Supplementary Information for "Black carbon emissions from in-use ships: a California regional assessment"

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Overview

The supplementary information provides: (1) details of the correction to the light absorption coefficient measured by the PSAP (2) a description of the weighted Pearson's r^2 calculations for the instrument comparison, (3) tables showing the EF_{BC} determined for each ship plume encounter, (4) a figure of the average normalized, number-weighted and mass-weighted size distributions for the rBC component of particles with a size-independent detection efficiency, (5) examples of time-series of black carbon (BC) and CO_2 concentrations across plumes and (6) consideration of the relationship between the absolute emission factors and the in-plume signal levels.

1 Correction for the PSAP

The light absorption coefficient of the particles deposited on the glass fibre filter of the PSAP is determined from the Beer-Lambert Law, after correction for instrument response and scattering within and by particles on the filter. Specifically,

$$\boldsymbol{b_{abs}(\lambda)} = \frac{\boldsymbol{0.85 \cdot \left(\frac{\lambda}{\lambda_0}\right)^{-1} \cdot b_{PSAP}(\lambda_0)}}{\boldsymbol{K_2}} - \frac{\boldsymbol{K_1 b_{sp}(\lambda)}}{\boldsymbol{K_2}}, \quad \text{(S1)}$$

where 0.85 is a filter area correction factor, K_2 (=1.2 ± 0.20) is the instrument response factor to absorption, K_1 (0.02 ± 0.02) is the instrument response to scattering, $b_{sp}(\lambda)$ is the scattering coefficient measured by a nephelometer at wavelength λ, and $b_{PSAP}(\lambda)$ is the absorption coefficient reported by the PSAP at wavelength λ_0 (Ogren, 2010;Bond et al., 1999). The ratio (λ/λ_0) in the first term adjusts the absorption coefficient from wavelength λ_0 (the measurement wavelength) to wavelength λ (the wavelength at which the nephelometer measured scattering), assuming an absorption Angstrom exponent (AAE) of 1 for the absorbing aerosol, where the AAE is defined as

$$AAE = \log\left(\frac{b_{abs,\lambda 1}}{b_{abs,\lambda 2}}\right) / \log\left(\frac{\lambda_1}{\lambda_2}\right) \quad \text{(S2)}$$

and where λ is wavelength. This particular PSAP operated at 3 different wavelengths, 467 nm, 530 nm, and 660 nm, while the nephelometer operated at 450 nm, 550 nm and 700 nm. Alternatively, the scattering coefficients could have been adjusted to the PSAP wavelengths using the observed scattering Angstrom exponent, defined similarly to the AAE. Use of the SAE to adjust the scattering would obviate the need to assume an AAE = 1, as was done above. However, we have confirmed that the AAE values determined from the PSAP measurements in the ship plumes were very close to unity (average = 1.04 ± 0.09), and thus the adjustment of the PSAP absorption coefficients to the nephelometer wavelengths is justified. Further, the use of AAE = 1.04 instead of 1 leads to differences of < 0.2% in the extrapolated b_{abs}, much smaller than the overall measurement uncertainty.

2 Weighted Pearson's r^2

Weighted Pearson's r^2 values were calculated for each instrument comparison, where the uncertainties of both instruments were used to weight the average x and y values in the calculation. The r^2 value is calculated as:

$$\boldsymbol{r^2} = \left(\frac{\sum(\boldsymbol{x_i}-\bar{\boldsymbol{x}})(\boldsymbol{y_i}-\bar{\boldsymbol{y}})}{\sqrt{\sum(\boldsymbol{x_i}-\bar{\boldsymbol{x}})^2(\boldsymbol{y_i}-\bar{\boldsymbol{y}})^2}}\right)^{\boldsymbol{2}} \qquad \text{(S3)}$$

where $\bar{x}$ and $\bar{y}$ are the uncertainty-weighted average EF_{BC} values for each of the instruments being compared, and x_i and y_i are the individual EF_{BC} values for each instrument. The uncertainty-weighted average is determined by:

$$\bar{\boldsymbol{x}} = \frac{\sum \boldsymbol{w_i x_i}}{\sum \boldsymbol{w_i}}, \qquad \text{(S4)}$$

where w_i is the weighting factor, calculated as

$$\mathbf{w_i} = \frac{\mathbf{1}}{\boldsymbol{\sigma}_\mathbf{i}}, \qquad \text{(S5)}$$

where σ_i is the uncertainty for a given EF_{BC}.

3 Vessel-specific BC Emission Factors

Vessel-specific BC emission factors were determined from each instrument measurement and then averaged and weight-averaged to get an overall EF_{BC} for each plume encounter. Not every instrument was sampling during each invidual plume intercept. Emission factors from vessels with slow speed diesel engines are shown in Table S1, from vessels with medium speed diesel engines in Table S2, and from vessels with high speed diesel engines in Table S3.

