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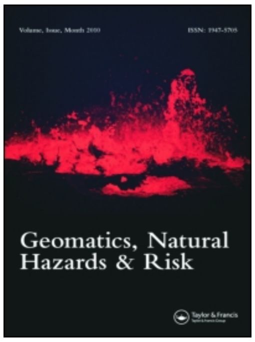

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Ecological response of phytoplankton to the oil spills in the oceans

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ABSTRACT

Oil spills in oceans have substantial influence on marine ecosystems. This study investigates 21 oil spills in the world. Analyzing Chlorophyll-*a* (Chl-*a*) from Moderate Resolution Imaging Spectroradiomerer (MODIS) data after Penglai oil spills on 4 June 2011, found a bloom with peak value of Chl-*a* (13.66 mg m^{-3}) spread over an area of 800 km^2 during 18–25 June 2011, and a pronounced increase in the monthly Chl-*a* concentration (6.40 mg m^{-3}) on June 2012 in the Bohai Sea. Out of the 21 oil spills, 14 blooms were observed, while 11 blooms associated with oil spills in the time interval of 3–10 months. In total, about 75% blooms occurred during June–August. Among all 14 blooms, 72% appeared when temperature was warm (20–30 °C), 7% appeared when temperature was low (10–20 °C), and the remaining 21% occurred when temperature was lower than 10 °C. This research concludes that the odds of a phytoplankton bloom after an oil spillage are higher at the time of higher temperature (>20 °C). The short-term impact of the oil spills on ecosystem could mainly depend on the quantity and composition of oil, while the long-term impact of the oil spills on ecosystem could be related to biodegradation of microorganisms.

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oil spills; phytoplankton; ecological; chlorophyll-a; bloom

1. Introduction

The oil spills in the ocean can have considerable influence on marine ecology, which have become one of the serious environmental concerns. Despite declining frequency, the risk of oil spills remains high in the global. According to the report of National Oceanic Administration in 2010, in average, an oil spill happens once every 4 days in coastal areas of China.

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The common list of reasons for oil spills in the ocean include offshore/onshore oil exploration, movements of oil tankers, ships, cruise sand submarines, and the waste produced due to general operation of the tankers (unloading, filling oil tankers, and cleaning) (Rogowska and Namieśnik 2010; Abbriano et al. 2011). Oil spills greatly impact shallow water and coastal resources, fishery resources, tourist industry, and ecosystems, especially mangroves, and salt marshes (Gundlach and Hayes 1978).

Cleaning up the affected ocean area is the first phase of the recovery in ocean conditions after an oil spill, and the recovery varies upon the ocean conditions. The BP Deepwater Horizon oil spills near the south-eastern Louisiana coast occurred on 20 April, 2010 was one of the worst oil spills in the global oil-spill history. About 200 million gallons of crude oil spilled in the Gulf of Mexico affected ocean ecology for a long time. Using ground and satellite data, photosynthetic activities and physiological status of the coastal salt marshes associated with oil spills were studied and marked decrease in photosynthetic activities (Mishra et al. 2012) were found. On 6 Jan 2018, the oil tanker 'SANCHI' collided with the Hong Kong bulk carrier CF Crystal" in the East China Sea. The 'SANCHI' was loaded with one million-barrel (about 136 thousand ton) gas condense that was a low-density mixture of hydrocarbon liquids of which the main component was a mixture of C5 to C11. The SANCHI's sinking resulted in a big leaking and burning of gas, with many oil spills areas observed from found satellite images.

Oil spills form oil films on the water with different thickness. These oil films prevent the interaction between ocean and atmosphere, including the exchanges of gas, heat and energy transfer (Mei and Yin 2009; Ivanov 2011) due to pronounced reduction in refractive index of ocean surface. As a result, phytoplankton photosynthesis intensity changes drastically, increasing or decreasing depending on the type and concentration of oil spills (Gordon Jr and Prouse 1973). Numerous parameters, for example, oil type and thickness of oil spills influence algae species and marine environment (Parsons et al. 1976; Davenport et al. 1982).

Oil spills have pronounced influence on zooplankton. High concentrations of petroleum hydrocarbons enhance toxicity of ocean water that can affect the brain, liver, kidneys, and cardiac performance of aquatic and terrestrial organism (Peterson et al. 2003; Milinkovitch et al. 2013), increase mortality rate and deformity (Abbriano et al. 2011), decrease the survival rate of eggs and larva, and influence the growth and reproduction of zooplankton (Yang et al. 2009). Oil spills also influence marine mammals, birds and benthos (Banks et al. 2008; Jung et al. 2011). After the oil spills, immediate cleaning is important to avoid adverse impact on ocean ecology for long period (Bernabeu et al. 2013).The use of dispersants further contaminants oil and ocean water complicate the marine ecosystems (Lan et al. 2012; Claireaux et al. 2013; Milinkovitch et al. 2013).

Although some studies show no obvious influence of oil spills on phytoplankton, especially on community structure (Moldan et al. 1985; Varela et al. 2006) many other studies found important influence of oil spills on marine phytoplankton. In a short term, the concentrations of phytoplankton reduced, and in the long term, outbreak of algal blooms occurred when the Chlorophyll-*a* (Chl-*a*) concentration increased (Lee et al. 2009; Sheng et al. 2011; Pan et al. 2012). High Chl-*a* concentration of phytoplankton were also found soon after the oil spills (Miller et al. 1978; Johansson et al. 1980; Hu et al. 2011). Similar conclusion can be drawn from different regional and environmental conditions: small amount of oil spills are unable to trigger blooms in the open ocean; but in closed or semi-closed ocean, serious oil spills would lead to blooms (Zhou et al. 2013). Changes in the Bohai Sea ecology associated with oil spills on phytoplankton are still not well understood. Here, we have made efforts to get better understanding of changes in ocean ecology caused by the Penglai oil spills. Based on the analysis of ground and satellite data, we present a possible mechanism of algal bloom initiation associated with the oil spills.

2. Study area, data and methods

2.1 Study area and the Penglai oil spills in June 2011

The Bohai Sea is one of the Chinese marginal seas, covering an area of approximately 77,000 km^2, with an average depth of 18 m. The sea surface temperature (SST) in the Bohai Sea varies in the range 0–28 °C (Tang et al. 2004b; 2006). During monsoon season (June–August) wind is southerly and during winter (December–February) season northerly wind is dominant (Sun et al. 2010). The Bohai Sea is covered by sea ice for about three months during November–February with relatively weak currents. Bohai Strait is only 150 km in width connect to the Bohai Sea and the Yellow Sea with water exchange between the two seas relatively feeble. Within the area of 37.12–41.00°N, 117.45–122.28°E that the Bohai Sea covers (Figure 2), we have considered four sampling sites (more than 5 m deep) to study the impact of oil spills (Figure 1b). In the Bohai Sea, oil exploitation started in 1967, the oil and gas production has been boosted from 10 million tons per $year^{-1}$ during 1967–2004 to 20 million tons $annually^{-1}$ during 2004–2009, and further increased to 30 million tons a $year^{-1}$in 2010. Consequently, rising oil production has led to frequent oil spills (Sun et al. 2009).

