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Oiling of the continental shelf and coastal marshes over eight years after the 2010 *Deepwater Horizon* oil spill★

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ABSTRACT

We measured the temporal and spatial trajectory of oiling from the April, 2010, *Deepwater Horizon* oil spill in water from Louisiana's continental shelf, the estuarine waters of Barataria Bay, and in coastal marsh sediments. The concentrations of 28 target alkanes and 43 target polycyclic aromatic hydrocarbons were determined in water samples collected on 10 offshore cruises, in 19 water samples collected monthly one km offshore at 13 inshore stations in 2010 and 2013, and in 16–60 surficial marsh sediment samples collected on each of 26 trips. The concentration of total aromatics in offshore waters peaked in late summer, 2010, at 100 times above the May, 2010 values, which were already slightly contaminated. There were no differences in surface or bottom water samples. The concentration of total aromatics declined at a rate of 73% y^{-1} to 1/1000th of the May 2010 values by summer 2016. The concentrations inside the estuary were proportional to those one km offshore, but were 10–30% lower. The oil concentrations in sediments were initially different at 1 and 10 m distance into the marsh, but became equal after 2 years. Thus, the distinction between oiled and unoiled sites became blurred, if not non-existent then, and oiling had spread over an area wider than was visible initially. The concentrations of oil in sediments were 100–1000 times above the May 2010 values, and dropped to 10 times higher after 8 years, thereafter, demonstrating a long-term contamination by oil or oil residues that will remain for decades. The chemical signature of the oil residues offshore compared to in the marsh reflects the more aerobic offshore conditions and water-soluble tendencies of the dissolved components, whereas the anaerobic marsh sediments will retain the heavier molecular components for a long time, and have a consequential effect on the ecosystems.

1. Introduction

The *Deepwater Horizon* (DWH) drilling platform collapsed in the northern Gulf of Mexico (GOM) April 20, 2010, killing 11 workers and injuring 17 other crew members. It subsequently released 4.9×10^6 barrels (bbl) of oil from 4000 m below the ocean floor in 1500 m water depth until completely closed off on September 19, 2010 (McNutt et al., 2012a, b). This marine oil spill was the largest in the world history, equal to 11 times the previous largest marine oil spill in the US until then from the grounding of the oil tanker *Exxon Valdez* in Alaska 21 years earlier (0.48×10^6 bbl; Paine et al., 1996), 25 times the oil discharges during Hurricane Katrina (Pine, 2006), but two-thirds of the land-based 1908 Dos Bocas spill in Veracruz, Mexico (Santiago, 2006). It followed the first large-scale subsurface spill in the Gulf of Mexico, the IXTOC I spill (3 million barrels), into the Bay of Campeche, Mexico, from June 1979 to March 1980. The DWH accident created a singularly massive oil spill delivered within the background of oil contamination from the Mississippi River, atmosphere, releases from ships, historic and recent oil

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production platforms and infrastructure, and natural seeps in the GOM (National Research Council, 2002). The daily release of DWH oil over 87 days was about 34–128 times higher than the daily hydrocarbon release from natural seeps over the entire GOM, which disperses and evaporates quickly ($0.16–0.6 \times 10^6$ bbl yr^{-1}; MacDonald et al., 2015).

The oil that entered the Gulf from the reservoir changed chemical composition and physical properties as a result of dissolution, photo-oxidation, incorporation of other elements, etc., to become what we call 'oil residues' herein. Burning, oil skimmers and 6965 m^3 of the dispersants COREXIT 9500A and COREXIT 9527A were applied in an attempt to disperse oil, hasten decomposition, and disrupt slicks (Kujawinski et al., 2011). Twenty-nine percent of the 4.9 mb DWH oil dispersed within the water column, another, 23% evaporated or dissolved, and 23% was unaccounted for (McNutt et al., 2012a). The oil and its residues were not rapidly flushed out of the GOM because the residence time of the deep water is 250 years (Rivas et al., 2015; Chapman, 2017). Oil residues spread away from the surface near the well head and were found on 1773 km of the 7058 km beaches and coastal marshes that were surveyed from Florida to Texas (Fig. 1; Michel et al., 2013). The oiled shoreline was predominantly in Louisiana (60.6%), and 38% of that remained visually evident after two years (Michel et al., 2013). The hydrocarbons interfacing with beaches and marshes were immediately toxic or subsequently generated chronic effects on the flora, fauna and human population as documented in an extensive Federal damage assessment (NRDA, 2016). The media produced disturbing visuals of oiled pelicans, dolphins, turtles, beaches and marshes (Fig. 1) (Morse, 2012). Much of commercial fishing was closed causing a >$1 billion deficit to the industry (NMFS, 2010; Carroll et al., 2016).

A massive cleanup effort by 47,489 people and over 6000 vessels ensued (Michaels and Howard, 2012; United States Coast Guard, 2011) to contain, remove and mitigate the damage estimated at $17 billion for natural resources (Bishop et al., 2017) and $37 billion for the combined environmental and economic losses (Smith et al., 2011). Tens of billion dollars ($US) in fines and restitution were made to Federal, State and regional governing bodies, non-governmental organizations, and private citizens. Multiple research efforts were funded (Cornwall, 2015; Murphy et al., 2016). Part of the general framework for the research was to document damages to the ecosystem, potential long-term threats and consequences, mitigation and restoration opportunities, and to prevent future spills. These damage valuations were partially dependent on knowing: 1) how much of the original oil and oil residue reached the continental shelf, then entered the estuaries, and were deposited in marshes, and, 2) if the amounts persisted or changed.

We document the petroleum contamination after the DWH oil spill in water on the Louisiana continental shelf, in estuarine waters, and in marsh sediments from May 2010 to June 2018. We compare the changes in total amounts of alkanes and aromatics over time, the coherence among concentrations in nearshore shelf and inshore estuarine waters, the initial accumulation in marsh sediments, and estimate the legacy of oil remaining within the marshes as of June 2018.

