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An Approach for Estimating the Nitrobenzene (NB) Emission Effect in Frozen Rivers: A Case Study of Nitrobenzene Pollution in the Songhua River, China

W. H. Xiao[1], J. H. Wang[1,*], Y. H. Huang[2,*], S. C. Sun[1,3], and Y. Y. Zhou[1]

[1]State Key Laboratory of Simulation and Regulation of Water Cycle in River Basin, China Institute of Water Resources and Hydropower Research, Beijing 100038, China
[2]Institute of Geographical Sciences and Natural Resources Research, Chinese Academy of Sciences, Beijing 100101, China
[3]Department of Environmental Science, Environmental Management College of China, Qinhuangdao 066004, China

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ABSTRACT. Clearly understanding the transport and fate of organic pollutants in frozen aquatic environments is necessary for implementing effective water quality management and public health risk assessment during the ice-bound period. In this paper, a nitrobenzene (NB) emission model coupling a modified hydrodynamic model with a water quality model was developed and applied to simulate a NB pollution emergency in the frozen Songhua River, China, that occurred on November 13, 2005. The proposed model is capable of simulating the NB emission effect in frozen rivers. The constants of the NB transfer radio from water to ice and the ice melting speed were obtained by laboratory experiments, which indicated that 10% of the NB would transfer to ice as the water froze. The quantitative statistical tests are adopted to evaluate the model performance based on ground-measured NB concentrations from April 11th to April 27th of 2006 (the ice melting period of the study area) in the Sujiatun segment of the Songhua River. The results generally show that the modeled and detected concentrations exhibit good consistency, with a RMSE of 0.6 μg/L and an R^2 of 0.877. The simulated NB released to the Amur River from the ice melt by this pollution event after April 11th is less than 12.5 μg/L, which is consistent with an absence of a negative influence on drinking water security along the River in the spring of 2006. The model performs satisfactorily for predicting the NB pollutant fate in the Songhua River and has the ability to supply the necessary information for controlling pollution events and for early warning, which could be applied to similar long frozen rivers.

Keywords: ice-bound period, nitrobenzene pollution, emission, Songhua River

1. Introduction

Surface water in natural rivers is always an important resource with a variety of uses related to recreation, water supply, and aquatic life (Huang et al., 2014). However, the deterioration of the water quality of river water bodies has been a serious ecological and social problem throughout the world (Huang et al., 2010). Organic toxicant pollution, especially industrial waste, discharged into rivers may lead to the deterioration of the water quality of rivers and may result in a great deal of inconvenience when the water body is considered to be a source of drinkable water (Ouellette and Wilhelm, 2003; Fleeger et al., 2003; Wang et al., 2012). Hence, the simulation of the transport and variability of the organic pollutions in river networks has become one of major issues for effectively implementing water quality management and risk assessment programs.

Although evaluating the environmental fate and ecological risk of chemicals through in situ samples and laboratory experiments is considered to be relatively accurate and credible, it is costly and time-consuming. Furthermore, the difficulty of achieving continuous water quality sampling is a tremensdous barrier in water quality monitoring and forecasting (Senay et al., 2001; Marinov et al., 2009). In contrast with conventional laboratory analysis methods, hydrodynamic and water quality modeling have been considered to be the only viable approaches for the rapid forecasting of the distribution and ecological impacts of hazardous substance released into the aquatic environment when monitoring data are limited (Peng et al., 2002). Benefiting from the development of practical analysis technology and numerical models, numerous complex hydrodynamic models, such as COHERENS, FATE-3D, AQUATOX and EFDC (Carafa et al., 2006; Kobayashi et al., 2006; Lei et al., 2008; Wu and Xu, 2011), were developed with applications in various surface waters. However, most studies were conducted and validated in unfrozen water. Systematic studies and the modeling of emissions during the special situation of ice formation and melting in a frozen river,

* Corresponding author. Tel. : +86 10 68781950 (J. H. Wang), +86 10 64889681 (Y. H. Huang); fax: +86 10 68483367(J. H. Wang), +86 10 68483367 (Y. H. Huang).
E-mail address: wjh@iwhr.com (J. H. Wang); huangyh@igsnrr.ac.cn (Y. H. Huang).