4 Black carbon size distributions

BC-specific size distributions determined from the SP2 instrument assuming a size-independent detection efficiency (= 0.7) are shown in Figure S1. These distributions can be compared with those shown in the main text, for which a size-dependent detection efficiency was used (Cappa et al., 2013).

5 Example plumes

Three independent plumes were intercepted from the slow speed diesel (SSD) vessel Dream Orchid. During the first intercept the Dream Orchid was travelling at ~7.4 knots during emission (Table S1) and was accelerating. During the second two intercepts the Dream Orchid was travelling at a steady ~11 knots. The weighted average EF_{BC} determined from the first, second and third plume intercepts were 2.9, 0.23 and 0.28 g-BC (kg-fuel)$^{-1}$. The absolute BC concentrations in the three plumes were quite different, with the concentration during the first plume much higher than the subsequent two. In contrast, the CO_2 concentrations were much more comparable in magnitude. Thus, the three Dream Orchid intercepts illustrate visually the temporal behaviour across a plume for cases of (i) large vs. small signals and (ii) large vs. small emission factors. Time-series of each of the intercepts are shown in Fig. S2.

6 Relationship between emission factors and absolute signal levels

The emission factors in this study were determined from the ratio between the area under the plumes for [BC] and [CO_2]. Although all plumes for which *EF*s are reported were subject to the constraint that the in-plume signal were above the plume-specific detection limit, defined here as $3\sigma/\mathrm{sqrt}(N)$ with σ the standard deviation across the background period and N the number of points across the plume, there is some utility in examining whether relationships exist between the determined emission factors and the absolute signal levels associated with BC and CO_2 in the plume and the standard deviation of the background signals. We use here measurements from the SP2 as an example, but given the good correspondence (i.e. large r^2 values) between the SP2 measurements and the other BC measurement techniques (Fig. 2 in the main text) these results can be considered general. The observationally determined BC *EF*s from the SP2 for all intercepts are shown versus both the background-substracted (i) area under the [rBC] time-series, A_{BC} and (ii) area under the [CO_2] time-series, A_{CO2}. There is some correspondence between the observed EF_{BC} and A_{BC} but no relationship between EF_{BC} and A_{CO2} (Fig. S3). Further, the BC *EF*s are shown versus A_{BC} and A_{CO2} after normalization by the standard deviation in the background signal, σ_{bgd}. Similar to the areas, the $A_{BC}/\sigma_{bgd,BC}$ exhibits some correspondence with EF_{BC} whereas $A_{CO2}/\sigma_{bgd,CO2}$ exhibits minimal relationship.

References

Bond, T. C., Anderson, T. L., and Campbell, D.: Calibration and intercomparison of filter-based measurements of visible light absorption by aerosols, Aerosol Science and Technology, 30, 582-600, 1999.

Cappa, C. D., Williams, E. J., Lack, D. A., Buffaloe, G. M., Coffman, D., Hayden, K. L., Herndon, S. C., Lerner, B. M., Li, S.-M., Massoli, P., McLaren, R., Nuaaman, I., Onasch, T. B., and Quinn, P. K.: The influence of operating speed on gas and particle-phase shipping emissions: results from the NOAA Ship Miller Freeman, Atmos. Meas. Tech., submitted, 2013.

Ogren, J. A.: Comment on “Calibration and Intercomparison of Filter-Based Measurements of Visible Light Absorption by Aerosols”, Aerosol Science and Technology, 44, 589-591, 2010, doi:10.1080/02786826.2010.482111.

Table S1. Black Carbon Emission Factors from Ships with Slow Speed Diesel Engines

