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Seasonal Hypoxia of Amursky Bay in the Japan Sea: Formation and Destruction

Pavel Ya. Tishchenko[1,*], Vyacheslav B. Lobanov[1], Vladimir I. Zvalinsky[1], Alexander F. Sergeev[1], Alexey Koltunov[2], Tatyana A. Mikhailik[1], Petr P. Tishchenko[1], Mariya G. Shvetsova[1], Sergey Sagalaev[1], and Tamara Volkova[1]

[1] V.I. Il'ichev Pacific Oceanological Institute, Far Eastern Branch, Russian Academy of Sciences, Vladivostok, Russia
[2] A.V. Zhirmunskiy Institute of Marine Biology, Far Eastern Branch, Russian Academy of Sciences, Vladivostok, Russia

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ABSTRACT

Based on detailed hydrological and hydrochemical surveys carried out in each of the four seasons of 2008, Amursky Bay in the north west quadrant of the Japan Sea was found to experience seasonal hypoxia. The primary process of hypoxia formation is a microbiological degradation of the "excess" amount of diatoms under rather low photosynthetic active radiation in bottom layer and weak water dynamics. The microbiological decay of dead diatoms under light deficient conditions intensively consumes dissolved oxygen and produces phosphates, ammonium, silicates, and dissolved inorganic carbon. Existence of a phytoplankton "excess" is caused by phytoplankton bloom resulting from nutrient pulses into Amursky Bay. There are two main sources of these nutrients: the waste waters of Vladivostok city and discharge from Razdolnaya River. The river delivers more than two times the amount of nutrients than the waste waters of Vladivostok. It is suggested that the phytoplankton "excess" might be caused by an enhanced supply of nutrients delivered into the surface layer resulting from the increased discharge of the river on a short time scale. Our data suggest that hypoxia is seasonal, with a peak at the end of summer. The upwelling of the Japan Sea water in the beginning of the fall season and its advection across the shelf is the primary process by which the hypoxia is destroyed. During the winter, strong vertical mixing due to termohaline convection makes the water column uniform and brings more oxygen into the water along with high primary production under the ice. Thus, during the winter season, the ecosystem of Amursky Bay recovers completely.

Key words: Hypoxia, Eutrophication, Hydrochemistry of estuary, Upwelling, Amursky Bay, Peter the Great Bay, Japan Sea

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1. INTRODUCTION

Dissolved oxygen concentrations (DO) in coastal waters have changed drastically over past decades leading to widespread hypoxia (Diaz 2001). The number of coastal sites where hypoxia has been reported has increased exponentially, therefore the hypoxia phenomena is now recognized as a global problem (Diaz 2001; Diaz and Rosenberg 2008; Breitburg et al. 2009; Zhang et al. 2010; Steckbauer et al. 2011). Hypoxia in coastal areas is governed by physical and biogeochemical processes. Some of the potential causes of hypoxia in the coastal area include enhanced input of nutrients and organic matter in areas with limited water exchanges, and human activity such as increasing usage of fertilizers, burning of fossil fuels, and waste water production that exports excess nutrients to coastal aquatic areas which leads to eutrophication (Nixon 2009; Rabalais et al. 2010). Excess nutrients provide more organic matter via photosynthesis, and then the excess organic matter is diverted to the lower trophic level, where microbes and microbial respiration create hypoxic conditions.

In Asian coastal waters, hypoxia has been reported in China, Japan, Korea and Russia (Lim et al. 2006; Tishchenko et al. 2008; Haraguchi et al. 2010; Ning et al. 2011). In Russia, a seasonal hypoxia in Amursky Bay (located in the northwestern Japan Sea) has been observed in recent years

* *Corresponding author*
E-mail: tpavel@poi.dvo.ru

(Tishchenko et al. 2008). This case is distinctly different from others in Asia because it is occurring in a sub-arctic climatic zone. Amursky Bay is located in the southern part of the Primorye Region of Russia (Fig. 1) and is characterized by a monsoon climate (e.g., Luchin 2005). The winter season is cold and dry and the northern part of the bay is covered with solid ice from the middle of December through March. Drifting ice is observed in the southern part of the bay. Summer is mild and wet with heavy rains often occurring at the end of a season resulting in an abrupt increase of river discharge.

Amursky Bay is a part of the Peter the Great Bay, located in the northwest Japan Sea. Its width is around 10 - 22 km, and it has an average depth of about 15 m. The Razdolnaya River flows into the northern part of the bay. This river originates in China and has an average annual discharge of 72 $m^3\ s^{-1}$. One of the largest cities of the Russian Far Eastern region, Vladivostok, is located on the eastern coast of the bay (Fig. 1). The first time a severe hypoxia in the bottom waters of Amursky Bay was observed in August 2007, when the lowest oxygen concentration was about 5 µM, (Tishchenko et al. 2008). After this finding, a seasonal investigation on hydrology and hydrochemistry was organized to study the formation and evolution of hypoxia in the bay. In this paper, we present our observations for seasonal variability of surface and bottom water parameters of Amursky Bay recorded over four successive seasons and discuss mechanisms of hypoxia formation and destruction.

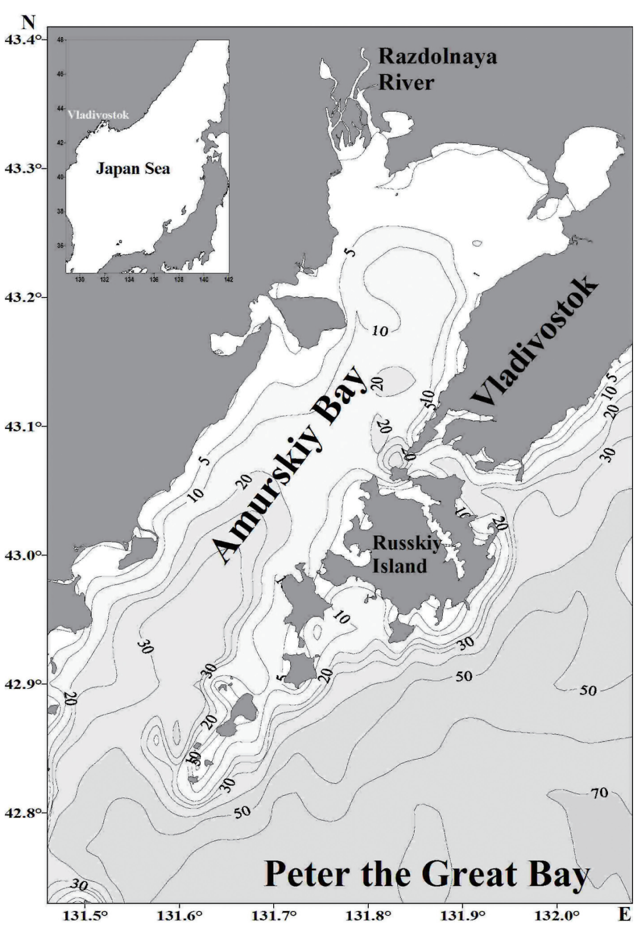

Fig.1. Amursky Bay is an upper part of Peter the Great Bay located in the northwestern Japan Sea. The city of Vladivostok with a population around 0.6 million located on its coast and Razdolnaya River entering from the north are two major sources of nutrients.

2. METHODS

Four oceanographic and hydrochemical surveys were carried out in each of the four seasons of 2008. During winter (February 19 - March 4), 54 stations were utilized. At this time, the northern part of the bay was covered by ice up to a thickness of 60 cm. The water samples were taken by drilling the ice and using plastic bottles. In the spring (May 21 - 27), summer (August 21 - 31), and autumn (October 17 - 26), we established 103, 140, and 84 stations respectively. In warmer periods, surveys were performed by the R/V Malakhit, and water samples were taken from surface and bottom layers using a 5 liter Niskin bottle. Typically, the bottom layer bottle is lowered to a depth of 1.0 to 1.5 meters above the floor sampling "bottom water" and the surface bottle is 1.0 to 1.5 meters below the surface sampling "surface water". Dissolved oxygen (DO) in the samples was "fixed" in oxygen flasks on board just after sampling by the addition of pickling reagents. Samples for pH and total alkalinity (TA) were poisoned by mercury chloride. All samples were kept in a dark cool room and delivered to a coastal laboratory. Analysis for DO concentration, pH, nutrients, and chlorophyll *a* filtration were carried out the same day. Analysis on TA, humic substances, and salinity were performed the next day. In warm periods, the transparency of the water column was determined by a Secchi disk at every station. CTD profiles were obtained at every station from the surface down to the bottom by a RBR XR 620 profiler using pressure, temperature, conductivity, chlorophyll fluorescence, and turbidity sensors for three seasons, winter, spring, and summer. In autumn, the CTD observations were carried out by SBE-19 plus profiler which had no fluorescence and turbidity sensors.

