

Characteristics of black carbon aerosol from a surface oil burn during the Deepwater Horizon oil spill

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[1] Black carbon (BC) aerosol mass mixing ratio and microphysical properties were measured from the NOAA P-3 aircraft during active surface oil burning subsequent to the Deepwater Horizon oil rig explosion in April 2010. Approximately 4% of the combusted material was released into the atmosphere as BC. The total amount of BC introduced to the atmosphere of the Gulf of Mexico via surface burning of oil during the 9-week spill is estimated to be $(1.35 \pm 0.72) \times 10^6$ kg. The median mass diameter of BC particles observed in the burning plume was much larger than that of the non-plume Gulf background air and previously sampled from a variety of sources. The plume BC particles were internally mixed with very little non-refractory material, a feature typical of fresh emissions from fairly efficient fossil-fuel burning sources and atypical of BC in biomass burning plumes. BC dominated the total accumulation-mode aerosol in both mass and number. The BC mass-specific extinction cross-section was 10.2 ± 4.1 and 7.1 ± 2.8 m²/g at 405 and 532 nm respectively. These results help constrain the properties of BC emissions associated with DWH and other large spills. **Citation:** Perring, A. E., et al. (2011), Characteristics of black carbon aerosol from a surface oil burn during the Deepwater Horizon oil spill, *Geophys. Res. Lett.*, 38, L17809, doi:10.1029/2011GL048356.

1. Introduction

[2] The explosion of the Deepwater Horizon (DWH) oil rig on 20 April 2010 resulted in the release of massive amounts of crude oil into the water column at a depth of ~1500 m and ~80 km offshore [Camilli et al., 2010]. A large portion of the oil accumulated at the surface where one of the main remediation strategies was controlled surface burning. An estimated 5% of the total leaked oil was burned [Lehr et al., 2010]. This led to the direct atmospheric emission of black carbon (BC), the refractory and most efficiently light absorbing component of soot, which affects both climate and human health. Such burns provide case studies to evaluate the microphysical properties (size, coating state, extinction efficiency) of BC aerosol and the overall yield of BC.

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[3] The particle size and the presence of internally mixed non-BC material (the “coating state”) influence the likelihood of BC removal through precipitation, which, in turn, affects the net mass loading and climate forcing impacts of BC in the atmosphere. Further, the mass-normalized absorption cross section of BC cores can be enhanced by up to 50% by the presence of non-refractory, non-absorbing coatings [Bond et al., 2006]. In addition to climate impacts, epidemiological studies indicate that particle number, surface area and the presence of organic coatings may be more important for assessing human health impacts than overall particle mass [Lighty et al., 2000]. These parameters generally cannot be determined with filter-based BC measurements. The properties of BC-containing aerosol, as derived from the single-particle data presented here, will provide models with detailed information relevant to predictions of the atmospheric lifetime and climate impacts of surface-oil burn BC for use in impact assessments of this oil spill remediation strategy.

[4] Here we describe observations of BC and other combustion products collected during a flight of the NOAA P-3 aircraft in the Gulf of Mexico on 8 June 2010. We focus on a single plume transect that occurred at 985 m altitude and ~5 km from an active burn site in which observed BC, CO and CO₂ concentrations were elevated well above background levels. We present emission ratios of CO, CO₂ and BC as well as the mass and number distributions, and coating states of the BC-containing particles produced.

2. Methods

[5] A map of the aircraft flight track is shown in Figure 1. The main plume intersection and background sampling regions are also shown. The aerosol and trace gas concentrations observed during the flight are publicly available at www.esrl.noaa.gov/csd/tropchem/2010gulf/ and have been discussed previously [de Gouw et al., 2011; Ryerson et al., 2011].

