



Impact of an accidental explosion in Tianjin Port on enhanced atmospheric nitrogen deposition over the Bohai Sea inferred from aerosol nitrate dual isotopes

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ABSTRACT

In recent years, emergent pollutant's accidents have occurred frequently in China, causing serious harm to the ecological environment. In this study, the impact of an accidental fire and explosion at Tianjin Port in 2015 on the atmosphere over the Bohai Sea was explored. Results showed sharp increases in the concentrations of several important components of fine particulate matter (e.g. NO_3^- , SO_4^{2-} , NH_4^+ , organic carbon, elemental carbon) over Beihuangcheng Island after the explosion. Among them, NO₃⁻ was most affected (about 10 days), with a maximum concentration of 16.45 μ g m⁻³. The δ^{15} N-NO₃ ranged from -1.58% to +8.74%, with an average of +2.79% \pm 3.32%. Influenced by the explosion, δ^{15} N-NO₃⁻ decreased significantly, which was in accordance with the industrial processes of explosives. The $\delta^{18}\text{O-NO}_3^-$ varied between +49.40% and +69.52%, and showed a marked increase ($+66.62\% \pm 3.92\%$) in the explosion-affected period. Using Monte Carlo simulation, the •OH pathway for NO_3^- formation was 51.79% \pm 10.94% at that time — much lower than in the regular period. The elevated dry deposition of NO₃ caused by the explosion was 266.08 μ mol N m⁻² d⁻¹ over the Bohai Sea — again, much higher than in the regular period. With the dry nitrogen deposition of NH₄⁺ (42.41 μ mol N m⁻² d⁻¹), the total nitrogen deposition increased by 308.49 μ mol N m⁻² d⁻¹, leading to severe ecological risk. Through the inverse computation of the dry deposition flux of NO₃⁻, the affected area over the Bohai Sea was less than 1.42×10^4 km², which is about 20% of the total area.

利用气溶胶硝酸盐双同位素评估天津港爆炸事故对渤海地区大气氮沉降的影响

近年来,我国突发性污染事故频繁发生,对生态环境造成了严重危害。本研究基于气溶胶硝酸 盐的氮氧同位素确认并评估了2015年天津港火灾爆炸事故对渤海地区大气氮沉降的影响研究 结果表明 δ^{15} N-NO $_3$ -与 δ^{18} O-NO $_3$ -范围分别为-1.58‰至+8.74‰与+49.40‰ 至+69.52‰ 受到火 灭爆炸影响, δ^{15} N-NO₃⁻显著下降,而 δ^{18} O-NO₃⁻明显上升。火灾爆炸导致渤海地区大气氦沉降量增加308.49 μmol N m⁻² d⁻¹,其中硝态氦为266.08 μmol N m⁻² d⁻¹,总影响区域约为渤海面积 的1/5。本研究显示同位素技术是污染事故研究中重要的示踪手段。

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1. Introduction

Environmental pollution events mainly include air pollution, water pollution, soil pollution, and other emergent pollution events or radiation pollution events (Fu, Wang, and Yan 2016). Since China's period of 'reform and opening up', total production and the scale of industrial enterprises in the country have expanded greatly. As such, the longterm accumulated environmental risks have evolved and intensified, and China has entered a period of high incidence of environmental pollution events (Huang and Zhang 2015). In recent years, all kinds of environmental pollution events have begun to occur more frequently (Text S1), causing huge economic and environmental losses. Among them, air pollution emergencies have a wider range of influences, and their secondary pollutants are more complicated. They are thus of relatively greater concern to the public and media compared with other types of pollution events (Chen, Tang, and Zhao 2015).

Tianjin Port is located at the mouth of the Haihe River — the intersection of the Beijing-Tianjin-Hebei urban agglomeration and the Bohai economic circle. Its throughput annual amounts to about hundred million tons, making it the largest port in the north of China (Zhang et al. 2019). At about 2330 LST (local standard time) 12 August 2015, a dangerous

goods warehouse belonging to Ruihai International Logistics Co. situated in the Dongjiang Bonded Port Area of Tianjin Binhai New Area exploded. The explosion can be divided into two phases: (1) a first explosion occurred at 2334, for which the local magnitude (ML) was about 2.3 — equivalent to 3 tons of TNT; and (2) a second explosion occurred after 30 s, for which the ML was about 2.9 — equivalent to 21 tons of TNT. The explosion led to 165 deaths, 8 people missing, and more than 800 people who were injured (Yu et al. 2016). The explosives included a large amount of NO₃⁻, such as NH₄NO₃ and KNO₃ (http:// news.china.com/zh_cn/focus/tjgbz/), which burned for more than 15 h and generated a large amount of polluting gas and atmospheric particulates. According to the forward air trajectories at that time, the nitrogen-containing pollutants were mostly transmitted to the Bohai Sea, but there remains a lack of understanding regarding the impacts over the region.

