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Black carbon concentrations and sources in the marine boundary layer of the tropical Atlantic Ocean using four methodologies

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Abstract

Black carbon (BC) is the highly carbonaceous byproduct of biomass burning and fossil fuel combustion with a composition ranging from thermally stable soot to less recalcitrant charcoal. Atmospheric particulate matter samples across the tropical Atlantic Ocean were quantified for BC using four different methods: chemothermal oxidation at 375 °C (CTO-375), pyrene fluorescence loss, thermal optical transmittance, and optical transmission attenuation. The highest BC concentrations were detected in the Caribbean Sea and off the African coast, with a regional average of $0.6 \mu\text{g m}^{-3}$ for both. The lowest average concentrations were measured off the coast of South America at 0.2 to $0.3 \mu\text{g m}^{-3}$. The thermally-based CTO-375 method generally detected lower BC concentrations than the other three methods. The ratio of soot-like BC, as defined by the CTO-375 method, relative to the broader BC combustion continuum, as defined by the pyrene fluorescence loss, was < 1 for all regions except for the Caribbean, supporting that charcoal was an important fraction of the aerosol BC. Regions impacted by biomass burning emissions should utilize multiple methods to better apportion the BC concentrations and sources.

1 Introduction

Black carbon (BC) is the byproduct of the incomplete combustion of biomass burning and fossil fuel combustion that is composed of highly condensed matrices of aromatic sheets (Goldberg, 1985). It has the potential for long range atmospheric transport which makes it ubiquitous in the environment (Ogren and Charlson, 1983; Chuang et al., 2002; Masiello, 2004). Atmospheric BC is an important forcing factor with respect to climate change, second only to carbon dioxide (Husain et al., 2007; Bond et al., 2013; Novakov and Rosen, 2013). Its optical properties make it a strong absorber of incident sunlight and its deposition to the ocean is a potential long-term sink for fixed carbon and organic pollutants (Schmidt and Noack, 2000; Forbes et al., 2006). BC is rarely

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5 measurements of transmission attenuation before and after firing the sample at a high temperature to remove the carbon component while retaining the mineral dust (Hansen et al., 1993). Comparative studies measuring EC and optically-defined equivalent black carbon (EBC) have detected both more and less BC on the same sample, emphasizing the need for multiple methods to be applied when measuring BC (Lavanchy et al., 1999; Currie et al., 2002; Park et al., 2006).

10 The tropical Atlantic Ocean is expected to have elevated BC concentrations due to a combination of intense biomass burning of the grassland biomes and the easterly winds which transport it to the Atlantic (Chester et al., 1979; Perry et al., 1997; Bond et al., 2004; Koch et al., 2009; Lohmann et al., 2009). There is also little information available on BC concentrations in remote marine environments, such as the tropical Atlantic Ocean. We evaluated four methods, CTO-375, TOT, pyrene fluorescence loss, and optical attenuation (OT-21), to measure different fractions of the BC combustion continuum (soot-like BC, EC, pyrene-BC, and EBC, respectively). This enabled us to gain a better understanding of the make-up of marine BC aerosols and their sources in the tropical Atlantic. Atmospheric BC samples were collected from four regions: the Caribbean (QFF 1–9; QFF 11–13), South America (QFF 9–10; QFF 14), the African Plume (QFF 15–19), and the subtropical Atlantic (QFF 20–24). We note that QFF-9 was placed in both the Caribbean and South America since we had crossed the inter-tropical convergence zone during sampling.

20 We hypothesized that (1) BC would be elevated in the tropical Atlantic compared to other remote environments, (2) BC concentrations for the same sample would vary by method, and (3) the use of multiple methods would offer unique insights into the forms of BC present.

2.5 Optical transmission attenuation (OT-21)

Filter samples were analyzed for EBC at Magee Scientific in Berkeley, California using the optical Transmissometer Model OT21. This instrument measured the light attenuation through a 25 mm diameter punch of each QFF at the wavelengths 880 and 370 nm, and compared it to that of a blank filter (Husain et al., 2007; Ahmed et al., 2009; Dutkiewicz et al., 2009). A wavelength of 880 nm was selected and an absorption coefficient of $16.6 \text{ m}^2 \text{ g}^{-1}$ was used to convert the absorbance to a EBC concentration. The detection limit was $0.06 \mu\text{g cm}^{-2}$.