Ship Name	Speed (kts)	PAS EF_{BC} g-BC (kg fuel)$^{-1}$	PSAP EF_{BC} g-BC (kg fuel)$^{-1}$	SP2 EF_{BC} g-BC (kg fuel)$^{-1}$	SP-AMS EF_{BC} g-BC (kg fuel)$^{-1}$	Weighted Average EF_{BC} g-BC (kg fuel)$^{-1}$	Average EF_{BC} g-BC (kg fuel)$^{-1}$
				Cargo Ships			
DREAM ORCHID	7.4	2.9 ± 0.38	2.9 ± 0.42	2.2 ± 0.55		2.9 ± 0.3	2.9 ± 0.4
DREAM ORCHID	11.2	0.52 ± 0.16	0.22 ± 0.033	0.2 ± 0.051		0.23 ± 0.03	0.37 ± 0.09
DREAM ORCHID	11		0.28 ± 0.04	0.2 ± 0.051		0.28 ± 0.04	0.28 ± 0.04
E.R. DARWIN	20		0.18 ± 0.03	0.067 ± 0.017		0.18±0.03	0.18 ± 0.03
VERRAZANO BRIDGE	8.1	0.21 ± 0.045	0.43 ± 0.063	0.21 ± 0.054		0.28 ± 0.04	0.32 ± 0.05
MOL ENDURANCE*	12	0.17 ± 0.036	0.16 ± 0.04	0.071 ± 0.018	0.16 ± 0.054	0.16 ± 0.02	0.17 ± 0.04
SUN RIGHT	12.1	0.74 ± 0.099	0.73 ± 0.11	0.49 ± 0.12	0.79 ± 0.24	0.74 ± 0.07	0.74 ± 0.2
SUN RIGHT	11.3	0.64 ± 0.095	0.66 ± 0.096	0.47 ± 0.12	0.71 ± 0.22	0.66 ± 0.07	0.7 ± 0.1
MSC CARACAS	11.6	0.42 ± 0.1	0.49 ± 0.071	0.28 ± 0.071		0.47 ± 0.06	0.45 ± 0.09
YM ORCHID*	0	6.1 ± 0.8	5.3 ± 0.76	3 ± 0.77		5.7 ± 0.4	5.7 ± 0.6
YM ORCHID	11.5		0.21 ± 0.031			0.21 ± 0.03	0.21 ± 0.03
AS PALATIA	11.4	0.25 ± 0.074	0.16 ± 0.024	0.11 ± 0.028		0.17 ± 0.02	0.21 ± 0.05
HANJIN BEIJING	11.6	0.21 ± 0.041	0.12 ± 0.017	0.072 ± 0.019		0.13 ± 0.02	0.16 ± 0.03
EVER SUMMIT	18.8			0.0042 ± 0.0025			
MARGRETHE MAERSK	12.1	0.12 ± 0.024	0.12 ± 0.017	0.05 ± 0.013	0.07 ± 0.024	0.11 ± 0.01	0.10 ± 0.02
MARGRETHE MAERSK	12.4	0.094 ± 0.015	0.079 ± 0.012	0.049 ± 0.012	0.08 ± 0.025	0.084 ± 0.009	0.08 ± 0.02
MARGRETHE MAERSK	12.3	0.1 ± 0.015	0.098 ± 0.014	0.048 ± 0.012	0.06 ± 0.019	0.092 ± 0.009	0.09 ± 0.02
MARGRETHE MAERSK	12.1	0.13 ± 0.02	0.089 ± 0.013	0.046 ± 0.012	0.08 ± 0.024	0.10 ± 0.01	0.10 ± 0.02
MARGRETHE MAERSK	12		0.25 ± 0.037	0.053 ± 0.015	0.14 ± 0.058	0.22 ± 0.03	0.20 ± 0.05
PACIFIC LINK	12	0.12 ± 0.034	0.1 ± 0.015			0.10 ± 0.01	0.11 ± 0.02
ZIM MEDITERRANEAN	8.2	0.87 ± 0.14	0.9 ± 0.23	0.86 ± 0.23		0.9 ± 0.1	0.9 ± 0.2
HORIZON TIGER*	19		0.11 ± 0.016	0.016 ± 0.016		0.11 ± 0.02	0.11 ± 0.02
LT CORTESIA	0	1 ± 0.14	1 ± 0.15	0.66 ± 0.17		1.0 ± 0.1	1.0 ± 0.1
WUXI DRAGON	10.1		0.26 ± 0.038	0.12 ± 0.031		0.26 ± 0.04	0.26 ± 0.04
STELLA PRIMA	5.7	0.47 ± 0.045	0.6 ± 0.062	0.38 ± 0.094		0.52 ± 0.04	0.54 ± 0.05
HK CHALLENGER	11.4	0.42 ± 0.088	0.085 ± 0.012	0.046 ± 0.016	0.11 ± 0.056	0.09 ± 0.01	0.20 ± 0.05
MATHILDE MAERSK	11.2		0.31 ± 0.046	0.17 ± 0.044		0.31 ± 0.05	0.31 ± 0.05

