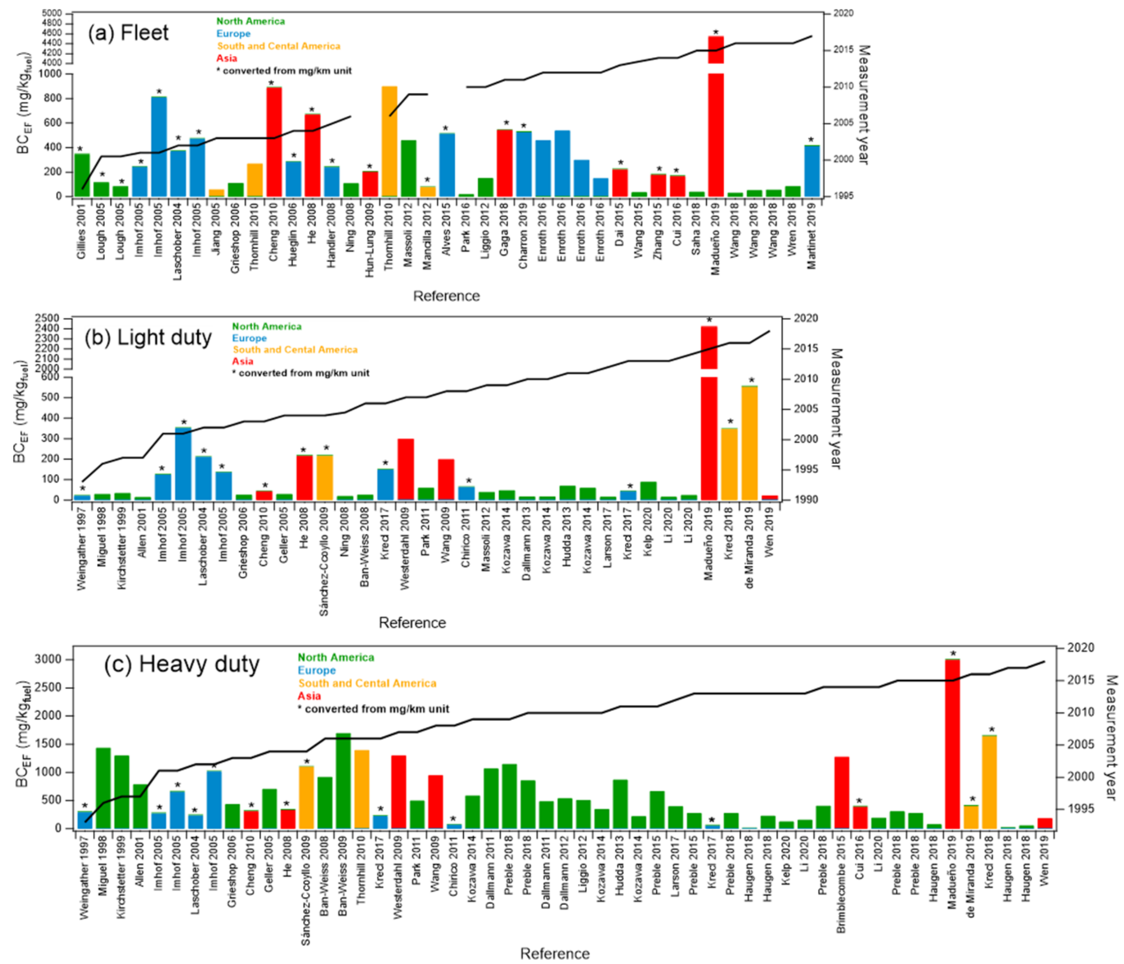

Figure 1. BC EFs obtained for total fleet (a), light-duty vehicles (b) and heavy-duty vehicles (c) by ambient air measurements. BC EFs are presented in the order of the measurement year shown in right vertical axis of the subfigures. The continent where each measurement has been conducted is indicated by color. Values that are converted from mg km^{-1} are marked with asterisk.

BC EFs based on tunnel measurements have been calculated differently, from the difference in the BC concentration in the entrance and exit of the tunnel. Additionally, in these investigations the ventilation of the tunnel, the distance from the tunnel entrance to the measurement point and the total number of vehicles passing the tunnel needs to be known (Handler *et al* 2008, Chirico *et al* 2011).

The determination of BC EFs separately for HD and light duty traffic was conducted by different methods. In many studies the traffic composition was characterized visually or by license plate survey (e.g. Miguel *et al* 1998, Kirchstetter *et al* 1999) and separate EFs were determined with linear regression (Weingartner *et al* 1997). Measurements were also carried out in tunnels or highways that were allowed to only one vehicle category (e.g. Allen *et al* 2001, Hudda *et al* 2013). Additionally, Thornhill *et al* (2010) and Krecl *et al* (2017) have utilized positive matrix factorization to separate the emissions from the heavy and light duty vehicles, and Larson *et al* (2017) have exploited absolute principal component scores to explore the heavy and light duty features of traffic emissions.

3.1.2. Ambient emission factor studies

Figure 1 and table S1 presents BC EFs determined for total fleet, HD vehicles and light-duty vehicles by measuring the BC and trace gases from ambient air. For the fleet, BC EFs varied significantly, from 20 to 4500 mg kg_{fuel}^{-1}, the average, median and standard deviation of BC EF being 400, 240 and 720 mg kg_{fuel}^{-1}, respectively. The smallest single BC EFs values were measured in North America and the largest in Asia. That is the trend also in general, the largest BC EFs were observed in Asia, Central and Southern America and Europe whereas measured BC EFs were smaller in Northern America. However, there was a wide variation also within the continents. Measurements were carried out in a total of 14 different countries. The measurements cover the time period from 1996 to 2017 but no clear trend in time for the fleet BC EFs was found. Four out of five largest single points were, however, measured before 2007, slightly indicating the decrease in BC EFs of traffic fleets. We did not find investigations conducted in Africa, Australia, and Oceania.

For HD vehicle traffic, the variation in BC EFs (36–3000 mg kg_{fuel}^{-1}) was slightly smaller than for the total fleet, the average, median and standard deviation of BC EF values being 620, 410 and 560 mg kg_{fuel}^{-1}, respectively. Different from the total fleet, for HD a decreasing time trend was observed excluding the three BC EF values measured in Asia and Central and South America after 2013. The decreasing trend was observed especially for BC EFs measured in North America but also in Europe.

For light duty vehicles, the measured BC EFs were in range 15–2400 mg kg_{fuel}^{-1} with average, median and standard deviation being 170, 54 and 400 mg kg_{fuel}^{-1}, respectively. Similar to the total fleet, measured BC EFs were in general smaller in North America than in Europe, Asia and Central and South America. For light duty vehicles, clear variation in time was not observed but the largest values were obtained in Central and South America and Asia after 2014.

The impact of season on BC EFs has been investigated in several studies. Saha *et al* (2018) measured slightly smaller BC EFs in winter (35 ± 3 mg kg_{fuel}^{-1}) than in summer (45 ± 3 mg kg_{fuel}^{-1}) in North Carolina. Kozawa *et al* (2014) reported smaller BC EFs in September (15 ± 11 mg kg_{fuel}^{-1}) than in May (21 ± 15 mg kg_{fuel}^{-1}) at a freeway in California in 2010, however, the difference between the years was even larger as in 2011 BC EF was larger in June (67 ± 31 mg kg_{fuel}^{-1}) than in September (54 ± 6 mg kg_{fuel}^{-1}). Wang *et al* (2018) have studied the temperature dependency in more detail and found that BC EF increased with increasing ambient temperature that was opposite trend to CO, NO_x and particle number EFs. In contrast, Li *et al* (2020) measured slightly larger EC EFs in winter than in spring both for HD (winter 197 ± 41 mg kg_{fuel}^{-1}, spring159 ± 34 mg kg_{fuel}^{-1}) and light duty vehicles (winter 24 ± 5.5 mg kg_{fuel}^{-1}, spring 16 ± 4.5 mg kg_{fuel}^{-1}) in a tunnel in Pittsburgh. BC EFs have been measured also separately at weekdays and weekends and at different diurnal hours but in those cases the driving force for the differences has typically been different contributions of heavy and light duty vehicles (e.g. Krecl *et al* 2018, Wang *et al* 2018). BC EFs have also been noticed to depend on the operating modes of the vehicles which are affected e.g. by the slope of the road; Mancilla and Mendoza (2012) determined EC EF for the traffic in uphill and downhill bores of a tunnel in Mexico, the EC EF being larger for the traffic in uphill bore (8.9 ± 2.1 mg km^{-1}) than for the traffic in downhill bore (2.5 ± 2.1 mg km^{-1}) for total vehicle fleet.

In addition to light-duty and HD vehicles, train and airplane EFs have been extracted from ambient data. Based on BC emission measurements performed next to railroad for 84 trains the average BC EF of 0.66 g kg_{fuel}^{-1} were obtained for diesel trains by Jaffe *et al* (2014). In the study made by Tang *et al* (2015), the average BC EF of 0.87 ± 0.66 g kg_{fuel}^{-1} was reported for the for a California commuter rail line fleet of diesel-electric passenger locomotives (29 locomotives), determined by simultaneous BC and CO_2 concentration measurements for the exhaust plumes of passing locomotives. They reported the BC EFs to be depended on engine load and speed; BC EFs were typically higher for accelerating locomotives traveling at higher speeds. Galvis *et al* (2013) used three methods for BC EF calculation (upwind–downwind difference in CO_2, wavelet analysis and regression method) for diesel train BC emissions at the vicinity of railyard. Depending on method, Galvis *et al* (2013) gained BC EFs of 0.16–0.73 g kg_{fuel}^{-1} for diesel trains. Shirmohammadi *et al* (2017) observed BC EFs of 0.12 and 0.11 g kg_{fuel}^{-1} for airplanes during takeoffs and landings near the Los Angeles International Airport (LAX, 150 m downwind of south runways).

3.2. BC EFs of road vehicles

3.2.1. Measurement techniques and calculation methods for BC EFs

The BC EFs of road traffic category were divided into the following sections: gasoline and diesel passenger cars, HD vehicles, and L-category vehicles.

