Dear Prof. Wajih Naqvi,

We would like to thank you and the anonymous reviewer and Prof. Carolin Löscher for their comments and suggestions which helped us improve our manuscript. Please find our final responses (in red) to all comments (in black) in this document. The line numbers mentioned by the reviewers refer to the original version of the manuscript while the line numbers in our replies refer to the revised version of the manuscript.

**Response to reviewer #1.**

Comments from reviewer #1 are in black while our response in red and changes in the manuscript are in blue.

The reviewer found the manuscript acceptable since it provided relevant findings. However, the following observations are pointed out:

1. The last sentence in the abstract seems spurious and unsubstantiated, because there was no part of the manuscript that reported research finds on $NO_x$ emissions from ships.

In our manuscript of Page 8 line 10-15, we compared the NO emission of the Yellow Sea and the Bohai Sea to the atmosphere based on our observation ($7.6\times10^7$ gN·month$^{-1}$) with the satellite-derived average $NO_x$ emission estimate (about $1.3\times10^{10}$ g N·month$^{-1}$) reported by Ding et al., 2018. Because Ding et al.'s (2018) estimate is dominated by the $NO_x$ emissions from ships' diesel engines, thus we concluded that the oceanic NO emissions to the atmosphere were negligible compared to anthropogenic $NO_x$ sources such as emissions from ships.

2. Judging from the distribution patterns of NO flux and the wind speed (Figure 5), one would expect an important inference and conclusive statement in the abstract and discussion. Previous investigators (Anifowose AJ, Sakugawa H. 2017. Determination of Daytime Flux of Nitric Oxide Radical (NO·) at an Inland Sea-Atmospheric Boundary in Japan. J Aquat Pollut Toxicol., 1:2) reported wind speed as

an important factor governing NO flux at the air-sea interface.

Yes, wind speed was an important factor governing NO flux at the air-sea interface judging from the distribution patterns of NO flux and the wind speed. We have supplemented it in the revised manuscript and cited this reference of Anifowose and Sakugawa (2017).

The comparison of wind speeds and flux densities reveal that the flux densities are mainly driven by the wind speed (Fig. 7) and they showed significant positive relationship (SPSS v.16.0). Anifowose and Sakugawa (2017) also found that wind speed was an important factor governing NO flux at the air-sea interface.

3. The analytical methods on the measurements of NO concentration and during irradiation experiments (photoformation rate) are not explicit enough. I understand that the authors referred to Liu et al. (2017), there is need for them to report detailed analytical procedures in the manuscript.

Thank you for your comment. We had supplemented the analytical methods on the measurements of NO concentration and during irradiation experiments in revised manuscript.

The photolysis experiments were conducted under natural light conditions on deck. Surface water samples were filtered with 0.45 μm Millipore membrane and transferred into 10 mL cleaned quartz vials wrapped with Al foil with no headspace. Then 200 μL $NaN_3$ solutions were added to remove the microbial influence. After adding 10 μL of $1\times10^{-3}$ mol $L^{-1}$ 2, 3-diaminonaphthalene (DAN, trap NO) solution and gentle mixing, the fluorescence of the mixed solution was measured before irradiation (Liu et al., 2017). Then the same sample without addition DAN of were capped with a Teflon-lined silicone septum and without Al foil, the vial was placed in a shallow circulating seawater bath. After 0.5 h irradiation time, 10 μL of $1\times10^{-3}$ mol $L^{-1}$ DAN were added and the fluorescence of the solution was analyzed. The NO concentrations were measured with the method described in Liu et al., (2017). The NO photolysis production rates were computed based on the time-dependent difference between the NO concentrations before and after irradiation. For dark controls, vials were wrapped in Al-foil. The quartz vials and syringe used in the experiment were soaked in a 10% (v/v) HCl bath for 24 h, rinsed with Milli-Q water and baked at 500°C for 4 h.

4. In view of comment 3 above, one would ask the precious question as to whether the measured concentration of NO during the irradiation experiment was steady-state concentration, even when the NO scavenging rate constant in the seawater (during the experiment) remained unknown?

During the irradiation of sunlight, production and consumption of NO occurs simultaneously in seawater bulk, for example, radicals from CDOM can scavenge NO, and thus the measured concentration of NO was a net value of NO production.

5. I think there should be comprehensive correlation plot (and its discussion in the manuscript) of relationship between NO and $NO_2$(a major NO source). While it is true that the authors presented Figure 3 to reflect this, we only have very scanty data plotted.

In this study, NO surface concentrations did not show any statistically significant relationship with $NO_2^-$ throughout the whole area, and similar variations of salinity, NO surface concentrations, and $NO_2^-$concentrations was found only from station B65 to station B70 where affected by the outflow of the Huanghe River in the southern BS (Figure 3). The photochemical production of NO was also affected by pH, temperature, irradiation density, concentration of CDOM, etc. besides $NO_2^-$ concentration. Therefore, a comprehensive correlation might be difficult to obtain by in situ observation.

6. Page 2, Line 1: 10 % should be 10%. This should be applicable in other relevant places in the manuscript.

We have correct them as suggested. Thank you for your suggestion.

This reaction may account for 10% of nitrite loss in surface waters of the central equatorial Pacific (Zafiriou et al., 1980).

Throughout the manuscript, the space between number and % were removed.

7. Page 3: we have interchangeable use of “h” and “hour(s)”. The authors should stick to “h” preferably.

We decide to stick to “h” as suggested.

The NO photolysis production rates were computed based on the time-dependent difference between the NO concentrations before and after irradiation. For dark controls, vials were wrapped in Al-foil. The quartz vials and syringe used in the experiment were soaked in a 10% (v/v) HCl bath for 24 h, rinsed with Milli-Q water and baked at 500°C for 4 h.

8. Page 6, Line 29: “$0.00 \times 10^{-11}$ mol $L^{-1}s^{-1}$” should be “not detectable”

We have corrected it as suggested.

The photoproduction rates of NO in the BS, the NYS, and the SYS varied from not detectable to $5.07 \times 10^{-11}$ mol $L^{-1}$ $s^{-1}$, 0.09 to $0.69 \times 10^{-11}$ mol $L^{-1}$ $s^{-1}$, and 0.32 to $1.54 \times 10^{-11}$ mol $L^{-1}$ $s^{-1}$, respectively.

9. Page 7: The statement between Lines 8 and 9 should read “…but would also generate reactive oxygen species like $O_2$•-, ROO• and other OH-related radicals, which in turn, would efficiently scavenge NO…”

We have revised it as suggested. Thank you for your suggestion.

The high illumination observed in the SYS (Table 2) does not only lead to enhanced NO photoproduction (see Table 2) but would also generate reactive oxygen species like $O_2$•-, ROO•, and other OH- related radicals, which in turn, would efficiently scavenge NO (Olasehinde et al, 2010)

10. Page 7, Line 17: 24h should be 24 h

We have corrected it as suggested. Thank you for your suggestions.

Together with the fact that that NO was supersaturated during daytime and nighttime during the 24 h station B65 this indicates that the BS and YS were a source of NO to the atmosphere.

11. Page 10, Line 19: 13(4), 1-31 were repeated in the reference

We have corrected it as suggested. Thank you for your comments and suggestions.

Liu, C.Y., Feng, W.H., Tian, Y., Yang, G.P., Li, P.F., and Bange, H.W.: Determination of dissolved nitric oxide in coastal waters of the Yellow Sea off Qingdao, Ocean Sci., 13(4), 1-31. https://doi.org/10.5194/os-2017-10, 2017.

**Response to Prof. Carolin Löscher.**

Comments from reviewer #1 are in black while our response in red and changes in the manuscript are in blue.

The manuscript by Ye Tian et al on NO distribution in the Bohai and Yellow Sea is a first report on the distribution of this intermediate of the nitrogen cycle, complemented with a model on production sources and pathways. The paper is well-written and straight forward to understand, it will certainly be interesting to the readers of Biogeosciences.

Thank you very much for your comments and suggestions. The manuscript was amended, and you will find a detailed description in how we took all the comments and suggestions into account in the preparation of the revised manuscript.

I have some comments of rather technical nature, as well as some questions to the authors, which I hope to be perceived constructive for the quality of the manuscript. Generally, I am wondering why there hasn't been any discussion of the NOx rates from anthropogenic sources as they were mentioned three times in the paper. This would possibly be important to do, particularly in the context of production pathways, which leads me to my second point.

In our manuscript 3.4 "sea-to-air fluxed of NO" part, we describe the anthropogenic source from the ship engine in the Bohai and Yellow Seas as "Ding et al. (2018) report a satellite-derived average $NO_x$ emission estimate for June over the BS and the YS in the period from 2007 to 2016 of about 1.3 ×$10^{10}$ g N $month^{-1}$. The obvious very large discrepancy between the satellite-derived emission estimate and the one presented here results from the fact that Ding et al.'s (2018) estimate is dominated by the $NO_x$ emissions from ships' diesel engines. This indicates that oceanic NO emissions to the atmosphere only account for a negligible fraction (~0.6%) of the $NO_x$ emissions observed over the BS and YS."

Most of the production seems to be photochemical in surface waters. A possible change in dust particles or a change in UV intensity could both alter this production, I assume, to a more or less significant extent- could that be discussed?

We have added the discussion about dust particles, however, we did not do the detailed researches about this, thus we cited Olasehinde et al. (2010) and Liu et al. (2017) to elaborate the influence of the dust particles.

Besides, Olasehinde et al. (2010) found that filtered and unfiltered seawater samples collected from the Seto Inland Sea showed no significant difference in NO• photoformation rates, which suggested a negligible contribution of NO• produced by photobiological processes from particle matter in seawater. However, Liu et al. (2017) reported that the rates difference between filtered (0.45 μm, $1.46\times10^{-12}$ mol $L^{-1}$ $s^{-1}$) and unfiltered ($1.52\times10^{-12}$ mol $L^{-1}$ $s^{-1}$) seawater samples from coastal waters of the Yellow Sea indicated that particles in seawater could increase the NO production rate. The difference might be due to the composition of sample, filter membrane, etc. Thus, further research is needed.

We used the UV data from ECMWF reanalysis data sets (ERA–5 hourly mean surface downward UV radiation flux data) and analyzed the photoproduction rates with UV data. The NO photoproduction rates showed a positive relationship with the mean surface downward UV radiation flux ($p < 0.01$, $r = 0.865$, $n = 26$),

Overall, the NO photoproduction rates showed a positive relationship with illuminance ($p < 0.01$, $r = 0.884$, $n = 26$) and the mean surface downward UV radiation flux ($p < 0.01$, $r = 0.865$, $n = 26$) indicating that the NO concentrations in the surface layer during daylight were dominated by photochemical production.

The average photoproduction rate in the NYS was obviously lower than those in the BS and the SYS, consistent with the average illuminances of 22450, 20433, and 27825 lx and the mean surface downward UV radiation flux 34.7, 32.1, and 40.6 W $m^{-2}$ for the BS, the NYS and the SYS, respectively.

In addition, there is this mysterious pathway producing NO during dark periods. One option would be nitrification, which is in some steps light sensitive- this needs to be discussed from my perception.

We have added some discussion like nitrification process and some chemical process into our manuscript.

This suggests that there was also a non-photochemical NO dark production in the surface layer like nitrification process or other chemical processes like the process of ammonium ($NH_4^+/NH_3$) oxidation into $NO_2^-$ and $NO_3^-$ (Joussotdubien and Kadiri, 1970). Caranto and Lancaster (2017) found that NO was an obligate bacterial nitrification intermediate produced by hydroxylamine oxidoreductase. Ward and Zafiriou (1988) also found that NO might play as an intermediate of a soluble byproduct of nitrification such as hydroxylamine.

The obvious pathway, denitrification, does not seem to contribute anything here- why is that?

In our study area, DO fluctuated from 178 to 271 μmol $L^{-1}$ in the surface water and 147 to 266 μmol $L^{-1}$ in the bottom water (added into the manuscript), which indicated that the water was well-oxygenated thus it seemed that denitrification could not occur. In our manuscript, “The water columns of the BS and YS were well-oxygenated during our study and, thus, no suboxic or anoxic conditions were detected. Therefore, we did not found any enhancement of NO concentrations in the bottom layers with low DO or $NO_2^-$ concentrations as observed in the OMZ of the eastern tropical South Pacific Ocean off Peru (Lutterbeck et al., 2018)”, which partly explained this.

Technical considerations:

P1

L. 12 change ‘pathways’ to ‘turnover’

We have corrected it and thank you.

however, our knowledge about its oceanic distribution and turnover is rudimentary.

L. 17 and throughout the text: There is a dot between mol and L, please remove

Thank you, we have removed the dot between mol and L throughout the text.

L. 21 and throughout the text, the commas are incorrect. Replace ‘unknown’ by ‘unidentified’.

We have corrected it and thank you.

The persistent nighttime NO supersaturation pointed to an unidentified NO dark production.

L. 23 The last sentence is repeated later in the manuscript and is not particularly informative as part of the abstract, I recommend removing it, here.

