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Review article

Wet deposition of black carbon: A synthesis

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ABSTRACT

Black carbon (BC) influences the global radiation budget and adversely affects air quality and human health. Removal of BC from the atmosphere can mitigate these effects. Although wet deposition (rainfall) is thought to be a dominant pathway for BC removal over land and oceans, studies quantifying rainfall BC concentrations and deposition are scarce. Here, we synthesize empirical measurements of BC in rainfall conducted between 1984 and 2016 from sites around the world. Data from 20 peer-reviewed publications and 42 unique sites were compiled. All observations were from the northern hemisphere, with the vast majority located in mid-latitudes with medium to high annual rainfall. Differences in rainfall BC concentrations and deposition among urban, rural, and remote sites were examined. Annual rainfall BC concentrations ranged 120-fold, from 2.8 μg L^{-1} at a remote site to 333 μg L^{-1} at an urban site, while annual BC deposition ranged 16-fold from 0.014 mg m^{-2} d^{-1} at a remote site to 0.22 mg m^{-2} d^{-1} at an urban site. The small number of observations, high measurement variability, and location relative to coast appeared to mask the effects of land use on BC in rainfall. However, characteristic seasonal patterns in BC concentrations and/or deposition were found for geographic regions with similar rainfall patterns, BC emission sources, and air mass trajectories. Annual rainfall BC deposition across sites was comparable to modeled estimates for the global oceans, but BC concentrations were somewhat higher than those measured in surface snow at central North American sites. Neither tropical regions nor urban areas (BC-emission hotspots) were well represented in the dataset, highlighting fundamental gaps in understanding of rainfall BC deposition. More extensive monitoring of rainfall BC deposition is imperative to develop improved global climate and carbon cycling models.

1. Introduction

Atmospheric black carbon (BC) is a minor constituent of the atmosphere with major significance for climate, air quality, and human health (Bond et al., 2013; Gustafsson and Ramanathan, 2016; Magalhaes et al., 2018; Meng et al., 2018; Zhang et al., 2018). Black carbon affects Earth's radiative balance by directly absorbing incoming solar radiation and has a total radiative forcing value of +1.1 W m^{-2}; of all anthropogenic pollutant emissions, only CO_2 has a greater estimated forcing (Bond et al., 2013). In addition, more than 90% of atmospheric BC exists as fine particulate matter with an aerodynamic diameter less than 2.5 μm ($PM_{2.5}$), contributing to particle pollution in both urban and fire-affected regions (Bond et al., 2013; Gao et al., 2018; McClure and Jaffe, 2018; Peng et al., 2016). These suspended BC particles have been shown to have negative effects on health in utero (Rychlik et al., 2019), adult health including lung and cardiovascular disease (Nichols et al., 2013; Ostro et al., 2015), and possibly decreased cognitive function in children (Suglia et al., 2007).

Black carbon is a primary pollutant emitted via fossil fuel and biomass combustion processes. Globally, biomass burning is the largest source of atmospheric BC, while fossil fuel combustion in diesel vehicles is the primary source in urban areas (Bond et al., 2013; Madhavi Latha and Badarinath, 2003). In the atmosphere, BC is distinguished by: (1) strong absorption of light in the visible spectrum; (2) its refractory nature, retaining its basic form at very high temperatures; (3) insolubility in water and organic solvents, including methanol and acetone; and (4) manifestation as aggregates of small carbon spheres (Bond et al., 2013). Although organic compounds can sorb to BC particles during atmospheric transport, BC is considered a useful tracer of anthropogenic air pollution given the chemical stability of the BC core (Andreae, 1983; Chen et al., 2012; Wang et al., 2011). Black carbon also has a short, one- to two-week atmospheric lifetime. As such, reducing

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BC emissions and increasing BC removal from the atmosphere can have a significant impact on near-term climate change and human health (Grieshop et al., 2009).

Once emitted into the atmosphere, wet (i.e., rain, snow, sleet, hail) and dry deposition are the only removal pathways for BC (Begam et al., 2016; Bibi et al., 2017). Dry deposition involves direct deposition of BC to receptor surfaces and in most models accounts for 15%–40% of total (wet + dry) BC deposition (Textor et al., 2006). Factors controlling dry BC deposition include particle size, aerodynamic resistance, and surface type (Liu et al., 2011; Wu et al., 2018). Wet deposition involves incorporation of BC into water droplets through two known mechanisms: nucleation scavenging and impact scavenging. Impact scavenging occurs when BC particles collide with water droplets as they fall through the air (Liu et al., 2013; Ohata et al., 2016). Nucleation scavenging is the process whereby aerosols act as cloud condensation nuclei (CCN) and seed the growth of cloud droplets (Ching et al., 2018; Ohata et al., 2016). Freshly emitted BC particles, especially from diesel emissions, are hydrophobic and not readily available to act as CCN (Ching et al., 2018; Liu et al., 2013; Weingartner et al., 1997). However, the ability of BC particles to act as CCN increases during atmospheric transport through condensation with hygroscopic particles, coagulation with more soluble particles, and through oxidation of coatings on the particles (Vignati et al., 2010).

Currently, wet deposition is thought to be the dominant pathway for BC removal from the atmosphere (Kondo et al., 2016; Taylor et al., 2014). Because wet removal influences the atmospheric lifetime of BC, and thus its distribution and dispersion in the atmosphere, better understanding of wet BC deposition is vital to resolve BC effects on local, regional, and global climate (Liu et al., 2013) and air quality. In addition, improved estimates of wet BC deposition rates are critical to constrain pools and fluxes of BC in terrestrial and aquatic ecosystems. Here, we synthesize empirical measurements of BC in rainfall specifically, from around the world with the following objectives: 1) quantify the mean and range of reported rainfall BC concentrations and deposition rates; and 2) examine differences in rainfall BC concentrations and deposition rates among urban, rural, and remote locations. Our synthesis focused on rainfall BC in terrestrial systems complements previous studies conducted on BC deposition to the global oceans (Jurado et al., 2008) and snow-dominated regions (Doherty et al., 2010, 2014; Hadley et al., 2010).

2. Controls on atmospheric wet BC deposition

Several factors affect the wet (i.e., rain, snow, sleet, hail) deposition of particles to the Earth's surface. These factors include emissions source strength and proximity to source areas, meteorology and climate, and particle characteristics (Kondo et al., 2016; Weathers and Ponette-González, 2011).