[6] BC in the atmosphere is generally found mixed with other materials within individual particles. Here this material is generically referred to as a “coating” and the BC-component as a “BC core”. Individual BC cores were measured using a Single Particle Soot Photometer (SP2) instrument [Schwarz et al., 2008a]. Briefly, an air jet containing ambient aerosol intersects an intense intracavity laser beam ($\lambda = 1064$ nm). The evolution of laser light scattered by individual BC-containing particles is used to identify the presence of coatings on BC cores and to optically size the unperturbed but dry particles prior to significant laser heating. After a particle with a BC core enters the laser beam, any associated

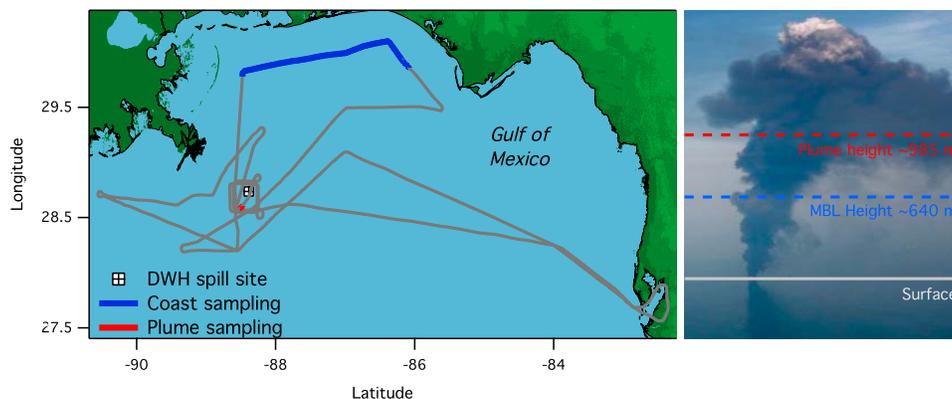


Figure 1. (left) P-3 Flight track on 8 June 2010. The white square shows the spill site. The main P-3 plume encounter is shown in red and the location of non-plume background sampling along the coast in blue. (right) Photograph of the oil burn plume taken from the P-3 with altitudes indicated for the boundary layer (determined using water vapor and temperature profiles) and plume encounter heights.

coating vaporizes and the BC component is then heated to its vaporization temperature (~ 4200 K). The hot BC material emits an amount of thermal radiation proportional to its refractory mass. The SP2 quantifies BC-core mass without sensitivity to its association with coatings or morphology [Cross *et al.*, 2010].

[7] The instrument as configured here quantified BC cores in a mass range of 0.7 to 1000 fg BC (a size range of 90–1000 nm volume equivalent diameter assuming a density of 2 g/cm^3). Summation of the individual BC-core masses within a measured quantity of air yields ambient mass mixing ratios with an uncertainty of $\sim 40\%$ dominated by calibration uncertainties. Calibrations of SP2 sensitivity to BC were performed using size-selected fullerene soot (Alfa Aesar Inc., Woodhill, MA) of 0.5–30 fg/particle and were stable to better than 5% between 2 May and 29 June. The SP2 response is non-linear for particles with high BC mass (>100 fg). An empirical correction for the non-linearity was applied based on instrument response data presented by Moteki and Kondo [2010]. Since the majority of observed BC mass was within the linear calibration range, this correction is small ($<5\%$ in the plume, typically $<1\%$). There was no bias due to coincident laser beam transits by multiple particles.

[8] Mixing ratios of CO and CO₂ were measured at 1 Hz with accuracies of $\pm 5\%$ and ± 0.25 ppm respectively. CO was detected with vacuum-ultraviolet resonance fluorescence [Holloway *et al.*, 2000] and CO₂ with a Picarro cavity-ring-down instrument [Chen *et al.*, 2010]. A wide suite of volatile organic carbon (VOC) compounds was measured using proton transfer mass spectrometry ($\pm 30\%$) [de Gouw and Warneke, 2007] and using gas chromatography on canister samples collected during flight ($\pm 5\%$) [Colman *et al.*, 2001]. The NMASS and UHSAS particle counters for particles in the range of 4 to 1000 nm diameter [Brock *et al.*, 2008], detected total submicron aerosol number ($\pm 15\%$) and volume ($\pm 40\%$) in the size range detected by the SP2. The aerosol extinction coefficient (σ_{ep}) was measured ($\pm 2\%$) in dried ambient air at 405 nm and 532 nm using a cavity ring-down spectrometer (CRDS) [Langridge *et al.*, 2011]. Non-refractory aerosol mass and composition was determined

($\pm 30\%$) at a 10 s resolution using an aerosol mass spectrometer (AMS) [Bahreini *et al.*, 2009].