*denotes average of ships sampled consecutively, at the same speed

Table S1. Continued

Ship Name	**Speed (kts)**	**PAS EF_{BC} g-BC (kg fuel)$^{-1}$**	**PSAP EF_{BC} g-BC (kg fuel)$^{-1}$**	**SP2 EF_{BC} g-BC (kg fuel)$^{-1}$**	**SP-AMS EF_{BC} g-BC (kg fuel)$^{-1}$**	**Weighted Average EF_{BC} g-BC (kg fuel)$^{-1}$**	**Average EF_{BC} g-BC (kg fuel)$^{-1}$**
			Tanker				
CORPUS CHRISTI	11.7	0.21 ± 0.032	0.19 ± 0.027	0.088 ± 0.023		0.20 ± 0.02	0.20 ± 0.03
ANDES	0	0.46 ± 0.15	0.97 ± 0.15	0.26 ± 0.071		0.7 ± 0.1	0.7 ± 0.2
TAIPAN	0.1	0.89 ± 0.12	0.78 ± 0.11	0.83 ± 0.21		0.83 ± 0.08	0.8 ± 0.1
CAP ROMUALD	14.7	0.24 ± 0.099	0.2 ± 0.029	0.066 ± 0.017	0.17 ± 0.077	0.20 ± 0.03	0.20 ± 0.07
BUNGA KELANA 10	0		0.17 ± 0.025	0.097 ± 0.035		0.17 ± 0.03	0.17 ± 0.03
BUNGA KELANA 10	0		0.097 ± 0.014	0.086 ± 0.027		0.10 ± 0.01	0.10 ± 0.01
BRITISH PURPOSE*	0.1	0.21 ± 0.043	0.2 ± 0.029	0.051 ± 0.014		0.14 ± 0.02	0.21 ± 0.03
BRITISH PURPOSE	0.1		0.11 ± 0.016	0.26 ± 0.071		0.11 ± 0.02	0.11 ± 0.02

*denotes average of ships sampled consecutively, at the same speed

Table S2. Black Carbon Emission Factors from Ships with Medium Speed Diesel Engines

Ship Name	Speed (kts)	PAS EF_{BC} g-BC (kg fuel)$^{-1}$	PSAP EF_{BC} g-BC (kg fuel)$^{-1}$	SP2 EF_{BC} g-BC (kg fuel)$^{-1}$	SP-AMS EF_{BC} g-BC (kg fuel)$^{-1}$	Weighted Average EF_{BC} g-BC (kg fuel)$^{-1}$	Average EF_{BC} g-BC (kg fuel)$^{-1}$
			Fishing				
MILLER FREEMAN	11.9	0.46 ± 0.056	0.48 ± 0.053	0.15 ± 0.038	0.5 ± 0.155	0.47 ± 0.04	0.48 ± 0.09
MILLER FREEMAN	4.8	0.29 ± 0.063	0.34 ± 0.088	0.18 ± 0.049		0.31 ± 0.05	0.32 ± 0.08
MILLER FREEMAN	2.9	0.09 ± 0.0094	0.04 ± 0.0051	0.02 ± 0.006	0.08 ± 0.0248	0.052 ± 0.004	0.07 ± 0.01
MILLER FREEMAN	6.9	0.14 ± 0.022	0.18 ± 0.024	0.08 ± 0.02	0.18 ± 0.0558	0.16 ± 0.02	0.17 ± 0.03
MILLER FREEMAN	6.9	0.23 ± 0.031	0.2 ± 0.031	0.11 ± 0.03	0.16 ± 0.0496	0.21 ± 0.02	0.2 ± 0.04
MILLER FREEMAN	10.2	0.43 ± 0.054	0.48 ± 0.059	0.24 ± 0.06	0.4 ± 0.124	0.45 ± 0.04	0.44 ± 0.08
			Tow				
LAGUNA	7.5		0.34 ± 0.05	0.13 ± 0.04		0.34 ± 0.05	0.34 ± 0.05
MIKE BRUSCO	11	0.15 ± 0.021	0.22 ± 0.032	0.09 ± 0.02		0.17 ± 0.02	0.18 ± 0.03
MIKE BRUSCO	4.2	1.5 ± 0.2	2.2 ± 0.32	0.9 ± 0.2		1.7 ± 0.2	1.9 ± 0.3
ROBYN J	6	0.25 ± 0.054	0.22 ± 0.033	0.14 ± 0.04		0.23 ± 0.03	0.24 ± 0.04
REBEL2	7.9	1.5 ± 0.21	1.4 ± 0.21	0.13 ± 0.03		1.4 ± 0.2	1.4 ± 0.21
REBEL2	7.8	1.2 ± 0.18	1.2 ± 0.18	1.0 ± 0.3		1.2 ± 0.1	1.2 ± 0.2
MIKIONA	6.8		0.13 ± 0.018	0.06 ± 0.02		0.13 ± 0.02	0.13 ± 0.02
			Tug				
LEADER	7.7		0.25 ± 0.063	0.07 ± 0.02		0.25 ± 0.06	0.25 ± 0.06
TIM QUIGG	0.1		0.11 ± 0.017	0.10 ± 0.02		0.11 ± 0.02	0.11 ± 0.02
JEFFREY M*	8.6		0.48 ± 0.07	0.44 ± 0.08		0.5 ± 0.07	0.5 ± 0.07
GOLIAH	7.2	0.38 ± 0.05	0.32 ± 0.046			0.35 ± 0.03	0.35 ± 0.05
ROYAL MELBOURNE	4.4	5.4 ± 0.73	2 ± 0.28	2.4 ± 0.61		2.4 ± 0.3	3.7 ± 0.5
DELTA CAPTAIN	7.8	0.25 ± 0.033	0.71 ± 0.1	0.15 0.04		0.3 ± 0.03	0.48 ± 0.07