The influence of source areas—areas actively emitting BC (e.g., industrial sites, roadways, active burn sites)—on wet BC deposition depends on particle emissions as well as on dispersion. In general, atmospheric BC concentrations are highest close to emission sources and decline exponentially as distance from source increases, with concentrations decreasing by as much as one-half anywhere from 50 to 150 m from the source (Polidori et al., 2010; Reponen et al., 2003; Zhu et al., 2002). However, wind dispersion directly affects atmospheric particle concentrations, and thus can contribute to either lower, or higher, concentrations in precipitation (Ouyang et al., 2015). For example, in Beijing, China, $PM_{2.5}$ concentrations in rainfall were lower when meteorological conditions caused increased dispersion of particles away from sources than when dispersion was low (Ouyang et al., 2015). Globally, most BC particles are thought to be removed by wet deposition near source areas within several days of transport (Oshima et al., 2012; Wu et al., 2018).

Although precipitation characteristics (e.g., amount, duration, intensity) influence atmospheric wet BC concentrations, the direction of these effects remains unclear. Studies report increasing, decreasing, and no change in concentration with variations in storm characteristics (Chýlek et al., 1999; Ogren et al., 1984; Witkowska et al., 2016). For example, a study conducted in Poland found that BC removal by rain and snow is most efficient when precipitation lasts longer than one day, with BC concentrations increasing with precipitation duration (Witkowska et al., 2016). In contrast, concentrations of BC in rainfall sampled in Nova Scotia did not change as a function of rainfall duration or with time from the beginning of the storm (Chýlek et al., 1999). Black carbon concentrations have also been found to vary with storm intensity. An early study in Seattle, U.S., showed that BC concentrations were higher during light rainfall and lower during more intense rainfall events (Ogren et al., 1984). The lack of conclusive evidence regarding the influence of storm duration and intensity on BC removal by rainfall supports studies suggesting that rainfall BC deposition is mainly due to nucleation processes (Ching et al., 2018; Ohata et al., 2016; Taylor et al., 2014), whereas impact scavenging may be more important in the case of snow BC deposition (Hadley et al., 2010).

The size, hygroscopicity, and mixing state of BC particles affect wet BC concentrations and deposition from the atmosphere as well. In Japan, BC particles in precipitation were primarily in the size range of 100–500 nm (Ohata et al., 2013), but can be larger when removed by impact scavenging. In the latter case, deposition is most efficient for particles/aggregates larger than 2 μm, as these are more easily intercepted by falling droplets (Andronache, 2003). The mixing state of BC, particularly with respect to the presence of sulfate coatings (which increase hygroscopicity), is an important factor determining the rate of deposition (Ogren and Charlson, 1984; Ohata et al., 2011). Freshly emitted BC is highly hydrophobic but undergoes transformations in the atmosphere during a process known as ageing. During the ageing process, BC particles shift from a hydrophobic to a more hydrophilic state due to condensation of soluble materials onto the particles, coagulation of BC with more soluble particles, and oxidation of organic materials that coat the particles (Vignati et al., 2010). In coupled atmospheric chemistry and climate models, the ageing process is approximated to be in the range of 1–2 days. However, in reality, the conversion rate of BC from hydrophobic to hydrophilic is not constant, introducing large uncertainties into modeled simulations of wet BC deposition (Liu et al., 2011). Finally, it has been hypothesized that emission source type could affect BC deposition via precipitation, with non-fossil BC (i.e., biomass-derived) more efficiently removed than fossil BC due to the presence of hydrophilic compounds coating non-fossil BC (Zhang et al., 2015). However, radiocarbon analysis of BC extracted from rainwater in Portugal showed that contributions of fossil and non-fossil sources to BC in rainfall were equal, with non-fossil sources accounting for 52%–58% of total BC (Zhang et al., 2015). In sum, wet BC concentrations and deposition are controlled by a combination of factors.

3. Material and methods

3.1. Literature search and coding process

To synthesize empirical measurements of rainfall BC concentrations and deposition, we conducted a literature search using Web of Science. The Boolean string “elemental carbon” OR “black carbon” AND “wet deposition” AND “rain*” was used to search for peer-reviewed publications. This search resulted in a total of 3940 articles. The title and abstract of each article were reviewed to determine if the article contained: 1) data on rainfall BC concentrations or BC deposition; 2) empirical rather than modeled data; and 3) measurements of more than one rainfall event per site.

A total of 25 publications were identified. Only 20 publications met the selection criteria and were included in the synthesis (Table S1). Separate studies conducted at the same site were included in the dataset if the measurements occurred in different land uses or during different time periods. For each selected publication, the following variables

were recorded: geographical location of the sampling site (i.e., coordinates, elevation (m asl), distance to coast (km)), land use (i.e., urban, rural, remote), location relative to coast (i.e., oceanic, coastal, inland), mean annual rainfall (mm) or annual rainfall (mm), collection year and month, collection season, sample size, sample water volume, rainfall BC concentration, rainfall BC deposition and, where available, atmospheric BC concentration (μg m^{-3}) and scavenging ratio. Where both rainfall and atmospheric BC concentration were reported, but the scavenging ratio was not, the volume-based BC scavenging ratio was calculated as:

$$\omega_v = C_p \, x \, \rho_w / C_a$$

where C_p is the concentration of BC in rain (μg kg^{-1}), ρ_w is the density of water (kg m^{-3}), and C_a is the concentration of BC in air (μg m^{-3}; Kasper-Giebl et al., 1999).

Geographic coordinates and Google Earth were used to obtain information on elevation and distance to coast for sites lacking these data. Distance to coast measurements were used to define sites as oceanic (i.e., islands), coastal (sites ≤10 km from open water), and inland (sites > 10 km from open water). Sites not classified as urban, rural, or remote in the original publication were assigned a land use by using distance from densely populated areas as a proxy (i.e., urban = within an urban area, rural = outside of an urban area, remote = isolated from population centers). For collection season, latitude was used to separate tropical (0–23.5°N) from temperate sites (23.5–66.5°N). For tropical sites, seasons were classified as either wet or dry, and for temperate sites, collection month was used to assign the sampling period to spring (Mar–May), summer (Jun–Aug), fall (Sep–Nov), or winter (Dec–Feb).

Collection method, analytical method, and species measured were also recorded. For rainfall collection, wet-only collectors refer to devices that open automatically when rainfall begins and close when rainfall ends. Bulk collectors remain open to the atmosphere between rainfall events. To determine BC concentrations in rainfall, various methods and instruments are available, including but not limited to the single particle soot photometer (SP2) and the laboratory organic carbon/elemental carbon (OC/EC) analyzer utilizing thermal optical transmission (TOT) and thermal optical reflectance (TOR) methods. The SP2 measurement is based on laser-induced incandescence while the OC/EC method is based on the thermal and optical properties of carbon. Petzold et al. (2013) recommend a nomenclature system based on method of detection of "black carbon", however, in this synthesis, the term "black carbon", or BC, will be used regardless of analytical method to avoid confusion.

