Study on Water Quality Dynamics and Residual Mercury Variation in Minamata Bay

Author(s): 周 長禄

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Graduate School of Engineering
Nagasaki University

Changlu Zhou
Abstract

Minamata Bay, located in the west coast of Kyushu Island, was heavily polluted by mercury-contaminated discharge from a chemical factory. Ever since the Minamata Disease caused by mercury poisoning was publicly recognized, water quality dynamics and residual mercury variation have been the research emphasis in the Minamata Bay and the larger Yatsushiro Sea. The transport and distribution of mercury are related to many factors such as wind, flow, sediment and so on. In addition to periodic in-situ measurements with field survey, numerical simulation models were established to present the dynamics of mercury and other ocean indexes effectively. Coupled with the Princeton Ocean Model (POM), an integrated three-dimensional mercury transport model was presented in this thesis to provide some new insights and further understanding of residual mercury variation in Minamata Bay. This model was integrated with the POM and a new precipitation module for providing hydrodynamic environment, and a cohesive sediment transport module for reproducing the mercury cycling between water column and sediments, incorporating oxidation, methylation and other reaction processes among different mercury species in different forms. Simulation results indicated that the integrated numerical model was generally feasible to reveal the temporal variations and spatial distributions of residual mercury and other factors. The present study achievements are divided into several chapters in this thesis.

Chapter 1 gives the introduction of research background in Minamata Bay firstly, the remediation project and some researches aiming at behavior of mercury and related factors in bay area are presented. After a brief review of mercury researches in global range, the study objectives and thesis outline are given.

Chapter 2 introduces the development of ocean numerical simulation and Princeton Ocean Model. Some common ocean circulation models are presented. After that is a detailed description of the Princeton Ocean Model from basic assumptions and algorithm to numerical scheme and solution procedures.

Chapter 3 shows the simulation results of hydrodynamic model and a new precipitation module. Simulated salinity and temperature on surface by original POM showed deviations with measured data, which was caused by the frequent precipitation during plum rain season and constant setting of surface thermal radiation boundary conditions. A new precipitation module was constructed with new thermal radiation boundary conditions which could change
temporally and spatially and integrated into POM, simulation accuracy showed significant improvement with this module. Cooling impact on surface temperature field of precipitation is analyzed, and the influence of river discharge and flow characteristic in research area are also discussed. This study indicates that the precipitation effect on numerical simulation in rainy season should not be neglected.

Chapter 4 performs a current induced cohesive sediment transport model based on the suspended solid distribution and flow magnitude in Minamata Bay. After the description of basic structures of sediment transport module, sensitivity tests are presented to determine the values of critical erosion and deposition shear stresses, and analyze the influence of wind and river discharge. Simulation results of the sediment transport module showed good agreements with measurements on different layers. The relationship between flow field and sediment concentration in bay area is also discussed.

Chapter 5 presents the integrated three-dimensional mercury transport model coupled with the hydrodynamic module and sediment transport module. Three mercury kinds obtained from in-situ measurements were selected for the comparison of simulation results. Simulated dissolved mercury kinds showed acceptable agreements with measurements and the vertical concentration profiles presented generally consistent distribution trends. Notable deviations occurred in the simulation of bottom particulate mercury, which were probably caused by the existence of abundant coarser particles. While mercury adsorbed by these sediments could barely suspend to upper layers. After the simulation verification part, mercury transport pattern over the large Yatsushiro Yea with mercury source from Minamata Bay is presented and discussed, together with the analysis of flow field.

Chapter 6 is the conclusions and recommendations for further studies. Main research achievements of this thesis are summarized in this chapter, some suggestions for improvement and potential directions of the following researches are also presented.
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1. Introduction

1.1 Study Background

Minamata Bay is located on the west coast of Kyushu Island, Japan, and includes an inner bay called Fukuro Bay in the southern part (Fig. 1.1). Total area of Minamata Bay is about 3.82 km² and average water depth is around 16.7 m. A small island called Kojiji Island creates a narrow channel with mainland in the north part of Minamata Bay, mass exchange of ocean water into and out of bay is able to happen on the north and west bay mouths. Minamata Bay is a part of the greater Yatsushiro Sea, which is also called the Shiranui Sea. The Yatsushiro Sea, surrounded by Kyushu Island and Amakusa Islands, is a semi-enclosed inland sea with an area of 1200 km². It borders the Kumamoto Prefecture and Kagoshima Prefecture, and is connected with the Ariake Sea in the north and the East China Sea in the south with some narrow straits. Water depth is getting deeper from north to south and average water depth is around 23 m. Dominant tidal current in the Yatsushiro Sea is semi-diurnal tide, while diurnal tide dominates around the Minamata Bay (TAI and YANO 2007). Flood tide flows to the northeast direction along with the coastline and ebb tide is in an opposite direction.

Fig. 1.1 Location of Minamata Bay and Yatsushiro Sea, Japan
Minamata Disease, which was publicly acknowledged by the government in 1956, was caused by mercury contaminated discharge from a chemical factory called the Shin Nippon Chisso Fertilizer Company (Akito et al. 2014). A sulfuric acid (H\textsubscript{2}SO\textsubscript{4}) solution containing mercury oxide (HgO) was used as catalyst during the production of acetaldehyde (Balogh et al. 2015). Untreated wastewater containing mercury byproducts was discharged directly into the Minamata Bay and this process lasted for more than 30 years from 1932. In the industrial wastewater, methyl-mercury is the most toxic form which could bio-accumulate in shellfish, fish and other marine organisms. Symptoms of the methyl-mercury poisoning include ataxia, numbness in limbs, general muscle weakness, and damage to hearing and speech. Consumption of methyl-mercury contaminated fish and seafood first led to the death of cats and birds in coastal villages in large numbers, animal effects were especially significant in cats and it was therefore named as “cat dancing disease” by local residents (Nabi 2014). In 1956, two young sisters were hospitalized due to the symptoms of having difficulties in walking and speaking. The hospital reported the discovery of “an epidemic of an unknown disease of the central nervous system”, making the first official recognition of Minamata Disease (Hachiya 2006). More individuals with the same symptoms suffering from the Minamata Disease were reported in succession, and most of them are the residents from fishing villages near the coast shore of Minamata Bay where was confirmed as the tragic source place gathering the mercury contaminants from wastewater discharge (Tokuomi 1961). Subsequent studies aiming at the mercury distribution in Minamata Bay were conducted.

![Fig. 1.2 Map of Minamata Bay dredging operations](image-url)
Investigation of mercury distribution in Minamata Bay suggested that total mercury release was around 70-150 tons, including 0.6-6 tons of methyl-mercury (Balogh et al. 2015). Large quantities of mercury were detected in fish, shellfish and sedimentary sludge in bay area. In order to prevent further diffusion of contaminants and to restore the coastal environment, the Minamata Bay Pollution Prevention Project was carried out as a remediation measure from 1977 to 1990 (Tomiyasu et al. 2014). This project involved the dredging and retreatment of contaminated sludge with mercury concentration greater than 25 ppm (Fig. 1.2). Dredging sludge was cast in filled area and covered with mountain soil to prevent spreading out of contaminated sludge. Before discharged sludge was confined with soil, reclaimed areas were kept in a submerged state to avoid methylation with air (Nakayama et al. 1998). Around 1.51 million m² area in sediment was dredged and filled in a land area of 0.58 million m². Compared to the removal standard of 25 ppm, the maximum mercury concentration was reduced to 8.75 ppm after the remediation project. However, the residual mercury pollution was still in a high level compared with the background level of this area (Tomiyasu et al. 2008). Investigation of mercury contamination in larger Yatsushiro Sea showed higher mercury values near the Minamata Bay, and concentrations decreased with increasing distance from source areas, which indicated that mercury pollutants had transported from Minamata Bay to Yatsushiro Sea (Nakata et al. 2008; Tomiyasu et al. 2000). Since mercury contamination was publicly recognized, residual mercury variation has become the research emphasis in the Minamata Bay and the Yatsushiro Sea. As important factors influencing the mercury transport, the dynamics of flow and sediment transport were studies by many researchers with field investigation or numerical simulation to analyze the relationship with mercury distribution. Murakami et al. (2004) investigated the characteristics of tidal current in Yatsushiro Sea with a three-dimensional model and indicated that the Nagashima channel on the south side played a dominant role in the ocean water exchange of Yatsushiro Sea. Tai et al. (2007) used the particle tracing method to present the tidal current behavior and suggested that the substances initially released into the Minamata Bay could spread over the whole area of the Yatsushiro Sea for one month. Lou Sha et al. (2012a) simulated the impact of river outflow and wind stress on the flow field and also presented the simulation about river discharge effect to the salinity distribution in Yatsushiro Sea (Lou et al. 2012b). Sediment researches were generally included in the study results of mercury because the transport of mercury was inseparable with sediment movement (Kudo et al. 1981; Masuda et al. 2011; Matsuyama et al. 2018; Oki and Tomiyasu 1998).

The speciation and transformation processes of different mercury species and forms in aquatic environment are complicated. In general, chemical speciation of mercury is known as
elemental, organic, and inorganic species, the physical speciation involves dissolved and particulate forms. In these major categories, there are numerous species of mercury compounds. Besides the transport process in water and sediment environments, mercury cycling in aquatic environment also includes the biological process. Mercury could be absorbed by plankton, periphyton and benthic invertebrates (Žagar et al. 2007; Lessard 2012). These marine organisms may remobilize the mercury pollutants with flow or cause mercury to enter the food chain through consumption from fish or other ocean creatures. The change of ocean factors like temperature may affect the activity and consumption of marine creatures, which could cause the response from ocean factors to the mercury transport. However, due to the limitation of data collection, the biological process of mercury transport was not considered in this study. Meantime the numerous mercury compounds are difficult to present with numerical simulation specifically, consequently, the numerous species of mercury were classified as the three major categories with two physical forms for modeling purposes. Dissolved mercury could move with the diffusion of ocean water and spread with flow, part of particulate mercury attached on suspended sediment also propagates in this way and mercury dynamics in different water depth display difference due to the changing currents and tides. Meantime, large amounts of the particulate mercury form deposited to the ocean bed as a main transmission source, and the pore water existed in bottom ocean sediment also has mercury exchange with water column. Complex and diverse forms of existence and transportation lead to various difficulties during the study of mercury and other related factors not only in Minamata Bay but also in a global range.