On 4 June 2011, the Penglai oil spills, occurred in the Bohai Sea and the spill continued until the end of August (Figure 1a). The oil spills slowly spread and seriously affected many coastal sites (Figure 1b). About 7,070 tons (http://china.caixin.com/2011-09-16/100305911.html) of crude oil were estimated in an area of 6,200km^2 in the Bohai Sea (Figure 1c).The disaster was unprecedented and imposed a serious threat to biological productivity and marine ecology and continued until 2013(source - Marine Bulletin from the State Oceanic Administration (SOA) of the People's Republic of China). On 15 July 2011—within 1 month—red tide (about 3.70 km long) was observed at a distance of ∼5.56 km from the Platform B of the Penglai Oil field and 3.70 km from Platform C (Figure 1d).

2.2. Study data

2.2.1. Details of the oil spills

In the present study, we used the historical data of the oil spills occurred in the Bohai Sea from various sources, such as the State Oceanic Administration of China (http://www.soa.gov.cn/), the China Oceanic Information Network (http://www.coi.

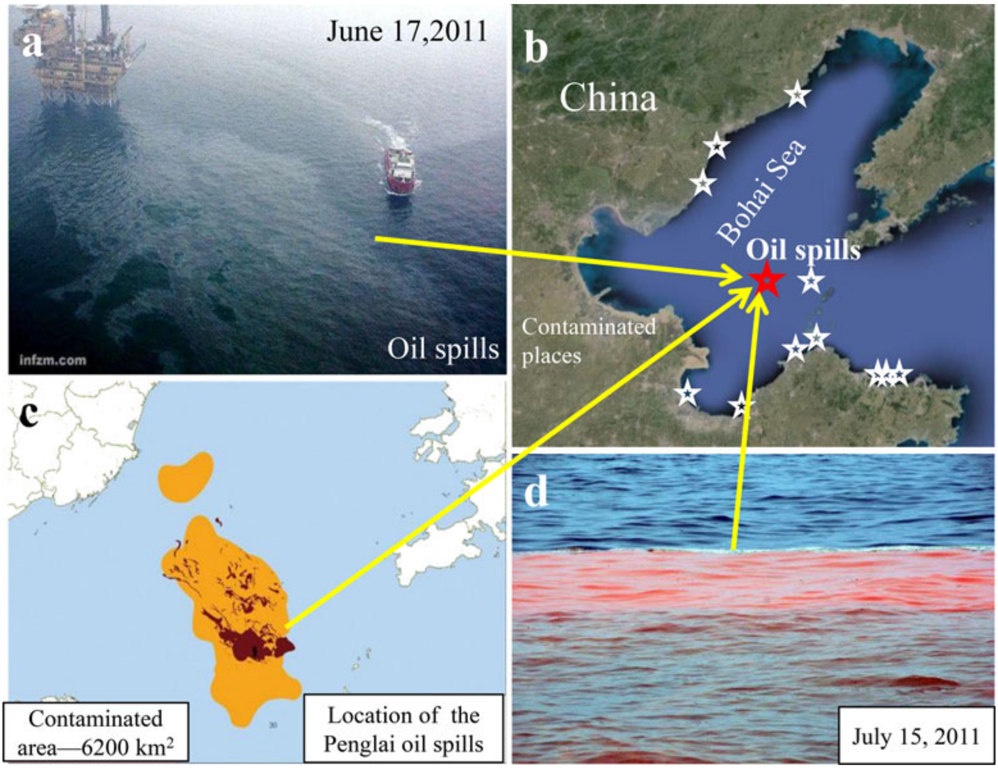

Figure 1. Penglai oil spills in the Bohai Sea. (a) Photos of the oil spills. (b) Affected beaches, cities and counties. (c) Polluted area due to the oil spills (from the State Oceanic Administration of the People's Republic of China). (d) Photo of red tide on July 15, 2011 (from news.sina.com.cn). Red pentagram is the location of the oil spills and white pentagrams are the affected cities or counties.

gov.cn/) and the Ministry of Environmental Protection of the of China (http://www.mep.gov.cn/). The historical data are of high quality as they are using measurements from ships.

2.2.2. Remote sensing data

The temporal and spatial variability of Chl-*a* and SST data is taken from the Moderate Resolution Imaging Spectroradiomerer (MODIS). We have used the Chl-*a* data from MODIS Terra level-3 monthly (4-km resolution), 8 days (4-km resolution) and level-2 daily (1-km resolution) data. The monthly Chl-*a* data for the period during February 2000–January 2013 and 8 days interval Chl-*a* data for the period during 7 April–5 September each year during 2000–2012.

The SST data are obtained from the Ocean Color website (http://oceancolor.gsfc.nasa.gov/) for the period 2 June–5 September each year during the period 2000–2012 (8-day composite data). Both monthly (June 2011–May 2012) and daily (4 June–31 August 2011) level-3 sea surface wind field data with spatial resolution of 25 km are taken from the Advanced Scatterometer (ASCAT) onboard the European MetOp satellite (http://manati.star.nesdis.noaa.gov/). These satellite data are processed using MATLAB code and SeaDAS Version 6.2 developed by NASA.

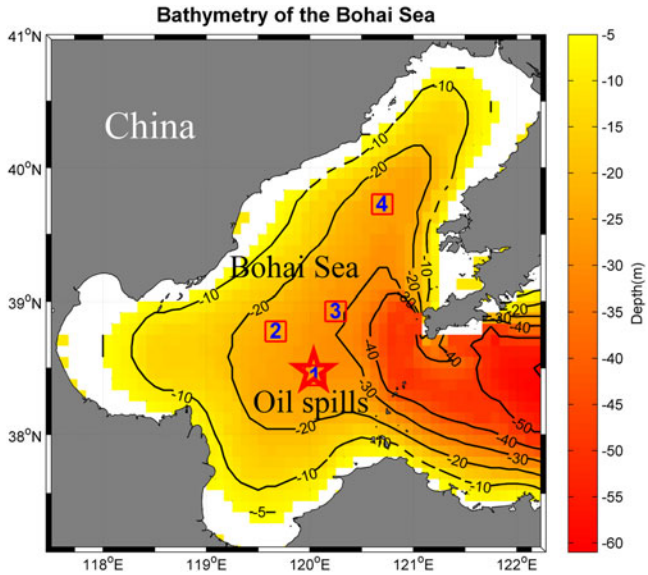

Figure 2. The study area with bathymetry of the Bohai Sea. Total 4 individual areas (4 red squares: Box 1, Box 2, Box 3, and Box 4) are included in the entire study area (region deeper more than 5 m). Red pentagram is the location of the 2011 oil spills, where Box 1 covers the oil spills. Blank region represents for depth shallower than 5 m. Black contours are the isobaths.