PH measurements were done by means of cell without liquid junction, using glass electrodes for hydrogen and sodium ions:

$$\text{glass-electrode-}Na^+ \left| \begin{array}{c}\text{test solution (ref-}\\ \text{erence solution)}\end{array} \right| H^+\text{-glass-electrode}$$

The buffer solution TRIS·HCl-TRIS-NaCl-H_2O (m_{NaCl} = 0.4; m_{TRIS} = $m_{TRISHCl}$ = 0.04) was used as a reference solution on the Pitzer pH scale. Then, obtained values were converted into a "total hydrogen concentration scale - pH_T". Measurements of pH were carried out at 5, 15, 20, and 15°C in winter, spring, summer, and autumn seasons, respectively. A detailed description of this method was published elsewhere

(Tishchenko et al. 2011). Comparison of this method with the commonly adopted spectrophotometric method was examined (Kang et al. 2011). Accuracy of the method is about ±0.004.

Analysis of TA was carried out by a direct colorimetric titration with hydrochloric acid in an open system using a mixed indicator (methylene blue and methyl red). The titration was carried out under a CO_2 free condition via the flow of CO_2-free air (or nitrogen). The change of the sample color from green to light-pink at the equivalence point was detected visually. The pH at the end point was about 5.4 - 5.5. This method is known as Bruevich's method (Bruevich 1944) and is described in English (Talley et al. 2004; Kang et al. 2011). Accuracy of the method is about ±0.15%. Humic substances, which are the main contributors of organic alkalinity, were determined by the spectrophotometric method; the organic alkalinity was calculated from knowledge of the humic substances concentration (Tishchenko et al. 2006). Then, the organic alkalinity was subtracted from the TA; the obtained corrected alkalinities (TA_{corr}) and pH_T were used for calculating $pH_{in\ situ}$, CO_2 partial pressure (pCO_2) and dissolved inorganic carbon (DIC), using a commonly known procedure (Dickson et al. 2007).

Dissolved oxygen samples were drawn from the Niskin bottle. They were analyzed on an automated oxygen titrator using Brinkman Dosimat automatic burette and photometric end-point detection. Details of the Winkler titration method, providing accuracy of the measurements of 0.5 to 1%, are described in Talley et al. (2004).

Phosphorous concentrations were determined photometrically as a phosphormolybdic acid complex, the absorbance of which was measured at 885 nm wave length. Silicate concentrations were determined photometrically as reduced by a silicomolybdate acid complex; the absorbance was measured at 810 nm wave length. Nitrite ions originally present in the samples, along with those produced by the near-quantitative reduction of nitrate by the packed bed cadmium reactor, are determined photometrically by diazotization with sulfanilamide and subsequent coupling with N-(1-naphthyl) ethylenediamine to form an intensely-colored azo dye (the Greiss reaction). Absorbance was measured at 540 nm wave length. Details of the methods used for nutrient analysis are given (Strikland and Parsons 1972). Ammonium concentrations were determined by the indophenol method. Details of this method are given by Grasshoff and Almgreen (1976).

The daily fluxes of nutrients supplied into Amursky Bay by the Razdolnaya River were calculated by multiplying the concentration of nutrients by values of water discharged from the river. These fluxes were obtained twice for each season and were approximated as a function of river discharge using the empirical equation of the second order. Using daily discharge data and empirical equations, the nutrient fluxes for each day of 2008 were estimated.

Thresholds of hypoxia are proposed in the literature to range broadly from 0.28 to 4 mg-O_2 l^{-1}, while most reports (55%) refer to a value of 2 mg-O_2 l^{-1} (Vaquer-Sunyer and Duarte 2008). There is a broad variability in the thresholds of oxygen concentrations for hypoxia among benthic organisms (Vaquer-Sunyer and Duarte 2008; Levin et al. 2009; Steckbauer et al. 2011). Therefore, there is no single definition of hypoxia which fits all organisms. Hypoxia is often defined as a content of DO concentration below 2 mg-O_2 l^{-1} (63 μM) O2 or 2 ml-O2 l^{-1} (89 μM) (Breitburg et al. 2009). The average value (76 μM) of these noted DO concentrations corresponds with the median lethal oxygen concentration for half of the tested species by Vaquer-Sunyer and Duarte (2008). This oxygen concentration was used as a threshold value for the assessment of the eutrophication status of Peter the Great Bay (NOWPAP CEARAC 2011) and will be accepted as a definition of hypoxia here.

3. RESULTS

Distribution of hydrological and hydrochemical parameters of surface and bottom waters in the Amursky Bay for the winter, spring, summer, and autumn of 2008 is presented at Figs. 2 - 7. In winter, both surface and bottom water temperatures are negative (-1.8 ~ -1.0°C) because of strong thermohaline convection caused by intense cooling and brine rejection during the ice formation process. Radiative heating, river inflow, and wind mixing increase water temperatures of the whole water column of the bay in the spring and summer seasons. It rises up to 11 - 14°C at the surface and to 5 - 8°C at the bottom layer by May, and reaches its maximum in August around 21°C at the surface and 15 - 21°C at the bottom (Fig. 2). The decrease in temperature is observed in autumn. In October 2008, the water temperature was around 13°C at the surface and 3 - 11°C at the bottom. It is necessary to note that from August to October we observed a gradual decrease of water temperature in the bottom layer, while not so much at the surface. This suggests that such an abrupt decrease of bottom water temperature had not been caused by atmospheric cooling but is a result of upwelling and cross shelf advection of open sea water. This is usually what happens along the northwestern coast of the Japan Sea in the period when the monsoon winds change in September and October. The distribution of salinity in the bay also shows pronounced seasonal variability (Fig. 3). In winter, salinity has a maximum for both surface and bottom layers, and reaches 34.5 - 35 or more. Such high salinity is caused by two factors: a) ice formation and brine rejection; and, b) low discharge of Razdolnaya River during this season (1 - 2 m^3 s^{-1}). Minimal salinity in the bay corresponds to summertime because of the maximum precipitation and correspondingly maximal Razdolnaya River discharge (Fig. 8). In this case, salinity of the bottom waters ranges between 28 to 33. During spring and autumn seasons, salinity values of bottom waters are intermediate,

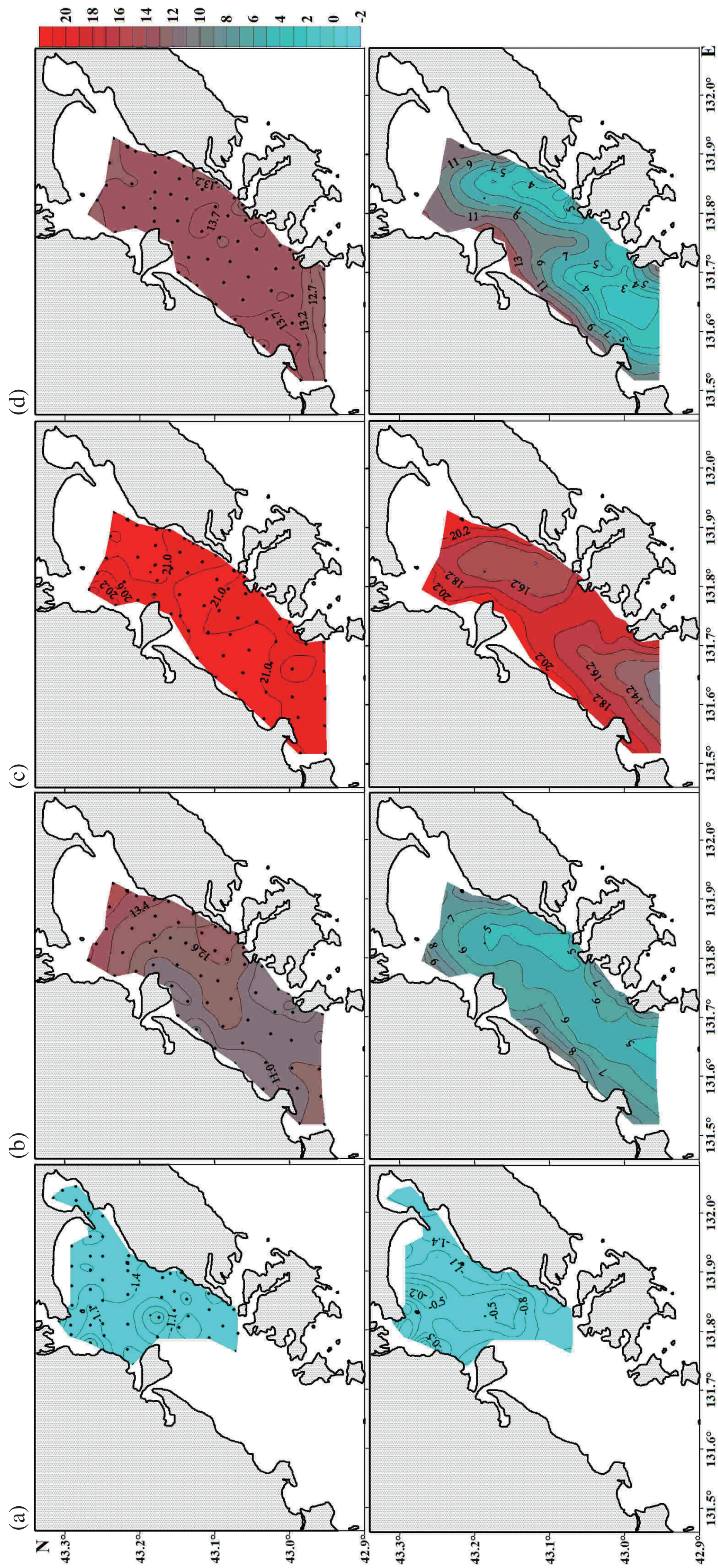

Fig. 2. Seasonal distributions of temperature (°C) in the surface waters (top panel) and bottom waters (bottom panel) of Amursky Bay in 2008. (a) February, (b) May, (c) August, (d) October.