We have removed it as you advised.

p. 2

L. 3 What would be the impact of NO in the atmosphere?

We have added the NO environmental influence to the revised ms.

Zafiriou and McFarland (1981) suggested that photochemically produced NO is a potential source of atmospheric NO during daylight, which could further lead to ozone hole, acid precipitation and photochemical smog (Bange, 2008).

L. 14 ‘The current understanding…’

We have revised it and thank you.

The current understanding of the oceanic NO distribution is mainly limited to the ocean surface (Zafiriou and McFarland, 1981; Olasehinde at al., 2009; 2010; Liu et al., 2017) and oxygen minimum zones (Ward and Zafiriou, 1988; Lutterbeck et al., 2018).

P. 3

L 13 ff. Please remove this list and show the stations on the map in Figure 1.

We have corrected it and thank you.

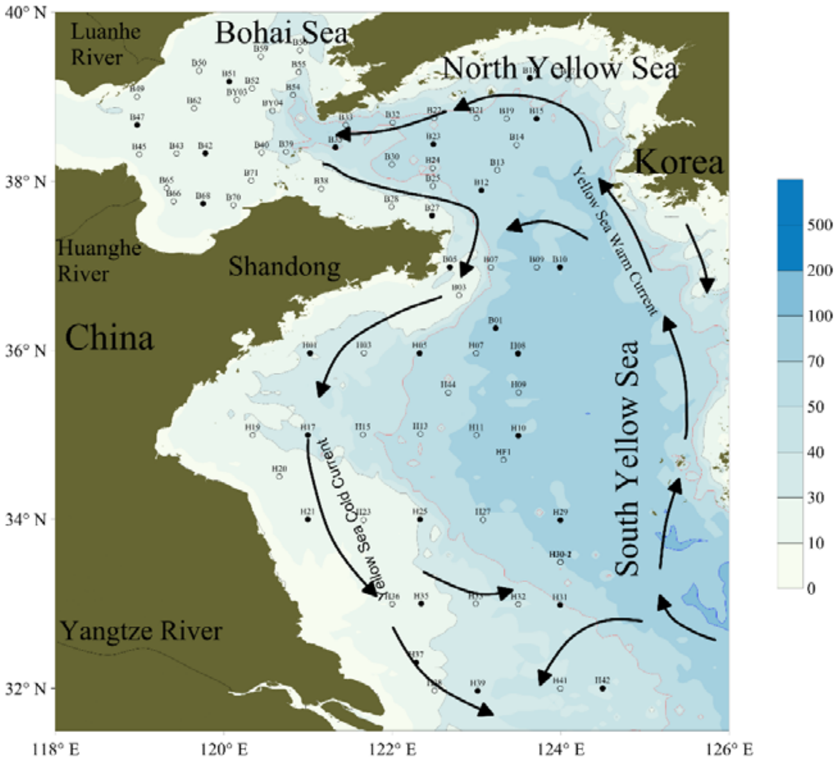

**Figure 1.** Locations of the sampling stations in the BS and the YS during summer. Solid dots (●) represent the stations for incubation experiments.

L. 22 Please explain what those chemicals are added for.

The $NaN_3$ solutions were added to remove the microbial influence while DAN were added to react with NO (process of trapping NO) and the fluorescence of the product was measured using a method described by Liu et al., 2017.

Then 200 μL $NaN_3$ solutions were added to remove the microbial influence. After adding 10 μL of $1\times10^{-3}$ mol $L^{-1}$ 2, 3-diaminonaphthalene (DAN, trap NO) solution and gentle mixing, the fluorescence of the mixed solution was measured before irradiation (Liu et al., 2017).

p. 4

L. 1 replace ‘were’ by ‘was’

We have corrected it and thank you.

The concentrations of dissolved inorganic nitrogen (nitrate, nitrite, and ammonium) was analyzed using a nutrient automatic analyzer (Auto Analyzer 3, SEAL Analytical, USA) in the laboratory.

L. 3 the statement of the precision sounds weird, rephrase please.

We have revised “higher” to “better than”.

The detection limits were 0.14 μmol $L^{-1}$ for nitrate, nitrite, and ammonium, with the precision of the method better than 3% (Liu et al., 2005).

p. 5

L. 16 This could still be explained, here and not only presented as a table, also the table content should be submitted to PANGAEA and a doi should go into the text, here.

We have added some information and explanation here and the data was submitted to Pangaea and we are still waiting for the doi (https://issues.pangaea.de/browse/PDI-21749).

In the study area, temperature varied from 8.8 to 21.7°C and salinity varied from 29.57 to 32.73‰ in the surface water. DO fluctuated from 178 to 271 μmol $L^{-1}$. The average concentrations of $NH_4^+$-N, $NO_2^-$-N, and $NO_3^-$-N were 2.11, 0.20, and 2.59 μmol $L^{-1}$, respectively. While temperature varied from 3.8 to 21.6°C, salinity varied from 30.00 to 33.71‰, and DO varied from 147 to 266 μmol $L^{-1}$ in the bottom water.

Fig. 2 The panels could be smaller and shown side by side, also a colored figure would be beneficial, here.

We have redrawn the colored figure and made them side by side.

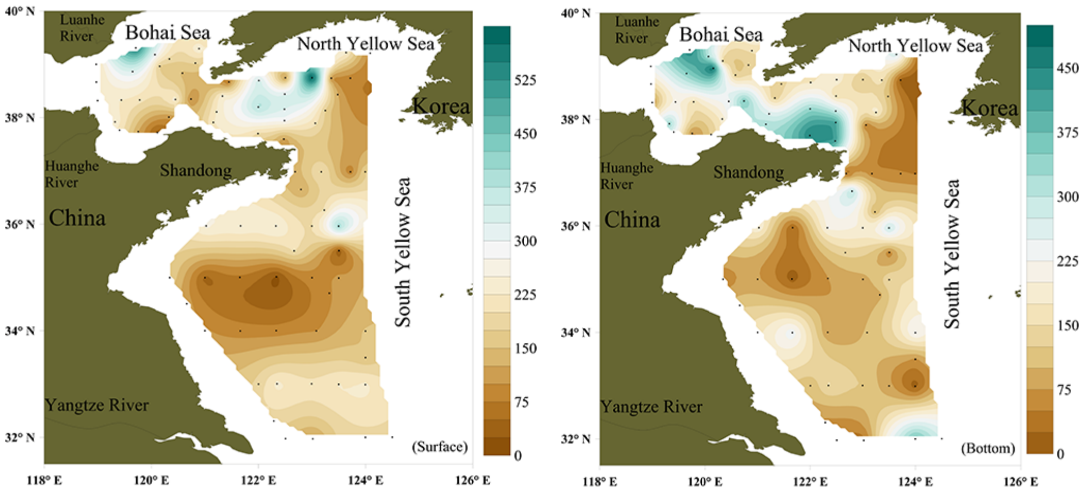

**Figure 2.** Horizontal distributions of NO (pmol $L^{-1}$) in the surface and bottom layers.

L. 26 How would a correlation to depth make sense if we are talking about surface samples?

We have removed the description of the correlation between the depth and the NO concentration in the surface water.

p. 7

L. 11 replace ‘was’ by ‘were’

We have corrected ‘was’ into ‘were’ and thank you.

Overall, the NO photoproduction rates showed a positive relationship with illuminance ($p < 0.01$, $r = 0.884$, $n = 26$) and the mean surface downward UV radiation flux ($p < 0.01$, $r = 0.884$, $n = 26$) indicating that the NO concentrations in the surface layer during daylight were dominated by photochemical production.

L. 13 Here, a discussion on the different biological pathways would fit in well.

Thank you for your suggestion. We have revised this part about the nitrification process here.

This suggests that there was also a non-photochemical NO dark production in the surface layer like nitrification process or other chemical processes like the process of ammonium ($NH_4^+/NH_3$) oxidation into $NO_2^-$ and $NO_3^-$ (Joussotdubien and Kadiri, 1970). Caranto and Lancaster (2017) found that NO was an obligate bacterial nitrification intermediate produced by hydroxylamine oxidoreductase, Ward and Zafiriou (1988) also found that NO might play as an intermediate of a soluble byproduct of nitrification such as hydroxylamine.

L. 19 dot after ) is missing.

We have corrected it and thank you.

p. 8

L. 17/18 Awkward sentence, please rephrase

We rephrased it as "The horizontal distribution of NO concentration in both the surface and bottom layers were highly variable, however, there was no significant difference between the mean NO concentrations in the surface and bottom layers."

L. 21, same comma situation as in the abstract. Again, it's only unidentified, but possibly known. As no genetic or biological data is presented such a statement is not possible.

We have corrected "unknown" into "unidentified" and the comma situation has also been revised.

The persistent nighttime NO supersaturation pointed to a non-photochemical (so far unidentified chemical and/or biological) NO dark production.

L. 24 This is the same sentence as in the abstract, the whole idea should be discussed, before, otherwise the statement is somewhat unfounded.

As mentioned above, $NO_x$ emissions from ship's diesel engines were described as "Ding et al. (2018) report a satellite-derived average $NO_x$ emission estimate for June over the BS and the YS in the period from 2007 to 2016 of about 1.3 ×$10^{10}$ g N month$^{-1}$. The obvious very large discrepancy between the satellite-derived emission estimate and the one presented here results from the fact that Ding et al.’s (2018) estimate is dominated by the $NO_x$ emissions from ships’ diesel engines. This indicates that oceanic NO emissions to the atmosphere only account for a negligible fraction (~0.6%) of the $NO_x$ emissions observed over the BS and YS.”

**Added references:**

Joussotdubien, J., and Kadiri, A.: Photosensitized Oxidation of Ammonia by Singlet Oxygen in Aqueous Solution and in Seawater, Nature, 227, 700-701, https://doi.org/10.1038/227700b0, 1970

# Nitric oxide (NO) in the Bohai and Yellow Seas

Ye Tian[1,3], Chao Xue[1,3], Chun-Ying Liu[1,3,*], Gui-Peng Yang[1,2,**], Pei-Feng Li[1,3], Wei-Hua Feng[4], and Hermann W. Bange[5]

[1]Key Laboratory of Marine Chemistry Theory and Technology, Ministry of Education, Qingdao, 266100, China
[2]Laboratory for Marine Ecology and Environmental Science, Qingdao National Laboratory for Marine Science and Technology, Qingdao 266071, China
[3]College of Chemistry and Chemical Engineering, Ocean University of China, Qingdao, 266100, China
[4]Key Laboratory of Engineering Oceanography, Second Institute of Oceanography, MNR, Hangzhou, 310012, China
[5]GEOMAR Helmholtz-Zentrum für Ozeanforschung Kiel, Kiel, 24105, Germany

*Correspondence to*: *Chun-Ying Liu (roseliu@ouc.edu.cn); **Gui-Peng Yang (gpyang@ouc.edu.cn)

删除的内容:

删除的内容: SOA

**Abstract.** Nitric oxide (NO) is a short-lived compound of the marine nitrogen cycle; however, our knowledge about its oceanic distribution and turnover is rudimentary. Here we present the measurements of dissolved NO in the surface and bottom layers at 75 stations in the Bohai Sea (BS) and Yellow Sea (YS) in June 2011. Moreover, NO photoproduction rates were determined at 27 stations in both seas. The NO concentrations in the surface and bottom layers were highly variable and ranged from below the detection limit (i.e. 32 pmol $L^{-1}$) to 616 pmol $L^{-1}$ in the surface layer and to 482 pmol $L^{-1}$ in the bottom layer. There was no significant difference between the mean NO concentrations in the surface (186 ± 108 pmol $L^{-1}$) and bottom (174 ± 123 pmol $L^{-1}$) layers. A decreasing trend of NO in bottom layer concentrations with salinity indicates a NO input by submarine groundwater discharge. NO in the surface layer was supersaturated at all stations during both day and night and therefore the BS and YS were a persistent source of NO to the atmosphere at the time of our measurements. The average flux was about $4.5\times10^{-16}$ mol $cm^{-2}$ $s^{-1}$ and the flux showed significant positive relationship with the wind speed. The accumulation of NO during daytime was resulting from photochemical production and photoprodcution rates were correlated to illuminance. The persistent nighttime NO supersaturation pointed to an unidentified NO dark production. NO sea-to-air flux densities were much lower than the NO photoproduction rates. Therefore, we conclude that the bulk of the NO produced in the mixed layer was rapidly consumed before its release to the atmosphere.

删除的内容: pathways

删除的内容: mol·

删除的内容: mol·

删除的内容: mol·

删除的内容: ,

删除的内容: so far unknown,

删除的内容: NO dark production.

删除的内容: Overall, the oceanic NO emissions to the atmosphere were negligible compared to anthropogenic $NO_x$ sources such as emissions from ships.