2. Methods

2.1. Study area description

The DWH spill occurred at 1500 m depth in the northern Gulf of Mexico (GOM), a marginal sea of the Atlantic Ocean bordered by five U.S. states on the northern and eastern border, five Mexican states on its western and southern border, and Cuba to the

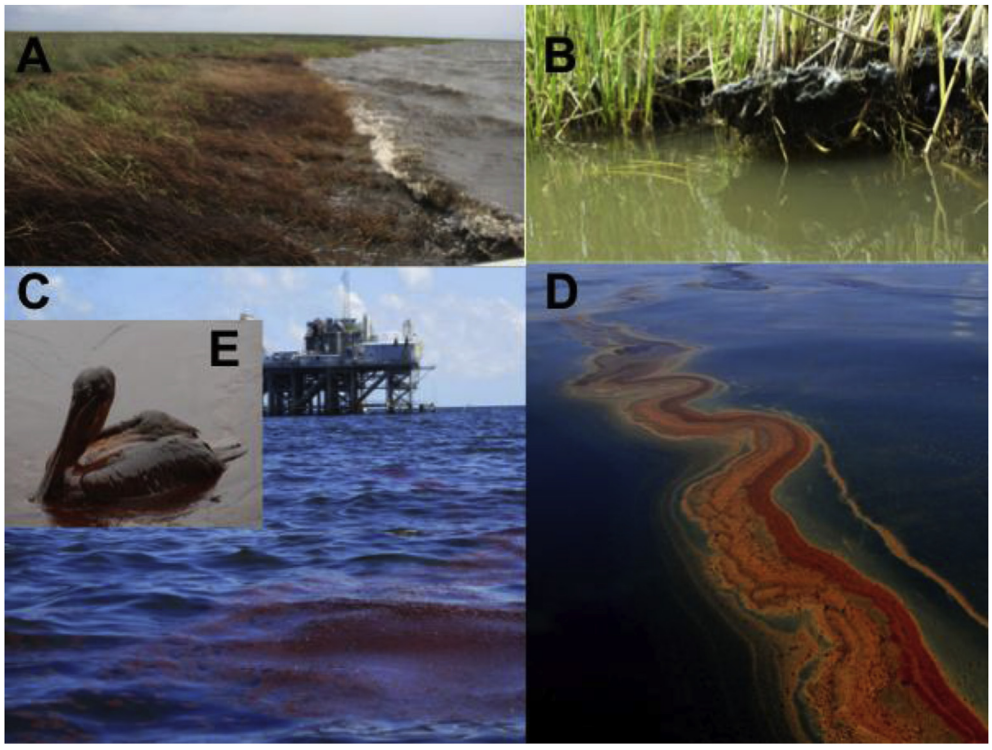

Fig. 1. Oil on the continental shelf and marsh. (A) Oil coating marsh vegetation at the shoreline in Bay Batiste, LA. (B) Oil residues, mostly black asphaltenes, at the base of salt marsh plants; note the overhang with exposed roots beneath. (C) The surface oil and mousse on surface waters in May 2010. D. A line of mousse and oil sheens off the Mississippi River delta. E. An oiled pelican (*Pelecanus occidental* - the Louisiana State Bird) covered in oil. A, B and C are photos by the authors. D is from NOAA, and E is from an open source (https://www.flickr.com/photos/49937499@N08/4667375844)

southeast. The average depth of the GOM is 1615 m and the maximum is 4400 m. There are broad continental shelves off Florida, the Yucatán Peninsula and the northwestern GOM. The volume flux of warm and salty Caribbean water entering through the Yucatán and Florida straits (Mooers and Maul, 1998) is equivalent to a 2.5 year refill rate for the GOM, compared to a 1/50,000th lower refill rate (weeks to months) for the shelf volume where coastal currents mix a smaller water volume (Barron and Vastano, 1994). There is, therefore, a relatively slower water turnover rate (years) in the center of the GOM, compared to turnover rates of weeks-to-months on the periphery that contains a smaller water volume of shallower depths.

The surface temperature of the warm and calm winds in summer GOM fuels hurricane winds, which have come ashore everywhere in the GOM. The transfer of water between continental shelf and deeper GOM waters is partially documented, or implied by strong inference (Fry et al., 2015; Turner and Rabalais, 2019). Ninety % of the water and 62% and 88% of the terrestrial riverine-derived N and P sources into the GOM, respectively, are from the Mississippi and Atchafalaya rivers on the northern boundary. The resulting stratification and enhanced nutrients drive the formation and maintenance of a low oxygen zone off the Louisiana and northern Texas coast, which is the second largest human-caused coastal hypoxic area in the global ocean (Rabalais et al., 2007). It extends from near shore to as much as 125 km offshore, and in water depths up to 60 m (Rabalais et al., 2007. Hydrocarbons from the DWH oil spill moved directly onto this shelf, and some continued into the estuaries of the northern GOM into the 10,000 km^2 + of wetlands (Turner and Rabalais, 2019).

The salinity in the northern GOM estuaries is influenced by water exchange between the estuarine entrance and the coastal zone, and local forcing (tidal advection, river discharge, precipitation) occurring within the estuary proper. The monthly tidal range in these estuaries averages 30 cm with a low in winter and high in late summer. Barataria Bay is northwest of the DWH spill site and is adjacent to the west bank of the Mississippi River in southeastern Louisiana, USA, and empties into the Gulf of Mexico and Terrebonne Bay is to the west (Fig. 2B). Some of the Mississippi River discharge enters the Barataria estuary from offshore to alter the salinity of the lower Barataria Bay and also brings nutrients into the estuary (Wiseman et al., 1990; Wissel et al., 2005; Turner et al., 2019). The majority of all oiled shorelines and most marsh oiling were in Louisiana (>60% and 95%, respectively; Michel et al., 2013).

The sample collections described below were used to determine the quantity and quality of oil in continental shelf water and one estuary near the DWH spill site, and in marsh sediments from the spring of 2010–2018.

2.2. Sample collection

Water samples without visible signs of oil slicks, sheens, tarballs or mousse were collected on the continental shelf from 17 to 19 May 2010 through July 2018 (Fig. 2). The 375 offshore water samples were sub-sampled from Niskin water bottles tripped within 0.5–1.0 m above the bottom or within 0.5 m of the surface from ten cruises across the hypoxia mapping area (Rabalais et al., 2018) (Fig. 2A). Six of these cruises were the summer shelf-wide hypoxia cruises with transects from shallow water to 50-m water depth that occurred in July 2010, 2013, 2014, 2015, 2017 and 2018 (Rabalais et al., 2018). Four cruises were conducted in May, June, August and October 2010 and covered the C and F transects with 155 samples (Fig. 2A). The average sample number for shelf-wide cruises was 40 per cruise (range n = 12 to 71). The sample distribution for these offshore samples is in Supplemental Table S2. Water samples were placed immediately into pre-cleaned amber glass bottles, refrigerated at 4 °C, and extracted within 14 days, as recommended by the US EPA (2007).