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such as the Songhua River, are not yet adequate yet. For example, NB pollution event that occurred on 13 November 2005, which was used as a case study in this paper, the results from several studies of the NB pollution based on laboratory research and numerical model simulations varied substantially (e.g., the half-lives of NB in the environment ranging from 2 to more than 625 d) (Zhu et al., 2007; Li et al., 2008). These varied results may be due to a mimicked natural environment instead of the real conditions in the river. In particular, the frozen period beginning in November in the Songhua River was ignored. The variation in simulation results makes it difficult to evaluate the fate of NB in the environment. Hence, we need a dynamic numerical pollutant transformation simulation model in high latitude and cold regions that considers the solidification and emission mechanisms of a pollutant during ice formation and melting.

On the basis of these results, this paper presents a numerical model for the ice-bound period to describe the distribution and fate of NB in the aquatic environment based on the combination of a hydrodynamic sub-model and a water quality sub-model. The nitrobenzene pollution emergency that occurred in the Songhua River on November 13th, 2005, was adopted as an application of the developed model, which was caused by an explosion that occurred at a petrochemical plant in Jilin Province, China. It was reported that over 100 t of NB and related compounds were released into the Songhua River and then transported into the Heilongjiang River, which is an international boundary river (Zhang et al., 2004; UNEP, 2005). To avoid contamination of drinking water, the water supply of the downstream Harbin City in Heilongjiang Province was cut off for 4 days. Fortunately, the Songhua River began to be freeze, and the urban reach in Harbin was completely frozen at 20:00 on Nov. 30th. The NB pollutant stayed in the frozen water and threatened the drinking water downstream when it melted in the spring of the next year (Sun et al., 2011). For the purpose of rapid implement effectively water quality management and risk assessment after chemical pollution accidents, a simple and maneuverable model with empirical parameters and significant factors of NB emission effect in frozen rivers is more valuable than pure experiment or mathematical model. The case study of the Songhua River in 2005, as used in this paper, could assist decision makers in rapidly investigating the fate of NB and assessing the water quality risks in frozen rivers.

2. Materials and Methods

2.1. Study Area

The Songhua River, with a full length of approximately 1,840 km, is the largest tributary of the Heilong River (Amur) in Northeast China, as shown in Figure 1. It flows from the Changbai Mountains through Jilin and Heilongjiang Provinces with an annual discharge of 2,463 cubic meters per second. The river finally flows into the Amur River Valley before entering Russia. The river freezes from late November until March, and it has its highest flows when the mountain snow melts during the spring thaw.