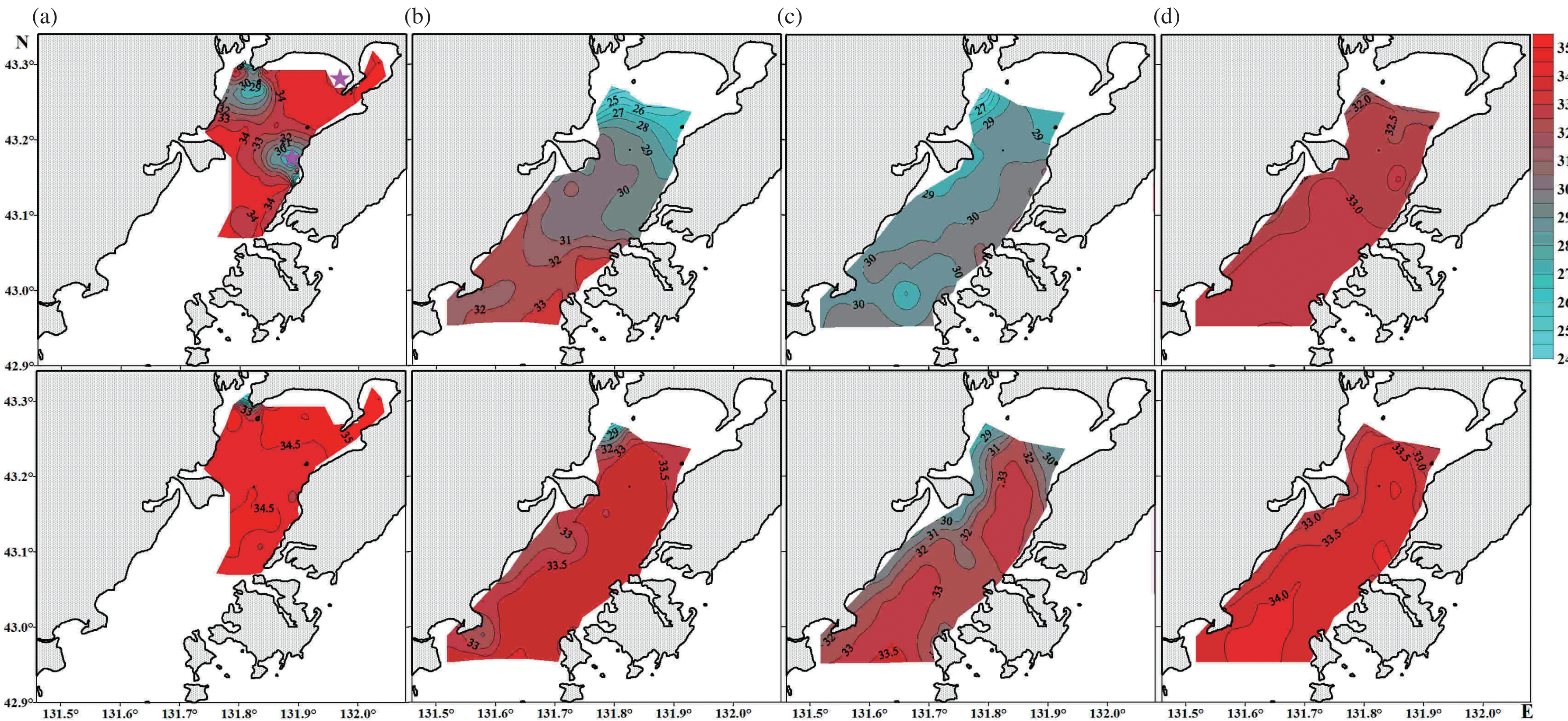

Fig. 3. Seasonal distributions of salinity in the surface waters (top panel) and bottom waters (bottom panel) of Amursky Bay in 2008. (a) February, (b) May, (c) August, (d) October. Stars indicate locations of major waste water inputs into the bay by Vladivostok city.

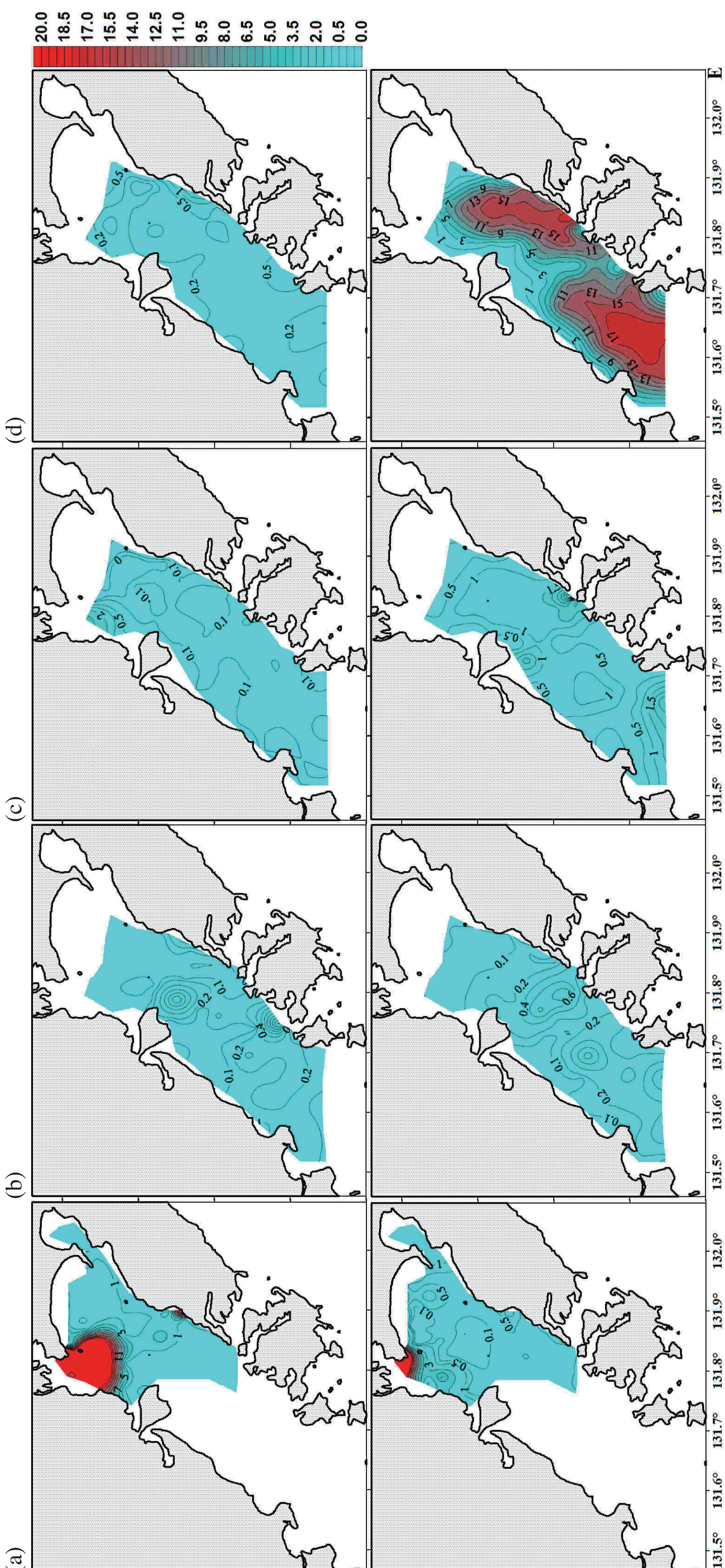

Fig. 4. Seasonal distributions of nitrate concentrations (μM) in the surface waters (top panel) and bottom waters (bottom panel) of Amursky Bay in 2008. (a) February, (b) May, (c) August, (d) October.