Water samples for oil analyses were also collected from a small boat in Barataria Bay (Fig. 2B) and 1 km offshore of the bay entrance on monthly transects during two periods: 1) from May to December 2010, and 2) for each month in 2013 as part of routine monthly sampling (Turner et al., 2019). Sample water (0.5 L and 1 L) was collected at 13 inshore stations (station numbers 1–14 in Turner et al., 2019) using a bucket rinsed with surface water, refilled and immediately put into a commercially-prepared amber glass bottle.

Marsh sediment samples were collected from fixed locations with vegetated cover dominated by *Spartina alterniflora*, *Juncus* spp. or *Schneoplectus americanus*. There were three sampling schemes: 1) five trips were made to sample 18–30 different sites in St. Bernard, Barataria and Terrebonne estuaries (n = 183). The May 2010 sampling occurred before oil was observed on beaches or behind barrier islands; 2) there were 14 biannual trips to east and west Barataria Bay and Terrebonne Bay from 2011 to 2018 (n = 326); and 3) there were seven trips to the north shore of Bay Batiste (these are the same sites used by McClenachan et al., 2013) from 2011 to 2018 (n = 210). The oiling of these marsh sites from 2010 to 2012 was reported in Turner et al. (2014a, b). Only samples collected at 1 and 10 m from the shoreline are reported here. All sediment samples were collected from the upper 5 cm of the sediment surface, stored in pre-cleaned amber jars on ice until delivery to the laboratory, and either immediately extracted or refrigerated at 4 °C for no more than 14 days until extraction, as recommended by the US EPA (2007).

2.3. Oil analyses

We targeted 28 alkanes and 43 polycyclic aromatic hydrocarbons (PAHs) and their respective alkyl homologs (18 parent PAHs, and 25 alkyl homolog groups) (herein, “alkanes” and “aromatics”) for quantitative analysis using GC/MS-SIM (gas chromatography/mass spectrometry in selective ion monitoring mode), as well as isoprenoid hydrocarbons pristane and phytane, and cyclic forensic petroleum biomarker compounds like the hopanes, steranes and triaromatic steroids (SIM ions 191, 217, 218 and 231 eluting between C23 and C31). The more complete methods description is found in the Supplemental Materials. Briefly, we used accepted standard operating and QA/QC procedures to prevent contamination and avoid sample degradation. The sediments were speed extracted with dichloromethane (DCM) and spiked with surrogate recovery standards to achieve a final standard concentration of 20 μg mL^{-1} in extracts. Water samples were liquid-liquid extracted with DCM. All extracts were concentrated by rotary evaporation and concentrated under a stream of nitrogen to a final volume of 1-μl and then 0.5 ml portions were used for injection. The target alkane and PAH concentrations were determined by an internal standard method and response factors calculated from a 5-point calibration curve using a commercially-available standard containing the normal alkanes from n-C10 through n-C35 and the parent PAH analytes of interest. The MS detector was tuned to PFTBA (perfluorotributylamine) before each set of analyses. The instrument was routinely checked for errors and a daily calibration standard and blank were analyzed with each sample batch to verify proper instrument performance. The identities of all analytes were confirmed using retention time comparisons from the ion chromatogram using full scanning mass spectral analysis of the riser source oil sample. The spectral data were processed by Chemstation Software (Agilent Technologies). The data are archived at https://data.gulfresearchinitiative.org/data/R1.x139.142:0004/.

Contamination from the oil spill was detected using two

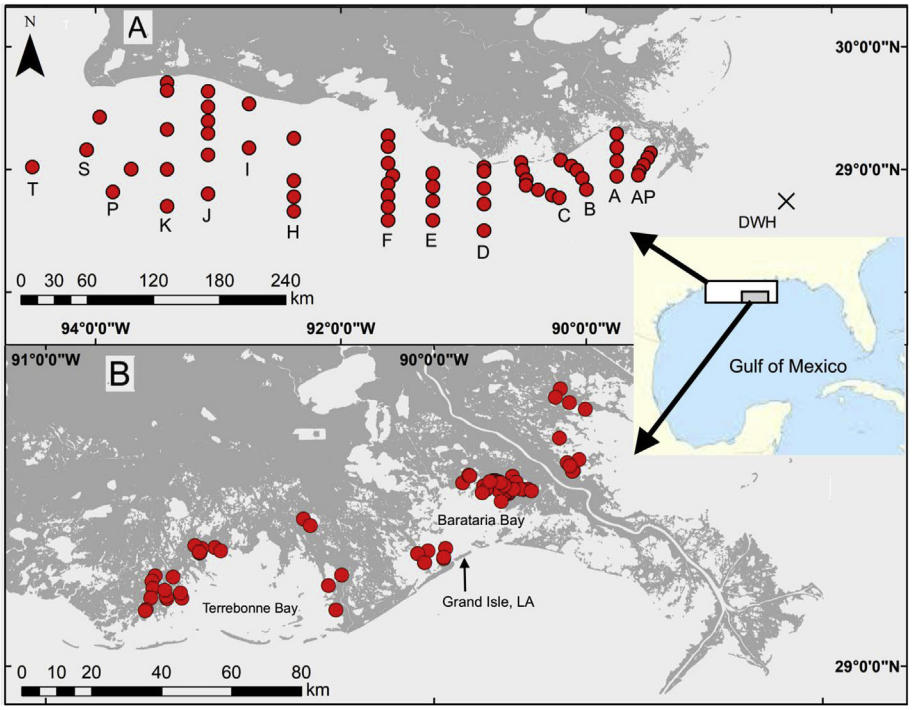

Fig. 2. Sample locations from offshore waters (A) and from marsh sediments (B). The DWH oil spill site is indicated with an X in 2A. The offshore station transects are identified with letters whose sampling frequency is in Supplemental Table S2.

methods. First, the concentrations of target aromatics and alkanes from petroleum has a specific composition, even after weathering, that is distinct from background target aromatic and alkane distributions within marsh samples. Second, samples containing oil residues within the marsh have detectable levels of petroleum biomarker compounds, such as the hopanes and steranes. In addition to the target alkanes and aromatic compounds measured in all chemical analyses, C30 hopane was quantified from the m/z 191 mass chromatogram as well as the chromatographic profile for the other hopanoid biomarkers. Additionally, the sterane biomarker profiles at m/z 217 and 218 mass chromatograms were also recorded. The chromatographic profiles of these biomarker compounds were visually paired with the same profiles in MC252 source oil for all samples to establish a link between the oil residues in samples with the spilled oil. Thus, marsh sediments were examined using these two analytical metrics to determine contamination from the DWH oil spill.

