

## Characterization of subsurface polycyclic aromatic hydrocarbons at the Deepwater Horizon site

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[1] Here, we report the initial observations of distributions of polycyclic aromatic hydrocarbons (PAH) in subsurface waters near the Deepwater Horizon oil well site (also referred to as the Macondo, Mississippi Canyon Block 252 or MC252 well). Profiles of *in situ* fluorescence and beam attenuation conducted during 9–16 May 2010 were characterized by distinct peaks at depths greater than 1000 m, with highest intensities close to the wellhead and decreasing intensities with increasing distance from the wellhead. Gas chromatography/mass spectrometry (GC/MS) analyses of water samples coinciding with the deep fluorescence and beam attenuation anomalies confirmed the presence of polycyclic aromatic hydrocarbons (PAH) at concentrations reaching  $189 \mu\text{g L}^{-1}$  (ppb). Subsurface exposure to PAH at levels considered to be toxic to marine organisms would have occurred in discrete depth layers between 1000 and 1400 m in the region southwest of the wellhead site and extending at least as far as 13 km. **Citation:** Diercks, A.-R., et al. (2010), Characterization of subsurface polycyclic aromatic hydrocarbons at the Deepwater Horizon site, *Geophys. Res. Lett.*, 37, L20602, doi:10.1029/2010GL045046.

### 1. Introduction

[2] The catastrophic explosion and sinking of the Deepwater Horizon drilling rig in the northern Gulf of Mexico [Schrope, 2010] resulted in a massive and unprecedented release of oil into the water column at a depth of approximately 1500 m. While observations of the presence of subsurface oil have been made in conjunction with both natural [Harvey et al., 1979] and human-induced [Boehm and Fiest, 1982; Elordui-Zapatarietxe et al., 2010; Johansen et al., 2003] releases, observations of subsurface distributions of oil and its chemical components in deep waters remain limited particularly for a release of the magnitude of the Deepwater Horizon event. A major environmental concern of large scale release of oil is toxicity to marine life, par-

ticularly that associated with the PAH fraction of the oil [Boehm and Page, 2007; National Research Council, 2003].

[3] Here, we summarize results from a cruise of opportunity conducted by the R/V *Pelican* in the vicinity of the Deepwater Horizon site approximately three weeks after the sinking of the oil rig. The R/V *Pelican* had been previously scheduled to carry out operations near the Deepwater Horizon wellhead site, in conjunction with a long term study of a region of nearby methane hydrate deposits [Boatman and Peterson, 2000]. This cruise provided the initial observations of deep subsurface optical signatures related to the released oil and associated elevated levels of PAH extending as far as 13 km from the wellhead site.

### 2. Methods

[4] The area of operations (Figure 1a) was in the vicinity of the Deepwater Horizon wellhead location. A series of 41 stations were occupied (see auxiliary material Table S1), but we focus here on two transects extending as far as 37 km from the wellhead (Figure 1b).<sup>1</sup> Vertical profiles of water column properties were acquired using a Seabird Electronics, Inc. SBE 911+ conductivity-temperature-depth (CTD) profiling package equipped with dual temperature and conductivity sensors, dual oxygen sensors, a WET Labs, Inc. C-Star transmissometer for the measurement of beam attenuation, and a WET Labs colored dissolved organic matter (CDOM) fluorometer. Heavy accumulations of surface oil were avoided with the profiling package so as not to contaminate the sampling gear or damage instruments.

[5] The profiling package was equipped with twelve 5 L General Oceanics Niskin bottles which could be triggered to sample at specified depths. Water samples for analysis of polycyclic aromatic hydrocarbons (PAH) were collected in 0.5 L amber glass bottles with Teflon caps. The sample bottles were thoroughly rinsed and filled with sample and amended with 8–10 mL of methylene chloride to inhibit sample degradation. Analyses were subsequently conducted according to methods previously described [Short et al., 1996].

