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Statistical-physical method for simulating the transport of microplastic-antibiotic compound pollutants in typical bay area ☆

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ABSTRACT

Microplastics and antibiotics are emerging pollutants in the environment and have received widespread attention globally. In coastal areas, microplastic and antibiotic pollution is ubiquitous and often overlapping. Microplastic-antibiotic compound pollutants that are formed through adsorption have thus become a major concern. However, modeling knowledge of microplastic transport in coastal areas is still limited, and research on the impact of compound pollutants caused by Polythene (PE)-antibiotics in such settings is in early stages. In this study, using a lattice Boltzmann method (LBM) and temporal Markov method (TMM) under a statistical-physical framework, we simulated pollutant transport and PE-antibiotic compound pollutants in coastal areas. First, a series of models are proposed, including an LBM wave-current coupling model, an LBM antibiotic transport model, an LBM particle-tracking model, a TMM microplastic transport model and the final LBM-TMM hybrid compound pollutant model. Then, the suitability and applicability of the models was validated using experimental data and numerical simulations. Finally, the models were applied to a study area, Laizhou Bay (China). The simulation results demonstrate that adsorption will reduce the concentration of antibiotics in the water environment. Within 44 days, the adsorbed antibiotic carried by PE particles migrate further, and the width of the pollution zone escalates from 234.2 m to 689.0 m.

1. Introduction

Microplastics are plastic particles with a size less than 5 mm, with the term first introduced in 1968 (Crawford and Quinn, 2017). At present, it has been recognized that microplastic pollution exists widely across a variety of environmental systems, including coastal areas where high levels have been detected as a result of human activity (Xu et al., 2023). Microplastic pollution in water environments mainly includes two aspects: direct and indirect pollution (Everaert et al., 2018). Direct pollution refers to pollution attributed to toxic and harmful substances released by microplastics, whereas indirect pollution refers to pollution caused by the chemical substances that can be carried by microplastics. Due to their high specific surface area and strong hydrophobicity, microplastics can adsorb heavy metals and persistent organic pollutants and act as carriers in aqueous environments (Guo et al., 2020), which poses potential risks to the environment and human health.

As an effective group of antibacterial drugs, antibiotics are extensively used by humans as pharmaceuticals as well as in agriculture and aquaculture. Only a small fraction of antibiotics is partially degraded in aquatic systems; most of them are residual (Kümmerer, 2003; Zhang et al., 2021). For instance, Andreozzi et al. (2003) found that the degradation coefficient of erythromycin is about 0.82% per day. Antibiotic residues can reach aquatic and terrestrial environments, where they can have detrimental effects (e.g., antibiotic resistance among pathogens infecting cultured animals and human) (Kümmerer, 2003, 2009; Rakib et al., 2023). Li et al. (2018) demonstrated that antibiotics can be adsorbed by microplastic particles. Additionally, antibiotics transform into different forms (cations, neutral ions and anions) under varying environmental conditions, especially pH. Such conditions affect the adsorption process between antibiotics and microplastics. Therefore, different types of antibiotics display distinct adsorption characteristics across varying environments (Li et al., 2018). Because antibiotics and microplastics are ubiquitous in aquatic environments, the adsorption

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of antibiotics by microplastics likely contributes to the large-distance transport of formative compound pollutants, increasing the risk to the aquatic environment (Imran et al., 2019).

Understanding microplastic-antibiotic compound pollution is still in its infancy, and relevant studies are limited, although some researchers have investigated the biological toxicity of compound pollution through field sampling, on-site monitoring and laboratory experiments. Zhang (2019) first confirmed the coexistence of microplastics and antibiotics in Taihu Lake in China and then evaluated the effect of microplastics on the distribution and bioaccumulation of roxithromycin in fish as well as their interactive biological effects. In laboratory experiments, Shan et al. (2020) studied the ability of ryegrass to remove ciprofloxacin in the presence of polystyrene, which increased the toxic effects of ciprofloxacin on plant growth. Additionally, there are relevant studies on the influence of microplastics on the antibiotic resistance genes in living organisms (Sathicq et al., 2021; Zhang et al., 2022). However, current research on these two emerging pollutants mainly focuses on either microplastics or antibiotics in the aquatic environment, whereas research on the impacts of microplastic-antibiotic compound pollution is still scarce. In particular, the influence of microplastics on the amount of antibiotics in waters through adsorption and the prediction of the spatial distribution of compound pollutants due to transport remain open questions. Therefore, it is useful to develop models for compound pollutant transport, which is a basis for understanding the extent of compound pollution in water environments.

In coastal areas, wave-current conditions, boundary conditions and topography make the hydrodynamic and pollutant transport processes complicated, with multiple important temporal and spatial scales playing a role (Cole et al., 2011). A convenient and efficient approach is to apply tested and validated numerical simulation tools to study hydrodynamics, pollutant transport and particle migration. Traditional fluid mechanics approaches are manifold, spanning Eulerian and Lagrangian methods. However, relatively speaking, computational costs can be quite high, especially for tracking the movement of large numbers of individual particles (Xu et al., 2014). Statistical physics approaches that build a bridge between micro- and macro-levels serve as tools to improve speed and efficiency (Huang, 2001). The lattice Boltzmann method (LBM) and Markov transport models are two common methods within such a statistical-physics framework that are widely used in various complex hydrological systems and play an increasingly important role in water environment simulations (Xing et al., 2020; Sherman et al., 2021).

LBM is a mesoscopic numerical simulation method, lying somewhere between macroscopic and microscopic ones (Zhou, 2002). It obtains the velocity distribution of particles at mesoscopic scales using a Lagrangian viewpoint and then uses principles from statistics to establish the relationship between the mesoscopic particle distribution and macroscopic physical quantities. LBM has developed rapidly in recent decades due to its advantages, such as ease of programming and ability to handle complex boundaries. To date, LBM has been extensively applied in various contexts relating to water environments, including hydrodynamics and pollutant transport (Zhou and Liu, 2013; Liu et al., 2020; Xing et al., 2020). While powerful, LBM still needs to process large sets of spatiotemporal distribution data. Therefore, regarding the transport rules of a large number of microplastics over long times and distances, we introduce another statistical simulation method based on Markov methods to improve efficiency. Markov methods are based on Markov chains and include many other frameworks of continuous-time random walk (CTRW) models, which can capture the motion characteristics of particles by sampling from a probability distribution to describe the large-scale transport process of a large number of particles efficiently and accurately (Borgne et al., 2011; Anna et al., 2013; Sherman et al., 2017). That is, given a spatiotemporal probability distribution containing the motion characteristics of a large number of representative particles, Markov models can predict their large-scale transport processes. This can considerably save the time required to perform calculations. Le Borgne at al. (2008) proposed a correlated CTRW, introducing the notion of a transition matrix to incorporate the velocity correlation effect of particles, which is regarded as ever present in many hydrological systems (Bolster et al., 2014). This correlated model can accurately capture particle transport in highly complex and heterogeneous hydrological environments (Le Borgne at al., 2008; Sherman et al., 2017, 2021), including in the context of microplastic transport in open channel flows (Xing et al., 2022).