In this study, the impacts of this accidental fire and explosion at Tianjin Port (hereafter referred to simply as 'Tianjin explosion') on the atmosphere over Bohai Sea were explored based on fine particulate (PM_{2.5}) data recorded on Beihuangcheng (BH) Island, which is located in the middle of the Bohai Strait. The objectives were to: (1) determine the affected time of Tianjin explosion on the Bohai Sea, based on the variation in the concentrations of PM_{2.5} and its species; (2) confirm the effects of Tianjin explosion through nitrogen/oxygen $(\delta^{15}N-NO_3^{-1}/\delta^{18}O-$ NO₃⁻) isotope analysis; and (3) assess the elevated dry nitrogen deposition caused by the explosion and estimate roughly the affected area over the Bohai Sea. To the best of our knowledge, this is the first attempt at interpreting the impact of Tianjin explosion on the atmosphere over the

Bohai Sea, and using isotopic analysis of NO₃⁻ in an explosion-affected period is also a first. The results should prove helpful in understanding the ecological risks of environmental pollution events.

2. Materials and methods

PM_{2.5} samples were collected on BH Island — an independent island located in the middle of the Bohai Strait from 31 July to 30 August 2015. It is a part of the annual atmospheric observations at the Environmental Monitoring Station of the State Ocean Administration of China (38°24'N, 120°55'E) (Zong et al. 2017). For particulate compositions, organic carbon (OC), elemental carbon (EC), and water-soluble ions, including Na⁺, K⁺, Ca²⁺, Mg²⁺, NH₄⁺, Cl⁻, NO₃⁻, and SO₄²⁻, were detected. In addition, δ^{15} N-NO₃⁻/ δ^{18} O-NO₃⁻ were also analyzed using the nitrous oxide (N2O) conversion method. Specific information about the sampling campaign and chemical analysis can be referred to in Text S2.

Forward trajectories starting at 2300 LST 12 August 2015 at the Tianjin explosion point were generated using the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model, which is a complete system for computing simple air parcel trajectories in complex dispersion simulations (Bressi et al. 2014). It is available on the National Oceanic and Atmospheric Administration's Air Resource Laboratory website (www.arl.noaa.gov/ready/hys plit4.html). As indicated by Jiang, Ji, and Teng (2017), the pollutants produced in the explosion were mainly raised to 1-2 km into the upper air. Therefore, the model was adopted to generate 72-h forward trajectories encountered

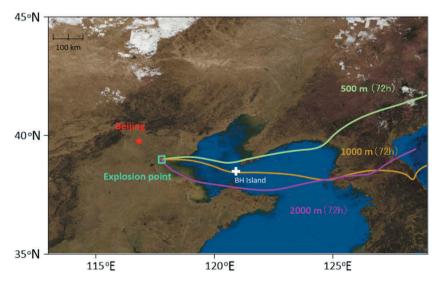


Figure 1. Geographical locations of the explosion point and BH Island, along with the characteristics of forward trajectories (72 h) at 500, 1000, and 2000 m, respectively, starting at 2300 LST 12 August 2015 at the explosion point.

at 500, 1000, and 2000 m, respectively, showing most of the pollutants spread to the middle of the Bohai Sea (Figure 1).

