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Long-Term Petroleum Hydrocarbons Pollution after a Coastal Oil Spill

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Abstract: The long-term status of petroleum hydrocarbons in both seawater and sediment contaminated by the Dalian New Port oil spill has been investigated since 2010. Seawater recovery is relatively swift and is complete within two years, while oil contamination persists in the coastal sediments for several years. Because of the slow degradation and low mobility in sediments, they serve as long-term reservoirs for residual oils. The erosion of sediments into the water column leads to an abrupt increase in hydrocarbons during storms. The cumulative results of hydrodynamic transport and ongoing industrial emissions lead to a spatial shift of hot spots with high petroleum hydrocarbon concentrations from the spill site to the inner corner of the bay. In addition to continuous petroleum hydrocarbon emissions from contiguous coastal outfalls, the regional oil contamination will persist indefinitely. The research provides comprehensive information for years to come to evaluate the long-term damage and multiphase medium impacts of a large oil spill.

Keywords: oil spill; petroleum hydrocarbons; environmental impacts; oil–sediment interaction; Dalian

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1. Introduction

Spilled oil has long been recognized as an ever-increasing problem for the oceans; it pollutes both beaches and sea surfaces and is consumed by fish, seabirds, and other creatures. The Dalian New Port oil spill (DLNPOS), occurred on 16 July 2010, released approximate 35,000 tonnes of crude oil into the coastal water [1]. Despite being of a smaller magnitude than the Deepwater Horizon Oil Spill in the northern Gulf of Mexico, the DLNPOS is regarded as the most serious oil spill disaster in the history of China [2]. Moreover, DLNPOS resulted in a more devastating disaster to coastal environments and marine organisms because of limited water self-clarification ability near the shoreline. Cleanup measures were immediately adopted to remove oil from the environment; however, a substantial portion of the spilled oil still remained in the marine environments. Over 100 km^2 sea area was covered with apparent slicks, and 20 km shoreline was heavily polluted by stranded oil–sediment aggregates [3]. The management in response to marine oil spills from ships usually pays close attention to mitigating acute rather than chronic environmental impact and natural resource damage [4]. Even if the visible oil film is quickly removed, the consequent damages of a significant leakage accident will have long-term effects including environmental quality, the marine life, and coastal ecosystem [5].

After the DLNPOS, many studies about its environmental impacts were carried out. Guo et al. (2014) simulated the short-term oil spill trajectory and fate considering transport processes under the action of wind-wave-current and weathering processes including evaporation, emulsification, and dissolution [2]. Numerical results indicated that most of the spilled oil had had entered into Dalian Bay and Dayao Bay and deposited at the surrounding shoreline. Polycyclic aromatic hydrocarbon concentrations after two months of the DLNPOS in seawater demonstrated a significant negative correlation with the distance away from the spill source, but not in sediments [6]. During the early stages of an oil spill, the sinking process of the buoyant residual oil from surface seawater to the benthic environment is not significant; meanwhile evaporation plays a primary role in removing oil from the sea in the first few days. In addition to dispersion and diffusion resulting from local turbulent mixing and current drift, petroleum hydrocarbon (PH) concentrations returned to normal one year after the DLNPOS [7]. Therefore, there are relatively few research results on the long-term tracking of petroleum pollution in water bodies.

Sediments which have accumulated residual oils can experience persistent contamination. PHs in tropical coastal ecosystems may exceed probable effect levels in the northeast of Brazil, which received a considerable quantity of spilled crude oil [8]. PHs levels in Bay Jimmy sediment 5 years after the Deepwater Horizon oil spill remained higher than in the pre-spill level [9]. More than half of sediment-adsorbed PHs are resistant to degradation, owing to a slow natural recovery rate. Oil pollution of coastal muds was detectable for over twenty years after the catastrophic Galeta oil spill [10]. Wang et al. (1998) found that degradation percentages of residual oil of surface sediments was less than a half by a field survey twenty-five years after spills [11]. A marked increase in sediment total petroleum hydrocarbon (TPH) concentration was recorded two years after the DLNPOS [12], and the high pollution level remained until six years later. Despite natural weathering, the diagnostic ratios of all the biomarkers remained stable 10 years after the Deepwater Horizon oil spill [13].

After the Dalian New Port oil spill, most field surveys were conducted before 2016. In order to assess the impact of the DLNPOS and the recovery of the marine environment, we conducted the most recent field survey to provide up-to-date information about the petroleum pollution status ten years after the spill.

2. Materials and Methods

2.1. Study Area and Sample Collection

A fuel pipeline exploded at the Dalian New Port on 16 July 2010, resulting in approximate 35,000 tonnes of crude oil being discharged into the coastal waters with more than 100 km^2 sea area seriously contaminated. Dalian New Port is located at the south end of Dagushan Peninsula, on the west coast of Dayao Bay, Yellow Sea, in China. We surveyed the spatial distribution of PHs in surface water and the sediments of several sites around the spill origin (38°58′15′′ N, 121°53′40′′ E) (Figure 1). The thirty-two sampling sites were divided into two groups, nearshore area (Sites N01–N12) and offshore area (Sites O01–O20). In 2003, 2006, and 2008, the PHs in seawater and sediment were investigated and analyzed to assess the coastal environment before the Dalian New Port oil spill (DLNPOS). Two months after the spill, a field sampling to assess the short-term impacts of oil spill was conducted. From 2010 to 2021, continuous field investigations were carried out each September, with the only exception being December 2020 due to the COVID-19 pandemic. Both seawater and sediment samples were gathered in a clean, acetone-rinsed glass bottle with Teflon-lined cap, and sent to the laboratory, and stored at −20 °C (sediment) and 4 °C (water) before sample treatment.