Laizhou Bay (37.65° N, 119.28° E ~ 37.68° N, 120.22° E) is one of the three largest bays in the Bohai Sea (Fig. 1). Favorable natural conditions and water quality make it rich in fishery resources. However, it also causes pollution, including microplastics and antibiotics. Additionally, along the coastline, there are more than 20 large rivers flowing into the bay. These rivers provide input routes for pollutants, resulting in a continuous terrestrial input of domestic and industrial wastewater. The Xiaoqing River, which is a typical river in this region, is the main pollution source of Laizhou Bay. The quality of its water will directly affect the regional economies and marine resources.

In order to simulate pollutant transport and PE-antibiotic compound pollutants in coastal areas, in this study, we will establish a series of water environment models, with a focus on a typical bay area. First, using LBM and temporal Markov methods (TMM), a two-dimensional (2D) wave-coupling hydrodynamic model, a 2D antibiotic transport model, a 2D particle-tracking model and a one-dimensional (1D) microplastic transport model were proposed. We then explore a 1D-2D LBM-TMM hybrid simulation method to investigate the transport of microplastic-antibiotic compound pollutants. After validation of the models, the hybrid simulation method is applied to our test area.

2. Model framework

2.1. LBM

2.1.1. Hydrodynamic model

As is typical in coastal areas, 2D shallow water equations, including the continuity equation and momentum equation, are commonly used to describe water flows and are stated as follows:

$$\begin{cases} \dfrac{\partial h}{\partial t} + \dfrac{\partial (hu_j)}{\partial x_j} = 0, \\ \dfrac{\partial (hu_i)}{\partial t} + \dfrac{\partial (hu_iu_j)}{\partial x_j} = -g\dfrac{\partial}{\partial x_i}(\dfrac{h^2}{2}) + \nu\dfrac{\partial^2 (hu_i)}{\partial x_j \partial x_j} + F_i, \end{cases} \tag{1}$$

where i and j represent the spatial direction indices following the Einstein summation convention; x_j represents the two Cartesian coordinates, x and y; u_j represents the velocity components, u and v, corresponding to those in the x and y directions, respectively; h represents the water depth; t is time; ν represents the eddy viscosity; and F_i represents the force term, and the calculation method can be found in Supplementary Information 1. We solve the above set of equations using an LBM approach, the details of which are included in Supplementary Information 2.

2.1.2. Advection-dispersion model

The processes of antibiotic dispersion, advection and decay can be described with the 2D advection-diffusion equation as follows:

$$\frac{\partial (hc)}{\partial t} + \frac{\partial (hcu_i)}{\partial x_i} = \frac{\partial}{\partial x_i}[D_{ij}\frac{\partial (hc)}{\partial x_i}] + S_c, \tag{2}$$

where c represents the depth-averaged antibiotic concentration; S_c represents a source term that can be calculated as $S_c = -hcD_d$, where D_d represents the decay coefficient including the degradation process of the antibiotic and adsorption process with suspended particles; and D_{ij} represents the dispersion coefficient. Likewise, we use an LBM approach to solve this equation, which is detailed in Supplementary Information 3.