2.3. Study methods

2.3.1. Phytoplankton blooms due to oil spills

First, selected single oil spill occurred in (the Penglai oil spills in the Bohai in 2011) and further collected 21 oil spills (listed in Table 1) for further analysis.

The time of oil spills from 1967 to 2018, the time scale was very long. The locations of oil spills in the Pacific, Atlantic, and Indian Ocean, the space was very wild. Thus, the analysis would more comprehensive.

2.3.2. Single oil spills in Bohai - According to the wind and flow data in the Bohai Sea, we identified the Penglai oil spills impacted area. We have considered four sampling locations (Figure 1, Box 1, Box 2, Box 3, and Box 4; Table 2) in and around the areas. The depths of all samplings are less than 30 m; Box 1 is the location of the Penglai oil spills, selecting for analysis sample of oil spills area. Box 2 is in the northwest of Box 1, selecting for analysis sample of near oil spills area, and Box 3 is in the northeast of Box 1 selecting for analysis sample of faraway oil spills area, (Figure 1). Box 4 is also in the northeast of Box 1. Box 4 is far away (about 180 km) from the oil spills, selecting for a comparison analysis. We used satellite products of 8-day-average Chl-*a* concentration date to make 1 month average Chl-*a* data.

2.3.3. 21 oil spills events in the world (Table 1)

Algal blooms events were observed after oil spills, we have carried out detailed analysis to study the occurrence time, season, and interval of phytoplankton blooms as well as temperature conditions.

Table 1. 21 serious oil spills in the world, with Information of location, time, related algal blooms and references.

Number	Name	Start time of oil spills	Duration (days)	Start time of oil algal blooms	Interval between algal bloom and the oil spill† (months)	References
1	Torrey Canyon	1967/3/18	12	–	–	Smith (1968)
2	Argo Merchant	1976/12/15	30	–	–	Atlas (1995), Kerr (1977)
3	Bravo	1977/4/22	9	–	–	Lännergren (1978)
4	Tsesis	1977/10/26	8	–	–	Johansson et al. (1980), Linden et al. (1979)
5	Amoco Cadiz	1978/3/16	15	1978/10	7	Riaux-Gobin (1985)
6	Ixtoc I	1979/6/3	295	1979/9, 1980/1	3 and 7	Jernelöv and Lindén (1981)
7	Castillo De Bellver	1983/8/6	7	–	–	Anderson et al. (1996), Weeks et al. (1991)
8	Exxon Valdez	1989/3/24		1989/11	8	Peterson (2001), Peterson et al. (2003)
9	M/T Haven	1991/4/11	3	–	–	Martinelli et al. (1995)
10	Braer	1993/1/5	7	–	–	Sheng et al. (2011)
11	Sea Empress	1996/2/15	7	–	–	Batten et al. (1998)
12	Jessica	2001/1/16	13	2011/9	8	Zhou et al. (2013)
13	Prestige	2002/11/13	148	2003/4	5	
14	Tasman Spirit	2003/7/27	42	2003/12	5	
15	Lebanon	2006/7/13	3	2007/5	10	Pan et al. (2012)
16	Hebei Spirit	2007/12/7	2	2008/3	4	Zhou et al. (2013)
17	Montara	2009/8/21	74	2010/5	9	Sheng et al. (2011)
18	Deepwater Horizon	2010/4/20	85	2010/8	About 21 days	Graham et al. (2010), Hu et al. (2011)
19	Penglai, this study	2011/6/4	89	2011/6, 2012/6, 2012/8	20 days,12 and 14 months	Zhou et al. (2013), Zhou et al. (2014)
20	Huangdao Explosion	2013/11/22	1	–	–	
21	The East China Sea	2018/1/6	15	–	–	

†Represents lag time when algalbloohe occurred after the oil spills - mean there is no bloom or no correlative data after the oil spills.

Table 2. Satellite data sampling area – the locations of four boxes.

Four sampling boxes	Geographic position	Distance from the location of the oil spills (km)	Area (km^2)
Box1	38.37-38.52°N, 119.95-120.15°E	Location of the oil spills	290
Box2	38.70-39.85°N, 119.57-119.77°E	49.35, northwest	290
Box3	38.85-39.00°N, 120.15-120.35°E	56.12, northeast	289
Box4	39.65-39.80°N, 120.60-120.80°E	152.96, northeast	285

3. Results

3.1. Southerly wind and SST variability after the oil spills

The movement and dispersion of oil spills in the ocean water are controlled by the surface wind, ocean currents and vertical mixing. We have used monthly sea surface wind field data form of ASCAT for the period June 2011–May 2012 to study the

Table 3. Oily sewage discharged and drilling mud discharge from 2004 to 2007.

	oily sewage discharged (ton)	drilling mud discharge (ton)
2004	3,940,000	7,410
2005	8,880,000	25,057
2006	9,940,000	25,690
2007	9,740,000	18,036