The passenger car (i.e. light-duty vehicle) BC EFs were covered in 17 articles which included studies for both gasoline and diesel vehicles tested on chassis dynamometer. The studies reported the effects of several different parameters including e.g. model year, fuel, exhaust aftertreatment, test cycle, on the BC EFs. For the BC measurement, variety of different instruments were used; MAAP (Giechaskiel *et al* 2010, Karavalakis *et al* 2014), AE33 (Timonen *et al* 2017, Pieber *et al* 2018, Pirjola *et al* 2019), AE51 (He *et al* 2018, Zheng *et al* 2019), AE31 (Chirico *et al* 2010, Deng *et al* 2020), thermal-optical EC/OC analyzer (May *et al* 2014), soot particle aerosol mass spectrometer (SP-AMS) (Karjalainen *et al* 2016), micro soot sensor (MSS) (Roth *et al* 2019, Yang *et al* 2019), and photoacoustic soot spectrometer (PASS-3) (Zimmerman *et al* 2016). In the studies, BC EFs were obtained mainly from constant volume sampler (CVS) measurements where the full exhaust flow was taken into the CVS, and the concentrations were measured in the controlled CVS flow. In general, associating this to the chassis roll data (vehicle speed as a function of time), e.g. temporal emission factor calculation is straightforward in that kind of experiments. Most of the studies reported BC EFs for a single or few vehicles whereas May *et al* (2014) comprised measurements of 66 vehicles.

The HD vehicle BC EFs were covered in eight articles. The starting temperatures, ladings, fuels, emission levels and emission control devices, as well as the test cycles on chassis dynamometer and the test routes for PEMS or chase measurement varied between the studies. Variety of different BC instruments were used in the studies; MAAP (Giechaskiel *et al* 2010), AE51, AE31, and AE33 (Gordon *et al* 2014, Book *et al* 2015, Zheng *et al* 2015, Järvinen *et al* 2019), reflectometer

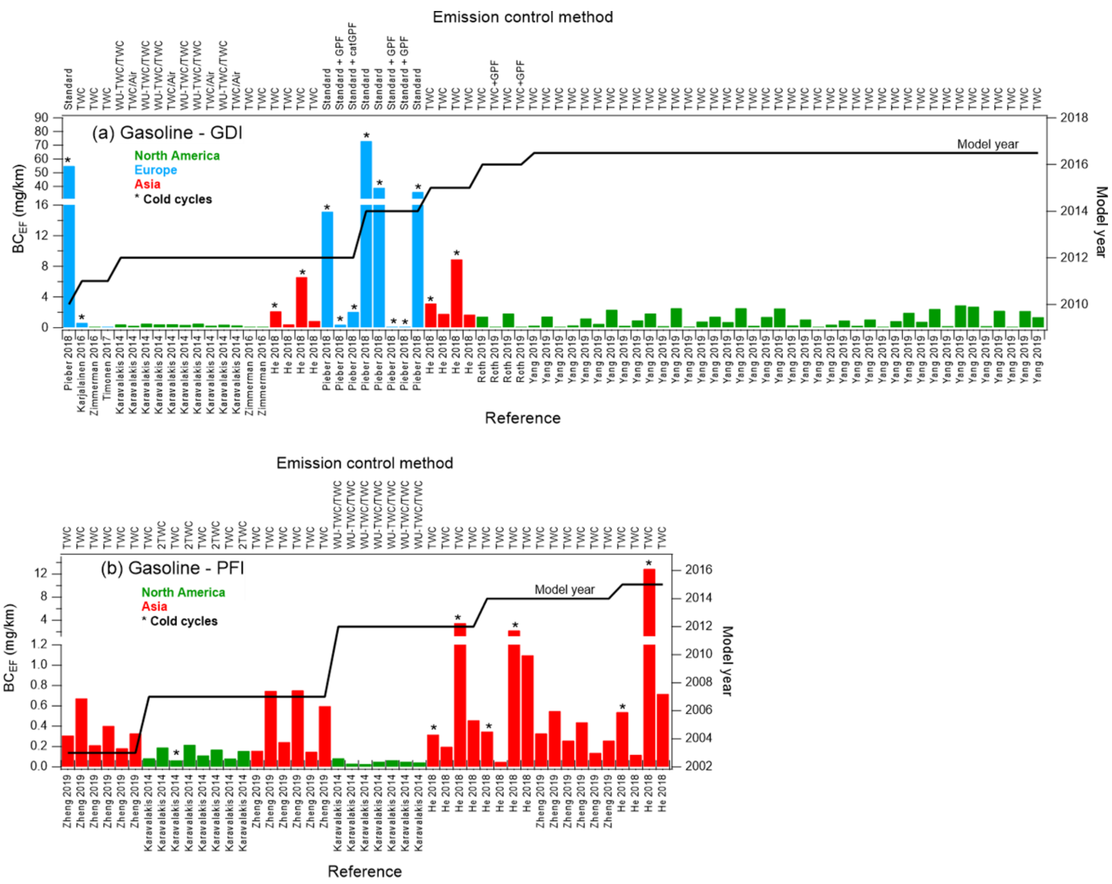

Figure 2. BC EFs (mg km^{-1}, left y-axis) for GDI and PFI gasoline passenger cars presented in the order of model year (right y-axis). The colors indicate the geographical area (North America, Asia, Europe) where the measurements were made. The exhaust aftertreatment devices of the vehicles are shown on upper x-axes, and cold test cycles are marked with asterisks.

(Oanh *et al* 2010) and sunset thermal-optical EC/OC analyzer (Oanh *et al* 2010, Hays *et al* 2017). For instance, Oanh *et al* (2010) investigated 29 HD buses and 25 HD trucks on a chassis dynamometer using exhaust dilution system (including CVS) following EU's Directive 70/220/EEC test protocol. They measured the BC concentrations from CVS using reflectometer and EC/OC analyzer and determined BC EFs by combining measured concentrations with chassis dynamometer data. We conducted the literature search also for non-road mobile machinery, typically equipped with diesel engines, but did not find studies reporting the measurement results of BC EFs.

The L-category vehicles BC EFs were found from two articles covering Euro 1 and Euro 2 emission levels in Europe (Clairotte *et al* 2012, Giechaskiel *et al* 2012). Euro 1 and Euro 2 L-category vehicles were tested using hot and cold ECE-47 cycles and two phases (1 and 2) of the Worldwide Harmonized Motorcycle Test Cycle (WMTC) cycle. One Euro 2 L-vehicle was tested with steady and acceleration points. In these studies, all the BC measurements were made with MAAP.

3.2.2. Road vehicle emission factor studies

The gasoline vehicle BC EFs (figure 2, table S2) were covered in ten articles. These studies originated from several geographical areas, i.e. North America, Asia, and Europe, that all have their own emission regulations for vehicles. The studies found from the literature included substantial variation of techniques; e.g. fuel injection (port fuel injection (PFI)/direct injection), fuel quality and ethanol content of fuel, exhaust aftertreatment and driving cycle varied between the individual BC emission measurements. Gasoline vehicles were typically equipped with three-way catalytic converters which have impact on gaseous emissions of the vehicles, but their effect on BC EFs can be assumed to be minor. In general, a large variation was observed in BC EFs of gasoline vehicles, starting from values close to zero up to the level of 80 mg km^{-1}. Although the emission regulations have been significantly tightened during past 30 years related to gaseous and particulate phase pollutants from on-road vehicles, it can be seen from figure 2 that this trend was not directly reflected to the observations of the BC emissions of gasoline vehicles.

Based on the reviewed studies, following observations can be made regarding BC EFs of gasoline vehicles: (a) gasoline direct injection (GDI) engines have potentially higher BC EFs than PFI gasoline engines using premixed fuel-air mixture (figure 2); (b) increase in the ethanol content of fuel decreases the BC EFs (Karavalakis *et al* 2014, Timonen *et al* 2017); (c) gasoline particulate filters are effective in reducing the BC EFs of gasoline vehicles (figure 2, see also Pieber *et al* 2018); (d) significantly higher BC EFs are measured for cold cycles, both with GDI and PFI cars. Interestingly, the BC EFs of Asian gasoline vehicles, that seem to stable or even increasing in respect of vehicles' model year, seem to be higher than the BC EFs from gasoline cars used in North America.

The BC emissions of diesel passenger cars were covered in four articles included in this review (table S2: Chirico *et al* 2010, Giechaskiel *et al* 2010, Pirjola *et al* 2019, Deng *et al* 2020), reporting the BC EFs in the units of mg kg_{fuel}^{-1} and mg km^{-1}. Those studies originated mainly from Europe, where diesel engines possess a significant share of the vehicle market. Similarly with gasoline vehicles above, the BC EFs has large variation starting from about ∼0 mg kg_{fuel}^{-1} levels up to the level of 25 mg kg_{fuel}^{-1}, Also, experimental condition and parameters potentially affecting the BC EFs, such like model year, exhaust aftertreatment and driving conditions (see table S2) varied significantly between the studies. For diesel vehicles, the key technological parameter concerning the BC emissions is the existence of the diesel particulate filter (DPF) in the exhaust pipe. The DPF systems are designed to efficiently reduce the concentrations of nonvolatile particle number for particles larger than 23 nm in the exhaust; for instance, the study conducted by Wihersaari *et al* (2020) reported 99.998% reduction efficiency of particle number for a DPF system used in diesel passenger car. Since the nonvolatile particle number limit mainly concerns BC-rich soot particles, the DPF systems are also efficient in BC removal, even so efficient that the BC EFs after the typical DPF can be assumed to be close to zero. However, reported measurements of BC EFs for diesel cars equipped with the DPF were not found. Another trend in diesel vehicle development has been the improvements of diesel engine combustion efficiency which has been achieved by improvement in combustion parameters like fuel injection pressure and injection timing (Lähde *et al* 2011), seen e.g. in figure 3 which compares the BC EFs of older and newer cars. Furthermore, renewable fuels produce less BC than fossil fuels which is mainly due to the fact that renewable fuels, in general, have higher fuel originated oxygen contents and contain less aromatic compounds (see e.g. Pirjola *et al* 2019). Both can lead to reduced BC formation in diesel combustion process and thus to lower BC EFs.