1.2 Researches of Mercury

Intense production activities of humanity without considering environmental burden contributed to various pollution issues on a global scale. As a global, persistent and bio-accumulative contaminant, mercury is present in environment as element mercury, inorganic mercury, and methyl-mercury. Mercury cycling processes are affected by many factors, such as wind and rain in atmosphere, hydrodynamics and sediment transport in aquatic systems and so on. Element mercury is a volatile mercury species that could transport within the atmosphere after mineral exploitation, fuel combustion or water evasion (Zhu et al. 2018). Compared with other mercury species, proportion of elemental mercury is small and supersaturation is common in surface water (Lessard 2012). Inorganic mercury in aquatic environment has two kinds of valences, monovalent and divalent, while divalent mercury is the
most abundant form of inorganic mercury in water systems. Therefore most mercury studies treated divalent mercury as substitute of inorganic mercury (Wang et al. 2004). When element mercury is oxidized to divalent form in atmosphere, it could be deposited to water, similarly, these transformation processes between two mercury species also happen in aquatic environment. With methylation processes, inorganic mercury could be transformed into methyl-mercury, which is the most toxic mercury compound leading to many kinds of poisoning incidents. Bio-accumulation and bio-magnification of methyl-mercury make it easily absorbed and accumulated in biological tissues and organs and lead to irreversible damage to the nervous system.

Compared to natural mercury emissions, human activities have aggravated the occurrence of mercury pollution. A few years after the Minamata Disease was officially acknowledged, a second outbreak of Minamata Disease occurred in Niigata, which was also caused by mercuric catalyst for acetaldehyde synthesis (Eto et al. 2010). In 1969, a river system in Canada was found to be contaminated by mercury emitted by a caustic soda factory, large amount of untreated mercury-contaminated wastewater was discharged from upstream (Harada et al. 2011). Mercury pollutants were discharged into local ecosystems due to the gold mining in Brazilian Amazon, evidence has shown that most of pollutants were exported to the Amazon River, in another effluent of Amazon called Tapajos River, high hair mercury levels were detected in local fishermen and their families (Harada et al. 2001; Nriagu et al. 1992). Industrialization processes all over the world, such as coal combustion, waste incineration, metal mining and chemical production (Li et al. 2009), have accelerated mercury emission to atmosphere and exacerbated pollutants cycle in global range. Due to the unique chemical characteristics of mercury, large amounts of mercury will deposit through a series of reaction processes during the global atmospheric transport, these deposition processes in marine environment is more significant and most of deposited mercury will incorporate into deep sediment or bio-accumulate in marine organisms, which become new sources for mercury transportation (Hylander and Goodsite 2006).

Researches of mercury are inseparable from field observation, continuous observations are essential for some seriously polluted areas. Through collecting polluted samples, transport and distribution patterns of mercury pollutants could be summarized after analyzing pollutant content in laboratory. While field observation is time-consuming and costly, it is difficult to obtain a long time distribution of pollutants. Compared with in-situ observation methods, numerical simulation is a more effective approach for understanding long-term distribution of different kinds of mercury species. Through establishing numerical models and combining with
observed data, some complex processes of mercury transportation could be reproduced. Simulation models of mercury could be divided into atmosphere and water categories due to the different existing forms. Mercury could also transport between these two systems by emission or deposition. Atmosphere plays an important role during the global dispersion of mercury. Mercury emissions to atmosphere are mainly element form, which is also the dominant form of mercury in atmosphere, and atmospheric observations indicated that the residence time of element mercury had reached to the order of one year (Poissant and Casimir 1998). Oxidation of mercury also happens in atmosphere, while oxidized mercury is highly water soluble and easily deposits into water systems through wet or dry deposition by rain or snow (Poissant and Pilote 1998).

Different numerical models have been developed and applied for mercury propagation in the atmosphere and between atmosphere and water systems. A modified model based on the Regional Lagrangian Model of Air Pollution (RELMAP) was developed to simulate different forms of atmospheric mercury and a wide variety of emission speciation, combinations of chemical and physical forms are evaluated (Bullock Jr et al. 1998). A non-steady-state compartmental box model for mercury cycling was described and used to simulate mercury fluxes on a global scale between atmosphere and ocean (Lamborg et al. 2002). An inter-comparison study was conducted to present atmospheric mercury transport and deposition in the North America region based on three regional-scale atmospheric mercury cycling models, the Community Multi-scale Air Quality model (CMAQ), the Regional Modeling System for Aerosols and Deposition (REMSAD) and the Trace Element Analysis Model (TEAM) (Bullock et al. 2008). Based on a three-dimensional ocean-atmosphere model of mercury called GEOS-Chem, a land-ocean-atmosphere model is developed to present mercury cycles between preindustrial and present biogeochemical conditions (Selin et al. 2008). In Mediterranean area, a modified mercury model of Weather Research and Forecasting model coupled with Chemistry (WRF/Chem) was used to simulate the atmospheric mercury emissions, concentrations and deposition with high spatial resolution (Gencarelli et al. 2014).

Mercury transport in aquatic environment involves multiple processes among hydraulics dynamic, sediment transport and atmosphere exchange, also the various reaction processes among different mercury species with different forms. Accordingly, it is difficult to simulate the integrated mercury transport processes in aquatic systems. Many transformation and reaction coefficients are unavailable due to the complicated processes, and approximations and empirical coefficients based on the limited collecting and experimental data are inevitable for the numerical simulation of mercury transport. Numerical models for mercury cycling in
aquatic systems could be separated into different categories:

(1) Lake system. A mass balance model modified from the Onondaga Lake Mercury Model was developed to simulate exchange into and out of plankton and fish in Onondaga Lake, New York, which was polluted by historical mercury discharges (Henry et al. 1995). Also in the Onondaga Lake area, by modifying the Water Quality Analysis Simulation Program, a model approach for simulating the fate of mercury was presented and model predictions for water column generally agreed with measured data (Kim et al. 2004). The Quantitative Water Air Sediment Interaction model was used to estimate mercury fluxes and concentrations of Big Dam West which was a remote lake, this model was able to provide reasonable mercury estimates with limited data collection (Ethier et al. 2008). A mass balance steady-state model called Hg Environmental Ratios Multimedia Ecosystem Sources was applied to predict mercury concentrations and fluxes in Lake Ontario, which was also applicable to a broad range of lake sizes (Ethier et al. 2012).

(2) River system. An integrated model combined three models: MERC4, WASP5 and RIVMOD was presented to simulate mercury transport in Carson River (Carroll et al. 2000), and this model also had a successful application during the modeling of mercury transport and transformation processes in the Idrijca and Soca river systems (Zagar et al. 2006). A regional mass balance model was developed for the St. Lawrence River near Cornwall, which described concentrations and transport of three mercury forms in a different environment (Lessard 2012). A seasonally-responsive dynamic systems model is used to analyze the sources of natural and anthropogenic mercury contributed to the Oregon’s Willamette River Basin, and to assess the impact of mercury contents on surface water, sediment and fish tissue (Hope 2006).

(3) Marine system. Compared with the river and lake systems, simulation of mercury cycling in coastal or marine areas is more complicated. With larger dimensions in horizontal and vertical directions, the dispersion processes are important and three-dimensional model is necessary to present a complete process of mercury cycling in marine system. A two-dimensional advection-dispersion mercury transport and fate model termed STATRIM was developed to simulate mercury transformation processes in the Gulf of Trieste (Širca et al. 1999), this model was upgraded to a three-dimensional model called PCFLOW3D with a series of improvements. PCFLOW3D was a non-steady state mercury transport model incorporated with hydrodynamic module, sediment transport, biogeochemical modules, and various mercury transportation processes, which had been widely applied in coastal and marine systems like the Gulf of Trieste (Rajar et al. 2000) and the Mediterranean Sea (Žagar et al. 2007). The PCFLOW3D model was also adopted to simulate the mercury mass balance in
Minamata Bay (Rajar et al. 2004a; Rajar et al. 2004b), the sources and sinks of total mercury and methyl-mercury in the bay area were evaluated and concluded, nevertheless, these simulations didn’t elaborate the details of distribution and transport process of different kinds of mercury species in various forms. To further understand the mercury transport pattern in Minamata Bay and Yatsushiro Sea, an integrated three dimensional numerical model for mercury dynamic simulation was developed and presented in this dissertation.

1.3 Study Objectives

During the several decades since the Minamata Disease was publicly acknowledged, relevant studies about water quality dynamics and mercury variation have been carried out continuously. However, these researches are mainly focused on regularity summarization based on field observation and experimental results. To advance the understanding of hydrodynamic environment and patterns of residual mercury distribution in Minamata Bay, an integrated three dimensional numerical model for mercury dynamic simulation was developed, coupling with an upgraded hydrodynamic model and a cohesive sediment transport module. This study aims to estimate impacts on water quality dynamics under different external forcing conditions, and to reproduce residual mercury variation and cycling in a more specific and precise approach by numerical simulation. The primary objectives of this thesis are presented in these following points:

(1) To improve the hydrodynamic model based on the Princeton Ocean Model to provide a more accurate simulation environment for the simulation of mercury transport;

(2) To construct a cohesive sediment transport module to present the transformation processes of mercury in different forms between solid sediments and water columns;

(3) To develop an integrated mercury transport model based on the hydrodynamic model and sediment module and validate simulation results through comparing the field observation data;

(4) To reproduce the mercury transport pattern by using the newly established mercury transport model and analyze the residual mercury dynamics.

1.4 Thesis Outline

Coupled with the Princeton Ocean Model, an integrated three-dimensional mercury transport model is presented in this thesis to provide some new insights and further
understanding of residual mercury variation in Minamata Bay. According to the construction orders of different modules, structures of this thesis are shown as follows:

Chapter 1 is the introduction of research background in Minamata Bay, researches about mercury in a global range, research objectives and outline of this thesis.

Chapter 2 introduces the development of ocean numerical simulation and some common ocean circulation models. The basic structure and algorithm of Princeton Ocean Model are described.

Chapter 3 shows the comparison of simulated and measured data with original Princeton Ocean Model, this hydrodynamic model was improved with a new precipitation module to adjust the rainfall impact on plum rain season, also with a new thermal radiation boundary condition which could change temporally and spatially.