As the fourth longest river in China, the Songhua River is

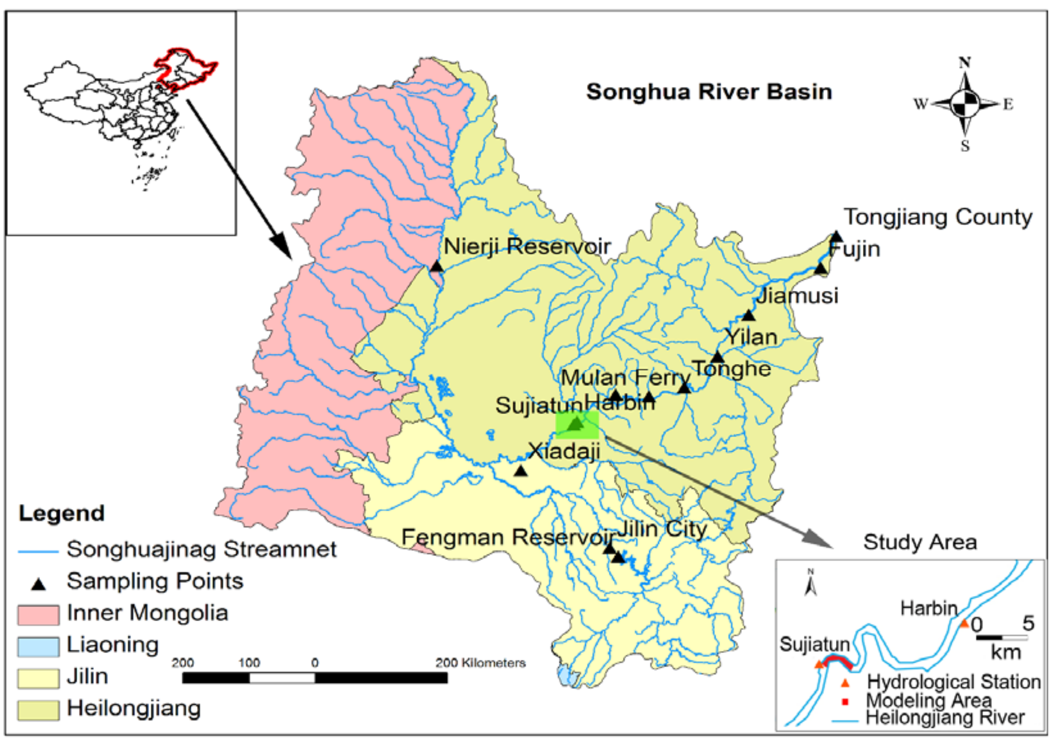

Figure 1. Location of study area.

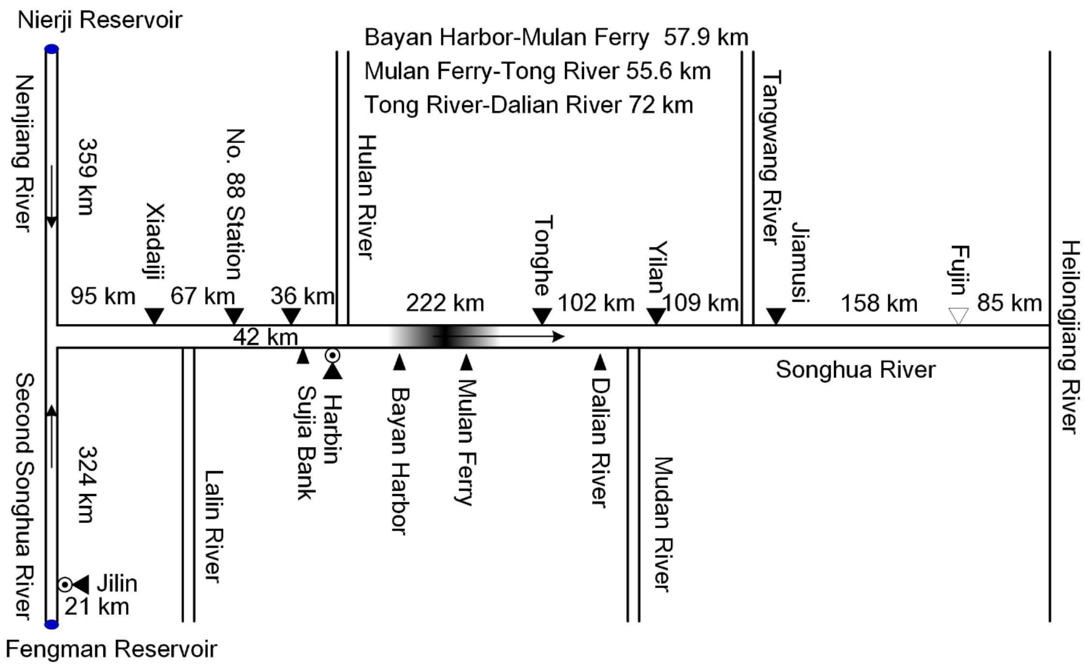

Figure 2. Conceptual illustration of the water system of the Songhua River pollution incident (The shaded part refers to the pollutant, and the arrow directs the water flow).

the main source of water for the cities and villages in the region. It provides an abundance of fishery resources (such as common carp, crucian carp and kaluga, and also contributes various utilities for human life, such as drinkable water, generated-electricity, water supply for industry and agriculture, flood prevention, shipping business, and environmental usage. Unfortunately, the Songhua River basin has suffered from frequent water pollution incidents in recent years. For example, the river was contaminated with NB in November of 2005 from the explosion at Jilin Petrochemical, leading to a shutdown of Harbin's water supply and threats of a Russian lawsuit against China. The spill stretched 80 kilometers and eventually reached the Amur River in Russia (Figure 2). In July 2010, several thousand barrels were washed away by floods from two chemical plants in China's Jilin City, which polluted the river with toxic chemicals, such as trimethylsilyl chloride and hexamethyldisiloxane. Hence, improving the water quality of the river is significant to the regional economy and pulic health in the northeast part of China.