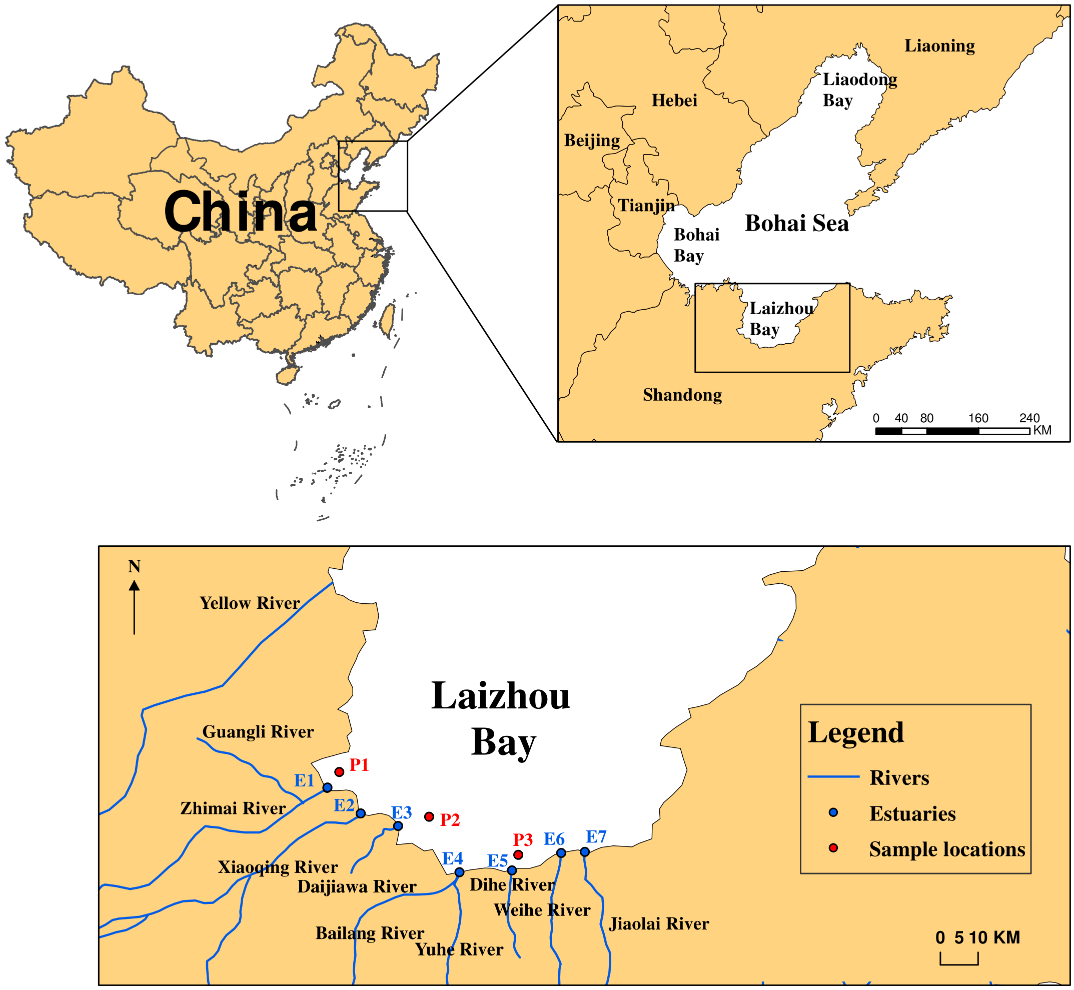


Fig. 1. Map of Laizhou Bay and its adjacent rivers and estuaries.

2.1.3. Particle-tracking model

Polyethylene (PE) in many forms is commonly used in the manufacturing of film, packaging materials, containers among others. In recent years, PE has become the largest category of total plastic production, accounting for 36% (Geyer et al., 2017). Meanwhile, spherical and cylindrical PE microplastics are widely present on the surface water of Laizhou Bay (Teng et al., 2020). Therefore, PE was selected as our target type of microplastics in this study. Considering their small size and hydrophobicity, PE particles can be regarded as conceptual Lagrangian particles. Each particle is displaced solving a second-order Runge-Kutta scheme applied to the following equations, which is derived from (Lavieville et al., 1995).

$$\begin{cases} \dfrac{\partial x_{pj}}{\partial t} = u_{pj}, \\ \dfrac{\partial u_{pj}}{\partial t} = \dfrac{3\nu C_D Re_p \rho}{4 d_p^2 \rho_p}(u_j - u_{pj}), \end{cases} \tag{3}$$

where C_D is the local drag coefficient. The empirical relation for C_D by Schiller and Naumann (1935) was employed (it should be noted that the Schiller-Naumann model applies to solid spherical particles), and details can be found in Supplementary Information 1 - Part (2); ν represents the eddy viscosity, as in Eq. (1); Subscript p represents the PE particle. Therefore, x_{pj}, u_{pj}, d_p and ρ_p represent the position, velocity, size and density of the PE particle, respectively; Re_p represents the Reynolds number of PE particles governed by $Re_p = (d_p|u_j - u_{pj}|)/\nu$; and u_j represents the velocity components as same as the u_j in Eq. (1).

It should be noted that the disintegration or aggregation of particles are not included in the LBM particle-tracking model. We do not account for buoyancy forces of particles as vertical displacements are not included with the shallow water equation. Particles can be thought of as moving along the sea surface. Additionally, the coupling between particles phase and water phase is one-way coupling.

Thus, the macroscopic $x_{pj}^{t+\Delta t}$ and $u_{pj}^{t+\Delta t}$ can be calculated to first-order in time using an explicit scheme as follows:

$$\begin{cases} x_{pj}^{t+\Delta t} = u_{pj}^{t}\Delta t + x_{pj}^{t}, \\ u_{pj}^{t+\Delta t} = \dfrac{3\nu C_D Re_p \rho \Delta t}{4 d_p^2 \rho_p}(u_j^t - u_{pj}^t) + u_{pj}^t. \end{cases} \tag{4}$$

2.2. Markov method – temporal Markov model

Predicting with the Markov method is based on a 1D random process. We project the transport process in the direction of the mean flow and then analyze the Lagrangian velocity statistics along the projected particle trajectories. The projection trajectory of each particle in time and space can be described by the Langevin equation,

$$\begin{cases} x^{n+1} = x^n + v_t^n \Delta t, \\ t^{n+1} = t^n + \Delta t, \end{cases} \tag{5}$$

where v_t^n represents a stochastic process of the Lagrangian velocity field along the particle's projected trajectory at an equidistant time Δt. Generally, v_t^n can be randomly sampled from the trajectories. However, particles may have a strong correlation between successive steps in many hydrological systems, namely, particles that make a fast/slow transition in one step may often make a fast/slow transition in the next step, and the correlation will have an effect on the behavior of particle transport on a macroscale level. Therefore, a temporal Markov method (TMM) model is applied in this work.

A spatial transition matrix $T_{i,j}$ is introduced here in order to describe the correlation, and the calculation of $T_{i,j}$ can be found in Supplementary Information 4.

After obtaining the transition matrix, we can acquire the breakthrough curves (BTCs) after certain steps, which are a very standard transport metric to characterize the motion properties of particles in complex hydrological systems. Therefore, BTCs are used to validate the TMM model in subsequent sections. Spatial distributions of particles at a given time can also readily be predicted with this model.

2.3. LBM-TMM hybrid model

By virtue of the above statistical-physical method, the results of the 2D antibiotic concentration from LBM and the 1D spatial distribution from TMM are collected and will be the basis for the 1D-2D LBM-TMM hybrid (LTH) model. To achieve this, we must first quantify the adsorption of antibiotics on PE, which is a bridge between the LBM and TMM methods. Generally, the equilibrium partition coefficient (K_d) of antibiotics between PE and water is used to represent the adsorption capacity (Velzeboer et al., 2014). In this work, a quantitative structure-property relationship (QSPR) model is applied to predict K_d. QSPR is an effective tool to reveal the mathematical relationship between the molecular structure of a compound and its environmental behaviors (Wei et al., 2017; Bakire et al., 2017) and relevant transport parameters, which are calculated theoretically using a variety of methods, including quantum chemistry (description of the QSPR model can be found in Supplementary Information 5). Then, based on the K_d calculated by the QSPR model, we apply them to the numerical simulation method for investigating the transport of PE-antibiotic compound pollutants.

To predict a large number of particles on long-distance transport efficiently, an LTH model is established in this work to intelligently combine the fine simulation of the 2D-LBM with the efficient simulation via the 1D-TMM. Using the idea of dimensional reduction, the microplastic number follows the normal distribution of the advection-diffusion equation in the vertical direction of the mean flow to simulate the transport of compound pollutants. The detailed implementation process of the LTH model can be found in Supplementary Information 6.