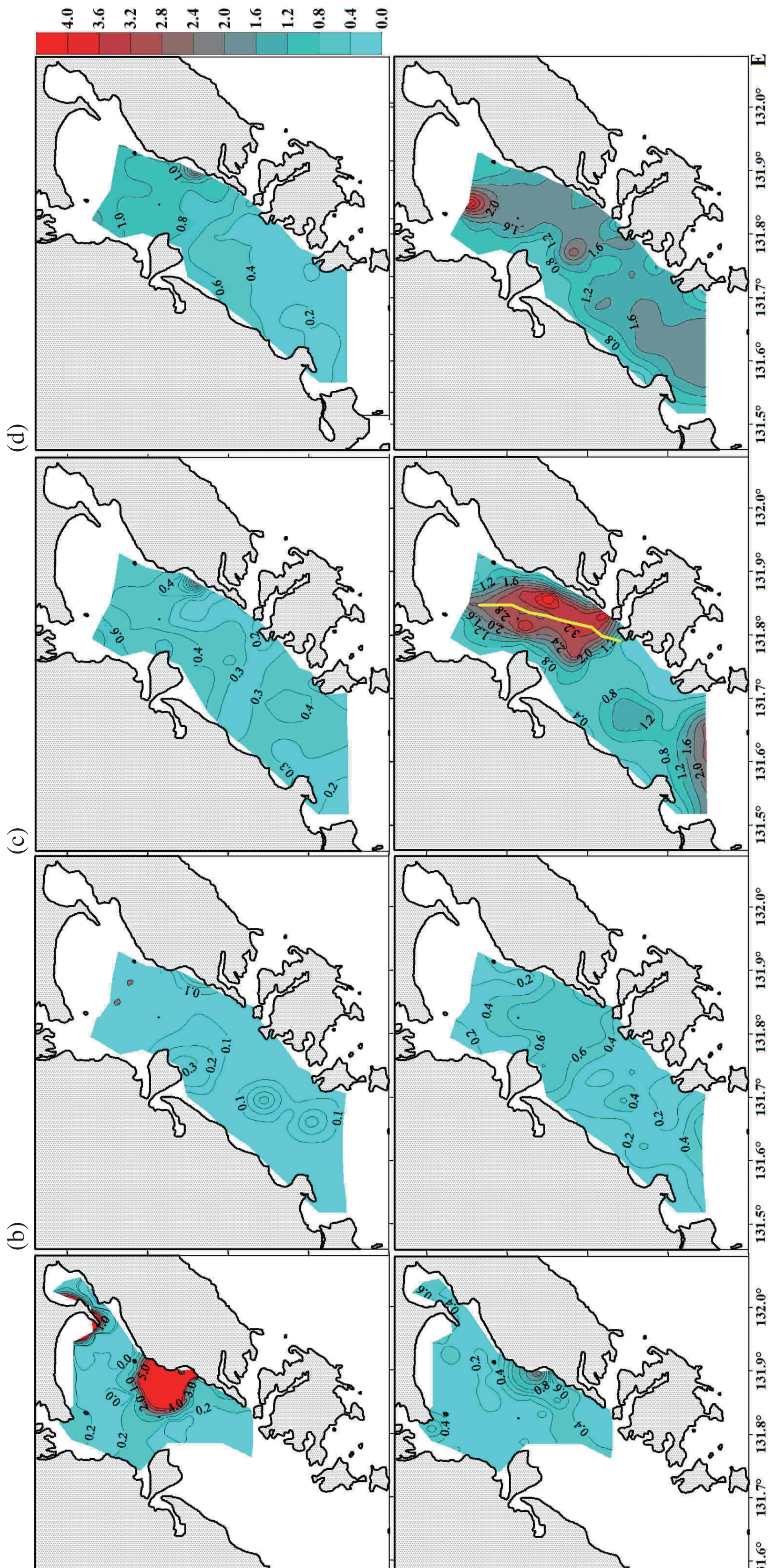

Fig. 5. Seasonal distributions of phosphate concentrations (μM) in the surface waters (top panel) and bottom waters (bottom panel) of Amursky Bay in 2008. (a) February, (b) May, (c) August, (d) October. The yellow line notes location of the CTD section presented at Fig. 10.

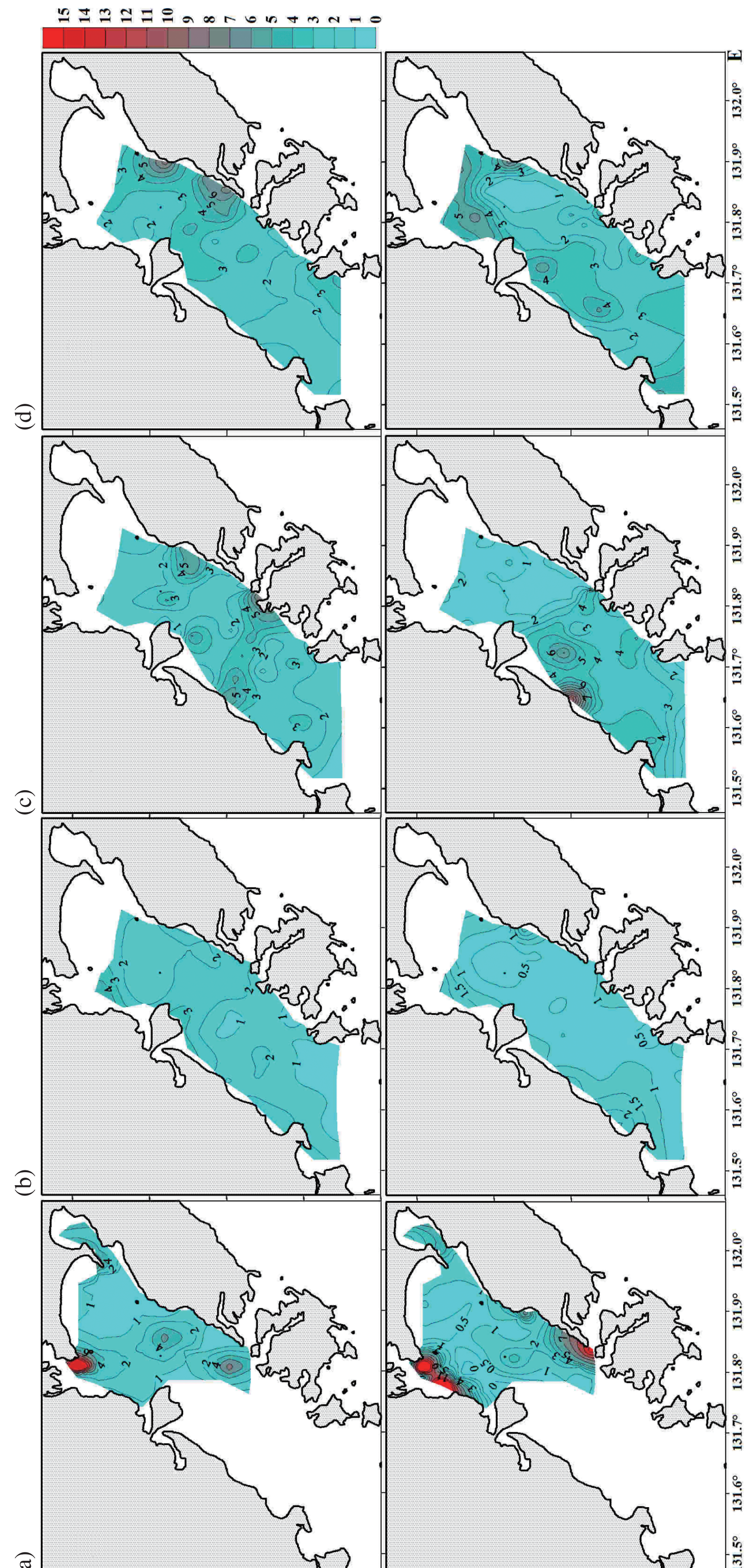

Fig. 6. Seasonal distributions of chlorophyll *a* concentrations (mg m^{-3}) in the surface waters (top panel) and bottom waters (bottom panel) of Amursky Bay in 2008. (a) February, (b) May, (c) August, (d) October.