2.2. Sample Collection and Analysis

To estimate the distribution of nitrobenzene in ice and the transfer of nitrobenzene from water to ice, a survey of the Songhua River was performed on Nov. 24th, 2005, from the upper reach of the main stream of the Songhua River at Sifangtai Bridge in Harbin. The 6 samples of water and ice were collected from four stations with equal distances from each other along the river. In the process of field monitoring and water sampling, the samples were stored in brown glass bottles in the dark at a temperature of approximately 4 °C and were taken back to the laboratory for extraction within 6 hours. Chromatographic analysis of NB was performed by comparing with the standard plots generated from the samples of two chromatographic columns within 24 hours after extraction. The water samples were divided to two parts named Sample A and Sample B. Sample A was extracted directly and treated using chromatographic analysis for nitrobenzene. Sample B was placed under the condition of -20 °C until half of the water was turned into ice; then, the water phase of Sample B (named Sample C) and the ice phase of Sample B (namely Sample D) were treated separately. Sample C was analyzed in the same way as Sample A, and Sample D was subjected to chromatographic analysis for nitrobenzene after melting (See Figure 3).

2.3. Model of Nitrobenzene Emission during the Ice-Bound Period

Modeling the fluid flow motion of surface runoff is performed prior to evaluating the nitrobenzene emissions in the river. Ice covers are common on rivers during the cold winter of northern China. The characteristics of different water flow types can significantly affect NB emissions. Therefore, two different hydraulic theories are presented according to the condition of ice cover: a pressure flow model for ice-covered river reaches and the Saint-Venant (SV) equations (Hamid and Naief, 2014).The system of equations is written as follows:

Continuity equation:

$$\frac{\partial A}{\partial t} + \frac{\partial Q}{\partial s} = q \tag{1}$$

Motion equation:

$$\frac{\partial Q}{\partial t}+\frac{\partial}{\partial s}(\frac{Q^2}{A})+gA\frac{\partial z}{\partial s}+g\frac{Q}{C^2}\frac{|Q|}{AR}=0 \quad (2)$$

where Q is water flow (m^3/s), s is the distance along the channel (m), t is time (s), q is the side inflow from melting ice, (m^3/s), A is the over-current section area (m^2), g is the acceleration of gravity (m/s^2), C is the Chézy coefficient ($m^{1/2}/s$), and R is the hydraulic radius (m).

According to the characteristics of the watercourse in a frozen river, the hydraulic parameter equations of the hydraulic radius, wetted perimeter and comprehensive roughness coefficient during the ice-bound period were modified as follows (Martin et al., 1999):

$$R = A/\chi \qquad \chi=\chi_{riverbed}+\chi_{riversheet} \quad (3)$$

$$C = R^{1/6}/n_c \qquad n_c=(\frac{n_b^{3/2}+n_i^{3/2}}{2})^{2/3} \quad (4)$$

where χ is the average wetted perimeter (m), $\chi_{riverbed}$ is the wetted perimeter of the river bed (m), $\chi_{riversheet}$ is the width of the ice sheet (m), n_c is the comprehensive roughness coefficient ($s/m^{1/3}$), n_b is the river bed roughness coefficient ($s/m^{1/3}$), n_i is the ice sheet roughness coefficient ($s/m^{1/3}$) , and n_c is the average value of n_b and n_i, reflecting the comprehensive effect of the two elements.

Furthermore, the water flow continuity equation and motion equation are discrete, with the 4 difference schemes of the weighted average implicit scheme of Preissmann (Wang, 1989; Yang, 1993; Wu et al., 2004; Li and Wang, 2005). The discrete system of linear equations system was then solved by the chasing method. As shown in Figure 4, the backwater effect caused by ice cover can be simulated by modifying the key parameters of the wetted perimeter and roughness coefficient.

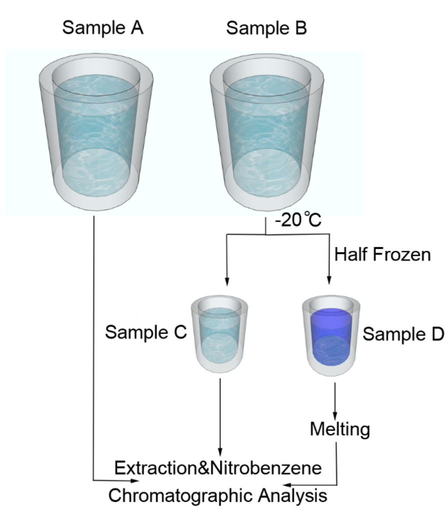

Figure 3. Flowchart of the nitrobenzene distribution test during ice formation.