2.6 Quality control and quality assurance

Quality control and assurance was assessed on all QFF samples using a combination of blanks and standard reference materials. BC was not detected on blank filters for all quantification methods except for the CTO-375 and TOT technique. We subtracted the average blank BC concentration of $1.7 \mu\text{g cm}^{-2}$ for the CTO-375 method and $0.04 \mu\text{g cm}^{-2}$ for the TOT method from each sample. We hypothesized that this carbon was entrained inside of the filters, and was liberated during the high temperature oxidation step. We confirmed in follow-up experiments that the blank associated carbon remained constant regardless of the air volume filtered.

Methodological quality control was assessed using the NIST standard reference material 1649a (urban dust). Risotto char was also used for the CTO-375 method only as suggested by the BC ring trial (Hammes et al., 2007). BC mass fractions for the CTO-375 method were within the expected range for 1649a at $2.7 \pm 0.1 \%$. An insufficient mass of the risotto char was recovered, as expected, since the CTO-375 is optimized to measure the soot fraction and chars are oxidized (Gustafsson et al., 1997; Flores-Cervantes et al., 2009). The absence of risotto char recovery suggests that charring was not occurring. Higher BC concentrations were detected with 1649a (7.9%) using the pyrene fluorescence loss method, which was expected (Flores-Cervantes et al., 2009). The TOT method produced an expected middle value of $4.2 \pm 0.4 \%$ (Currie

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et al., 2002) A precombusted quartz sand blank was also used for the CTO-375 and pyrene fluorescence loss methods with no BC detected. Statistical analyses were conducted using the data analysis tool in Microsoft EXCEL.

3 Results and discussion

3.1 Caribbean Sea

Total organic carbon concentrations from CTO-375 and TOT were significantly correlated (p value = $3E^{-5}$; $R^2 = 0.84$) and ranged between $0.3\text{--}5.8\ \mu\text{g m}^{-3}$ and $0.4\text{--}6.1\ \mu\text{g m}^{-3}$, for the CTO-375 and TOT, respectively (Table 1). Modeled organic matter concentrations for the Caribbean were mostly lower around $0.5\ \mu\text{g m}^{-3}$ (Liousse et al., 1996). The ratio of BC to the total organic carbon was highly variable, ranging from 0% to 86% for the CTO-375 method and 3% to 58% for the TOT method with averages of 45% and 16% respectively (Fig. 1). The qualitative color description of each filter for the Caribbean region was mostly pale yellow to tan, which was unique to this region and could be from mineral dust or biomass burning particles.

The highest BC concentrations measured in this study occurred in the Caribbean Sea, particularly between the highly developed and populated islands of the Dominican Republic and Puerto Rico (Table 2; Fig. 2). The BC concentrations between the four methods ranged from below detection limit to $3.4\ \mu\text{g m}^{-3}$. The pyrene fluorescence loss method measured the highest pyrene-BC concentration and had a regional average of $1.1\ \mu\text{g m}^{-3}$ with four of the samples exceeding $1\ \mu\text{g m}^{-3}$. Similarly, the CTO-375 method had a maximum soot-like BC concentration of $3.2\ \mu\text{g m}^{-3}$ and a regional average of $0.8\ \mu\text{g m}^{-3}$. The pyrene fluorescence loss and optical attenuation were found to be significantly correlated (p value = 0.048; $R^2 = 0.37$) as was the optical attenuation and TOT method (p value = 0.007; $R^2 = 0.62$). Optical attenuation and the TOT methods both did not detect their highest overall BC concentration in this region, but

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concentrations were still elevated up to $0.7 \mu\text{g m}^{-3}$ for optical attenuation and $0.6 \mu\text{g m}^{-3}$ for TOT.