2.4. Statistics

The percent of the average analyte composition for each sampling trip was computed for comparison purposes. The mean and standard error of the mean ($\mu \pm 1$ SE) for the total alkane and total aromatics were computed for each sampling trip collecting water or marsh sediments, and a linear regression calculated for the log10 transformation of concentration vs year. A linear regression was derived between the total aromatics in the Barataria Bay inshore stations and one offshore station for 2010 and 2013. A *t*-test was run using Prism software to test if there was a difference between the concentrations of aromatics in surface and bottom water samples collected on each offshore cruise. A Welch's correction for unpaired samples was applied. The statistical significance was $p < 0.05$ for all linear regression slopes and comparisons.

3. Results and discussion

3.1. Offshore

The concentration of the total aromatics was not different in surface and bottom water samples within each sampling trip ($p > 0.05$), but was different among dates. The average concentration of target aromatics in all stations was 3–9 $\mu g\,L^{-1}$ on the first three trips in 2010 (May, June, July), and then rose to 153 $\mu g\,L^{-1}$ in August and 323 $\mu g\,L^{-1}$ in September, after which it declined until it was below 0.01 $\mu g\,L^{-1}$ by 2015, 2017 and 2018 (Fig. 3A). These values are a plausible consequences of the spill on this shelf. For example, if all of the DWH oil (density 0.839) were equally distributed throughout the entire water mass of the GOM after one-half was volatilized, then the concentration of all target aromatics would equal 1453 $\mu g\,L^{-1}$, equal to nine times the concentration of dissolved target aromatics measured on the Louisiana shelf in July 2010.

The decline in total target aromatics in the July cruises from 2010 to 2018 was 73% y^{-1} ($Y = -1.329{*}X + 2676$; $p = 0.014$; $F = 17.1$; $R^2 = 0.81$), which is a first order decay rate, meaning that the concentration was declining without regard to the decay rate itself or dilution (Fig. 3B). There was no significant change in the total target alkane concentrations over the same interval ($p > 0.45$). The average ratio of target aromatics to total alkanes (by measured weight; $\mu g\ L^{-1}/ng\ L^{-1}$) was about 7 in May 2010, quickly climbed 100-fold by September, and fell below 0.1 from 2015 to 2018 (Fig. 3C). Several water samples collected in August and September of 2010 contained trace levels of petroleum biomarker compounds, indicating that these water samples contained entrained small residues of oil, not just the more water soluble components from the oil.

The average concentration of total target aromatics in May 2010 was three orders of magnitude higher than in July 2018 (Fig. 3A), and it is necessary to ask if the July 2018 concentration could be a

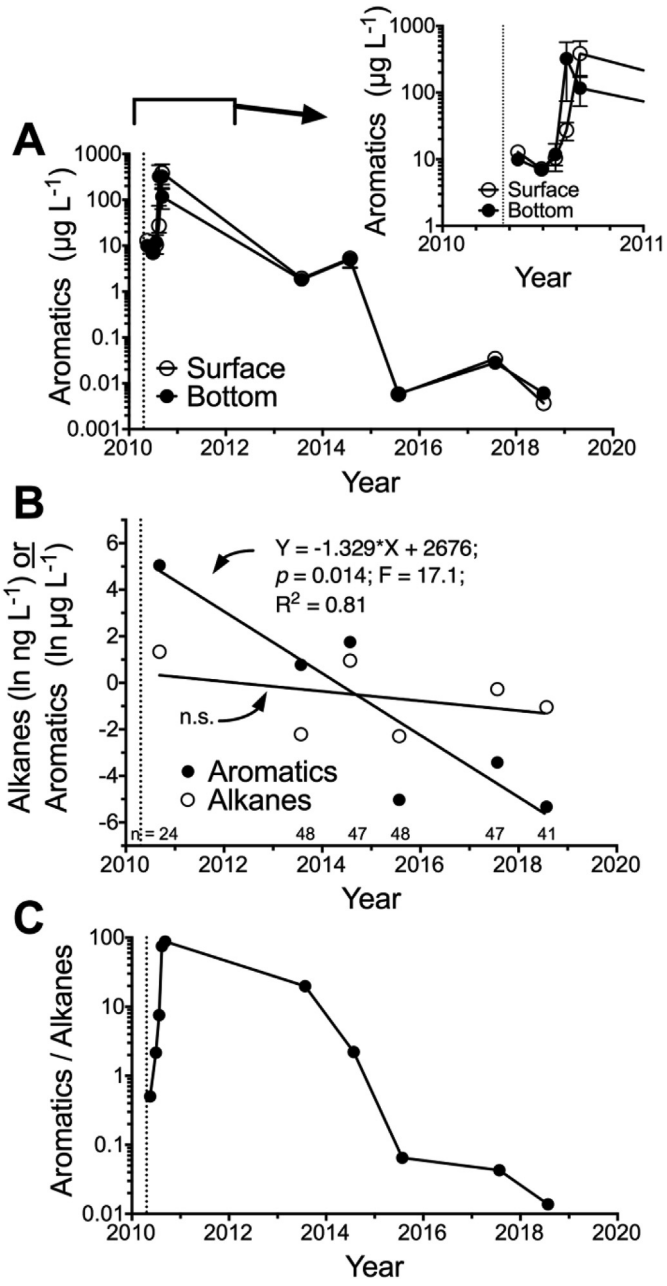

Fig. 3. The concentration of total aromatics and total alkanes in offshore water samples taken on the continental shelf from May 2010 through July 2018. A. Average amounts of aromatics in surface and bottom water samples (µ ± 1 SE). B. The natural log of the average concentration of total aromatics and total alkanes for the July samples taken on the annual hypoxia mapping cruises from the mouth of the Mississippi River to the Texas coast. C. The ratio of total aromatics/total alkanes for each sampling trip (by measured weight; µg L^{-1}/ng L^{-1}). The vertical dotted line is the beginning of the oil spill offshore.