### 3. Results and Discussion

#### 3.1. Subsurface CDOM Fluorescence and Beam Attenuation Features

[6] Vertical sections along both the northern and southern transects (Figure 2) were characterized by numerous layers of

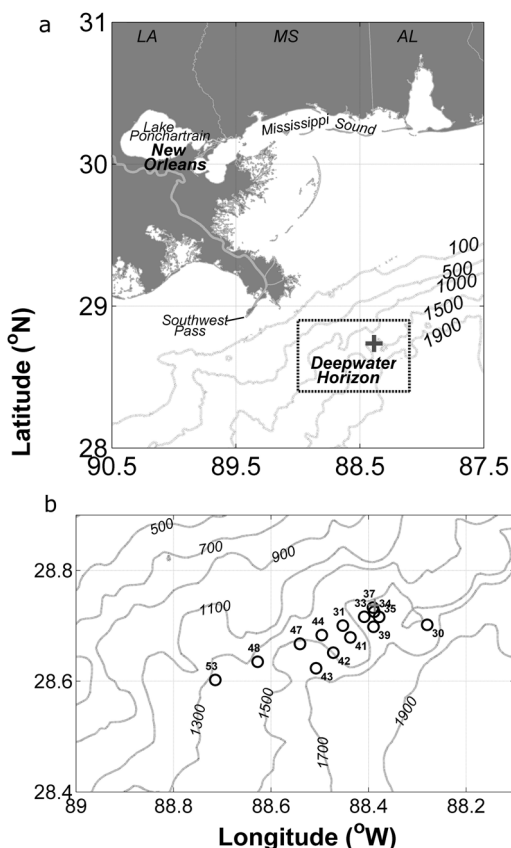
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**Figure 1.** (a) The Deepwater Horizon wellhead site indicated by the ‘+’ was located at a depth of approximately 1500 m at a location approximately 103 km southeast of Southwest Pass, Louisiana, U.S.A. Units of bathymetric contour lines are in m. The dashed box delineates the region enlarged in Figure 1b. (b) A series of stations were occupied in northern and southern transects extending from the wellhead site and vertical profiles of optical properties as well as discrete water samples were obtained from these stations. The “northern transect” included stations 37, 33, 31, 44, 47, 48 and 53; stations 34, 35, 39, 41, 42 and 43 comprised the “southern transect.”

elevated CDOM fluorescence (Figures 2a and 2b) and beam attenuation (Figures 2c and 2d) at water depths greater than 1000 m. Highest values were closest to the wellhead site, but signals above background were detected as far as 13 km from the site. The *in situ* CDOM fluorometer had an excitation/emission wavelength pairing of 370/460 nm, which was sensitive at  $\mu\text{g L}^{-1}$  concentrations to a wide range of fluorescent organic compounds and effective for detecting dispersed oil in seawater [Fuller *et al.*, 2005]. Beam attenuation is an optical property related to water clarity; higher values of beam attenuation are associated with higher levels of both absorption and scattering of light, in this case at a wavelength of 660 nm. Laboratory analyses of water samples found relatively low levels of absorption at 660 nm (data not shown). Consequently, we attributed the observed features in beam attenuation to particle scattering (e.g., dispersed droplets of oil). The pattern of beam attenuation along the two transects

was generally consistent with the CDOM fluorescence, although patterns differed particularly with increasing distances from the wellhead site (Figures 2a–2d). In general, the features observed tended to be distributed along isopycnal surfaces (black horizontal lines in Figures 2a–2d). This pattern was consistent with isopycnal (along density surface) transport of materials.