*denotes average of multiple plumes sampled consecutively, at the same speed

Table S3. Black Carbon Emission Factors from Ships with High Speed Diesel Engines

Ship Name	Speed (kts)	PAS EF_{BC} g-BC (kg fuel)$^{-1}$	PSAP EF_{BC} g-BC (kg fuel)$^{-1}$	SP2 EF_{BC} g-BC (kg fuel)$^{-1}$	SP-AMS EF_{BC} g-BC (kg fuel)$^{-1}$	Weighted Average EF_{BC} g-BC (kg fuel)$^{-1}$	Average EF_{BC} g-BC (kg fuel)$^{-1}$
			High Speed Craft				
JET CAT EXPRESS	28.4		0.48 ± 0.07	0.21 ± 0.06		0.48 ± 0.07	0.48 ± 0.07
JET CAT EXPRESS	28.4				0.44 ± 0.45	0.4 ± 0.5	0.4 ± 0.5
JET CAT EXPRESS	32.1	0.11 ± 0.099	0.11 ± 0.02	0.1 ± 0.03		0.11 ± 0.02	0.11 ± 0.02
JET CAT EXPRESS	28.8	0.16 ± 0.078	0.12 ± 0.02	0.095 ± 0.03		0.12 ± 0.02	0.14 ± 0.05
CATALINA JET	25.5		0.5 ± 0.08	0.23 ± 0.07		0.50 ± 0.08	0.5 ± 0.08
CATALINA JET	21.4		0.74 ± 0.19			0.74 ± 0.19	0.74 ± 0.19
CATALINA JET	26.3	0.8 ± 0.1	0.44 ± 0.07	0.43 ± 0.11	0.83 ± 0.29	0.55 ± 0.05	0.69 ± 0.15
CATALINA JET	24.6	0.44 ± 0.066	0.43 ± 0.06	0.37 ± 0.09		0.43 ± 0.05	0.43 ± 0.06
CATALINA JET	29	0.25 ± 0.13	0.17 ± 0.03	0.13 ± 0.04		0.18 ± 0.03	0.21 ± 0.08
STARSHIP EXPRESS	27.7		0.18 ± 0.06	0.11 ± 0.03		0.18 ± 0.06	0.18 ± 0.06
STARSHIP EXPRESS	7		0.46 ± 0.07	0.26 ± 0.26		0.46 ± 0.07	0.46 ± 0.07
STARSHIP EXPRESS	30.1		0.69 ± 0.1	0.57 ± 0.16		0.69 ± 0.10	0.69 ± 0.10
CAPT.T.LE	15.9		1.1 ± 0.15	0.8 ± 0.2		1.1 ± 0.2	1.1 ± 0.2
			Pilot Boat				
PILOT BOAT WHITE	14.8		0.71 ± 0.11			0.71 ± 0.1	0.71 ± 0.1
PILOT BOAT WHITE	0		0.19 ± 0.03	0.11 ± 0.03		0.19 ± 0.03	0.19 ± 0.03
PILOT BOAT BANNING	5.5	0.9 ± 0.2		0.63 ± 0.16		0.90 ± 0.2	0.9 ± 0.2
PILOT BOAT DRAKE	7		0.27 ± 0.04	0.031 ± 0.01		0.27 ± 0.04	0.27 ± 0.04
P/V GOLDEN GATE	19.2		0.12 0.03	0.07 ± 0.02		0.12 ± 0.03	0.12 ± 0.03

