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[1] Extensive measurements of the emission of light absorbing carbon aerosol (LAC) from commercial shipping are presented. Vessel emissions were sampled using a photoacoustic spectrometer in the Gulf of Mexico region. The highest emitters (per unit fuel burnt) are tug boats, thus making significant contributions to local air quality in ports. Emission of LAC from cargo and non cargo vessels in this study appears to be independent of engine load. Shipping fuel consumption data (2001) was used to calculate a global LAC contribution of $133(\pm27)$ Ggyr^{-1}, or $\sim1.7\%$ of global LAC. This small fraction could have disproportionate effects on both air quality near port areas and climate in the Arctic if direct emissions of LAC occur in that region due to opening Arctic sea routes. The global contribution of this LAC burden was investigated using the MOZART model. Increases of $20-50$ ng m^{-3} LAC (relative increases up to 40%) due to shipping occur in the tropical Atlantic, Indonesia, central America and the southern regions of South America and Africa. **Citation:** Lack, D., B. Lerner, C. Granier, T. Baynard, E. Lovejoy, P. Massoli, A. R. Ravishankara, and E. Williams (2008), Light absorbing carbon emissions from commercial shipping, *Geophys. Res. Lett.*, *35*, L13815, doi:10.1029/2008GL033906.

1. Introduction

[2] Black carbon (BC) aerosol is produced during combustion of fossil and biofuels and has strong source regions including urban centers, tropical forests and the high latitudes during spring agricultural burning and summer boreal wildfires [*Koch and Hansen*, 2005]. Compared to the radiative forcing contributions of CO_2 (+1.56 Wm^{-2}) BC is estimated to contribute +0.44 Wm^{-2} [*Intergovernmental Panel on Climate Change (IPCC)*, 2007], one quarter of which occurs in environmentally sensitive regions like the Arctic due to biofuel and anthropogenic BC decreasing the albedo of snow and ice surfaces [*Flanner et al.*, 2007; *IPCC*, 2007]. In addition to the climate impacts, BC (as an aerosol emission) contributes to adverse health effects, premature mortality and visibility reduction [*Park et al.*, 2003; *Corbett et al.*, 2007a].

[3] Large uncertainties in the climate effect of BC exist and can be partially attributed to uncertainties in the global inventories [e.g., *Bond et al.*, 2004]. About one-third of global BC emissions is from fossil fuel combustion and estimates of BC emission from shipping vary from $19-132$ Gg yr^{-1} (or $\sim0.2\%$ to $\sim5\%$ of anthropogenic emissions) [*Sinha et al.*, 2003; *Bond et al.*, 2004; *Eyring et al.*, 2005a; *Wang et al.*, 2008; T. C. Bond, personal communication, 2007]. Although a seemingly small contribution to the total, commercial shipping is an uncertain contributor due to the use of low quality unregulated fuels [*Bond et al.*, 2004], the intensity of shipping near landfall [*Corbett and Fishbeck*, 1997] and emissions of cloud condensation nuclei that alter cloud properties in pristine marine environments [*Ferek et al.*, 1998; *Schreier et al.*, 2007]. Fuel consumption by commercial shipping is expected to grow $2\%-6\%$ yr^{-1} [*Eyring et al.*, 2005b; *Corbett et al.*, 2007b] while spatial changes in shipping routes are also expected. With increasing understanding of the climatic importance of BC, particularly in the Arctic [*McConnell et al.*, 2007], and the potential for an increase of direct emissions of BC into the Arctic [*Roach*, 2005; *Granier et al.*, 2006; *Stroeve et al.*, 2007] it is essential to better understand BC emissions from shipping. Two studies of the direct emission of BC from three ships have been published [*Sinha et al.*, 2003; *Petzold et al.*, 2008]. We report an extensive set of measurements of light absorbing carbon aerosol (a quantity functionally similar to BC) emissions from commercial vessels taken onboard the NOAA research vessel Ronald H. Brown (RHB) during the 2006 Texas Air Quality Study/Gulf of Mexico Atmospheric Composition and Climate Study (TexAQS/GoMACCS). Figure 1a shows the study area encompassing the Gulf of Mexico to the Port of Houston (10th for total cargo tonnage globally) [*Port of Houston Authority*, 2008], RHB track and areas where most vessels were encountered. We apply measured emission factors to estimate a global burden and investigate the impacts on BC surface concentrations using the MOZART global model [*Horowitz et al.*, 2003; L. K. Emmons, personal communication, 2008].