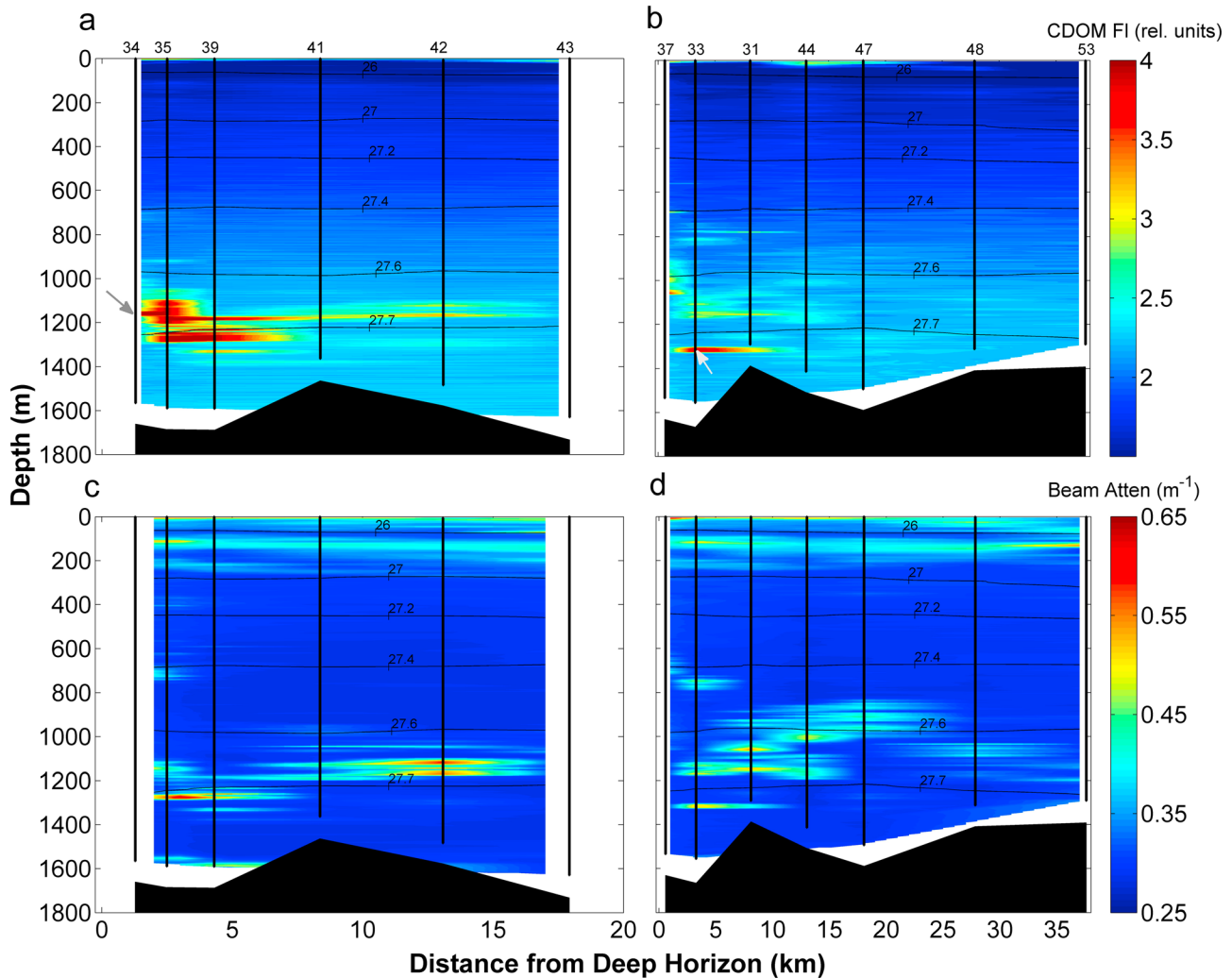
### 3.2. Horizontal Distribution in CDOM Fluorescence

[7] A pronounced fluorescence anomaly associated with the wellhead site was evident in contour maps of maximum values of fluorescence for depths below 800 m (Figure 3a). Relatively high values of fluorescence were evident as a plume emanating as far as 13 km from the wellhead site in a southwestwardly direction and reaching 9.6 km in width (Figure 3a). The fluorescence plume extended vertically over a depth range of approximately 500 m and was oriented along bathymetric features as seen in a three dimensional scatter plot of the fluorescence anomaly (Figure 3b). Elevated CDOM fluorescence levels were detectable over a horizontal area of approximately 100 km<sup>2</sup>.