3. Validation and application

In this work, we have proposed a novel 1D-2D hybrid model to investigate the transport of PE-antibiotic compound pollutants in a representative shallow coastal region. Thus, the lattice Boltzmann model for simulating antibiotic transport and the temporal Markov model for simulating PE particle migration should be validated first. Then, the proposed LTH model is applied to our representative test case – Laizhou Bay.

3.1. Validation of the lattice Boltzmann model

3.1.1. Data acquisition and model inputs

The bottom elevation of Laizhou Bay can be plotted, as shown in Fig. S4. Tidal-level data in Laizhuo Bay were extracted from Liu et al. (2017). To validate the advection-diffusion model, field data of antibiotics in Laizhou Bay were also collected. We obtained the experimental data from the Zhang et al. (2012), in which four types of antibiotics with high prevalence were selected, including erythromycin (ETM), enoxacin (ENO), trimethoprim (TMP) and sulfamethoxazole (SMX), and the sample sites included the estuaries of adjacent rivers and the sampling locations (Fig. S4).

Degradation is an important decay process for environmental antibiotics, and it depends on the chemical structure and environmental factors. Meanwhile, adsorption between antibiotics and suspended sediment is also a major removal process of antibiotics from water. The adsorption process is generally expressed by the partition coefficient (K_d). The Decay details of the target antibiotics ETM, ENO, TMP and SMX are shown in Table S2 (Supplementary Information).

Detailed and specific parameter setting and boundary conditions can be found in Supplementary Information 7.

3.1.2. Validation

The hydrodynamic model is the basis for all the model frameworks throughout the paper and must first be validated. As shown in Fig. 2(a) and Fig. 2(b), the maximum velocity occurs northwest of the bay mouth, and the minimum value occurs along the coastline. The proposed hydrodynamic model was validated to be able to generate reliable results compared with the previous results (Lv et al., 2017; Xing et al., 2020).

Then, simulated concentrations are compared to the vertically integrated measurements (Zhang et al., 2012) at the near-shore locations P1, P2, and P3 (Fig. 1) to validate the lattice Boltzmann model. Fig. 2(c) shows the comparison between the simulations and measurements for different antibiotics and locations after 42 hours. All the absolute value of percentage errors are less than 15%. Since the samples are acquired at approximately 50 cm below the water surface, this inevitably involves some errors in validating the 2D depth-averaged model. Therefore, the results obtained by the proposed model are deemed sufficiently accurate for dealing with the transport of antibiotics in Laizhou Bay.

3.2. Validation of the temporal Markov model

3.2.1. Particle-tracking data acquisition

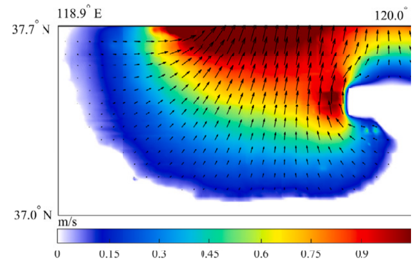
The temporal Markov model is employed to simulate the transport of the PE particles. To this end, we need to collect the trajectories of a large number of particles to determine the spatial transition matrix, which can provide enough information about particle movements. The particle-tracking model based on LBM is used to generate the trajectories of particles.

In the LBM particle-tracking model, 10^6 PE particles were released simultaneously in the first time step at the estuary of the Xiaoqing River. Based on the previous literature and field research of the study area (Teng et al., 2020), the size and density of the PE particles in this study are determined as 1.66 mm and 0.956 g/cm^3, respectively. The model was simulated for 24 hours in real-world time, and particle-position data were recorded every minute (details on the data analysis for particle-tracking data can be found in Supplementary Information 8).

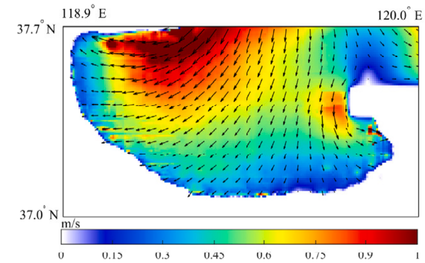
3.2.2. Validation

Considering the two reciprocating processes in the particle-tracking model, the entire simulated time can be regarded as two continuous temporal increments. The stable segments of projected trajectories during each reciprocating process provide us with an isochronal time, namely, $\Delta t = 30000$ s. Therefore, the spatial distribution $\psi(x)$ of particles can be obtained at Δt (Fig. S6 (a)). Then, the spatial transition matrix can be calculated (Fig. 2(d)), there is an obvious diagonal tendency, reflecting a strong correlation. We use $k = 20$ classes in this study, which has been shown to be sufficient to acquire correlated effects and generate reliable results (Borgne et al., 2011).

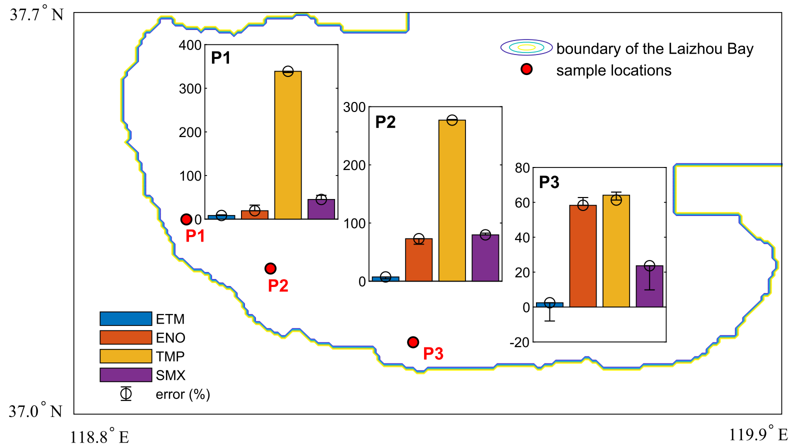
The comparison at $2\Delta t$ between the predicted BTC from the TMM and the LBM simulations is shown in Fig. 2(e). At this point, the mean absolute percentage error between the TMM predictions and LBM simulations is 6.76%, which demonstrates that the TMM model can accurately reproduce the early, peak and late arrivals. Therefore, the TMM