## Introduction

Nitric oxide (NO) is a short-lived intermediate of the oceanic nitrogen cycle (Bange, 2008). It received limited attention so far because its determination in seawater is challenging (Zafiriou and McFarland, 1980; Lutterbeck and Bange, 2015; Liu et al., 2017). NO in surface seawater can be produced via the photolysis of nitrite ($NO_2^-$) (Zafiriou and McFarland, 1981; Olasehinde et al., 2009; 2010; Liu et al., 2017):

$$NO_2^- + H_2O \xrightarrow{h\nu} NO + OH^-.$$

This reaction may account for 10% of nitrite loss in surface waters of the central equatorial Pacific (Zafiriou et al., 1980). Zafiriou and McFarland (1981) suggested that photochemically produced NO is a potential source of atmospheric NO during daylight, which could further lead to ozone hole, acid precipitation and photochemical smog. Apart from the photochemical production, various microbial pathways of NO have been identified including denitrification, nitrification and anammox (Schreiber et al., 2014; Martens-Habbena et al., 2015; Caranto and Lancaster, 2017; Kuypers et al., 2018). Additionally, NO is a messenger molecule in marine organisms: phytoplankton does not only response to exogenous NO (Zhang et al., 2005), but also produce NO during their growth (Zhang et al., 2006a, b; Kim et al., 2006, 2008). Chen et al. (2015) reported that calmodulin (a messenger protein expressed in eukaryotic cells) of the tropical sea cucumber participates in the production of NO during immune response. Morrall et al. (1998; 2000) characterized the NO synthase activity in the tropical sea anemone *Aiptasia pallida*, suggesting that NO and NO synthase can act as ecotoxicological biomarkers in the tropical marine environment. Moreover, the characterization of NO synthase gene in the intertidal copepod *Tigriopus japonicas* has been found and the intracellular production of NO in shrimp haemocytes has been observed (Xian et al., 2013; Jeong et al., 2016). Thus, NO seems to be widespread with different functions in marine organisms.

删除的内容:

删除的内容: .

The current understanding of the oceanic NO distribution is mainly limited to the ocean surface (Zafiriou and McFarland, 1981; Olasehinde at al., 2009; 2010; Liu et al., 2017) and oxygen minimum zones (Ward and Zafiriou, 1988; Lutterbeck et al., 2018). Only recently, the distribution of NO as well as its seasonal variation in the Jiaozhou Bay and adjacent waters were studied (Feng et al., 2011; Xue et al., 2012; Tian et al., 2016).

删除的内容: C

In this study, we present first measurements of dissolved NO in the Yellow Sea (YS) and the Bohai Sea (BS). The overarching objective of our study was to decipher the biogeochemical fluxes of NO in the BS and YS. The specific objectives were (i) to determine the spatial variation of dissolved NO concentrations in the water column, (ii) to determine NO photoproduction rates, and (iii) to estimate the sea-to-air gas exchange fluxes of NO.

## 2 Materials and methods

### 2.1 Study area

The BS and the YS are marginal seas of the western Pacific Ocean. The BS is a shallow and almost enclosed sea with a surface area of $77\times10^3$ km$^2$ and a volume of $1.39\times10^3$ km$^3$. Its maximum depth is 83 m with an average depth of only 18 m. The Huanghe River is a major source of freshwater to the BS whereas the YS is the source of salt for the BS through water exchange via the Bohai Strait. The YS has a surface area of $380\times10^3$ km$^2$ and total volume of $16.7\times10^3$ km$^3$. Maximum depth is 140 m with an average depth of 44 m, and like the BS, it is a continental shelf sea. The Yangtze River at the southwest corner of the YS is the major source of freshwater for the southern YS and the East China Sea. The hydrographic properties of this region are mainly influenced by the Yellow Sea Cold Water Mass (YSCWM) on the shelf (Lü et al., 2010; Li et al., 2016) and the Yellow Sea Coastal Currents on the western side of the basin (Su, 1998; Lee et al., 2002; Zhang et al., 2004). The BS and YS are regions surrounded by areas of high population growth and economic development in China and Korea (Zhan et al., 2010; Jiang et al., 2014).

### 2.2 Sampling

Samples were collected from 13 to 28 June 2011 on board of the R/V "Dong Fang Hong 2" in the BS and the YS. Sampling for NO from both the surface (at 1 m) and the bottom layer (= 1 m above the ocean bottom) were performed at 75 sampling stations (including one 24 h anchor station: B65) shown in Fig. 1 and listed in Table 1. Water samples were collected using 8-liter Niskin bottles equipped with silicon O-rings and Teflon-coated springs and mounted on a Sea-Bird CTD (conductivity, temperature, depth) instrument (Sea-Bird Electronics, Inc., USA). A 500 mL Wheaton glass serum bottle was rinsed with in situ seawater three times, and then was filled with seawater quickly through a siphon. When the overflowed sample reached the half volume of the bottle, the siphon was withdrawn rapidly, 0.5 mL saturated $HgCl_2$ (aq) solution was added, and the bottle was sealed quickly. The surface water samples were immediately analyzed after collection, and samples from other depths were temporarily placed in dark in a water bath with a surface seawater circulation system and analyzed within 1 h (Liu et al., 2017). Photoproduction rates were determined in surface water samples from 22 stations in the YS and 4 stations in the BS (shown in Figure 1), respectively.

删除的内容: our

删除的内容: (H01, H05, H08, H10, H17, H21, H25, H29, H31, H35, H37, H39, H42 B01, B05, B10, B12, B15, B18, B23, B27, and B35)

删除的内容: (B42, B47, B51, and B68)

### 2.3 Analytical procedures

Analysis for dissolved NO was conducted with the improved method of Liu et al. (2017) by a combined purge-and-trap and fluorometric detection method. The precision of the analytical method was better than ± 7% and the limit of detection (LOD) was 32 pmol $L^{-1}$ in field experiment (Liu et al., 2017).

删除的内容:

The photolysis experiments were conducted under natural light conditions on deck. Surface water samples were filtered with 0.45 μm Millipore membrane and transferred into 10 mL cleaned quartz vials wrapped with Al foil with no headspace. Then 200 μL $NaN_3$ solutions were added to remove the microbial influence. After adding 10 μL of $1\times10^{-3}$ mol $L^{-1}$ 2, 3-diaminonaphthalene (DAN, trap NO) solution and gentle mixing, the fluorescence of the mixed solution was measured before irradiation (Liu et al., 2017). Then the same sample without addition of DAN were capped with a Teflon-lined silicone septum and without Al foil, the vial was placed in a shallow circulating seawater bath. After 0.5 h irradiation time, 10 μL of $1\times10^{-3}$ mol $L^{-1}$ DAN were added and the fluorescence of the solution was analyzed. The NO concentrations were measured with the method described in Liu et al., (2017). The NO photolysis production rates were computed based on the time-dependent difference between the NO concentrations before and after irradiation. For dark controls, vials were wrapped in Al-foil. The quartz vials and syringe used in the experiment were soaked in a 10% (v/v) HCl bath for 24 h, rinsed with Milli-Q water and baked at 500°C for 4 h.

删除的内容: the addition

删除的内容: of

删除的内容: mol·

删除的内容: T

删除的内容: after irradiation for 0.5 hour

删除的内容:

删除的内容:

删除的内容: ours

Chlorophyll *a* (Chl-*a*) was fluorometrically measured by an F-4500 fluorescence spectrophotometer after filtration of 200 mL seawater through a Whatman glass fiber filter and extraction in 90% acetone according to Strickland and Parsons (1968). The wind speeds were measured at a height of 10 m above the sea surface using a Model 27600-4X ship-borne weather instrument

删除的内容:

(Young, USA). Illuminance was measured by a digital illuminometer (TES-1330A, Shenzhen, China). Dissolved oxygen (DO) was measured in discrete water samples by the Winkler method (Grasshoff et al., 2009). The concentrations of dissolved inorganic nitrogen (nitrate, nitrite, and ammonium) was analyzed using a nutrient automatic analyzer (Auto Analyzer 3, SEAL Analytical, USA) in the laboratory. The detection limits were 0.14 μmol $L^{-1}$ for nitrate, nitrite, and ammonium, with the precision of the method better than 3% (Liu et al., 2005).

删除的内容: were

删除的内容: mol·

删除的内容: exceeding

删除的内容:

## 2.4 Calculation of NO flux and NO saturation

Fluxes of NO across the sea-to-air interface were estimated following the approach of McGillis et al. (2000) for a sparingly soluble gas which is also moderately reactive in the atmosphere:

$F = k_{sea} (c_{\mathrm{sea}} - p\mathrm{NO}_{\mathrm{air}} \times H^{cp})$,

here $F$ stands for the flux density (mass area$^{-1}$ time$^{-1}$) across the air-sea interface, $k_{sea}$ is the gas transfer velocity (length time$^{-1}$), and $c_{\mathrm{sea}}$ is the measured concentration of NO in the surface seawater (mass volumn$^{-1}$). The partial pressure of the atmospheric NO ($p\mathrm{NO}_{\mathrm{air}}$) was calculated as:

$p\mathrm{NO}_{\mathrm{air}} = x'\mathrm{NO}_{\mathrm{air}} \times (p_{\mathrm{ss}} - p_{\mathrm{w}})$,

where $x'\mathrm{NO}_{\mathrm{air}}$ is the mole fraction of atmosphere NO (dimensionless). We used the value of 2.13 ppb for $x'\mathrm{NO}_{\mathrm{air}}$ which is the average atmospheric NO mole fraction over the YS (Hu Min, Peking University, personal communication, 2018). $p_{\mathrm{ss}}$ is the barometric pressure at sea surface which was set to 1 atm as the average pressure and $p_{\mathrm{w}}$ is the water vapor pressure at sea surface which was calculated after Weiss and Price (1980):

删除的内容: And

$\ln p_{\mathrm{w}} = 24.4543 - 6745.09 / (T + 273.15) - 4.8489 \times \ln (T + 273.15) / 100) - 0.000544 \times S)$.

$H^{cp}$ is the Henry's law constant which is calculated as:

$H^{cp}(\mathrm{T}) = H^{\ominus} \times \exp (-\Delta \mathrm{sol}\, H / R \times (1 / T - 1 / T^{\ominus})$

where $-\Delta sol \frac{H}{R} = \frac{d\ln H}{d\ln(\frac{1}{T})}$.

$H^{\ominus}$ and $-\Delta \mathrm{sol}\, H / R$ are tabulated in Sander (2015).

删除的内容:

删除的内容:

删除的内容:

删除的内容: ,

$k_{sea}$ was calculated as:

$k_{sea} = k_{\mathrm{w}} (1 - \gamma_{\mathrm{a}})$,

$\gamma_{\mathrm{a}} = 1 / (1 + (k_{\mathrm{a}} / (H^{cc} \times k_{\mathrm{w}}))$,

$H^{cc} = H^{\mathrm{cp}} \times RT$,

$k_{\mathrm{a}} = 659 \times u \times (\mathrm{M}_{\mathrm{NO}} / \mathrm{M}_{\mathrm{H_2O}})^{-1/2}$,

$k_{\mathrm{w}} = 0.251 \times u^2 \times (\mathrm{Sc} / 660)^{-1/2}$,

where $k_{\mathrm{w}}$ is the water side air-sea gas transfer coefficient for sparingly soluble gases (length time$^{-1}$) calculated according to Wanninkhof (2014), $\gamma_{\mathrm{a}}$ is the fraction of the entire gas concentration gradient across the airside boundary layer as a fraction of the entire gradient from the bulk water to the bulk air (dimensionless) (McGillis et al., 2000), $k_{\mathrm{a}}$ is the air side air-sea gas transfer coefficient (length time$^{-1}$) according to McGillis et al. (2000), $H^{cc}$ is the Henry coefficient (dimensionless) (Sander, 2015 ), $M_{NO}$ and $M_{H_2O}$ are relative molecular mass of NO and $H_2O$ (dimensionless), and $u$ is the wind speed at 10 m height under neutral boundary conditions (length time$^{-1}$).

The Schmidt number ($Sc$) is the kinematic viscosity of water divided by the molecular diffusion coefficient of the gas in (sea)water (Jähne et al., 1987; Wanninkhof, 2014). Seawater dynamic viscosity ($\mu_{sw}$) is a function of temperature ($T$) and salinity ($S$) and was estimated using the following equations (Sharqawy et al., 2010):

$\mu_{sw} = \mu_w\ (1 + A\ S + B\ S^2\ )$,

$A = 1.541 + 1.998\times10^{-2}\ T - 9.52\times10^{-5}\ T^2$,

$B = 7.974 - 7.561\times10^{-2}\ T + 4.724\times10^{-4}\ T^2$,

$\mu_w = 4.2844\times10^{-5} + (0.157\ (T + 64.993)^2 - 91.296)^{-1}$.

Seawater density was estimated using Millero's empirical equation (Millero et al., 1976), and NO diffusion coefficient $D_L$ ($\times 10^{-5}$ cm$^2$ s$^{-1}$) in water was calculated according to Wise and Houghton (1968):

$D_L = 0.9419\ \exp(0.0447\ T)$.

