Numerical studies on biogeochemical cycle of mercury in the East China Seas

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1 Purposes

Mercury, a persistent heavy metal pollutant, exists in the atmosphere in gaseous form and undergoes long-range transport via atmospheric circulation, making it a global environmental concern (O'Driscoll et al., 2005). The mid-20th century Minamata disease incident in Japan prompted the adoption of the Minamata Convention on Mercury in 2013, an international effort to reduce mercury emissions and usage, safeguarding human health and ecosystems from mercury pollution.

Oceans are critical to the global mercury cycle, acting as both a source and sink (Mason and Sheu, 2002). The ocean is also a key site for mercury transformation, accumulation, and release in biogeochemical processes. In coastal waters, mercury primarily exists as elemental mercury (Hg^0), divalent mercury (Hg^{2+}), methylmercury (MeHg), particulate divalent mercury (Hg^P), and particulate methylmercury ($MeHg^P$), with Hg^{2+} and Hg^P being the dominant forms (Spokes and Liss, 1996; Bowman et al., 2016). MeHg, a potent neurotoxin, bioaccumulates in marine fish, posing significant risks to human health and ecosystems (Clarkson and Magos, 2006).

Coastal waters, key areas for fisheries, are a major source of human mercury exposure through seafood consumption (Watson and Tidd, 2018). Mercury pollution in these regions thus poses dual threats to ecosystems and public health. Located downwind of the East Asian monsoon, China experiences elevated atmospheric mercury levels. These levels significantly increase coastal mercury concentrations through deposition. Rapid industrialization has driven a continuous rise in terrestrial mercury emissions, more than doubling from 1984 to 2013 (Liu et al., 2016), leading to coastal mercury levels surpassing global averages (Chen et al., 2022). In critical zones such as major river estuaries, mercury pollution has reached alarming levels, requiring enhanced monitoring and management to mitigate risks to ecosystems and human health.

2 Methods

The migration and transformation of mercury are driven by biological, chemical, and physical processes. Current research methods include cultivation experiments, field investigations, and numerical models. Numerical models are particularly useful for analyzing multi-factor interactions and predicting future marine mercury risks.

The East China Seas, including the Bohai Sea, the Yellow Sea, and the East China Sea, are one of the world's most productive and human-impacted coastal regions. Rapid industrialization and urbanization in the late 20th century led to a significant increase in mercury loads, from 110 tons in 1984 to 250 tons in 2013 (Liu et al., 2016; Meng et al., 2014). This project aims to develop a 3D coupled hydrodynamic-biogeochemical mercury model for the East China Seas. The model will study mercury migration and transformation, with a focus on MeHg uptake, accumulation, and ecological effects. It will also simulate and predict long-term mercury pollution risks in the region.

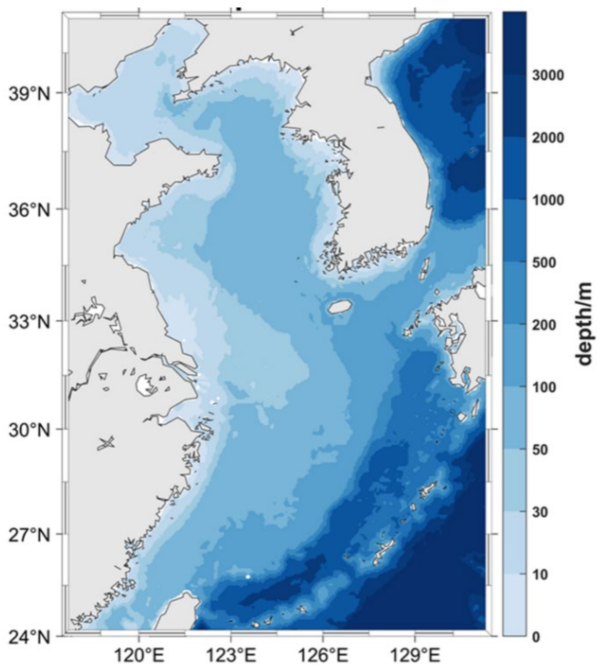

Fig. 1 Bathymetry (shading) of the East China Seas

The proposed 3D coupled hydrodynamic-biogeochemical mercury model integrates three modules: hydrodynamic, ecosystem, and mercury cycling. The hydrodynamic module provides physical parameters (e.g., water temperature, flow velocity), while the ecosystem module supplies biological variables (e.g., plankton biomass, detritus) to study mercury bioaccumulation.