Chapter 4 presents a cohesive sediment transport model based on the suspended solid distribution in Minamata Bay. As an important factor affects the sediment transport, bottom shear stresses were analyzed under different external conditions like wind and river discharge. The sediment transport model was validated by field observation data.

Chapter 5 describes the mercury transport model, integrating the hydrodynamic model and cohesive sediment transport module. Different kinds of mercury species in various forms were simulated and compared with measured data. Transport and distribution patterns of mercury in large Yatsushiro Sea were presented with mercury source from Minamata Bay.

Chapter 6 is the conclusions and recommendations for further studies. Research achievements of this thesis are summarized in this chapter, some suggestions for improvement and potential directions of the following researches are also presented.
2. Review of Hydrodynamic Models and Description of Princeton Ocean Model

2.1 Development of Ocean Circulation Model

With the development of computer technology and fluid mechanics theory in the 60s and 70s of last century, Ocean numerical models have been widely developed and applied. Various numerical prediction models for waves, tides and other marine indexes have been established successively. The development of ocean simulation has experienced changes from horizontal or vertical two dimensional to three dimensional, the technology of coordinate system in vertical direction and grid application in horizontal direction has also been improved. With the continuous improvement of researchers, numerical models for ocean circulation have been constantly optimized with more abundant simulation modules for different research applications, more refined mesh arrangement, more streamlined code structures, and more accurate simulation results.

2.1.1 Introduction of Coordinate Systems and Grid Types

In vertical direction, z-coordinate was widely applied in early ocean models, while it is generally difficult to get accurate simulation results with large topographic variations, the step-like representation of topography will make flow field on bottom inconsistent with reality (Ezer and Mellor 2004). In order to solve these defects, terrain-following coordinate was developed and utilized in many following ocean models. \( \sigma \)-coordinate used in ocean models has improved the simulation capability in complex terrain environment. In a \( \sigma \)-coordinate system transformed from the z-coordinate, vertical layers keep same in all simulation domain irrespective of the depth of water column. It enables higher resolution both in deep and shallow water, and allows continuous fields to be represented smoothly at bottom layers. However, the use of \( \sigma \)-coordinate will cause numerical baroclinic pressure gradient errors, the horizontal pressure gradient numerical scheme leads to the increased diapycnal diffusion or truncation errors (Auclair et al. 2000; Mellor et al. 1998). The isopycnic coordinate uses potential density referenced to a given pressure as the vertical coordinate, which has better behavior for simulating tracer transport. Nevertheless, this coordinate is not suitable for the surface mixed layer regions where density could be changed by thermodynamic processes. To counter
different disadvantages, a hybrid coordinate system combined with the z-coordinate and σ-coordinate was developed, which was called s-coordinate. S-coordinate permits uniformly high resolution near surface of z-coordinate and preserves the terrain following characteristic of σ-coordinate (Song and Haidvogel 1994). A more complex hybrid coordinate system was designed through applying different coordinate systems in different terrain areas. This coordinate system was used in the Hybrid-Coordinate Ocean Model, which adopted isopycnic coordinate in deep stratified ocean, z-coordinate near surface within the mixed layers, and σ-coordinate in shallow coastal regions with higher resolution (Chassignet et al. 2007).

In horizontal direction, grid types could be divided into different categories depending on different methods or simulation areas. Many ocean circulation models use finite-difference method to solve the basic governing equations of fluid dynamics. Correspondingly, rectangular grids with transformed or stretched coordinates become the most common horizontal grid system. Many different schemes have been developed based on rectangular grids which have the advantage of simple structure, convenient calculation. Non-uniform rectangular grids or nested grids allow for higher resolution focus on a small research area in large ocean domain without expending computer resources. However, like the z-coordinate in vertical direction, the rectangular grids are difficult to reproduce accurate terrain in boundary areas with complex and varied coastlines, and the structured grids will be jagged in the curve coastline regions which can lead to unrealistic flow disturbance in these areas. Therefore irregular grids have been presented to resolve complex coastlines features. The orthogonal curvilinear coordinate system was developed for modeling coastal areas through coordinate transformation technology, which provided flexibility to refine grid in regions of large gradients (Blumberg and Herring 1987). Development of finite volume method and finite element method has motivated the application of unstructured triangular grids. This grid system allows the continuous change of triangle size throughout whole simulation domain, accordingly, higher resolution could be applied in specified zone or complex coastlines with better accuracy.

2.1.2 Common Ocean Circulation Models

After decades of improvement and development, numerous open source or commercial numerical models have been successfully applied for the simulation of ocean circulation. Ocean circulation models are usually based on Boussinesq approximation, hydrostatic momentum, mass balances, ocean water equation, tracer conservation and so on. Open-access source programs are easier to obtain via internet freely compared to the high price cost of commercial models, and users could modify the source code or build new modules depending
on different research applications. Some open source models have been widely used for scientific researches and these models have been continuously improved with the addition of more researchers.

(1) The Princeton Ocean Model (POM), which is also the basic hydrodynamic model in this thesis, has been a pioneering model in ocean research since the early 1980s. POM is capable of dealing with wide range of simulation from small-scale estuaries and coastal areas to global ocean climate change (Bao et al. 2000; Brenner et al. 2007; Chau and Jiang 2002). A detailed description of POM will be introduced in next section.

(2) Finite-Volume Community Ocean Model (FVCOM) is a prognostic coastal ocean circulation model with the unstructured-grid. FVCOM was originally developed for estuaries and coastal region with complex irregular geometry and steep topography (Chen et al. 2003), with continuous upgrades, FVCOM has been used for global, regional and coastal ocean as well as estuarine applications (Bai et al. 2013; Chen et al. 2011). It also combines the advantages of finite element method through using flexible grids and finite-difference method with numerical efficiency, the use of triangular grid enables FVCOM to resolve complex geometry and bathymetry (Chen et al. 2008). Unstructured meshes and simplicity of coding structure improve the simulation accuracy and applicability of FVCOM (Chen et al. 2009).

(3) Regional Ocean Modeling System (ROMS) is a three-dimensional coastal ocean circulation model that includes several sub-models for simulating different ocean factor dynamics (Warner et al. 2008). ROMS is versatile in diverse applications in different systems and regions, especially for mesoscale systems (Haidvogel et al. 2008). Different model components could be included or excluded via C-preprocessor in a different simulation environment. The 4-dimensional variational data assimilation capability integrated into the model structure has been widely applied (Moore et al. 2011; Powell et al. 2008).

Open source ocean models are publicly available for non-commercial academic researches and education, however, operation of the numerical model codes is complicated which requires deep learning and understanding of code structures, operating system, and programming software without a friendly operation interface. Pre-processing needs multiple files to specify topography, computational grids, external forcing, initial conditions, and other information. Many variables or parameters require reset according to different research objectives. Post-processing also needs targeted output for different research contents and understanding of various kinds of analysis and drawing software. Appearance of commercial software provides other choices for ocean circulation simulation with convenient visualization and easy operation features.
(1) Estuarine and Coastal Ocean Model with Sediments (ECOMSED) is enhanced from the commercial version of POM which is called Estuarine and Coastal Ocean Model (ECOM) for the simulation of shallow water environments (Blumberg 2002). Combing with the hydrodynamic module, sediment transport module, and wind-induced wave module, ECOMSED is capable of simulating currents, different kinds of suspended sediments transport, dissolved tracers and neutrally-buoyant particles in estuarine and coastal ocean systems. Among these functions, the simulation for sediment transport and concentration is especially powerful (Jian-wei 2008; Qun 2007).

(2) DELFT- 3D is an integrated numerical simulation software suite which has several modules for simulating flows, sediment transport, waves and water quality (Lesser et al. 2000). DELFT- 3D has a serious computer software suite for three-dimensional simulation of river, coastal and estuarine areas (Qinghua 2006). Coordinate systems are abundant for different applications and functions of different modules are coupled in a good way which allows users to operate effectively.

(3) MIKE is a range of software products for simulating different type of researches in water environments (Warren and Bach 1992). MIKE is able to deal with simulations from one dimension to three dimensions, hydrodynamic to water environments and ecosystem, and the simulation range covers areas from small scale like river, lake and estuarine to offshore and deep ocean. The hydrodynamic module provides the basis for computations of processes performed in many other modules (Ting 2010; Xu et al. 2012).

2.2 Introduction of Princeton Ocean Model

The Princeton Ocean Model (POM) is a powerful numerical ocean circulation model created by Alan Blumberg and George Mellor around 1977 at Princeton University (Blumberg and Mellor 1987). With continuous and innovative developments of many researchers, POM has been developed to deal with a wide range of simulation from small-scale coastal processes to global ocean climate change and has been successfully applied in numerous oceanographic problems. POM has attributes such as: mode splitting combined with two and three-dimensional modes, a σ-coordinate in vertical direction and curvilinear orthogonal coordinates in horizontal direction, free surface, complete thermodynamics. The embedded level 2.5 Mellor and Yamada turbulence closure model for vertical mixing is widely used in various ocean numerical models. POM has provided the basic theory and simulation framework for many following numerical models, some modules and features can all be
traced back to the initial POM.

2.2.1 Basic Assumptions of POM

The direct analytic solution of the great Navier-Stokes equations is still unattainable in fluid mechanics, while the numerical solutions are available based on several assumptions or approximations. To solve the primitive equations in POM, some simplifying approximations and hypothesis are used:

(1) Hydrostatic approximation. Compared with the horizontal scale, some factors such as water particle acceleration and turbulent friction term in vertical scale are small. Correspondingly, the magnitude of gravitational acceleration is large and the vertical pressure gradient is assumed as the product of density times the gravitational acceleration. The calculation of vertical momentum equation could be simplified through this approximation.

(2) Boussinesq approximation. Although fluid dynamic of ocean is assumed to be incompressible, small perturbations of water density still exist. The approximation ignores density different unless the differences appear in terms multiplied by gravitational acceleration and replaces the density with reference density.

(3) Rotta and Kolmogorov hypothesis. The turbulence closure model used in POM is based on the energy redistribution hypothesis of Rotta and the Kolmogorov hypothesis of local, small-scale isotropy (Mellor and Yamada 1982).