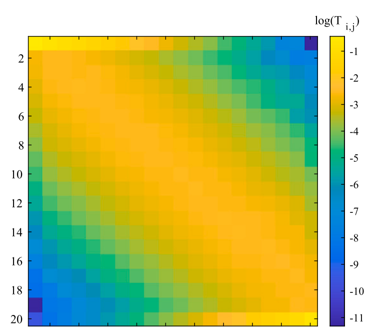
(a) The ebb peak of Laizhou Bay in a tidal period



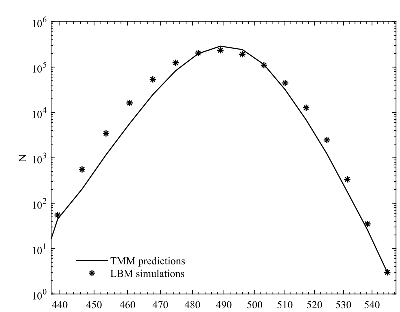
(b) The flood peak of Laizhou Bay in a tidal period



(c) Comparison of simulations and measurements



(d) Spatial transition matrix



(e) Comparison of BTC between the TMM predictions and LBM simulations

Fig. 2. Validation of the model framework: in subfigure (c), P1, P2 and P2 are sample locations corresponding to those in Fig. 1; histograms represent the concentrations (ng/L) of four antibiotics; and the percentage error (%) between the simulations (C) and measurements (C_m) is calculated by $(C - C_m)/C \times 100\%$ (it should be noted here the y-axis represents the concentrations of antibiotics as well as the percentage error. (For interpretation of the colors in the figure(s), the reader is referred to the web version of this article.)

model appears effective at capturing the motion properties of a large number of PE particles in a computationally efficient manner.

3.3. Setting up a scenario in Laizhou Bay

Under the proposed statistical physical framework, we established an LBM-TMM hybrid model to investigate the transport of PE-antibiotic compound pollutants. To apply the proposed hybrid model in Laizhou Bay, the following scenario is assumed:

i. Transport of antibiotics: Considering a sewage treatment plant near the estuary of the Xiaoqing River, the effluent was discharged continuously into Laizhou Bay for 30 minutes. The concentration of all the target antibiotics in the effluent was 100 mg/L.

ii. Transport of PE particles: As in Section 3.2.1, a number of 10^6 PE particles were released simultaneously at the estuary of the Xiaoqing River.

4. Results and discussion

4.1. Equilibrium partition coefficient of antibiotics

K_d can be determined by Eq. (S15) and is also shown in Table S1 (Supplementary Information). The K_d of ETM, ENO, SMX, TMP and

Table 1
Transport of PE-antibiotic combined pollutants in Laizhou Bay.

Day	Antibiotic	Max.CP (%)	W (m)	x (m)	$Before$ (g)	$After$ (g)	PI (%)
14th	ETM	5.64			-	-	0
	SMX	4.96	234.2	9835	-	-	0
	TMP	4.97			-	-	0
24th	ETM	2.26			8.29×10^{7}	8.31×10^{7}	0.24
	SMX	0.11	495.0	16960	0.2568	0.2569	0.039
	TMP	0.61			6.848×10^{-7}	6.852×10^{-7}	0.058
44th	ETM	1.22			6.03×10^{6}	5.95×10^{6}	1.36
	SMX	0.06	689.0	31200	1.873×10^{-9}	1.874×10^{6}	0.05
	TMP	0.35			1.421×10^{-19}	1.423×10^{-19}	0.14

CPFX are 238.78 L/kg, 174.58 L/kg, 8.87 L/kg, 50.12 L/kg and 216.77 L/kg, respectively. We find that $logK_d$ of ETM, ENO and CPFX are the same order of magnitude; therefore, ETM is chosen as a typical antibiotic among the three referred types of antibiotics to investigate the transport of compound pollutants. As a result, the target antibiotics selected for this study were ETM, SMX and TMP.

It has been proven that the adsorption equilibrium between PE particles and the aqueous phase is achieved after 80 hours (Yu et al., 2020); therefore, according to the calculation process of the 1D-2D LTH model, the equilibrium time was assumed to be the fourth day.

4.2. Impacts on antibiotic concentration

Due to the adsorption process between the antibiotic and PE particles, the antibiotic concentration of each grid is reduced. For the same grid at the same time, the adsorption amount of ETM is maximum, and the adsorption amount of TMP is minimum. Moreover, their equilibrium partition and decay coefficients satisfy the following relationship:

$$\begin{cases} K_{dETM} > K_{dTMP} > K_{dSMX}, \\ K_{deTMP} > K_{deSMX} > K_{deETM}, \end{cases} \tag{6}$$

which demonstrates that the decay process of antibiotics plays a greater role in the adsorption amount. For the same kind of antibiotic within 44 days, it is obvious that the concentration changes of TMP and SMX decrease significantly over time, which also emphasizes the importance of the decay process of antibiotics. However, the concentration changes in ETM (larger K_d but smaller D_d) increase slightly in the southeastern grids over time, which indicates that the accumulation effect of antibiotics with a smaller decay coefficient is stronger in the southeastern coastal area where the flow velocity is low. This will lead to a higher potential ecological risk in this area.

Furthermore, the percentage changes (CP) in the antibiotic concentration were calculated as

$$CP = \frac{\Delta c}{c(i,j)} * 100\% \tag{7}$$

The results within 44 days are shown in Fig. 3, and the maximum CP (Max.CP) of the three days for antibiotics are shown in Table 1.