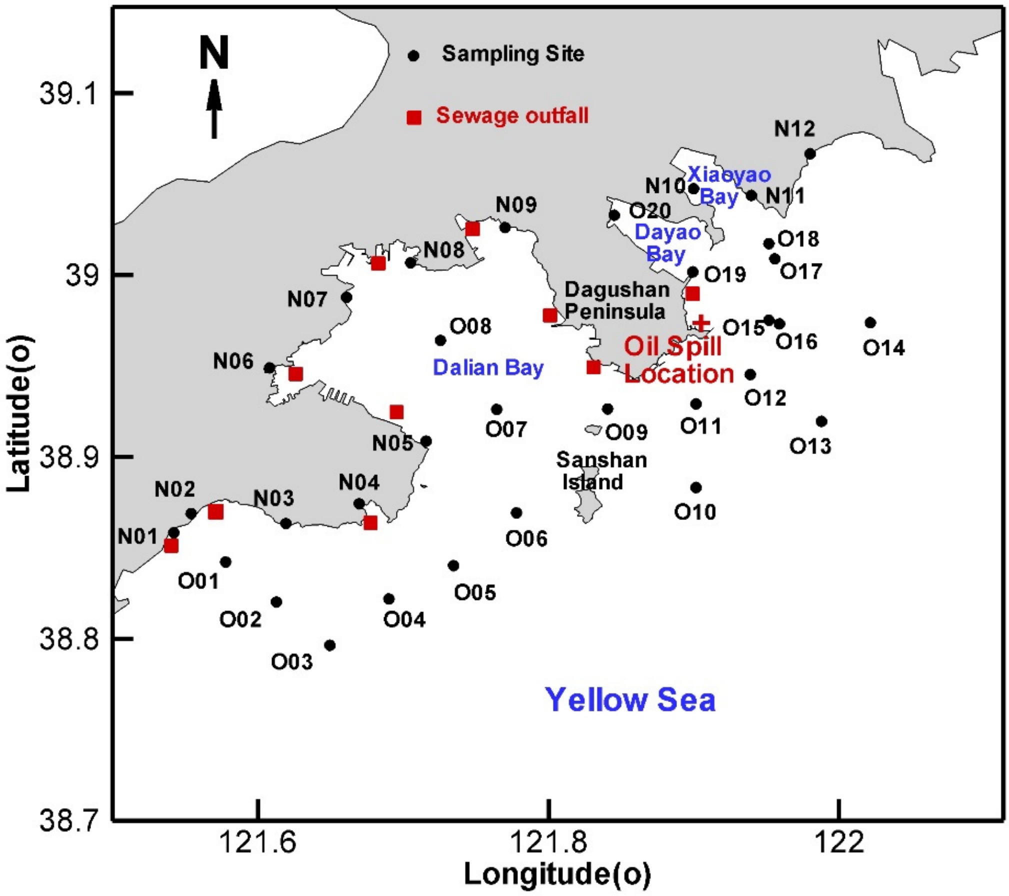

Figure 1. Sampling locations around the Dalian New Port oil spill site.

2.2. Analytical Method

The unfiltered seawater samples were analyzed by UV spectrophotometry for TPH according to the specification of China National Standards [14]. Each seawater sample was acidified to weak acidity with 1:3 sulfuric acid, then extracted with 10 mL × 2 hexane, the extract was sealed and stored at (5 ± 2) °C away from light, and its PH concentration was determined by UV spectrophotometry. Dissolved petroleum hydrocarbon (DPH) concentration can be determined by using the same approach to treat water samples after filtration through acid-treated Millipore filters (0.45 µm filter). Particulate petroleum hydrocarbon (PPH) level in seawater is obtained as the difference between TPH and DPH.

An accurate 1 g of air-dried sediment sample was weighed and extracted with 15 mL × 2 hexane, and then the hexane was washed with 20 mL sodium sulfate solution 3 times. The extract was sealed and stored at (5 ± 2) °C away from light and analyzed by UV spectrophotometry to obtain PHs content in sediment determined.

All sample collection, storage and monitoring were strictly conducted through quality assurance (QA) and quality control (QC). Recoveries of the standard samples range from 92.8 to 107.5%, and the method detection limits were 3.5 µg/L for seawater and 3 mg/kg for sediment. The relative standard deviation (RSD) for three parallel samples was <10%.

3. Results and Discussions

3.1. PHs in Seawater

The findings showed that total petroleum hydrocarbon (TPH) concentration in seawater varies both temporally and spatially (Figure 2). In September 2010, two months after the DLNPOS, the highest TPH concentration was up to 5.126 mg/L near the spill site. The concentration in Dayao Bay and Dalian Bay, once heavily polluted by oil slicks, was evidently higher than that in other sea areas. The hydrodynamic model provides important insights into PH transport processes under the action of sea currents and winds. Southerly winds prevail in Dalian from July to September, causing the highest concentrations of TPH in the north of the spill site. Tides in the North Yellow Sea account for the primary part of sea current. Due to tidal current asymmetries in velocity, residual oil slicks turned southwestward at the southern tip of the Dagushan Peninsula (Figure 3). Oil slicks between the Sanshan Islands and the Dagushan Peninsula had split up into two branches: one branch enters into the Dalian Bay, the other keeps flowing southwestward. The samples we collected in September 2010 showed slightly higher oil pollution levels than values from other surveys [15]. This is because some of the sites we investigated are located near the coastline, while all the sites where Guo et al. (2017) collected water samples are located offshore. Some untreated oil film still stayed along the low-energy coast. Due to extensive hand and mechanically assisted recovery techniques used after DLNPOS, the water quality of the polluted waters quickly returned to normal (Figure 2b). In September 2015, the TPH of contiguous coastal waters dropped to around 0.01 mg/L. There were some highly polluted areas at the top of Dalian Bay, Dayao Bay and Xiaoyao Bay due to the poor local water exchange ability. The values we obtained are in good agreement with those from previous surveys of Guo et al. (2017) [15]. TPH levels were evidently high in the sea area over 30 km southwest of the spill site. This result can be explained by assuming that PHs are carried by the southwestward residual current from upstream. Another non-ignorable reason is that the routine discharge from coastal sewage outfalls has been increasing with the development of local society. The discharge standard of PHs from marine outfalls should not exceed 12 mg/L. Even if the discharged sewage meets the standard, the cumulative discharge of a large number of sewage outlets will lead to the increase in the concentration of coastal petroleum pollutants.