2.2.2 Basic Equations of POM

Basic equations in the circulation model include continuity equation, ocean momentum equations, temperature and salinity equations, and the turbulence kinetic energy and mixing length equations. All the basic equations have been transformed from conventional Cartesian coordinate to the bottom following sigma coordinate as shown in Fig. 2.1. Transformation equations are given as:

\[ x^* = x, \quad y^* = y, \quad t^* = t, \quad \sigma = \frac{z - \eta}{D}, \quad D = H + \eta \]  \hspace{1cm} (2.1)

where \( x, y, z \) are space variables of the conventional Cartesian coordinate and \( t \) is the time variable; \( x^*, y^*, \sigma \) are space variables of the sigma coordinate and \( t^* \) is time variable; \( D \) is total water depth, \( H \) is bottom topography and \( \eta \) is free surface elevation. From water surface \( z = \eta \) to bottom \( z = -H \), \( \sigma \) ranges from \( \sigma = 0 \) to \( \sigma = -1 \). On the basis of approximations and hypothesis, the basic equations of POM in sigma coordinate could be presented as:
\[
\frac{\partial UD}{\partial x} + \frac{\partial VD}{\partial y} + \frac{\partial \omega}{\partial \sigma} + \frac{\partial \eta}{\partial t} = 0
\]  

(2.2)

\[
\frac{\partial UD}{\partial t} + \frac{\partial U^2D}{\partial x} + \frac{\partial UD}{\partial y} + \frac{\partial UD}{\partial \sigma} - fVD + gD\frac{\partial \eta}{\partial x} + \frac{gD^2}{\rho_0} \int_0^\rho \left[ \frac{\partial p}{\partial x} - \frac{\sigma}{D} \frac{\partial D}{\partial x} \frac{\partial \rho}{\partial \sigma} \right] d\sigma' = \frac{\partial}{\partial \sigma} \left[ K_M \frac{\partial U}{\partial \sigma} \right] + F_x,
\]  

(2.3)

\[
\frac{\partial VD}{\partial t} + \frac{\partial UD}{\partial x} + \frac{\partial V^2D}{\partial y} + \frac{\partial VD}{\partial \sigma} + fUD + gD\frac{\partial \eta}{\partial y} + \frac{gD^2}{\rho_0} \int_0^\rho \left[ \frac{\partial p}{\partial y} - \frac{\sigma}{D} \frac{\partial D}{\partial y} \frac{\partial \rho}{\partial \sigma} \right] d\sigma' = \frac{\partial}{\partial \sigma} \left[ K_M \frac{\partial V}{\partial \sigma} \right] + F_y,
\]  

(2.4)

\[
\frac{\partial TD}{\partial t} + \frac{\partial TUD}{\partial x} + \frac{\partial TVD}{\partial y} + \frac{\partial T\omega}{\partial \sigma} = \frac{\partial}{\partial \sigma} \left[ K_H \frac{\partial T}{\partial \sigma} \right] + F_T - \frac{\partial R}{\partial \sigma},
\]  

(2.5)

\[
\frac{\partial SD}{\partial t} + \frac{\partial SUD}{\partial x} + \frac{\partial SVD}{\partial y} + \frac{\partial S\omega}{\partial \sigma} = \frac{\partial}{\partial \sigma} \left[ K_H \frac{\partial S}{\partial \sigma} \right] + F_S,
\]  

(2.6)

\[
\frac{\partial q^2}{\partial t} + \frac{\partial Uq^2}{\partial x} + \frac{\partial Vq^2}{\partial y} + \frac{\partial \omega q^2}{\partial \sigma} = \frac{\partial}{\partial \sigma} \left[ K_q \frac{\partial q^2}{\partial \sigma} \right] + \frac{2K_M}{D} \left[ \left( \frac{\partial U}{\partial \sigma} \right)^2 + \left( \frac{\partial V}{\partial \sigma} \right)^2 \right] + \frac{2g}{\rho_0} K_H \frac{\partial \rho}{\partial \sigma} - \frac{2Dq^2}{B_I} + F_q,
\]  

(2.7)

\[
\frac{\partial q^2tD}{\partial t} + \frac{\partial Uq^2tD}{\partial x} + \frac{\partial Vq^2tD}{\partial y} + \frac{\partial \omega q^2tD}{\partial \sigma} = \frac{\partial}{\partial \sigma} \left[ K_q \frac{\partial q^2tD}{\partial \sigma} \right] + E_l \left( K_M \left[ \left( \frac{\partial U}{\partial \sigma} \right)^2 + \left( \frac{\partial V}{\partial \sigma} \right)^2 \right] + E_j \frac{g}{\rho_0} K_H \frac{\partial \rho}{\partial \sigma} \right) - \frac{Dq^2t}{B_I} W + F_i,
\]  

(2.8)

Fig. 2.1 The sigma coordinate system

where \( U, V, \omega \) represent the current velocity components along horizontal and vertical directions, respectively, the transformation of vertical velocity between Cartesian and sigma
coordinate is shown in Eq. 2.9 and Eq. 2.10; \( D = H + \eta \), sum of bottom topography depth and free surface elevation; \( f \) is the Coriolis parameter. \( \rho' = \tilde{\rho} - \rho_0 \), represents the density disturbance, and the subtraction of initial density field \( \rho_0 \) from density \( \tilde{\rho} \) could reduce the truncation errors associated with the calculation of the pressure gradient term in sigma coordinate in steep topography; \( T \) is water temperature and \( S \) is salinity; \( K_M \) is vertical kinematic viscosity and \( K_H \) is the vertical eddy diffusivity coefficient of temperature and salinity; \( \partial R / \partial \sigma \) is the heat radiation forcing term; \( q^2 \) is twice the turbulence kinetic energy and \( l \) is the turbulence length scale; \( K_q \) is the vertical diffusivity coefficient of turbulence kinetic energy; \( \phi \) represents \( T, S, q^2 \) or \( q^2 l \); \( A_M \) is horizontal kinematic viscosity and \( A_H \) is horizontal heat diffusivity, in order to maintain a valid bottom boundary simulation with large horizontal diffusion, the Smagorinsky diffusivity formula is used.

\[
W = \frac{dz}{dt} = \frac{\partial z}{\partial x} + U \frac{\partial z}{\partial x} + V \frac{\partial z}{\partial y} = D \frac{\partial \sigma}{\partial t} + DU \frac{\partial \sigma}{\partial x} + DV \frac{\partial \sigma}{\partial y} + \frac{\partial \bar{\rho}}{\partial \sigma} - c_s^2 \frac{\partial^2 \bar{\rho}}{\partial \sigma^2} + \frac{\partial \rho'}{\partial \sigma} \tag{2.9}
\]

\[
DW = \frac{\partial \sigma}{\partial z} + \frac{\partial \eta}{\partial \sigma} + \frac{\partial D}{\partial \sigma} + U \left( \frac{\partial \sigma}{\partial x} + \frac{\partial \eta}{\partial x} \right) + V \left( \frac{\partial \sigma}{\partial y} + \frac{\partial \eta}{\partial y} \right) + \frac{\partial \rho'}{\partial \sigma} \tag{2.10}
\]

\[
W = D \frac{d \sigma}{dt} + \frac{\partial \eta}{\partial t} + \frac{\partial D}{\partial t} + U \left( \frac{\partial \sigma}{\partial x} + \frac{\partial \eta}{\partial x} \right) + V \left( \frac{\partial \sigma}{\partial y} + \frac{\partial \eta}{\partial y} \right) \tag{2.11}
\]

\[
F_x = \frac{\partial}{\partial x} \left[ A_M H \left( \frac{\partial U}{\partial \sigma} + \frac{\partial V}{\partial \sigma} \right) \right] + A_M H \left( \frac{\partial U}{\partial \sigma} + \frac{\partial V}{\partial \sigma} \right) \tag{2.12}
\]

\[
F_y = \frac{\partial}{\partial y} \left[ A_M H \left( \frac{\partial U}{\partial \sigma} + \frac{\partial V}{\partial \sigma} \right) \right] + A_M H \left( \frac{\partial U}{\partial \sigma} + \frac{\partial V}{\partial \sigma} \right) \tag{2.13}
\]

where \( \phi \) represents \( T, S, q^2 \) or \( q^2 l \); \( A_M \) is horizontal kinematic viscosity and \( A_H \) is horizontal heat diffusivity, in order to maintain a valid bottom boundary simulation with large horizontal diffusion, the Smagorinsky diffusivity formula is used.

\[
A_M = C \Delta x \Delta y \left[ \frac{1}{2} \left( \frac{\partial U}{\partial x} \right)^2 + \frac{1}{2} \left( \frac{\partial V}{\partial x} + \frac{\partial U}{\partial y} \right)^2 + \left( \frac{\partial V}{\partial y} \right)^2 \right]^{\frac{1}{2}} \tag{2.14}
\]
where $C$ is Smagorinsky diffusivity coefficient, normally in the range from 0.1 to 0.2, while under the circumstance that the grid spacing is small enough, $C$ could be 0; $TPRNI$ is the inverse, horizontal, turbulence Prandtl number which takes the value of 0.2 or less.

Vertical boundary conditions for Eq. 2.2 without freshwater inflow are

$$\omega(0) = \omega(-1) = 0 \quad (2.16)$$

and surface boundary conditions for Eq. 2.3 and Eq. 2.4 are

$$\frac{K_m}{D} \left( \frac{\partial U}{\partial \sigma} \frac{\partial V}{\partial \sigma} \right) = - \left( \langle w(0) \rangle \langle w(0) \rangle \right), \sigma \to 0 \quad (2.17)$$

where right terms are the input of wind stress components. The bottom boundary conditions are

$$\frac{K_m}{D} \left( \frac{\partial U}{\partial \sigma} \frac{\partial V}{\partial \sigma} \right) = C \left[ U^2 + V^2 \right]^{\frac{1}{2}}, \sigma \to -1 \quad (2.18)$$

these boundary conditions could be derived by matching the numerical solution to the law of the wall. The boundary conditions for Eq. 2.5 and Eq. 2.6 are

$$\frac{K_m}{D} \left( \frac{\partial T}{\partial \sigma} \frac{\partial S}{\partial \sigma} \right) = - \left( \langle w(0) \rangle \right), \sigma \to 0 \quad (2.19)$$

$$\frac{K_m}{D} \left( \frac{\partial T}{\partial \sigma} \frac{\partial S}{\partial \sigma} \right) = 0, \sigma \to -1 \quad (2.20)$$

The boundary conditions of Eq. 2.7 and Eq. 2.8 are calculated with the turbulence closure constants and friction velocity at the top or bottom.