baseline value to use for conditions before the oil spill residues reached the inner Louisiana continental shelf. There are four supportive logic points to support the view that the May samples were already contaminated with oil and do not represent baseline concentrations: 1) tar balls and sheens were found on the Grand Isle beach on May 21, 2010 (Rioux, 2010). A dive team (N.N. Rabalais, personal observation) at the 20-m isobath near the entrance to Barataria Bay in mid-May 2010 encountered oil residues in and just below the water surface (Fig. 1D). The shipboard water samples collected in May 2010 were sourced or indicative of dissolved oil contamination from the DWH oil spill; 2) there were three years of offshore samples in 2015, 2017 and 2018 that were below the detection limit for target aromatics and alkanes, and these were considered to be unlikely 'oiled' conditions; 3) the ratio of target aromatics/alkanes was lower in the same last three sampling years compared to in May 2010; and 4) a minimum sustained current speed of 0.4 cm s^{-1} for 30 days would have been sufficient to transport the oil from the DWH spill site to this continental shelf location, assuming it took the most direct path. This speed was observed in Alabama by Mulabagal et al. (2013) who determined the chromatographic signatures of petroleum biomarkers to identify source material of tar balls found on Alabama beaches after the DWH oil spill. While there were no tar balls observed about 2 weeks after the spill and 3 weeks before beaches were oiled, mousse was found in Alabama beaches on June 11, 2010, 8 weeks after the spill began. Beazley et al. (2012) found oil residues in Alabama surface sediments in marshes on June 8, 2010 (50 µg g^{-1}). A current speed of 0.4 cm s^{-1} over 30 days would be sufficient to take oil to the Alabama coast from the well head, assuming it went the most direct route.

Algae can produce alkanes (Schirmer et al., 2010; Gelin et al., 1999) which add to the pool of target alkanes. Bottom waters in the inner and mid Gulf of Mexico continental shelf off southeastern Louisiana are hypoxic because of the excess production of phytoplankton in surface waters that eventually sinks to the seabed and through decomposition by bacteria and the waters become devoid of dissolved oxygen (Rabalais et al., 2007). The algal populations in surface waters vary among years and seasons (Walker and Rabalais, 2006; Parsons et al., 2015). It is not surprising, therefore, that the total target alkane concentration in surface waters among years demonstrates no trends in summer. However, the distribution of target alkanes in May 2010 water samples is distinct from the alkane distribution seen from typical biogenic sources, having no evidence of odd carbon preference in the (Cn, n = 20–30) range (see Fig. S1).

3.2. DWH oil exchange through tidal passes

The concentration of target aromatic hydrocarbons in water samples taken within Barataria Bay changed in concert with the concentrations in nearshore waters (Fig. 4). The concentration of total target aromatics within Barataria Bay, for example, rose sharply in September 2010, compared to the previous three months (Fig. 4A) – a rise similar to that in offshore waters (Fig. 3A). The concentration of total aromatics within the estuary each month was proportional to the amount found 1 km off of the estuary entrance, but 15–35% lower (Fig. 4B). Concurrent fluctuations between the salinity offshore and in Barataria Bay are well-documented (Wiseman et al., 1990; Turner et al., 2019). We conclude that what is in the water column offshore can be assumed to enter the estuary – including dissolved petroleum hydrocarbons and oil residues.

3.3. Oil residues in the marsh

Our first sampling in the southeastern Louisiana marshes was 10 m from the shoreline in May 2010, one month after the beginning of the DWH oil spill. At this time, only occasional samples with detectable concentrations of target aromatics in May 2010 (before oiling of the marshes) were found (Fig. 5). The peak concentration was in February 2011, when there was a thousand-fold higher concentration of total target aromatic concentrations in marsh surficial sediments, compared to May 2010 (Fig. 5). Thereafter, the petroleum hydrocarbon concentrations of both target aromatics

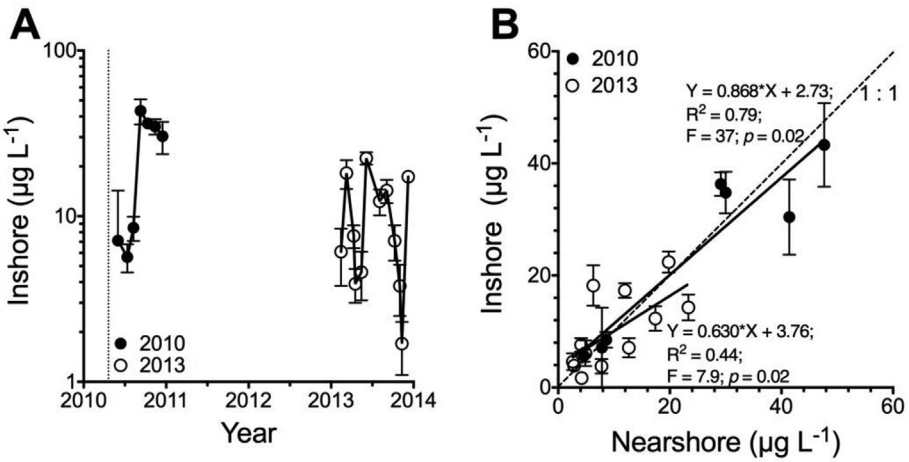

Fig. 4. The concentration of aromatics in 2010 (dark circles) and 2013 (open circles) at the offshore station 1 km outside of Barataria Bay and five stations (µ ± 1 SE) within lower Barataria Bay measured on a single day. A. Concentration versus date. The dotted line is the start of the *Deepwater Horizon* oil spill. B. The relationship between the concentrations within 1 km offshore and inshore for each month. The linear regression lines for each year are shown separately and the dotted line is the 1:1 correspondence line.