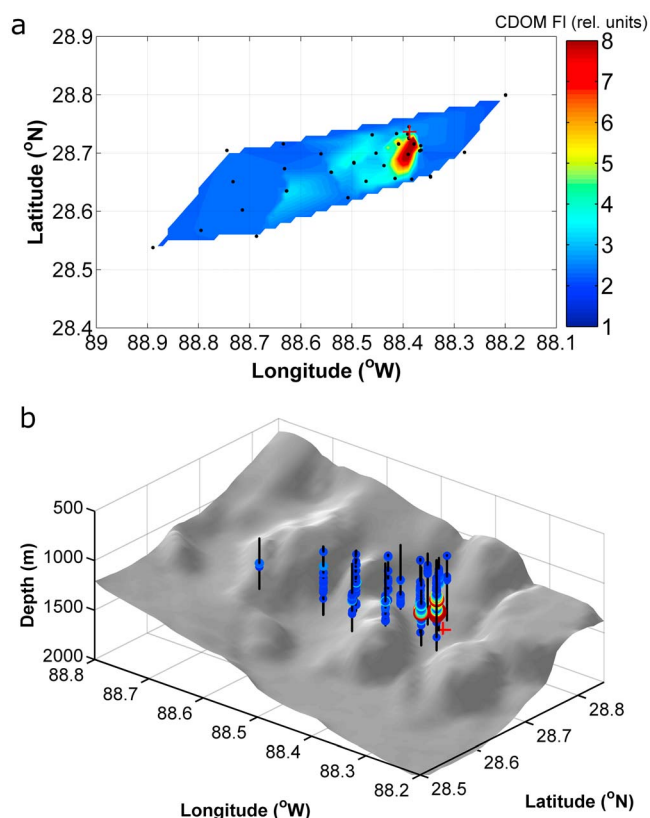
[8] The southwesterly and along isobath orientation of the plume evident in Figure 3 was consistent with observations of subsurface currents in this region. The Shelf Energetics and Exchange Dynamics experiment conducted a one-year time series of observations in this region using bottom-mounted acoustic Doppler current profilers [Carnes *et al.*, 2008; Teague *et al.*, 2006]. Their results revealed a complex current structure with variability dominated by both local and remote wind forcing as well as cyclonic eddies derived from the Loop Current. Mean currents at 900 m were generally along isobaths toward the southwest [Carnes *et al.*, 2008] consistent with the observed distribution of fluorescence (Figure 3). Average current speeds along the 900 m isobaths ranged from 1.9–2.8 cm s<sup>-1</sup>. At these speeds, dispersed and dissolved subsurface oil could have been transported as far as 34–50 km in three weeks, which is consistent with the observed horizontal extent of optical signatures in Figure 3.

### 3.3. PAH Analyses

[9] GC/MS analyses confirmed that the fluorescence anomalies were related to oil as evidenced by elevated PAH at depths where high fluorescence was observed. For samples coinciding with deep fluorescence features from two representative stations near the wellhead site, concentrations of total PAH were 189  $\mu\text{g L}^{-1}$  at 1320 m at Site 33 and 29  $\mu\text{g L}^{-1}$  at 1160 m at Site 34 (see arrows in Figure 2). Crude oil consists of a complex suite of compounds with different fluorescence fingerprints depending on its origins [Bugden *et al.*, 2008], and the form and composition of released oil changes as it mixes with water and is transformed by chemical and biological processes. In addition, natural sources of organic matter will contribute to a background level of CDOM fluorescence. Therefore, establishing a quantitative relationship between *in situ* fluorometry and oil-related compounds requires an empirical calibration based on chemical analyses of oil in discrete samples. For analyses of PAH determined in samples collected from depths greater than 800 m, the correlation of total PAH with CDOM fluorescence was highly significant ( $r^2 = 0.980$ ,  $N = 8$ ,  $p < 0.001$ ; see auxiliary material Figure S1). The derived regression relationship between CDOM fluorescence and



**Figure 2.** (a) The CDOM fluorescence section for the southern transect in Figure 1b exhibited a region of relatively high fluorescence at depths greater than 1000 m extending from the wellhead site as far as 13 km. The horizontal lines in all cases represent contours of sigma-t (related to potential density of seawater) in units of  $kg\ m^{-3}$ . The arrow indicates the station and depth for which GC/MS analyses of PAH are provided in Table 1. (b) CDOM fluorescence for the northern transect was similarly characterized by high values in close to the wellhead site and extending out as far as 13 km. Arrow as in Figure 2a. Patterns of beam attenuation for the southern (c) and northern (d) transects showed elevated features in deep water similar to that seen for CDOM fluorescence. Values of beam attenuation were more broadly distributed both horizontally and vertically than was evident for CDOM fluorescence.