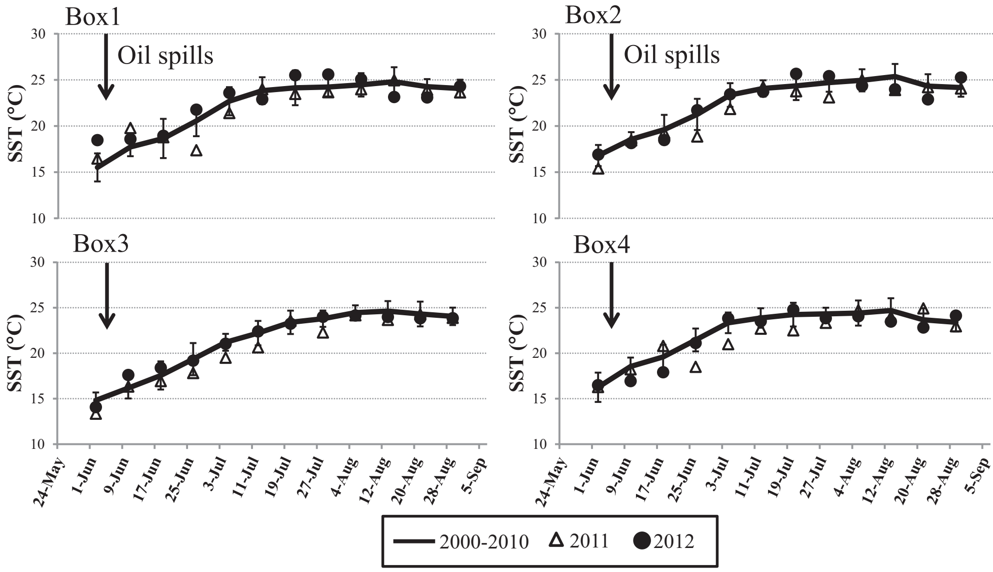

Figure 3. The variability curve of SST from June 2–September 5 (2000–2012). The continuous line is the average value from 2000 to 2010. Black vertical arrow stands for the date of beginning of the oil spills.

extent of dispersion of the oil spills. The wind direction is almost southeasterly during spring (March–May) and summer (June–August), and northerly wind during autumn (September–November) and winter (December–February). Daily wind data shows southward direction during 4 June–31 August 2011. After the oil spills, the wind was mainly southeastward during June to August. It is mainly because of the summer monsoon during this period.

Monthly SST data do not show any pronounced changes in the Bohai Sea during February 2000–January 2013. After the oil spills, a small reduction in SST (about 2 °C) was observed in Box 1 and 4 during 25 June–3 July 2011 (Figure 3), compared to the same periods during 2000–2010, and small changes in SST were observed in Box 2 and 3.

3.2. Variation of monthly Chl-a concentration

Figure 4a shows variations of MODIS-derived monthly Chl-*a* during February 2000–January 2013. Time-average Chl-*a* is shown by the dotted blue curve for the period of 2000–2010 (Figure 4b). A small change in Chl-*a* occurred in the Bohai Sea, with the maximum value (3.37 mg m^{-3}) in the month of March and the lowest value

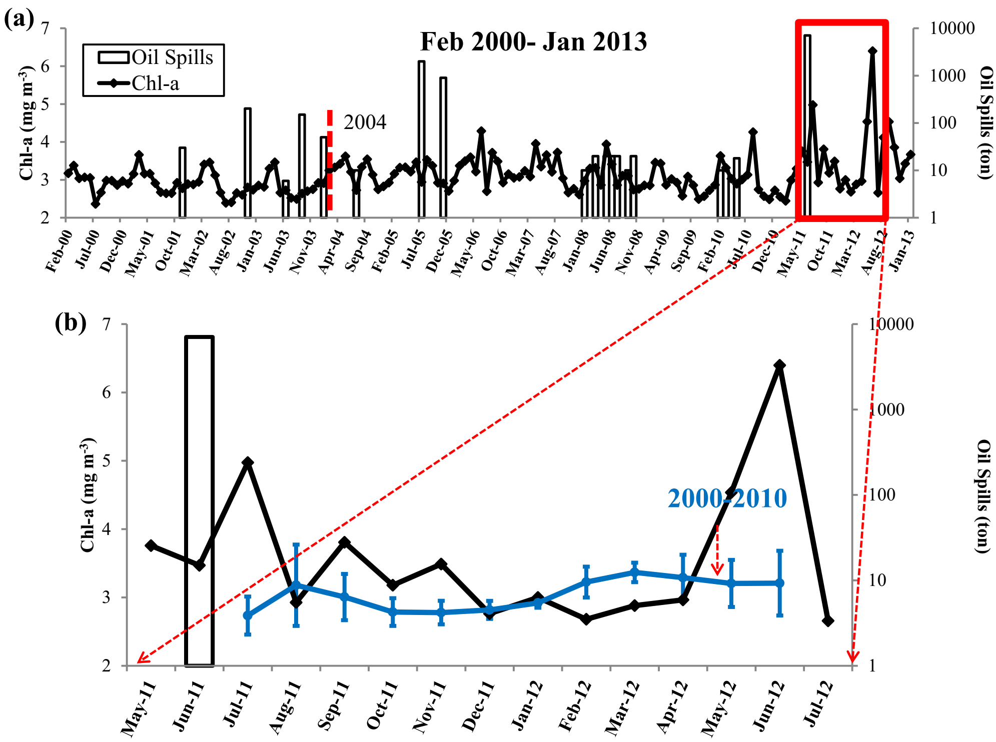

Figure 4. (a) Variability of Chl-*a* with the dates of oil spills marked by a red vertical box. The left x-axis stands for monthly mean Chl-*a* and the right *x*-axis is the amount of oil spills. (b) Blue average curve (with error bars) represents monthly mean Chl-*a* during 2000–2010. Blue short line means standard deviation. Black curve is the Chl-*a* from May 2011 to June 2012.

(2.73 mg m^{-3}) in the month of July. Before 2004, Chl-*a* was almost constant (Figure 4a), which is similar to the time-average value (Figure 4b); after 2004, the Chl-*a* exhibits high variance in its annual maximum and minimum values. Monthly Chl-*a* shows a maximum (4.97 mg m^{-3}) in the month of July 2011 (one month after the Penglai oil spills) (Figure 4b). In June 2012, a pronounced increase in Chl-*a* (6.40 mg m^{-3}) was observed, nearly doubling the time-average value of Chl-*a* (3.21 mg m^{-3}) in the month of June during 2000–2010. In addition, the Chl-*a* concentration in the month of June 2012 was also the highest during February 2000-January 2013.

3.3. Variations of Chl-a (8-day mean) at four data sampling sites

3.3.1. RS detect oil spills in the Penglai 19-3 oilfield area

Envisat Advanced Synthetic Aperture Radar (ASAR) image which shows the location of oil spills was used to detect the oil spills. On 14 June 2011, the Envisat ASAR image clearly shows oil slicks in the northeast, after 10 days of the Penglai oil spills (Figure 6b).

The oil pollution appeared near the Penglai 19-3 Oilfield area on May 27, 2009 (Figure 6c), which was also confirmed by the archived data.