The water quality equation during ice melting can be expressed as follows (Carroll et al., 2000; Kashefipour and Falconer, 2002; Lane et al., 2008; Kumar et al., 2010):

$$\frac{\partial c}{\partial t}+u\frac{\partial c}{\partial x}=E\frac{\partial^2 c}{\partial x^2}-kc+\frac{q_{ice}C_{ice}}{A} \quad (5)$$

where c is the pollutant density (μg/L), x is the vertical length of the reach (m), u is the average horizontal flow speed (m/s), E is the discretion index with the Fisher formula (m^2/s), k is the attenuation coefficient (s^{-1}), q_{ice} is the lateral inflow of melted ice (m^2/s), and c_{ice} is the pollutant density (μg/L).

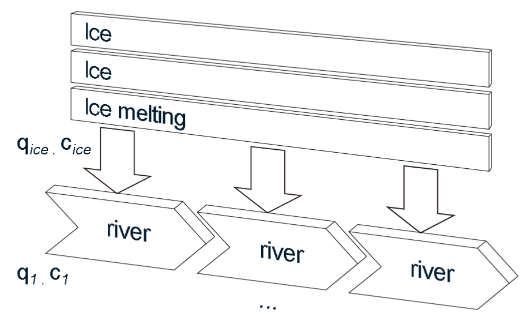

Figure 4. Flowing process of pollutant melted from ice.

3. Results and Discussion

3.1. Nitrobenzene Distribution from In Situ Samples

According to the Standard of the P.R.C. of "*Water Quality-Determination of Nitroaromatics by Gas Chromatography (HJ648-2013)*" and the laboratory experiment mentioned above, nitrobenzene concentrations were analyzed in three types of samples, including original water, water phase and ice phase. As shown in Table 1, the nitrobenzene concentrations are the highest in samples of the water phase, ranging from 0.491 to 0.693 μg/L. The nitrobenzene concentrations are the lowest in samples of the ice phase, ranging from 0.018 to 0.044 μg/L. This clearly indicates that the nitrobenzene concentrations in the ice phase samples were only approximately 10% of those in the original water. Therefore, we defined μ, which is the ratio of the nitrobenzene concentration in ice to that in water in a polluted river segment, as 0.1 in this paper (Pujol and Sanchez-Cabeza, 1999; Li et al., 2008; Liu et al., 2008; Zhang et al., 2010; Wang et al., 2012).

Table 1. Nitrobenzene Concentrations in Original Water Samples, Water Phase Samples and Ice Phase Samples

No.	Original water		Water phase		Ice phase		μ
	Volume (mL)	Concentrations (μg/L)	Volume (mL)	Concentrations (μg/L)	Volume (mL)	Concentrations (μg/L)	
1	200	0.39	175	0.693	225	0.044	0.113
2	200	0.37	180	0.650	220	0.040	0.108
3	200	0.33	200	0.533	200	0.040	0.121
4	200	0.32	160	0.590	240	0.043	0.134
5	200	0.29	175	0.537	225	0.018	0.062
6	200	0.27	180	0.491	220	0.027	0.100

3.2. Simulation

For the nitrobenzene pollution emergency that occurred in the Songhua River on Nov. 13th, 2005, it would take approximately 25 days for the pollutant to travel from the petrochemical plant (the site of the explosion) to the outlet of the river basin. However, the nitrobenzene remaining in the ice deteriorated the water quality as the ice started melting in April of 2006. In this paper, we simulated the nitrobenzene emission during the ice-bound period from April 11th to April 27th of 2006. We defined the attenuation coefficient and the initial density of nitrobenzene at the upper reach (c_0) as 0. Furthermore, the nitrobenzene emission was assumed to be consistent and commensurate with ice melting. Additionally, the density and melting speed were set to the highest level, and the simulation reach started from the Sujiatun section, which was 16 km above to 2.5 km below the drinking water intake of Harbin City (Figure 1). The maximum nitrobenzene concentration for the ice in the simulation is:

$$C_{ice} = \mu \times C_{\max} \tag{6}$$

where C_{max} is the peak value of nitrobenzene that the pollutant reached (μg/L or g/m^3) and μ is the ratio of the nitrobenzene concentration in ice to that of river segments contaminated by the pollutant. In the Sujiatun segment, the nitrobenzene concentrations reached a maximum of 0.58 μg/L at midnight on Nov. 25th, 2005. As a result, the maximum value of the nitrobenzene concentrations was:

$$C_{ice} = 0.58\mu g/L \times 0.1 \approx 0.06\mu g/L \tag{7}$$

According to the Hydraulic Yearbook of Songhua River 2006, the ice melting speed and river width were set to 1.25 cm/d and 360 ~ 400 m. Then, the lateral inflow of the melted ice of a river reach of 1-km length was calculated as:

$$\begin{aligned} q_{ice} &\approx 1.25 \times 10^{-2} (m \cdot d^{-1}) \times 400(m) \\ &= 5(m^2 \cdot d^{-1}) = 5.8 \times 10^{-2} (m^2 \cdot s^{-1}) \end{aligned} \tag{8}$$

Based on the laboratory results and the simulation of nitrobenzene emissions during the ice-bound period, the variations in the nitrobenzene concentration of the 2.5 km downstream Sujiatun segment after April 11th are shown in Figure 5. The increase in the nitrobenzene concentration was 0.02 ~ 0.025 μg/L. Since the nitrobenzene pollution emergency located at Jilin City, which was 1,250 km from the end of the Songhua River in the Tongjiang segment, the upper limit of the nitrobenzene concentration in water flowing into the Amur River should be:

$$C_{max} = 1250(km)/2.5(km) \times 0.025(\mu g/L) = 12.5(\mu g/L) \tag{9}$$

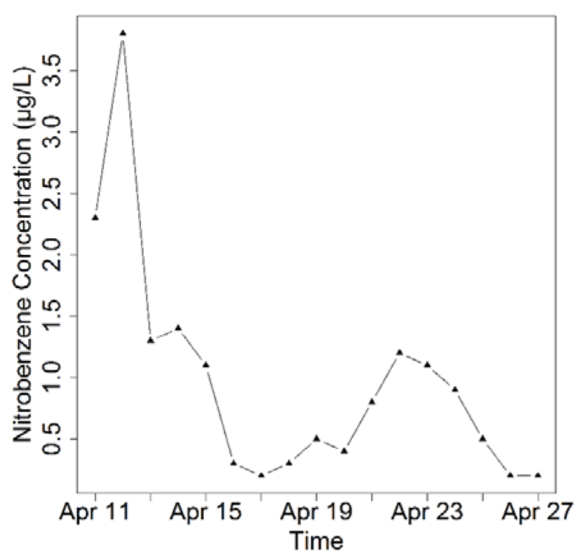

Figure 5. Nitrobenzene concentrations variations at 2.5 km downstream from the Sujiatun segment.

3.3. Validation of the Simulation

To evaluate the accuracy of the simulation more precisely, a comparison was conducted between the evaluation value and the in situ measurement value. The ground-based measured nitrobenzene concentrations of the Sujiatun segment were measured by the Environmental Protection Bureau of Heilongjiang Province from April 11th to April 27th of 2006.

Table 2 shows the deviation between the nitrobenzene concentrations simulated by the proposed algorithm and the results from ground-based measurements in the Sujiatun segment from April 11th to April 27th of 2006. Although the rela-

Table 2. Accuracy Evaluation of the Simulation at 2.5 km Downstream from the Sujiatun Segment

Date	Simulation (μg/L)	In-situ measurement (μg/L)	Error (μg/L)	Relative Error (100%)
2006-4-11	2.3	4	-1.7	-42.50
2006-4-12	3.8	5	-1.2	-24.00
2006-4-13	1.3	2	-0.7	-35.00
2006-4-14	1.4	1	0.4	40.00
2006-4-15	1.1	0.9	0.2	22.22
2006-4-16	0.3	0.7	-0.4	-57.14
2006-4-17	0.2	0.7	-0.5	-71.43
2006-4-18	0.3	0.6	-0.3	-50.00
2006-4-19	0.5	0.5	0	0.00
2006-4-20	0.4	0.4	0	0.00
2006-4-21	0.8	0.6	0.2	33.33
2006-4-22	1.2	1	0.2	20.00
2006-4-23	1.1	1	0.1	10.00
2006-4-24	0.9	0.8	0.1	12.50
2006-4-25	0.5	0.7	-0.2	-28.57
2006-4-26	0.2	0.5	-0.3	-60.00
2006-4-27	0.2	0.4	-0.2	-50.00