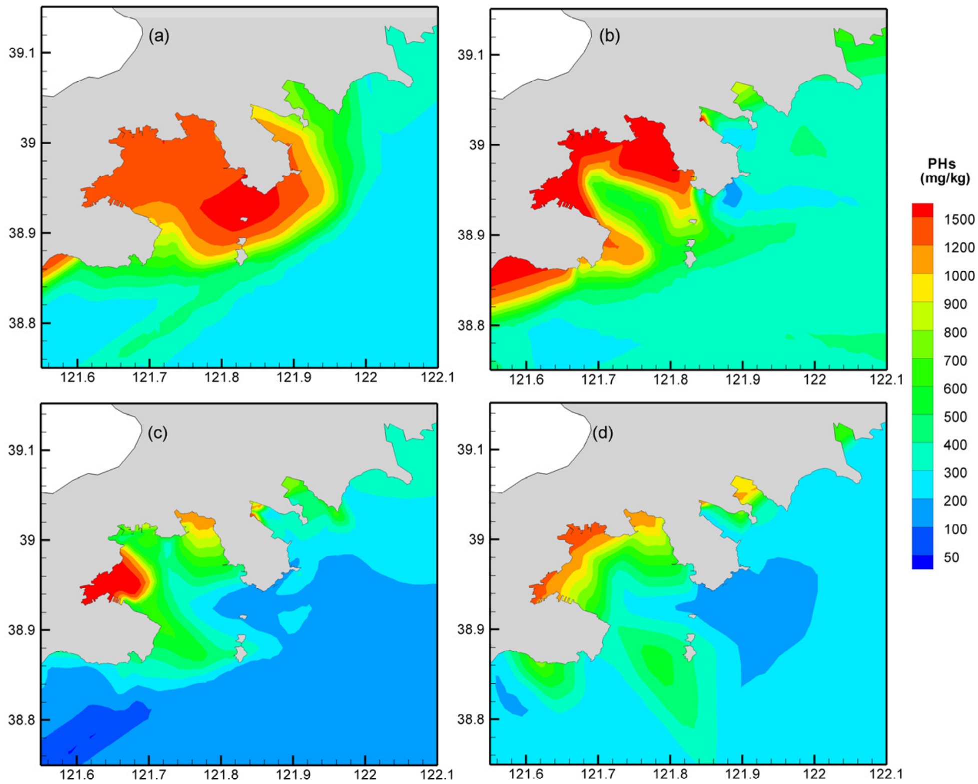

Figure 2. Spatial distribution of TPH in seawater in different years, (**a**) September 2010, (**b**) September 2015, (**c**) December 2010, and (**d**) September 2021.