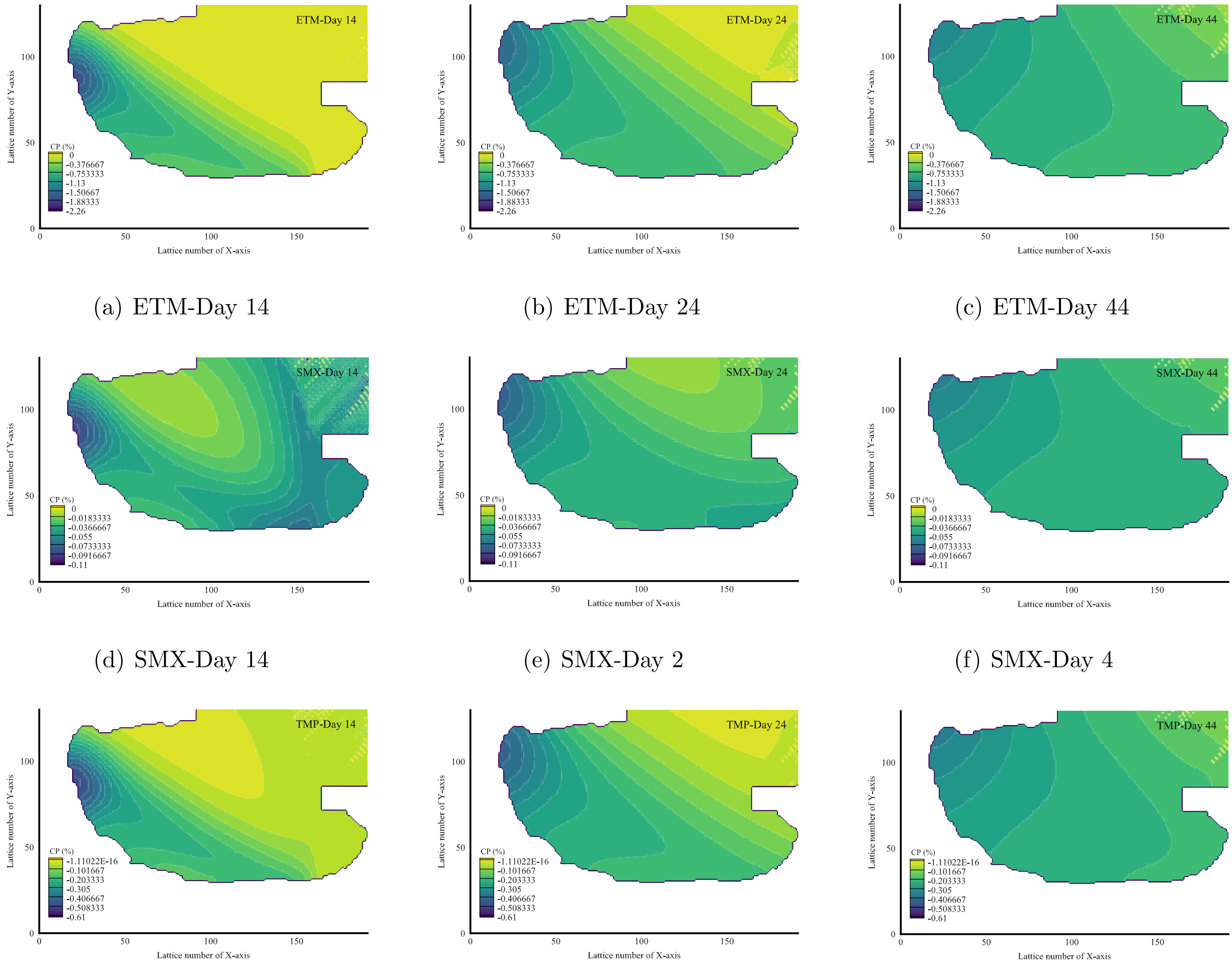
There is a decreasing trend of antibiotic concentration over time as the particles travel towards the outer part of the bay due to the hydrodynamic conditions including the turbulence due to terrain conditions and tidal boundaries. Among the three antibiotics, PE particles have the maximum effects on the percentage changes in the ETM concentration as well as the minimum effects on SMX, which is caused by the different equilibrium partition coefficients of the three antibiotics (Eq. (6)); that is, antibiotics with larger equilibrium partition coefficients will have greater percentage changes in their concentration.

4.3. Transport of compound pollutants

To investigate the transport of PE-antibiotic compound pollutants, the spatial distribution of antibiotic mass adsorbed by PE particles was plotted as shown in Fig. 4. The adsorption amount of PE particles by ETM is maximum, and that by TMP is minimum. This demonstrates that the adsorption amount depends on the decay coefficient of the antibiotic. During the transport process, all the curves gradually flatten due to the impacts of the advection and dispersion on PE particles. The head and tail of all the curves show fluctuations (especially on the 44th day, in which there are breakpoints at the head of the curves), as there are fewer particles at the edge of the contamination zone due to advection and dispersion. Then, we calculated the width of the contaminated zone (W), which can be defined by the distance difference between the slowest and fastest moving particles (shown in Table 1). Within 44 days, the adsorbed antibiotic carried by particles will migrate further with the transport of microplastics. The width of the pollution zone escalates from 234.2 m to 689.0 m.

Then, we compared the total amount of antibiotics in Laizhou Bay before and after adsorption to discuss the long-distance transport of the compound pollutants. Before adsorption, the total amount of antibiotics can be calculated from the LBM advection-diffusion model. After adsorption, the total amount is the sum of the antibiotic mass remaining in the aqueous phase and carried by PE particles. To compare with the spatial distribution results under the 1D Markov system, it is necessary to reduce the dimensionality of the antibiotic concentration results under the 2D Euler system; that is, the 2D antibiotic simulations obtained by LBM need to be reduced to 1D results along with the mean flow direction (projected line). As shown in Fig. 5, the antibiotic mass in Laizhou Bay gradually declines with increasing x (recall x represents the distance from the initial position of the particles), which results from the function of transport and diffusion. The slight fluctuations are attributable to the boundaries of the computational domain.

As illustrated in Fig. 5, the antibiotic mass in Laizhou Bay after adsorption was visibly less than that before adsorption due to the adsorption process and the decay of antibiotics. Then, we focus on some crucial segments whose x-values correspond to the peaks in Fig. 4. As shown in Table 1, $Before$ and $After$ represent the total antibiotic mass in Laizhou Bay. PI represents the percentage increase, which can be calculated by $(After - Before)/After \times 100\%$. On the 14th day, at approximately $x = 9835$ m, the total antibiotic mass between $Before$ and $After$ is basically the same. Over time, the antibiotic masses of the three antibiotics increased by different magnitudes around the x-values we focus on, which indicates that the compound pollutants raise the ecological risk in these areas. Meanwhile, antibiotics with larger equilibrium partition coefficients will have a greater PI. Taking the ETM as an example, PI increases by approximately six times from the 24th day to the 44th day. Therefore, microplastics can be used as carriers for long-distance transport to adsorb antibiotics, providing the formation of compound pollutants. Given enough time, the PI of the antibiotic amount near the location of the compound pollutants will rise



(a) ETM-Day 14 (b) ETM-Day 24 (c) ETM-Day 44

(d) SMX-Day 14 (e) SMX-Day 2 (f) SMX-Day 4

(g) TMP-Day 14 (h) TMP-Day 2 (i) TMP-Day 44

Fig. 3. The percentage changes in the antibiotic concentration in Laizhou Bay.

markedly, so there will be an increasing ecological risk of antibiotics in the sea.