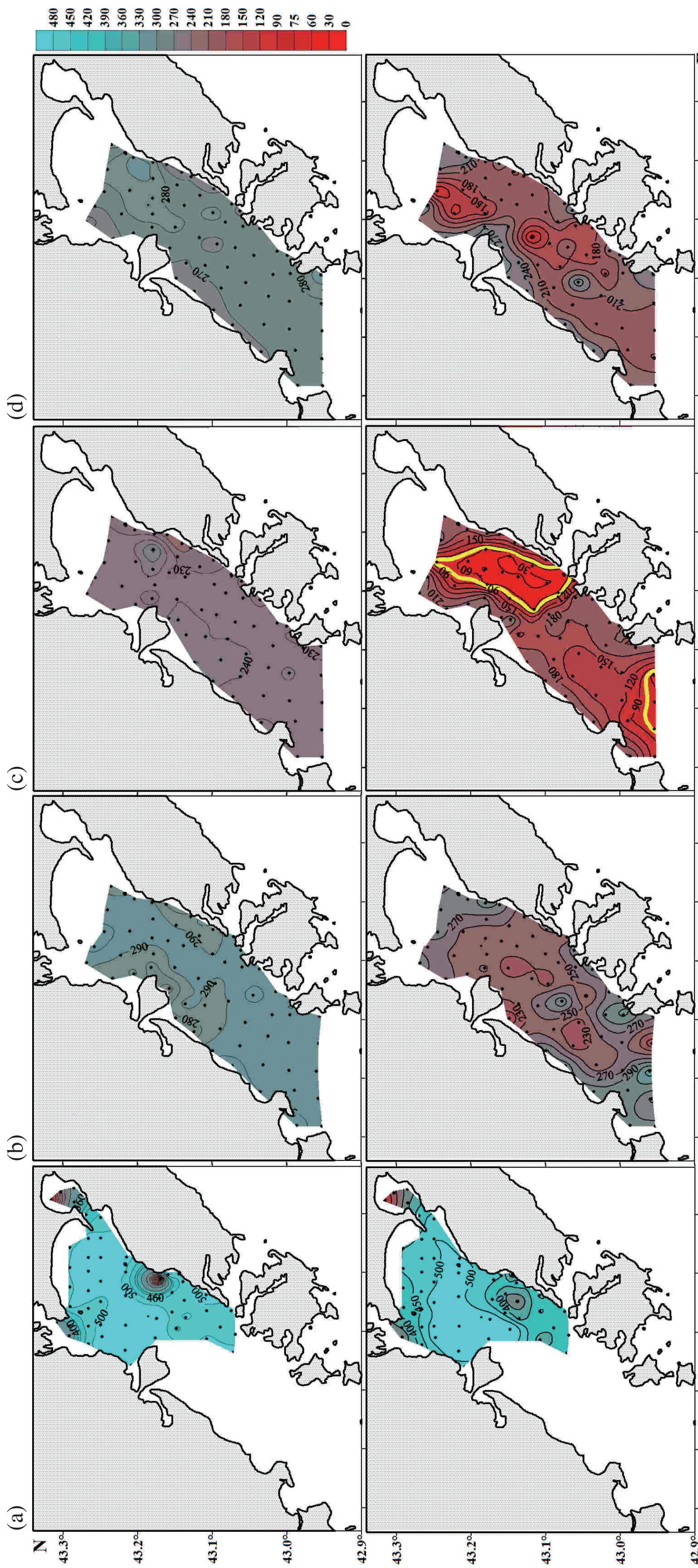

Fig. 7. Seasonal variability of dissolved oxygen concentration (μM) in the surface waters (top panel) and bottom waters (bottom panel) of Amursky Bay in 2008. (a) February, (b) May, (c) August, (d) October. The yellow line corresponds to DO concentration of 76 μM which is accepted as hypoxia.

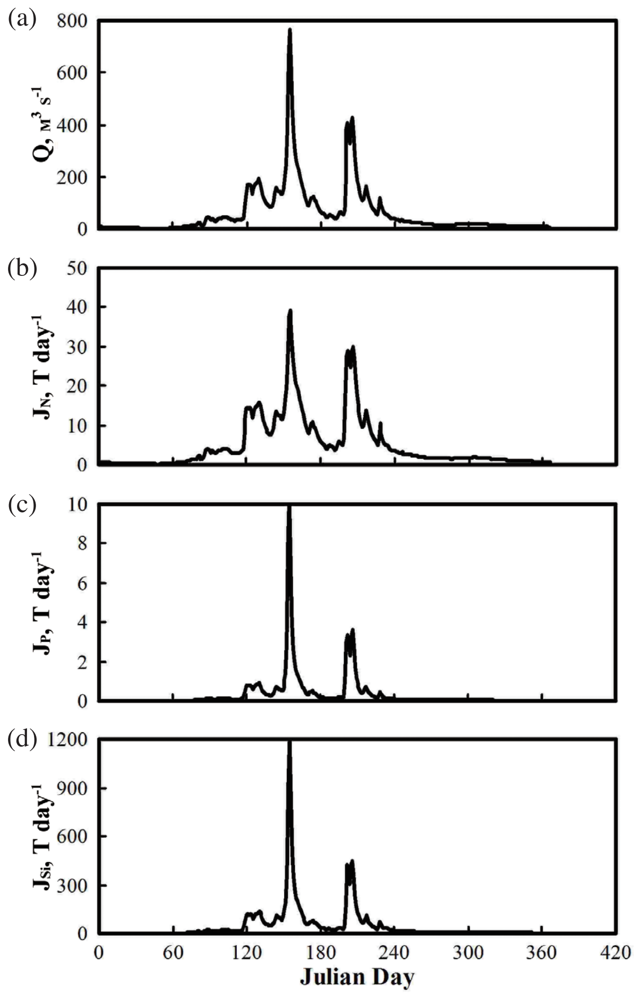

Fig. 8. Daily discharge of the Razdolnaya River (a) and fluxes of dissolved inorganic nitrogen (b), dissolved inorganic phosphorous (c) and dissolved inorganic silicates (d) into Amursky Bay by the Razdolnaya River as a function of Julian Days (redrawn from Mikhailik et al. 2011).

between those present for winter and summer seasons. At the moment of our observations in 2008, they were around 29 close to Razdolnaya river mouth and 33.0 - 34.0 over the major part of the bay.

Seasonal distribution of nitrate ions in the bay demonstrates generally low concentrations (0.1 - 1 μM) over all seasons with exception of two cases of high nitrate content (Fig. 4). One of them was observed in winter near the mouth of the Razdolnaya River (Fig. 4a). It may be explained by river input as river water has the highest concentrations of nitrate (up to 229 μM) in the winter. Another case of observed high nitrate concentrations (up to 18 μM) was in bottom waters during October (Fig. 4d, low panel).

Seasonal distribution of phosphorous concentrations is shown in Fig. 5. In winter, there is a large positive anomaly in the middle part of the bay, visible in the surface and bottom layers, and another less visible anomaly in the northern part of the bay (Fig. 5a). They are associated with waste water discharge from Vladivostok city and from small villages located in the north. The concentration of phosphorous has been increasing slightly from spring to autumn, both in surface and in bottom waters. It is also important to note that high phosphorous concentrations (up to 4 μM) were observed in the bottom waters in August (Fig. 5c, low panel).

Seasonal distribution of chlorophyll *a* demonstrates moderate concentrations of about 1 - 4 μg l^{-1} for all seasons in the surface and bottom waters (Fig. 6). These observations suggest that Amursky Bay is a high productive area in all seasons, even in winter when the area is covered with ice.

DO concentrations are widely varied in bottom waters. The maximal and minimal concentrations of DO were observed in winter (up to 598 μM) and summer (lowest value is 17 μM) (Fig. 7). The most important feature of this distribution is the occurrence of hypoxia in bottom waters in August (Fig. 7c, low panel). The yellow line on Fig. 7c (low panel) corresponds to the DO concentration, equal to the critical value of 76 μM, below which the conditions are considered to be hypoxia. The area with hypoxic conditions occupies a central part of the bay, with depths more than 15 m. Distributions of ammonium, normalized dissolved inorganic carbon (NDIC = DIC·35/S), CO_2 partial pressure, and silicates in the bottom waters have very similar structures to the distribution of DO and phosphate concentrations. Figure 9 shows a good correlation between phosphates, ammonium, NDIC, silicates concentrations, CO_2 partial pressure, and oxygen concentrations. It also shows dependence of DIN {$[NO_3]$ + $[NO_2]$ + $[NH_4]$} as a function of phosphates concentrations (Fig. 9f).

4. DISCUSSION

The high amplitude of seasonal variations of oceanographic parameters is most obvious and is as might be expected (Luchin et al. 2005). Detailed hydrological and hydrochemical surveys carried out over each of the four seasons of 2008 detected seasonal characteristics regarding hypoxia in Amursky Bay. Obviously, there are processes that induce and destroy hypoxia, and which have both natural and human influences. We will try to distinguish the role of natural and anthropogenic processes in formation and destruction of hypoxia.