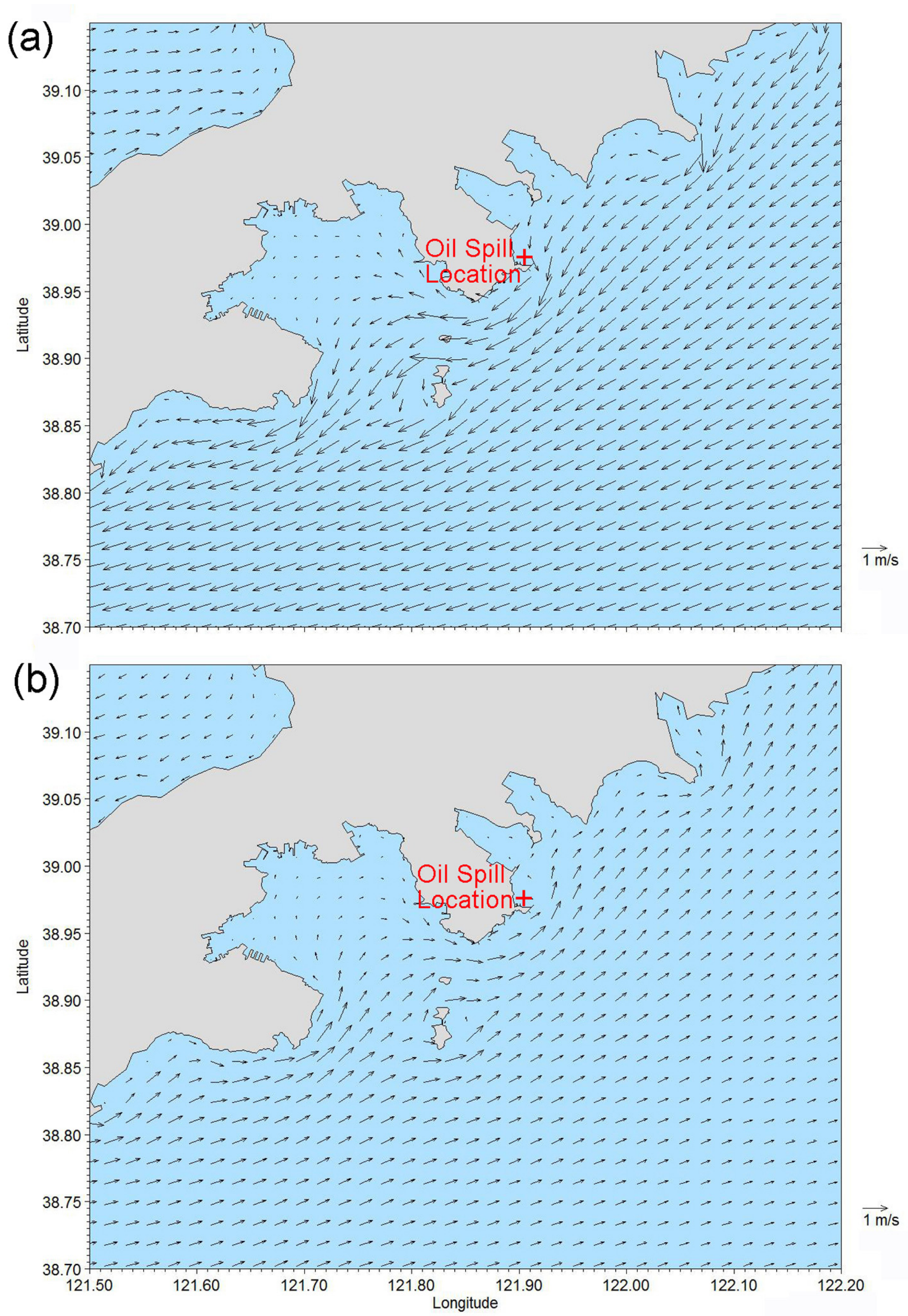

Figure 3. Tidal currents near Dalian New Port oil spill site during maximum (**a**) flood and (**b**) ebb. Arrow length represents flow intensity, and arrow direction means current direction.

Levels of TPH in water ranged from 0.024 to 0.063 mg/L with a mean of 0.035 mg/L in December 2020, and ranged from 0.016 to 0.121 mg/L with a mean 0.068 mg/L in September 2021. Both of mean values of TPH in September 2020 and December 2021 were remarkably lower than 2.512 mg/L in September 2010. The highest concentrations were found in seawater samples from FJZ site near a sewage outlet in December 2020 (Figure 2c). Owing to the terrigenous loads and weak exchange ability, the contamination level in the northwest corner of Dalian Bay is also generally high. Surprisingly, the survey results of December 2020 and September 2021 were in sharp contrast to each other in terms of

TPH concentrations. First, the average concentration of TPH increased about twofold in less than one year. Second, high pollution levels occurred in the outside open sea area rather than in the inner corners of Dalian Bay, Dayao Bay, and Xiaoyao Bay. Moreover, TPH concentration in September 2021 reached a peak during the recent eight years, while no oil spill event had happened during this period. There is no large river input, and seasonal factors will not lead to such drastic variations. Sediment resuspension must be the most probable source of TPH to the water column. In most cases, marine environmental recovery is relatively swift. By 2015, the apparent impact of the oil spill in the surface waters had vanished. However, oil contamination can persist in the marine bed sediments, and this is confirmed by the trend of PH contents in sediments. Residual oil deposited on the seabed will re-enter the water column with the suspension of sediment during storms, causing the secondary pollution of seawater.

Field surveys were usually conducted on a calm day. We collected samples on 23 September 2021, which was also a calm day. However, three days before the field survey, extreme weather occurred in Dalian with maximum wind force up to 26.6 m/s. The maximum significant wave heights reached 3.5 m off the coast of Dalian, approximately triple the usual (Figure 4). During storms, such large waves easily erode bed sediment and keep sediment in suspension. Average suspended particulate matter (SPM) concentration on 23 September 2021 was 41.6 mg/L, more than four times of 10.1 mg/L on 8 December 2020. The wave height rise inside the Dalian Bay, Dayao Bay, and Xiaoyao Bay is not as significant as that in the offshore area, and correspondingly, the increase in TPH concentration was comparatively small. One of the evidence supporting this assumption is that spilled oil penetrated 16 cm on the seabed within 3 years after the accident [3]. The surge of oil level in water caused by storms is not a unique case. On 20 August 2018, when the typhoon "Rumbia" passed by Dalian, the wild waves stirred up the oil substances on the sea floor, resulting in the oil concentration in the water body elevating to 5 mg/L [16].

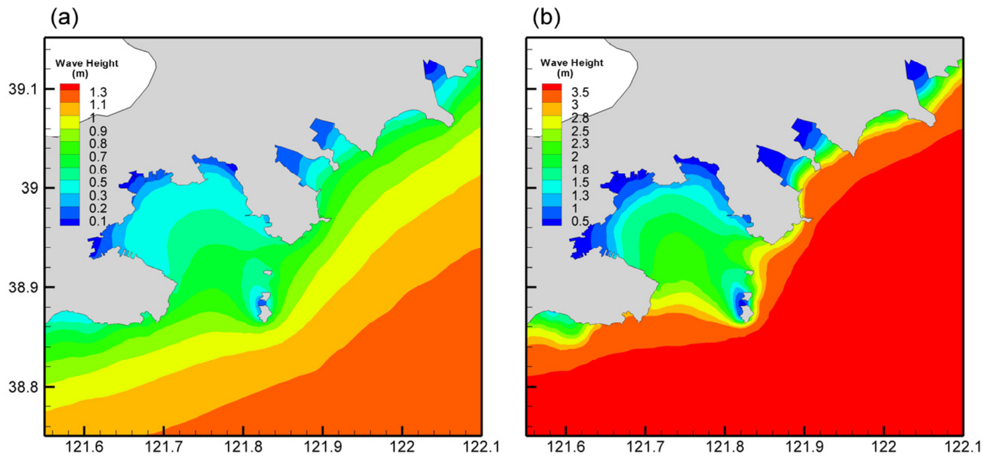

Figure 4. Annual mean significant wave height distribution (**a**) and significant wave heights on 20 September 2021 (**b**).