The BC EFs of HD vehicles are shown in figure 4 and in table S3. In general, the variation of BC EFs was several orders of magnitude; the range of BC EF values was <0.02–2600 mg km^{-1}, and average value was 130 mg km^{-1}, median was 35 mg km^{-1}, and the standard deviation of the values was 270 mg km^{-1}. The highest BC EF values, larger than 2000 mg km^{-1}, were seen with very old vehicle technologies but, however, this kind of high-emitters are currently not frequently met in traffic. Although the older model vehicles are a minority of vehicle fleets, they can account for the large fraction of the BC emissions and, in general, their role should be included e.g. in emission estimates and inventories. In terms of mitigation, removing these older vehicles from traffic is generally seen as an effective and efficient strategy to reduction on-road traffic BC emissions.

The BC EFs for HD vehicles clearly confirms the crucial role of DPF in BC emission reduction; while the above-mentioned relatively high BC EFs were for the HD vehicles without the DPF, BC EFs were lower than 2 mg km^{-1} for all vehicles with the DPF. Importantly, in part of the experiments the BC EFs of HD diesel vehicles were even below the detection limit of the measurement systems used in the studies. In principle, the exhaust aftertreatment based emission reduction methods for gaseous compounds (diesel oxidation catalyst (DOC), NO_x absorber catalyst (NAC), SCR) may not very significantly affect the BC emissions of HD diesel vehicles. However, e.g. Preble *et al* (2018) have reported lower BC EFs for the HD vehicles with DPF and SCR than for the HD vehicles equipped with DPF only. They discussed that the observation can be linked with engine age, engine management strategies, or changes in DPF system durability. It should be kept in mind that these other emission control devices can have other effects on particulate emissions, e.g. by affecting the co-emitted compounds, i.e. semivolatile compounds (see e.g. Rönkkö *et al* 2013) and compounds that can form secondary aerosol particles in the atmosphere (e.g. Gordon *et al* 2014).

In addition to the effects of DPF, figure 4 indicates that the BC EFs of HD diesel vehicles are affected by other technological development also. This can be seen as a decreasing trend of BC EFs measured for HD diesel vehicles in Asia; clear decrease was seen as a function of model year, except one experiment with a modern vehicle. Because the other exhaust aftertreatment devices than DPF are not significantly affecting the BC emissions, this declining trend may be linked to the development of engines and/or fuels.

In addition to the BC EFs in mg km^{-1} shown in figure 4, the BC EFs of HD diesel vehicles have been reported in mg kg_{fuel}^{-1} (e.g. dynamometer study by Gordon *et al* (2014), chase study by Järvinen *et al* (2019)). Järvinen *et al* (2019) have reported BC EFs for buses with different emission levels; enhanced environmentally friendly vehicle (EEV) bus utilizing EGR, two Euro VI buses (one using EGR, DPF and SCR, the other using DPF and SCR) and two EEV buses that were equipped with retrofitted exhaust aftertreatment systems (DPF + SCR). The experiments were made with warm engine and on urban driving conditions, measuring the BC with AE33 installed to the chasing mobile laboratory. The reported EF BCs were 100, 10, 0, 10 and 10 mg kg_{fuel}^{-1},

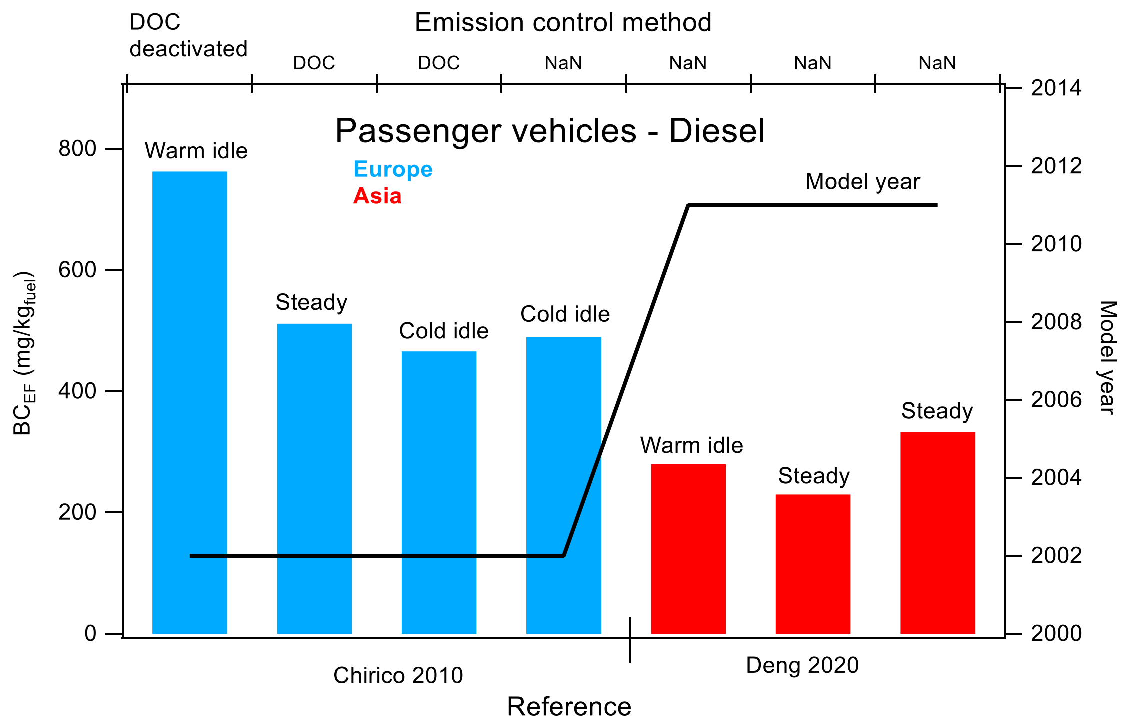

Figure 3. Comparison of BC EF's (mg kg^{-1} fuel) of diesel passenger cars without DPF. The colors indicate the geographical area (Asia, Europe) where the measurements were made, the y-axes on the left shows the BC EF value and the y-axes on the right shows the model year of the cars. Please note that the modern diesel passenger cars are typically equipped with DPF which decreases the BC EF values close to zero (see e.g. Wihersaari *et al* 2020).

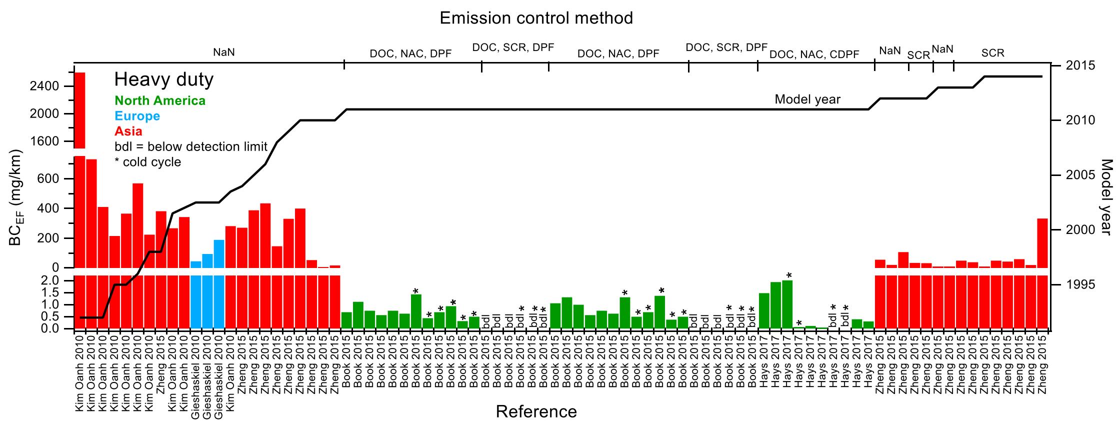

Figure 4. BC EFs (mg km^{-1}) for individual heavy-duty (HD) trucks and buses with different model year. The color indicates the continent (North America, Asia, Europe) where measurements have been made. The emission control methods are shown on upper x-axes; DOC is the diesel oxidation catalyst, NAC is NO_x absorber catalyst, DPF is diesel particulate filter, SCR is selective catalytic reduction, none means that there are no aftertreatment devices, and NaN means that the article does not contain the information. The experiments, where the BC EF was not able to be determined due to the low emissions compared to the measurement methodology sensitivity, are indicated by BDL (below detection limit).

respectively, showing the variation in the BC EFs and demonstrating the effects of technology level on real-world BC emissions. In the study of Gordon *et al* (2014), four HD trucks with different combinations of exhaust aftertreatment devices (DOC, DPF and SCR; DOC and DPF; none; none) and two medium-duty vehicles (DOC; none) have been studied at different driving conditions. The reported BC EFs by Gordon *et al* (2014) were zero for the HD vehicles with DPF, 579.9 mg kg_{fuel}^{-1} and 218.3 mg kg_{fuel}^{-1} for the HD vehicles without the DPF, and 881.9 mg kg_{fuel}^{-1} and 1041.6 mg kg_{fuel}^{-1} for the medium-duty vehicles, all measured using an AE. Thus, the results of these two studies are in line with the values presented in figure 4, indicating again the important role of DPF in mitigation of BC emissions from traffic.