4.4. Computational benefits and model limitations

We employed two statistical-physical methods in this work: LBM and TMM. Regarding LBM, the seemingly chaotic movements of a large number of particles can be statistically averaged to obtain a velocity distribution, which is the only unknown quantity when the particles' movements are at equilibrium. Given the velocity distribution, macroscopic quantities can be calculated. Although LBM uses a statistical method to solve the velocity distribution, in essence, it still tracks the movements of numerous discrete particles to obtain the macroscopic flow field. Therefore, its computational efficiency is much lower than that of TMM, which does not need to resolve large bodies of spatio-temporal data. The required CPU time between the two methods is calculated in MATLAB. The LBM particle-tracking model takes $\mathcal{O}(10^6)$ CPU seconds to run 10^6 particles in 24 hours, whereas the TMM model costs $\mathcal{O}(10^{-1})$ CPU seconds to predict the movements of particles in the same situation. This very large gap highlights the great advantage of the high efficiency of the TMM model. However, without LBM simulations, an efficient TMM model cannot be properly implemented since there is no model input. Therefore, the complete reduction in the computational cost has not been truly realized. However, since the input spatial distribution of particles can be captured before completing a full LBM simulation, the TMM model still has computational advantages over LBM.

The innovative 1D-2D hybrid model uses an approach of dimensional reduction to simplify microplastic transport into an average behavior in the mean flow direction. Then, its influence on the antibiotic quantity is investigated. This indeed greatly reduces the computing cost, making it extremely adaptive for efficiently predicting the transport of a large number of microplastics for a longer time. However, the 1D Markov model is quite inadequate, as the 2D Markov model (Mose et al., 2019) will have much broader application prospects. Therefore, we could involve the 2D Markov model in the statistical-physical framework, which is worth further study.

PE particles, which are prevalent in Laizhou Bay, were selected as our target type of microplastic to proceed with the research. However, the physical properties (such as the size, density, shape, and surface texture) or aging effects of the microplastics detected in coastal water may vary significantly depending on the category of polymer and the du-

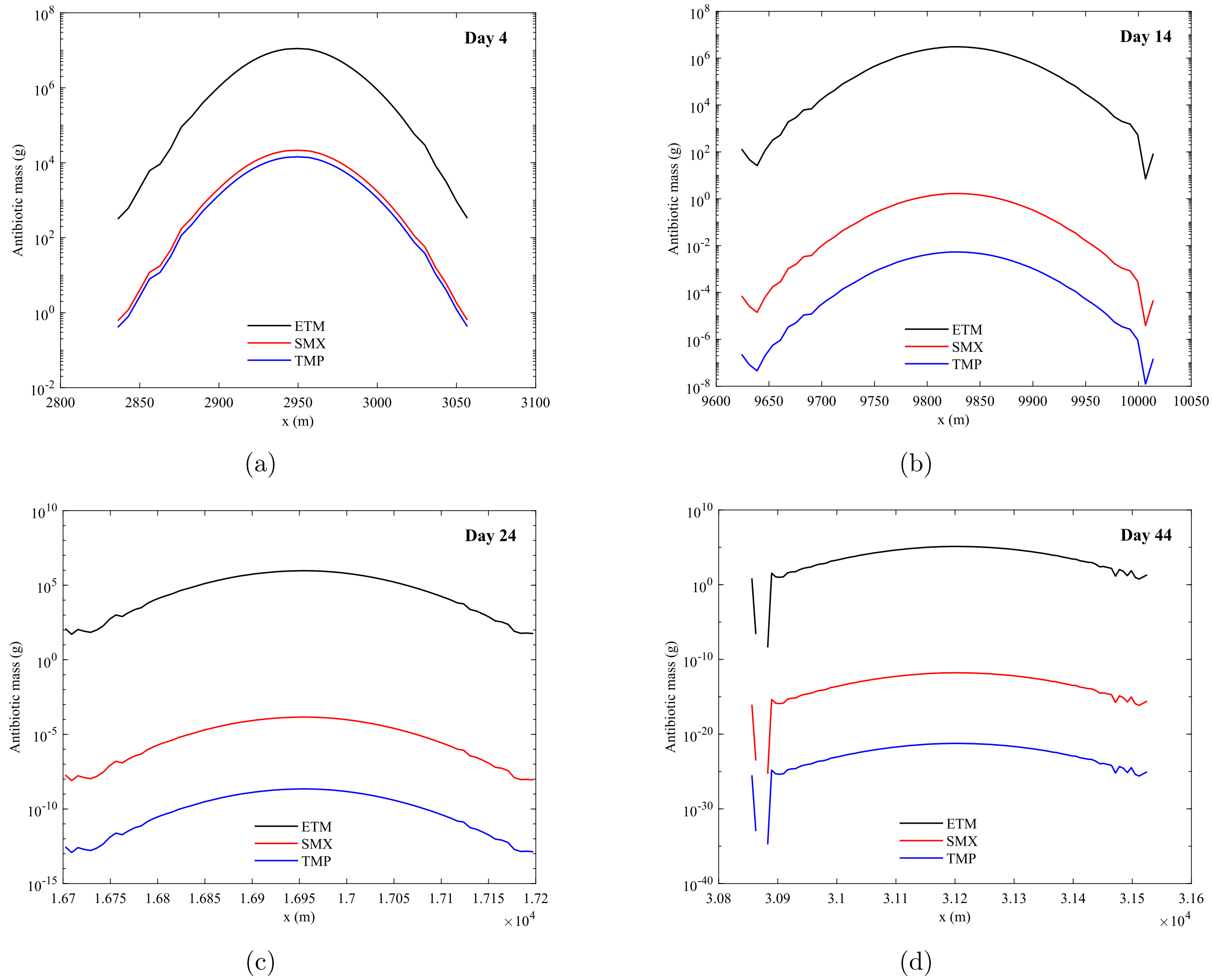


Fig. 4. The spatial distribution of PE-antibiotic compound pollutants.

ration of its exposure in the environment (Bao et al., 2022; Liu et al., 2022). The referred properties have a powerful influence on hydrodynamic processes such as suspension and settlement, thus affecting the transport process. The physical and dynamic properties of microplastics have been poorly investigated to date. Moreover, the properties also have impacts on the adsorption process; for instance, particles with larger specific surface areas can adsorb more antibiotics (Xuan and Jwa, 2019). In addition, while the degradation of antibiotics is considered in this study, a rigorous way to consider the degradation of microplastics should be sought in future research efforts.