The Caribbean Sea has had very few reported BC measurements. Typical July EBC measurements in this region were between $0.014\text{--}0.019 \mu\text{g m}^{-3}$ (Hansen et al., 1990). Similarly, large scale model estimates of atmospheric BC concentrations from both fossil fuel combustion and biomass burning have predicted BC concentrations in the summer months to be $< 0.010 \mu\text{g m}^{-3}$ (Lioussé et al., 1996). We caution that model estimates are generally integrated over larger vertical and horizontal scales than the direct measurements from this study; hence model estimates may be diluted in comparison to our measurements. Our data cannot differentiate if these elevated BC concentrations are typical for the summer, or a time specific anomaly, such as the occurrence of a nearby wildfire.

The average $\delta^{13}\text{C}$ value for the total organic carbon fraction was -22‰ , which indicates that marine plankton was a main source for the organic carbon (Table 1). The average soot-like BC $\delta^{13}\text{C}$ value was more depleted at -34‰ , which is indicative of terrestrial plants that utilize the C_3 carbon fixation pathway for photosynthesis. C_3 plants include most trees, so this could indicate a soot-like BC source such as a forest fire. The large fractionation between the total organic carbon and soot-like BC also implies that the charring of organic matter was likely insignificant. The intensity of open forest fires during the sampling time was retrieved from the global fire emissions data base (Giglio et al., 2003; Davies et al., 2004). Elevated concentrations of BC could be partly due to forest fires, specifically in Cuba. Ten-day backward wind trajectories also displayed the potential of distal BC sources originating from Eastern Europe and North East Africa (Fig. 1). The average atmospheric residence time of soot-like BC in the Northern Hemisphere is 5.4 days, suggesting that some BC particles could be transported from Europe to the Caribbean (Chuang et al., 2002).

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pleted to be solely marine plankton (Table 1). The backward wind trajectories showed that the air originated from a distal source in southern Africa, but local burning activities in the Amazon rainforest could also be a source of the organic material (Fig. 1). The average $\delta^{13}\text{C}$ value of the soot-like BC pool was also within the C_3 plant range of -28‰ . The small variation between the carbon pools suggests that total organic carbon and soot-like BC could be from the same source, such as the same wildfire event. We hypothesized that C_4 grass combustion (the carbon fixation pathway optimized for drier climates such as the African Savanna) was not an important source of this soot-like BC pool, thus distal BC inputs from Africa were minimal.

3.3 African plume

A primary objective of this study was to sample the African aerosol and dust plume to determine if the tropical Atlantic could be receiving significant inputs of BC. The MO-GUNTIA global transport model has estimated that up to 70% of the global BC from biomass emissions originates from Africa, most of which are transported to the Atlantic Ocean (Cooke and Wilson, 1996; Perry et al., 1997). We sampled this plume during the Southern Hemispheric dry season when large scale Savanna fires are frequent (Cahoon et al., 1992). The global fire emissions database showed that our sampling occurred during widespread grassland burnings in Southern Africa. Approximately 13% of the air parcels originated from the African continent as predicted by the mean backward wind trajectories. The HySPLIT backward trajectory model does not take into account eddy diffusion, thus may underestimate the transport of air from land into the marine boundary layer.

Total organic carbon concentrations in the African Plume were significantly correlated and ranged from 0.1 to $1.5\ \mu\text{g m}^{-3}$ for CTO-375 and 0.2 to $1.7\ \mu\text{g m}^{-3}$ for TOT (p value = 0.002; $R^2 = 0.98$; Table 1), which agreed with the modeled organic carbon concentrations of 0.5 to $2.0\ \mu\text{g m}^{-3}$ (Liousse et al., 1996). The regional average of the fraction of BC composing the total organic carbon pool was 13% for CTO-375 and

73% for TOT, suggesting that there was a form of BC that could be detected by the TOT method, but was oxidized as organic carbon in the CTO-375 method (Fig. 1).