3.2. Calculations and data analysis

Rainfall BC concentrations and deposition rates were recorded in the units presented in the original publication. If BC concentrations and deposition were included in a graph, but the actual values were not reported in the text, the GETDATA Graph Digitizer 2.26 software was used to determine numerical values. Data extraction from figures was conducted for Sase et al. (2012) and Zhang et al. (2015).

Some studies reported data for samples collected sequentially during a single rain event as individual values. In these instances, a volume-weighted mean (VWM) of the sequential samples was calculated using the following equation:

$$VWM = \frac{\sum (BC\ conc \times precip)}{\sum precip}$$

where BC conc (μg L^{-1}) represents the reported rainfall BC concentration and precip (L) represents the water volume of each individual sample.

If rainfall BC concentration and water volume were reported for a site, but deposition was not reported, then deposition rate (mg m^{-2} d^{-1}) was calculated as

$$Rainfall\ Deposition = \left(\frac{BC\ conc\ (\mu g\ L^{-1}) \times precip\ (mm)}{1000} \right) \Big/ sample\ days$$

We analyzed the dataset for differences in rainfall BC concentrations and deposition rates among urban, rural, and remote sites on both annual and seasonal timescales. Studies consisting of at least nine months of consecutive sampling were considered to provide sufficient data for estimates of annual deposition. If the average concentration for two distinct seasons (e.g., wet season and dry season; Ducret and Cachier, 1992) was reported in a publication, the median was calculated and used as an annual mean and included in the annual analysis (after Iavorivska et al., 2016). Several studies reported annual and seasonal values for rainfall BC concentration and deposition. These studies were included in both the annual and seasonal analyses. Studies with no annual data or clear seasonal definition were not included in the summary statistics. To compare rainfall BC concentrations, all concentration measurements were converted to μg L^{-1}. Deposition rates were converted to mg m^{-2} d^{-1}. Additional details on unit conversions and calculations can be found in Table S1.

Mean ± standard error, medians, and ranges were used to assess the average and variation in BC concentration and deposition within and among land use classes. To determine if there were annual or seasonal differences among urban, rural, and remote sites, or if there were seasonal differences within a land use class, a Mann-Whitney test or a Wilcoxon Each Pair nonparametric test was used depending on the number of sites per class ($\alpha = 0.05$). All calculations and statistical analyses were performed using JMPv14.

4. Results

4.1. Dataset on rainfall BC concentrations and deposition

Of the 20 studies included in the synthesis, data were compiled for 42 unique sampling sites across the northern hemisphere (Fig. 1). The majority of these sites were located in the mid-latitudes (23.5–66.5°N), with just five sites in the tropics (0-23.5°N) and no sites north of the Arctic Circle. Urban sites comprised 26%, rural sites 62%, and remote sites 12% of the locations sampled (Table 1). In addition, 62% of the sites in the dataset were classified as inland, with a smaller proportion of coastal (26%), and oceanic (12%) sites. All but three sites were low in elevation (< 500 m) and most of the sites for which annual rainfall data were available were relatively wet (> 800 mm).

From the standpoint of time of sampling, twice as many sites were sampled during spring and summer ($n = 24$) compared to fall and winter ($n = 11$). This was due to a major sampling campaign conducted in Sweden by Ogren and Charlson (1984). Few sites had measurement periods spanning one to several years (Table 2).

There was some variation in collection and analytical methods used in the measurement of BC in rainfall. Bulk and wet-only collectors were each deployed at about half of the study sites, while thermal-optical methods were the most commonly used approach for quantifying BC (~80% of sites). The SP2 was employed at one site (Mori et al., 2014), while at two sites BC concentration was determined using a photometer at 528 nm (Budhavant et al., 2016; Granat et al., 2010). Other methods were employed at six sites and included an elemental analysis system (Hwang et al., 2004), a thermal/titration method (Ducret and Cachier, 1992), and a thermal-optical method employing a spectrometer and LiCOR CO_2 analyzer (Hadley et al., 2010).

4.2. Rainfall BC concentrations

Annual rainfall BC concentrations ranged ~120-fold, from 2.8 μg L^{-1} at a remote oceanic site to 333 μg L^{-1} at an urban inland site (Fig. 2a). The annual mean BC concentration for urban sites was 113 ± 74 μg L^{-1} (median = 50 μg L^{-1}), approximately 2.5-fold and 7.5-fold higher compared to annual BC concentrations for rural

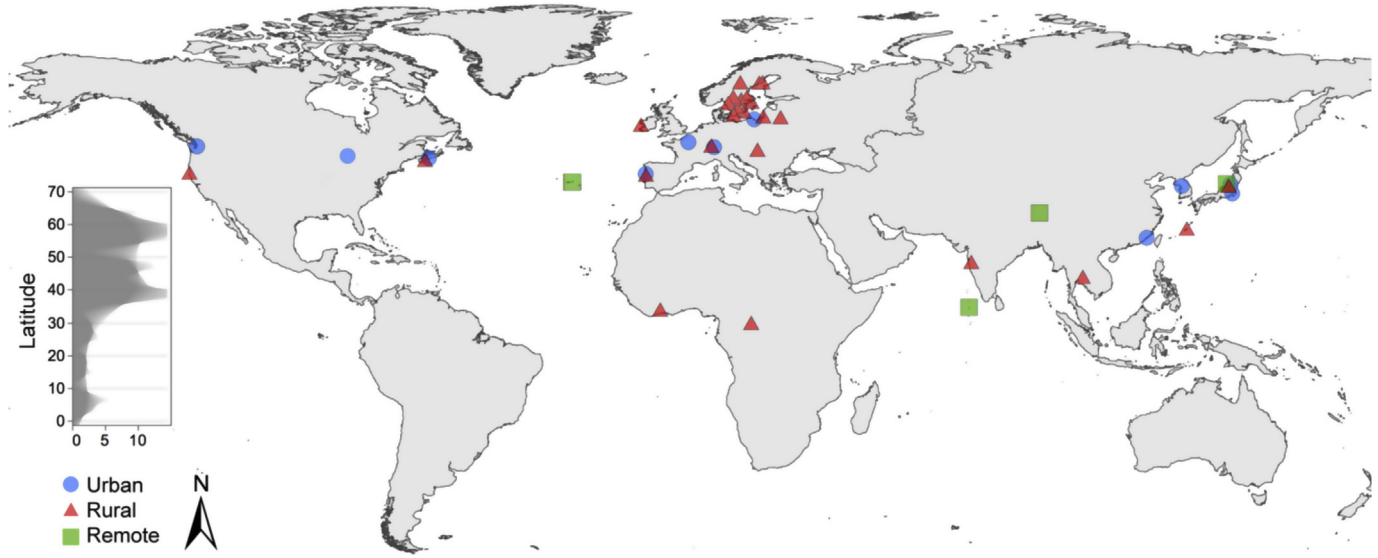

Fig. 1. Geographical location of 42 urban, rural, and remote sites included in this synthesis where measurements of black carbon in rainfall were conducted between 1984 and 2016. The latitudinal distribution of sites is shown on the left.