In our literature searches, we did not find measurements of BC EFs of non-road mobile machinery. In general, due to the large use of diesel engines in

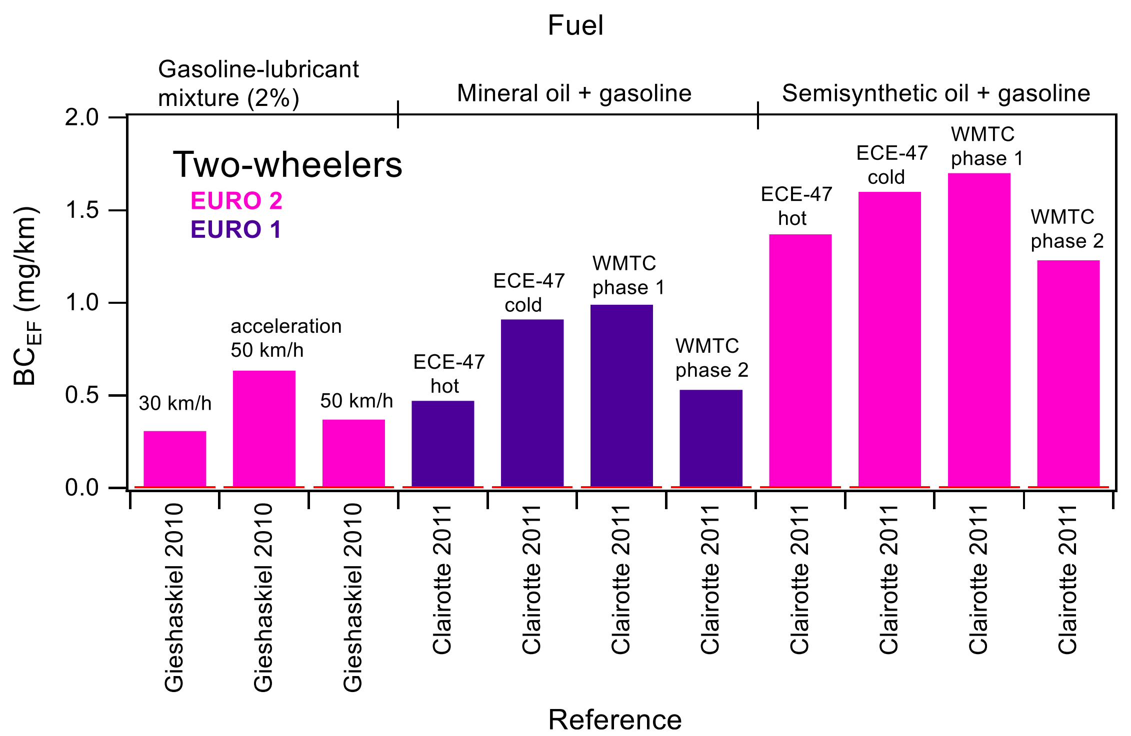

Figure 5. BC EFs (mg km^{-1}) of EURO1 and EURO2 two-wheelers using different fuels (gasoline-lubricant mixture (2%), mineral oil + gasoline and semisynthetic oil + gasoline) for different speeds, cycle phases and initial engine temperature points.

these machines, their BC emissions can be assumed to be largely controlled by the age of the machine and use of DPF in exhaust aftertreatment system. For instance, in the study of Karjalainen *et al* (2019), the BC emissions of diesel engine have been measured for several exhaust aftertreatment scenarios and for two fuels, also demonstrating the BC emissions from non-road mobile machinery.

The BC EFs of two-wheelers (L-category vehicles) have been reported in very limited number of studies. In the studies by Giechaskiel *et al* (2010) and Clairotte *et al* (2012), the BC emission factor range was 0.4–1.7 mg km^{-1}, average BC EF was 0.9 mg km^{-1}, median BC EF was 0.9 mg km^{-1}, and the standard deviation of BC EFs was 0.4 mg km^{-1} (figure 5, table S4). In general, the BC emissions of two-wheelers were relatively low, especially when reporting in kilometer-based units; the values are in the same range with the diesel cars with the DPF. However, the BC EFs of two-wheelers can be significantly affected e.g. by driving conditions (higher emissions for cold starts and acceleration) and the fuel-lubricant mixture (figure 5). Furthermore, it should be kept in mind that the two-wheelers' BC EFs per passenger kilometer can be high when compared to vehicles with multiple passengers.

3.3. Marine traffic

3.3.1. Measurement techniques and calculation methods for BC EFs

In marine emission studies, the analysis of BC has been mostly based on filter sampling and thermal optical OC/EC analysis (Sunset Laboratories or DRI method). In some studies, also methods and instruments including filter smoke number, MAAP, and AE33 AE have been applied. Corbin *et al* (2020) used various methods to determine BC emissions, and from their study, the results from LII measurements were selected for this review. BC measurement methods used in reviewed publications are given in the table S5. BC EFs were originally reported in various units, mostly per kg_{fuel} or per kWh. Conversion procedure to per MJ_{fuel} is specific for each paper and reported in the study by Aakko-Saksa *et al* (2021).

3.3.2. Marine traffic emission factor studies

The international ship traffic is currently motored mainly by diesel engines burning heavy fuel oil. The engines used in ocean going vessels include both two- and four-stroke diesel engines that can be categorized as medium and slow speed engines. Typically, vessels have many engines that can be run according to the different power needs at open seas and harbors. Recently, the use of cleaner diesel fuels (marine gas oil, marine diesel oil) and natural gas have increased, and dual-fuel engines have become available.

The global ship traffic is governed by the IMO which has set targets under pollution prevention treaty (MARPOL) to reduce global greenhouse gas emissions from shipping by 50% by 2050 compared to 2008. Also, emission limits for BC have been discussed by the IMO, especially for the Arctic area. Currently, emissions from shipping have been mitigated by limiting the sulfur content of marine fuels to 0.5%

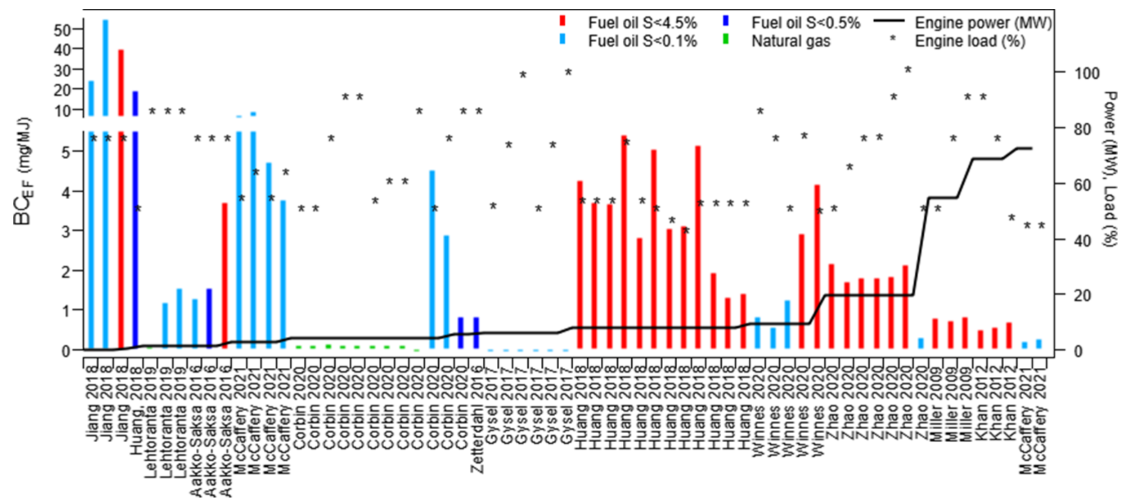

Figure 6. BC emission factors of marine engines of various power consuming residual fuels. Jiang *et al* (2018) was high-speed engine, other either medium- or low-speed engines. All results depict engine operation at high load points (load >40% of maximum continuous rating) representing typical marine engine operation. The solid line marks maximum engine power whereas the asterisks indicate engine load.

S and by establishing sulfur and NO_x emission control areas in certain marine regions where the sulfur limit is 0.1%. In addition, regional limits exist e.g. by the EU and China. Particle number emissions of ship engines are only limited for inland waterway vessels in the EU. The existing legislation has driven ship operators to adopt new fuels and aftertreatment technologies such as scrubbers, SCR, and DOC, but diesel particle filter (DPFs) are not yet adopted by the shipping sector.

The studies included in this review (figure 6) reported emissions from marine engines applied in shipping which are typically compression ignited medium or low-speed engines operating with either four-stroke or two-stroke cycle. The included engines had maximum continuous rating of power between 0.2 and 68.5 MW representing variation from engines applied in shipping boats to engines applied in ocean-going vessels.

The studies reporting BC EFs for marine engines with maximum rated powers between 187 kW and 72 MW are shown in figure 6 and table S5. BC emissions from marine engines varied between 0.07 and 52 mg MJ_{fuel}^{-1}, with average, median and standard deviation of 3.8, 1.3 and 8.6 mg MJ^{-1}, respectively. The highest BC emissions were reported by Jiang *et al* (2018) for a two-stroke marine engine manufactured in 1980s. Majority of the values, considering only the medium and low-speed engines, were between 0.5 and 5 mg MJ_{fuel}^{-1}.

In selected publications, engines consumed various fuels: residual and distillate fuels and natural gas with ignition by aid of pilot fuel. The fuels were classified primarily according to their type and residual fuels according to sulfur content. The studies were categorized according to fuel type used by the engine and engine loads above 40%, corresponding to cruising operations at open sea, were included. Engine load affects BC emissions from marine engines. Medium to high engine loads (>40%) typically produce the lowest BC emissions. If engines are operated at low loads, BC emissions may increase significantly (Zhao *et al* 2020).

In general, BC EFs of marine engines depend on the maximum engine power, i.e. engine size (indicated by black line in figure 6). The largest engines seem to emit less BC than smaller ones, which is likely related to more lenient NO_x regulation (discussed in detail by Aakko-Saksa *et al* 2021). In addition, the engines with the highest maximum power are typically two-stroke engines operating at low rotation speed, whereas the medium-sized engines (often auxiliary engines) are typically medium-speed, and four-stroke.