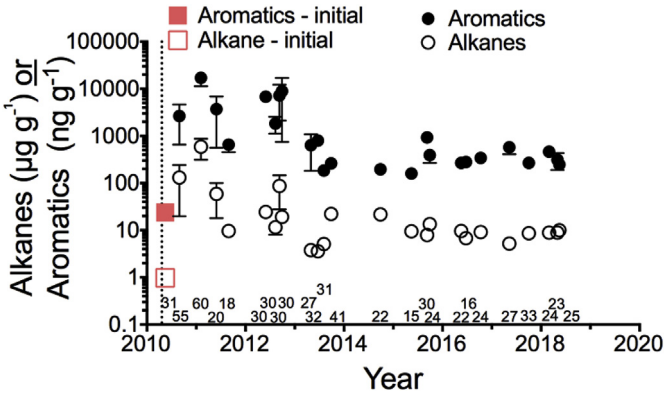

Fig. 5. The concentration of total aromatics (filled symbols, ng g^{-1} ±1 SE) and total alkanes (open symbols, µg g^{-1} ±1 SE) in marsh sediments for each sampling trip from May 2010 to May 2018. Proportional amounts of aromatics are in Supplemental Table S3. The red symbols are the samples taken before oil reached the marsh. The vertical dotted line is the beginning of the DWH oil spill offshore. The sample number averaged for individual trips are below each symbol (along the x-axis) and are the same for alkanes and aromatics.

and alkanes dropped 100 fold (i.e., to about 10 times higher than the May 2010 sediment sample concentrations) and remained higher through spring 2018. The target alkane concentrations increased approximately 100 times the May 2010 values in the marsh sediments compared to a 1000–fold increase observed for the offshore water samples. The standard error of the mean (SE) was greater in the first few years after the oil spill than in later years (2013–2018), indicating the spotty nature of hydrocarbon contamination in the immediate aftermath of oiling, but was so small in later years, when oil residues were more dispersed and evenly distributed, that the SE is obscured by the data point (Fig. 5).

The concentration of target aromatics and alkanes in surficial sediments was at least one order of magnitude higher 1 m versus 10 m into the marsh during the first 12 months after the oil spill (Fig. 6A and B). After the spring of 2011, however, the concentration at 1 and 10 m and their ratios were about equal (Fig. 6C). The oil distribution within sites and among estuarine basins became similar within two years. A June and September (2012) sampling along a 100-m transect into the marsh could not document an attenuation of concentrations within marsh sediments with distance (Turner et al., 2014a) (also Fig. 6A and B).

The only comparable study of oil residue concentrations along transects leading into the marsh was by Hester et al. (2016). Hester et al.'s first sample was in fall 2011, 18 months after the spill started and after considerable oiling of the marsh. The sites were divided into five classes of oiling, which were based on a visual estimation, not analytically. They estimated that there were lower amounts 100 m into the marsh than at the edge. There was a one order of magnitude reduction after four years, which is similar to the average oiling change demonstrated by our results (Fig. 6). Our conclusion is that sites that were unoiled became contaminated with petroleum hydrocarbons and from oil residues within two years. Further, defining sites using a binary choice, such as 'oiled' or 'not oiled,' at the beginning of the intrusion of oil residues into the marsh is not valid after 18 months without an analytical confirmation.

3.4. Differing compositions offshore and inshore in the first year

Lehr et al. (2010) estimated that the sum of collected, direct recovery, burned and skimmed oil from offshore represented 23% of the total oil spilled. Their estimate leaves the remaining 77% potentially in the GOM, where another 25% either evaporated or dissolved, and 10% was dispersed. The quality of oil buried in reservoirs was transformed after dissolution, evaporative losses, natural and chemical dispersion, particle attachment, photo- and biodegradation, and re-complexations (Fig. 1). A portion went to the seabed as mostly fossilized carbon from the spill, which Chanton et al. (2015) estimated was 3–5%. The term 'fossilized carbon' refers to "dead carbon" from the spilled DWH oil that was converted into biomass residues that ultimately ended up on the seafloor. Some of this biomass came from mineralization of oil on the seafloor, but much came from mineralization of oil hydrocarbons in the water column and on the surface that ended up being transported to the seafloor associated with sinking marine particles (marine snow). Thus, a considerable portion of oil residue remained in and on the water surface.

The proportional amount of naphthalene and alkyl naphthalene(s) declined from the peak of 58% of all target aromatics at the well head source oil (measured using the analyte molecular weight) to shelf and inshore waters and then on the marsh sediments (Fig. 7) (Supplemental Table S1). The percent dropped to 22% in the May 2010 water samples from offshore, and 1% at the highest

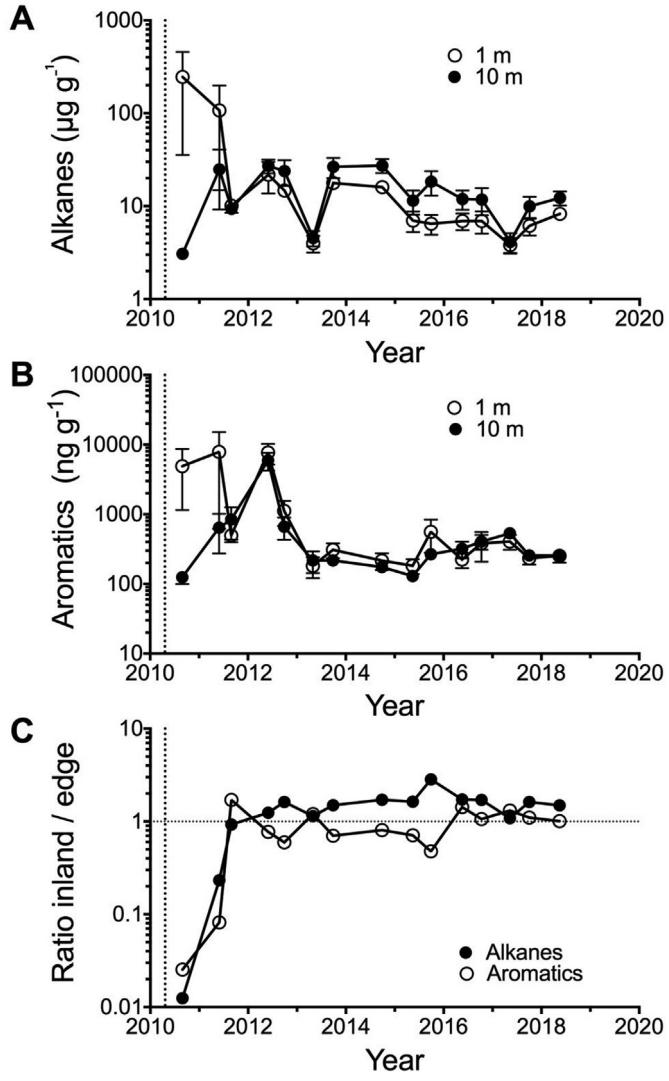

Fig. 6. The concentration for the marsh sediment samples taken 1 m and 10 m into the marsh by year. The vertical dotted line is the start of the oil spill offshore. A. Total alkanes. B. Total aromatics. C. The ratio between the concentration of alkanes and aromatics in the 1- and 10-m samples. The average ($\mu \pm 1$ SE) is shown for each sampling trip.