We monitored changes in suspended matter concentration, and it was found that there was a complicated correlation between PHs and suspended matter (Figure 5). Although the general pattern of PHs is dependent on SPM, Figure 5 demonstrates that PHs tend to increase with SPM concentration between 10 and 50 mg/L. A sudden increase in the SPM concentration is caused by the resuspension of bottom sediments, which brings pollutants adsorbed in sediments into the water column. When SPM concentration is too high, the particulates show a strong adsorption effect on the hydrophobic PHs as a scavenger to control the pollution level. Once SPM is below 10 mg/L, PHs usually do not decrease with it. Low SPM often occurs in calm seas, where low-energy hydrodynamic conditions hinder the dilution and degradation of PHs. Strictly speaking, the interactions between spilled oil and SPM are complex. Suspended particles with the ability to bind hydrophobic compounds lead to a significant amount of oil sedimentation. Field studies indicated that as much as a half of the insoluble hydrocarbon had been removed from the sea surface to the benthic environment [17]. To some extent, sediment can be regarded as an oil remover in the water column. However, contamination will persist indefinitely due to the more stable properties of the hydrocarbon after oil sedimentation through agglomeration. A much larger area on the seafloor contained residues of Deepwater Horizon spilt oil than previously recognized was published in the literature [18]. Residual oil deposited on the seabed will re-enter the water column with suspension of sediment during storms, causing the secondary pollution of seawater; that is the reason why TPH in September 2021 was so high.

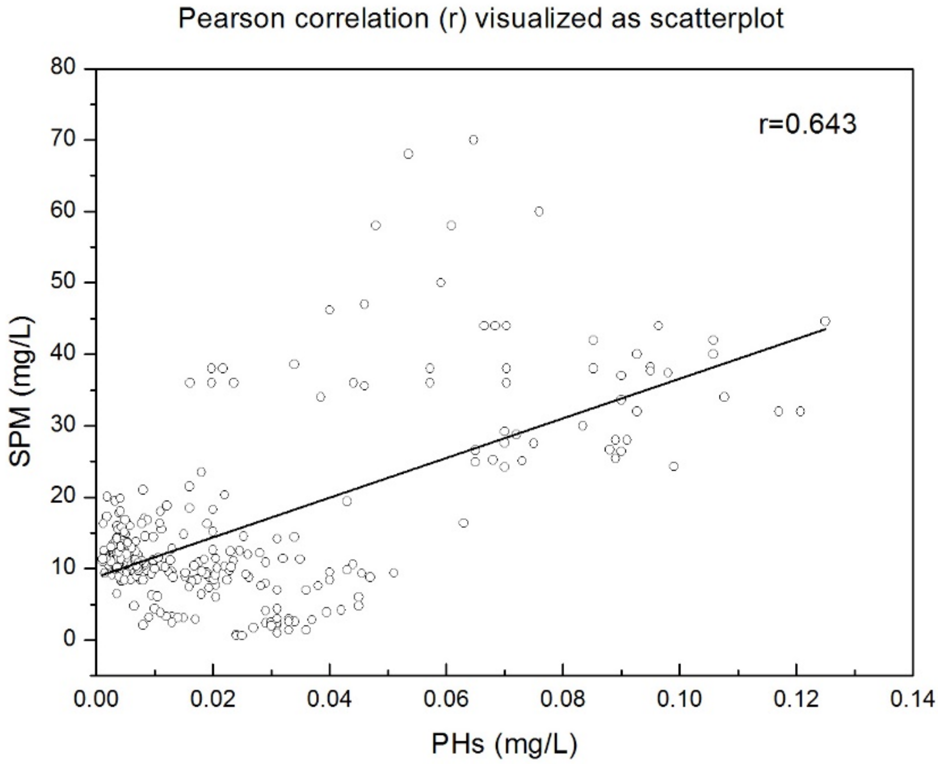

Figure 5. A scatterplot of the SPM levels as a function of the PH concentrations from historical monitoring results.

Similar incidents occurred after the Deepwater Horizon oil spill. Carcinogenic equivalents in Gulf menhaden tissues increased from 2012 to 2013; however, total polycyclic aromatic hydrocarbons in their tissues did not increase during the same interval, indicating an oil re-suspension event rather than exposure to a new source of oil [19]. The water near Dalian New Port is much shallower, and the deposited oil is theoretically easier to resuspend in the water. However, due to low frequency of storm surge in Dalian and quick sediment re-deposition after storms, the resuspension of deposited hydrocarbons have been ignored in previous surveys.

PHs are transported in the water column in complex forms, including particulate, colloidal, and dissolved fractions. For the water samplings collected in 2020 and 2021, the physical separation of particulate and dissolved phase PHs was based on in situ filtering 1 L water samples in-line through capsule filters with 0.45 μm pore size. On the one hand, the concentration of particulate petroleum hydrocarbons (PPH) increased sharply from 0.008 mg/L sampled in December 2020 to 0.055 mg/L in September 2021 with the presence of high suspended sediment concentration (Figure 6). On the other hand, the mean dissolved petroleum hydrocarbon (DPH) level in the particles dropped from 0.027 mg/L in December 2020 to just 0.013 mg/L in September 2021. Sediment entering into the water column enhances the TPH contamination levels while it plays a role as adsorbent for

DPH. Meanwhile, the average PH content in the suspending particles decreased from 2473 to 1422 mg/kg with increasing SPM level, because PHs are preferentially absorbed by fine-grained particles, whose fraction remains considerably elevated during or a few days after a storm event. It is thought that erosion of sediments and release of hydrocarbons into the overlying water column leads to a water-borne hydrocarbons increase [20]. Our findings support this judgment and clarify the increase in particulate components rather than the dissolved.