The saturation factor ($\alpha$) is defined as $\alpha = c_{sea}\ /\ (p\mathrm{NO}_{air}\times H^{cp})$, $\alpha > 1$ represents NO was supersaturated and the flux was from sea to air.

## 3 Results and Discussion

### 3.1 NO in the surface and bottom layers

The NO concentrations from the surface and bottom layers of the BS and the YS as well as the local sampling time, bottom depth (D), temperature (T), salinity (S), Chl-*a*, wind speed (u), and DO are listed in Table 1. In the study area, temperature varied from 8.8 to 21.7°C and salinity varied from 29.57 to 32.73‰ in the surface water. DO fluctuated from 178 to 271 μmol L$^{-1}$. The average concentrations of $NH_4^+$-N, $NO_2^-$-N and $NO_3^-$-N were 2.11μmol L$^{-1}$, 0.20 μmol L$^{-1}$, and 2.59 μmol L$^{-1}$, respectively. While in the bottom layer, temperature varied from 3.8 to 21.6°C, salinity varied from 30.00 to 33.71‰, and DO varied from 147 to 266 μmol L$^{-1}$.

The surface concentrations of NO ranged from below the LOD to 616 pmol L$^{-1}$ with an overall average of 186 ± 108 pmol L$^{-1}$ and exhibited a considerable spatial variability (Fig. 2a). The mean NO surface concentrations in the BS (203 ± 107 pmol L$^{-1}$) and northern YS (NYS) (212 ± 130 pmol L$^{-1}$) were higher than the mean NO concentration in the southern YS (SYS) (159 ± 84 pmol L$^{-1}$). The NO concentrations in the bottom layer ranged from below the LOD to 482 pmol L$^{-1}$, with an overall average of 174 ± 123 pmol L$^{-1}$. The mean concentrations of NO in the bottom layers of the BS and NYS were 228 ± 116, 210 ± 138, respectively and were higher than the mean (127 ± 98 pmol L$^{-1}$) for the SYS. The maximum NO surface and bottom concentrations were measured at stations B21 and B28 in the NYS, respectively (Fig. 2b), whereas the lowest NO surface and bottom concentrations were measured in the center of the SYS (Table 1). Overall, there were no statistically significant differences between the mean NO concentrations in the surface and bottom layers.

带格式的：字体：倾斜

带格式的：字体：倾斜

带格式的：字体：倾斜

带格式的：字体：倾斜

带格式的：字体：倾斜

删除的内容：mol·

删除的内容：mol·

删除的内容：mol·

删除的内容：mol·

删除的内容：mol·

NO surface concentrations did not show any statistically significant relationship with sea surface temperature, salinity, DO, Chl-*a*, and illuminance. Trends of NO concentrations with salinity and DIN were only found for the stations affected by the outflow of the Huanghe River in the southern BS (Fig. 3) where we found an inverse relationship between salinity and NO surface concentrations. Here high NO concentrations were associated with high DIN concentrations (data not shown, see Liu et al., 2015; Yang et al, 2015) indicating that DIN, especially $NO_2^-$, was a prerequisite for enhanced NO concentrations.

The water columns of the BS and YS were well-oxygenated during our study and, thus, no suboxic or anoxic conditions were detected. Therefore, we did not found any enhancement of NO concentrations in the bottom layers with low DO or $NO_2^-$ concentrations as observed in the OMZ of the eastern tropical South Pacific Ocean off Peru (Lutterbeck et al., 2018). However, NO concentrations in the bottom layer showed negative correlations with salinity ($p < 0.05$, $r = -0.272$, $n = 60$) and bottom depth ($p < 0.05$, $r = -0.298$, $n = 61$) indicating a decrease of NO concentrations from the coast toward offshore waters. It is known that both the BS and YS are affected by submarine ground water discharge (Kim et al., 2005; Liu et al., 2017a; 2017b; Taniguchi et al. 2008). Moreover, NO has been detected in groundwater and aquifers (Smith et al., 2004, Smith and Yoshinari, 2008). Therefore, we suggest that input of NO from submarine groundwater discharge contributed to the distribution of NO bottom concentrations as well.

An overview of published NO surface concentrations is given Table 3. The average NO surface and bottom concentrations from this study are comparable to the concentrations measured in the Jiaozhou Bay, in the waters off Qingdao and in the Seto Inland Sea. However, our mean concentrations are considerably higher than the NO concentrations reported from the central equatorial Pacific and then eastern tropical North Pacific Ocean. The maximum concentration reported here is at the lower end of the NO concentrations recently reported from the anoxic oxygen minimum zone off Peru. Overall, NO surface concentrations seem to be generally higher in coastal waters compared to those found in offshore waters.

### 3.2 Diurnal variability

The diurnal variability of surface NO concentrations, illuminance, DO, and Chl-*a* were investigated at the anchor station B65 (Fig. 4). NO concentrations varied from 64 to 424 pmol $L^{-1}$, exhibiting a significant diurnal variation with the maximum concentration eightfold higher than the minimum concentration. The NO concentrations reached the maximum concentration in the early afternoon (about 13:00 local time, LT) and then decreased to the minimum concentration at 22:00 LT. A less pronounced second maximum (209 pmol $L^{-1}$) was reached at 04:00 LT. DO showed a similar diurnal cycle but shifted by 3 h with maxima at 16:00 LT and 07:00 LT. Chl-*a* concentrations peaked at 19:00 LT and 07:00 LT. The illuminance had its maximum at 13:00 LT coinciding with the NO maximum indicating that the first NO maximum was indeed resulting from a photochemical production during daytime (Zafiriou and McFarland, 1981). However, the second maximum of NO at 04:00 when it was still dark, must have resulted from an alternative chemical and/or biological production.

删除的内容: depth,

删除的内容: was

删除的内容: H

带格式的: 字体: 倾斜

删除的内容: *R*

带格式的: 字体: 倾斜

删除的内容: 258

删除的内容: p < 0.05, R = -0.292

删除的内容: NO

删除的内容: mol·

删除的内容: three

删除的内容: ours

删除的内容: h

### 3.3 Photoproduction rates

The results of the NO photoproduction experiments are listed in Table 2. The photoproduction rates of NO in the BS, the NYS, and the SYS varied from not detectable to 5.07×$10^{-11}$ mol $L^{-1}$ $s^{-1}$, 0.09 to 0.69×$10^{-11}$ mol $L^{-1}$ $s^{-1}$, and 0.32 to 1.54×$10^{-11}$ mol $L^{-1}$ $s^{-1}$, respectively. The average photoproduction rate of the whole study area was 1.14 ± 1.37×$10^{-11}$ mol $L^{-1}$ $s^{-1}$. The photoproduction rates from the BS and YS are in good agreement with the rates reported from Seto Inland Sea (Olasehinde et al, 2009; 2010). However, the mean NO photoproduction rates are higher than those from the central equatorial Pacific Ocean (Zafiriou and McFarland, 1981) and lower than those from Kurose River, Japan (Olasehinde et al, 2009) (Table 2) which mirrors the available $NO_2^-$ concentrations which are low in the open ocean and but high in a river.

The average photoproduction rate in the NYS was obviously lower than those in the BS and the SYS, consistent with the average illuminances of 22450, 20433, 27852 lx and the average mean surface downward UV radiation flux 34.7, 32.1, 40.6 W $m^{-2}$ (ECMWF reanalysis data) for the BS, the NYS and the SYS, respectively. Enhanced photoproduction rates (≥ 2.00 ×$10^{-11}$ mol $L^{-1}$ $s^{-1}$) occurred in the SYS, especially in the central part of the SYS and stations influenced by the Yellow Sea Cold Current (see Fig. 1). This is apparently in contrast to the distribution of NO which showed lowest concentrations in the central SYS (see above). The high illumination observed in the SYS (Table 2) does not only lead to enhanced NO photoproduction (see Table 2) but would also generate reactive oxygen species like $O_2$•-, ROO•, and other OH-derived radicals, which in turn, would efficiently scavenge NO (Olasehinde et al, 2010). Overall, the NO photoproduction rates showed a positive relationship with illuminance ($p < 0.01$, $r = 0.884$, $n = 26$) and the mean surface downward UV radiation flux ($p < 0.01$, $r = 0.865$, $n = 26$) indicating that the NO concentrations in the surface layer during daylight were dominated by photochemical production. However, we did not find a significant difference between the mean NO concentrations sampled during day (179 ± 80 pmol $L^{-1}$) and night (195 ± 140 pmol $L^{-1}$). This suggests that there was also a non-photochemical NO dark production in the surface layer like nitrification process or other chemical process like the process of ammonium ($NH_4^+/NH_3$) oxidation into $NO_2^-$ and $NO_3^-$ (Joussotdubien and Kadiri, 1970). Caranto and Lancaster (2017) found that NO is an obligate bacterial nitrification intermediate produced by hydroxylamine oxidoreductase, Ward and Zafiriou (1988) also found that NO might play as an intermediate of a soluble byproduct of nitrification such as hydroxylamine.

Besides, Olasehinde et al. (2010) found that filtered and unfiltered seawater samples collected from the Seto Inland Sea showed no significant difference in NO photoformation rates, which suggested a negligible contribution of NO produced by photobiological processes from particle matter in seawater. However, Liu et al. (2017) reported that the rates difference between filtered (0.45 μm, 1.46×$10^{-12}$ mol $L^{-1}$ $s^{-1}$) and unfiltered (1.52×$10^{-12}$ mol $L^{-1}$ $s^{-1}$) seawater samples from coastal waters of the YS indicated that particles in seawater could increase the NO production rate. The difference might be due to the composition of sample, filter membrane, etc. Thus, further research is needed.

**删除的内容:** 0.00

**删除的内容:** mol·

**删除的内容:** mol·

**删除的内容:** mol·

**删除的内容:** mol·

**删除的内容:** , which were

**删除的内容:** and

**删除的内容:** 2

**删除的内容:** S

**删除的内容:** mol·

**删除的内容:** southern

**删除的内容:** can

**带格式的:** 字体：倾斜

**删除的内容:** *R*

**带格式的:** 字体：倾斜

**删除的内容:** was

**带格式的:** 非上标/ 下标

### 3.4 Sea-to-air fluxes of NO

In the present study, the NO supersaturation was ubiquitous at all investigated sites. The supersaturation factors varied from 8 to 154, with an average of 47. Together with the fact that that NO was supersaturated during daytime and nighttime during the 24 h station B65 this indicates that the BS and YS were a source of NO to the atmosphere. The sea-to-air flux densities ranged from $5.8\times10^{-19}$ to $3.6\times10^{-15}$ mol cm$^{-2}$ s$^{-1}$, with an average value of $4.5\times10^{-16}$ mol cm$^{-2}$ s$^{-1}$ (Fig. 7). The comparison of wind speeds and flux densities reveal that the flux densities are mainly driven by the wind speed (Fig. 7) and they showed significant positive relationship (SPSS v.16.0). Anifowose and Sakugawa (2017) also found that wind speed was an important factor governing NO flux at the air-sea interface. Our flux densities from the BS and YS were similar to those computed for the central equatorial Pacific Ocean and the Seto Inland Sea, while they are slightly lower than those computed for of Jiaozhou Bay waters (Table 2). Based on the YS area of $380\times10^3$ km$^2$ and the BS area of $77\times10^3$ km$^2$, the emission of NO to the atmosphere was estimated to be $9.0\times10^8$ g N yr$^{-1}$ or 7.6 $\times10^7$ g N month$^{-1}$. Ding et al. (2018) report a satellite-derived average NO$_x$ emission estimate for June over the BS and the YS in the period from 2007 to 2016 of about 1.3 $\times10^{10}$ g N month$^{-1}$. The obvious very large discrepancy between the satellite-derived emission estimate and the one presented here results from the fact that Ding et al.'s (2018) estimate is dominated by the NO$_x$ emissions from ships' diesel engines. This indicates that oceanic NO emissions to the atmosphere only account for a negligible fraction (~0.6%) of the NO$_x$ emissions observed over the BS and YS.

### 3.5 NO mixed layer budget

In order to estimate the contribution of different source and sinks of NO in surface layer of the BS and YS we applied a simple box model. We assume that the surface layer is represented by the mixed layer with a mean water depth of 15 m depth (Qiao et al., 2004). At steady state the loss of NO by air-sea exchange ($F_{ase}$) must be equal to the sum of the photoproduction rate ($F_{pp}$) in the mixed layer and the input from below into the mixed layer by diapycnal diffusion ($F_{dia}$) and other production or consumption pathways ($F_{poc}$) and advection into or out of the BS/YS mixed layer ($F_{adv}$). To this end, $F_{ase}$ is given by

$$F_{ase} = F_{pp} + F_{dia} + F_{poc} + F_{adv}.$$

The mean $F_{ase}$ was $4.5\times10^{-16}$ mol cm$^{-2}$ s$^{-1}$ (see above). We assume that the NO photoproduction decreases linearly from the average rate of $1.15 \pm 1.47 \times10^{-11}$ mol L$^{-1}$ s$^{-1}$ at the surface (see above) to 0 at 15 m. The mean $F_{pp}$ was calculated to be $8.6\times10^{-12}$ mol cm$^{-2}$ s$^{-1}$ in mixed layer. Since the mean surface NO concentration in the surface layer was statistically not different from the mean NO concentration in the bottom layer (see above) it is reasonable to assume that $F_{dia}$ = 0. Moreover, it seems reasonable to assume that advection of NO into or out of the BS/YS surface layer is zero. Since $F_{ase} < F_{pp}$ we conclude that $F_{poc}$ should be negative indicating that the bulk of the produced NO was rapidly consumed in the surface layer before its release to the atmosphere. Please note that we have some indications that there is also a dark production of NO (see above), so that the 'true' NO surface production might be even higher. Chemical reactions with DO, OH, or ROO• etc. are potential sinks for NO in the surface layer of the BS and YS (Ford et al., 1993; Olasehinde et al, 2010; Carpenter and Nightingale, 2015).