concentration of total target aromatics in September 2010, before rising to 27% in July 2014 (Supplemental Table S1). In contrast, the average of the naphthlalene and alkyl naphthalenes for each of the marsh sediment sampling trips was $\leq$8% of the total aromatics, including <2% at the peak concentration in February 2011 (Supplemental Table S3). Sixty percent of the petroleum hydrocarbons in oil and oil residues contaminating water samples on the coast at the peak concentration were comprised of three rings (mol. wt. < 234) (Table S1), whereas it was 28% in the marsh sediment samples. The low proportional amounts of naphhthalene and alkyl naphthalenes indicates significant volatilization and biodegradation, which is higher for smaller target aromatic hydrocarbons. The low amounts of naphthalenes in marsh sediments is consistent with the highly depleted concentrations in oil residues that reached the marsh shoreline, and left residues containing mostly the heavily-weathered oil residues, such as alkyl chrysenes isomers, asphaltenes, and resins which are the heavier by-products of oiling that accumulated on the marsh surface. Further, these oil residues also contain petroleum biomarker compounds, some of which were

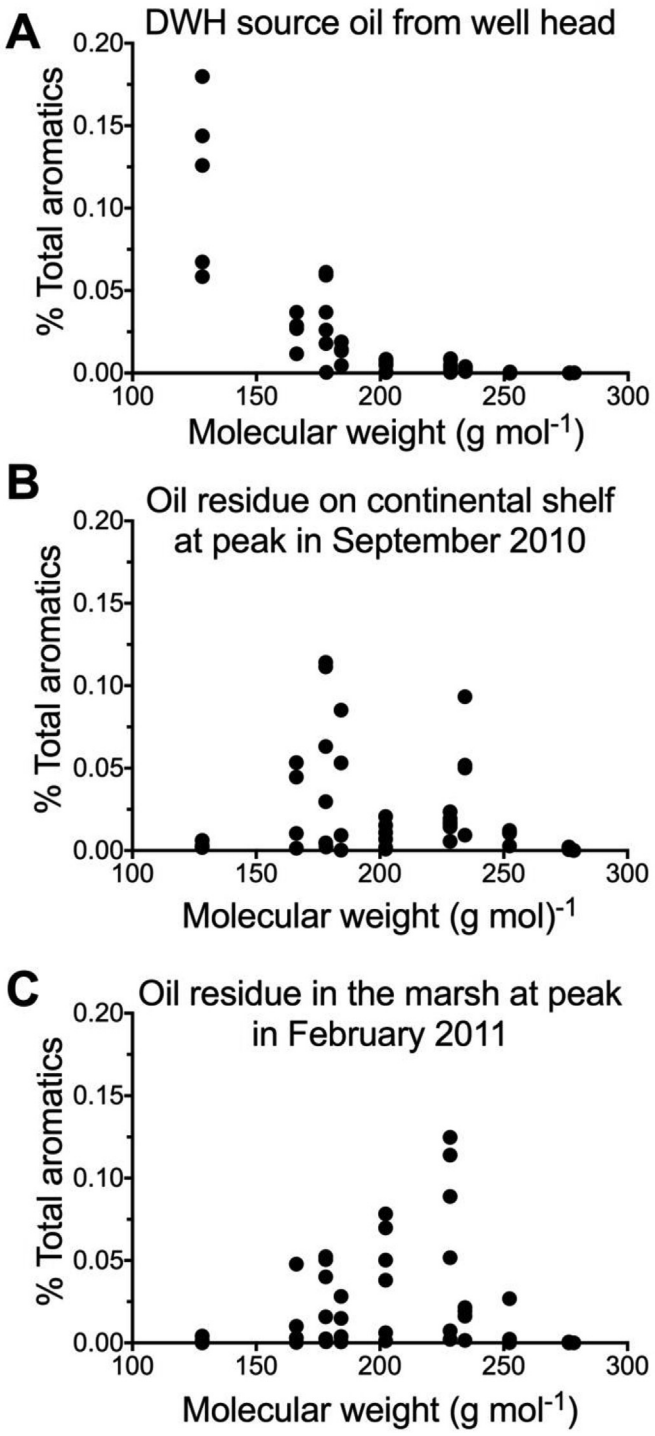

Fig. 7. The percent of the total aromatics by molecular weight for (A) the DWH source oil at the well head (Table S3); (B) during the peak concentration of oil residues in continental shelf waters in September 2010; and, (C) during the peak concentration of oil residues in marsh sediments in February 2011. Naphthalene and naphthalenes have a molecular weight less than 130 g mol^{-1}.

weathered and indicative of the composition found in the source oil from the well head (Meyer et al., 2018).

3.5. Degradation rates

The degradation rate of target alkanes and aromatics is affected

by a variety of factors, especially the molecular weight or carbon number (C_n), volatility, molecular solubility, eH (oxidation potential) and pH, sediment organic content, sediment type, oxygen availability and temperature. The target alkanes have a lower molecular weight and are more easily volatilized than alkanes with higher molecular weights, whereas the mid-length ($C_n = 14$ to 20) alkanes are generally non-polar liquids with minimal water solubilities (Overton et al., 2016). The target aromatics are, in general, 100 times more soluble in water than alkanes of the same carbon number (McAuliffe, 1966), which means that target aromatics are more prone to evaporative losses than are the alkanes. The oxygenated water plume near the well-site was enriched with water soluble components indicating some decomposition of DWH oil (Reddy et al., 2012). The evaporative losses of target aromatics moving from the well head to estuary were about 64% of the 10- and 12-carbon naphthalenes (Liu et al., 2012). In general, the higher the molecular weight of PAHs indicates a higher hydrophobicity and toxicity, and a slower degradation rate in coastal marshes. The effect of these and other physiochemical properties means that mid-length target aromatic hydrocarbons ($C_n = 14$ to 20) that are not readily volatilized or leached from sediments can remain biologically active unless degraded. Further, the decomposition of organics in anaerobic sediments, in general, is considerably slower than in aerobic sediments (Hambrick et al., 1980; Delaune et al., 1981). Laboratory studies by Bauer and Capone (1985) revealed no re-mineralization of either anthracene or naphthalene in intertidal sediments without oxygen and a temperature dependency. The results of other laboratory studies by Boyd et al. (2005) showed that the oxygen supply was the determining factor controlling PAH degradation. The result is that target alkanes and aromatics will only very slowly degrade once accumulated in anaerobic areas of marsh sediments, and much faster in aerobic waters or aerated surface sediments. Reddy et al. (2002), for example, found that pristane, phytane and other branched alkanes remained 30 years after the West Falmouth, MA oil spill in 1969 and predicted that "hydrocarbon contamination will persist indefinitely in the sedimentary record." This is in sharp contrast to the rapid degradation of the DWH oil's target alkanes and aromatics found in lab weathering studies under ideal mixing and nutrient levels using waters collected from the Louisiana marsh shoreline (Olson et al., 2017).