3. Results

3.1. Combustion Efficiency of Surface Burning

[9] Deliberate burning of surface oil is a common method of spill remediation [Evans *et al.*, 2001]. Since previous observations of the products under ambient conditions have been sparse, product yields and particle characteristics are not well quantified. Although near-field sampling of plumes by the P-3 was complicated by flight restrictions near active burn zones, one intense plume was encountered and sampled for ~ 30 s at an altitude of 985 m. Enhancements were observed in BC, CO and CO₂ abundances, shown as a function of cross-wind distance in Figure 2 (left). The flight legs just prior to the plume transit had $\sim 100 \text{ ng BC/m}^3$ air, higher than typical continental background air. The largest enhancements correspond to 151 ng-C/g-air for BC and 200 ppbv (83 ng C/g-air), and 8 ppmv (3300 ng-C/g-air) for CO and CO₂ respectively. Units of carbon mass mixing ratios are given in parentheses for facility in calculating fractional yields.

[10] Observed enhancements of benzene (0.75 ppbv), ethane (1.5 ppbv) and ethyne (3.8 ppbv) are negligible with respect to the total carbon budget of the burn plume. The NOAA P-3 was not configured to measure gas-phase polycyclic aromatic hydrocarbons (PAHs) other than naphthalene, however a previous study of ocean-surface burns [Ross *et al.*, 1996] found that the sum of all non-methane VOCs in the gas- and particle phases comprised $<2\%$ of the carbon released to the atmosphere. Thus, the unmeasured carbon mass is presumed to be small. The aerosol mass of non-BC-containing particles estimated with the SP2 scattering channels indicates that BC is the principal ($>90\%$ by mass) aerosol product arising from the combusted oil. In addition the SP2 data show no evidence of coatings on the BC. Due to intermittent sampling, AMS data is unavailable during the most intense 3 s of plume sampling but, in support of the above conclusion, there is no discernible enhancement above background in total non-refractory aerosol mass during the wider

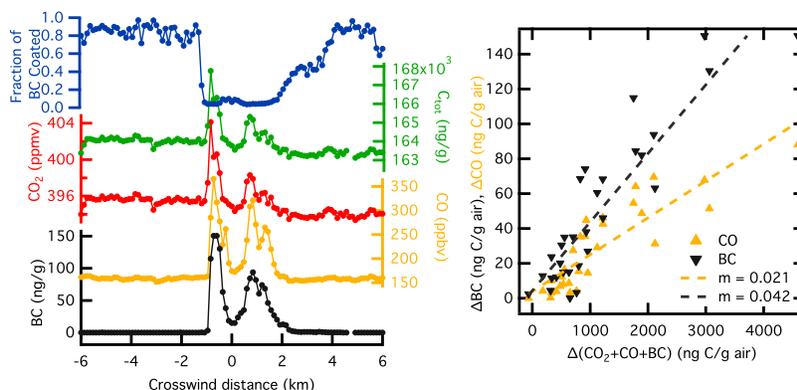


Figure 2. (left) Traces of BC (black), CO (yellow), CO₂ (red), total C (green), and BC coated fraction (blue) as a function of crosswind distance during plume transit (outlined by grey box). (right) BC (black, downward triangles) and CO (yellow, upward triangles) vs. total carbon emitted ($C_{\text{tot}} = \text{CO}_2 + \text{CO} + \text{BC}$). The slopes (m) of the regression lines yield the fractions of C_{tot} represented by BC (4.2%) and CO (2.1%) respectively.

transit. The combined abundance of BC, CO and CO₂ is therefore estimated to represent >98% of the carbon mass introduced to the atmosphere from this burning event.