3. Results and discussion

3.1 Component characteristics of PM_{2.5} before, during, and after the explosion

Figure 2 illustrates the chemical features of PM_{2.5} on BH Island from 31 July to 30 August 2015. Apparently, sharp increasing concentrations of several important components (e.g. NO₃⁻, SO₄²⁻, NH₄⁺, OC, and EC) were observed beginning at the time of Tianjin explosion, while other species, such as Cl⁻, Na⁺, K⁺, and Mg²⁺, did not change significantly. This indicates that the explosion inflicted a marked impact on secondary particulate components and carbonaceous species in the Bohai region. For better revealing the impacts of the explosion, the sampling period was divided into three phases: (1) before the explosion (named BE, 31 July-9 August); (2) during the explosion (DE, 12 August-21 August); and after the explosion (AE, 24 August-30 August). The average concentrations of PM_{2.5}, NO₃ $^-$, SO₄ $^{2-}$, and NH₄ $^+$ were 61.11 \pm 10.06 $\mu g \text{ m}^{-3}$, 14.04 \pm 2.34 $\mu g \text{ m}^{-3}$, 9.33 \pm 0.81 $\mu g \text{ m}^{-3}$, and 6.42 \pm 3.64 µg m⁻³, respectively, in the DE period — much higher than those in the BE period (23.01 \pm 10.02 μ g m⁻³, $3.02 \pm 2.52 \ \mu g \ m^{-3}$, $4.90 \pm 2.16 \ \mu g \ m^{-3}$, 2.35 ± 1.48 $\mu g \ m^{-3}$) and AE period (23.41 \pm 10.24 $\mu g \ m^{-3}$, 2.52 \pm

1.18 μ g m⁻³, 4.32 \pm 1.63 μ g m⁻³, 2.46 \pm 1.02 μ g m⁻³). Comparatively, the concentrations of these secondary constituents in the BE and AE phases were not much different (p > 0.05), reflecting the basic feature of this season, while the high concentrations in the DE period implied a considerable influence of the explosion on BH Island. Different from the continuous tendency of high concentration for secondary constitutes, carbonaceous species, such as OC and EC, only peaked on 12 August, and then declined rapidly. In addition, the concentration of Ca²⁺, which had been stable, increased on 21 August accompanied by a decrease in NH₄⁺ concentration.

Based on the Geostationary Ocean Color Imager (Jiang, Ji, and Teng 2017), the explosive gas mass had transported to and passed BH Island in about 24 h. The time was exactly consistent with the time of the increase and decrease for OC and EC concentrations. This may indicate that the high contents of OC and EC were mostly primary products of Tianjin explosion (Liu et al. 2014). However, judging from the variation of secondary components, we speculate that the impact of Tianjin explosion on the Bohai Sea region could have lasted about 10 days. In the BE and AE periods, the concentration of NO₃⁻ was mostly lower than that of SO₄²⁻, but the trend reversed in the DE period, suggesting that the effect of Tianjin explosion on NO₃⁻ was the most significant in terms of single components in PM_{2.5}. The highest concentration of NO_3^- was 16.45 $\mu g m^{-3}$ in the DE

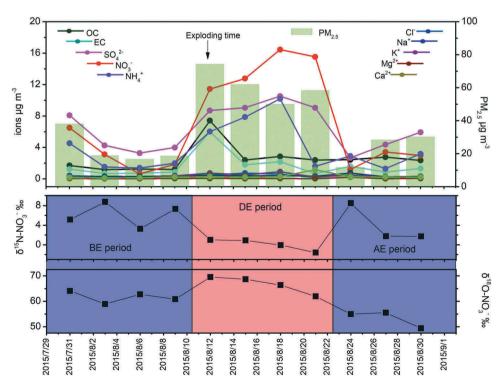


Figure 2. Variation in the concentrations of PM_{2.5} components (OC, EC, SO₄²⁻, NO₃⁻, NH₄⁺, Cl⁻, Na⁺, K⁺, Mg²⁺, and Ca²⁺) and the time series of δ^{15} N-NO₃⁻ and δ^{18} O-NO₃⁻ in PM_{2.5} observed on BH Island from 31 July to 30 August in 2015.

period — about 25 times the lowest value in the BE and AE periods; and taking the average value of the BE and AE stages (2.81 μ g m⁻³) as the background value of the Bohai Sea, Tianjin explosion caused the concentration of NO₃⁻ on BH island to rise by 13.64 μ g m⁻³. As reported, the explosives mainly included NaCN (700 tons), NH₄NO₃ (800 tons), KNO₃ (500 tons) (Jiang, Ji, and Teng 2017), and possible chemical reactions occurred via the following chemical equations:

$$NaCN + H2O \rightarrow HCN + NaOH; 4HCN + 9O2 \xrightarrow{light} 2H2O + 4NO2 + 4CO2$$
(1)

$$4NH4NO3 \xrightarrow{400^{\circ}C} 3N2 + 2NO2 + 8H2O$$
 (2)

$$2\text{KNO}_{3} \stackrel{\text{light}}{\longrightarrow} 2\text{KNO}_{2} + \text{O}_{2}; 2\text{KNO}_{2} \rightarrow \text{NO} + \text{NO}_{2} + \text{K}_{2}\text{O} \tag{3}$$