Also, fuel type was observed to affect emissions significantly which can be seen by comparing the bars colored differently in figure 6 (high sulfur fuel oils indicated by red, lower sulfur contents by blue, and nearly sulfur free natural gas by green). It can be seen from the figure that, in general, high sulfur fuel oils tend to produce higher BC EFs than low sulfur fuels (see also Zhao *et al* 2020, Aakko-Saksa *et al* 2016, Huang *et al* 2018, McCaffery *et al* 2021). Especially the studies that report BC emissions from same engine burning multiple fuels can be used to compare the emission effects of fuels, and they indicate relatively clearly that shifting to lower sulfur fuel oils reduces BC emissions (Zhao *et al* 2020, Aakko-Saksa *et al* 2016). High sulfur fuels are mostly heavy fuel oils containing also other impurities and heavy hydrocarbons in significant numbers, whereas low sulfur fuels are often comparable to diesel fuels used in road traffic. Thus, the differences in the BC emissions may not be explained by sulfur content itself but overall properties of the fuels. Compared to fuel oils, natural gas offers significant reduction in BC emissions (figure 6). Natural gas engines are often dual fuel engines, where gas is ignited by injecting pilot oil and engine may also run entirely on fuel oil such as in Lehtoranta *et al* (2019) and Corbin *et al* (2020). When run with fuel oil, dual fuel engines do not offer any BC emission advantages.

It should be noted that compared to road vehicles, particle filtration technologies have not yet been implemented in large scale in marine engines and their effects in emissions have not been studied widely. Also, newer engines with more advanced technology and injection system most likely emit less BC, but old engines are still in use in large scale and relevant to marine BC emissions. Furthermore, also the other harbor activities and smaller vessels can have significant BC emissions and relatively large EFs; e.g. in the recent study of Schlaerth *et al* (2021), the BC EFs of 0.56 ± 0.86 g $\mathrm{kg_{fuel}}^{-1}$ (passenger boats), 0.64 ± 0.29 g $\mathrm{kg_{fuel}}^{-1}$ (tugboats with loads), 0.48 ± 0.67 g $\mathrm{kg_{fuel}}^{-1}$ (tugboats without loads), 0.36 ± 1.2 g $\mathrm{kg_{fuel}}^{-1}$ (fishing boats) were reported. In the study of Sugrue *et al* (2022) for the BC emissions of in-use excursion vessels and ferries, the mode-weighted mean BC EF values for each engine tier varied from 0.05 g $\mathrm{kg_{fuel}}^{-1}$ (Tier 4, active SCR) to 0.68 g $\mathrm{kg_{fuel}}^{-1}$ (Tier 0).

3.4. Aviation

3.4.1. Measurement techniques and calculation methods for BC EFs

The BC EFs from aviation were covered in eight publications which reported results from ground-based aircraft engine measurements. Typically, the BC emissions were sampled either using a probe directly at the engine exhaust outlet and diluting and cooling the sample applying ejector diluters (Chan *et al* 2013, 2015, Durdina *et al* 2014, 2017) or alternatively by sampling the exhaust downwind the engine, allowing natural cooling and dilution of the exhaust (Kinsey *et al* 2011, 2019, Moore *et al* 2015, Yu *et al* 2017). BC detection techniques in aviation studies include various methods; MAAP, AE, MSS, and LII instruments. In most cases, BC EFs were reported for various engine thrust conditions, somewhat adapting to the landing and take-off (LTO) emission test cycle defined by the International Civil Aviation Organization (ICAO). An LTO cycle covers two aircraft operations, landing (approach, landing, and taxi-in to the gate) and take-off (taxi-out onto the runway, take-off and climb-out) (ICAO 2021). The studied engines were classified either as turbofan (turbine jet engine) or as turboprop (turbine engine driving propeller) type.

3.4.2. Aviation emission factor studies

The BC EFs reported for aviation engines ranged from 0.1 to 554 mg $\mathrm{kg_{fuel}}^{-1}$, with average, standard deviation and median at 77, 106 and 33 mg $\mathrm{kg_{fuel}}^{-1}$, respectively (figure 7, table S6). The emissions are strongly dependent on the engine thrust condition as well as studied fuel. Typically, the BC EFs follow a U-shaped curve, having high emissions at idle conditions, and again at high thrust percentages, corresponding to engine operation during cruising, climb-out and take-off. Cruise corresponds to 80% thrust (Chan *et al* 2015), whereas climb-out requires 85% (Durdina *et al* 2017, Kinsey *et al* 2019), and take-off 95%–100% of the nominal engine thrust (Chan *et al* 2015, Durdina *et al* 2017, Kinsey *et al* 2019). Durdina *et al* (2017) found that for a 1 h flight, over 70% of the BC emissions came from the climb phase and over 25% dispersed under 3000 ft altitude. The BC emissions from cruise operations were found to reach 50% of the total BC emissions when the flight time was increased to 4 h.

The studies consider various aviation fuels, ranging from conventional kerosene-based jet fuels to synthetic and paraffinic kerosenes produced through Fischer–Tropsch (FT) synthesis from fossil or biogenic sources. The most widely used aviation fuel are kerosene type Jet A-1 and Jet-A, which is used in the USA and has a lower maximum freezing point. Wide-cut jet fuel (Jet B) still is used in some parts of Canada and Alaska as it is suited to cold climates. JP-8 (the US military) and F34 (NATO) are military jet fuels, similar to commercial aviation's Jet A-1, but with the addition of corrosion inhibitor and anti-icing additives (Chevron 2007). The alternative fuels in the studies include synthetic FT kerosene produced from natural gas (FT (NG), Kinsey *et al* 2019), synthetic paraffinic kerosene (SPK) produced from coal (FT (coal), Kinsey *et al* 2019), and fully synthetic FT SPK (Chan *et al* 2015). The studies also reported BC EFs for three renewable fuels; (a) fully synthetic kerosene with aromatics (CH-SKA, Chan *et al* 2015) from industrial oilseeds carinata (*Brassica carinata*) produced through catalytic hydrothermolysis process, (b) 50% volume mixed hydro-processed esters and fatty acid (HEFA) paraffinic kerosene produced from camelina (*Camelina sativa L.*) oilseeds through a hydro-treating and cracking process, mixed with 50% Jet A-1 (HEFA-SPK, Chan *et al* 2015), and (c) camelina-based hydro-processed bio-jetfuel by 50% in volume mixed with F34 (C-HEFA, Chan *et al* 2013).

The synthetic kerosenes produced either from fossil or renewable sources lead to lower BC EFs from aviation engines. Chan *et al* (2013) found that the C-HEFA blend reduced BC emissions by 50% at idle and 32% at take-off when compared to F34. In the study by Chan *et al* (2015) the use of 50% HEFA-SPK led to significant reduction in BC mass emissions compared to Jet A-1 by 58%–85% for various engine load conditions. For the 100% FT-SPK fuel, reductions of BC emissions compared to Jet A-1 emissions of 70%–97% were achieved. Moore *et al* (2015)

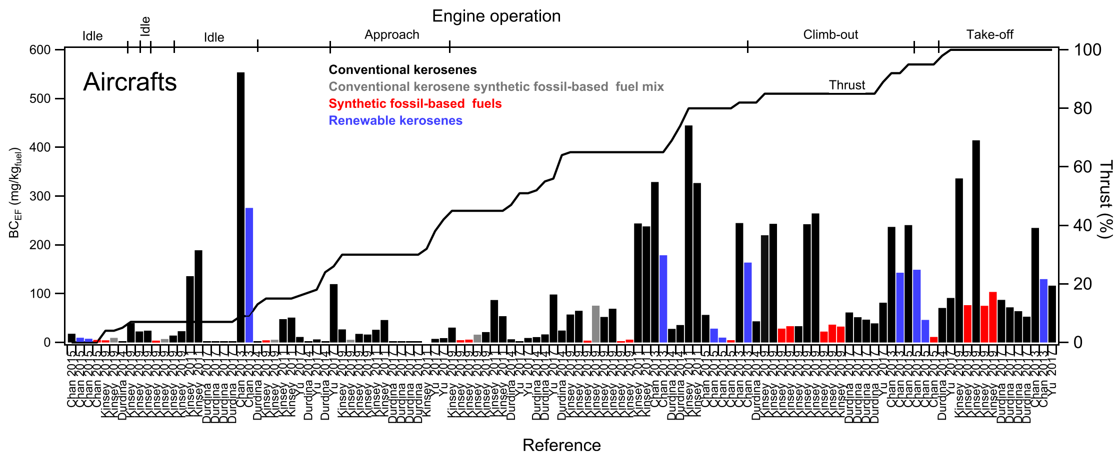

Figure 7. BC EFs emissions from aircrafts with various fuels. Jet-A and Jet-A1 denote conventional kerosene fuels, and JP-8, and F34 their military alternatives. FT (coal), FT (NG), and FT-SPK denote synthetic kerosenes, which are produced through FT synthesis from natural gas or coal and contain very low levels of sulfur or aromatics. CH-SKA, HEFA-SPK, and C-HEFA denote synthetic kerosenes derived from biogenic sources, which apart from CH-SKA have low sulfur and aromatic contents. Conventional kerosenes are marked with black color, synthetic fossil-based fuels with red, and renewable kerosenes by blue color.

provided meta-analysis of several aviation studies (with $N = 439$) and concluded that while the fuel aromatic and sulfur content mostly affect the volatile aerosol fraction, the naphthalenic content of the fuel determined the magnitude of the BC EFs (Moore *et al* 2015). Naphthalene is the simplest two-ring (dicyclic) aromatic that can be found in Jet-A and Jet-A1 fuels in up to 3% by volume (Chevron 2007).

Yu *et al* (2017) reported BC EFs for two different engine types both burning Jet-A fuel. They found that the turboprop engine generally emitted more BC. Also, the BC EFs reported by Chan *et al* (2013) are higher than for turbofan engines at low thrust conditions. In addition to main propulsion engines, auxiliary power units (APUs) contribute to BC emissions from aircrafts. Kinsey *et al* (2012) found the BC EFs from APU to vary between 20 and 450 mg $kg_{fuel}{}^{-1}$, depending on fuel and operation condition.

3.5. Residential combustion

Combustion of fossil and renewable fuels are utilized in households for cooking, heating and pleasure (saunas, visual effects of fire), resulting to substantial BC emissions from these sources. In general, residential BC sources are characterized by large variation of different fuels and combustion appliances and several applications, as well as a lack of standardized emission measurements; in scientific literature most of the experiments have been designed independently and separately, and international emission and measurement standards have not typically affected the study designs. This has led to the situation where the comparability of BC EFs is not ideal. On the other hand, the existing BC EF information is versatile, due to the different society levels and habits, available and affordable fuel sources and combustion technologies.