(mean = 46 ± 14 μg L^{-1}; median = 28 μg L^{-1}) and remote (mean = 15 ± 6.7 μg L^{-1}; median = 15 μg L^{-1}) sites, respectively (Fig. 3). Rainfall BC concentrations measured in Paris, France, strongly influenced the annual mean for urban sites. When this site was excluded, the annual mean BC concentration was 40 ± 19 μg L^{-1}, similar to that for rural sites. On an annual basis, there were no significant differences in BC concentrations among land use classes.

No differences in rainfall BC concentrations were detected among urban, rural, and remote sites during summer, fall, or winter. A statistically significant difference among land use classes was detected in spring, when concentrations were higher at rural compared to urban sites (p = 0.0079; Fig. 3).

The urban sites did not exhibit significant seasonal differences in rainfall BC concentrations, with mean values ranging from 24 ± 13 μg L^{-1} in fall (median = 19 μg L^{-1}) to 69 ± 29 μg L^{-1} in winter (median = 25 μg L^{-1}; Fig. 3). In contrast, at rural sites, mean rainfall BC concentrations ranged 17-fold among seasons. Rainfall BC concentrations were higher in spring (mean = 237 ± 51 μg L^{-1}; median = 200 μg L^{-1}) compared to summer (mean = 94 ± 21 μg L^{-1}; median = 83 μg L^{-1}; p = 0.048), fall (mean = 14 ± 6.3 μg L^{-1}, median = 11 μg L^{-1}; p = 0.0079), and winter (mean = 19 ± 8.01 μg L^{-1}; median = 24 μg L^{-1}; p = 0.018). Summer rainfall BC concentrations were also higher than in fall (p = 0.024). At the remote sites, differences among seasons were not significant, with mean BC concentrations ranging from 9.4 ± 3.9 μg L^{-1} during fall (median = 12 μg L^{-1}) to 28 μg L^{-1} during winter (two sites only). Mean rainfall BC concentrations at rural tropical sites were 102 ± 46 μg L^{-1} (median = 69 μg L^{-1}) and 155 μg L^{-1} (one site) during the wet and dry seasons, respectively. At the sole tropical remote site, concentrations were lower: 24 μg L^{-1} during the wet and 84 μg L^{-1} during the dry season.

4.3. Rainfall BC deposition

Annual rainfall BC deposition ranged ~16-fold, from 0.014 mg m^{-2} d^{-1} at a remote oceanic site to 0.22 mg m^{-2} d^{-1} at a rural inland site (Fig. 2b). Annual mean deposition for the urban sites was 0.15 ± 0.28 mg m^{-2} d^{-1} (median = 0.16 mg m^{-2} d^{-1}). At rural and remote sites, mean deposition was 0.11 ± 0.029 mg m^{-2} d^{-1} (median = 0.096 mg m^{-2} d^{-1}) and 0.044 ± 0.17 mg m^{-2} d^{-1} (median = 0.047 mg m^{-2} d^{-1}), respectively (Fig. 4). On an annual basis, there were no significant differences in rainfall BC deposition among land use classes.

Rainfall BC deposition in urban areas ranged 5.5-fold from 0.1 mg m^{-2} d^{-1} in the fall to 0.55 ± 0.15 mg m^{-2} d^{-1} (median = 0.41 mg m^{-2} d^{-1}) in the winter (Fig. 4). Differences among spring, summer, and winter seasons were not significant. Rural site averages ranged 37-fold with fall deposition low (mean = 0.0076 mg m^{-2} d^{-1}) compared to spring (mean = 0.28 ± 0.067 mg m^{-2} d^{-1}; median 0.13 mg m^{-2} d^{-1}), summer (0.18 ± 0.022 mg m^{-2} d^{-1}, median = 0.20 mg m^{-2} d^{-1}) and winter (mean = 0.15 ± 0.028 mg m^{-2} d^{-1}). Mean deposition values for spring and summer seasons were similar at these rural sites. Rainfall BC deposition at the only remote site where seasonal measurements were conducted ranged 42-fold from 0.002 mg m^{-2} d^{-1} in the summer to 0.085 mg m^{-2} d^{-1} in the winter (Fig. 4). Seasonal data were only available for one tropical rural site; BC deposition was 0.04 mg m^{-2} d^{-1} during the wet and 0.09 mg m^{-2} d^{-1} during the dry season.

5. Discussion

5.1. Spatial variability in rainfall BC concentrations and deposition

Our data synthesis revealed large spatial variability in annual rainfall BC concentrations and deposition among sites primarily located in the mid-latitudes and with medium to high annual rainfall. Annual rainfall varied little among remote sites and ~4-fold within urban and rural land use classes, while BC concentrations ranged 7- to 17-fold within land use classes (Table 2). Thus, variability in rainfall BC deposition was due to variation in BC concentrations. Several factors likely masked the effects of land use on these rainfall BC concentrations: the small size of the dataset, changes in BC emissions over time, use of diverse analytical methods (Cerqueira et al., 2010), and the influence of such factors as distance to coast on hygroscopicity (Jurado et al., 2008).

Only 17 of 42 sites had observations of BC in rainfall that spanned one or more years, even fewer of atmospheric BC concentrations (Table 2). Compared to rural sites, very few urban (n = 4) and remote (n = 3) sites had either annual concentration or deposition data. There were similarly low sample sizes (i.e., number of sites) for each season of the year, making it difficult to examine differences by land use class on both annual and seasonal timescales. Furthermore, measurements were conducted over a period of more than three decades (Table 1). The highest rainfall BC concentrations were from the late 1980s and early 1990s. Since then, atmospheric BC concentrations have decreased in

Table 1
List of sites included in this synthesis of rainfall BC concentrations and deposition. n.d. indicates no data.