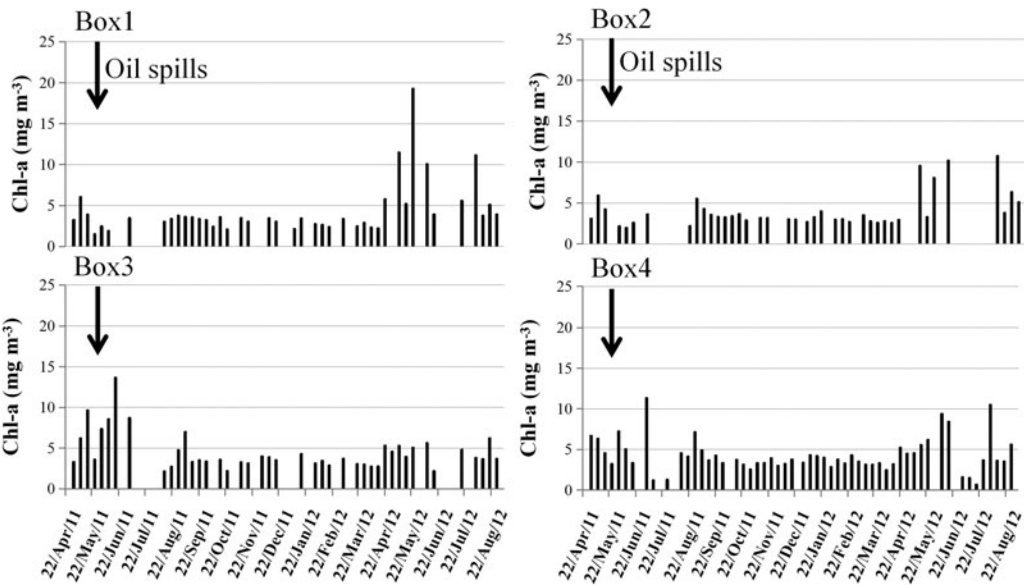

Figure 5. Change of Chl-*a* after Penglai oil spills. Black arrows indicate the beginning date of the oil spills.

3.3.2. The 8-day-averaged Chl-a show an change in four sites

The 8-day-averaged data clearly show small changes in Chl-*a* for a few days after the oil spills in Box 1 and 2. Afterwards, the Chl-*a* concentration reached to a maximum (about 19.25 mg m^{-3} in Box 1, 10.19 mg m^{-3} in Box 2) in late May and early June 2012, specially in Box 1 (Figure 5). Chl-*a* concentration increased (13.66 mg m^{-3}) in Box 3 after about 20 days (Figure 5), and a phytoplankton bloom appeared (MODIS image, Figure 6a) on 24 June 2011 covering 800 km^2 area. This bloom disappeared after 8 days. A small increase in Chl-*a* concentration was observed in Box 4 in early 2011 July (11 mg m^{-3}) and 2012 mid-June (10.5 mg m^{-3}) (Figure 5, Box 4).

The 8-day-averaged Chl-*a* shows an enhancement in Box 1 and 2 after the oil spills during late May and early 2012 June (Figure 7), and similar enhancement was observed in early 2012 August. Large increase in Chl-*a* concentration was observed in Box 3 during late June 2011. In Box 4, small changes in Chl-*a* was clearly observed in early 2011 July (5 mg m^{-3}) and 2012 August (10 mg m^{-3}) (Figure 7), compared to the average value (3.03 mg m^{-3}) of Chl-*a* during 2000–2010.

3.4. The interval between algal bloom and the oil spill

After the Penglai oil spills, three algal bloom events were observed (Table 2). Table 1 lists major oil spill events in the world. Out of the 20 oil spills, 11 algal bloom events were detected after 11 oil spills (Figure 8). The time interval between blooms and oil spills are found in the interval of 3 to 10 months. In some cases the interval was within 1 month, and in some other cases it was more than 12 months.

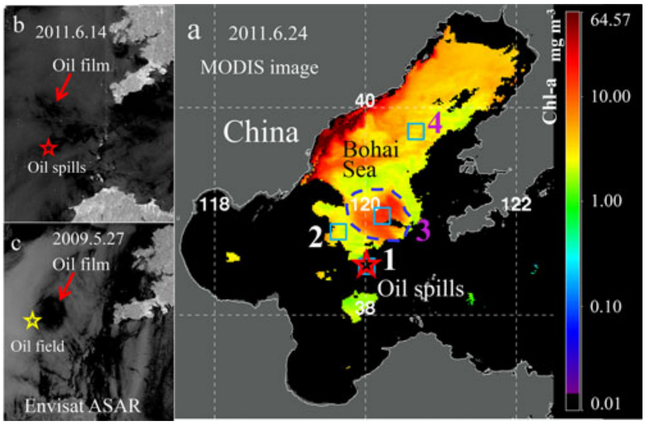

Figure 6. Satellite images. (a) MODIS image of Chl-*a* on June, 24 2011, 20 days after the Penglai oil spills. Black areas are covered by clouds, or missing data. Gray area indicates the land. Thin white curve represents coastline. Numbers 1–4 represent the four boxes. Both red pentagram and "1" indicate the location of the oil spills. The algal bloom is marked by the blue dashed line. (b) Envisat ASAR image of the location of some oil slick on 14 June 2011. (c) Envisat ASAR image on 27 May 2009. Red pentagram is the location of the oil spills and the red arrow points to the location of oil slick. Yellow pentagram is the Penglai 19-3 Oilfield.