[11] Orthogonal distance regressions of the plume data (Figure 2) indicate that $4.2 \pm 1.7\%$ of the measured atmospheric carbon released from the combusted oil was in the form of BC, $2.1 \pm 0.1\%$ was released as CO and the remainder as CO₂. The ranges given incorporate both the instrumental uncertainties and statistical fit uncertainties. These fractional yields are converted to emission factors (g-C produced/kg-fuel consumed) for comparison to previous work using the mass fraction of carbon in the fuel, 850 g/kg, equivalent to a hydrocarbon mixture with an average of 18 carbon atoms per molecule [Wang *et al.*, 2003]. This yields emission factors for the present study of 36 ± 14 g-C/kg-fuel for BC and 18 ± 1 g-C/kg-fuel for CO. By comparison, Ross *et al.* [1996] report a BC emission factor of 66 ± 18 g-C/kg-fuel for marine surface oil burning. Total particle emissions of 35–80 g-C/kg-fuel have also been reported for varying oil pool depths [Benner *et al.*, 1990] and of 60–180 g-C/kg-fuel for varying pool surface areas [Koseki and Mulholland, 1991]. Although the impact of weathering on surface-oil burn product yields is unknown we note that the combusted oil in the present work is likely more weathered than that used in previous studies of intentionally orchestrated surface-oil burns.

[12] From the P-3 data outlined above, we can estimate the total amount of BC introduced to the atmosphere in the Gulf during the period of active surface burning. The volume of oil burned between 20 April and 19 July is estimated to be 220,000–313,000 barrels (bbls) consumed in 410 separate burns [Schaum *et al.*, 2010]. If we then assume 1) an oil density of 0.9 g/cm³ and 2) 90% of the oil mass burned was released to the atmosphere [Evans *et al.*, 2001], we calculate the mass of BC emitted as follows:

$$\begin{aligned} \text{Oil burned (g)} &= (220 - 313 \times 10^3 \text{ bbls}) \times (1.6 \times 10^5 \text{ cm}^3/\text{bbl}) \\ &\quad \times (0.9 \text{ g/cm}^3) \\ &= 3.2 - 4.6 \times 10^{10} \text{ g} \end{aligned}$$

$$\begin{aligned} \text{BC emitted (g C)} &= (3.2 - 4.6 \times 10^{10} \text{ g}) \times (0.9) \\ &\quad \times (0.022 - 0.050 \text{ g-C-as-BC/g-oil}) \\ &= 0.63 - 2.07 \times 10^6 \text{ kg C} \end{aligned}$$

Our derived BC emission factor of 36 ± 14 g-C/kg-fuel therefore suggests that surface oil burning introduced $(1.35 \pm 0.72) \times 10^6$ kg of BC to the atmosphere during the 9-week period of active burning. For comparison, this is approximately equivalent to the total BC emissions of all the maritime shipping in the Gulf region over the same time period [C Wang *et al.*, 2008]. The microphysical properties of the BC-containing aerosol produced, however, differ substantially from those emitted from ships. In addition, the buoyant energy of the plume resulted in lofting above the marine boundary layer (Figure 1), higher than typical ship emission injection heights.

3.2. Microphysical Properties of BC

[13] Figure 3 shows mass and number distributions as functions of BC-core volume equivalent diameter (D_{Veq}) observed in the surface burn plume. The distributions are centered at larger mean diameters (388 nm and 206 nm for

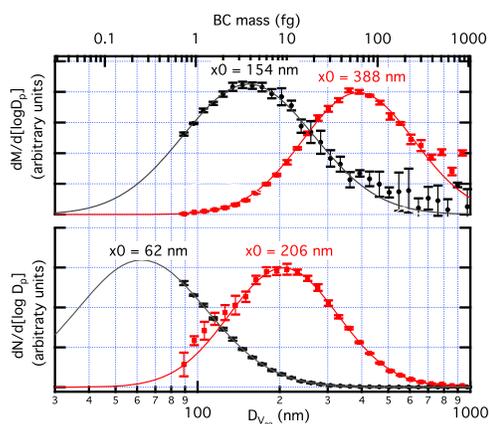


Figure 3. (top) BC mass (M) in log-normal diameter bins ($dM/d(\log D_p)$) vs volume equivalent diameter (D_{Veq}) for the burning oil plume (red) and for particles encountered along the coast (black). (bottom) BC number (N) in log-normal diameter bins ($dN/d(\log D_p)$) vs D_{Veq} from the plume (red) and the coastline (black). Distributions are fit with log-normal functions (solid traces) and labeled with the mean diameters (x_0). Error bars show standard deviation of separate distributions derived for quintiles of the data.