2. Measurement of Light Absorbing Carbon Emission Factors

[4] A photoacoustic technique [*Lack et al.*, 2006] was used to measure aerosol light absorption (b_{abs}) (<1 μm, relative humidity < 30%, 532 nm) seconds to minutes downwind of ship plumes. This technique measures optical absorption of BC and 'brown carbon' [*Andreae and Gelencser*, 2006], which combined we refer to as light absorbing carbon (LAC). For fresh emissions the majority of absorption will be from BC however LAC is a more appropriate definition when using a light absorption technique (refer to the auxiliary material for a more detailed definition

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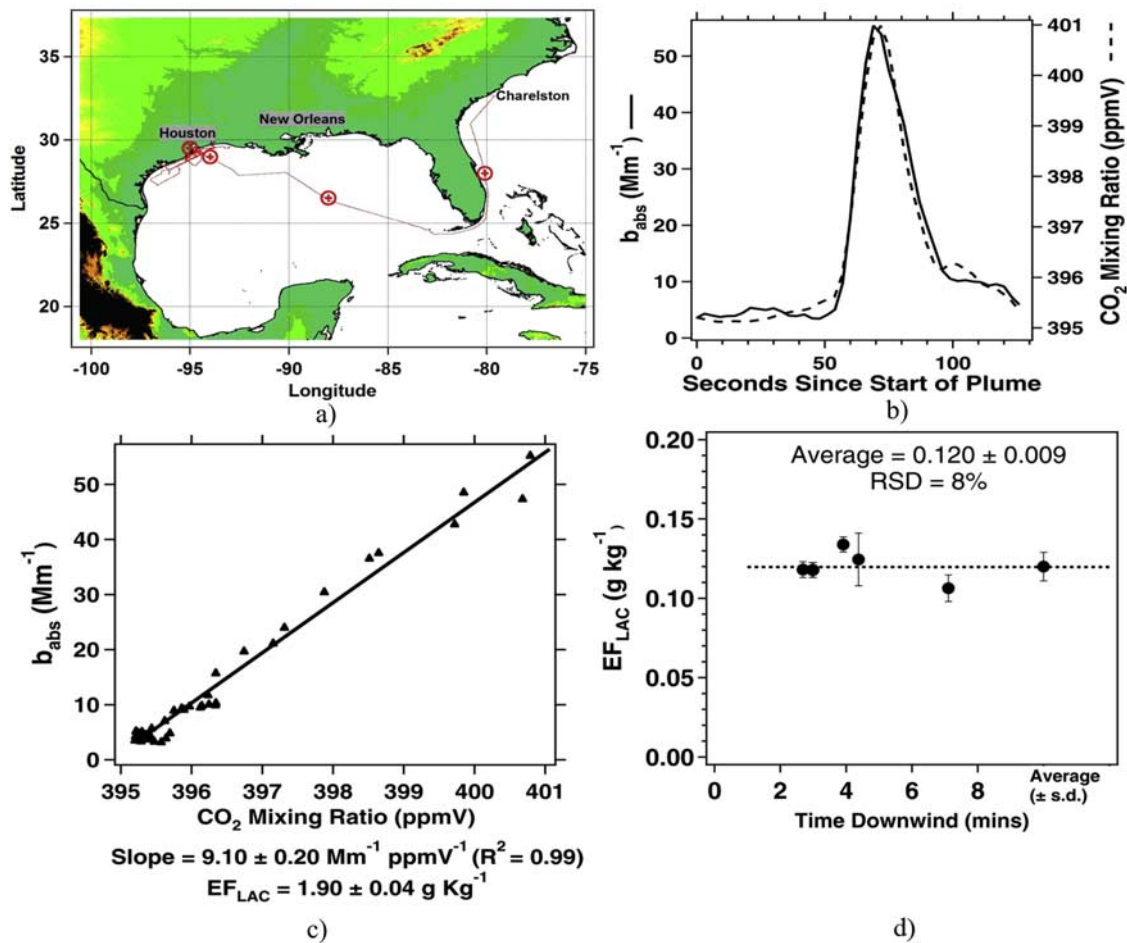[4]Now at Lockheed Martin Coherent Technologies, Longmont, Colorado, USA.