Assuming that the above chemical reactions occurred completely, we can infer that the amount of NO_x emission from the explosion was approximately 1600 tons. The NO_x would have been discharged into the atmosphere with more diffusivity, longer duration, and a more extensive diffusion area compared with solid pollutants, which might explain the high concentration of NO₃ observed after 12 August – the time of the explosion and the transmission to BH Island of explosive gas masses. Although there are no precursors of NH_4^+ in the explosion, the high concentrations might be attributable to the promotion of NO_x. NO₃ formed due to the explosion would have increased the acidity of the atmosphere and greatly promoted the conversion of gaseous NH₃ to particulate NH₄⁺ (Pan et al. 2016). We can speculate that the NH₃ content in the Bohai region was very low at that time, although there are no direct observational data. However, it can be inferred by the increase in Ca²⁺ concentration and decrease in NH₄⁺ on 21 August, as the high acidity gas would have continued to acidify the Ca material to balance the acidity in the case of insufficient NH₃ in the atmosphere (Xiao et al. 2017).

3.2 Nitrogen and oxygen isotope characteristics impacted by the explosion

In order to further confirm the impact of Tianjin explosion on the atmosphere over the Bohai Sea, we carried out $\delta^{15}N$ and $\delta^{18}O$ analysis for NO_3^- , which was most severely affected by the explosion. During the observation period, $\delta^{15}N-NO_3^-$ ranged from -1.58% to +8.74%, with an average of $+2.79\% \pm 3.32\%$ (Figure 2). In the DE period, the mean $\delta^{15}N-NO_3^-$ was $+0.06\% \pm 1.19\%$, which was significantly lower than that of the BE period

(+4.63% \pm 3.05%) and the AE period (+3.99% \pm 3.90%). Similar to the concentration tendency, there was no significant change between the BE period and AE period, indicating the local characteristics of the Bohai Sea in August (about +4%), while the decrease of δ^{15} N-NO₃⁻ in the DE period indicated that the high concentration of NO₃⁻ was caused by the explosion. Generally, atmospheric NO₃⁻ is mainly derived from the conversion of NO_x, of which δ^{15} N of different anthropogenic and natural sources usually varies over a large range (Elliott et al. 2019). For example, NO_x from coal combustion owns a higher $\delta^{15}N$ value, while $\delta^{15}N$ from microbial processes is more negative (Hastings, Sigman, and Lipschultz 2003). From the source apportionment result in our previous research based on an improved Bayesian model, the sources of NO_x in August on BH Island were coal combustion, biomass burning, mobile sources, and microbial processes, with contributions of $32.66\% \pm 6.67\%$, $31.84\% \pm 3.65\%$, $22.16\% \pm 4.87\%$, and $13.34\% \pm 4.61\%$, respectively (Zong et al. 2017). However, this method is not suitable for the source apportionment of NO_x during the DE period. As mentioned above, NO_x mostly originated from the explosion reaction of NaCN, NH₄NO₃, and KNO₃, which were all industrial products with $\delta^{15}N$ characteristics significantly different from those of conventional emission sources. According to the reaction principles (S₃–S₆), the nitrogen in the explosives was primarily from the N₂ in the atmosphere. Although a certain degree of isotope fractionation would have occurred during the transformation (Walters, Simonini, and Michalski 2016), the $\delta^{15}N$ value should be similar to the isotopic value of N₂ (about 0‰) in the atmosphere. This was consistent with the average of the δ^{15} N-NO₃⁻ observed herein. Notably, δ^{15} N-NO₃⁻ showed a trend of continuous decline in the DE period, which may have been due to the divergence of heavy nitrogen as the diffusion time became longer. With the decrease of NH₃ concentration in the Bohai region, nitric acid (HNO₃) would have combined with Ca substances to form calcium nitrate [Ca(NO₃)₂] with larger particle size, and the deposition was more remarkable, leading to more negative δ^{15} N-NO₃⁻ (Luo et al. 2018).