mass and number distributions respectively) than is typically seen in fresh urban emissions, biomass burning or aged aerosol [Moteki *et al.*, 2007; Schwarz *et al.*, 2008b]. The BC distributions observed off the coastline in the absence of distinct DWH burning are shown for comparison. Using a log-normal fit to the observed BC mode, the SP2 detects >95% of the mass and >90% of the number of BC cores in the accumulation mode within the plume. The particle counters show a population of smaller Aitken-mode particles (30–90 nm). If this mode constituted entirely of BC cores, its contribution to the overall BC mass in the plume would be negligible. The contribution to number could be significant.

[14] In the surface oil burn plume, BC-containing particles detected by the SP2 account for >90% of the total particle number measured by the UHSAS between 0.1 and 1 μm . This predominance of BC in the plume allows for the calculation of a mass-specific extinction efficiency from measured enhancements in the aerosol extinction (σ_{ep}) and the observed BC mass loadings. BC is typically a small fraction of total aerosol mass in the atmosphere and therefore rarely dominates σ_{ep} . The integrated enhancements in σ_{ep} across the plume were 13550 Mm^{-1} at 532 nm and 19420 Mm^{-1} at 405 nm. Dividing these numbers by the integrated enhancement in BC, $1900 \mu\text{g}/\text{m}^3$, yields BC mass-specific extinction (MSE) coefficients of $7.1 \pm 2.8 \text{ m}^2/\text{g}$ and $10.2 \pm 4.1 \text{ m}^2/\text{g}$ at 532 nm and 405 nm respectively. This is within the range (6.4–10.8 m^2/g) of previously reported values of MSE for fresh BC emissions from a variety of sources at 532 nm [Roessler and Faxvog, 1980]. It is lower than might be suggested by Mie theory and recent measurements of the mass specific absorption in the range of 6–8 m^2/g [e.g., Schwarz *et al.*, 2008b]. Given that the MSE is the sum of absorption and scattering, our results imply that either the scattering or the absorption is lower for the plume BC than expected for typical BC particles of comparable size.

[15] Basic coating information can be extracted from SP2 data using the relative timing of the peak incandescence signal and the peak scattering signal as outlined by Moteki and Kondo [2007]. Due to saturation of the scattered light detector the coating state can be determined only for BC cores <275 nm D_{veq} (57% of the BC cores in the plume), however there is no observed size dependence of coating state over the detectable range. We therefore assume that the coated number fraction for cores <275 nm D_{veq} is representative of the whole distribution of particles in the plume. Figure 2 shows the number fraction of particles classified as thickly coated during the plume transit. Just outside of the plume the thickly coated fraction is ~80% while within the plume almost none of the particles have a detectable coating and the thickly coated fraction is <5%. For comparison, 50–80% of BC particles observed in the background Gulf of Mexico, unaffected by DWH surface burning, are categorized as thickly coated.

[16] Based on Mie theory calculations as by Schwarz *et al.* [2008a] for a spherical shell-and-core configuration (assumed (1.95,0.79) core and (1.5,0) coating indices of refraction), a 200 nm D_{veq} BC core will be classified as “thickly coated” by this technique if it is coated by >15 nm of non-BC material. For a hydrocarbon with a density of $1 \text{ g}/\text{cm}^3$ a BC core of this size classified as uncoated thus comprises at least 79% of the total particle mass. Given that essentially

none of the BC particles observed in the plume showed any indication of being coated, this limiting case likely overestimates the actual amount of non-BC material associated with each core by a wide margin.