**Figure 3.** (a) Maximum values of CDOM fluorescence for the lower 800 m portion of the water column are shown in a contour map in relationship to the wellhead site location, which was designated by the red '+'. Maximum values of CDOM fluorescence for depths greater than 800 m were characterized by high values extending southwest from the wellhead site and covered a region of approximately 200 km<sup>2</sup>. (b) A three dimensional representation of the deep fluorescence plume shows relatively high CDOM fluorescence over a depth range of approximately 400 m and distributed along bathymetric features. Values of CDOM fluorescence < 2.5 were excluded from the plot. Bathymetric data were acquired from the General Bathymetric Chart of the Ocean's website (The GEBCO\_grid, version 20091120, <http://www.gebco.net>).

total PAH was  $\text{PAH } (\mu\text{g L}^{-1}) = 42.0 (\text{CDOM fluorescence}) - 99.2$ . Based on this relationship, fluorescence units of 6 and higher that were observed in the proximity of the wellhead site (Figure 3) corresponded to total PAH levels in excess of  $150 \mu\text{g L}^{-1}$ . Camilli *et al.* [2010] have made subsequent independent chemical observations confirming the existence of a deep hydrocarbon plume, which corroborates our results.

[10] The composition of PAH in the deep plumes consisted predominately of methyl-naphthalenes (Table 1). In contrast, a surface sample was found to have a larger proportion of three ring fluorenes, phenanthrene, anthracenes, and dibenzothiophenes (Table 1). This pattern was consistent with a water partitioning of more soluble PAH compounds during transport to the surface [Gonzalez *et al.*, 2006].

[11] Prior studies have demonstrated both lethal and sublethal effects of 1,2 dimethyl-naphthalene at concentrations of  $161 \mu\text{g L}^{-1}$  for a coastal copepod species, *Paracartia (Acartia) grani* [Calbet *et al.*, 2007]. Moreover, toxic effects of mixtures of different PAH compounds on copepod species are additive [Barata *et al.*, 2005]. Other work has documented physiological effects on embryonic and larval marine fish [Carls *et al.*, 2008; Couillard *et al.*, 2005], even at concentrations of total PAH of less than  $5 \mu\text{g L}^{-1}$  [Carls *et al.*, 2005]. The addition of dispersants that occurred in the case of the Deepwater Horizon oil release event could have substantially increased the PAH levels in the water column, particularly in the case of more hydrophobic compounds, and thus increase the exposure effects [Couillard *et al.*, 2005; Yamada *et al.*, 2003].

[12] Our findings suggest that toxicity effects of the subsurface PAH compounds could have extended at least as far as 13 km from the wellhead site at the time these data were acquired, with highest concentrations confined to depths between 1000 and 1400 m. A key consideration in assessing the ecological impact of this event will be the persistence of these chemicals in the natural environment [Boehm and Page, 2007]. Results from mesocosm experiments provide evidence that loss rates of PAH compounds containing less than three aromatic rings is rapid, occurring largely within two days [Yamada *et al.*, 2003]. Reduced dissolved oxygen levels that coincided with the locally high fluorescence and beam attenuation features observed in vertical profiles (see auxiliary material Figure S2) were consistent with chemical oxidation or biodegradation by marine microorganisms [Gertler *et al.*, 2009; Venosa and Holder, 2007]. Recent investigations have confirmed that reduced oxygen saturation levels within the deep plume corresponded to enhanced microbial activity consistent with degradation of petroleum hydrocarbons [Hazen *et al.*, 2010].

#### 4. Conclusions

[13] The findings reported here represent the earliest known observations of the distributions of deep subsurface petroleum hydrocarbons related to the Deepwater Horizon oil release, and are consequently important for establishing an overall temporal sequence of hydrocarbon exposure. Subsequent studies have confirmed the presence of this feature [Camilli *et al.*, 2010; Hazen *et al.*, 2010] (see <http://ecowatch.ncddc.noaa.gov/JAG/reports.html>). Based on our findings, subsurface exposure to PAH resulting from the Deepwater Horizon oil release was likely to be associated with acute toxicity effects in discrete depth layers between 1000 and 1400 m in the region southwest of the wellhead site and extending at least as far as 13 km. A comparison of the chemical nature of the PAH compounds between surface and deep waters suggested a partitioning of oil components during transport from sea floor to surface, with predominantly two ring PAH in the deep plumes. These compounds may experience relatively rapid loss rates, but this has yet to be demonstrated. From the time that these observations were made, there was an extensive release of additional oil and dispersants at the site. Therefore, the effects on the deep sea ecosystem may be considerably more severe than supported by the observations reported here.