The $\delta^{18}\text{O-NO}_3^-$ on BH Island varied between +49.40% and +69.52‰, with a mean of +61.18‰ \pm 6.15‰, which was well within the broad range of values previously reported (Fang et al. 2011). Generally, the oxygen atoms of atmospheric NO_x are rapidly exchanged with O₃ in the NO/NO₂ cycle (S₇–S₉), and then NO₂ is translated to HNO₃ based on the •OH pathway (S₁₀) or O₃ pathway (S₁₁–S₁₃). Thus, the $\delta^{18}\text{O-NO}_3^-$ value is determined by its generation pathways, and can be adopted to explore the conversion process of NO_x to NO₃⁻ in the atmosphere. In the DE period, $\delta^{18}\text{O-NO}_3^-$ showed a significant increase (+66.62‰ \pm

3.92%), which was higher than those in the other two stages (BE: $+61.65\% \pm 2.24\%$; AE: $+53.29\% \pm 3.38\%$). This indicated that the explosion had a certain influence on the δ^{18} O-NO₃⁻, and impacted on the conversion pathway from NO_x to NO₃⁻. For exploring this effect, we adopted the assumption that 2/3 of oxygen atoms of NO₃⁻ may have derived from O₃ and 1/3 from •OH in the •OH generation pathway, and 5/6 of oxygen atoms were from O₃ and 1/6 from •OH in the O₃ pathway (Hastings, Sigman, and Lipschultz 2003), and then assessed the respective contributions of the two-generation pathways via a Monte Carlo simulation (Text S3) (Goulden et al. 1996). The results showed that the average contributions for the •OH generation pathway were 76.29% \pm 7.10%, 51.79% \pm 10.94%, and 91.76% ± 2.48% in the BE period, DE period, and AE period, respectively. Obviously, the conversion ratio of the O₃ pathway increased significantly in the DE period, which may have been due to the massive generation of O₃ as a byproduct during the chemical processes in Tianjin explosion. Besides, the increase of $PM_{2.5}$ mass (61.11 \pm 10.06 μ g m⁻³) could also have facilitated the O₃ pathway through the N₂O₅ heterogeneous chemistry (Qu et al. 2019).

3.3 Elevated dry nitrogen deposition triggered by the explosion

Generally, an increase in the nitrogen concentration (e.g. NO₃⁻, NH₄⁺) in the atmosphere will inevitably elevate the amount of nitrogen deposition (Duce et al. 1991). Here, we evaluated the characteristics of atmospheric dry deposition over the Bohai Sea during the explosion. Adopting dry deposition velocities used for PM_{2.5} samples of 0.017 m s $^{-1}$ for NO $_3$ $^-$ and 0.0022 m s $^{-1}$ for NH $_4$ $^+$

(Nakamura, Matsumoto, and Uematsu 2005), the NO₃⁻ and NH₄⁺ resulting from dry deposition in the DE period was estimated to be 332.68 \pm 55.39 μ mol N m⁻² d⁻¹ and $67.75 \pm 38.41 \,\mu\text{mol N m}^{-2} \,\text{d}^{-1}$, respectively. Whereas the average NO_3^- and NH_4^+ deposition was 66.60 \pm 45.64 μ mol N m⁻² d⁻¹ and 25.34 \pm 12.66 μ mol N m⁻² d⁻¹, respectively, in the BE and AE period. If the averages were taken as the background values of August 2015 in the Bohai Sea region, the elevated dry nitrogen deposition of $\mathrm{NO_3}^-$ and $\mathrm{NH_4}^+$ would have been 266.08 (Figure 3) and 42.41 µmol N m⁻² d⁻¹, respectively; the total nitrogen deposition increased by 308.49 µmol N m⁻² d⁻¹. This indicated that a great amount of nitrogen had entered the Bohai Sea region and may have caused serious damage to its ecosystem because marine areas are usually nitrogen-limited (Jickells et al. 2005). For example, an increasing nitrogen input has changed the N:Si ratio in the Yellow Sea, contributing to shifts in phytoplankton assemblages from diatoms to nondiatoms, as well as harmful algal blooms of nondiatoms (Liu and Glibert 2018); and enhanced atmospheric deposition of inorganic nitrogen in the Bohai Sea region caused a 56.5% increase in the phytoplankton biomass on average (Shou et al. 2018). Specifically, NO₃ was directly produced by the explosion, while NH₄⁺ was generated indirectly by the high concentration of NO_x. The high deposition of NH₄⁺ may have been due to the change from gaseous NH₃ to particulate NH₄ with a more intense trend of deposition (Park et al. 2019). Correspondingly, the deposition would have decreased after the event, but the total NH₄⁺ deposition was elevated. Therefore, it can be seen that attention should be paid to the direct emissions as well as the byproducts in