3.5.1. Measurement techniques and calculation methods for BC EFs

The BC EFs from residential combustion were covered in 27 publications which reported results from large variation of different BC emission measurements. Several experimental setups have been used; the flue gas from the combustion has been led to hood and then to dilution tunnel or flue gas transfer lines, and then sampled to the actual measurement (Chen *et al* 2005, 2006, 2009, Zhi *et al* 2008, Arora *et al* 2020, Li *et al* 2021), or the flue gas has been led to the chimney and sampled from there to measurement instruments (Heringa *et al* 2012, Fachinger *et al* 2017, Zhang *et al* 2021), or directly to the lines where the sample was taken to instruments and e.g. to smog chambers (Heringa *et al* 2011, Bertrand *et al* 2017). In certain experiments, the whole combustion appliance has been closed to the test chamber (Champion *et al* 2017, Li *et al* 2021). Shen *et al* (2012) have led the flue gas from the combustion to mixing chamber where the sampling to instruments was conducted. The BC containing flue gas has been sampled using a probe directly to dilution tunnel or sample line and by diluting and cooling the sample applying ejector diluters (Heringa *et al* 2011, 2012, Bertrand *et al* 2017, Nielsen *et al* 2017), FPS-4000 diluter (Dekati Oyj) (Sun *et al* 2017), porous tube diluter (Tissari *et al* 2019) or by axial diluter (Goetz *et al* 2018). E.g. in the study of Islam *et al* (2021), the flue gas has been sampled directly from plume 1–1.5 m above the cookstove or above the chimney's exit. Differences in BC EF results obtained by different measurement setups have not been extensively studied. However, Li *et al* (2021) have studied how the flue gas residence time in the measurement system affects the EFs, observing that the EC EFs were not systematically changed. From values provided by Li *et al* (2021), we included the

EFs measured with the shortest residence time (1 s) to this review.

BC detection techniques utilized in residential emission studies include MAAP (Heringa *et al* 2011, 2012, Fachinger *et al* 2017), AE (Zhi *et al* 2008, Heringa *et al* 2011, Bertrand *et al* 2017, Nielsen *et al* 2017, Goetz *et al* 2018, Tissari *et al* 2019), reflectometer (Johnson *et al* 2008), and SP-AMS (Nielsen *et al* 2017). The optical transmissometers (Quinones-Reveles *et al* 2021, Zhang *et al* 2021) and the thermal-optical carbon analyzers has been used to analyze EC from quartz fiber filters (Chen *et al* 2005, 2006, 2009, Sippula *et al* 2007, Zhi *et al* 2008, Shen *et al* 2010, 2012, 2015, 2020, Champion *et al* 2017, Tissari *et al* 2019, Islam *et al* 2021, Li *et al* 2021, Quinones-Reveles *et al* 2021). The integrating sphere method has also been used (Sun *et al* 2017) and, in addition, e.g. Arora *et al* (2020) have used in their studies a cell phone-based monitoring system, based on automated calculation of BC concentration from the flue gas exposed quartz filters.

In some cases, BC EFs have been reported for various combustion conditions (e.g. Nielsen *et al* 2017) and for different phases of the combustion process (warm start, flaming, smoldering, see e.g. Fachinger *et al* 2017). Standardized protocols have been used in some of the measurements (Bertrand *et al* 2017, Champion *et al* 2017, Li *et al* 2021); e.g. Li *et al* (2021) have studied the emissions using the water boiling test and Champion *et al* (2017) have used 'Standard Test Method for Determining Particulate Matter Emissions from Wood Heaters, ASTM test' (Cordwood Annex from the American Society for Testing and Materials (ASTM) E-2780).

The studied combustion appliances have been technically very different owing different technology levels; different types of cookstoves typical for developing countries (Arora *et al* 2020), open fire and patsari stoves used in Mexico (Johnson *et al* 2008), mud stoves, chimney stoves and several other appliances typical in Nepal (Goetz *et al* 2018), the residential wood stoves used in Navajo homes (Champion *et al* 2017), mud stoves, tandoor stoves, liquefied petroleum gas (LPG) stoves typical for northern and southern India (Islam *et al* 2021), different types of cooking and heating stoves used in China (Shen *et al* 2020, Zhang *et al* 2021), coal stoves used in China (Chen *et al* 2005, 2006, 2009, Shen *et al* 2015, Sun *et al* 2017, Zhi *et al* 2008, 2009), camp stoves (Quinones-Reveles *et al* 2021), brick wok stoves and movable cast-iron stoves (Shen *et al* 2010), log wood burners and wood pellet burners (Heringa *et al* 2011, 2012), biofuel gasifier stove (Shen *et al* 2015), wood stoves (Bertrand *et al* 2017, Fachinger *et al* 2017, Nielsen *et al* 2017), sauna stoves (Tissari *et al* 2019, Savolahti *et al* 2020, and references therein) and different stoves fueled by pellets (Sippula *et al* 2007, Shen *et al* 2012, Bertrand *et al* 2017).

Also, the fuel types and fuel qualities used in BC EF studies varied significantly from natural biomasses to processed bio-based fuels and different types of fossil fuels. Fuels have included wood, sticks, dung, mustard stalk (Johnson *et al* 2008, Champion *et al* 2017, Goetz *et al* 2018, Arora *et al* 2020, Islam *et al* 2021, Li *et al* 2021); logwood and wood pellets (Sippula *et al* 2007, Heringa *et al* 2011, 2012, Nielsen *et al* 2017, Bertrand *et al* 2017, Tissari *et al* 2019, Quinones-Reveles *et al* 2021; references in Savolahti *et al* 2020); different types of coal (anthracite, bituminous coal, raw coal and coal chunks, coal briquettes made from different raw coals) (Chen *et al* 2005, 2006, 2009, Zhi *et al* 2008, Sun *et al* 2017, Champion *et al* 2017, Goetz *et al* 2018, Zhang *et al* 2021); sawdust (Goetz *et al* 2018); biogas (Goetz *et al* 2018) and LPG (Islam *et al* 2021); rice straw, corn straws, corn stover, corn cobs, corn stalks, wheat straw, crop residues, biomass pellets, cotton stalks, branches of poplar and other wood, wood logs, sesame straws, soybean straws, and bamboo (Shen *et al* 2010, 2012, 2020, Li *et al* 2021, Zhang *et al* 2021).

3.5.2. Residential combustion emission factor studies

Based on this review, there are many experimental studies where the BC EFs have been measured and reported for residential combustion applications. A clear result of this review was that the variation of residentially used fuels and combustion appliances is large, depending e.g. on the purpose of the combustion (heating, cooking, saunas, etc.), locally available fuels and apparently also on economical and societal context. In addition, as mentioned above, no uniform or standardized methods exist in the studies, meaning that the procedures of combustion experiments, flue gas sampling systems and BC measurement techniques vary significantly. As a result of all these factors, the BC EFs of residential combustion have large variation.

We present the BC EFs here separately for fossil coal (figure 8) and renewable biomass (figures 9 and 10). Both fuel types include many different combinations of fuels, combustion appliances and combustion situations, and although the highest BC EFs were found for the coal combustion, the EFs varied even more than three orders of magnitude meaning that the fossil coal combustion in households cannot be said to be cleaner than the biomass combustion, from the viewpoint of BC emissions. In respect of total climate warming effects, it is clear that the CO_2 emissions from coal combustion are crucial and the coal combustion is more harmful for the climate than biomass combustion.

For the residential coal combustion, the measured BC EFs were in range 0.03–13.25 mg kg_{fuel}^{-1} with average, median and standard deviation being 1.08, 0.208 and 2.38 g kg_{fuel}^{-1}, respectively (figure 8, table S7). We observed that the quality of the coal significantly affects the BC emissions; this can be seen

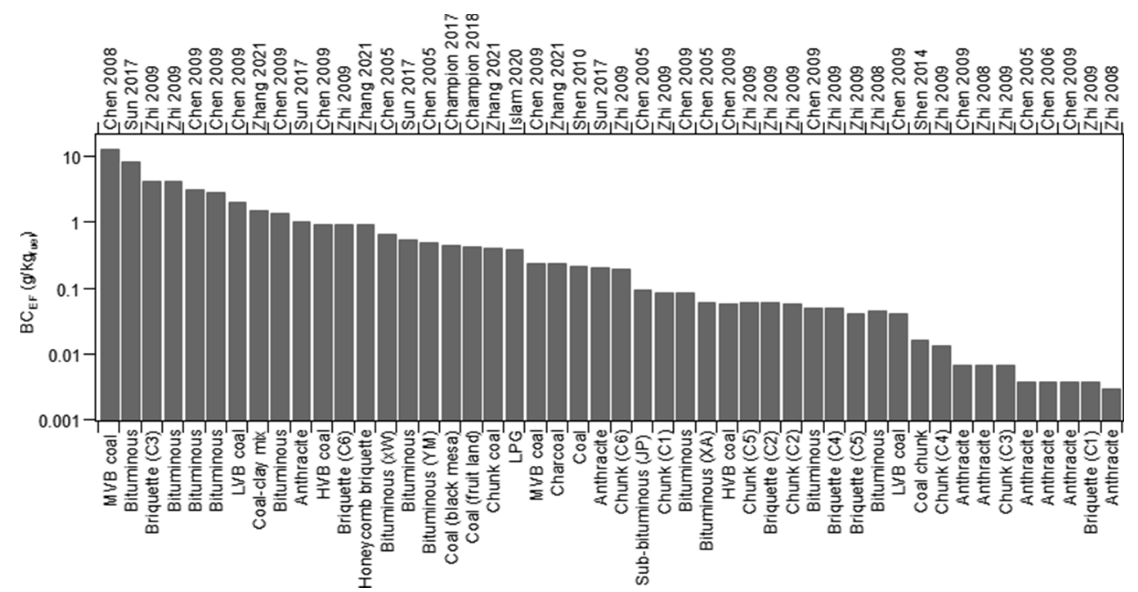

Figure 8. The BC EFs (g kg^{-1} fuel) for residential fossil fuel combustion. In most of the studies the fuel was different coal types and variety of different residential coal stoves were studied (see table S7 for burning device). In the Islam *et al* (2021), the burning device was LPG burner and in Shen *et al* (2015) the burning device was a gasifier stove.