4. Summary

[17] Single-particle measurements of the BC mass mixing ratio and the microphysical state of BC-containing aerosol produced from controlled burning of surface oil from the Deepwater Horizon Spill were obtained using the NOAA SP2. BC is found to be $4.2 \pm 1.7\%$ and CO is found to be $2.1 \pm 0.1\%$ of the combustion carbon in the plume, which correspond to emission factors of 36 ± 14 and $18 \pm 1 \text{ g-C}/\text{kg-fuel}$ respectively. The total amount of BC introduced to the atmosphere via this pathway between 20 April and 19 July 2010 is estimated to be $(1.35 \pm 0.72) \times 10^6 \text{ kg}$. BC represented >95% of the total accumulation mode particle mass and mass-specific extinction cross-sections of 10.2 ± 4.1 and $7.1 \pm 2.8 \text{ m}^2/\text{g}$ at 405 and 532 nm, respectively, are derived. BC cores from fresh surface oil burning are found to be remarkably large compared to previously sampled urban and biomass burning emissions and most are associated with only small amounts of non-refractory materials. These properties influence the atmospheric lifetime and climate impact of BC emitted from surface oil burns. The overall yield and the microphysical properties of the BC aerosol presented here will inform the science and policy communities about the consequences of oil burns conducted in response to accidental oil releases in marine environments.

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References

- Bahreini, R., et al. (2009), Organic aerosol formation in urban and industrial plumes near Houston and Dallas, Texas, *J. Geophys. Res.*, *114*, D00F16, doi:10.1029/2008JD011493.
- Benner, B. A., et al. (1990), Polycyclic aromatic hydrocarbon emissions from the combustion of crude-oil on water, *Environ. Sci. Technol.*, *24*(9), 1418–1427, doi:10.1021/es00079a018.
- Bond, T. C., G. Habib, and R. W. Bergstrom (2006), Limitations in the enhancement of visible light absorption due to mixing state, *J. Geophys. Res.*, *111*, D20211, doi:10.1029/2006JD007315.
- Brock, C. A., et al. (2008), Sources of particulate matter in the northeastern United States in summer: 2. Evolution of chemical and microphysical properties, *J. Geophys. Res.*, *113*, D08302, doi:10.1029/2007JD009241.
- Camilli, R., et al. (2010), Tracking hydrocarbon plume transport and biodegradation at Deepwater Horizon, *Science*, *330*(6001), 201–204, doi:10.1126/science.1195223.
- Chen, H., et al. (2010), High-accuracy continuous airborne measurements of greenhouse gases (CO_2 and CH_4) using the cavity ring-down spectroscopy (CRDS) technique, *Atmos. Meas. Tech.*, *3*(2), 375–386, doi:10.5194/amt-3-375-2010.
- Colman, J. J., et al. (2001), Description of the analysis of a wide range of volatile organic compounds in whole air samples collected during PEM-Tropics A and B, *Anal. Chem.*, *73*(15), 3723–3731, doi:10.1021/ac010027g.
- Cross, E. S., et al. (2010), Soot particle studies instrument inter-comparison project overview, *Aerosol Sci. Technol.*, *44*(8), 592–611, doi:10.1080/02786826.2010.482113.
- de Gouw, J., and C. Warneke (2007), Measurements of volatile organic compounds in the earths atmosphere using proton-transfer-reaction mass spectrometry, *Mass Spectrom. Rev.*, *26*(2), 223–257, doi:10.1002/mas.20119.