4.1 Formation of Hypoxia

The monsoon climate of the Primorye Region is the main influencing factor on the seasonal characteristics of atmospheric precipitation. A major part of atmospheric precipitation occurs during the summer. Heavy rains may cause occasional flooding (Fig. 8). Increase of atmospheric temperatures and increased fresh water discharge from rivers result in a strong vertical stratification of the water column

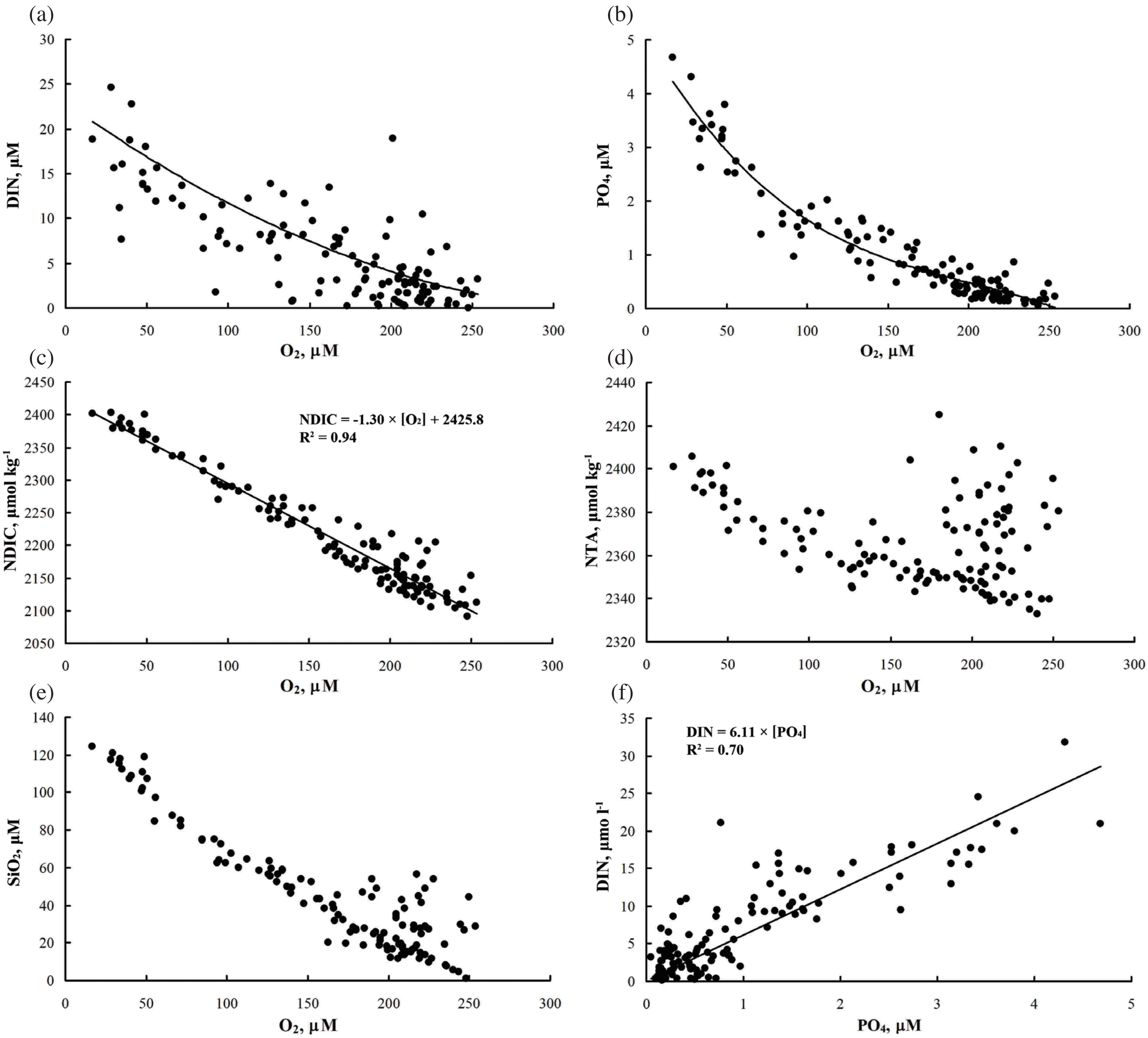

Fig. 9. Relationships between the bottom waters hydrochemical parameters obtained in August 2008 in Amursky Bay. (a) DIN ([NO_3^-] + [NO_2^-] + [NH_4^+]) vs DO, the solid line is obtained by the least squares method [Eq. (7) in text]; (b) PO_4^- vs DO, the solid line is obtained by the least squares method [Eq. (8) in text]; (c) NDIC (35 × DIC/S) vs DO; (d) NTA (35 × TA/S) vs DO; (e) [SiO_2] vs DO; (f) DIN vs PO_4^-.

during the summer (Figs. 2, 3, 10). The topography of Amursky Bay reveals a depression in its central portion (Fig. 1) which limits horizontal advection and water exchange in the bottom layer. These natural features of the bay cause weak dynamics in the bottom waters during the summer. Vertical sections of potential density anomaly and the fluorescence of chlorophyll a (Fig. 10) suggest that the depth of photic layer varies between 10 and 15 m. This is in agreement with the estimation of the photic layer by Secci disc (photic layer = 2.7 × Seccidepth).

As we suggested earlier (Tishchenko et al. 2008), the main process of hypoxia formation is by microbiological degradation of the "excess" amount of phytoplankton at depths exceeding 15 m under rather low photosynthetic active radiation in the bottom layer during the weak water dynamics period. Existence of "excess" phytoplankton is created by a phytoplankton bloom, which is the result of the nutrient enrichment of Amursky Bay, or its eutrophication (see the recent review by Lutaenko and Vashenko 2008). Moderate chlorophyll *a* concentrations during each season (Fig. 6) suggest a high primary production throughout the year due to the high eutrophication of the bay.

There are two main sources of nutrients in the Amursky Bay: (a) waste waters from the city of Vladivostok and (b) discharge from the Razdolnaya River. Annual fluxes of nutrients are summarized in Table 1. Waste waters from the

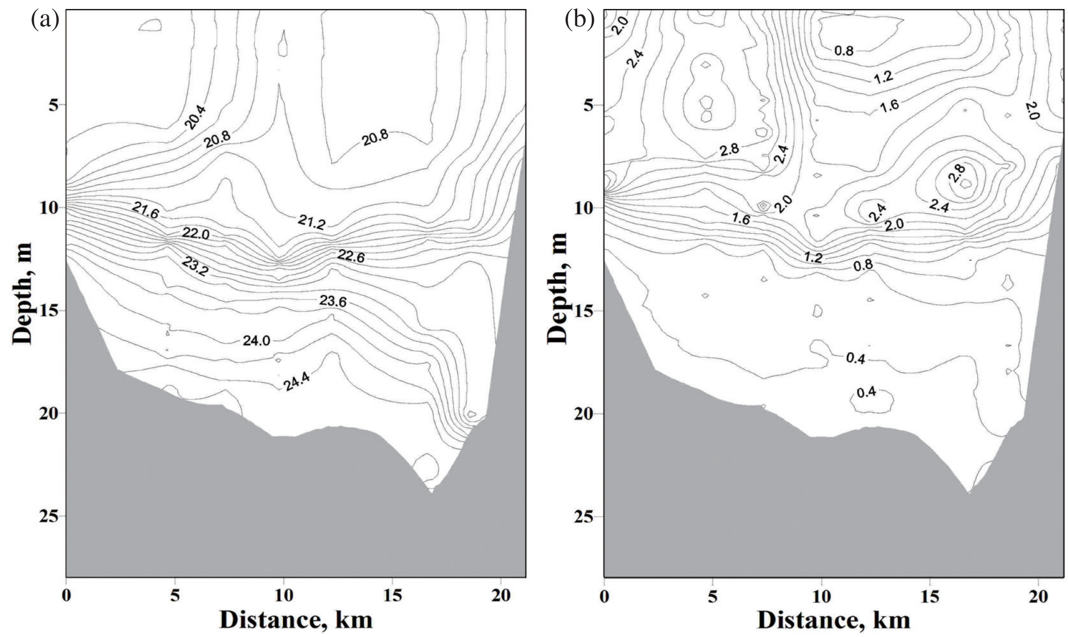

Fig. 10. Sections of density anomaly (kg m^{-3}) (a) and fluorescence of chlorophyll (mg m^{-3}) (b) in Amursky Bay, August 2008. Location of the section is noted by a yellow line on Fig. 6c, low panel; the left side of the section corresponds to the northern part of the bay.

Table 1. Annual loads (T year^{-1}) of nutrients (DIN - Dissolved Inorganic Nitrogen; DIP - Dissolved Inorganic Phosphorous), COD (Chemical Oxygen Demand), SS (Suspended Solids), BOD_5 (Biochemical Oxygen Demand with five days exposition) into Amursky Bay from river runoff and waste waters of Vladivostok (NOWPAP CEARAC, 2011).