The hydrodynamic module, based on the Princeton Ocean Model (POM), simulates the East China Seas (117.5–131.5°E, 24–41°N), including the Bohai Sea, the Yellow Sea, the East China Sea, and part of the Sea of Japan. It features a horizontal resolution of 1/18° (~6 km) and 21 vertical sigma layers, forced by ERA5 reanalysis data from ECMWF.

The ecosystem module includes 10 variables: dissolved inorganic nitrogen, phosphate, silicate, dissolved organic nitrogen, dissolved organic phosphorus, dinoflagellates, diatoms, zooplankton, nitrogen-phosphorus detritus, and biogenic silica.

The mercury cycling module tracks 5 variables: Hg^0, Hg^{2+}, MeHg, Hg^P, and $MeHg^P$. Key processes include oxidation, reduction, methylation, demethylation, adsorption, desorption, and sediment-water exchange (diffusion, sedimentation, resuspension). MeHg is absorbed and released by phytoplankton and transferred through the food chain.

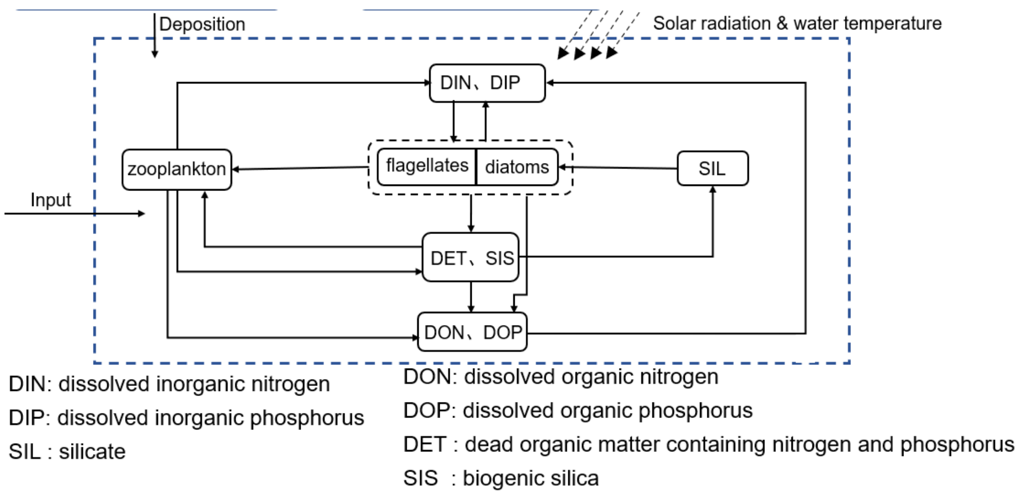

Fig. 2 Ecosystem Module

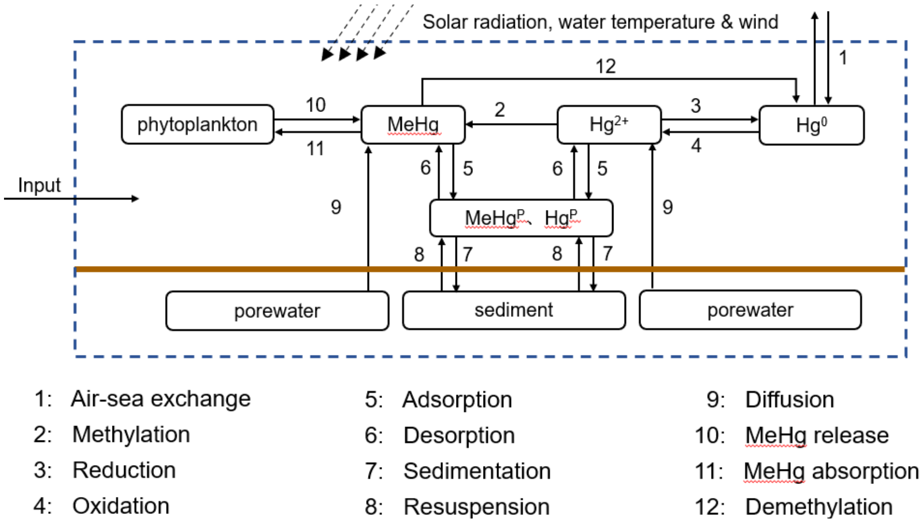

Fig. 3 Mercury Cycling Module

3 Results

1) Mercury Biogeochemical Cycle Box Model

The mercury biogeochemical cycle is simulated using a box model. Fig. 4 shows the seasonal variation of total mercury concentration. The concentration is lower in summer and higher in winter, consistent with previous findings. According to the research of Chen et al. (2022), there are two main possible reasons: In summer, higher temperatures and solar radiation enhance the evasion of Hg^0; In winter, stronger wind-driven water mixing increases the resuspension of particulate mercury (Hg^P and $MeHg^P$) from sediments.