BC concentrations in the African Plume region resulted in distinctly different concentrations between the CTO-375 and the other three methods. Concentrations between all four methods ranged from 0 to $1.4 \mu\text{g m}^{-3}$, with CTO-375 detecting the lowest concentrations and the TOT and optical attenuation detecting the greatest concentrations, both with regional averages of $0.8 \mu\text{g m}^{-3}$ (Table 2). Concentrations for the CTO-375 method ranged from 0 to $0.2 \mu\text{g m}^{-3}$, which are similar to BC concentrations found in the Canadian Arctic (Sharma et al., 2004). BC concentrations, omitting the CTO-375 method, ranged from 0.2 to $1.3 \mu\text{g m}^{-3}$. The TOT method measured the overall highest concentration of EC in the region ($1.4 \mu\text{g m}^{-3}$), which was close in magnitude to the EBC concentration measured by the optical attenuation method ($1.3 \mu\text{g m}^{-3}$) and pyrene fluorescence loss of ($0.9 \mu\text{g m}^{-3}$). The pyrene fluorescence loss and optical attenuation were found to be significantly correlated to each other (p value = 0.01; $R^2 = 0.90$) as well as the TOT method and optical attenuation (p value = 0.006; $R^2 = 0.94$).

The optically-based (optical attenuation and, in part TOT) and chemically-based (pyrene fluorescence loss) methods yielded BC concentrations expected for this region while the chemothermal method yielded lower concentrations (Table 2). We thus conclude that less thermally stable forms of BC, such as char and charcoal, dominated in this region. The ratio of soot to BC (CTO-375/pyrene fluorescence loss) was 0.3 ± 0.2 (Fig. 1). Our results imply that in areas with high biomass burning inputs, charcoal may be present in concentrations comparable to or greater than the soot fraction.

The $\delta^{13}\text{C}$ value for the total organic carbon pool was enriched compared to the other regions in this study (average of -21‰ ; Table 1). We interpret this enrichment as a mix of marine plankton and C_4 plant material, presumably from the grasslands of southern Africa. This is supported by the backward wind trajectories (Fig. 1) and the global fire emissions data base detecting widespread open burns during sampling (Aux information). The average $\delta^{13}\text{C}$ value for the soot-like BC was -17‰ , which strongly indicates

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that the soot-like BC was a byproduct of grass combustion. Underlying sediments in this region, such as from the Niger Delta, have been measured to be similarly enriched at -17‰ , which agrees with our data (Holtvoeth et al., 2005).

3.4 Subtropical Atlantic

Northern Hemispheric air originating from the open ocean and the Arctic was sampled in the subtropical Atlantic (Fig. 1). Concentrations of BC were low in the subtropical Atlantic, as expected, but increased upon approaching the United States coast. Previous modeling studies have suggested that the clean Arctic air inputs would result in lowered BC concentrations, as compared to urban regions, with BC concentrations $\geq 0.05 \mu\text{g m}^{-3}$ (Koch and Hansen, 2005).

The CTO-375 method detected lower concentrations than the other three methods (Fig. 2). The CTO-375 method measured soot-like BC ranging from $0.1\text{--}0.2 \mu\text{g m}^{-3}$, where the greatest soot-like BC measurement occurred in the last sample that approached the eastern United States coast. The pyrene fluorescence loss technique ranged in pyrene-BC concentrations from $0.1\text{--}0.6 \mu\text{g m}^{-3}$ and had an apparent trend of being highest near the African and United States coast and low across the subtropical gyre. The highest pyrene-BC concentration was also on the last filter, but nearly an order of magnitude higher than previously modeled concentration of $< 0.1 \mu\text{g m}^{-3}$ (Cooke and Wilson, 1996).

The optical attenuation method measured EBC concentrations ranging from $0\text{--}0.3 \mu\text{g m}^{-3}$ and was the only method not to detect elevated EBC concentrations on the last filter. The last filter was qualitatively described in color as tan, whereas the other filters in this region were varying shades of gray. Although we cannot correct the color of each filter for the variable volumes of filtered air, we can speculate that the optical attenuation did not detect EBC due to its color (Lavanchy et al., 1999; Andreae and Gelencser, 2006; Feng et al., 2013). The two samples analyzed by the TOT method detected a median EC concentration between the CTO-375 and the other two, non-thermal, methods (Fig. 2).