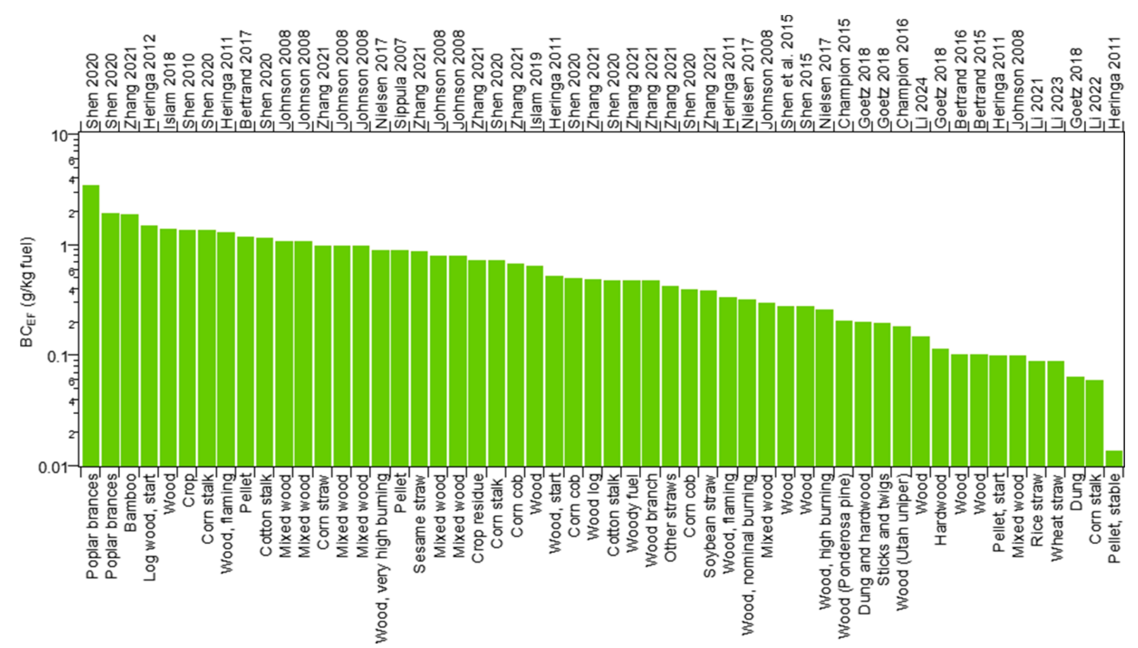

Figure 9. BC EFs for residential combustion (g $kg_{fuel}{}^{-1}$) when burning variety of different biomass-based fuels. The BC EF values and devices are given in table S8 and figure S2.

especially by comparing the BC EFs from combustion of bituminous coal with anthracite coal (figure 8): these two qualities of coal have different composition (bituminous coal contains significantly tar-like substances while the anthracite coal is mostly EC) and their heating values are different, approximately 33 MJ kg^{-1} for anthracite and >27 MJ kg^{-1} for bituminous coal. In addition, the other clearly influencing factor was the form and structure of coal, so that the BC EFs were relatively high for the raw coal and the chunk coal, while the emissions factors were significantly lower for the coal briquettes. Furthermore, the design of the combustion device affected the BC emissions; for example, the study of Zhi *et al* (2009) showed that the BC EFs for similar coal were significantly lower with improved stove when compared to normal residential stove.

The variation of the fuel quality was in the studies of BC EFs from residential biomass combustion even larger than in the studies focusing on residential coal combustion. For the residential biomass combustion, the measured BC EFs were in range

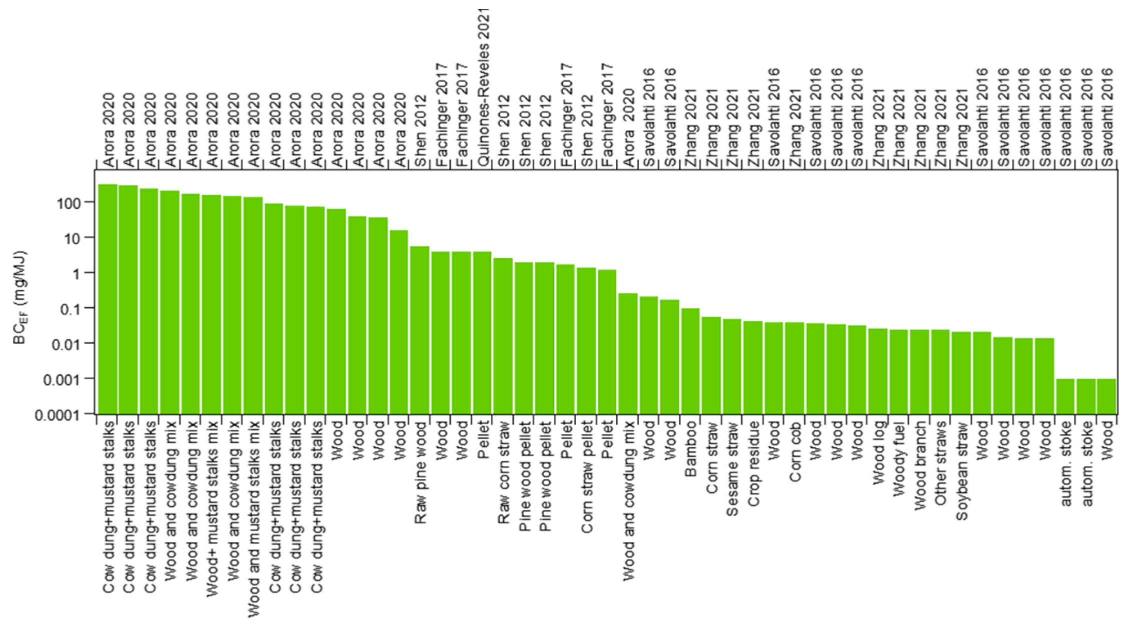

Figure 10. BC EFs for residential combustion (g $kg_{fuel}{}^{-1}$) when burning variety of different biomass-based fuels. The BC EF values and devices are given in table S8 and figure S3.

0.014–3.51 g $kg_{fuel}{}^{-1}$ with average, median and standard deviation being 0.68, 0.49 and 0.63 g $kg_{fuel}{}^{-1}$, respectively (figures 9, 10 and table S8). For the studies where the energy content was reported instead of fuel mass, the measured BC EFs were in the range of 0.001–316 mg MJ^{-1} with average, median and standard deviation being 44, 1.2 and 82 mg MJ^{-1}, respectively. As mentioned above and seen in figure 9, biomass fuels varied from cow dung and straws to different wood fuels. In addition, e.g. BC EFs from wood combustion have been studied varying both the wood types and structures, log wood and pellets being the most typical studied wood fuels. As a result of this variation, also the reported EFs varied significantly, from values approximately 0.001 mg MJ^{-1} to the values close to 10 mg MJ^{-1}. Interestingly, the review indicated that the type of fuel (e.g. log-wood vs. wood pellet) was not the most important factor affecting the BC emissions from combustion; instead, the design of combustion appliance seemed to affect much more, which can be seen e.g. by comparing the results for wood pellet combustion presented by Shen *et al* (2012) and Sippula *et al* (2007).

Regarding the future studies on BC emissions from residential combustion, this review study indicated that it is important to report the EF values in the units which enable correct comparisons with other studies. The current literature reports the EFs typically normalized by heating value of the combustion or by mass of fuel. Unit conversions and the correct comparison require typically the information on heating value of the fuel but also water content of the fuel. We did not make the unit conversions in this study in order to avoid the decrease of accuracy; however, large number of fuel specific heating values are available in the literature.

3.6. Power and heat generation

Due to their relatively high contribution to anthropogenic atmospheric emissions in general, it could be assumed that also large-scale production of electricity and heat would emit substantial amount of BC. In large power plants, the typical fossil fuels include coal, oil, natural gas and sometimes peat. In recent decades power plants have been designed and constructed also for waste fuels and biomasses. However, the information on BC EFs from large scale power and heat production units is scarce.

The BC EFs of large scale power plants have been reported only by two articles (Mylläri 2018, 2019). Those studies measured the BC for from coal-fired combined heat and power (CHP) plant with an AE AE33 (Drinovec *et al* 2015) after direct sampling of flue gas from flue gas duct followed by controlled dilution and cooling of the flue gas before the measurement instrument. The reported BC EFs for the power plant, in normal situation equipped with electrostatic precipitators (ESPs) and combined unit for flue-gas desulphurization (FGD) and bag house filters (BHF), were 3.6 mg MJ^{-1} when measured after ESP but before the FGD and BHF, and 0.014 mg MJ^{-1} when measured after all flue-gas cleaning systems. From these values, the latter EF represents the emission to the atmosphere. BC EFs from the same CHP plant but fired with the mixture of industrial pellets and coal (share of pellets was 10.5%) were 11.74 mg MJ^{-1} when measured after the ESP but before the FGD and BHF, and 0.014 mg MJ^{-1} after the flue-gas cleaning systems, the latter representing again the atmospheric emission of the studied power plant (Mylläri *et al* 2019). The BC EFs for power and heat generation have been collected to table S8.

3.7. Discussion

Based on the review results above, a significant amount of literature about the BC emissions and BC EFs has accumulated over the past decades. The literature contains EFs for several categories of anthropogenic emission sources. However, it was seen that while e.g. the vehicle emissions and residential combustion emissions have been studied relatively widely, e.g. the BC emission factor information related to large scale combustion applications is presented only in very few scientific publications. In addition, the comparability of the measurement results is often difficult due to the differences in BC measurement methods; large variety of measurement devices based on optical, thermal and other measurement technologies, as well as instruments designed for atmospheric studies (requires dilution when applied in tailpipe emission measurements) and emission studies were used for BC and EC quantification. Furthermore, the EFs have been calculated using different approaches and methods.