Figure 1. (a) Map of the study area. RHB ship track shown as red line, red points indicate areas of most vessel interactions. (b) Representative time series of b_{abs} and CO_2 measured in a ship plume. (c) Regression analysis of b_{abs} and CO_2 data. (d) Emission factors for LAC from a sequence of encounters of the exhaust plume from the tanker Patriot.

of LAC).[1] A cavity ring down spectrometer [*Baynard et al.*, 2007] measured 532 nm gas phase optical absorption interference (on average, a 15% correction to b_{abs}). We determine LAC emission factors (EF$_{LAC}$: grams of LAC per kilogram of fuel burnt) using equation 1.

$$EF_{LAC}\left(\text{g kg}^{-1}\right) = \frac{b_{abs}\left(\text{Mm}^{-1} at\ STP\right)}{CO_2(\text{ppmV})} \times \frac{1}{MAC(\text{m}^2\ \text{g}^{-1})} \times f_{fuel} \quad (1)$$

The ratio of b_{abs} to CO_2 normalizes b_{abs} to plume dilution (Figures 1b and 1c), and then divided by a mass absorption coefficient (MAC) produces the EF$_{LAC}$. Representing the optical absorption per unit mass of LAC, MACs have been measured for absorbing aerosol from a range of fuel types and combustion efficiency. MACs for LAC and BC were reviewed by *Bond and Bergstrom* [2006], who reported a MAC for fresh fossil-fuel combustion aerosol of 7.5 ± 1.2 m^2 g^{-1} at 550 nm (7.75 m^2 g^{-1} used here, MAC converted to 532 nm using λ^{-1} [*Kirchstetter et al.*, 2004]). The conversion factor f_{fuel} ($1.62 * 10^6$ m^3 ppmv kg^{-1})

includes 1) the fraction of fuel that is carbon (0.865 by weight) [*Lloyd's Register*, 1995] and 2) the conversion of CO_2 mixing ratio to concentration of carbon. For the 101 plumes analyzed the average b_{abs}/CO_2 R^2 was 0.93 ± 0.05. Gas phase measurements discussed in this paper (CO, NO$_Y$, SO$_2$) are described by E. Williams et al. (Emissions of NO$_X$, SO$_2$ and CO from commercial marine shipping, manuscript in preparation, 2008) and briefly described in the auxiliary material.