Table S3. Continued

Ship Name	Speed (kts)	PAS EF_{BC} g-BC (kg fuel)$^{-1}$	PSAP EF_{BC} g-BC (kg fuel)$^{-1}$	SP2 EF_{BC} g-BC (kg fuel)$^{-1}$	SP- AMS EF_{BC} g-BC (kg fuel)$^{-1}$	Weighted Average EF_{BC} g-BC (kg fuel)$^{-1}$	Average EF_{BC} g-BC (kg fuel)$^{-1}$
				Passenger			
ALAN T	0.2		3.3 ± 0.48			3.3 ± 0.48	3.3 ± 0.48
MAJESTIC	11.8		0.06 ± 0.02	0.034 ± 0.013		0.06 ± 0.02	0.06 ± 0.02
MARINER OF THE SEAS	18		0.05 ± 0.01	0.036 ± 0.01		0.049 ± 0.007	0.049 ± 0.007
MARINER OF THE SEAS	15.7	0.1 ± 0.014	0.1 ± 0.02	0.075 ± 0.02		0.10 ± 0.01	0.10 ± 0.01
CAT EXPRESS	26	0.51 ± 0.075	0.49 ± 0.07	0.34 ± 0.09		0.5 ± 0.05	0.5 ± 0.07
NAPA	28.9	0.24 ± 0.032	0.17 ± 0.03	0.1 ± 0.03		0.20 ± 0.02	0.21 ± 0.03
SOLANO	33.4	0.51 ± 0.07	0.49 ± 0.07	0.32 ± 0.08		0.5 0.05	0.5 ± 0.07
MARIN	22.1	0.49 ± 0.064	0.45 ± 0.07	0.31 ± 0.08		0.47 ± 0.05	0.47 ± 0.06
ENCINAL	22.9	1 ± 0.14	0.83 ± 0.12	0.63 ± 0.16		0.91 ± 0.09	0.93 ± 0.13
				Cruise Ship			
CARNIVAL SPLENDOR	20.6	0.11 ± 0.022	0.08 ± 0.01	0.024 ± 0.008	0.067 ± 0.03	0.08 ± 0.01	0.09 ± 0.02
CARNIVAL SPLENDOR	11.1	0.047 ± 0.016	0.04 ± 0.01	0.022 ± 0.006		0.044 ± 0.006	0.04 ± 0.01
CARNIVAL PARADISE	12.3	0.2 ± 0.059	0.07 ± 0.01	0.05 ± 0.01	0.14 ± 0.05	0.08 ± 0.01	0.14 ± 0.04