- de Gouw, J. A., et al. (2011), Organic aerosol formation downwind from the Deepwater Horizon oil spill, *Science*, 331(6022), 1295–1299, doi:10.1126/science.1200320.
- Evans, D. D., et al. (2001), In situ burning of oil spills, *J. Res. Natl. Inst. Stand. Technol.*, 106(1), 231–278.
- Holloway, J. S., R. O. Jakoubek, D. D. Parrish, C. Gerbig, A. Volz-Thomas, S. Schmitgen, A. Fried, B. Wert, B. Henry, and J. R. Drummond (2000), Airborne intercomparison of vacuum ultraviolet fluorescence and tunable diode laser absorption measurements of tropospheric carbon monoxide, *J. Geophys. Res.*, 105(D19), 24,251–24,261, doi:10.1029/2000JD900237.
- Koseki, H., and G. W. Mulholland (1991), The effect of diameter on the burning of crude oil pool fires, *Fire Technol.*, 27(1), 54–65, doi:10.1007/BF01039527.
- Langridge, J. M., et al. (2011), Aircraft instrument for comprehensive characterization of aerosol optical properties, part I: Wavelength-dependent optical extinction and its relative humidity dependence measured using cavity ringdown spectroscopy, *Aerosol Sci. Technol.*, 45, 1305–1318, doi:10.1080/02786826.2011.592745.
- Lehr, B., et al. (2010), Oil budget calculator: Deepwater Horizon, technical documentation, Fed. Interagency Solut. Group, Washington, D. C.
- Lighty, J. S., et al. (2000), Combustion aerosols: Factors governing their size and composition and implications to human health, *J. Air Waste Manage. Assoc.*, 50(9), 1565–1618.
- Moteki, N., and Y. Kondo (2007), Effects of mixing state on black carbon measurements by laser-induced incandescence, *Aerosol Sci. Technol.*, 41(4), 398–417, doi:10.1080/02786820701199728.
- Moteki, N., and Y. Kondo (2010), Dependence of laser-induced incandescence on physical properties of black carbon aerosols: Measurements and theoretical interpretation, *Aerosol Sci. Technol.*, 44(8), 663–675, doi:10.1080/02786826.2010.484450.
- Moteki, N., Y. Kondo, Y. Miyazaki, N. Takegawa, Y. Komazaki, G. Kurata, T. Shirai, D. R. Blake, T. Miyakawa, and M. Koike (2007), Evolution of mixing state of black carbon particles: Aircraft measurements over the western Pacific in March 2004, *Geophys. Res. Lett.*, 34, L11803, doi:10.1029/2006GL028943.
- Roessler, D. M., and F. R. Faxvog (1980), Optical properties of agglomerated acetylene smoke particles at 0.5145- μ m and 10.6- μ m wavelengths, *J. Opt. Soc. Am.*, 70(2), 230–235, doi:10.1364/JOSA.70.000230.
- Ross, J. L., R. J. Ferek, and P. V. Hobbs (1996), Particle and gas emissions from an in situ burn of crude oil on the ocean, *J. Air Waste Manage. Assoc.*, 46(3), 251–259.
- Ryerson, T. B., et al. (2011), Atmospheric emissions from the Deepwater Horizon spill constrain air-water partitioning, hydrocarbon fate, and leak rate, *Geophys. Res. Lett.*, 38, L07803, doi:10.1029/2011GL046726.
- Schaum, J., et al. (2010), Screening level assessment of risks due to dioxin emissions from burning oil from the BP Deepwater Horizon Gulf of Mexico spill, *Environ. Sci. Technol.*, 44(24), 9383–9389, doi:10.1021/es103559w.
- Schwarz, J. P., et al. (2008a), Coatings and their enhancement of black carbon light absorption in the tropical atmosphere, *J. Geophys. Res.*, 113, D03203, doi:10.1029/2007JD009042.
- Schwarz, J. P., et al. (2008b), Measurement of the mixing state, mass, and optical size of individual black carbon particles in urban and biomass burning emissions, *Geophys. Res. Lett.*, 35, L13810, doi:10.1029/2008GL033968.
- Wang, A. Z., et al. (2003), Characteristics of spilled oils, fuels, and petroleum products: 1. Composition and properties of selected oils, *Rep. EPA/600/R-03/072*, Natl. Exposure Res. Lab., Research Triangle Park, N. C.
- Wang, C., et al. (2008), Improving spatial representation of global ship emissions inventories, *Environ. Sci. Technol.*, 42(1), 193–199, doi:10.1021/es0700799.

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