Nutrients, COD, SS	DIN	N-tot	DIP	P-tot	COD	DISi	SS	BOD_5
River runoff	1800	4200	120	450	36560	17040	117840	37800**
Waste-water	700	1150*	100	140*	8000***	-	2156**	1733**

Note: (*) N-tot and P-tot values were calculated assuming that organic forms of nitrogen and phosphorus are 40 and 30% from the total of its contents, respectively (Henze et al. 1992); (**) (Gavrilevsky et al. 1998); (***) (POMRAC Technical Report 2006).

city of Vladivostok have a nearly constant rate over each season, and can be identified by a few local points, especially visible in the surface waters during winter (matched stars, Fig. 2). These points are characterized by relative low salinity (Fig. 3), high concentrations of phosphorous (Fig. 5), and ammonium. These locations are easily defined under the ice due to the absence of wind/wave mixing and a relatively low rate of photosynthetic nutrient immobilization. It is the opposite during warmer periods. Figure 6 does not show any high chlorophyll *a* concentration in the spring or summer, which is likely caused by a bloom in phytoplankton. Moreover, comparatively low concentrations of nitrate and phosphates in surface water are demonstrated by Figs. 4 and 5. These results suggest the high efficiency of the "biological pump". This pump transports nutrients supplied by non-salty waters (waste water and river water) from the surface water to the bottom water of the bay via photosynthesis, with the consequence of a degradation biomass in phytoplankton. It should be noted that our surveys were carried out during periods when the Razdolnaya River had comparatively low discharges (150 and 50 m^3 s^{-1} in May and August, respectively). We think that the local nutrients inputs play a minor role in hypoxia formation because (a) according to Table 1 these sources supply only about 30% of the loaded nutrients and (b) have an almost constant rate of nutrient flux to the bay. The energy from primary production flows consistently at higher trophic levels through all of the trophic food web chain.

In contrast with the city waste water, local inputs from the Razdolnaya River mostly originate from non-local sources (agriculture fields, atmospheric transport, and precipitation), which are highly variable in summer. Unexpected fluxes caused by heavy precipitation can be considered as nutrient pulses. These pulses play an important role in the formation of seasonal hypoxia (Anderson and Taylor 2001). Using daily nutrient fluxes (Fig. 8), it is easy to estimate that during the May - August period, the Razdolnay River supplies 80, 90, 87 and 92% of the annual fluxes of nitrogen, phosphorus, silicates, and suspended substances, respectively. We suggest the following scenario for hypoxia formation in the Amursky Bay:

(a) Due to heavy rains in June and July, water discharge may increase some of the time, and a large amount of nutrients and suspended matter are supplied into the surface layer of Amursky Bay by the Razdolnaya River during high water periods. In 2008, such eutrophication pulses occurred on June 2 and July 19 (Fig. 4; Mikhailik et al. 2011).

(b) After the settling of suspended matter, a phytoplankton bloom occurred as revealed by satellite images (Fig. 11).

(c) Due to the fast rate of the process, zooplankton and fish are excluded from the food chain, therefore the phytoplankton dies and sinks to the bottom.

(d) In the absence of higher trophic levels (mostly mobile fauna that fled), energy that was previously used to sustain more complex food webs is diverted to lower trophic levels (microbes). Microbiological treatment of autochthonic organic matter under low light conditions forms the near-bottom hypoxic layer.

(e) Due to the strong stratification of the water column in summertime caused by heating and freshening of surface waters, vertical mixing is limited and maintains a near-bottom hypoxic layer.

In August, there are strong correlations between DO and the concentration of DIN ($DIN = NO_3^- + NO_2^- + NH_4^+$), phosphates, NDIC, NTA, and silicates in the bottom water (Fig. 9). We assume that distinct correlations between the decrease of oxygen and the increase of NDIC, NTA, DIN, phosphorous and silicates contents (Fig. 9), and the similar shapes of spatial distributions of these anomalies (Figs. 5c, 7c, low panels), prove that these are the result of one process that governs hydrochemical features observed in the bottom waters of the bay during August. This process is a microbiological degradation of the "excess" phytoplankton, the main part of which is diatoms. Phylogenic studies show that the microalgae population in the area of the Razdolnaya River mouth and the adjacent waters of Amursky bay is dominated in population density by diatoms and cryptophytes (64 and 27%, respectively) and in biomass by diatoms (94%) (Stonik et al. 2009).

At normal oxic conditions, the degradation of "excess" phytoplankton can be expressed by the following scheme (Friedrich et al. 2002):

$$(CH_2O)_{106}(NH_3)_{16}H_3PO_4 + 138 \cdot O_2 \rightarrow \\ 106 \cdot CO_2 + 122 \cdot H_2O + 16 \cdot HNO_3 + H_3PO_4 \quad (1)$$

We used Redfield's stoichiometry of organic matter in scheme (1) (Redfield et al. 1963). However, there was a low level of nitrate concentration (0.1 - 1 μM; Fig. 5) and moderate concentrations of ammonium observed in the hypoxic area in both 2007 and 2008. At low oxygen concentrations, nitrate ions can be used by denitrifying bacteria for degradation of the organic matter. This process may be schematically expressed as follows (Friedrich et al. 2002):

$$(CH_2O)_{106}(NH_3)_{16}H_3PO_4 + 84.8NO_3^- + 99.8H^+ \rightarrow \\ 106CO_2 + 148.4H_2O + 16NH_4^+ + 42.4N_2 + H_2PO_4^- \quad (2)$$

Thus, a coupled process of nitrification-denitrification explains existence of this high anomaly of ammonium concentration. Mass-balance of nitrification-denitrification (schemes 1 and 2) can be written as:

$$(CH_2O)_{106}(NH_3)_{16}H_3PO_4 + \frac{7314}{63} \cdot O_2 + \frac{97}{63} \cdot H^+ \rightarrow \\ 106 \cdot CO_2 + \frac{160}{63} \cdot NH_4^+ + \frac{424}{63} \cdot N_2 + \frac{7950}{63} \cdot H_2O + H_2PO_4^- \quad (3)$$

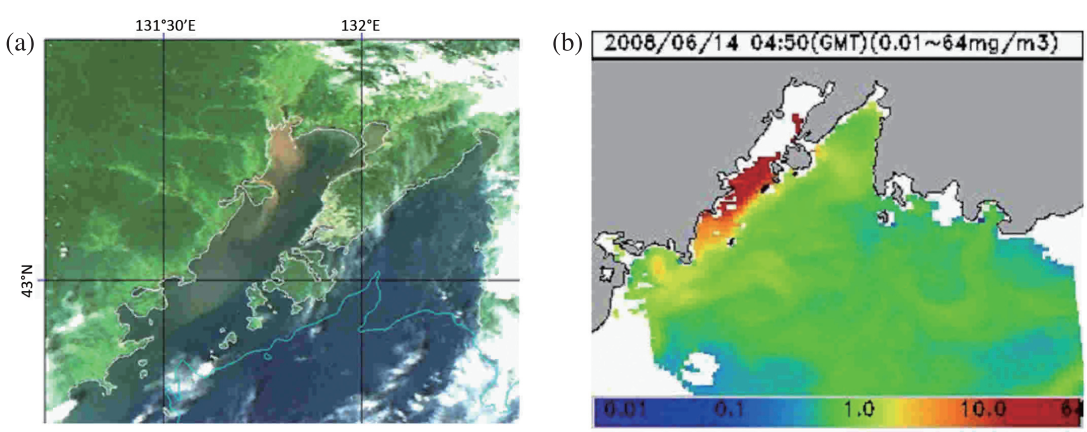

Fig. 11. Color satellite images from MODIS showing high content of suspended material from the Razdolnaya River (a) and then high Chl-a concentration (b) in Amursky Bay in Summer. Sources: http://rapidfire.sci.gsfc.nasa.gov/ and http://www.nowpap3.go.jp.

However, a mass-balance of reactions (1, 2 and 3) does not agree with our observations. According to (3), we expect N:P ratio to be about 2.5. Our averaged data (plot on Fig. 9f) give an N:P ratio value of about 6. Therefore, we suggest that for explanation of observed hydrochemical data, a microbiological pathway of dissimilatory nitrate reduction to ammonium ions (DNRA) occurs (An and Gardner 2002). Using Redfield stoichiometry of organic matter, this process can be schematically written as (Friedrich et al. 2002):

$$\begin{aligned}&(CH_2O)_{106}(NH_3)_{16}H_3PO_4 + 53NO_3^- + 122H^+ \rightarrow \\ &106CO_2 + 53H_2O + 69NH_4^+ + H_3PO_4\end{aligned} \tag{4}$$

Mass-balance of reaction (1) and (4) can be presented as follows:

$$\begin{aligned}&(CH_2O)_{106}(NH_3)_{16}H_3PO_4 + 106 \cdot O_2 + 16H^+ \rightarrow \\ &106 \cdot CO_2 + 106 \cdot H_2O + 16 \cdot NH_4^+ + H_3PO_4\end{aligned} \tag{5}$$