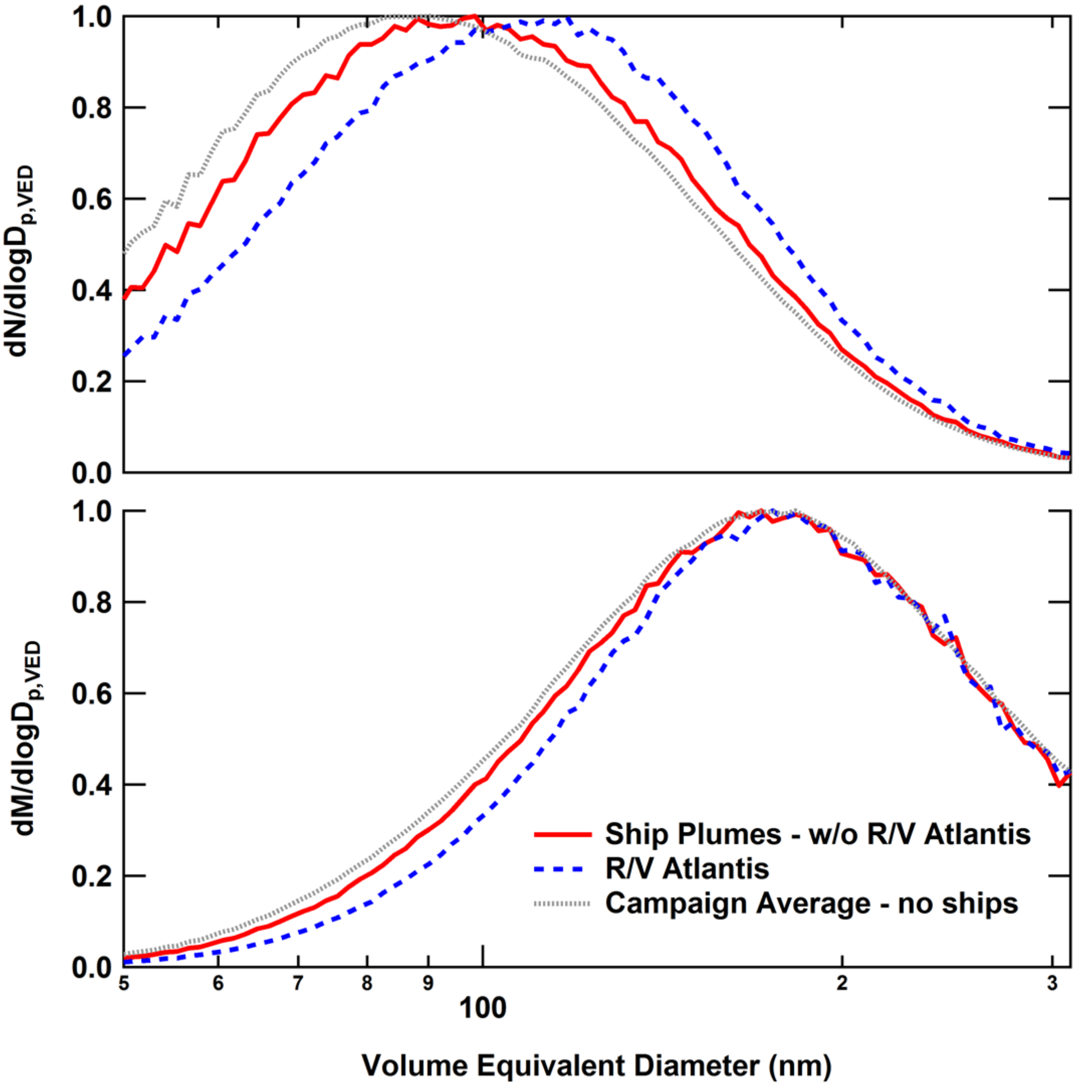

Figure S1. Average normalized, number-weighted (top) and mass-weighted (bottom) size distributions for the rBC component of particles. These are the same size distributions as shown in Figure 3 in the main text, but where a size-independent detection efficiency of 0.7 has been applied.

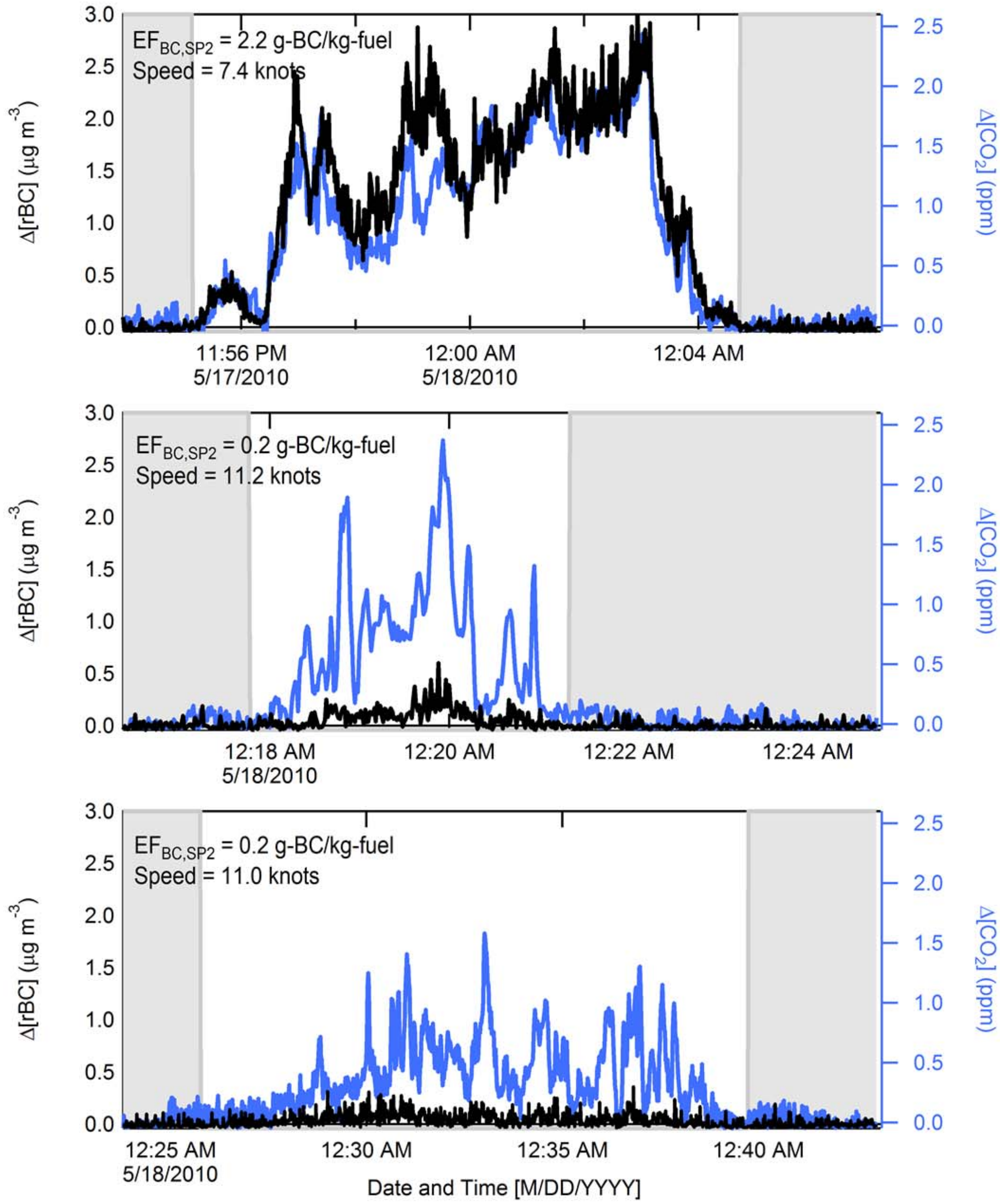

Figure S2. Time-series of Δ[rBC] (black, left axis) from the SP2 instrument and Δ[CO_2] (blue, right axis) for three plume intercepts for emissions from the vessel Dream Orchid. The white areas indicate the plume period and the gray the background period. The Δ indicate that the reported concentrations have been background-subtracted.