Site	Land use	Sampling period	Collector type	Sample size	Reference
Aveiro, Portugal	urban	Feb–Apr 2011	wet-only	7	Zhang et al. (2015)
Dübendorf, Switzerland	urban	Apr 2012–Mar 2013	wet-only	12	Zhang et al. (2015)
Gdynia, Poland	urban	Jan 2012–Feb 2013	wet-only	33	Witkowska et al. (2016)
Halifax, Canada	urban	Apr 1995, Jun 1995 Sept 1995, Oct 1995	wet-only	1,4,1,1	Chýlek et al. (1999)
Paris, France	urban	Jan 1988–Aug 1990	wet-only	58	Ducret and Cachier (1992)
Seattle, U.S.	urban	Dec 1980–Jan 1981	wet-only	7	Ogren et al. (1984)
Seoul, South Korea	urban	Mar–Jun 1998	n.d.	5	Hwang et al. (2004)
Shibata, Japan	urban	Jan–Dec 2010	bulk	n.d.	Sase et al., 2012
Tokyo, Japan	urban	Jun 2011–Jun 2012	wet-only	24	Huo et al. (2016)
Warren, U.S.	urban	Jan–Apr 1984, Dec–Apr 1985	wet-only	15, 25	Dasch and Cadle, 1989
Xiamen Island, China	urban	Apr–Aug 2014	wet-only	33	Zhao et al., 2016
Arup, Sweden	rural	Apr–Aug 1981	bulk	4	Ogren and Charlson (1984) [a]
Aveiro, Portugal	rural	Jan 2003–Mar 2004	bulk	33	Cerqueira et al. (2010)
Bredkalen, Sweden	rural	Apr–Aug 1981	bulk	4	Ogren and Charlson (1984) [a]
Bridgewater, Canada	rural	Aug 1995–Nov 1995	wet-only	10	Chýlek et al. (1999)
Cape Hedo, Japan	rural	Apr 2010–Mar 2013	wet-only	33	Mori et al. (2014)
Enyele, Congo	rural	Jun 1988–Jun 1989	wet-only	45	Ducret and Cachier (1992)
Forshult, Sweden	rural	Apr–Aug 1981	bulk	4	Ogren and Charlson (1984) [a]
Granan, Sweden	rural	Apr–Aug 1981	bulk	4	Ogren and Charlson (1984) [a]
Grimso, Sweden	rural	Apr–Aug 1981	bulk	4	Ogren and Charlson (1984) [a]
Hedesunda, Sweden	rural	Apr–Aug 1981	bulk	4	Ogren and Charlson (1984) [a]
K-Puszta, Hungary	rural	Sept 2002–Jun 2004	wet-only	19	Cerqueira et al. (2010)
Kassjo, Sweden	rural	Apr–Aug 1981	bulk	4	Ogren and Charlson (1984) [a]
Lamto, Ivory Coast	rural	Jun–Oct 1990	wet-only	21	Ducret and Cachier (1992)
Mace Head, Ireland	rural	Oct–Nov 1989	wet-only	18	Ducret and Cachier (1992)
Molėtai, Lithuania	rural	Apr 1987–June 1990	bulk	n.d.	Armalis (1999)
Niigata, Japan	rural	Jun 2011–Jun 2012	wet-only	24	Huo et al. (2016)
Preila, Lithuania	rural	Dec 1986–Jun 1990	bulk	n.d.	Armalis (1999)
Ricklea, Sweden	rural	Apr–Aug 1981	bulk	4	Ogren and Charlson (1984) [a]
Rorvik, Sweden	rural	Apr–Aug 1981	bulk	4	Ogren and Charlson (1984) [a]
Ryda Kungsgard, Sweden	rural	Apr–Aug 1981	bulk	4	Ogren and Charlson (1984) [a]
Sakaerat Biosphere Reserve, Thailand	rural	Jan–Dec 2010	bulk	n.d.	Matsuda et al. (2012)
Schauinsland, Germany	rural	Feb 2003–Aug 2004	bulk	44	Cerqueira et al. (2010)
Sinhagad, India	rural	Jun 2008–Oct 2010	wet-only	61	Budhavant et al. (2016)
Sjoangen, Sweden	rural	Apr–Aug 1981	bulk	4	Ogren and Charlson (1984) [a]
Smedby, Sweden	rural	Apr–Aug 1981	bulk	4	Ogren and Charlson (1984) [a]
Trinidad Head, U.S.	rural	Feb–Apr 2006	wet-only	14	Hadley et al. (2010)
Azores, Portugal	remote	Feb 2003–Jun 2004	bulk	7	Cerqueira et al. (2010)
Azores (Terceira Island), Portugal	remote	Dec 2009–Oct 2010	wet-only	103	Custodio et al. (2014)
Hanimaadhoo, Maldives	remote	May 2005–Feb 2007	wet-only	58	Granat et al. (2010)
Nam Co, China	remote	Jul–Oct 2006	wet-only	12	Ming et al. (2010)
Sado, Japan	remote	Jun 2011–Jun 2012	wet-only	24	Huo et al. (2016)

[a] Ogren and Charlson (1984) collected five samples during the sample period, however only one rain-dominated spring sample was included in the analysis. The other sample was dominated by snow.

Europe, Japan, and the U.S. (Huo et al., 2016; Kirchstetter et al., 2017; Witkowska et al., 2016), likely translating into lower rainfall BC concentrations.

There has also been progress in procedures and analytical methods for the measurement of carbon (Iavorivska et al., 2016), and BC specifically, in rainwater (Torres et al., 2014). Variability in rainfall BC concentrations may arise from these differing protocols. For example, rainwater filtration of BC particles has been reported to result in collection efficiencies of 50%–95% (Chýlek et al., 1999; Hadley et al., 2010; Ogren et al., 1983). Torres et al. (2014) suggest that these are likely overestimated, with collection efficiency decreasing with increasing BC concentration. With respect to analytical method, most studies included in this synthesis employed thermal-optical methods for determination of BC. Even so, the TOT method can result in overestimation of BC mass when OC becomes charred and is mistakenly classified as BC (Hadley et al., 2010). This is typically, but not always, corrected by monitoring light transmission through the sample to differentiate between OC and EC (Hadley et al., 2010). For instance, Chýlek et al. (1999) used the TOT technique, but BC measurements were based on filter optical characteristics and not on the mass of carbon evolved from samples (Cerqueira et al., 2010). The thermal method used by Ducret and Cachier (1992) also does not account fully for pyrolytic conversion of OC to EC and thus may result in overestimates of BC concentration. The uncertainty of the SP2 method for measurement of BC in rainfall is estimated to be ± 10% (Torres et al., 2014).