删除的内容: mol·
删除的内容: mol·
删除的内容: ·
删除的内容: ·
删除的内容: n
删除的内容: ·
删除的内容:
带格式的: 字体: 倾斜
带格式的: 字体: 倾斜
带格式的: 字体: 倾斜
带格式的: 字体: 倾斜
带格式的: 字体: 倾斜
带格式的: 字体: 倾斜
带格式的: 字体: 倾斜
带格式的: 字体: 倾斜
带格式的: 字体: 倾斜
带格式的: 字体: 倾斜
带格式的: 字体: 倾斜
带格式的: 字体: 倾斜
删除的内容: mol·
删除的内容: mol·
带格式的: 字体: 倾斜
删除的内容: mol·
带格式的: 字体: 倾斜
带格式的: 字体: 倾斜
带格式的: 字体: 倾斜
带格式的: 字体: 倾斜

## 4. Conclusions

This study reports the distribution and photoproduction rates of dissolved NO measured during a cruise in June 2011 to the Bohai and Yellow Seas. The NO concentration distribution in both the surface and bottom layers were highly variable however there was no significant difference between the mean NO concentrations in the surface and bottom layers. NO concentrations in the bottom layer showed significant decreasing with salinity indicates NO input by submarine groundwater discharge. NO in the surface layer was supersaturated at all stations during day and night. The accumulation of NO during daytime was resulting from photoproduction and the measured NO photoproduction rates were correlated to illuminance. The persistent nighttime NO supersaturation pointed to a non-photochemical (so far unidentified chemical and/or biological) NO dark production. On the basis of a simple box model calculation we conclude that the bulk of the NO produced in the surface layer was rapidly consumed before its release to the atmosphere. Overall, the BS and YS were a persistent source of NO to the atmosphere at the time of our measurements. However, the oceanic NO emissions were negligible compared to the $NO_x$ emissions from ships diesel engines.

*Data availability.* The data can be accessed by email request to the corresponding authors and have been submitted to the open-access database PANGAEA (data submission 25 September 2019, https://issues.pangaea.de/browse/PDI-21749).

*Author contributions.* YT, CX, CL, GY, PL and WF prepared the original manuscript and designed the experiments; HWB made many modifications and gave a lot of suggestions on design of figures and the computing method. All authors contributed to the analysis of the data and discussed the results.

*Competing interests.* The authors declare that they have no conflict of interest.

*Acknowledgements.* We thank the captain and crew of the R/V “Dong Fang Hong 2” for their support and help during the cruise. We thank the editor and two reviewers for their thoughtful feedback on the manuscript.

*Financial support.* This research was supported by the National Natural Science Foundation of China (Nos.41676065 and 40706040), the National Key Research and Development Program of China (Grant No. 2016YFA0601301), and the Fundamental Research Funds for the Central Universities (No. 201762032).

## References

Anifowose, A. J., and Sakugawa, H.: Determination of Daytime Flux of Nitric Oxide Radical (NO•) at an Inland Sea-Atmospheric Boundary in Japan, J. Aquat. Pollut. Toxicol., 1(2), 1-6, 2017.

Bange, H.W.: Chapter 2 - Gaseous Nitrogen Compounds (NO, $N_2O$, $N_2$, $NH_3$) in the Ocean. Nitrogen in the Marine Environment (Second Edition), Elsevier, Amsterdam, Netherlands, 51-94, 2008.

**删除的内容:** s

**删除的内容:** . T

**删除的内容:** ,

**删除的内容:** so far unknown, non-photochemical (chemical and/or biological) NO dark production.

**删除的内容:** **Acknowledgements** .
We thank the captain and crew of the R/V “Dong Fang Hong 2” for their support and help during the cruise. This research was supported by the National Natural Science Foundation of China (Nos.41676065 and 40706040), the National Key Research and Development Program of China (Grant No. 2016YFA0601301), the Fundamental Research Funds for the Central Universities (No. 201762032), and the AoShan Talents Program of Qingdao .

**带格式的:** 英语(英国)

Caranto, J.D. and Lancaster, K.M.: Nitric oxide is an obligate bacterial nitrification intermediate produced by hydroxylamine oxidoreductase, PNAS., 114, 8217, https://doi.org/10.1073/pnas.1704504114, 2017.

Carpenter, L.J. and Nightingale, P.D.: Chemistry and Release of Gases from the Surface Ocean. Chemical Reviews, Chem. Rev., 115(10), 4015-4034, https://doi.org/10.1021/cr5007123, 2015.

Chen, T., Ren, C., Li, W., Jiang, X., Xia, J., Wong, N.K., and Hu, C.: Calmodulin of the tropical sea cucumber: gene structure, inducible expression and contribution to nitric oxide production and pathogen clearance during immune response, Fish Shellfish Immunol., 45(2), 45(2), 231-238. https://doi.org/10.1016/j.fsi.2015.04.013, 231-238, 2015.

Dang, H., Zhou, H., Zhang, Z., Yu, Z., Hua, E., Liu, X., and Jiao, N.Z.: Molecular detection of *Candidatus Scalindua* pacifica and environmental responses of sediment anammox bacterial community in the Bohai Sea, China, Plos One, 8, e61330, https://doi.org/10.1371/journal.pone.0061330, 2013.

Ding, J. R., Ronald, V.D.A., Mijling, B., Jalkanen, J.P., Johansson, L. and Levelt, P.F.: Maritime NOx emissions over Chinese seas derived from satellite observations, Geophys. Res. Lett., 45(9), https://doi.org/10.1002/2017gl076788, 2018.

Feng, W.H., Liu, C.Y., Yang, G.P., Li, P.F., and Xue, C.: Chemiluminescene detection of nitric oxide in seawater, Acta Oceanol. Sin., 33 (6), 93-98, 2011. (In Chinese with English abstract)

Ford, P.C., Wink, D.A., Stanbury, D.M.: Autoxidation kinetics of aqueous nitric oxide, Febs Lett., 326(1-3):1, https://doi.org/10.1016/0014-5793(93)81748-o, 1993,

Grasshoff, K., Kremling, K., and Ehrhardt, M.: Methods of seawater analysis, John Wiley & Sons, New York, American, 2009.

Jeong, C.B., Kang, H.M., Seo, J.S., Park, H.G., Rhee, J.S., and Lee, J.S.: Identification and molecular characterization of nitric oxide synthase (NOS) gene in the intertidal copepod Tigriopus japonicas, Gene, 577(1), 47, https://doi.org/10.1016/j.gene.2015.11.019, 2016.

Jiang, X., Teng, A., Xu, W., and Liu, X.: Distribution and pollution assessment of heavy metals in surface sediments in the Yellow Sea, Mar. Pollut. Bull., 83 (1), 366-375, https://doi.org/10.1016/j.marpolbul.2014.03.020, 2014.

Joussotdubien, J., and Kadiri, A.: Photosensitized Oxidation of Ammonia by Singlet Oxygen in Aqueous Solution and in Seawater, Nature, 227, 700-701, https://doi.org/10.1038/227700b0, 1970.

Jähne, B., Heinz, G., and Dietrich, W.: Measurement of the diffusion coefficients of sparingly soluble gases in water, J. Geophys. Res.: Oceans, 92(C10):10767-10776, https://doi.org/10.1029/jc092ic10p10767, 1987.

Kim, D., Yamaguchi, K., and Oda, T.: Nitric oxide synthase-like enzyme mediated nitric oxide generation by harmful red tide phytoplankton, J. Plankton Res., 28(6), 613-620, https://doi.org/10.1093/plankt/fbi145, 2006.

Kim, D., Yang, S.K., Lee, Y., Yamaguchi, K., Matsuoka, K., Lee, K.W., Choi, K.S., and Oda, T.: Detection of nitric oxide (NO) in marine phytoplankters, J. Biosci. Bioeng., 105 (4), 414-417, https://doi.org/10.1263/jbb.105.414, 2008.

Kim, G., Ryu, J. W., Yang, H. S., and Yun, S. T.: Submarine groundwater discharge (SGD) into the Yellow Sea revealed by 228 Ra and 226 Ra isotopes: Implications for global silicate fluxes, Earth Planet, Sci. Lett., 237 (1-2), 156-166, https://doi.org/10.1016/j.epsl.2005.06.011, 2005.

Kuypers, M.M.M., Marchant, H.K., and Kartal, B.: The microbial nitrogen-cycling network, Nat. Rev. Microbiol., 16 (5), 263-276, https://doi.org/10.1038/nrmicro.2018.9, 2018.

Lee, H.J., Jung, K.T., So, J.K., and Chung, J.Y.: A three-dimensional mixed finite-difference Galerkin function model for the oceanic circulation in the Yellow Sea and the East China Sea in the presence of $M_2$ tide, Cont. Shelf Res., 22 (1), 67-91, https://doi.org/10.1016/s0278-4343(01)00068-1, 2002.

Li, J., Li, G., Xu, J., Dong, P., Qiao, L., Liu, S., Sun, P., and Fan, Z.: Seasonal evolution of the Yellow Sea Cold Water Mass and its interactions with ambient hydrodynamic system, J. Geophys. Res., 121 (9), 6779-6792, https://doi.org/10.1002/2016jc012186, 2016.

Liu, C.Y., Feng, W.H., Tian, Y., Yang, G.P., Li, P.F., and Bange, H.W.: Determination of dissolved nitric oxide in coastal waters of the Yellow Sea off Qingdao, Ocean Sci., 13(4), 1-31. https://doi.org/10.5194/os-2017-10, 2017.

Liu, J., Du, J., and Yi, L.: Ra Tracer-Based Study of Submarine Groundwater Discharge and Associated Nutrient Fluxes into the Bohai Sea, China: A Highly Human-Affected Marginal Sea, J. Geophys. Res., 122 (1), 139-152, 2017a.

Liu, J. A., Su, N., Wang, X., and Du, J.: Submarine groundwater discharge and associated nutrient fluxes into the Southern Yellow Sea: A case study for semi-enclosed and oligotrophic seas-implication for green tide bloom, J. Geophys. Res.: Oceans, 122 (11), 8646-8660, https://doi.org/10.1002/2017jc013095, 2017b.

Liu, S.M., Zhang, J., Chen, H.T., and Zhang, G.S.: Factors influencing nutrient dynamics in the eutrophic Jiaozhou Bay, North China, Prog. Oceanogr., 66 (1), 66-85, 2005.

Liu, S.M.: Response of nutrient transports to water-sediment regulation events in the Huanghe basin and its impact on the biogeochemistry of the Bohai, J Marine Syst., 141, 59-70, https://doi.org/10.1002/2016jc012282, 2015.

Lutterbeck, H.E. and Bange, H.W.: An improved method for the determination of dissolved nitric oxide (NO) in seawater samples, Ocean Sci., 11 (6), 959-981, https://doi.org/10.5194/osd-12-959-2015, 2015.

Lutterbeck, H.E., Arévalo-Martínez, D.L., Löscher, C.R., and Bange, H.W.: Nitric oxide (NO) in the oxygen minimum zone off Peru, Deep-Sea Res. Part II, in press, 2018.

Lü, X., Qiao, F., Xia, C., Wang, G., and Yuan, Y.: Upwelling and surface cold patches in the Yellow Sea in summer: Effects of tidal mixing on the vertical circulation, Cont. Shelf Res., 30 (6), 620-632, https://doi.org/10.1016/j.csr.2009.09.002, 2010.

Martens-Habbena, W., Qin, W., Horak, R.E., Urakawa, H., Schauer, A.J., Moffett, J.W., Armbrust, E.V., Ingalls, A.E., Devol, A.H., and Stahl, D.A.: The production of nitric oxide by marine ammonia-oxidizing archaea and inhibition of archaeal ammonia oxidation by a nitric oxide scavenger, Environ. Microbiol., 17(7), 2261-2274, https://doi.org/10.1111/1462-2920.12677, 2015.

Mcgillis, W.R., Dacey, J.W.H., Frew, N.M., Bock, E.J., and Nelson, R.K.: Water-air flux of dimethylsulfide, J. Geophys. Res., 105(C1), 1187–1193, 2000.