**Table 1.** Analyses of Polycyclic Aromatic Hydrocarbons

PAH Compounds	Site 30 Surface		Site 33 1320 m		Site 34 1160 m	
	Micrograms Per Liter	Percent	Micrograms Per Liter	Percent	Micrograms Per Liter	Percent
Naphthalene	0.13	0.15	1.22	0.65	0.31	1.07
C1-Naphthalenes	0.71	0.84	32.1	17.0	2.28	7.75
C2-Naphthalenes	1.73	2.04	70.4	37.3	8.45	28.7
C3-Naphthalenes	3.22	3.80	56.6	29.9	11.2	38.2
C4-Naphthalenes	2.28	2.69	24.3	12.9	4.41	15.0
Biphenyl	0.11	0.13	0.16	0.08	0.06	0.20
Acenaphthylene	0.03	0.03	nd	nd	0.01	0.03
Acenaphthene	0.23	0.27	0.01	0.00	0.01	0.03
Fluorene	0.30	0.36	0.26	0.14	0.33	1.13
C1-Fluorenes	1.90	2.25	0.43	0.23	0.54	1.83
C2-Fluorenes	3.97	4.68	0.41	0.22	0.51	1.75
C3-Fluorenes	3.53	4.16	0.24	0.13	0.21	0.71
Phenanthrene	2.70	3.18	0.32	0.17	0.20	0.67
Anthracene	0.16	0.19	0.02	0.01	0.01	0.02
C1-Phenanthrenes/Anthracenes	14.2	16.8	0.62	0.33	0.31	1.06
C2-Phenanthrenes/Anthracenes	16.3	19.3	0.57	0.30	0.17	0.59
C3-Phenanthrenes/Anthracenes	8.78	10.4	0.33	0.18	0.07	0.23
C4-Phenanthrenes/Anthracenes	2.83	3.34	0.11	0.06	0.03	0.09
Dibenzothiophene	0.36	0.43	0.07	0.04	0.03	0.12
C1-Dibenzothiophenes	2.66	3.14	0.15	0.08	0.06	0.21
C2-Dibenzothiophenes	5.35	6.31	0.18	0.10	0.05	0.18
C3-Dibenzothiophenes	3.68	4.34	0.10	0.05	0.03	0.09
Fluoranthene	0.11	0.13	0.00	0.00	0.00	0.01
Pyrene	0.34	0.41	0.02	0.01	0.00	0.02
C1-Fluoranthenes/Pyrenes	1.06	1.26	0.06	0.03	0.01	0.05
C2-Fluoranthenes/Pyrenes	1.28	1.51	0.09	0.05	0.02	0.05
C3-Fluoranthenes/Pyrenes	1.03	1.22	0.06	0.03	0.01	0.03
Benzo(a)anthracene	2.28	2.69	0.05	0.03	0.01	0.05
Chrysene	nd	nd	nd	nd	nd	nd
C1-Chrysenes	2.95	3.48	0.08	0.04	0.02	0.07
C2-Chrysenes	nd	nd	0.06	0.03	0.01	0.05
C3-Chrysenes	nd	nd	nd	nd	nd	nd
C4-Chrysenes	nd	nd	nd	nd	nd	nd
Benzo(b)fluoranthene	nd	nd	nd	nd	nd	nd
Benzo(k)fluoranthene	nd	nd	nd	nd	nd	nd
Benzo(e)pyrene	0.37	0.43	nd	nd	nd	nd
Benzo(a)pyrene	nd	nd	nd	nd	nd	nd
Perylene	0.09	0.11	nd	nd	nd	nd
Indeno(1,2,3-c,d)pyrene	nd	nd	nd	nd	nd	nd
Dibenzo(a,h)anthracene	nd	nd	nd	nd	nd	nd
Benzo(g,h,i)perylene	nd	nd	nd	nd	nd	nd
Total PAH	84.8	100.0	189.0	100.0	29.4	100.0

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