2.2.3 Numerical Scheme of POM

The dynamics of coastal circulation contains fast moving external gravity waves and slow-moving internal gravity waves, in order to make rational use of computational ability, a mode splitting technique was adopted in the numerical scheme of POM (Blumberg and Mellor 1987). The mode splitting technique divides the calculation processes into two parts: external mode and internal mode. External mode is two-dimensional with short time step and this mode is integrated from three-dimensional internal mode with a longer time step. The short time step of external mode was limited by the Courant-Friedrichs-Levy (CFL) computational stability condition (Courant et al. 1967) due to the explicit form in horizontal direction. The consumption of computing time in external mode is effectively reduced through this technology. The
external mode mainly provides surface elevation to the internal mode and the internal mode provides integrals of momentum advection, density and bottom stress to the external mode.

2.2.3.1 External Mode

The velocity equations of external mode are obtained by integrating the equations of internal mode, Eq. 2.2 to Eq. 2.4, from \( \sigma = -1 \) to \( \sigma = 0 \) with using the boundary conditions. Basic equations for external mode without vertical velocities are:

\[
\begin{align*}
\frac{\partial \bar{U}_D}{\partial x} + \frac{\partial \bar{V}_D}{\partial y} + \frac{\partial \eta}{\partial t} &= 0 \\
\frac{\partial \bar{U}_D}{\partial t} + \frac{\partial \bar{U}_D}{\partial x} + \frac{\partial \bar{U}_D}{\partial y} - F_x - f \bar{V}_D + gD \frac{\partial \eta}{\partial x} &= \langle wu(0) \rangle + \langle wu(-1) \rangle \\
&+ G_x - \frac{gD}{\rho_0} \int_{-1}^{0} \left[ D \frac{\partial \bar{p}}{\partial x} - \frac{\partial D}{\partial x} \sigma \frac{\partial \bar{p}}{\partial \sigma} \right] d\sigma d\sigma \\
\frac{\partial \bar{V}_D}{\partial t} + \frac{\partial \bar{V}_D}{\partial y} + \frac{\partial \bar{U}_D}{\partial x} - F_y + f \bar{U}_D + gD \frac{\partial \eta}{\partial y} &= \langle wv(0) \rangle + \langle wv(-1) \rangle \\
&+ G_y - \frac{gD}{\rho_0} \int_{-1}^{0} \left[ D \frac{\partial \bar{p}}{\partial y} - \frac{\partial D}{\partial y} \sigma \frac{\partial \bar{p}}{\partial \sigma} \right] d\sigma d\sigma
\end{align*}
\]

the over-bar variables mean vertically integration. With these equations, the external mode is in charge for the calculation of free elevation and vertically averaged velocities.

2.2.3.2 Internal Mode

The internal three-dimensional mode calculates the velocities, temperature, salinity and the turbulence quantities. A numerical method called the operator-splitting method is used during the calculation of internal equations. The variables are separated into a vertical diffusion step with implicit form to accommodate high resolution in vertical near the surface, and an advection plus horizontal diffusion step with explicit form. Taking the temperature equation as an example, the Eq. 2.5 could be presented as:

\[
\frac{\partial T_D}{\partial t} + \text{Adv}(T) - \text{Diff}(T) = \frac{1}{D} \frac{\partial}{\partial \sigma} \left( K_{\nu} \frac{\partial T}{\partial \sigma} \right) - \frac{\partial R}{\partial \sigma}
\]

(2.24)

where \( \text{Adv}(T) \) and \( \text{Diff}(T) \) are the advection and horizontal diffusion terms. The equation could be separated into two formulas. The first is the differencing of the advection and horizontal diffusion parts:

\[
\frac{DT - D^{n-1}T^{n-1}}{2\Delta t} = -\text{Adv}(T^n) + \text{Diff}(T^{n-1})
\]

(2.25)

and the second is the vertical diffusion part:
\[ \frac{D^{n+1}T^{n+1} - DT}{2\Delta t} = \frac{l}{D^{n+1}} \frac{\partial}{\partial \sigma} \left( K_H \frac{\partial T^{n+1}}{\partial \sigma} \right) - \frac{\partial R}{\partial \sigma} \]  \hspace{1cm} (2.26)

where $DT$ could be any three-dimensional variable. While the time differencing scheme called “leap frog” from $n-1$ to $n+1$ could cause divergence between odd and even time steps. A weak filter is adopted to remove divergence and smooth solution at each time step:

\[ T_s = T^n + \frac{\alpha}{2} \left( T^{n+1} - 2T^n + T^{n-1} \right) \]  \hspace{1cm} (2.27)

The interaction processes between external mode and internal mode are shown in Fig. 2.2. “DTE” and “DTI” represent the time interval of external mode and internal mode, respectively. The suffixes B and F appended to variables denote the time $t^{n-1}$ and $t^{n+1}$. The integrals parameters from internal mode at time $t^n$ are supplied to the external mode as labeled “Feedback”, and these values keep constant for the calculation in external mode from $t^n$ to $t^{n+1}$. The horizontal velocities sending to the internal mode are time averages over the interval from external variables, and the calculated free surface elevation is also provided to the internal mode.

**Fig. 2.2 Interaction processes between external mode and internal mode**

### 2.2.3.3 Differencing Scheme

The horizontal time differencing of POM is explicit and the vertical differencing is implicit, spatially the horizontal finite difference scheme uses the Arakawa C-grid and the grid arrangement is shown in Fig. 2.3. Although the model uses the finite differencing method, the advection operators in the primitive formulas are described in a finite volume form, take the
momentum equation Eq. 2.3 as an example, and the advection operator is in the same form separated as the temperature differencing equation Eq. 2.24, the advection term for $U$ is

$$-\text{Adv}(U)h_xh_y = \delta_x\left(Dh_xUU\right) + \delta_y\left(Dh_yUV\right) + h_xh_y\frac{\delta_x(oU)}{\delta\sigma} - fV Dh_xh_y \quad (2.28)$$

$$f = \frac{V\delta_x(h_y)}{h_xh_y} - \frac{U\delta_y(h_x)}{h_xh_y} \quad (2.29)$$

where $h_x$ and $h_y$ are the space variables for the orthogonal curvilinear grid, corresponding to $dx$ and $dy$ in rectangular grid; $\delta_x$ and $\delta_y$ represents the difference through the opposing faces of volume element; $\tilde{f}$ is curvature term caused by the transformation from rectangular to curvilinear grid. In the source code arrangement of POM, Eq. 2.28 is also calculated separately, horizontal advection, curvature term and the horizontal diffusion term $Dif(U)$ as shown in Fig. 2.24 for temperature equation are calculated first, so their vertical averages could be supplied and used first in the external mode. The vertical advection part is included and calculated in the following subroutine combined with the pressure gradient and Coriolis terms for the internal mode calculation.

![Fig. 2.3 Locations of variables and the grid arrangement](image)
The solution technique for vertical diffusion part as shown in Eq. 2.26 for temperature equation is from Richtmeyer and Morton (Mellor 1998), surface and bottom boundary conditions are also derived from this technique. The modification of boundary conditions in the POM source code needs to understand the detailed calculation processes. The calculation and setting of vertical diffusion and boundary conditions are included in a subroutine called proft in the source code and the derivation procedure of this the solution for vertical diffusion is presented as follows. With reference to Fig. 2.3, finite difference form of vertical diffusion could be described as:

\[
\frac{Df_k - Df_i}{2\Delta} = \frac{1}{D} \frac{\partial}{\partial \sigma} \left( K_H \frac{\partial T_i}{\partial \sigma} \right) - \frac{\partial R}{\partial \sigma}
\]

(2.30)

\[
f_k - f_i = \frac{2\Delta}{D^2} \frac{\partial}{\partial \sigma} \left( K_H \frac{\partial T_i}{\partial \sigma} \right) - \frac{2\Delta}{D} \frac{\partial R}{\partial \sigma}
\]

(2.31)

where \( k \) is the vertical layer number from second layer to bottom minus 1 layer. It is notable that \( D \) could be set to \( D \) as long as the same value of \( D \) is used in advection and horizontal diffusion equation, like Eq. 2.25. \( R \) here means not only the radiation term of temperature equation, but also some source or sink terms for other transport equations, such as turbulence, sediment or pollutants. An approximation second-order derivation of central difference scheme, as shown below, is used for the differencing of first term on the right side of Eq. 2.31.

\[
\left( \frac{\partial^2 F}{\partial x^2} \right)_k = \frac{F_{k+1} - 2F_k + F_{k-1}}{(\Delta x)^2} + O(\Delta x)^2
\]

(2.32)

Based on the variables arrangement of Arakawa C-grid, Eq. 2.31 could be further expanded:

\[
f_k - f_i = \frac{2\Delta}{D^2} \frac{1}{dz_k} \left( \frac{K_H}{dz_{k-1}} (f_{k-1} - f_k) - \frac{\partial}{\partial \sigma} (f_k - f_{k-1}) \right) - \frac{2\Delta}{D} \frac{1}{dz_k} (R_k - R_{k-1})
\]

(2.33)

through merging the similar terms:

\[
-f_{k-1} \left[ -\frac{2\Delta}{D} \frac{1}{dz_k} \frac{K_H}{dz_{k-1}} \right] + f_k \left[ -\frac{2\Delta}{D^2} \frac{1}{dz_k} \frac{K_H}{dz_{k-1}} - \frac{2\Delta}{D} \frac{1}{dz_k} \frac{K_H}{dz_{k-1}} - 1 \right] - f_{k+1} \left[ -\frac{2\Delta}{D^2} \frac{1}{dz_k} \frac{K_H}{dz_{k+1}} \right] = \frac{2\Delta}{D} \frac{1}{dz_k} (R_k - R_{k+1}) - f_k
\]

(2.34)

which could be written in the form as:

\[
-f_{k-1} c_k + f_k (c_k + a_{k-1}) - f_{k+1} a_k = d_k
\]

(2.35)

where
\[ a_k = -\frac{2\Delta t}{D^2} \frac{1}{dz_k} \frac{(K_H)_{k+1}}{dzz_{k+1}} \]
\[ c_k = -\frac{2\Delta t}{D^2} \frac{1}{dz_k} \frac{(K_H)_k}{dzz_{k-1}} \]
\[ d_k = \frac{2\Delta t}{D} \frac{1}{dz_k} (R_k - R_{k+1}) - f_k \]  