Millero, F.J., Gonzalez, A., and Ward, G.K.: Density of seawater solutions at one atmosphere as a function of temperature and salinity, J. Mar. Res., 34(1), 61-93, 1976.

删除的内容: , 13 (4), 1-31

Morrall, C.E., Trapido-Rosenthal, H.G., Knap, A.H., and Depledge, M.H.: Development of nitric oxide and nitric oxide synthase as ecotoxicological biomarkers in the tropical marine environment, Mar. Environ. Res., 46, 429-432, https://doi.org/10.1016/s0141-1136(97)00080-9, 1998.

Morrall, C.E. and Galloway, T.S., Trapidorosenthal, H.G., Depledge, M.H.: Characterisation of nitric oxide synthase activity in the tropical sea anemone Aiptasia pallida, Comp. Biochem. Physiol., Part B: Biochem. Mol. Biol., 125 (4), 483-491, https://doi.org/10.1016/s0305-0491(00)00157-7, 2000.

Olasehinde, E.F., Takeda, K., and Sakugawa, H.: Development of an Analytical Method for Nitric Oxide Radical Determination in Natural Waters, Anal. Chem., 81 (16), 6843-6850. https://doi.org/10.1021/ac901128y, 6843-6850, 2009.

Qiao, F.L., Xia C.S., Shi J.W.: Seasonal variability of thermocline in the Yellow Sea, Chin. J. Oceanol. Limnol., 22 (3):299-305, 2004.

Sander, R.: Compilation of Henry's law constants (version 4.0) for water as solvent, Atmos. Chem. Phys., 14 (21), 29615-30521, https://doi.org/10.5194/acp-15-4399-2015, 2015.

Schreiber, F., Stief, P., Kuypers, M.M.M., and De Beer, D.: Nitric oxide turnover in permeable river sediment, Limnol. Oceanogr., 59 (4), 1310-1320, https://doi.org/10.4319/lo.2014.59.4.1310, 2014.

Sharqawy, M. H., V, J. H. L., and Zubair, S. M.: Thermophysical properties of seawater: a review of existing correlations and data, Desalination & Water Treatment, 16 (1-3), 354-380, https://doi.org/10.5004/dwt.2010.1079, 2010.

Smith, R. L., Böhlke, J. K., Garabedian, S. P., Revesz, K. M., and Yoshinari, T.: Assessing denitrification in groundwater using natural gradient tracer tests with 15 N: In situ measurement of a sequential multistep reaction, Water Resources Res, 40 (7), 1-17, https://doi.org/10.1029/2003wr002919, 2004.

Smith, R. L., and Yoshinari, T.: Occurrence and turnover of nitric oxide in a nitrogen-impacted sand and gravel aquifer, Environ. Sci. Technol., 42 (22), 8245, https://doi.org/10.1021/es801290v, 2008.

Strickland, J.D.H. and Parsons T.R.: A practical handbook of seawater analysis. Fisheries Research Board of Canada, Ottowa, 1968.

Su, J.: Circulation dynamics of the China seas: north of 18° N, The Sea., 6, 13-19, 1998.

Taniguchi, M., Ishitobi, T., Chen, J., Onodera, S. I., Miyaoka, K., Burnett, W. C., Peterson, R., Liu, G., and Fukushima, Y.: Submarine groundwater discharge from the Yellow River Delta to the Bohai Sea, China, J. Geophys. Res.: Oceans, 113, C06025, https://doi.org/10.1029/2007jc004498, 2008.

Tian, Y., Xue, C., Liu, C.Y., and Li, P.F.: Distribution and influencing factors of nitric oxide in the surface water of Jiaozhou Bay, Res. Environ. Sci., 29 (10), 1451-1458, 2016. (In Chinese with English abstract)

Wanninkhof, R.: Relationship between wind speed and gas exchange over the ocean revisited, Limnol. Oceanogr., 12, 351-362, https://doi.org/10.4319/lom.2014.12.351, 2014.

Ward, B.B. and Zafiriou, O.C.: Nitrification and nitric oxide in the oxygen minimum of the eastern tropical North Pacific, Deep-Sea Res., Part I, 35(7):1127-1142, https://doi.org/10.1016/0198-0149(88)90005-2, 1988.

Weiss, R.F. and. Price, B.A.: Nitrous oxide solubility in water and seawater, Mar. Chem., 8(4):347-359, https://doi.org/10.1016/0304-4203(80)90024-9, 1980.

Wise, D.L. and Houghton, G.: Diffusion coefficients of neon, krypton, xenon, carbon monoxide and nitric oxide in water at 10-60°C. Chem. Eng. Sci., 23, 1211-1216, https://doi.org/10.1016/0009-2509(68)89029-3, 1968.

Xian, J.A., Guo, H., Li, B., Miao, Y.T., Ye, J.M., Zhang, S.P., Pan, X.B., Ye, C.X., Wang, A.L., and Hao, X.M.: Measurement of intracellular nitric oxide (NO) production in shrimp haemocytes by flow cytometry, Fish Shellfish Immunol., 35, 2032-2039, https://doi.org/10.1016/j.fsi.2013.10.014, 2013.

Xue, C., Liu, C.Y., Yang G.P., Zhu, C.J., Chen, J., and Zhang, H.H.: Distribution and Controlling Factors of Nitric Oxide Concentrations in Surface Seawater of Jiaozhou Bay and Adjacent Waters, Chin. J. Environ. Sci., 33 (4), 1086-1090, 2012. (In Chinese with English abstract)

Yang, J., Yang G.P., Zhang, H.H., and Zhang, S.H.: Spatial distribution of dimethylsulfide and dimethyl-sulfoniopropionate in the Yellow Sea and Bohai Sea during summer, Chin. J. Oceanol. Limnol., 33 (4), 1020-1038, https://doi.org/10.1007/s00343-015-4188-5, 2015.

Zafiriou, O.C., Mcfarland, M., and Bromund, R.H.: Nitric oxide in seawater, Science, 207 (4431), 637-639, https://doi.org/10.1126/science.207.4431.637, 1980.

Zafiriou, O.C. and Mcfarland, M.: Nitric oxide from nitrite photolysis in the central equatorial Pacific, J. Geophys. Res.: Atmos., 86 (C4), 3173-3182, https://doi.org/10.1029/jc086ic04p03173, 1981.

Zhan, S., Peng, S., Liu, C., Chang, Q., and Jian, X.: Spatial and Temporal Variations of Heavy Metals in Surface Sediments in Bohai Bay, North China, Bull. Environ. Contam. Toxicol., 84 (4), 482-487, https://doi.org/10.1007/s00128-010-9971-6, 2010.

Zhang, J., Yu, Z.G., Raabe, T., Liu, S.M., Starke, A., Zou, L., Gao, H.W. and Brockmann, U.: Dynamics of inorganic nutrient species in the Bohai seawaters, J. Mar. Syst., 44 (3-4), 189-212, https://doi.org/10.1016/j.jmarsys.2003.09.010, 2004.

Zhang, Z.B., Lin, C., Liu, C., and Lei, X.: Study on patterns and chemical features of NO effect on marine phytoplankton growth. Sci. China: Chem., 48 (4), 376-384, https://doi.org/10.1360/03yb0166, 2005.

Zhang, Z.B., Xing, L., Wu, Z.Z., Liu, C.Y., Lin, C., and Liu, L.S.: Discovery of nitric oxide in marine ecological system and the chemical characteristics of nitric oxide. Sci. China: Chem., 49 (5), 475-480, https://doi.org/10.1007/s11426-006-2017-6, 2006a.

Zhang, Z.B., Liu, C.Y., Wu, Z.Z., Xing, L., and Li, P.F.: Detection of nitric oxide in culture media and studies on nitric oxide formation by marine microalgae, Med. Sci. Monit., 12, 75-85, 2006b.

**Figure Captions**

**Figure 1.** Locations of the sampling stations in the BS and the YS during summer. Solid dots (●) represent the stations for incubation experiments.

**Figure 2.** Horizontal distributions of NO (pmol L$^{-1}$) in the surface water and bottom water.

**删除的内容:** mol·

**Figure 3.** Variations of salinity, NO surface concentrations, and $NO_2^-$ concentrations from station B65 to station B70.

**Figure 4.** Diurnal variations of NO concentrations, illuminance (I), DO, and Chl-*a* concentrations in the surface water at the anchor station B65.

**Figure 5.** Wind speeds and flux densities of NO in the Bohai and Yellow Seas.

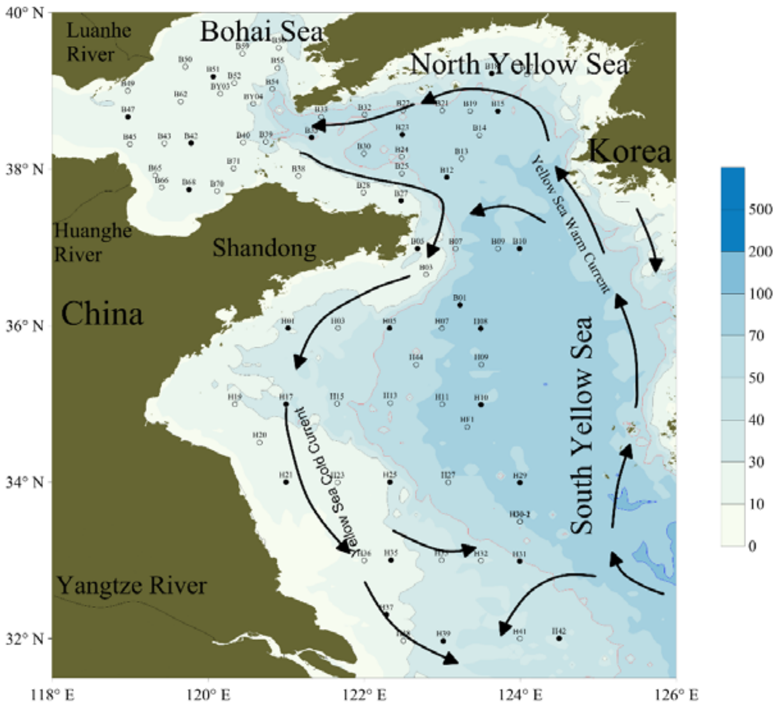

**Figure 1.** Locations of the sampling stations in the BS and the YS during summer Solid dots (●) represent the stations for incubation experiments.

带格式的: 居中

删除的内容: .

带格式的: 英语(美国)

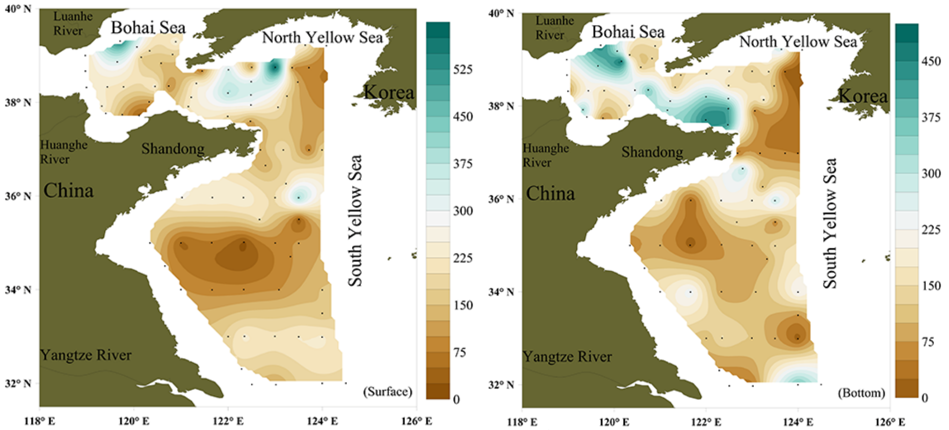

**Figure 2.** Horizontal distributions of NO (pmol L$^{-1}$) in the surface and bottom layers.

带格式的：居中

删除的内容：mol·

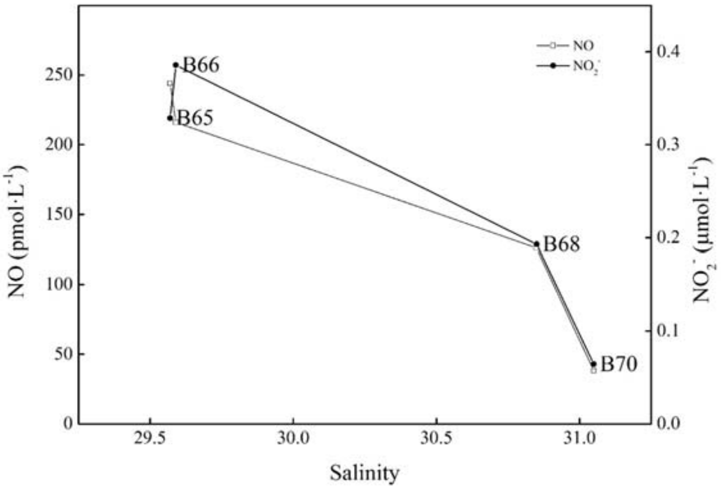

**Figure 3.** Variations of salinity, NO surface concentrations, and $NO_2^-$concentrations from station B65 to station B70.