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hypothesized that the CTO-375 method isolated only the soot-like fraction of BC and TOT may include other carbonaceous products. We suggest here that TOT may include other portions of the BC combustion continuum in addition to soot or that the CTO-375 method may oxidize a portion of the soot fraction along with the organic carbon.

4 Conclusions

Four distinctly different methods were used to measure black carbon concentrations across the Atlantic Ocean, covering different portions of the black carbon combustion continuum. Non-thermal methods consistently detected more black carbon than the chemothermal oxidation and thermal optical transmittance methods. This implies that less thermally stable combustion-derived byproducts (i.e. charcoal) were an important fraction to the marine aerosols in the tropical Atlantic. Our results confirm the importance of biomass burning on emitting black carbon. In particular, black carbon concentrations were elevated (0.1 to $0.8 \mu\text{g m}^{-3}$) over the tropical Atlantic when air masses came off Africa. Concentrations of charcoal were comparable to those of the soot across the tropical Atlantic, averaging up to 80 % of the black carbon, implying efficient long-range transport for both black carbon fractions. Similar field measurements should be conducted in other remote regions for both the summer and winter seasons to better understand the global atmospheric concentrations and composition of black carbon. Future work should aim to both measure the entire black carbon combustion continuum and to apportion charcoal vs. soot when evaluating carbonaceous aerosols.

Supplementary material related to this article is available online at <http://www.atmos-chem-phys-discuss.net/13/29785/2013/acpd-13-29785-2013-supplement.pdf>.

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Table 2. Average black carbon concentrations by region ($\mu\text{g m}^{-3}$) using four different methods: the chemothermal oxidation at 375 °C (CTO-375), thermal optical transmittance (TOT), pyrene fluorescence loss (PFL), and optical attenuation (OT-21).

Regional Average	CTO-375	TOT	PFL	OT-21
Caribbean	0.8 ± 0.3	0.3 ± 0.2	1.1 ± 0.3	0.2
South America	0.2 ± 0.1	0.2 ± 0.0	0.3 ± 0.2	0.3
African Plume	0.1 ± 0.1	0.8 ± 0.1	0.5 ± 0.3	0.8
Subtropical Atlantic	0.1 ± 0.1	0.2 ± 0.0	0.4 ± 0.1	0.2

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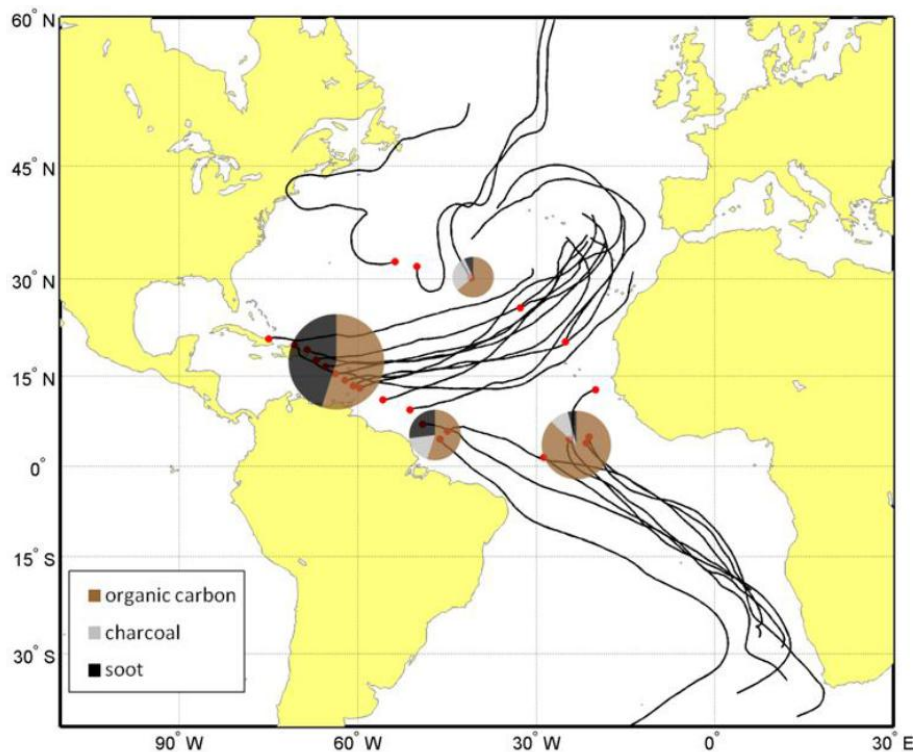