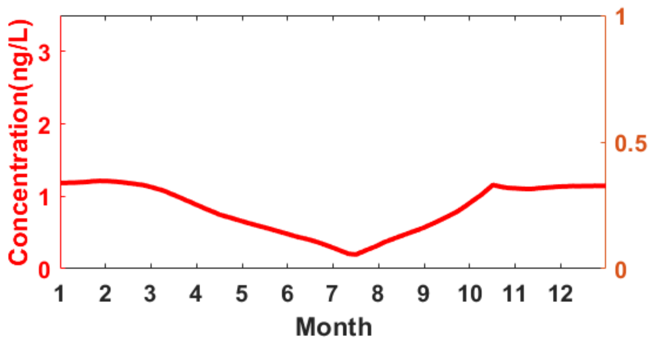

Fig. 4 Seasonal Variations of Total Mercury Concentration

2) 3D Coupled Hydrodynamic-Biogeochemical Mercury Model

Completed work includes the development and validation of the 3D Hydrodynamic-Ecosystem Coupled Model, along with the preliminary integration of the mercury cycling module. The hydrodynamic module (POM), extensively validated and widely used, ensures reliability.

In shallow coastal areas, high suspended particulate matter concentrations often compromise satellite chlorophyll data accuracy. Thus, this study focused on validating surface chlorophyll simulations in East China Seas regions deeper than 40 meters. Fig. 5 compares satellite-observed (MODIS: https://oceancolor.gsfc.nasa.gov/l3/order/) surface chlorophyll concentrations (top four panels) during the 2016 spring bloom (March–June) with model simulations (bottom four panels). The model accurately simulates spring blooms in the Yellow Sea and near the Yangtze River estuary. In the Yellow Sea, blooms begin in March and peak in April, while near the Yangtze River estuary, they persist from early spring to summer, peaking in May. The simulations align well with observations in terms of timing, spatial distribution, and bloom evolution. This demonstrates the model's reliability in simulating ecological processes.

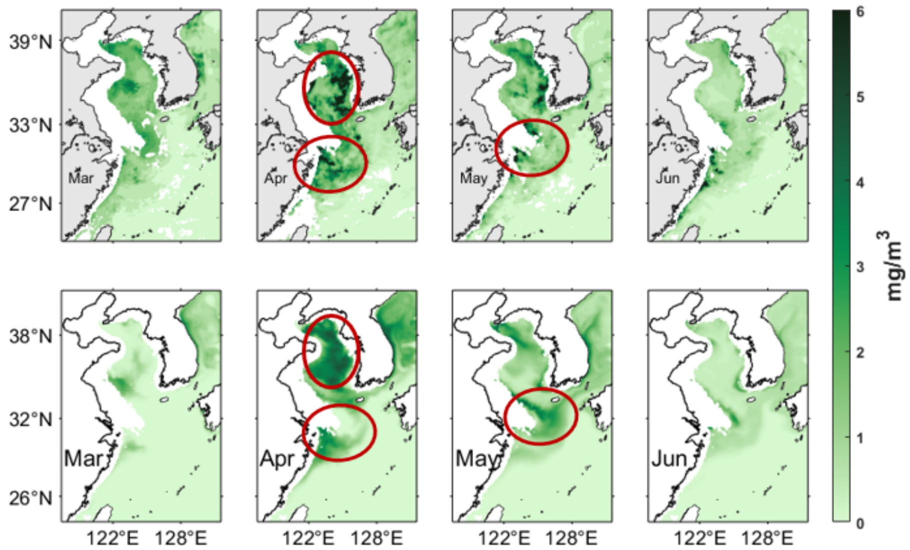

Fig. 5 Validation of Surface Chlorophyll Concentrations During the 2016 Spring Bloom (Top: MODIS; Bottom: Model Results)