According to reaction (5), expected N:P ratio is 16. These processes are more complicated and evolving via the formation of an intermediate product - nitrite ions. Simultaneous existence of nitrite and ammonium ions provides another microbiological pathway - anoxic ammonium oxidation (ANAMMOX; Thamdrup and Dalsgaard 2002):

$$NO_2^- + NH_4^+ \rightarrow N_2 + 2H_2O \tag{6}$$

It has been found that NO_3^--dependent respiration is dominant over oxic respiration only when DO drops to below 2 - 4 μM, though denitrifying activities may occur at up to 20 of DO (Lam and Kuypers 2011). According to our data, such conditions can be realized on a sediment/water interface and/or in surface layer of sediments of the bay. Nonlinear correlations of DIN and phosphates content are from the decreasing of DO concentration (Figs. 9a, b), suggesting the enhancement of sediment-water ammonium and phosphates fluxes. By least-square fitting, we obtain equations as follows:

$$DIN / \mu M = 22.96 - 0.1307[O_2] + 1.8238 \cdot 10^{-4}[O_2]^2 \tag{7}$$

$$\begin{aligned}PO_4^{3-} / \mu M = 5.06 - 5.265 \cdot 10^{-2}[O_2] + \\ 2.2245 \cdot 10^{-4}[O_2]^2 - 3.67377 \cdot 10^{-7}[O_2]^3\end{aligned} \tag{8}$$

From relation (7), we found that molar ratios of [N]:[O] vary from 1.5:138 (at maximal oxygen contents) to 17:138 (at minimal oxygen contents). It is noted that Redfield's ratio is 16:138. Similar calculations for [P]:[O] ratios give 1.1:138 and 6.3:138. Redfield's ratio is 1:138. These calculations demonstrate significant denitrification at moderate oxygen concentration in bottom waters. In this case, a nitrification-denitrification process [reaction (3)] is probably dominant. To explain observed [P]:[O] ratios that exceed Redfield's ratio more than six times, we first suggest the sulphatereduction pathway of degradation of organic matter:

$$\begin{aligned}&(CH_2O)_{106}(NH_3)_{16}H_3PO_4 + 53SO_4^{2-} \rightarrow \\ &38H_2S + 16NH_4^+ + H_2PO_4^- + 106HCO_3^- + 15HS^-\end{aligned} \tag{9}$$

This pathway is realized into sediments. Second, we have to suggest enhanced vertical fluxes of nutrients from sediments into the overlying water. Such fluxes have been observed and discussed elsewhere (Forster et al. 1995; Testa and Kemp 2012). Actually, an enhancement of sediment-water ammonium and phosphorous fluxes was observed when the overlying water contained a DO concentration below 50 μM. One of the reasons for the enhancement is the intensification of bioturbation under hypoxic conditions (Forster et al. 1995). In our case, an average water volumetric respiration rate in the bottom water between March 1 and August 21 can be estimated as 3.3 μM day^{-1}. This is close to the results of measured volumetric respiration rates ranging from 4.1 to 10.8 μM day^{-1} on the Louisiana continental shelf (Murrell and Lehrter 2011). We have to note that pathways (4) and (9) of the degradation organic matter increase TA in surrounding water, as is demonstrated by Fig. 9d. However, with increasing oxygen concentrations, scattering on plots 9d and e are increased as well. These scattering points correspond to the samples with relative low salinity (about 29, 30). It should be noted that river water has a high silicate concentration and high NTA values. The cause of these scattering points is the contribution of river water properties into studied samples. Microbiological degradation of died "excess" of phytoplankton is a very complicated process and there are many possible pathways. Realization of the pathways is dependent on local conditions, primarily the DO concentration. Hydrochemical anomalies, which were observed in August 2007 and 2008 in the bottom waters of Amursky Bay, where low DO concentrations and N:P ratios, high concentrations of silicates, phosphates, ammonium, and NDIC all suggest that the degradation of expired phytoplankton is mostly governed by processes presented by schemes (1) - (9).

4.2 Destruction of Hypoxia

Formation of hypoxia on the shelf in tropical and subtropical areas caused by upwelling in the eastern boundary current system has been reported in many studies (e.g., recent review by Zhang et al. 2010). In our case, upwelling plays an opposing role and may interrupt coastal hypoxia.

Over the northwestern Japan Sea, monsoon winds change their phase from southern - northern direction on western - eastern or northwestern - southeastern direction which usually happens during September and October. These winds induce the development of upwelling along the Primorye coast and advection of the Japan Sea water onto the shelf of Peter the Great Bay (Zhabin et al. 1993; Zvalinsky et al. 2006; Zuenko 2008). Thus, in 2008, an upwelling and advection of cold open sea water in the bottom layer of the bay occurred just before our October survey (Fig. 2d, low panel). As a result, the bottom water temperature decreased to 3°C, while temperature of the surface water was still around 13°C. Nitrate concentrations significantly increased, up to 18 µM in October (Fig. 4d, low panel). They were around 0.1 - 1 µM in the winter, spring, and summer seasons. It should be noted that the shapes of cold water and nitrate distributions in the bottom waters are very similar (Figs. 2d, 4d). This could be only explained by advection of the Japan Sea intermediate water, which has high DO concentration (about 280 µM) as well and thus destroys hypoxia in the near-bottom layer of the bay. According to its characteristics (Talley et al. 2004) we suggest that this water could upwell from 100 - 300 m depths.

At the same time, changes of the monsoon phase are accompanied by changes in precipitation. During the dry season (September through March), the Razdolnaya River discharge gradually declines and the contribution of nutrients supplied to the Amursky Bay becomes insignificant (Fig. 8).

The third physical process, which finally destroys hypoxia, is winter convection. Due to cooling and brine rejection during ice formation, the water column of the Amursky Bay becomes well mixed and vertically uniform. Convection intensifies photosynthesis under the ice during the winter season. High photosynthetic activity results in low concentrations of nutrients, both in the surface and in the bottom waters. Moreover, the whole water column becomes supersaturated by DO (130 - 150%) and under-saturated by dissolved CO_2 (usually, pCO_2 was around 140 µatm) for the main part of the studied area.

It is also important that in the winter season, the prevalence of anticyclonic, unclouded weather provides high radiation over the region, and because of frozen rivers and the ice cover, any nutrient pulses are minimal. This period is very good for fishing, for both people and animals (Larga seals) (Trukhin 2005). Obviously, in the winter, energy flows through the food chain, from a low trophic level (phytoplankton) to the upper layer (animals). Winter is the best season for the Amursky Bay when its ecosystem is revitalized.

4.3 The Long Term Trend of Hypoxia

Published data demonstrates that the lowest values of DO concentrations obtained in the summer in the bottom waters of Amursky Bay have been decreasing systematically with time over the last eighty years (Table 2). The eutrophication of the Amursky Bay is not only a process affecting the individual regional ecosystem, but is also a part of a global phenomenon. The driving forces at the global scale include human population growth (mostly around the East China Sea), increased anthropogenic emission of reactive nitrogen species into the atmosphere (mostly through agriculture, the increase in automobile use, oil exploration, and deforestation), increased atmospheric CO_2 (global acidification), and climate change (Duarte 2009). It is well documented that the exponential increase of fossil fuel combustion, production of N-fixing crops, and the industrial production of fertilizers corresponds to periods of exponential spreading of coastal eutrophication (Boesch 2002; Rabalais et al. 2010; Zhang et al. 2010; Kim et al. 2011). There is a period between 1960s - 1980s, in which Amursky Bay became hypoxic during the summer, most likely originating in the 1970s (Table 1). This could be the result of global processes.

5. SUMMARY

There are both natural and human induced drivers of hypoxia in the Amursky Bay. The natural factors include a monsoon climate with sharp seasonal variations in river discharge, dominant winds, stratification, water dynamics, and topography of the bay. The human factors include local and non-local sources of nutrients and fluxes of suspended substances. The primary influencing factor regarding the formation of hypoxia in the bay during the summer is a synergism between the monsoon climate of the Primorye Region and eutrophication of the coastal area; in effect, unpredictable

Table 2. Lowest DO concentrations (saturation degree by air oxygen) in the bottom water of Amursky Bay which was obtained in surveys implemented in the summer over a period between 1928 - 2011 (1 - Voronkov 1941; 2 - Lastovetsky and Veshcheva 1964; 3 - Redkovskaya 1980; 4 - Rodionov 1984; 5 - Podorvanova et al. 1989; 6 - 11 our data).

[O2], %	**60**	**40**	**36**	**23**	**30**	**22**	**18**	**1.7**	**6.7**	**21.7**	**5.4**	**6.1***
Year	1928	1961	1973	1975	1975	1978	2005	2007	2008	2009	2010	2011
Reference	1	2	3	3	4	5	6	7	8	9	10	11

Note: (*) Concentrations printed by bold mean hypoxic conditions.

nutrient fluxes caused by heavy precipitations provide "excess" phytoplankton. During the autumn and winter, natural drivers such as upwelling, decrease of river runoff, and winter convection destroy hypoxia. The winter season is the best season for Amursky Bay, as it is when its ecosystem is replenished.

Obviously, the natural drivers have been active in the area over many years. However, analysis of available published data and our observations suggests that a negative tendency in DO content in the bottom water of Amursky Bay has started only in the second half of the last century. This could be explained by an increasing role of non-local sources of nutrients over time. This is in agreement with the conclusion of Rabalais et al. (2009), that eutrophication of coastal waters by non-local sources of nutrients is a part of global change. Presently, a lack of efficient management of non-local nutrient loading is a global social problem at the present time.

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