(2.36)

Now assume a general solution for **Eq. 2.35** with the form:
\[ f_k = ee_k \times f_{k+1} + gg_k \]  

(2.37)

through this equation to get \( f_{k-1} \) and insert \( f_{k-1} \) into **Eq. 2.35** and adjust it to the general solution format, the parameters in general solution could be obtained.
\[ ee_k = \frac{a_k}{a_k + c_k (1 - ee_{k-1})} - 1 \]  

(2.38)
\[ gg_k = \frac{c_k \times gg_{k+1} + d_k}{a_k + c_k (1 - ee_{k-1})} - 1 \]  

(2.39)

Values of \( a_k, c_k \) and \( d_k \) are calculated by **Eq. 2.36**, use the recursion from \( ee_1 \) and \( gg_1 \) to get \( ee_{k-1} \) and \( gg_{k-1} \), and the general solution will be solved, while the value of \( ee_1 \) and \( gg_1 \) on the surface layer are necessary, which are the surface boundary conditions. On surface layer where \( k=1 \), **Eq. 2.33** is presented as:
\[ f_i - f_i - \frac{2\Delta t}{D^2} \frac{1}{dz_i} \left[ (K_H)_i (f_0 - f_i) - \frac{(K_H)_i}{dzz_i} (f_i - f_1) \right] - \frac{2\Delta t}{D} \frac{1}{dz_i} (R_i - R_2) \]
\[ = - \frac{2\Delta t}{D} \frac{1}{dz_i} \left[ - \frac{(K_H)_i}{Ddzz_i} (f_0 - f_i) + (R_i - R_2) \right] - \frac{2\Delta t}{D} \frac{1}{dz_i} \left[ wtsurf + (R_i - R_2) \right] \]  

(2.40)

where the pink part is called \( wtsurf \) in the POM code, which means variables’ surface fluxes;

Blue part is \( a_1 \) as shown in **Eq. 2.36**. The **Eq. 2.40** could be further changed into:
\[ f_i = \frac{a_i}{a_i - 1} f_2 - \frac{1}{a_i - 1} f_i + \frac{1}{a_i - 1} \left[ \frac{2\Delta t}{D} \frac{1}{dz_i} wtsurf + (R_i - R_2) \right] \]  

(2.41)

Compared with the general solution of **Eq. 2.37**, on surface layer, the parameters of surface boundary condition are obtained:
\[ ee_i = \frac{a_i}{a_i - 1} \]  

(2.42)
\[ gg_i = \frac{1}{a_i - 1} \left[ \frac{2\Delta t}{D} \frac{1}{dz_i} wtsurf + (R_i - R_2) \right] - f_i \]  

(2.43)
At bottom zero heat or other fluxes are specified, the bottom boundary conditions could be derived by using the same method:

\[ f_{b-1} = \frac{c_{b-1} \times g b_{b-2} - f_{b-1} + \frac{2\Delta t}{D} \frac{I}{dz} (R_1 - R_2)}{c_{b-1}(I - ee_{b-2}) - I} \]  \hspace{1cm} (2.44)

The source or sink terms on surface layer of temperature are heat fluxes \( w_{surf} \) and short wave radiation penetration \( R \), for other factors, surface boundary conditions are calculated through corresponding \( w_{surf} \) and bottom boundary conditions are calculated through changing the values of \( R_1 \) and \( R_2 \) for source or sink terms.

### 2.2.4 Numerical Solution Procedures

Main program in POM is called \textit{pom2k}, which contains model initialization and definition of variables in the beginning. The pre-processing works before the simulation include the preparations for different input files like initial condition, topography depth, grid distribution, external forcing and so on. All the input files and subroutines will be called by the main program. The cycling times of internal mode are based on the setting of internal model time step setting and total simulation duration. The external two-dimensional mode is embedded in the internal mode loop, and cycling times are the quotient between external mode and internal which is defined as \textit{ISPLIT} in source code.

Before the start of external loop, internal mode will calculate the horizontal portions of momentum advection first and integrate them in the vertical direction for the use in external mode. After the external mode finishes the loop of calculation for free surface elevation and vertically averaged velocities, calculation results will be sent to the internal mode for further computation. Due to the different truncation errors between the internal and external modes, the vertical integrals may depart slightly from calculated averaged velocities with longtime calculation. The internal velocities will be adjusted as the mean value over the calculation interval to match the computation of external mode.

After the calculation of the vertical velocity and verifying the vertical boundary conditions, the internal mode solves the 2.5 Mellor and Yamada turbulence closure model, temperature equation, salinity and momentum equations, sequentially. All the calculated variables will be limited with an open boundary condition subroutine and user could modify the open boundary condition settings according to the research objectives, the POM source code also provides a variety of open boundary conditions for selection. The flow chart of the code is shown in Fig. 2.4 and the boxes with sidebars contain subroutines.
Fig. 2.4 Flow chart of POM source code
3. Hydrodynamic Simulation and Precipitation Module

3.1 Field Observation and Simulation Domain

Although numerical simulation is a more cost-effective approach for hydrodynamic researches, field observation is necessary not only to provide basic data for simulation, but also for the verification of simulated results. It can reflect the real situation of water quality with measuring equipment and is also the only way to study the dynamics of water quality without better numerical model. A periodic joint observation has been carried out for water and sediment sampling since 2006 in Minamata Bay by National Institute for Minamata Disease, Nagasaki University and Kyushu University (Matsuyama et al. 2010). Centralized observations in summer or winter were taken every year depending on weather conditions, while lower observation frequency in other seasons. Three fixed observation points named station 1 to 3 (ST.1, ST.2, and ST.3) in Minamata Bay were chosen for sample collection, and station 4 and station 5 (ST.4 and ST.5) were added in recent years, as shown in Fig. 3.1. An integrated electrode sensor shown in Fig. 3.2 was used to measure several ocean water parameters, such as water depth, dissolved oxygen, water temperature, salinity, pH, chlorophyll-a, and turbidity. The data were measured on boat through a connecting cable and recorded in a handy controller, meantime, water sampling was conducted. The measurements of ocean water indexes along with sediment were conducted in the whole water depth, while water sampling for analyzing mercury (also includes sediment analysis) was carried out on water surface, 6 m, 10m, bottom+1 m, and bottom +0.1 m. All water parameters were measured continuously until to the water bottom and the value on integer depth would be averaged after returning to the laboratory. The specific positions located with GPS device and water sampling depths at five stations are shown in Table 3.1.

The simulation domain covering all the Yatsushiro Sea is shown in Fig. 3.1. Uniform grid arrangement of 119 × 119 was taken and horizontal resolution was about 500 m in both longitude and latitude directions. Bold lines in the lower left quarter of Fig. 3.1 represent the position of open boundary conditions for tidal elevation during simulation. ST.1 to ST.3 are located in the bay area, ST.4 is located near the middle between ST.1 and ST.5, and ST.5 is closed to the mouth of the Komenotsu River. Water depths shown in the figure demonstrate south area is deeper than north area in general. Measuring sequence is from south to the north side (ST.5, ST.4, ST.1, ST.2, and ST.3) on a fishing boat, which will take around two hours.
Fig. 3.1 Research region and five sample collection stations

Table 3.1 Location of observation stations and sampling depths

<table>
<thead>
<tr>
<th></th>
<th>ST.1</th>
<th>ST.2</th>
<th>ST.3</th>
<th>ST.4</th>
<th>ST.5</th>
</tr>
</thead>
<tbody>
<tr>
<td>Latitude</td>
<td>32º11'9&quot; N</td>
<td>32º11'33&quot; N</td>
<td>32º12'7&quot; N</td>
<td>32º09'17&quot; N</td>
<td>32º08'11&quot; N</td>
</tr>
<tr>
<td>Longitude</td>
<td>130º21'46&quot; E</td>
<td>130º22'13&quot; E</td>
<td>130º22'16&quot; E</td>
<td>130º20'56&quot; E</td>
<td>130º19'50&quot; E</td>
</tr>
<tr>
<td>Water Sampling depths (m)</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td></td>
<td>6</td>
<td>6</td>
<td>6</td>
<td>6</td>
<td>6</td>
</tr>
<tr>
<td>Bottom+1</td>
<td>Bottom+1</td>
<td>Bottom+1</td>
<td>Bottom+1</td>
<td>Bottom+1</td>
<td>Bottom+1</td>
</tr>
<tr>
<td>Bottom+0.1</td>
<td>Bottom+0.1</td>
<td>Bottom+0.1</td>
<td>—</td>
<td>—</td>
<td>—</td>
</tr>
</tbody>
</table>

Because numerical simulation requires large amounts of continuous measurement data for comparison and verification, field observation data with longer interval time could not be adopted. Due to the typhoon or other weather effects, centralized observations were not able
Fig. 3.2 Integrated electrode sensors

to be carried out every year, therefore, simulation duration for simulating temperature and salinity was chosen from July 5th to July 20th, 2016, six times’ field observations on July 5th, 7th, 9th, 11th, 15th, and 17th have been conducted during this period, and observation data of last five times were selected for comparing the simulated results.

3.2 Hydrodynamic Simulation with Original POM

3.2.1 Model Setting for Original POM

The σ-coordinate was used to deal with topographical variability in vertical direction and divided into 10 layers with 3 logarithmic layers on ocean surface. Due to the mode splitting technique used in POM, time interval was set to two values, 1 second for external two-dimensional mode and 5 seconds for internal three-dimensional mode. The surface elevation was chosen as the tidal forcing on two open boundaries. Due to the lack of tidal gauge data, a tidal prediction method (LOUSHA 2013) based on historical data was applied to obtain the harmonic constants of four tidal constituents (M2, S2, K1, O1) on July 5th, 2016, and the predicted harmonic constants were interpolated on two open boundaries by using inverse distance weighted method. Wind velocity was obtained from the Japan Meteorological Agency, wind stress was calculated during POM pre-processing work and interpolated into each internal time step by a linear interpolation method. Thermal radiation boundary condition in the first simulation was set to constant with averaged heat fluxes. Three rivers exist in the simulation domain, Kuma River in the north and two B-class rivers, the Minamata River and Komenotsu
There are no river discharge records for B-class river, therefore, the discharge of two B-class rivers was set to a constant value of 40 m³ s⁻¹. Recorded real-time discharge of Kuma River from Japan Ministry of Land, Infrastructure, Transport, and Tourism was interpolated into each internal time step. Five stations’ measured temperature and salinity data on July 5th were interpolated on the whole simulation domain as initial conditions by using the inverse distance weighted method.