Table 1. LAC Emission Factor Statistics by Vessel Category

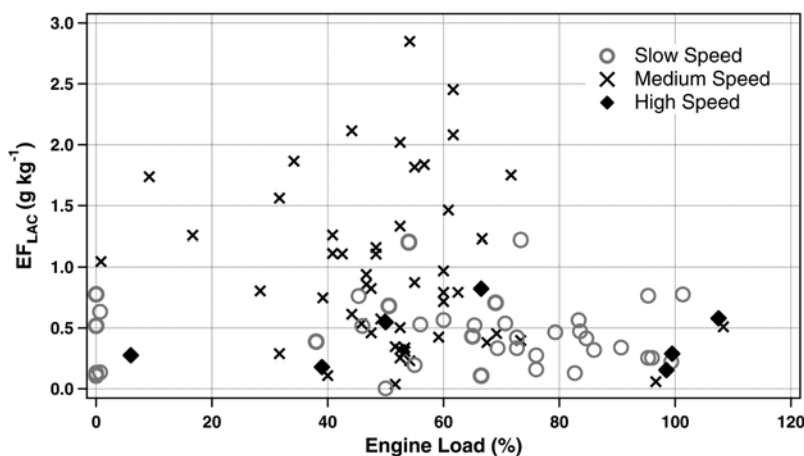|	LAC (g kg^{-1}) Avg. ± S.D.	Points
Vessel Engine Classification		
Slow Speed Diesel (SSD)	0.41 ± 0.27	42
Medium Speed Diesel (MSD)	0.97 ± 0.66	51
High Speed Diesel (HSD)	0.36 ± 0.23	8
Vessel Type		
Tankers (SSD)	0.38 ± 0.27	31
Container (SSD)	0.80 ± 0.23	4
Cargo Carriers (SSD)	0.40 ± 0.23	4
Bulk Carriers (SSD)	0.38 ± 0.16	3
Tug Boats (MSD)	0.97 ± 0.66	51
Passenger Boats (HSD)	0.36 ± 0.23	8

Figure 2. Emission factors for LAC from slow, medium and high speed diesel vessels versus inferred engine load.

[5] The precision of EF_{LAC} measurements was determined using repeated encounters with an anchored vessel (Patriot, tanker ship) which occurred at night under constant wind conditions (~ 7 ms^{-1}). The plume transit time of the closest and farthest encounters was ~ 2 and ~ 7 minutes respectively (Figure 1d). Assuming constant operating conditions for Patriot we attribute the EL_{LAC} variability to imprecision in b_{abs} and CO_2, aerosol processing during plume transit and calculation uncertainties. The standard deviation (SD) in these measurements is 8%. Propagation of uncertainties in the EF_{LAC} gives EF_{LAC} ±20%, dominated by the MAC uncertainty (15.5%), average SD in b_{abs}/CO_2 (10%), precision in EF_{LAC} (8%), uncertainty in fuel carbon content (1%) and total carbon conversion to CO_2 (2%). Vessel identification, speed and unique plumes were identified from the automated identification system (AIS), a collision avoidance transmission required on vessels exceeding 300 metric tons.

3. Results

[6] We present EF_{LAC} data for 101 vessel encounters in the open ocean, transit channels and ports. Vessels included tanker, container, cargo and bulk ships operating slow speed diesel (SSD) engines; tug and large fishing boats operating medium speed diesel (MSD) engines, and passenger vessels (e.g. ferries, pilot boats) operating high speed diesel (HSD) engines. On average MSD vessels emit more LAC aerosol per unit fuel consumed than the other vessels by almost a factor of two (Table 1). Importantly, it is these vessels (tug boats), operating in busy ports near populated areas, that emit the most LAC aerosol per unit fuel consumed, contributing to air quality issues for these areas. The ratio of AIS vessel speed to vessel service speed [*Corbett et al.*,

2006; *Lloyd's Register*, 2006] was used as an estimate of engine load. Figure 2 shows that within our dataset EF_{LAC} appear independent of engine load. This is inconsistent with *Petzold et al.* [2004], who suggest an inverse relationship due to inefficient combustion at low speeds producing more LAC. Studies of aerosol emissions with engine load show large variability [*Lloyd's Register*, 1995]. The variability within our data and of vessels types may not exclude a LAC:engine load link for individual vessels. This suggests that more research is required to explore LAC emissions with engine load for vessels of the international fleet. It is clear that laboratory bench tests will not capture the variability of global vessel emissions.

