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学 位 論 文 要 約
Dissertation Abstract

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Development and application of transport models for organic pollutants in the shelf seas

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Organic pollutants in the environment include nonpersistent organic pollutants and persistent organic pollutants (POPs). A mass of organic pollutants enter seawater from rivers and the atmosphere. In addition, the pollutants accumulated in the sediment (in particular, legacy POPs) return to the sea through the resuspension process. Note that the physical and biochemical conditions in shelf seas are generally complex; therefore, the behaviors of organic pollutants cannot be fully understood based on observations alone. The numerical model is a useful tool for conducting studies on the transport and biogeochemical cycles of organic pollutants in the ocean. In this study, we developed a hydrodynamic-ecosystem-suspended particulate matter (SPM)-pollutant coupled model, to study the spatiotemporal patterns of organic pollutants in shelf seas and investigate the reasons for these variations. The model was established by coupling the pollutant, ecosystem, and SPM modules to the hydrodynamic module in sequence.

First, we developed a three-dimensional (3D) hydrodynamic-antibiotic model, to investigate the transport and dilution of sulfamethoxazole (SMX) in the Upper Gulf of Thailand (UGoT). Antibiotics are typical nonpersistent organic pollutants that can rapidly decompose in seawater. As a result, the adsorption and desorption of antibiotics by the particles in seawater are not important, and the ecosystem and SPM modules are not necessary for the study of antibiotics. The simulation produced a spatially averaged annual mean SMX concentration of 0.58 μg m^{-3}, which varied slightly between seasons assuming a temporally constant river SMX loading observed in August. In contrast, the horizontal distribution of SMX concentrations varied strongly with the season, because of the change in the residual currents. In addition, SMX is diluted to the concentrations lower than 10 % of those in river waters within a short distance offshore of estuaries. To better understand such behaviors, we examined the relationship between salinity and SMX concentrations in the UGoT and conclude that a linear relationship can be assumed in the areas mainly affected by a single river. The annual budget demonstrates that 98 % of SMX in the UGoT is removed by

natural decomposition. As the concentrations of fluvial pollutants in the UGoT depend on their river loading and decomposition rates, we derived a function to predict the pollutant concentrations and flushing times, based on the river input flux and half-life.

The POPs are generally hydrophobic and are easily adsorbed by the particles in the ocean. Therefore, biological particles and SPM should be considered when studying the behavior of POPs in seawater. Dynamic marine conditions can affect the spatial distribution of POPs. Interesting phenomena were indicated by the observations of POPs in seas, e.g., high-concentration patches in the offshore area, different horizontal patterns between the surface and bottom seawaters, and different vertical patterns of various POPs, which are generally accompanied by the appearance of a bottom cold water mass (BCWM). Therefore, we studied the relationship between these special phenomena and BCWM, using a hydrodynamic-ecosystem-POP coupled model, for an idealized ocean with BCWM. Generally, dissolved POPs are transferred from the upper layer to the lower layer and accumulated in the BCWM, as they get adsorbed to the upper biological particles, and then, sink during the spring and summer seasons, resulting in differences in their horizontal distributions in the upper and lower layers. Notably, the high concentration patches in winter are generally induced by the release of POPs from the BCWM caused by intense vertical mixing, large air-to-sea flux that results from low water temperature, and the “biological pump” of POPs across the season. Owing to the different ability of sorption to the biological particles, not all types of POPs can accumulate in the BCWM during summer. This leads to different vertical patterns for different types of POPs in the season. Notably, in this study, we developed a prediction function for the accumulation rate of POPs in the BCWM based on their biochemical parameters. The results indicate that the accumulation trend of POPs in the BCWM increases with the Henry’s law constant (H) and bioconcentration factor (BCF).

Besides the research in the idealized sea, we also investigate the effect of BCWM on the transport of POPs in the real sea. A 3D hydrodynamic-ecosystem-POP coupled model was applied to investigate the effects of physical and biological conditions on the transport of POPs in the central Yellow Sea. Two POPs with different physical and biochemical properties were selected as the targets: polychlorinated biphenyl congener 153 (PCB-153) and decabromodiphenyl ether (BDE-209). The concentration of dissolved PCB-153 was high in late spring and low in autumn, whereas that of biological particle-bound PCB-153 was higher in early spring than other seasons. For BDE-209, the dissolved concentration was high in summer and low in winter, and the biological particle-bound concentration was high in early spring. In addition, the dissolved PCB-153 and BDE-209 were accumulated at the sea bottom and sea surface during summer, respectively. The different seasonal and spatial patterns of these two POPs are caused by the larger Henry’s law constant and bioconcentration factor of PCB-153, which leads to larger fugacity from sea to air and stronger absorption by the biological particles of PCB-153 than BDE-209. The balanced mass concentration of POPs in biological particles is the product of the BCF and the concentration of dissolved POPs. The mass concentration of BDE-209 was closer to balance than that of PCB-153, because of its larger depuration and desorption rates of the biological particles. In addition, the combination with biological particles of POPs reduces the dissolved concentration and tends to enhance the diffusive flux from air to sea. This effect is more apparent for PCB-153 than BDE-209, owing to the more balanced air-sea diffusive exchange of PCB-153.

Note that SPM concentration is generally high in coastal seas and can affect the transport and fate of POPs in seawater. Therefore, we added the SPM module to the hydrodynamic-ecosystem-POP coupled model, to investigate the effect of SPM on the behavior of POPs in the ocean. The Bohai Sea was selected as the target area, owing to its high turbidity. The SPM in the Bohai Sea mainly originates from the resuspension of sediment, which is related to bottom stress. The bottom stress is high in the nearshore area, owing to the shallow depth; in winter, it is high because of the strong wave. In this study, we divided SPM into three types, namely, clay, silt, and sand, with clay and silt being the dominant types. We selected BDE-209 as the target pollutant, because its concentrations are high in the atmosphere, rivers, sediments, and the seawater of the Bohai Sea. The dissolved concentration of BDE-209 was higher in summer than winter, affected by the adsorption of dissolved BDE-209 by the SPM. Furthermore, the concentration of particle-bound BDE-209 was generally higher in winter than other seasons, due to both large atmospheric deposition and resuspension from the sediment. However, the seasonal variations in BDE-209 in different areas of the Bohai Sea were different. This may be related to its different sources and processes in these areas. The flux in each biogeochemical process of BDE-209 was calculated using the model results.

In this study, we developed a simulation scheme for the behaviors of organic pollutants in shelf seas, which can be useful for evaluating the increasingly serious organic pollution in the ocean and is important for the health of marine organisms and humans. Note that the coupled model applied in this study is more effective and accurate than those applied in previous studies. A comparison of biogeochemical cycles of different organic pollutants was conducted in this study, along with investigating the reasons and impact factors for the accumulation of POPs in the bottom cold water and analyzing the effect of SPM on the transport of POPs, using the coupled model. In brief, the coupled model developed in this study can be used to quantitatively calculate the transport and biogeochemical processes of marine organic pollutants and determine the control mechanism of their spatiotemporal variation.