BC EFs are often presented in different units e.g. mg km^{-1} (vehicles), mg MJ^{-1} (energy production and shipping), mg $kg_{fuel}{}^{-1}$ (aviation and residential combustion). Most of the unit formats are logical and justified based on the application and applicability of the EFs. The most contradictory application is residential combustion, where the literature used either mg $kg_{fuel}{}^{-1}$ or mg MJ^{-1}. Both the different measurement methods and conversion to different units can increase the uncertainty of BC EFs found in peer-reviewed literature and complicate the subsequent use of values in the inventories (see e.g. Meyer 2012) and modeling.

Our review highlights the importance of society level emission regulation in BC emission mitigation; especially in North America, a clear BC emission reduction was observed in long-term trend of ambient studies for road traffic influenced by HD diesel vehicles. This can be seen as a consequence of strict emission regulations forcing the use of DPFs in vehicles. In general, the DPFs remove efficiently the BC particles from engine exhaust which was seen in studies where the BC EFs were determined by direct tailpipe measurements of individual diesel-powered vehicles. The gasoline particle filter will likely also reduce the BC EFs of gasoline vehicle fleet in near future. However, we note that vehicles with broken or removed particle filters may be important source of BC emissions also in future as well as old vehicles that remain in the fleet for decades. These old or malfunctioning vehicles should be included in emission inventories, and in terms of BC emission mitigation, removing these vehicles from traffic is generally seen as an effective and efficient strategy to reduction on-road traffic BC emissions (see e.g. Preble *et al* 2018, Boveroux *et al* 2019). Importantly, the progress in vehicle technologies seems to produce opposite trend in gasoline vehicle fleets after c. 2010; the BC emissions were observed to be higher for gasoline vehicles with direct fuel injection (GDI) than for vehicles with PFI being opposite to the trend in CO_2 emissions with these technologies.

Regarding the BC EFs from residential combustion, we observed large variation in EFs, indicating that fuel type and quality as well as combustion appliances influence significantly to BC EFs. Some of the measurements were focusing on emissions of open fires places, whereas in other studies the emissions were measured from the combustion appliance either in laboratory or the or at the household either inside or at the top of chimney. Variation was large in experimental setups so that some of the studies were done in controlled and repeatable manner and some of the studies studied BC emissions of households in their everyday life. Also, the used fuels varied from different forms of coal to wood, biomass residues (sticks, hay etc) and dung. However, due to lack of binding regulation and lack of aftertreatment systems suitable for household use, the decrease in BC EFs can be achieved mainly by improved and more efficient combustion technologies and by correct fuel choices and adaption of clean burning procedures by individuals.

In the case of shipping, a relatively clear correlation was seen between the engine size and BC EFs so that the fuel specific BC EFs of the largest engines were the lowest. This result as well as the observations related to the effects of marine engine fuels on BC emissions should be taken into account e.g. when the IMO considers a limitation to BC emissions of ships, as well as when the engine manufacturers are developing technologies with lower atmospheric emissions and lower climate impact. Engine load (thrust) and fuel affected also the BC EFs of aircraft engines, so that the highest values were at idle and again at high thrusts corresponding to cruising, climb-out and take-off. Regarding aviation, synthetic fuels were reported to reduce BC EFs compared to traditional kerosene fuels. Thus, taking the challenges of exhaust cleaning systems into account, especially the fuel related technological development could diminish the BC emissions and emissions' impacts both from marine traffic and aviation.

Although the BC EFs have been reported in several publications and for large number of combustion aerosol sources, it is hard to say how the EFs have developed in certain geographical areas; e.g. Africa and South America are underrepresented in BC EF studies, both related to the individual BC sources and ambient studies. Globally, the largest data gaps were in BC EFs of large-scale energy production which can be seen crucial for estimating global radiative forcing potential of anthropogenic BC emissions. Only a few articles contained EFs measured

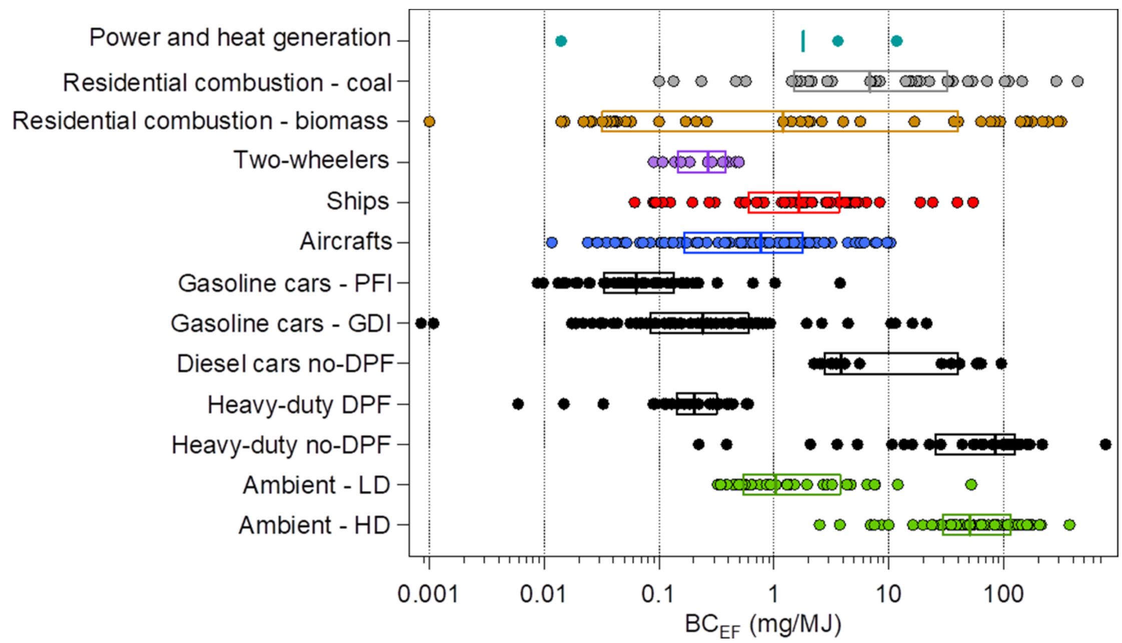

Figure 11. Individual BC EFs of different anthropogenic sources (circles), median values for each subcategories (vertical lines), and 25–75 percentiles (boxes) of the values. Percentiles were not calculated for power and heat generation as there were only four values available. More detailed information can be found from the sections above, from tables presented in supplementary material, and from original articles.

in the full-scale energy production site. These studies were for modern power plants. Thus, it would be utmost important to increase the knowledge on emissions from different power plants with different ages and technologies, such as fuel and flue gas cleaning devices. In addition, much more research is needed to improve the global coverage of BC EFs of large-scale power and heat production.

In addition to the amount of BC emissions, also the location of the BC emission is an important factor when the health and climate impacts of atmospheric BC are considered. E.g. the road traffic BC emissions take typically place in the vicinity of people, and thus the decrease of BC EFs of diesel vehicles and road traffic in general is likely reducing significantly the BC concentrations and exposure of citizens to BC in urban areas (e.g. Luoma *et al* 2021). Regarding power plants and several industries, high stacks of power plants and more distant locations reduce their role in respect of human health but, however, not totally solving that problem. When considering the climate impacts of BC emissions, the geographical location (near the snow-covered areas) and the season (winter vs summer) are more important. The BC EF information collected here is applicable e.g. for future simulation or modeling studies where the focus could be, for instance, to understand atmospheric processes affecting the optical processes of atmospheric aerosols, or the effects the BC can have on climate warming e.g. when deposited on ice and snow cover. Anyway, this review showed that the BC emission can be reduced technologically and as indicated by decreases in BC EFs seen especially in North America and Europe, the technological development and the utilization of low-emission techniques can be accelerated by emission regulation, i.e. by society level actions.

4. Summary and conclusions

Figure 11 presents a summary of the BC EFs found from the literature for different sectors. The differences in observed EFs were large, even up to six orders of magnitude within the same category depending on the combustion device, fuel, and aftertreatment systems. It is important to notice that the BC EFs of diesel passenger cars are not included in the figure 11, because of the underrepresented share of DPF-equipped vehicles in scientific literature concerning BC EFs. However, we think that the BC EFs of HD vehicles demonstrate relatively well the BC EFs of diesel vehicles in general, i.e. the EFs have large variation and they are significantly affected by filtration.

BC is an important pollutant and significant efforts need to be done in near future in order to increase the understanding of the sources of BC, most efficient mitigation methods as well as its impacts to climate, air quality and health. By including accurate BC EFs in emission inventories as well as in climate and health models, it would be easier to effectively mitigate the adverse impacts and subsequent societal costs caused by BC emissions. For that purpose, it is utmost important to collect and maintain accurate and up-to-date BC emission factor information. The trends observed in BC EFs are important information both for the development of combustion and flue gas cleaning technologies as well as to politicians in charge of setting emission limits and air quality guidelines.

Future research needs include expansion of spatial and temporal variation of BC EF studies of power and heat generation especially to include different types of power plants, studies on modern vehicles as well as contribution of non-exhaust sources (e.g. brakes). Furthermore, this review indicates that it would be utmost important to focus research and technological development on residential combustion to find potential ways to mitigate residential combustion emissions; those emissions have potential to significantly influence human health and, on the other hand, the current technological level is not at the level of many other combustion applications. Due to the variability of measurement techniques, more studies are needed comparing the results of different BC instruments, to improve comparability of the results from different research fields and to diminish the measurement related uncertainties of the climate and air quality related evaluations. It is also clear that the BC concentrations will be monitored more in the future, which increases the need for reliable and user-friendly devices. However, in health effect assessment and climate studies, it is important to conduct comprehensive physical and chemical characterization of BC containing aerosols; e.g. particle size resolved EFs and information on co-emitted species and other light-absorbing species are important when evaluating the climate impacts of BC. This kind of additional information linked with BC EFs could efficiently improve utilization of results of BC emission research.

Data availability statement

No new data were created or analyzed in this study.

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