Fresh BC emissions in urban areas are more hydrophobic than BC particles that have undergone ageing during transport, and thus, in theory, lower scavenging rates could occur in polluted urban air with higher BC concentrations than in rural or remote areas (Ogren et al., 1984). For the few sites with data, atmospheric BC was higher at urban compared to remote sites, but we found no difference in mean scavenging ratios among land use classes (Table 2). Instead, oceanic and inland sites generally had higher scavenging ratios than coastal sites (Fig. 5). An obvious exception is the site at Mace Head, Ireland, a coastal site exposed to the ocean from nearly all directions. These limited data suggest that there may be hidden interactions between distance to coast and land use class influencing rainfall BC removal; such interactions warrant further attention in the literature on rainfall BC deposition.

5.2. Seasonal variability in rainfall BC concentrations and deposition

Although spring BC concentrations were higher at rural compared

Table 2
Annual rainfall (mm), BC concentrations in air (μg m^{-3}) and rainfall (μg L^{-1}), scavenging ratios, and rainfall BC deposition (mg m^{-2} d^{-1}) for urban, rural, and remote sites included in this synthesis with one or more years of data. 'n.d.' indicates no data.

Site	Land use	Rainfall (mm)	BC in air (μg m^{-3})	BC in rain (μg L^{-1})	Scavenging ratio (x10^6)	BC deposition (mg m^{-2} d^{-1})
Dübendorf	urban	1086	n.d.	22	n.d.	n.d.
Gdynia	urban	563[a]	2.3[c]	20	0.02	0.1
Paris	urban	637[a]	2.2	333	0.15	n.d.
Shibata	urban	2242	n.d.	n.d.	n.d.	0.20
Tokyo	urban	1438	0.83	79	0.10	0.16
Aveiro1	rural	1095	1.2	14	0.02	0.02
Cape Hedo	rural	2124	0.18	33	0.18	0.18
Enyele	rural	n.d.	2.3	100	0.11	n.d.
K-Puszta	rural	511	0.91	24	0.04	0.03
Molėtai	rural	683[a]	n.d.	n.d.	n.d.	0.22
Niigata	rural	1535	0.64	25	0.04	0.09
Preila	rural	830[a]	n.d.	100	n.d.	0.21
Sakaerat	rural	1354[b]	1.6	n.d.	n.d.	0.05
Schauinsland	rural	1570	0.29	28	0.17	0.10
Azores1	remote	1095	0.04	2.8	0.20	0.01
Azores2	remote	1133	n.d.	15	n.d.	0.05
Sado	remote	1087	0.38	26	0.08	0.07

[a] Mean annual rainfall.
[b] Rainfall data from Sase et al. (2012).
[c] BC concentration in $PM_{2.5}$.

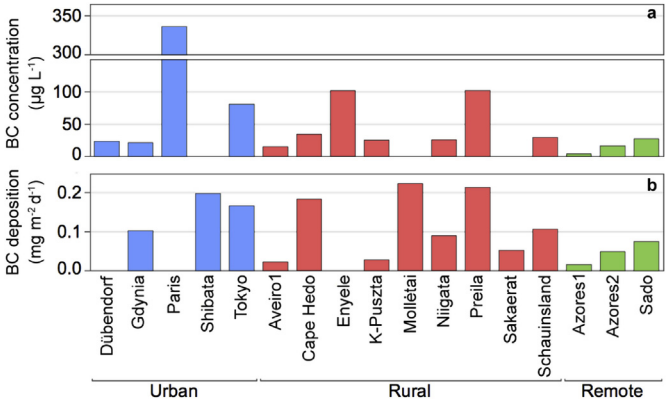

Fig. 2. Annual mean rainfall (a) black carbon (BC) concentrations (μg L^{-1}) and (b) black carbon (BC) deposition rates (mg m^{-2} d^{-1}) measured at urban, rural, and remote sites included in this synthesis. No bars indicate that data for the site were not available.

to urban sites (Fig. 3), the median was biased upward by elevated rainfall BC concentrations (≥200 μg L^{-1}) at nine Swedish sites. Excluding these Swedish sites resulted in a median rainfall BC concentration for rural sites (65 μg L^{-1}) that was not statistically different from that of urban sites (38 μg L^{-1}). Our inability to detect other seasonal differences within and among land use classes resulted from the same factors that contributed to high spatial variability (e.g., sample size, analytical methods) discussed in the previous section.

In their study on particulate carbon in precipitation at European sites, Cerqueira et al. (2010) highlight the lack of information on seasonal variability. In this synthesis, there were 15 sites with data on rainfall BC for two or more seasons. Given that rainfall patterns, major BC emission sources, and air mass trajectories often characterize broad regions, we used these data to explore the effect of geographic context on seasonal variations in BC concentrations and deposition (Fig. 6).

All East Asian sites, except Tokyo, demonstrate spring and winter peaks in either rainfall BC concentrations or deposition. These sites are affected by long-range transport of BC from northern and northeast Asia in spring and winter (Huo et al., 2016; Kondo et al., 2011; Mori et al., 2014; Zhao et al., 2016), while elevated spring rainfall at Cape Hedo and Xiamen Island drives high spring deposition. In Tokyo, high summer fossil fuel emissions obscure this spring and winter signal, leading to a summer maximum in BC concentrations (Huo et al., 2016). In contrast to more polluted East Asian sites, sites in the North Pacific and North Atlantic regions experience lower rainfall BC concentrations due to few local emission sources and the influence of clean maritime air masses (Chýlek et al., 1999; Custodio et al., 2014; Hadley et al., 2010). Deposition rates also exhibit low seasonality, although spring and winter peaks can be seen due to long-range transport (e.g., from Asia, Hadley et al., 2010; North America and Europe, Custodio et al., 2014) coupled with, in some cases, a Mediterranean rainfall regime. The continentally-influenced sites in Southwestern and Central Europe show higher rainfall BC concentrations due to fossil fuel and biomass combustion, but overall little seasonality (Zhang et al., 2015). The tropical sites show elevated BC concentrations or deposition during the dry season when biomass burning occurs in Central Africa (Bauters et al., 2018; Ducret and Cachier, 1992), South (Granat et al., 2010) and Southeast Asia (Matsuda et al., 2012; Ponette-González et al., 2016). Biomass burning aerosol has high BC content (Jurado et al., 2008), resulting in similar or higher dry season rainfall BC concentrations at tropical compared to mid-latitude sites. Overall, different geographic regions exhibit characteristic seasonal BC concentration and deposition patterns.