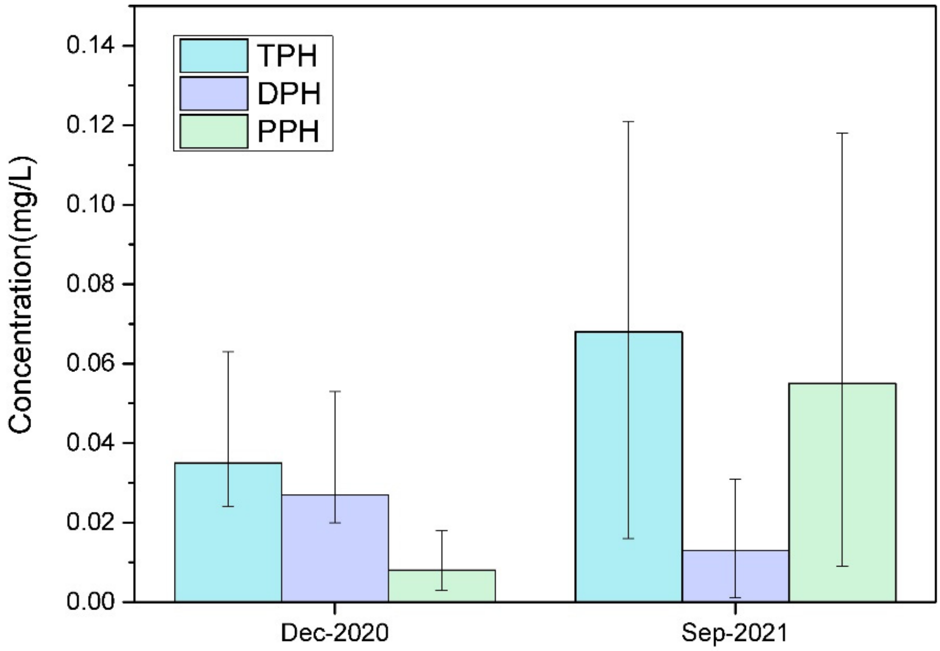

Figure 6. Concentration of different PH components in Dalian coastal waters. Bars represent the mean concentration and error bars represent the maximum and minimum of the data from each survey.

3.2. PHs in Sediment

Spilled oil is subject to a variety of weathering processes, including evaporation, dissolution, emulsification, photo-oxidation, biodegradation, and oil–sediment interactions [21]. In recent years, the sediments have been increasingly recognized as a major sink of pollutants [22]. Dissolved oil components and dispersed oil droplets interact with sediment through either adsorption or direct aggregation in the sediment phase [23].

There are two ways for oil to enter into sediments: oil stranded on the shoreline and oil sinking in the subtidal zone. The extensive pollution of the intertidal sediments after DLNPOS was of great concern. Cleanup was carried out by mechanical removal, so the oiled shoreline was soon restored to a semblance of normality. Oil–mineral aggregate (OMA) formation is also instrumental in the natural removal of stranded oil from the coastal environment under the action of physical mixing and sediment relocation [24]. With a large amount of oil pushed up against the shoreline for several days, OMA offshore transport into the subtidal zone was a significant possibility.

Despite extensive hand and mechanically assisted removal techniques used during the initial clean-up for DLNPOS, the remaining oil impracticable to remove, an estimation of 10% total spill amount was deposited to accumulate in the sediments [25]. When the light component volatilizes, the heavy component with high density also enters the water body easily under the action of waves. Dispersed oil droplets may aggregate readily with or be adsorbed on the suspended sediments, then sink to the seabed once the sediment-carrying capacity of the water column decreases [26]. Even though invisible on the sea surface, the chemical stability of the sediment-associated hydrocarbons would be enhanced after sinking to the seafloor. The extent and effect of oil on the floor after DLNPOS remain unknown, which becomes the blind spot of environmental management after an accident.

Along the extension of the tidal current, the maximum content of PHs in sediment (1928.1 mg/kg) was found between the Dagushan Peninsula and Sanshan Island just two months after DLNPOS (Figure 7a). The highest petroleum content in sediments in September 2010 were higher than values from Gu et al. (2021) [12], whose sampling sites were not arranged in this region. Similarly, the average value 669 mg/kg of our survey results was higher than the values of Gu et al. (2021). The average value of our survey results was closer to that of Guo et al. (2017). It should be noted that Guo's survey results included a site with 2856 mg/kg PHs content, showing high intersample variability. Deposited sediments remain stable at small flow velocities, but once the flow velocity or wave height became large enough, the sediments resuspend into the waterbody. The subtidal groove of the Peninsula and Sanshan Island is a deep sea area with strong current and better water exchange, which contributes to pollutant dispersion.