tive error range is from -71 to 40%, with a MRE of -16.5%, nearly all of the absolute errors were less than 0.5 μg/L, with a RMSE of 0.6 μg/L. The absolute value of error is low for water quality monitoring. It is shown in Figure 6 that the differences are slight between the nitrobenzene concentration values estimated by the model and the ground-based measurements in the Sujiatun segment. Their slope and R^2 of a y = x line analysis were 1.318 and 0.877, respectively. With consideration of the error associated with the in situ measurement and the advantage of the temporal and spatial continuity of the results, it is concluded that the results of the algorithm are acceptable.

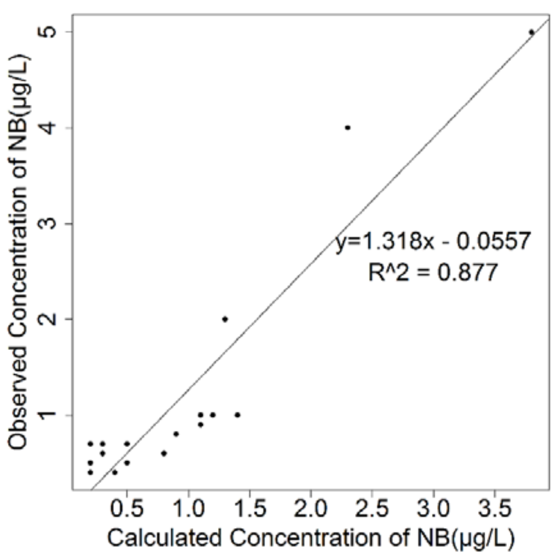

Figure 6. Scatter plot of the estimated NB concentration versus in-situ measurements in the Sujiatun segment from April 11th to April 27th of 2006.

4. Conclusions

In recent years, several events resulting in the spillage of serious toxic substances (e.g., NB) occurred in the Songhua River may pose a danger to the aquatic environment as well as public health. However, the NB pollution simulation results varied substantially in frozen rivers, such as the Songhua River. In this study, we developed a NB transport model that was appropriate for estimating NB emissions in frozen rivers. The model couples the modified pressure flow model, the Saint-Venant (SV) equations and water quality equations during the ice-bound period. The results of the analysis showed that the NB concentrations in water are higher than in ice and that the transfer radio of NB from water to ice was approximately 10%. Based on the in situ measured parameters of the NB concentrations in ice and the melting speed of the ice, the model was applied to a case study of NB water pollution in the Songhua River, China, in 2005, and its performance was generally satisfactory compared with the field observations of NB. The evaluation of the time-series accuracy of the simulation at 2.5 km downstream from the Sujiatun segment showed that the RMSE between the simulated NB concentrations and ground-based measurements was 0.6 μg/L. The slope and R^2 of a y = x line analysis were 1.318 and 0.877, respectively. The predicted up-limited value of NB emission from ice melt to the Amur River was 12.5 μg/L, which is consistent with the water quality monitoring results in the spring of 2006. These results indicate that the developed model is capable of assessing the spatial and temporal variability of the fate of NB in frozen rivers.

In this paper, we proposed a NB emission effect model for frozen rivers based on a petrochemical plant explosion, which is useful to emergency response plan for NB accident pollution. However, to substantiate its significance of extension to the other frozen rivers in China, even if NB accidents and related data were scarce, further comparison among different frozen rivers is very important. Furthermore, for the aims of emergency response plan for NB accident pollution, the model is simple and maneuverable for rapid government decision. Herewith, we ignored several factors such as climatic parameters of precipitation and temperature, which would affect the emission radio or concentration radio of NB in frozen rivers by water flow. They are necessary to be improved in further research. In spite of the limitations mentioned above, the validation showed that the model could be considered to be an auxiliary assessment tool for providing a necessary data reference for ecological risk and human health assessments of water pollution occurring during an ice-bound period.

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