<table>
<thead>
<tr>
<th>River systems</th>
<th>Latitude</th>
<th>Longitude</th>
</tr>
</thead>
<tbody>
<tr>
<td>Kuma River</td>
<td>32°30'01&quot; N</td>
<td>130°33'54&quot; E</td>
</tr>
<tr>
<td>Minamata River</td>
<td>32°13'25&quot; N</td>
<td>130°23'45&quot; E</td>
</tr>
<tr>
<td>Komenotsu River</td>
<td>32°07'25&quot; N</td>
<td>130°20'04&quot; E</td>
</tr>
</tbody>
</table>

### 3.2.2 Simulation Results of Original POM

In order to provide a basic environment for the simulation of sediment and mercury transportation, temperature and salinity field were simulated first through the original POM model. Total running time of hydrodynamic model is about four hours. Ocean water in vertical direction was roughly divided into three layers, surface layer, middle layer, and bottom layer, in order to facilitate comparison of measured and simulated data. Comparisons of simulated and measured results of temperature and salinity at five stations are shown from Fig. 3.3 to Fig. 3.5. Simulation results of middle and bottom layers on Fig. 3.4 and Fig. 3.5 show good agreement, most of the errors between measurement and simulation are within 1 °C or PSU, only during the later simulation period of middle layer small divergence occurs. However, large deviations appear at surface layers during simulation on Fig 3.3, simulated surface temperature was significantly lower than measured data of five observation stations, while the simulated surface salinity was abnormally higher than measurements, deviation even reached to 10 °C or PSU at some certain times. This abnormal situation indicates that the surface boundary conditions of salinity and temperature are inconsistent with reality. Through consulting historical datum, a large amount of precipitation as shown in Fig. 3.6 was found in the research area during the simulation period, which was caused by the plum rain season. From May to July every year Minamata area, Kumamoto Ken, is under the plum rain season, heavy rainfall will
Fig. 3.3 Comparison of measured and simulated data on surface layer

Fig. 3.4 Comparison of measured and simulated data on middle layer
Fig. 3.5 Comparison of measured and simulated data on bottom layer

Fig. 3.6 Precipitation during simulation period in Yatsushiro City and Minamata City

cause dilution of surface ocean water, correspondingly, both surface temperature and salinity will be changed. The simulation results indicated that surface boundary conditions of temperature and salinity need to be modified and the precipitation effect on surface salinity and
temperature in the Yatsushiro Sea couldn’t be neglected, especially during the rainy season. Therefore a precipitation module was developed and added into POM. Meanwhile, new thermal radiation boundary conditions which could change temporally and spatially were presented instead of the constant setting of thermal radiation boundary condition for providing a more accurate simulation of diurnal and regional temperature change.

3.3 Precipitation Module and New Thermal Radiation Boundary Conditions

Ocean researches related to the rainfall impact have been carried out in recent years. Jones et al. (2016) presented near surface salinity profile with rainfall events by a rain impact model. Ho et al. (2017) analyzed the influence of rainfall in different degrees on sea surface salinity based on satellite remote sensing data. Li et al. (2016) improved the rainfall prediction from the sea surface salinity change by establishing linkage between these two factors. The Finite-Volume Community Ocean Model has the rainfall module to for the calculation of precipitation fields to drive simulation provided by Fifth-Generation Penn State/NCAR Mesoscale Model (MM5) (Chen et al. 2005). As to POM, researches about precipitation effect are rarely carried out. Origin POM applied the river discharge to represent the rainfall effect on salinity field simulation, while previous researches on the Yatsushiro Sea showed the river discharge influence was normally around the river mouse areas. Among these five observation stations, only ST.5 which located on the Komenotsu River mouth was significantly affected by the river inflow, however, the effect on other four stations was relatively weaker. Simulation results in Fig. 3.3 showed the necessity of adding the rainfall precipitation effect into the model in rainy season. Consequently, a precipitation module was developed under new thermal radiation boundary conditions and coupled with the POM hydrodynamic model.

3.3.1 Precipitation and River Inflows Forcing

Precipitation data were treated as vertical velocity on surface ocean layer according to the setting rules for non-zero flow across the sea surface in POM source code, precipitation data of Minamata City in Fig. 3.6 was adopted and interpolated into the whole simulation domain to improve the simulation accuracy in Minamata Bay. Surface salinity flux between ocean and atmosphere was calculated by following equation.

\[ W_{surf} = vflux \times (S_A - S_{surf}) \]  (3.1)
where $W_{surf}$ is surface salinity flux, $v_{flux}$ is the volume flux through water column surface and specified as surface layer velocity, $S_A$ is fresh water salinity and $S_{surf}$ is ocean surface salinity. River discharge of B-class river is still a calibration value due to the lack of discharge records. Two B-class rivers’ discharge was set to a constant value of 70 m$^3$/s$^{-1}$ and doubled if the rain rate was larger than 4 mm after calibration. Similar with the previous original simulation, recorded discharge of Kuma River was used in simulation. Surface salinity and temperature flux boundary condition for river inflows are shown below.

$$\frac{K_H}{D} \left( \frac{\partial S}{\partial \sigma}, \frac{\partial T}{\partial \sigma} \right) = \left( \frac{SR}{A} \frac{TR}{A} \right)$$

(3.2)

where $K_H$ is the vertical turbulent diffusivity coefficient; $S$ is salinity; $T$ is temperature; $R$ is river discharge and $A$ is grid area where the rivers locate.

### 3.3.2 New Thermal Radiation Boundary Conditions

Field measurements for the heat fluxes are difficult and these values are generally parameterized by using the commonly available hydro-meteorological data. Constant setting of thermal radiation boundary conditions in the previous simulation caused unreasonable simulation results of ocean surface temperature, meantime the effect of precipitation on surface temperature should also be considered. Thus new heat flux boundary conditions were developed which could vary temporally and spatially, also including the precipitation effect on temperature. In the present simulation of POM, surface heat fluxes consist of four major heat flux components and the net heat flux $Q$ on ocean surface is represented by

$$Q = Q_S - (Q_L + Q_{SE} + Q_{LA})$$

(3.3)

where $Q_S$ is the short wave solar radiation penetration reaches on ocean surface, $Q_L$ is the net long wave radiation which emitted from the ocean surface, $Q_{SE}$ is the sensible heat flux and $Q_{LA}$ is latent heat flux. In the source code of POM, input of thermal radiation boundary conditions was separated into two part, parameter “swrad” for the short wave radiation penetration $Q_S$ and parameter “wtsurf” for the sum of net long wave radiation $Q_L$, sensible heat $Q_{SE}$, and latent heat $Q_{LA}$. Following Rosati and Miyakoda (Rosati and Miyakoda 1988), a series of formulas have been adopted for the calculation of $Q_S, Q_L,$ and $Q_{LA}$.

(a) Short wave solar radiation flux

$$Q_S = Q_t \left( 1 - 0.62C + 0.0019 \varphi \right)(1 - \alpha)$$

(3.4)

$$Q_t = Q_0 \tau + \left[ \frac{(1 - \alpha)Q_o - Q_o \tau}{2} \right]$$

(3.5)
\[ Q_o = \frac{\cos z \times D_f \times J_o}{a^2} \]  \hspace{1cm} (3.6)

\[ \varphi = 90^\circ - |\theta + \delta| \]  \hspace{1cm} (3.7)

\[ \cos z = \sin \theta \sin \delta + \cos \theta \cos \delta \cosh \]  \hspace{1cm} (3.8)

\[ h = 15 \left( t + \frac{(\bar{\theta} - 135^\circ)}{15} \right) - 12 \]  \hspace{1cm} (3.9)

\[ \delta = 23.45^\circ \frac{\pi}{180^\circ} \sin \left[ 2\pi \left( \frac{284 + n}{365} \right) \right] \]  \hspace{1cm} (3.10)

where \( Q_r \) is total solar radiation reaching on ocean surface under clear sky which is approximated by the sum of direct component of solar radiation reaching the ocean surface and diffuse sky radiation under cloudless conditions; \( C \) is fraction of cloud cover which was obtained from satellite pixel coverage with the value of 75% on July, 2016; \( \alpha \) is ocean surface albedo, set to 6% (Hirose et al. 1996); \( Q_o \) is solar radiation at top of atmosphere; \( \tau = 0.7 \) is atmospheric transmission coefficient; \( A_a = 0.09 \) is ozone absorption coefficient; \( D_f \) is daylight fraction with month averaged value 50%; \( J_o \) is solar constant which equals 1353 Wm\(^{-2}\); \( a \) is the radius vector of earth, calculation method was referred to the ECOMSED and MM5 models; \( \varphi \) is solar noon altitude in degrees, \( \theta \) is latitude and \( \delta \) is sun declination angle; \( z \) is zenith angle, \( h \) is the sun’s hour angle, \( \bar{\theta} \) is longitude and \( t \) is time on simulation day; \( n \) is the simulation day number in 2016. Although the calculation of short wave solar radiation flux is quite complicated, the adoption of different solar angle change parameters could reflect the real sun movement in daytime and night. The simulated short wave solar radiation will be closer to the actual value in various locations to reproduce more accuracy diurnal and regional temperature change.

(b) Net long wave radiation flux

\[ Q_L = \varepsilon S_B T_s^4 \left( 0.39 - 0.05 E_v^{1/2} \right) \left( 1 - BC \right) + 4 \varepsilon S_B T_s^4 \left( T_s - T_A \right) \]  \hspace{1cm} (3.11)

\[ E_v = r E_{\text{sat}} \left( T_A \right) \]  \hspace{1cm} (3.12)

\[ E_{\text{sat}} \left( T_A \right) = 6.1078 \exp \left[ 7.5 T_A / \left( T_A + 237.3 \right) \right] \]  \hspace{1cm} (3.13)

where \( \varepsilon = 0.97 \) is ocean emissivity; \( S_B = 5.67 \times 10^{-8} \) is the Stefan-Boltzmann constant; \( E_v \) is vapor pressure and computed by a polynomial approximation from Lowe (Lowe 1977); \( 1 - BC \) is the cloudiness correction fraction and \( B \) is a latitude varying constant (Fung et al. 1984), set to 0.63;
$T_S$ and $T_A$ are temperatures of ocean and atmosphere; $r$ is relative humidity and $E_{sat}(T_A)$ is saturation vapor pressure. The last term on the right side of Eq. 3.11 is an additional term due to air-sea temperature difference.