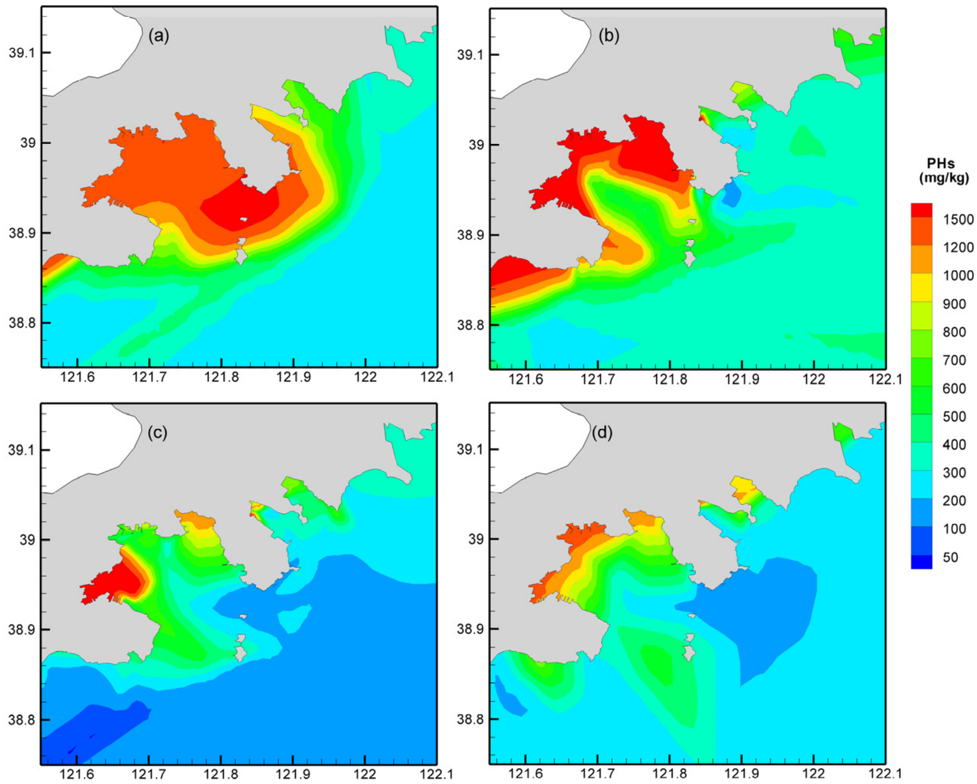

Figure 7. Spatial distribution of PHs in bed sediment in different years, (**a**) September 2010, (**b**) September 2015, (**c**) December 2010, and (**d**) September 2021.

Sediments are kept in suspension due to the presence of turbulent fluctuations until the sediment carrying capacity decreases. Most of sediments directly enter the head of Dalian Bay and are deposited in nearshore areas under the action of wind-driven and tidal currents. The weaker waves and flow intensity in the corner of the bay cause less resuspension and more deposition of sediment. The continuous onshore sediment transport produces a PH accumulation over 1500 mg/kg in Dalian Bay, especially close to the shoreline (Figure 7b).

In December 2020, the maximum PH concentration occurred in nearshore sediments at the northwest corner of Dalian Bay, one order of magnitude higher than other subtidal regions (Figure 7c). This was attributed to more than one PH source in this zone. Even before DLNPOS, background PH concentrations were quite high due to anthropogenic emissions and the low self-purification ability of environment.

Large waves lead to increased suspended sediment concentration during windy periods and cause a redistribution of sediment from high to low pollution areas (Figure 7d). Although these extreme events last for only a few days, they play an important role in the local PH's re-distribution and fate.

The spilt oil type in Dalian New Port event was crude oil. After the light component volatilized rapidly, the heavy component entered the water column after emulsification. There was almost no oil film on the sea surface just one month after the spill accident. The oil droplets entering the water body continued to sink to the seabed, and the amount of oil in the sediments near Dalian New Port has been increasing slowly over the following years. It is generally agreed that the outflow discharge of oils from port and shipping are heavy oils and bunker fuel, and industrial sewage mainly contains light oil.

Apart from weathering, dissolution, evaporation and other physical processes affecting the oil, the types of coastline are also an important factor on the impact of spilt oil on a region [27–29]. The coastal landforms near Dalian New Port are mainly bedrock type and sandy type. Most of the deposited oil was removed quickly after stranding. Residual buried oil flocculated aggregates of a solid-stabilized emulsion where oil droplets were coated with mineral fines and transported by tidal washing and wave erosion. Once oil–SPM agglomerates settle to the bottom in the subtidal zone, a different set of weathering processes are expected. Microbial degradation becomes the major approach to the removal of hydrocarbons, and it is much slower than photochemical degradation near the surface.

3.3. PHs Temporal Trends

Figure 8 summarizes the PH contamination levels in both seawater and sediment sampled on control sites (12 nearshore and 20 offshore) near the spill source from 2003 to 2021. The annual evolution trend of petroleum concentration is consistent with the survey results of Guo et al. (2017) and Gu et al. (2021).

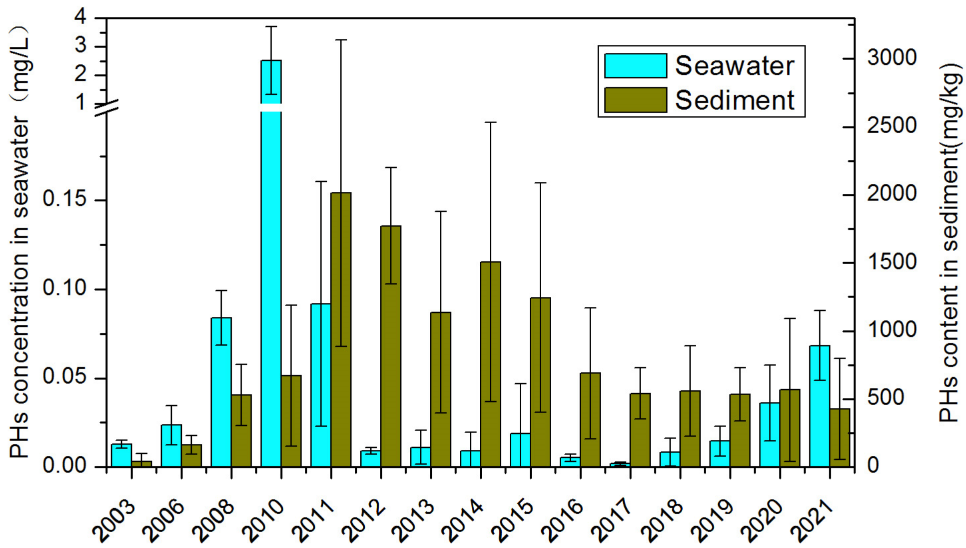

Figure 8. Petroleum hydrocarbon concentrations in seawater (cyan bar) and petroleum hydrocarbon content of sediment (dark yellow bar) in Dalian coastal sea from 2003 to 2021.