5. Conclusions

In this work, we simulated the transport of PE-antibiotic compound pollutants using a statistical-physical framework including lattice Boltzmann and Markov methods. The simulation results show that the concentration of antibiotics in Laizhou Bay (which does not include the antibiotics adsorbed by microplastics) was reduced by the adsorption between the PE particles and antibiotics. Concurrently, the compound pollutants will migrate over a larger distance with time. Moreover, with further movement of the compound pollutants, the percentage increase in the antibiotic quantity in its vicinity becomes greater, which raises environmental risks in Laizhou Bay. The proposed hybrid model does not only predict the influence of microplastics on the abundance of antibiotics, but also efficiently predicts the spatial distribution of compound pollutants. The proposed statistical-physical framework can be applied to simulating the transport of microplastic-antibiotic compound pollutants in other bay areas. Basically, we need to obtain the data set of the study area including topographic data, boundary conditions, tidal period, etc. Once obtained they should be readily implementable within our more general framework, which will help us better predict movement trends of pollutants and improve assessment of environmental risks in the future.

CRediT authorship contribution statement

Liming Xing: Writing – original draft, Validation, Methodology, Conceptualization. **Haifei Liu:** Writing – review & editing, Supervision, Resources, Methodology, Conceptualization. **Diogo Bolster:** Writing – review & editing, Methodology.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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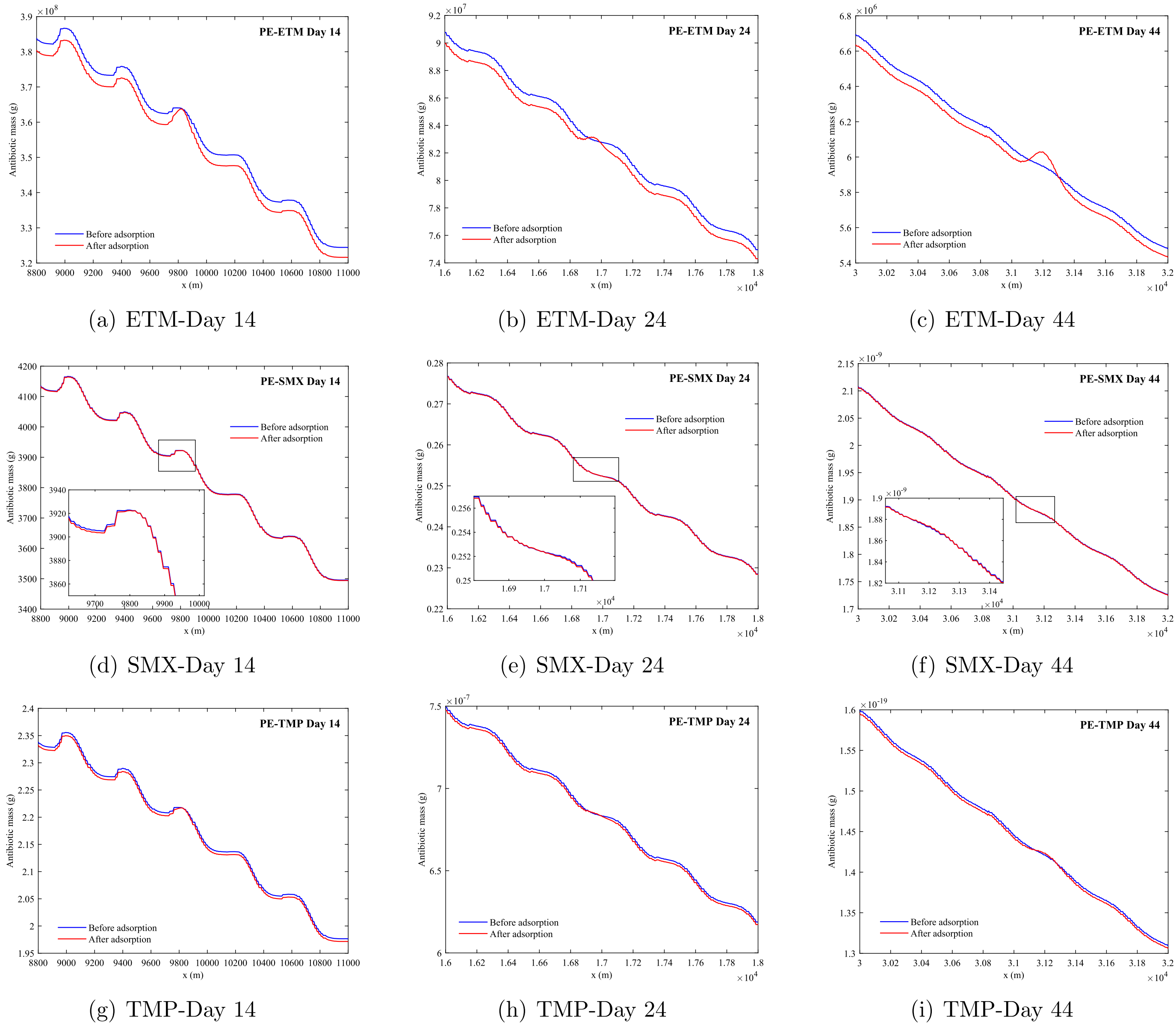

(a) ETM-Day 14 (b) ETM-Day 24 (c) ETM-Day 44

(d) SMX-Day 14 (e) SMX-Day 24 (f) SMX-Day 44

(g) TMP-Day 14 (h) TMP-Day 24 (i) TMP-Day 44

Fig. 5. Long-distance transport of PE-antibiotic pollutants (in the case of SMX, crucial segments are magnified as individual boxes to show detailed information).

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Appendix A. Supplementary material

Supplementary material related to this article can be found online at https://doi.org/10.1016/j.envpol.2024.123339.

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