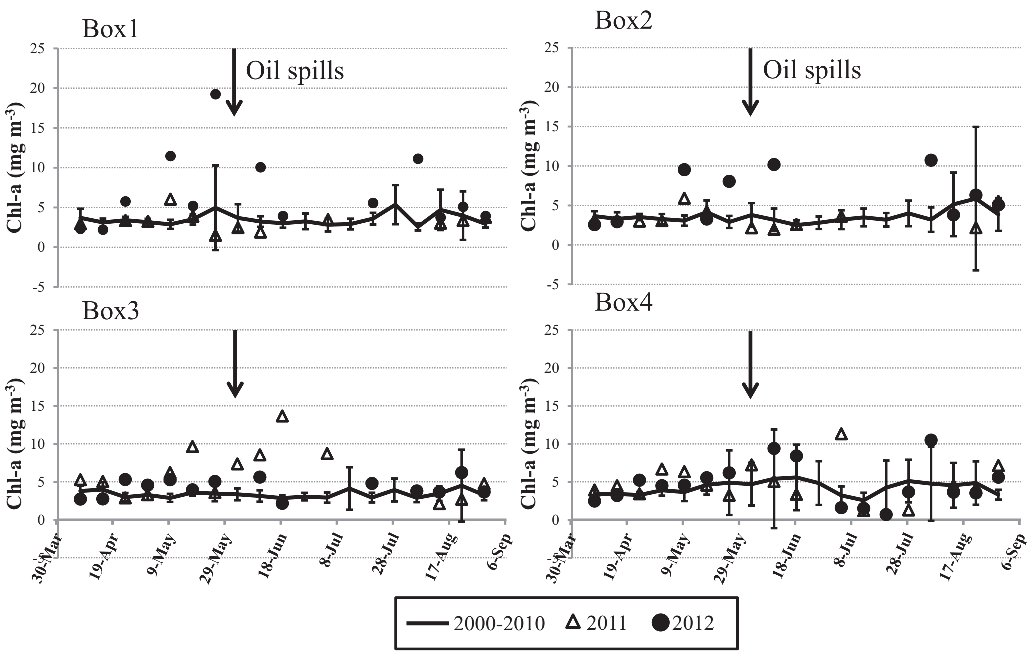

Figure 7. Variation of Chl-*a* from 7 April–5 September 5 (2000–2012 mean, 2011 and 2012). The Black arrow marks the beginning date of the oil spills.

3.5. The temperature after oil spills and occurrence of blooms

After the oil spills, most frequent blooms occurred during March–May, June–August, and September–November. Three out of four blooms occurred during June–August

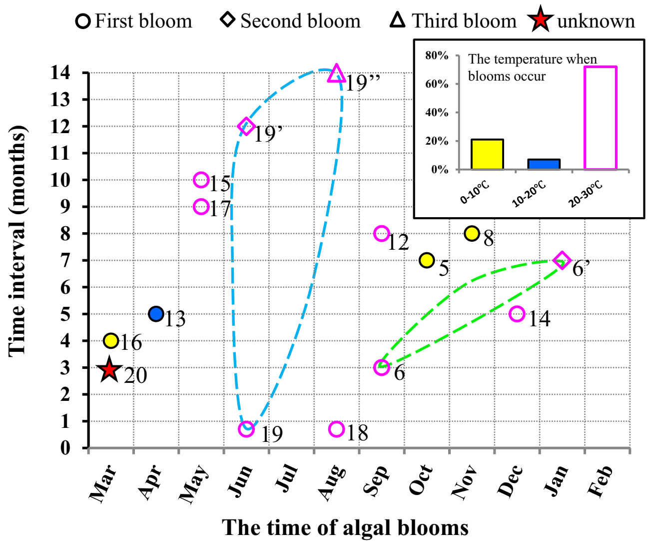

Figure 8. The interval between algal bloom and the 20 oil spills and occurrence time of algal blooms. The dashed blue line covers the blooms after the Penglai oil spills and the green dashed line covers the blooms after the Ixtoc I oil spills.

after the Penglai oil spills. About 72% phytoplankton blooms occurred when temperature was warmer (20–30 °C), except in one case temperature was low (10–20 °C), and three blooms occurred when temperature was less than 10 °C. In Huangdao, the spills occurred on 22 Nov 2013 (Table 1), while the temperature was 9–13 °C. After the oil spills, the monthly average Chl-*a* concentration was 2.51–3.54 mg m^{-3} for the period Nov 2013–Nov 2014.

4. Discussion

4.1 Dispersion of oil spills

After oil spills, oil films formed different thicknesses over the ocean water and the dispersion of oil spills are mainly determined by the surface wind, ocean salinity, oil properties (density and dielectric properties), and SST (Singh 1982; 1984).The oil films undergo a series of physical and chemical weathering processes, such as spreading, dissolution, evaporation, drifting, sinking, emulsification, photo-oxidation, and microbial de-gradation (Medina-Bellver et al. 2005; Ivanov 2011; Joo et al. 2013). Wind and ocean currents are the main factors influencing oil diffusion (Le Henaff et al. 2012).

The surface ocean currents of the Bohai Sea, mainly from the Yellow Sea Warm Current (YSWC), show distinct in different seasons (Zou et al. 2005). The YSWC flows into two branches in the Bohai Sea, one branch enters the Liaodong Bay and forms an anti-clockwise circumfluence with the coastal current, the other branch goes along the Bohai Bay (Zou et al. 2005). Combined the wind direction, which was mostly southeast during the 3 months after the oil spills and the direction of the YSWC, the oil slicks drifted northwest and northeast of the location of the oil spills.

Guo et al. (2013) also observed dispersion of oil in the eastern and north-western parts of the Bohai Sea at different times from satellite (Guo et al. 2013). Based on the contaminated area (Figure 2c), wind direction, currents, satellite data (Guo, Liu and Xie 2013) and the oil films (Figure 6b), we selected four boxes (Figure 1b): Box 1 for oil spills area; Box 2 and Box 3 for contaminated area; Box 4 for uncontaminated area.

4.2. Changes in SST

A small reduction in SST appeared in Box 1 and 4, and no change in SST was found in Box 2 and 3 (Figure 3). Since Box 1 is the location of the oil spills, while Box 4 is not polluted, the small reduction at these two sites clearly show that oil spills has no impact on SST. The growth of phytoplankton depends on various factors, for example, nutrients, light, SST, and wind (Tang et al. 2003; 2004a;Wang et al. 2010). Changes in SST was not found in enhancing Chl-*a* concentration, therefore, oil spills have no notable impact on SST.

4.3. Phytoplankton blooms and the oil spills

The Chl-*a* concentration before 2004 was found to be close to the mean value, while large changes in Chl-*a* concentration was detected afterwards (Figure 4a). The amount and growth rate of oily wastewater, mud and cuttings in the Bohai sea have increased every year after 2004 (marine bulletin from SOA, http://www.soa.gov.cn/zwgk/hygb/zghyhjzlgb/). The increase in oil pollution (circular shape) near the Penglai 19-3 Oilfield in the year 2009 could be due to seepage from oil platforms. Therefore, the big change in Chl-*a* concentration after 2004 is likely due to the considerable increase in oil pollution. There are two reasons for speculation which are domestic sewage and petroleum exploitation (Table 3), according China Marine Environmental Quality Bulletin. (State Oceanic Administration, 2004, 2005, 2006, 2007).

After the Penglai oil spills in the month of June 2011, changes in Chl-*a* concentration in the Bohai Sea are found about 1 month after the oil spills. Petroleum hydrocarbons influence the growth and reproduction of plankton, and zooplankton was found to be more sensitive as compared to phytoplankton to the oil. The quantity of zooplankton would greatly decrease after the oil spills. As zooplankton are mainly predators for phytoplankton, therefore, the Chl-*a* concentration could be increase in July 2011. Pronounced increase in the Chl-*a* concentration was found in June 2012, with the highest concentration (6.40 kg m^{-3}) during February 2000–January 2013. Some ingredients of oil could persist for a long time since it is difficult for them to evaporate, such as heavy petroleum hydrocarbons (Oudot and Chaillan 2010; Natter et al. 2012). The oil spills could be one of the major contributors to the changes in Chl-*a* concentration in 2012.