It should be noted that even before DLNPOS, the PH concentration in seawater has increased annually with the development of the shipping industry and aggravation of industrial and domestic sewage. The steep increase in the PH concentration of seawater up to 2.512 mg/L was attributed to DLNPOS. Similar to the quick fall in water-borne hydrocarbons after the Exxon Valdez spill [30] and Deepwater Horizon spill [31], the PH level had dropped to 0.092 mg/L in the next year, 2011, attributed to the immediate cleanup operation and marine dilution capacity. In 2012, the concentration of PHs in the water was completely restored with a mean value 0.009 mg/L, even lower than that from before the accident. The level of PHs in water has remained at a low level since 2012, yet it increases year by year. However, there are a variety of PH sources, excluding accidental or voluntary discharge, around industrialized coastal cities, resulting in an evident upward trend of PH levels since 2017. The average concentration 0.068 mg/L in September 2021 was the highest value in nearly a decade. Year-to-year levels of PHs varied in two distinct phases, consistent with rapid loss in the water column while slow loss after deposition to the seafloor.

Though there have been no recent large-scale oil spill accidents responsible for the sharp increase, deposited oil-contaminated sediments can be identified as a secondary pollution source under extreme conditions. The International Convention for the Prevention of Pollution from Ships (MARPOL) regulation prohibits ships from discharging disinfected and comminuted sewage when they are three nautical miles from the nearest coastline, while the restrictions on graywater are lacking. With the increase in shipping traffic, wastewater discharge from ships becomes inconvenient [32]. The new global regulations allow for installations of exhaust gas cleaning systems for the use of less expensive heavy fuel oils at the expense of discharging large quantities of petroleum hydrocarbons directly to the marine environment [33]. Heavy fuel oil combustion with scrubbers could aggravate the local environmental load.

Compared to seawater, the PH content in coastal sediments has an evident time lag. The PH concentration in bottom sediments in 2010 increased by 20% compared with 2008, but did not reach the peak. The investigation in 2010 was conducted two months after the oil spill. Although there was no visible oil film on the sea surface at that time, the residual oil did not settle to the seabed, but suspended in the water column as neutrally buoyant oil–sediment aggregate. When oil–sediment aggregates form after the processes of oil sediment collision, they may stay in the water for a long time [34]. Once their density exceeds that of seawater as the composition changes, they will fall to the seabed. This large-scale settlement occurred intensively during a few months after DLNPOS accident, generating the peak PH concentration in the sediment appearing in 2011. A considerable part of the residual oil was stranded along the coast as oil–sediment aggregate and then transported from the intertidal zone to the subtidal zone through sediment movements. Oil partitioning into sediments is much less prone to degradation, since photodegradation of PHs in seabed sediments is probably not a significant process where little light can penetrate [20,35].

Due to the slow degradation of petroleum in sediments, the PH concentration did not recover to the roughly equivalent pollution level of 528.4 mg/kg in 2008 until 2017. There has been no significant change of PHs in sediment since 2017, while the concentration of PHs in seawater is increasing. It implies that daily discharge has become the main PH pollution source. Although municipal waste discharges and operational discharges from ships exceed emissions from damaged oil tankers, oil pipelines and drilling platforms, a large amount of spilled oil from accidental spills in a short time has a dramatic and persistent environmental impact. PHs entering the sea through routine operations immediately transported by wind, current and wave processes disperse naturally. Later, their concentration may quickly be drawn down by evaporation, chemical/biological degradation, and physical dilution without polluting the seabed [36]. In contrast, water-in-oil emulsification may increase droplet effective volume and then improve the chance of collision with suspended sediments to produce oil–mineral aggregates. Once the oil–sediment aggregate occurs, it transports residual oil to the seabed with a density heavier than both seawater and crude oil.

4. Conclusions

Our continuing follow-up survey results suggest that there are reasons for both optimism and concern about the consequences of DLNPOS on adjacent coastal environment. On one hand, the contamination level of PHs in the water column returned to its previous state after just one year, due to the degradation and dilution effect. On the other hand, the quality of seawater may improve after an on-site cleanup, but it does not mean that pollution has disappeared or that the ocean has returned to its original state.

The recent rise of seawater PHs may be related to new sources discharging industrial and domestic sewage rather than the incident of 2010, due to the long time interval. With the increase in oil-bearing wastewater discharge in the surrounding sea area, the outlook for PH pollution status is not optimistic. Moreover, the quality of seawater is only one aspect of the impact of oil spill pollution on the ocean. PHs adsorbed by suspended sediments sink on the seabed and undergo long-term environmental persistence. Residual oil deposited on the seabed will re-enter the water column with a suspension of sediment during storms, causing the secondary pollution of seawater. Variable degradation rates occur in two distinct phases and the exchange process between them results in environmental quality fluctuations. These findings are of importance for comprehensively evaluating the long-term environmental impact and conducting effective remediation measures.

In this paper, we focused on the overall fate of PHs in seawater and sediment. Future reports will address other aspects generated by the DLNPOS event, such as the persistent organic pollutants. Further, an investigation on the bioaccumulation and trophic transfer of oil residues around the spill site should be carried out in the following research.

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