Oil legacies

The results of multiple studies suggest that some of the spilled oil and its residues may persist for decades and continue to negatively affect coastal ecosystems (Teal et al., 1992; National Research Council, 2002; Reddy et al., 2002; Culbertson et al., 2007a, b, 2008). Peacock et al. (2007) documented the persistence of oil 25 years after the Bouchard 65 oil barge spill, and oil persisted from the *Arrow* oil spill for more than 20 years (Vandermeulenn and Singh, 1994). Linden et al. (2004) reported that salt marshes oiled in the 2001 Gulf War had not recovered after 10 years because of the "absence of physical energy, wave action, and the anaerobic environment," which are the same conditions in the northern GOM salt marshes. We observed fresh oil in crab burrows on our sampling trips. D'Sa (2016) examined the fluorescent components of dissolved organic matter offshore before, during and after the spill and found that the variable fluorescence amounts and intensities in 2013 were consistent with the long-term persistence of the oil residues in the dissolved carbon pool until that time.

An oiling legacy can affect organisms through a complex suite of subtle interactions, perhaps with threshold effects, or dependencies from one species in both offshore (Joye et al., 2016) and inshore ecosystems (Rabalais and Turner, 2016). Peterson et al. (2003) provided several striking outcomes arising from a 10-year analysis of the 1989 *Exxon Valdez* oil spill. One of the strongest observations was that the impacts were more than from the immediate effect of toxins, but also from the unexpected persistence of toxins creating chronic stressors that continued to affect wildlife through indirect cascades of food web relationships that postponed recovery. The foundational species for many organisms is the emergent marsh vegetation that is used in many metrics of marsh restoration (Fleeger et al., 2018; Zengel et al., 2018). Michel and Rutherford (2014), for example, conducted a review of marsh oiling and concluded that six of 32 marshes on five continents had not completely recovered from an oil spill after 10 years. Lin et al. (2016) reported that in their heavily-oiled sites in Louisiana, which experienced near complete plant mortality in 2010, were somewhat recovered 42 months after the DWH spill with live aboveground biomass only 50% of reference values and belowground biomass in the upper 12 cm only 24% of reference values. Further, shoreline erosion accelerated as a result of oiling (McClenachan et al., 2013; Hester et al., 2016; Turner et al., 2016; Beland et al., 2017).

Some hydrocarbons from the spilled oil do make their way into food webs (Bonisoli-Alquati et al., 2016; Wilson et al., 2016). The indirect rather than toxic effects may be substantial and diverse, although not visually obvious. The concentrations of target PAHs in June 2013 marsh sediments remained at levels that affected the reproduction and growth of resident fish in laboratory experiments (Whitehead et al., 2012). The phytoplankton community shifted in 2010 along with a loss of abundance in some groups compared to a 19-y baseline (Parsons et al., 2014, 2015); fledged nests of the resident seaside sparrows were less frequent in marsh plots determined to be oiled versus those determined as unoiled (Bergeon Burns et al., 2014); and marsh periwinkle population size in heavily-oiled marshes remained reduced for at least five years after the spill (Zengel et al., 2017). Insect communities were depressed after this oiling (McCall and Pennings, 2012; Pennings et al., 2014; Bam et al., 2018). Fleeger et al. (2018) found that the meiofauna, nematodes, copepods, and juvenile annelids, excluding the polychaete *Manayunkia aestuarina*, recovered in about three years in near synchrony with *Spartina alterniflora* stem density, but other common taxa either recovered more slowly (i.e., juvenile bivalves and amphipods) or not at all. The microbes have not re-established to pre-spill compositions as of 2016 (Summers Engel et al., 2017). However, Rabalais et al. (2018) could find no difference in the low oxygen concentrations or mid-summer hypoxic area of offshore waters in 2010 compared to a 27-y history.

4. Conclusions

The oil released at 1500 m during the DWH oil spill created a spike in oil concentrations on the continental shelf which did not return to pre-spill concentrations for more than 5 years. The petroleum constituents found offshore were temporally synchronous with the residual oil in the Barataria Bay estuary, implying transport and mixing. The oil residues in the water fraction, having lost much of the lighter fractions, were not identical to those deposited on marsh vegetation and sediments. The initial oiling was widespread throughout Louisiana coastal marshes and eastwards, but not in all areas. The smearing of oil residues into the marsh after two years suggests that the total area oiled was larger than the initial oil distribution. These oil residues in sediments were in the form of a viscous fluid emulsion with heavier petroleum constituents that were further transformed under anaerobic conditions. The total target alkanes and total target aromatics remaining in marsh sediments in the eight years after oiling are each at least one order of magnitude greater than the background levels in

sediments immediately before the oiling. Some of these DWH oil residues will be re-suspended and distributed in marsh sediments throughout the estuary, perhaps to be released when the marsh erodes. The consequences of the remaining oil residues in the ecosystem will affect more than the emergent plant biomass to include bacteria, crabs, mussels, and perhaps fish, for example. A point of understanding these oil spill impacts, therefore, is to appreciate the nuanced set of interactions within ecosystems and to value decisions necessary for quick containment, effective damage evaluation and appropriate mitigation. A repeat of the DWH oil spill is undesirable, as the various courts have stipulated in criminal and civil judgments. Improved operations and better containment should be a mandate to avoid all oil spills of any size in petroleum recovery and production activities.

Declarations of interest

None.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2019.05.134.

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