5.3. Comparisons with other systems

Despite the variability in our dataset, we find that the magnitude of annual rainfall BC deposition is comparable to yearly averaged BC fluxes to the global oceans modeled by Jurado et al. (2008). In that study, BC deposition to 0-30°N and 30-60°N was 0.18 and 0.10 mg m^{-2} d^{-1}, respectively. In this synthesis the mean across 17 sites (38% coastal and oceanic) was 0.12 mg m^{-2} d^{-1} for 0-30°N and 0.11 mg m^{-2} d^{-1} for 30-60°N.

Doherty et al. (2010, 2014) present regional averages of estimated mass of BC per mass of surface snow for central North America as well as the Arctic. In the Arctic, Doherty et al. (2010) report median values that range from 1 to 34 μg L^{-1}. These values are lower than the median rainfall BC concentration for all 37 mid-latitude sites in this synthesis, 62 μg L^{-1}. With the exception of Canadian sites (median = 19 μg L^{-1}), regional surface snow averages for central North America (Doherty et al., 2014) are more comparable: 35 μg L^{-1} in the Pacific Northwest; 37 μg L^{-1} in the Intramountain West; and 44 μg L^{-1} in the Northern US Plains. Such comparisons should be treated with caution, however. As Doherty et al. (2010) note, there is some evidence that their estimates are considerably higher than those obtained with thermal-optical methods.

Few direct comparisons of BC in rainfall and snow have been conducted in mid-latitude sites. Those that have indicate that snow BC concentrations and deposition are often higher than in rain, as opposed to lower which our comparisons with those of Doherty et al. (2014) might suggest. Hadley et al. (2010) measured slightly higher BC deposition via snow compared to rain in the Sierra Nevada Mountains, while Witkowska et al. (2016) measured snow deposition rates in Gdynia, Poland, that were 32-fold higher compared to those by rain. In Halifax and Seoul, BC deposition rates were not reported but snow:rain enrichment ratios ranged from 3 to 4 (Chýlek et al., 1999; Hwang et al., 2004). Regardless, BC deposition may be higher in terrestrial systems experiencing seasonal snowfall than in systems with seasonal rainfall alone.

5.4. Importance of wet deposition of black carbon

Current model simulations of BC deposition using the Petroff and Zhang scheme implemented in the Community Atmospheric Model

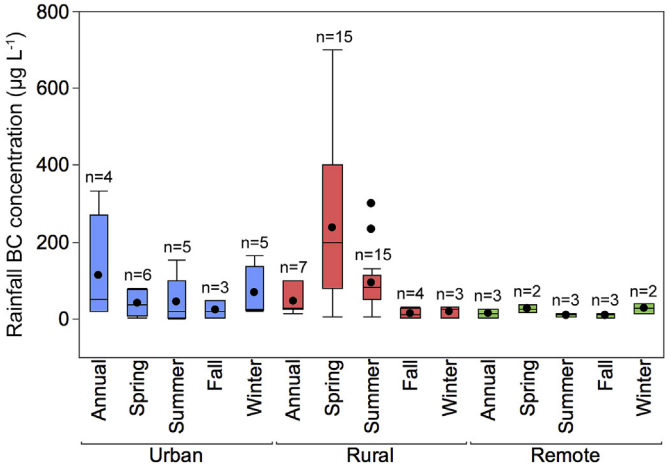

Fig. 3. Boxplots of rainfall black carbon (BC) concentrations (µg L^{-1}) by season for urban, rural, and remote mid-latitude sites included in this synthesis. Black points represent mean values. N represents the number of sampling sites.

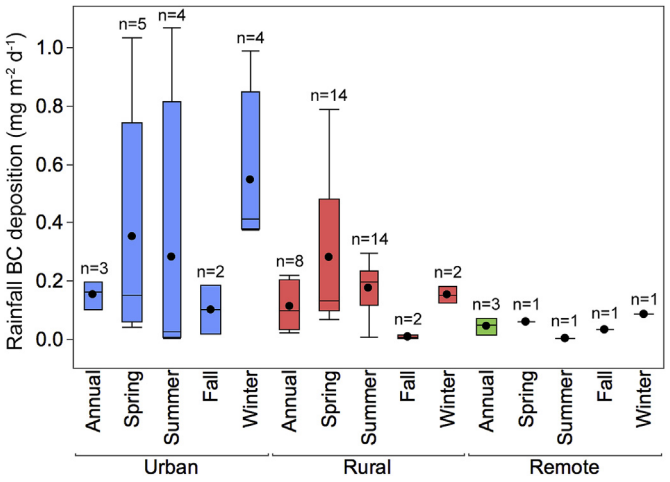

Fig. 4. Boxplots of rainfall black carbon (BC) deposition rates (mg m^{-2} d^{-1}) by season for urban, rural, and remote mid-latitude sites included in this synthesis. Black points represent mean values. N represents the number of sampling sites.

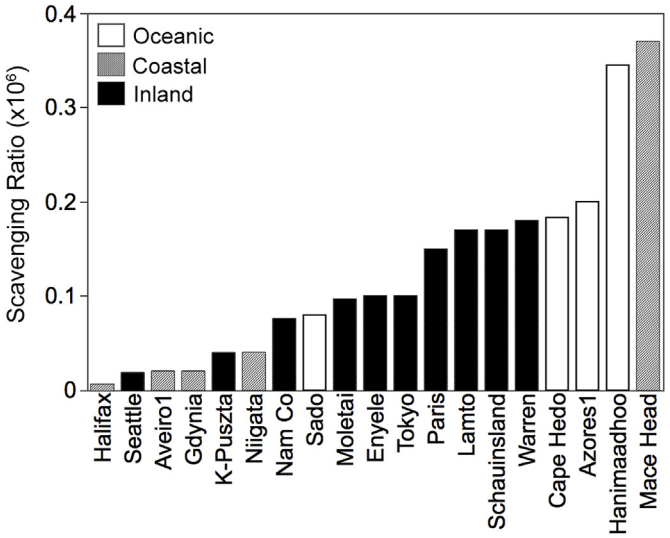

Fig. 5. Volume-based scavenging ratios (x10^6) for oceanic, coastal, and inland locations included in this synthesis.

version 5 (CAM5) show that wet deposition (deposition by any form of precipitation, not just rainfall) represents ~80% of total (wet + dry) BC deposition globally (Wu et al., 2018). Additional models indicate that wet deposition is responsible for up 83% of total deposition to the global oceans (Jurado et al., 2008) and 78%–90% of total BC deposition in the northern hemisphere and Arctic, respectively (Ting-Feng and Cun-De, 2016). Thus, removal of BC from the atmosphere via wet deposition has major implications for climate change, air quality, and terrestrial and aquatic ecosystems (Bond et al., 2013; Ostro et al., 2015).