Box 1, 2, and 3 are contaminated locations. Box 1 is the location of highly contaminated area due to the oil spills. Box 2 is in the main path of oil movement by wind and the YSWC, belong to highly polluted area, and Box 3 is part of the contaminated area. According to the satellite data (Figures 5 and 7), the variability of Chl-*a* concentration enhanced over the historical Chl-*a* concentration after the Penglai oil spills, and is very likely that the increase of the Chl-*a* is related with the oil spills, instead of other environmental factors.

About 1 month after the oil spills, the Chl-*a* concentration in Box 1 and Box 2 show small changes, while Box 3 shows a largest bloom (Figures 5–7). In a short period after the oil spills, most marine ecology could be influenced because of the toxicity of oil, depending on the type and concentrations of the oil spills. Harrison et al. (1986) found that low concentrations (<100 mg/l) of polycyclic aromatic hydrocarbon (PAHs) enhance phytoplankton growth and higher concentrations (>100 mg/l) of PAHs inhibit the growth of phytoplankton; complicated hydrocarbon or high-molecular-weight would also inhibit the growth and photosynthesis of algae (Vargo et al. 1982; Harrison et al. 1986; Yang et al. 2009). At the same time, as zooplankton are more sensitive to oil spills due to the lack of oxygen in the water surface, quantity of zooplankton greatly decreased after oil spills (Fefilova 2011). The Chl-*a* bloom in Box 3 might be related to the decrease of predators and stimulation of oil on phytoplankton (Dunstan et al. 1975; Parsons et al. 1976).

About 12 months after the Penglai oil spills, the Chl-*a* in Box 1 and 2 increased and a phytoplankton bloom was observed (Figures 5 and 7). Our earlier results also show pronounced algal blooms several months after the oil spills (Sheng et al. 2011; Pan et al. 2012; Zhou et al. 2013), and some of the studies have shown that the algal blooms, occurred in a long time after oil spills, that could be associated with the oil spills (Jernelöv and Lindén 1981; Riaux-Gobin 1985; Peterson et al. 2003). After two months, another bloom appeared in Box 1 and 2 (Figure 7), and the intensity of the second bloom was larger compared to the first one. Microorganism could degrade petroleum hydrocarbons (Tazaki et al. 2006; Almeida et al. 2013; Ribeiro et al. 2013), and the decomposition products could enhance the phytoplankton growth, which might lead to algal blooms (the first bloom-Box 1 and Box 2). Since the start time of degradation and the ability to degrade are different, depending on microorganism and environment (Atlas 1995; Van Hamme et al. 2003), the times of phytoplankton blooms are also found to be different (Atlas 1995).The appearance of the second bloom might be linked to bio degradation from sunken oil, which was possibly re-suspended by some ocean dynamic processes (Moonkoo et al. 2013). Also, sea ice, a unique feature in the Bohai Sea, was likely to affect biodegradation, which could delay the process of degradation. The characteristics of Chl-*a* and phytoplankton blooms in Box 1 and 2 are found to be similar (Figures 5 and 7).

The area shown in Box 4, near the Liaodong Bay, about 180 km away from the oil spills, may not be influenced by the oil spills and instead was more likely affected by coastal human activities. In this area, strong variations in Chl-*a* is observed at the time of red tide every year (Lin et al. 2008), which is possibly connected to the red tides in the Bohai Sea. Specifically, a large area (3452 km^2) near Box 4 was affected by a red tide event during 8 June–20 August. Therefore, the change in Chl-*a* concentration in Box 4 was mainly due to strong red tides.

Comparisons of algal blooms occurred during three-time intervals at three locations (Table 4). The first bloom was the largest, which occurred in Box 3 and the

Table 4. The three phytoplanktonblooms associated with the Penglai oil spills in Bohai Sea in the year 2011.

Blooms after oil spills	The location of Blooms	Chl-*a* concentration (mg m^{-3})	Average Chl-*a* in the same time from 2000 to 2010	Interval between algal bloom and the oil spill		Influence factors	Driving factors
First bloom	Box 3	13.66	2.86	About 20 days	Short term	Quantity of oil and thickness of oil films	Self-diffusion or currents
Second bloom	Box 1	19.25	4.96	12 months	Long term	Biodegradation	Wind, currents
	Box 2	10.18	3.18				
Third bloom	Box 1	11.13	2.57	14 months		Biodegradation, oil re-suspension, sea ice	Wind, currents
	Box 2	10.73	3.20				

time interval of which was about 20 days after the oil spills. The second bloom appeared in Box 1 and 2, and its time interval was 12 months. Enhancement in Chl-*a* concentration (in the second bloom) was very small in Box 1 and 2. After about 2 months following the second bloom, the third bloom occurred. The favorable conditions for the third bloom are the quantity and thickness of oil films, biodegradation, re-suspension of oil, and sea ice (Table 4). The possible driving factors behind the blooms, could be self-diffusion, wind and water currents (Table 4). It is observed that the blooms occur after a long-time interval of the oil spills, and the locations of those blooms might be far away from the original location depending on dispersion of oil spills, which were largely determined by the ocean currents and meteorological parameters during that period.

4.4. Time intervals and seasons

The Bohai Sea is a semi-enclosed sea that restricts disappearance of the oil spills. The oil spills can persist for long time at the same location. This may cause several blooms.

Among all the blooms after the oil spills, the time intervals are different and most blooms appear after 3–10 months. Soon after the oil spills, the extent of affected regions depends on the wind and ocean temperature. Zooplankton was more sensitive to the lack of oxygen, it is one of the reasons that the blooms occurred about 1 month after the oil spills. After several months, the production of biodegradation of microorganisms favored phytoplankton and blooms. The time of degradation by different microbial varies in different seas and the degradation process takes several months (Atlas 1995). The other marine driving forces (e.g., currents or/and the process of freezing/thawing of sea ice) may delay the speed of degradation and put off the occurrence of blooms, extending the time intervals to be more than 12 months. In winter, as the oil was frozen in the ice, the oil was prevented from moving, sinking, spreading, and so decomposing. Only until spring when the temperature rose and the ice thawed, the oil then started moving again. In such way, the oil degradation was delayed.