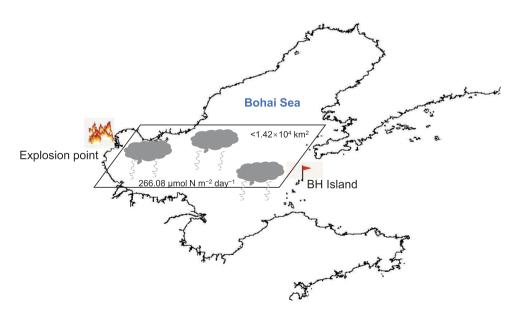


Figure 3. Elevated deposition characteristics of NO₃⁻ and the area of influence in the Bohai Sea resulting from Tianjin explosion.

the event of an emergency, especially for atmospheric incidents with complex reaction mechanisms.

Bohai is a semi-enclosed sea with an area of 7×10^4 km². Based on the deposition volume and time of influence (about 10 days indicated by the DE period), the dry deposition flux of N-NO₃⁻ induced by Tianjin explosion was estimated at 2607 tons, which significantly exceeded the amount of N-NO_x produced (about 527 tons). Such a large difference may have been caused by the total area of the Bohai Sea adopted in the above calculation. Assuming that all the NO₃⁻ generated uniformly and deposited into the Bohai Sea, the area affected by the explosion in the Bohai Sea can be reverse-estimated. The re-estimated area was approximately 1.42×10^4 km², which is about 20% of the Bohai sea. This was also in accordance with the forward trajectories, because most of the air flow was mainly transmitted to the middle of the Bohai Sea (Figure 1). The actual impact area, of course, should be smaller than the computation, because the NO₃ produced could not enter into the Bohai Sea completely. Chung and Kim (2015) proposed that the smoke plumes of Tianjin explosion could have transported to the Korean Peninsula, based on a variety of satellite remote sensing data.

4. Conclusion

The concentrations of $PM_{2.5}$, NO_3^- , SO_4^{2-} , and NH_4^+ were 61.11 \pm 10.06 μ g m⁻³, 14.04 \pm 2.34 μ g m⁻³, 9.33 \pm 0.81 $\mu g m^{-3}$, and 6.42 \pm 3.64 $\mu g m^{-3}$, respectively, in the DE period — much higher than those in the BE and AE period. This confirms the effect of Tianjin explosion on the Bohai Sea region, and the effect could have lasted about 10 days, as inferred by the duration of the DE period. Compared with some alkali ions, secondary particulate components and carbonaceous species were more affected. Specifically, NO₃⁻ was most affected in PM_{2.5}, with an increase of 13.64 μ g m⁻³. The mean δ ¹⁵N- NO_3^- in the DE period was $+0.06\% \pm 1.19\%$, which was similar to the isotopic value of N₂ in the atmosphere. Based on the industrial processes of explosives, the observed δ^{15} N-NO₃ proved that the high concentration of NO₃⁻ in the DE period mainly came from Tianjin explosion. The δ^{18} O-NO₃⁻ on BH Island varied between +49.40% and +69.52% with a mean of +61.18% \pm 6.15%, and it showed a significant increase (+66.62%) ± 3.92‰) in the DE period. Using Monte Carlo simulation, the •OH generation pathway of NO₃ was 51.79% ± 10.94% at that time — much lower than in the BE and AE period, indicating an increase in the conversion ratio of the O₃ pathway for NO₃⁻ formation. The elevated dry nitrogen deposition of NO₃⁻ and NH₄⁺ caused by Tianjin explosion was 266.08 and 42.41 μ mol N m⁻² d⁻¹, respectively, in the Bohai Sea. Through inverse computation of the dry deposition flux of NO₃⁻, the affected area of the Bohai Sea was less than 1.42×10^4 km². It is undeniable that there are some uncertainties in the calculation, but the results of this paper truly reflect the impact of Tianjin explosion on the Bohai Sea, which will inevitably help with its ecological recovery in the future.

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