The presence of BC in the troposphere can significantly increase absorption of incoming solar radiation, leading to positive radiative forcing (Bibi et al., 2017). In an urban environment in Pakistan, the contribution of BC to total aerosol radiative forcing was greater than 84% over a 2-year observational period, and up to 93% during a one-month period (Bibi et al., 2017). Moreover, previous studies have shown that increased wet BC deposition at low- and mid-latitudes leads to decreased aerosol transport to high-latitude remote regions. A decrease in aerosol transport lowers aerosol concentrations and decreases aerosol absorption of solar radiation at high latitudes (Wu et al., 2018). Therefore, if BC is removed from the atmosphere by deposition, BC forcing will decrease (Bond et al., 2013).

However, once removed from the atmosphere, deposited BC could have major ecosystem implications, some of which remain relatively unknown. For example, in the open ocean where rainfall BC deposition is the primary pathway for BC input, Mari et al. (2019) were the first to assess the impacts of rainfall BC on nutrient concentrations and particle dynamics. They found that rainfall BC inputs were associated with higher nutrient concentrations in the surface microlayer and sorption of dissolved organic matter. The effects of BC deposition onto snow and ice are better understood; BC on snow and ice surfaces lowers surface albedo, leading to increased warming at the surface (Bond et al., 2013; Zhang et al., 2017). Further research is needed to determine the biogeochemical consequences of BC deposition in forested, grassland, and aquatic ecosystems.

5.4.1. *Measurement gaps*

Our compilation highlights major gaps in the measurement and monitoring of rainfall BC concentrations and deposition. All observations were from the northern hemisphere, with the majority of sites located in Europe (Fig. 1). However, this is not representative of global anthropogenic BC emissions. According to a recent inventory, total anthropogenic BC emissions are highest in India, East Asia, and equatorial Africa; these are approximately 2.5-fold, 4-fold, and 2-fold greater, respectively, than anthropogenic BC emissions in Europe and Russia combined (Klimont et al., 2017). Rainfall in these high BC-emission regions is seasonally wet to very wet, with rainfall BC deposition likely to be high as well, highlighting the need for year-round studies in these regions.

Urban sites were poorly represented in the dataset, especially in the U.S. and Europe where the transport sector (on-road diesel engines) is the most important source of BC to the atmosphere (Bond et al., 2013; Klimont et al., 2017). Recent research shows that in highly polluted urban environments the light absorption and radiative impacts of BC can be enhanced by the presence of pollutants that can coat BC particles (Bai et al., 2018; Peng et al., 2016; Zhang et al., 2018). As urban populations continue to grow, it is critical to understand how BC is removed from the atmosphere and how this removal influences radiative forcing and air quality in urban areas (Scovronick et al., 2017).

Due to the importance of wet BC deposition for understanding the global radiation budget and its effects on climate and ecosystems, more long-term observations of BC in rainfall are needed, especially in tropical and urban environments. Including measurements of BC in rainfall in the routine analysis of samples collected at deposition monitoring networks (e.g., NADP, EANET) would allow for a more robust understanding of the influence of emissions and precipitation on BC

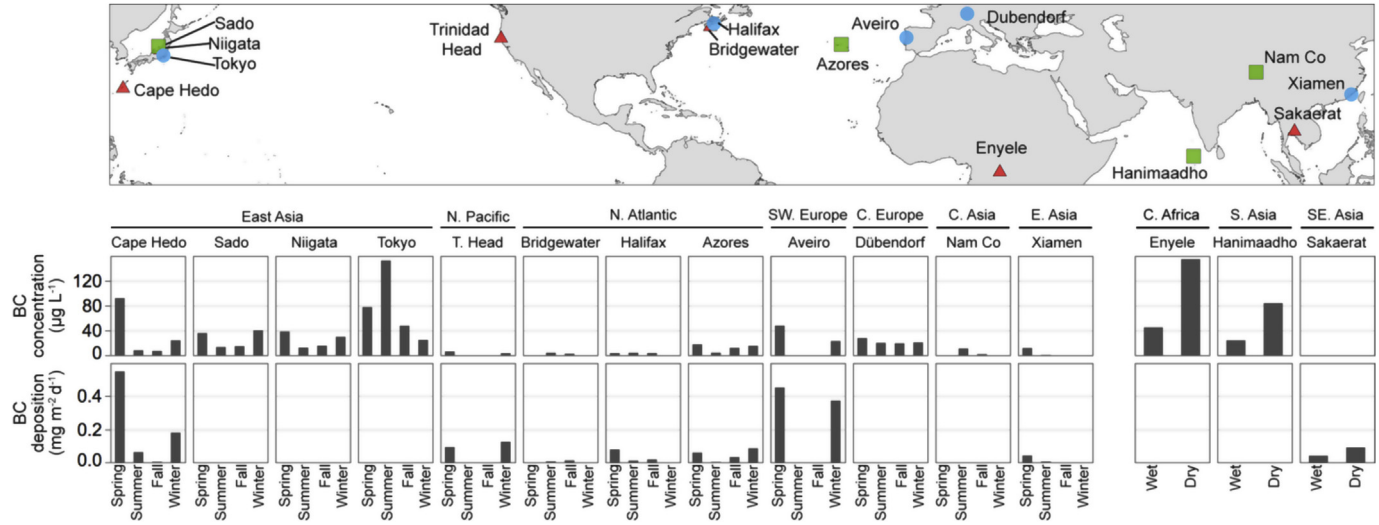

Fig. 6. Seasonal rainfall black carbon (BC) concentrations (μg L^{-1}) and deposition (mg m^{-2} d^{-1}) at 12 mid-latitude and three tropical sites included in this synthesis. No bars indicate that data for the site were not available. Sites are categorized by region and organized by longitude (left to right for mid-latitude and tropical sites).

deposition on a global scale. Long-term observations would also advance understanding of pools and fluxes of BC in the global BC cycle.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.atmosenv.2019.06.033.

Declaration of interests

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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