In the northern hemisphere, where spring (March–May) and autumn (September–November) are opposite to the southern hemisphere, most of the blooms occurred after the oil spills during spring and autumn (Figure 8; Diaz and Rosenberg

2008). This was due to favorable conditions of warmer SST (about 20–30 °C; Robarts and Zohary 1987).

Based on our past research results (Sheng et al. 2011; Pan et al. 2012; Zhou et al. 2013), it is clear that oil spills influence the growth of plankton that could result in algal blooms. Meanwhile, the time interval, between oil spills and algal blooms, could be more than 10 months. The blooms after oil spills are related to growing seasons of phytoplankton and warmer SST (Zhou et al. 2013). Based on detailed data analysis, we have discussed possible processes and mechanisms (Figure 9) associated with the short- and long-term impacts on marine ecosystem due to oil spills and the factors for algal blooms. The probable causes could include: the stimulation of oil on phytoplankton, toxicity on zooplankton, biodegradation of microorganism, re-suspension of oil by wind and ocean surface currents, and the delayed biodegradation by sea ice, as well as the temperature of oil spills and growing seasons of phytoplankton (Figure 9). The present study shows impacts of oil spills on marine ecosystems in the Bohai Sea, Three blooms occurred successively with variances in occurrence time, severity and location, depending on many factors. A concept model shows impacts on marine ecological environment (Figure 9) in short and long periods after the oil spills.

The nutriment released from the oil decomposition was absorbed by phytoplankton and then boosted the growth of phytoplankton. The time of oil decomposition could last for a long time. Besides, there were the indirect impact of the post-oil-spill blooms, which was that the oil killed zooplankton and marine animal. Muhammad and Ahmad (2017) studied the impact of pH from 8.5 to 6.5 on toxicity of crude oil WAF to white-leg shrimps, *L. vannamei*. Muhammad and Ahmad (2017) found that the pH of 6.5 increased the toxicity of crude oil water accommodated fraction (WAF). Declining predators also contributed to the phytoplankton boom. Therefore bloom occurred a while after oil spills (Figure 5).

4.5 The temperature of the blooms occurred

During high temperature conditions in summer, the blooms are common, 14 phytoplankton blooms were observed after 21 oil spills. Ten (about 72%) blooms occurred when temperature was warmer (20–30 °C). The probability of post-oil-spill blooms was low when temperature was cooler, with only one bloom occurrence in the temperature of 10–20 °C, and other three blooms occurring when temperature was less than 0–10 °C. Blooms were not observed (Figure 6c) in May 2009 when the average temperature was 16 °C (<20 °C). We also have observed oil spills on the 6 January 2018, caused by the collision of oil tankers (Table 1) in the East China Sea, blooms did not occur because of low temperature (about 0 °C).

4.6. Biological actions play important role

Some of marine environmental factors, like water temperature, salinity, nitrogen and phosporus (Table 3), could affect the content of Chl-*a*. In the present study, oil is organic that may decompose to carbon and hydrogen, but cannot decompose to salinity, nitrogen and phosphorus. During oil spills, oil reduced the number of predator of phytoplankton,

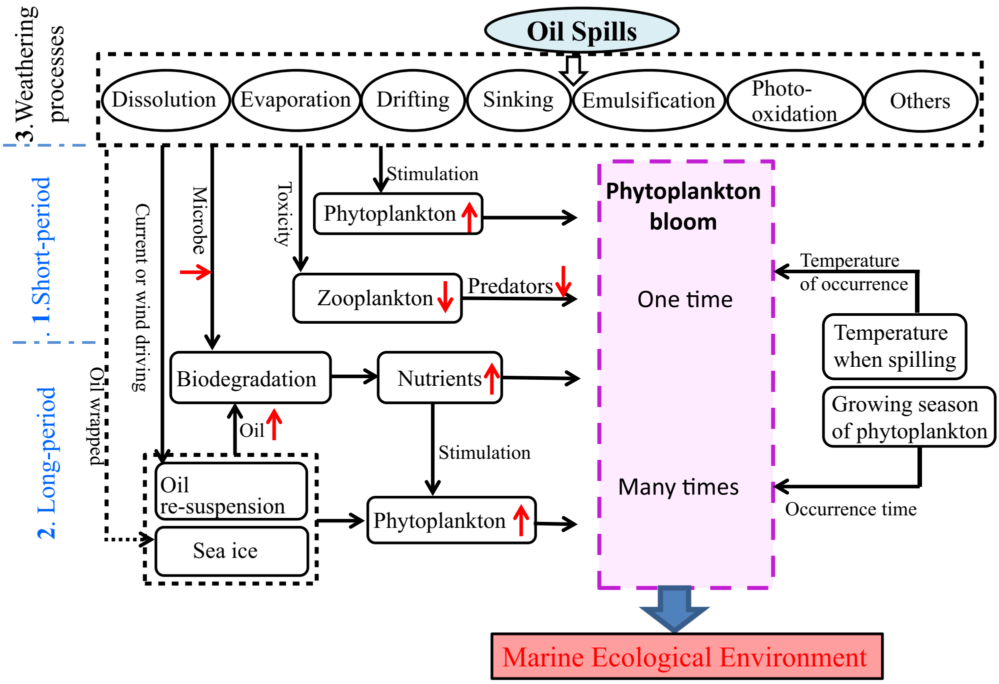

Figure 9. The concept model showing both short and long period impact of oil spills on marina ecological environment. Solid black arrows indicate relations between the two sides, and dashed black arrows suggest the potential relations.

and when oil spill decomposition at last, the ingredient of decomposition can cultivate phytoplankton, biological action more adaptable to high temperature (>20 °C), so phytoplankton blooms occurred in high temperature (>20 °C) condition after oil spills. It clearly shows that the biological actions play an important role in oil spill cases.

5. Summary

The short-term impact of oil spills on ecosystem could mainly depend on the quantity and composition of oil spills, the thickness of oil slicks, and the toxicity of hydrocarbons on zooplankton (1 in Figure 9).

The long-term impact could be the biodegradation of indigenous microorganisms. Many other parameters could also affect the ecosystem, including the degree of surface wind, the strength of ocean currents, and the extent of oil spills frozen in the sea ice (2 in Figure 9).

The time interval between the accident of oil spills and the occurrence of algal blooms varies. Phytoplankton blooms generally occur during the growth season of phytoplankton or/and under warmer weather conditions.

The extent of regions affected by the oil spills can be mainly subjected to the direction of ocean wind, the strength of ocean currents and the degree of temperature. More detailed short- and long-term data of various parameters are required to understand better the speed of dispersion of oil spills, the pace of chlorophyll blooms and their impacts on the marine ecology (3 in Figure 9).

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