[7] The comparison between EF_{LAC} and emission factors for some gas phase species (EF_{CO}, EF_{SO2} and EF_{NOY}) (refer to Figure S1) allowed for association of LAC production with production of primary gas phase pollutants, which might suggest LAC emission control strategies. A direct relationship between EF_{LAC} and EF_{CO} for MSD engines was observed [$R^2 = 0.6$] (not for SSD). This relationship indicates that LAC emission from MSD vessels result from inefficient combustion (e.g., from aging or poor maintenance) [*MAN B&W Diesel*, 2007], and large variability in the quality of the tug fleet in the study area. Statistically significant links between EF_{SO2} or EF_{NOY} and EF_{LAC} for MSD or SSD engines were not observed. We have determined an average EF_{LAC} for SSD vessels of 0.41 (±0.27) g kg^{-1}, for MSD vessels 0.97 (±0.66) gkg^{-1} and for HSD vessels 0.36 (±0.23) g kg^{-1}. The average EF_{LAC} of SSD vessels at dock (4 vessels) was 0.52 (±0.28) g kg^{-1}, which indicates that EF_{LAC} from vessels at dock are not necessarily less than the SSD average. This is critical for considerations of switching docked vessels to clean shore power [*Kay and Caeser*, 2007].

Table 2. Estimated Annual Emission of LAC by Ship Type[a]

	Tanker	Lg Cargo	Bulk Cargo	Gen. Cargo	Non Cargo	Total
FC (Mt yr^{-1})	56.8	42.7	39.4	68.9	46.2	254
EF_{LAC} (kg t^{-1})	0.38	0.60	0.38	0.38	0.97	
LAC (Gg yr^{-1})	21.6	25.6	15.0	26.2	44.8	133 (±27)

[a]Fuel consumption (FC) is in millions of metric tons and the LAC emission factors are in kg per metric ton of fuel. Global ship type and FC profiles from *Eyring et al.* [2005a]. All but the 'Non Cargo' category are classed as Slow Speed Diesels. Lg Cargo included Container and Cargo Carriers.
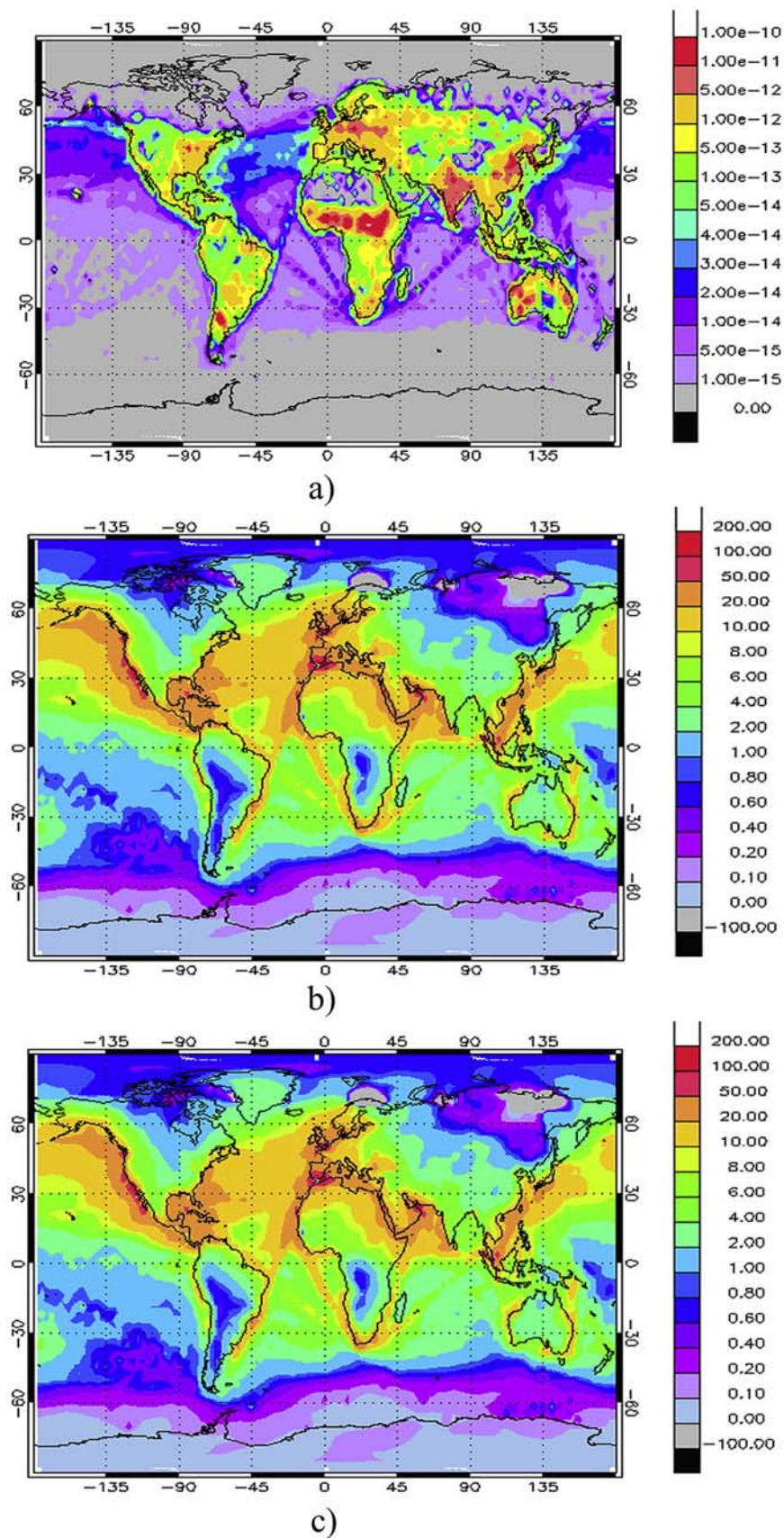
Figure 3. (a) Global surface emission of LAC (kgm^{-2} s^{-1}) with shipping emissions shown over ocean regions. (b) Absolute difference in LAC surface concentrations (ngm^{-3}) from shipping after transport. (c) Percentage difference in LAC surface concentrations from shipping after transport (for January).