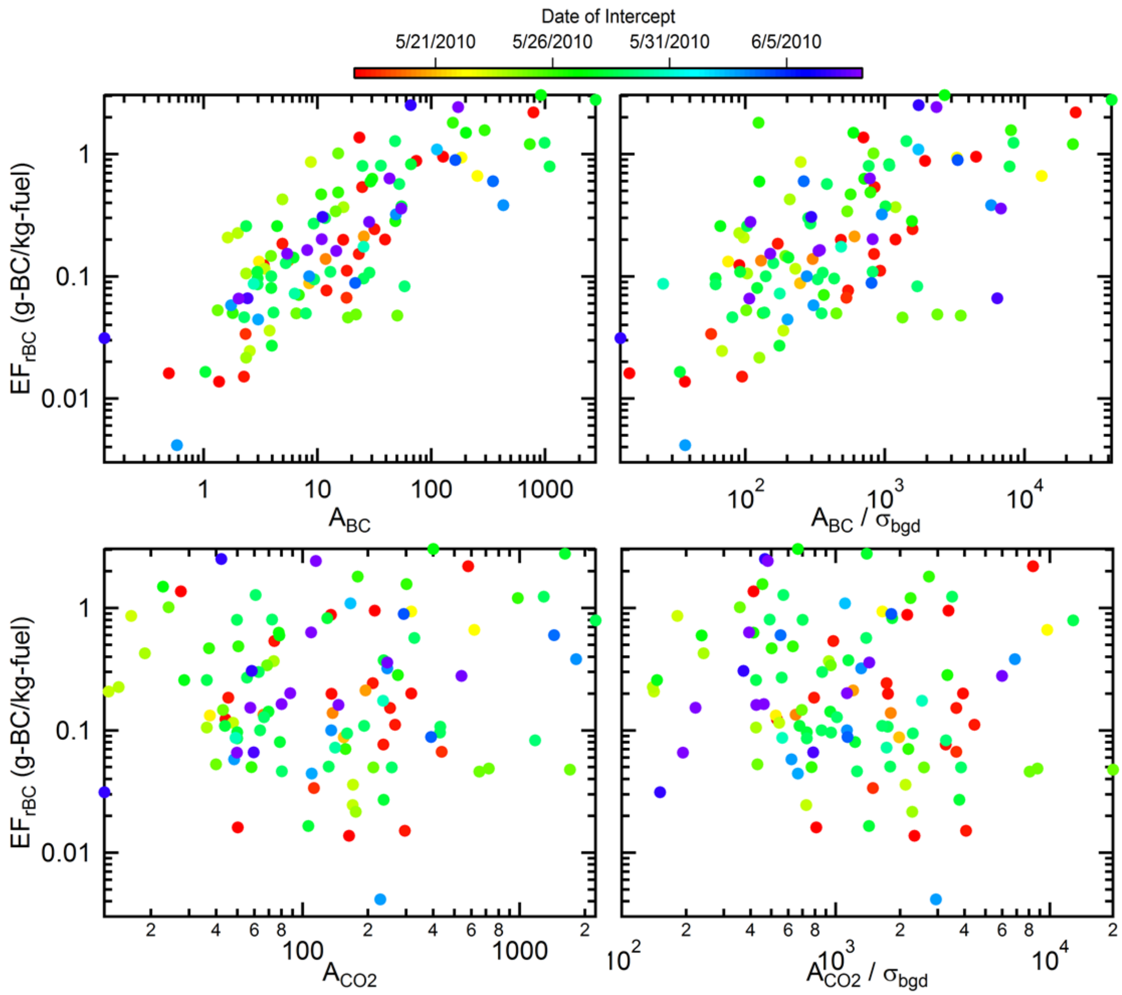

Figure S3. The observed BC emission factor for each plume intercept, as determined from the SP2 instrument as a function of the area under the background subtracted (top, left) [rBC] and (bottom, left) [CO_2] time-series, or the background subtracted areas normalized by the standard deviation in the signal during the background period (right panels). Points are colored according to the time of the plume intercept.