带格式的: 居中

带格式的: 居中

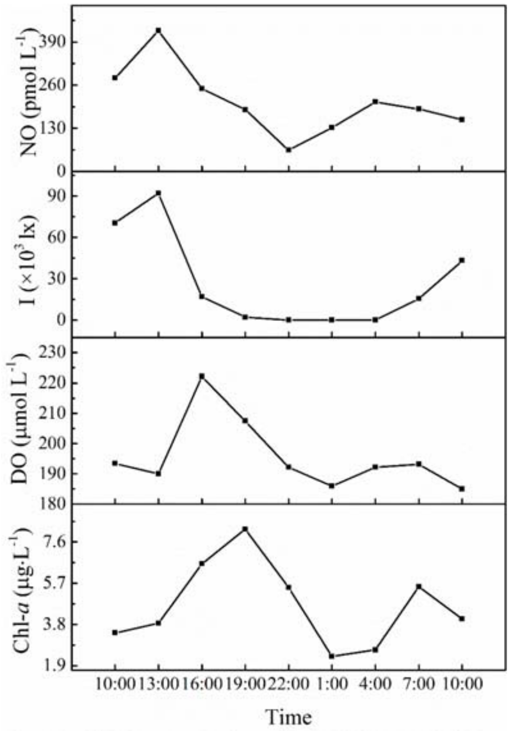

**Figure 4.** Diurnal variations of NO concentrations, illuminance (I), DO, and Chl-*a* concentrations in the surface water at the anchor station B65.

**带格式的:** 制表位： 25.09 字符, 居中

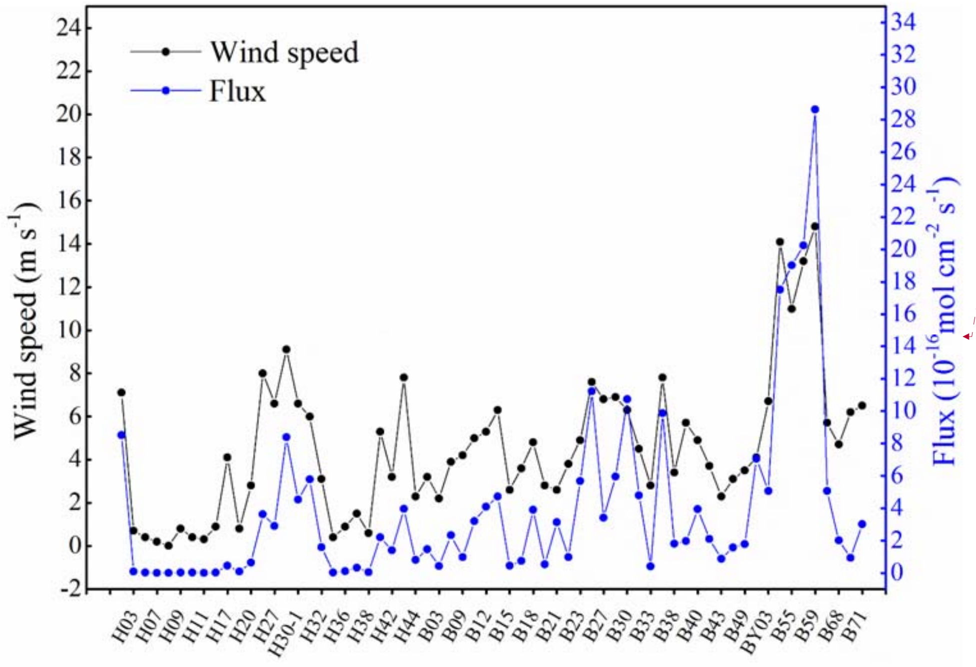

**Figure 5.** Wind speeds and flux densities of NO from the Bohai and Yellow Seas.

**带格式的:** 居中

**Table Captions**

**Table 1** Description of sampling stations and seawater temperature (T), salinity (S), Chl-*a* concentrations, DO, illuminance (I), and NO concentrations in the BS and the YS.

**Table 2** Description of sampling stations and their seawater temperature, illuminance, mean surface downward UV radiation flux (UV), and photoproduction rates in the BS and the YS.

删除的内容: tion intensity

**Table 3** NO Concentrations of different regions in literatures.

**Table 1** Description of sampling stations and seawater temperature (T), salinity(S), Chl-*a* concentrations,

| Station | Location | Depth (m) | Local time | $T_{surf}$ (°C) | $T_{bott}$ (°C ) | $S_{Surf}$ ‰ | $S_{bott}$ ‰ | Chl-$a_{surf}$ (μg L$^{-1}$) | $DO_{surf}$ (μM) | $DO_{bott}$ (μM) | I (lx) | $[NO]_{surf}$ (pM) | $[NO]_{bott}$ (pM |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| H01 | 121.03° E, 35.97° N | 33 | 1928 | 15.0 | 9.7 | 31.72 | 31.76 | 0.565 | 233.35 | 265.84 | 169 | 257 | 196 |
| H03 | 121.67° E, 35.97° N | 37 | 2250 | 18.3 | 8.8 | 31.73 | 31.68 | 0.726 | 212.8 | 242.69 | 32 | 248 | <LOD |
| H05 | 122.33° E, 35.97° N | 55 | 0238 | 17.1 | 6.3 | 31.50 | 32.25 | 0.207 | 231.82 | 203.8 | 32 | 253 | 206 |
| H07 | 123.00° E, 35.97° N | 71 | 0618 | 17.0 | 7.7 | 31.55 | 32.93 | 0.446 | 226.54 | 199.21 | 19090 | 195 | 195 |
| H08 | 123.50° E, 35.96° N | 75 | 0929 | 17.1 | 7.7 | 32.35 | 32.99 | 7.674 | 222.67 | 194.6 | 57700 | 407 | 295 |
| H09 | 123.50° E, 35.50° N | 76 | 1240 | 18.3 | 7.8 | 32.20 | 33.05 | 7.284 | 215.41 | 202.61 | 101500 | 32 | 57 |
| H10 | 123.50° E, 34.99° N | 77 | 1546 | 18.7 | 7.8 | 32.53 | 33.16 | 3.206 | 218.32 | 197.55 | 13200 | 110 | 148 |
| H11 | 123.00° E, 35.00° N | 72 | 1842 | 19.1 | 8.1 | 31.97 | 33.36 | 2.587 | 220.06 | 196.44 | 58 | 90 | 135 |
| H13 | 122.34° E, 35.01° N | 62 | 2242 | 19.0 | 7.7 | 31.67 | 32.87 | 0.521 | 215.07 | 199.33 | 32 | <LOD | 78 |
| H15 | 121.65° E, 35.00° N | 46 | 0235 | 19.6 | 6.3 | 31.59 | 32.00 | 0.627 | 207.00 | 213.24 | 32 | 35 | <LOD |
| H17 | 121.00° E, 35.00° N | 38 | 0612 | 18.4 | 10.5 | 31.19 | 31.86 | 2.400 | 224.96 | 229.62 | 33900 | 41 | 81 |
| H19 | 120.34° E, 35.00° N | 28 | 1013 | 17.2 | 15.3 | 31.62 | 31.71 | 1.375 | 220.03 | 218.6 | 98700 | 172 | 55 |
| H20 | 120.66° E, 34.51° N | 20 | 1354 | 19.0 | 18.8 | 30.01 | 30.00 | 0.470 | 216.86 | 207.34 | 77400 | 113 | 122 |
| H21 | 121.00° E, 34.00° N | 19 | 1733 | 20.4 | 20.1 | 31.00 | 30.88 | 0.597 | 191.46 | 194.87 | 6490 | NA | NA |
| H23 | 121.66° E, 34.00° N | 20 | 2104 | 19.1 | 19.1 | 31.96 | 31.96 | 4.417 | 197.10 | 196.78 | 32 | NA | 244 |
| H25 | 122.33° E, 34.00° N | 40 | 0112 | 19.3 | 9.8 | 32.00 | 32.51 | 5.632 | 211.41 | 225.14 | 32 | 86 | 78 |
| H27 | 123.08° E, 34.00° N | 70 | 0618 | 18.9 | 8.8 | 32.73 | 33.36 | 4.938 | 205.1 | 204.93 | 10122 | 99 | 104 |
| H29 | 124.00° E, 33.99° N | 82 | 1257 | 19.2 | 9.1 | 32.63 | 33.71 | 13.933 | 208.46 | 180.25 | 65183 | 153 | 232 |
| H30-1 | 124.00° E, 33.50° N | 69 | 1615 | 18.4 | 10.1 | 31.79 | 33.65 | 1.047 | 226.07 | 198.98 | 12780 | 153 | 70 |
| H31 | 123.99° E, 32.99° N | 49 | 1912 | 18.6 | 11.9 | 31.68 | 32.72 | 1.593 | 222.42 | 191.59 | 11330 | 231 | <LOD |
| H32 | 123.50° E, 33.00° N | 39 | 2130 | 18.4 | 13.8 | 31.65 | 32.27 | 15.783 | 229.45 | 191.93 | 32 | 226 | NA |
| H33 | 122.99° E, 33.00° N | 36 | 2352 | 17.6 | 14.8 | 31.91 | 32.15 | 7.636 | 224.52 | 198.59 | 32 | NA | 124 |
| H35 | 122.35° E, 33.00° N | 36 | 0358 | 17.6 | 17.6 | 31.69 | 31.71 | 3.195 | 210.63 | 211.31 | 32 | 233 | 154 |
| H36 | 122.00° E, 33.00° N | 14 | 0610 | 18.4 | 18.4 | 31.98 | 31.99 | 2.272 | 198.29 | 197.87 | 44800 | 183 | 102 |
| H37 | 122.29° E, 32.31° N | 25 | 1037 | 19.1 | 19.0 | 30.95 | 31.08 | 5.725 | 181.50 | 180.75 | 22200 | 189 | 68 |
| H38 | 122.50° E, 31.97° N | 27 | 1328 | 19.2 | 18.3 | 29.65 | 31.28 | 6.407 | 177.64 | 147.26 | 9620 | 179 | 237 |
| H39 | 123.02° E, 31.97° N | 38 | 1600 | 18.6 | 18.5 | 31.89 | 31.89 | 3.406 | 180.44 | 178.31 | 18140 | 114 | 36 |
| H41 | 124.00° E, 32.00° N | 43 | 2125 | 17.2 | 17.1 | 31.41 | 31.45 | 1.090 | 202.38 | 201.91 | 32 | NA | 376 |
| H42 | 124.50° E, 32.00° N | 43 | 0030 | 18.7 | 15.4 | 31.90 | 32.24 | 0.897 | 225.02 | 199.71 | 32 | 188 | 271 |
| HF1 | 123.32° E, 34.71° N | 78 | 1635 | 20.3 | 8.2 | 32.38 | 33.40 | 0.671 | 203.16 | 191.03 | 10940 | 97 | 92 |