[8] *Sinha et al.* [2003] and *Petzold et al.* [2008] are the only existing studies that measured EF_{LAC} (or BC) for shipping exhaust plumes. Both studies used a light absorption technique and an assumed MAC to calculate EF_{LAC}. *Sinha et al.* [2003] sampled the exhaust of a large tanker and a container ship (both SSD engines) in the open ocean and derived an EF_{LAC} of 0.18 (±0.02) g kg^{-1}. *Petzold et al.* [2008] sampled a container ship at 85% of maximum power and determined an EF_{LAC} of 0.17 (±0.04) g kg^{-1} from 10 plume passes. These results are lower than that determined for SSD in this study by a factor of two. The inventory of *Bond et al.* [2004] used an indirect emission factor for BC from ships of 1.02 g kg^{-1}. Propagating the uncertainties of the contributing parameters for this estimate gives in an uncertainty of ca. 1 g kg^{-1}. Thus from literature and our data it is clear that EF_{LAC} from shipping vessels vary widely.

[9] We make a global assessment of LAC emission from shipping based on the fuel consumption data (for 2001) of *Eyring et al.* [2005a], which is in close agreement with the most recent inventory of *Wang et al.* [2008]. Table 2 shows the results, where we have combined our emission factors to match global vessel type and fuel usage distributions from *Eyring et al.* [2005a]. Our estimated total LAC emission from shipping is 133(±27) Gg yr^{-1} (2001). For comparison *Eyring et al.* [2005a] estimated 50 Gg yr^{-1} (2001 fuel usage, EF_{LAC} of *Sinha et al.* [2003]) while *Bond et al.* [2004] and T. C. Bond (personal communication, 2007) report a value of 132 Gg yr^{-1} of BC for shipping (1996). The most recent inventory of *Wang et al.* [2008] estimated 71.4 Gg yr^{-1}. Within all estimates lies an assumption that the EF_{LAC} used represent that for the average engine load of all vessels. Within this study we sampled a wide range of engine loads and given the apparent independence of EF_{LAC} and engine load (Figure 2) we propose that our EF_{LAC} represent the fleet average.