Fig. 1. HySPPLIT estimates of a 10 day backward wind trajectory at the end of each filter sample at a height of 20 m and the average regional fraction of black carbon within the total organic carbon (IRMS). Black carbon is further divided between soot (CTO-375) and charcoal (pyrene fluorescence loss). The size of each pie chart is in accordance to regional average total organic carbon concentration ranging from 0.2 to 1.8 $\mu\text{g m}^{-3}$.

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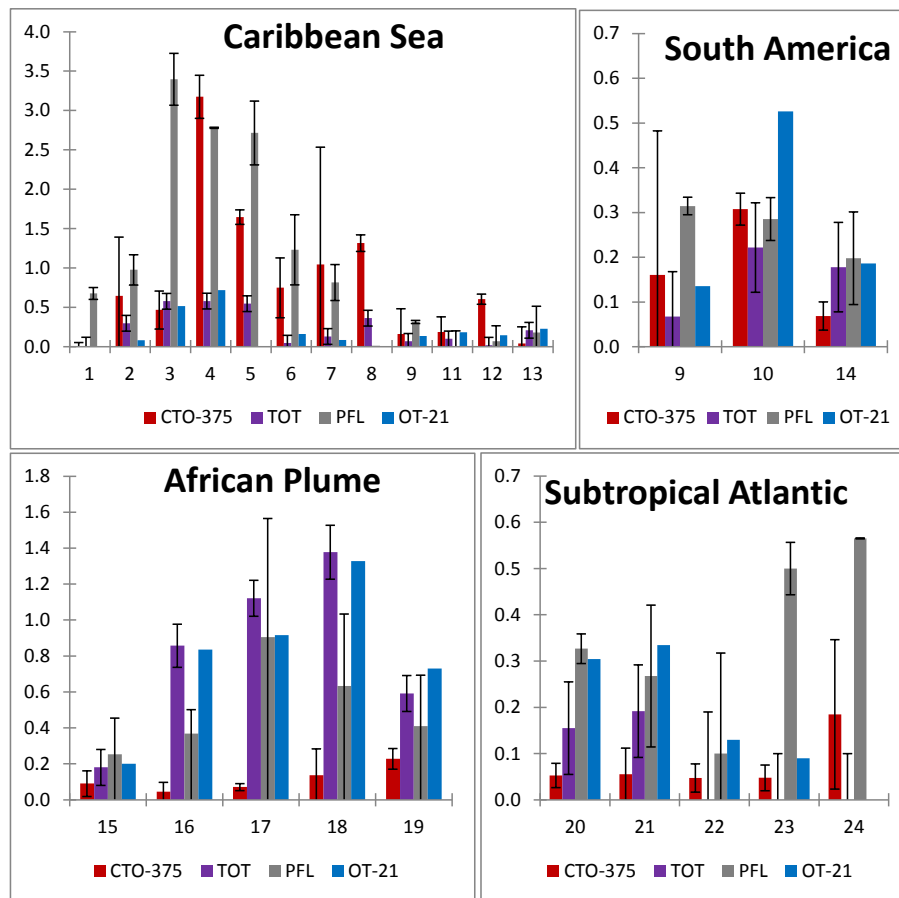


Fig. 2. Comparison of black carbon concentrations (μgm^{-3}) from the four selected methods group by region with one standard deviation.

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