带格式的：居中

删除的内容：·

| | | | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| H44 | 122.67° E, 35.50° N | 69 | 2348 | 19.7 | 7.6 | 31.51 | 32.91 | 3.712 | 222.87 | 185.19 | 32 | 205 | NA |
| B01 | 123.23° E, 36.26° N | 75 | 0328 | 16.9 | 7.3 | 31.56 | 32.80 | 0.605 | 240.44 | 204.71 | 528 | 201 | 95 |
| B03 | 122.79° E, 36.65° N | 25.7 | 0753 | 15.4 | 11.7 | 31.38 | 31.48 | 0.236 | 258.34 | 243.82 | 54200 | 126 | 311 |
| B05 | 122.69° E, 36.98° N | 41 | 1136 | 15.5 | 11.5 | 31.59 | 31.62 | 1.494 | 247.11 | 235.10 | 100000 | NA | <LOD |
| B07 | 123.17° E, 36.98° N | 63 | 1443 | 20.5 | 6.7 | 31.57 | 32.23 | 0.309 | 222.31 | 237.81 | 76500 | 208 | 60 |
| B09 | 123.72° E, 36.98° N | 76 | 1811 | 20.0 | 6.8 | 31.92 | 32.42 | 0.441 | 211.24 | 236.46 | 2440 | 79 | NA |
| B10 | 123.99° E, 36.98° N | 77 | 1947 | 21.0 | 6.8 | 31.95 | 32.42 | 0.312 | 204.13 | 227.81 | 922 | 177 | 48 |
| B12 | 123.06° E, 37.89° N | 62 | 0200 | 18.8 | 6.4 | 31.64 | 32.14 | 0.149 | 217.03 | 235.66 | 32 | 207 | 49 |
| B13 | 123.25° E, 38.13° N | 65 | 0440 | 18.9 | 5.9 | 31.73 | 32.09 | 0.398 | 212.71 | 257.77 | 203 | 172 | 191 |
| B14 | 123.48° E, 38.43° N | 66 | 0700 | 19.2 | 7.1 | 31.76 | 32.15 | 0.427 | 218.36 | 262.51 | 2500 | NA | NA |
| B15 | 123.72° E, 38.74° N | 59 | 0918 | 19.4 | 8.7 | 31.79 | 32.11 | 0.274 | 216.53 | 262.56 | 16640 | 90 | <LOD |
| B17 | 124.09° E, 39.21° N | 41 | 1300 | 11.3 | 10.9 | 31.62 | 31.65 | 0.663 | 260.47 | 259.75 | 12930 | 89 | <LOD |
| B18 | 123.64° E, 39.22° N | 49 | 1528 | 19.7 | 8.7 | 31.39 | 31.84 | 0.600 | 223.91 | 256.77 | 5140 | 236 | 276 |
| B19 | 123.36° E, 38.74° N | 57 | 1852 | 19.7 | 7.3 | 31.57 | 32.13 | 2.276 | 216.04 | 264.95 | 1010 | 94 | 161 |
| B21 | 123.00° E, 38.75° N | 54 | 2052 | 19.7 | 5.2 | 31.29 | 31.98 | 0.527 | 214.72 | 264.49 | 32 | 616 | 123 |
| B22 | 122.50° E, 38.75° N | 55 | 2348 | 17.4 | 3.9 | 30.81 | 32.03 | 1.067 | 246.19 | 249.64 | 32 | 98 | 192 |
| B23 | 122.49° E, 38.44° N | 55 | 0210 | 20.1 | 4.9 | 31.66 | 31.93 | 0.579 | 207.68 | 245.58 | 32 | 327 | 154 |
| B25 | 122.48° E, 37.94° N | 49 | 1616 | 18.4 | 5.2 | 31.17 | 31.79 | 0.090 | 224.02 | 239.04 | 8720 | 286 | 407 |
| B27 | 122.47° E, 37.60° N | 27 | 1903 | 18.0 | 14.1 | 31.16 | 31.47 | 0.369 | 229.34 | 188.49 | 203 | 110 | 417 |
| B28 | 121.99° E, 37.70° N | 22.8 | 2143 | 13.3 | 9.4 | 31.47 | 31.60 | 2.080 | 270.02 | 247.82 | 32 | 194 | 453 |
| B30 | 122.00° E, 38.20° N | 56 | 0147 | 18.7 | 3.8 | 31.18 | 32.02 | 1.128 | 217.62 | 246.15 | 32 | 387 | 338 |
| B32 | 122.01° E, 38.70° N | 53 | 0550 | 16.1 | 4.6 | 30.61 | 31.91 | 4.697 | 240.34 | 251.92 | 25000 | 339 | 171 |
| B33 | 121.43° E, 38.67° N | 61 | 0752 | 18.1 | 5.6 | 30.82 | 31.81 | 2.737 | 221.37 | 252.02 | 28700 | 72 | NA |
| B35 | 121.33° E, 38.40° N | 50 | 1052 | 17.0 | 6.3 | 30.81 | 31.75 | 4.616 | 237.4 | 252.03 | 30900 | 244 | 128 |
| B38 | 120.74° E, 38.35° N | 22 | 1530 | 15.2 | 12.1 | 31.19 | 31.47 | 1.945 | 251.83 | 255.95 | 15000 | 223 | 315 |
| B39 | 120.45° E, 38.34° N | 29 | 2021 | 13.8 | 12.3 | 31.19 | 31.30 | 0.706 | 241.53 | 240.32 | 32 | 94 | 363 |
| B40 | 119.78° E, 38.33° N | 30 | 2208 | 14.4 | 12.1 | 30.97 | 31.28 | 0.382 | 254.71 | 245.62 | 32 | 243 | 241 |
| B42 | 119.44° E, 38.33° N | 26.8 | 0550 | 19.0 | 12.6 | 30.64 | 31.37 | 1.151 | 214.26 | 216.44 | 19700 | 211 | 157 |
| B43 | 119.00° E, 38.32° N | 24 | 0740 | 18.8 | 13.9 | 30.93 | 31.32 | 2.667 | 226.67 | 188.85 | 42400 | 224 | 208 |
| B45 | 118.97° E, 38.67° N | 20.5 | 1022 | 19.4 | 17.2 | 31.07 | 31.22 | 1.612 | 236.45 | 208.28 | 81900 | NA | 60 |
| B47 | 118.97° E, 38.67N | 25 | 1325 | 17.4 | 13.0 | 31.18 | 31.47 | 0.133 | 251.91 | 212.27 | 91900 | 227 | NA |
| B49 | 118.97° E, 39.00° N | 21.4 | 1610 | 17.6 | 16.3 | 31.31 | 31.37 | 0.596 | 270.9 | 240.59 | 29000 | 204 | 236 |
| B50 | 119.71° E, 39.31° N | 26 | 2010 | 20.7 | 11.8 | 31.25 | 31.37 | 0.135 | 215.16 | 212.47 | 32 | 565 | 452 |

| | | | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| B51 | 120.07° E, 39.18° N | 24 | 2220 | 19.3 | 14.8 | 31.21 | 31.31 | 0.625 | 220.94 | 213.47 | 32 | NA | 307 |
| BY03 | 120.16° E, 38.96° N | 21 | 0008 | 17.7 | 17.5 | 31.15 | 31.15 | 0.843 | 242.26 | 246.04 | 32 | 167 | 482 |
| B52 | 120.34° E, 39.10° N | 22 | 0201 | 18.6 | 17.6 | 31.23 | 31.22 | 1.114 | 245.12 | 231.72 | 32 | NA | 141 |
| BY04 | 120.58° E, 38.84° N | 37 | 0515 | 16.8 | 15.8 | 31.24 | 31.27 | 0.423 | 218.19 | 206.65 | 1463 | 147 | 125 |
| B55 | 120.89° E, 39.29° N | 33 | 0929 | 16.9 | 13.2 | 31.16 | 31.36 | 0.210 | 231.28 | 226.55 | 39500 | 248 | NA |
| B56 | 120.91° E, 39.55° N | 31.6 | 1124 | 17.5 | 14.0 | 31.29 | 31.40 | 0.967 | 214.96 | 191.25 | 70500 | 189 | 204 |
| B59 | 120.44° E, 39.48° N | 27 | 1357 | 17.4 | 12.7 | 31.25 | 31.41 | 0.232 | 215.45 | 192.18 | 86200 | 217 | 185 |
| B65 | 119.65° E, 38.86° N | 16 | 0957 | 20.8 | NA | 29.57 | NA | 0.323 | 193.56 | NA | 31800 | 244 | 335 |
| B66 | 119.32° E, 37.92° N | 14 | 1225 | 21.7 | 21.6 | 29.59 | 30.03 | 0.493 | 190.04 | 174.75 | 35500 | 216 | 244 |
| B68 | 119.41° E, 37.73N | 16.6 | 1442 | 21.5 | 21.1 | 30.85 | 30.86 | 0.757 | 195.03 | 179.67 | 30000 | 126 | 63 |
| B70 | 120.12° E, 37.72N | 17 | 1717 | 20.7 | 19.1 | 31.05 | 31.17 | 0.988 | 202.73 | 187.42 | 109 | 38 | 131 |
| B71 | 120.22° E, 38.01N | 19.9 | 1939 | 17.8 | 17.2 | 31.18 | 31.19 | 0.344 | 226.72 | 212.02 | 53 | 106 | 184 |

NA: not available
<LOD: below the limit of detection

**Table 2** Description of sampling stations and their seawater temperature, illuminance, mean surface downward UV radiation flux (UV), and photoproduction rates in the BS and the YS.

| | Station | I<br>(lx) | UV<br>(W m$^{-2}$) | T<br>(°C) | photoproduction rate<br>($10^{-11}$ mol L$^{-1}$s$^{-1}$) |
|---|---|---|---|---|---|
| SYS | H01 | 7310 | 6.5 | 15.0 | 0.21 |
| | H05 | 57700 | 91.6 | 17.1 | 3.18 |
| | H08 | 57700 | 91.6 | 17.1 | 2.92 |
| | H10 | 57700 | 91.6 | 18.7 | 5.07 |
| | H17 | 57700 | 91.6 | 18.4 | 2.79 |
| | H21 | 57700 | 91.6 | 20.4 | 4.37 |
| | H25 | 16790 | 21.5 | 19.3 | 1.28 |
| | H29 | 16790 | 21.5 | 19.2 | 0.00 |
| | H31 | 16790 | 21.5 | 18.6 | 0.51 |
| | H35 | 16790 | 21.5 | 17.6 | 0.22 |
| | H37 | 16790 | 21.5 | 19.1 | 1.17 |
| | H39 | 18140 | 23.7 | 18.6 | 1.29 |
| | H42 | 18140 | 23.7 | 18.7 | 0.16 |
| | B01 | 9720 | 10.5 | 16.9 | 0.28 |
| | B05 | 9720 | 10.5 | 15.5 | 0.05 |
| | B10 | 9720 | 10.5 | 21.0 | 0.09 |
| | Average | 27825 | 40.6 | 18.2 | 1.47 |
| NYS | B12 | 15200 | 22.2 | 18.8 | 0.45 |
| | B15 | 15200 | 22.2 | 19.4 | 0.09 |
| | B18 | 15200 | 22.2 | 19.7 | 0.22 |
| | B23 | 15200 | 22.2 | 20.1 | 0.69 |
| | B27 | 30900 | 52.0 | 18.0 | 0.27 |
| | B35 | 30900 | 52.0 | 17.0 | 0.10 |
| | Average | 20433 | 32.1 | 18.3 | 0.33 |
| BS | B42 | 30900 | 52.0 | 19.0 | 1.54 |
| | B47 | 14000 | 17.5 | 17.4 | 0.88 |
| | B51 | 14000 | 17.5 | 19.3 | 0.32 |
| | B68 | 30900 | 52.0 | 21.5 | 1.50 |
| | Average | 22450 | 34.7 | 19.3 | 1.14 |

带格式的：居中

带格式表格

删除的内容：mol·

带格式的：字体：（中文）+中文正文（宋体），（中文）中文(中国)

带格式的：字体：（中文）+中文正文（宋体），（中文）中文(中国)

带格式的：字体：（中文）+中文正文（宋体），（中文）中文(中国)

带格式的：字体：（中文）+中文正文（宋体），（中文）中文(中国)

带格式的：字体：（中文）+中文正文（宋体），（中文）中文(中国)

带格式的：字体：（中文）+中文正文（宋体），（中文）中文(中国)

带格式的：字体：（中文）+中文正文（宋体），（中文）中文(中国)

带格式的：字体：（中文）+中文正文（宋体），（中文）中文(中国)

带格式的：字体：（中文）+中文正文（宋体），（中文）中文(中国)

带格式的：字体：（中文）+中文正文（宋体），（中文）中文(中国)

带格式的：字体：（中文）+中文正文（宋体），（中文）中文(中国)

带格式的：字体：（中文）+中文正文（宋体），（中文）中文(中国)

带格式的：字体：（中文）+中文正文（宋体），（中文）中文(中国)

带格式的：字体：（中文）+中文正文（宋体），（中文）中文(中国)

带格式的：字体：（中文）+中文正文（宋体），（中文）中文(中国)

带格式的：字体：（中文）+中文正文（宋体），（中文）中文(中国)

带格式的：字体：（中文）+中文正文（宋体），（中文）中文(中国)

**Table 3** NO concentrations and flux densities from different regions.

| Regions | [NO] (pmol L$^{-1}$) | NO fluxes (mol cm$^{-2}$ s$^{-1}$) | Sampling date | Sampling depth | Reference |
|---|---|---|---|---|---|
| Jiaozhou Bay | 157 | 7.2×10$^{-16}$ | June, July and August, 2010 | Surface water | Tian et al., 2015 |
| Jiaozhou Bay and its adjacent waters | 160 ± 130 | 10.9×10$^{-16}$ | March 8-9, 2011 | Surface water | Xue et al., 2012 |
| Central equatorial Pacific | 46 | > 2.2×10$^{-16}$ | July 14 to August 16, 1978 | Surface water | Zafiriou and McFarland, 1981 |
| Eastern tropical North Pacific Ocean | 0-65 | - | November, 1983 | 0-3500m | Ward and Zafiriou, 1988 |
| Eastern tropical South Pacific Ocean off Peru | <500-9500 | - | February 6 to March 11, 2013 | Surface-327 m | Lutterbeck et al., 2018 |
| Coastal water off Qingdao | 260 ± 140 | - | November, 2009 | Surface water | Liu et al.,2017 |
| Seto Inland Sea, Japan | 24-320 | 3.55 ×10$^{-16}$ | October 5-9, 2009 | Surface water | Olasehinde et al, 2010 |
| Yellow Sea and Bohai Sea | Surface: 186 ± 108<br>Bottom: 174 ± 123 | 4.5×10$^{-16}$ | June 13-28, 2011 | 1 m and 1 m above the bottom | This study |

**带格式的:** 居中

**删除的内容:** mol·