[10] There are many implications to an improved quantification of global LAC emissions and here we use our global estimate to briefly investigate one, the spatial distribution of LAC from shipping (Figure 3a), by using the MOZART chemical transport model [*Horowitz et al.*, 2003; L. K. Emmons, personal communication, 2008] (refer to auxiliary material for model details). We use the same model parameters as detailed by *Granier et al.* [2006], LAC inventory of *Bond et al.* [2004] and the spatial distribution of emissions from *Endresen et al.* [2003], and scale the LAC intensity for shipping to 133 Gg yr^{-1}. We recognize some uncertainty may be introduced by using the spatial distributions used by *Endresen et al.* [2003] (Automated Mutual-assistance VEssel Rescue) compared to that suggested by *Wang et al.* [2008] (International Comphensive Ocean-Atmosphere DataSet). Figure 3a shows this distribution for LAC (for January) and reveals the major shipping routes in use. By assessing the surface concentrations of LAC with and without shipping emissions in the MOZART model we see that there can be a significant absolute increase in atmospheric concentrations of LAC due to shipping activity (20–100 ngm^{-3}, Figure 3b). Shipping between North America and Asia significantly increases the amount of LAC in the Alaska region, thus being a potential contributor to Arctic pollution. The most dramatic relative increases in LAC concentrations (up to 40%, Figure 3c) are

in cleaner regions; e.g. the East coasts of North America and China do not show significant relative changes due to shipping, presumably due to the dominance of terrestrial sources of LAC.

4. Summary

[11] Based on 101 encounters of 96 unique vessels in the Gulf of Mexico we determine mass based emission factors of light absorbing carbon (EF_{LAC}) of 0.41 (±0.27) g kg^{-1}, 0.97 (±0.66) g kg^{-1} and 0.36 (±0.23) g kg^{-1} for slow (SSD), medium (MSD) and high speed diesel (HSD) powered vessels respectively. We provide a 20% uncertainty for each EF_{LAC} based on contributing uncertainties. EF_{LAC} was found to be independent of an engine load proxy, which is not consistent with limited previous data [*Petzold et al.*, 2004]. EF_{LAC} and EF_{CO} were correlated for MSD vessels indicating LAC emission related to engine efficiency. EF_{LAC} for MSD (mostly tug boats) were double any other class; significant for local air quality near busy ports and vessel traffic lanes. EF_{LAC} for SSD vessels, were more than double that of limited previous estimates [*Sinha et al.*, 2003; *Petzold et al.*, 2008]. This difference and the variability in EF_{LAC} emphasize the difficulty in applying a single emission factor to all shipping. Given this variability, investigation of the links between EF_{LAC} and engine load on commercial shipping under a variety of conditions is needed to better quantify emissions; such measurements are planned for future field studies. Using shipping fuel consumption data of *Eyring et al.* [2005a] and our EF_{LAC} we calculate a global LAC contribution from shipping of 133(±27) Ggyr^{-1}, or ~1.7 % of total LAC for 2001, which compares favorably to one previous estimate currently used in many global models [*Bond et al.*, 2004]. Although contributing ~1.7% of total LAC we point out that shipping tends to be concentrated in trade routes and ports near populated areas, evidenced by the largest absolute (20–50 ng m^{-3}) and relative (up to 40%) increases in LAC in Figures 3b and 3c. Future emission increases into sensitive areas, such as the Arctic may produce substantial local effects through climate feedback mechanisms. Thus the small fraction of LAC emitted globally by shipping may mask the considerable significance for climate, air quality, and health on local and regional scales.

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