4 Future challenges

The 3D hydrodynamic-mercury biogeochemical coupled model has been successfully developed and is operational. However, the mercury cycling module currently relies on random data for riverine inputs and atmospheric deposition, used only for initial framework setup. Future work will involve collecting real observational data for the East China Seas from literature and databases. These data will be used to optimize and refine the module. Model validation against field observations will ensure its accuracy and reliability in simulating mercury cycling processes. This will provide a robust tool for studying mercury pollution and its ecological impacts.

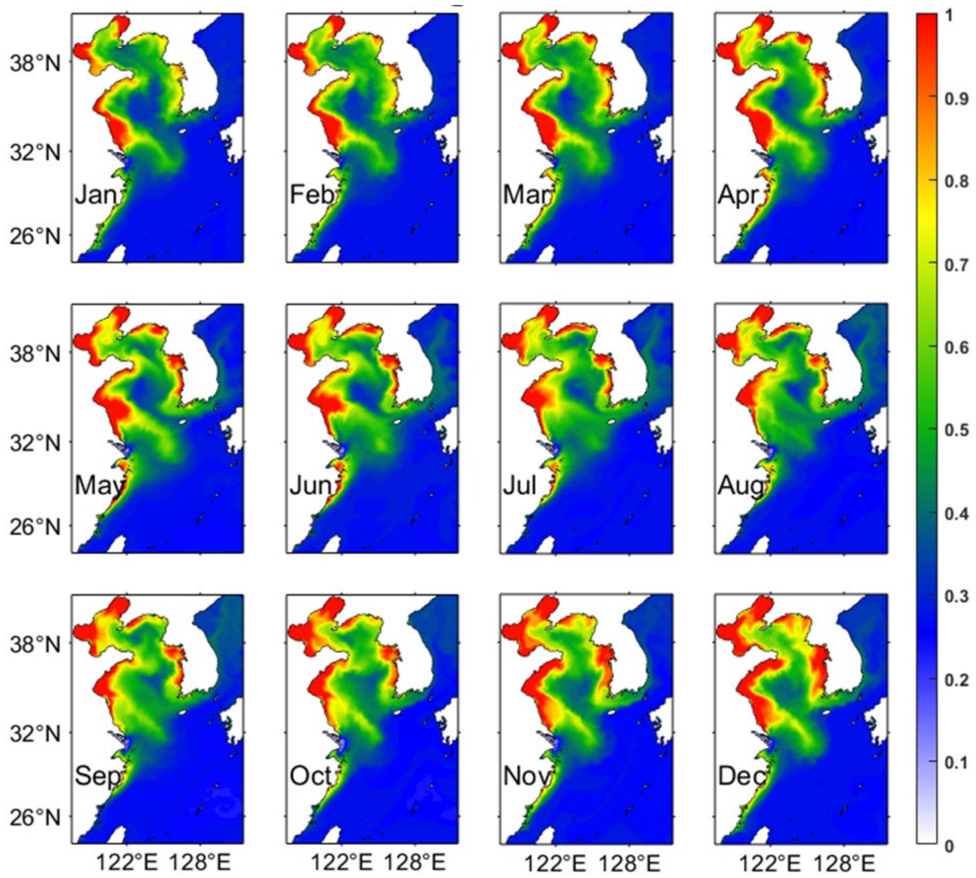

Fig. 6 Seasonal Distribution of Surface Total Mercury in the East China Sea in 2016 (Normalized Results)

Significant progress has been made in understanding marine mercury cycling and ecological risks. However, research on MeHg, a major threat to marine ecosystems, remains limited. The impacts of climate change (e.g., warming and deoxygenation) on mercury transformation, migration, and bioaccumulation in coastal ecosystems are still poorly understood. As global mercury reduction policies are implemented, understanding how climate change may undermine these efforts is a critical scientific challenge.

This model enables long-term simulation and prediction of mercury pollution risks in the East China Seas. It assesses the combined impacts of climate change and emission reduction policies. The study is essential for understanding mercury's behavior in food webs and assessing ecological risks. It also provides scientific support for mercury pollution control and policy development in China's coastal regions.

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