(c) Latent heat flux

\[
Q_{LA} = \rho_A L C_A [V_w E_{sat}(T_s) - r E_{sat}(T_A)] (0.622 / P)
\]

(3.14)

where $\rho_A = 1.225 \text{ kgm}^{-3}$ is air density; $L = 2.501 \times 10^6 \text{ Jkg}^{-1}$ is latent heat of vaporization of water, $C_A = 1.1 \times 10^3$ is the turbulent exchange coefficient; $V_w$ is the wind magnitude; $E_{sat}(T_s)$ and $E_{sat}(T_A)$ are the saturation vapor pressure at ocean and atmosphere temperature; $P = 1010\text{ mb}$ is the surface air pressure.

(d) Sensible heat flux

Effect of rainfall to temperature is mainly through the sensible heat, hence the sensible heat flux forcing should consider the cooling of surface ocean caused by mixing of cooler precipitation. Gosnell et al. (1995) used theoretical models to calculate the sensible heat flux into ocean induced by precipitation. Fairall et al. (1996) analyzed the bulk variables related to air-sea fluxes and the algorithm was used in the Finite Volume Community Ocean Model (FVCOM) which coupled with the MM5 model (Chen et al. 2005) for the calculation of precipitation fields to drive simulation. To simulate the precipitation effect on the temperature field, sensible heat flux was separated into two parts in the new improved POM, heat conduction by turbulent transfer from ocean surface to the atmosphere and the cooling of precipitation.

\[
Q_{se} = Q_{TU} + Q_p
\]

(3.15)

where $Q_{TU}$ is the turbulent transfer heat flux calculated by

\[
Q_{TU} = \rho_A C_A C_W [V_w (T_s - T_A)]
\]

(3.16)

here $C_A = 1005 \text{ Jkg}^{-1}\text{K}^{-1}$ is the specific heat capacity of air.

$Q_p$ is the sensible heat flux caused by precipitation, formulas are given as

\[
Q_p = -RC_W \beta (T_s - T_A) (I + B^{-1})
\]

(3.17)

where $R$ is rainfall rate; $C_W = 4186 \text{ Jkg}^{-1}\text{K}^{-1}$ is water heat capacity; $\beta$ is the Clausius-Clapeyron wet bulb factor given in Eq. 3.18; $B$ is bulk Bowen ratio calculated in Eq. 3.22.

\[
\beta = (1 + \left( \frac{Ld \eta}{dC_s dT} \right))^{-1}
\]

(3.18)

\[
d_c = 2.11 \times 10^{-5} \left( \frac{T}{273.16} \right)
\]

(3.19)
where $d_v$ and $d_h$ are diffusivities of water vapor and heat, both parameters are calculated by using the same formulas of MM5 and FVCOM model (Chen et al. 2005); $q_s$ is the saturation specific humidity, $T = T_A + 273$ is used to changes degree Celsius to degree Kelvin, and term $dq_s/dT$ is solved through the Clausius-Clapeyron relation (Fairall et al. 1994). By using the Clausius-Clapeyron equation (Wallace and Hobbs 2006), the following relationship could be obtained.

$$
\frac{dq_s}{dT} = \frac{q_s(T_A) \times (0.622L)}{RT^2} 
$$

(3.21)

where $q_s(T_A)$ is saturation humidity at air temperature, which is the product of relative humidity and specific humidity at saturation calculated from Buck (Buck 1981); $R=287$ is the gas constant for dry air. The bulk Bowen ratio is calculated by:

$$
B = \frac{C_A(T_s - T_A)}{L[q_s(T_s) - q_s(T_A)]} 
$$

(3.22)

where $q_s(T_s)$ is specific saturation humidity of ocean.

Except for the impact on sensible heat flux, precipitation could also affect the ocean surface stress, which needs the wind speed above 10 meters ocean surface for calculation. The research of Fairall (1996) indicates averaged 5m/s wind speed and 0.5mm/h rainfall have 2% effect to surface stress, while in this case averaged wind speed and rainfall are 1.3 m/s and 1mm/h respectively, consequently the slight precipitation effect on ocean surface momentum is not considered in this study.

### 3.3.3 Simulation Results Coupled with New Precipitation Module

Based on the new equations for surface boundary conditions of temperature and salinity, some subroutines for calculating new boundary conditions were programmed and coupled into the original POM structure and different real-time variables are read and calculated through the main program. With the input of precipitation data and new boundary conditions, the temperature and salinity fields were simulated again. The new model was carried out under the condition with rainfall and without rainfall, separately, in order to present the improvement of new thermal boundary conditions under sunny days. Simulation results under different precipitation conditions were compared with the previous original POM, with the same elevation open boundary conditions, initial conditions, grid arrangement and other external
forcing with previous setting.

3.3.3.1 Model Verification

Due to the lack of specific measured data of tide and current velocity, only tidal elevation was adopted for the verification of model simulation results. Japan Meteorological Agency predicted elevation (minus the datum surface value) and hourly measured elevation of Kuma River from Japan Water Information System were used as the contrasting data. According to the predicted location provided by the Japan Meteorological Agency, simulated elevation of ST.3 was chosen for comparison with Japan Meteorological Agency predicted elevation. As shown in Fig. 3.7, simulated elevation of ST.3 based on the tidal prediction method matched well with the official predicted data, simulated elevation range may slightly less than measurements at the initial stage. Nevertheless, Because Kuma River which belongs to class A river classification in Japan is a large river system which has many branches, the Japan Water Information System provides various measured elevation data in different observation stations, we tried to choose the station which is closest to the Kuma River location set in the simulation grid points. In addition to slight deviations at middle stage of simulation, agreement of simulated results with measurements is acceptable at Kuma River.

Fig. 3.7 Verification of elevation at station 3 and Kuma River
3.3.3.2 Comparison of Simulation Results

Comparison of five stations’ measured and simulated surface data during five times’ field observation are shown in Fig. 3.8. Four kinds of data are presented: black straight lines are measured surface data, black dotted lines are simulated surface data by previous original POM, blue straight lines are the simulated surface results of new model with precipitation input and red straight lines are simulated surface results of new model without precipitation.

Fig. 3.8(a) shows the comparison results of surface salinity. Under the circumstance of no precipitation, the surface salinity simulated by the new model had a slight decrease compared with previous simulation (red straight lines and black dotted lines) after amending the freshwater boundary condition. Two simulated results of the new model (red and blue straight lines) overlapped on July 7th because no rainfall happened. The rainfall decrease on surface salinity of new model is evident on other observation days under new model. Simulation results showed significant improvement under the precipitation condition, which caused 2-7 PSU reduction of five stations’ salinity in comparison with the previous model (blue straight lines and black dotted lines), especially on the last two observation days simulated results show good
agreement. The abnormal simulated data of ST.5 on July 7\textsuperscript{th} was caused by the constant setting of river discharge which had an obvious impact on the river mouse area, where ST.5 located.

Fig. 3.8(b) shows the comparison results of surface temperature. Even with no rainfall input, the modification of heat flux boundary conditions caused about 2-4 degrees increase of temperature compared with previous model (red straight lines and black dotted lines), and the promotion was more visible in the middle and late stages of simulation. Precipitation impact to the surface temperature field is about averaged 0.2 degrees during the two simulations of new model (blue and red straight lines), the small scale cooling of rainfall was probably due to the daylight field observation when short wave solar radiation heat flux dominated, while the turbulent sensible heat flux fluctuation caused by precipitation is about an order of magnitude less than the latent heat flux and solar radiation flux. Another reason is that although rainfall is frequency during simulation period, the selected observation duration is most during sunny days, ocean surface temperature rises soon after the cooling effect of precipitation due to the high radiation in summer. However, compared with the previous constant heat boundary conditions, new heat flux forcing can reproduce surface temperature well and the numerical results under precipitation input showed good agreement with measured data. Small error of ST.5 was caused by lack of the accurate temperature boundary condition for river inflow.

Fig. 3.9 and Fig. 3.10 show comparison results of middle and bottom layers. Compared with surface layer, variation of salinity and temperature and impact from surface boundary conditions are reasonably small, correspondingly, the display interval of vertical coordinates is appropriately enlarged compared the presentation of Fig. 3.4 and Fig. 3.5. Three kinds of data are presented: black straight lines are measured surface data, black dotted lines are simulated surface data by previous original POM, blue straight lines are the simulated surface results of new model with precipitation input, and the simulation results of new model without rainfall input is not presented for middle and bottom layers. Comparison results of middle and bottom layers’ salinity shown in Fig. 3.9 (a) and Fig. 3.10 (a) demonstrate that new model with precipitation slightly improves the salinity on lower layers. Simulated salinity on both laters further approaches the measurements. The decrease of salinity on surface layer with precipitation doesn’t cause the ensemble salinity reduction in vertical directions, which illustrates surface boundary condition effect is chiefly concentrated in upper layers. The sudden drops of ST.3 on July 9\textsuperscript{th} and July 11\textsuperscript{th} are difficult to achieve during numerical simulation without specific origin. The temperature comparison of middle and bottom layers in Fig. 3.9 (b) and Fig. 3.10 (b) reveals temperature increase in the whole simulation areas with external heat radiation fluxes input, and simulation results show good agreement in lower layers. Although
(a) Salinity (PSU)  
(b) Temperature (°C)  

Fig. 3.9 Comparison of measured and simulated data on middle layer

(a) Salinity (PSU)  
(b) Temperature (°C)  

Fig. 3.10 Comparison of measured and simulated data on bottom layer
with a small temperature and salinity fluctuation at lower layers, simulation results are still more accuracy compared with previous model.

**Fig. 3.11** and **Fig. 3.12** present temperature and salinity difference on surface layer at noontime between new model with precipitation and previous model after 5 and 10 simulation days. In both figures: (a) 12:00 on July 10th during slack tide after flood tide and (b) 12:00 on July 15th during slack tide after ebb tide. As shown in **Fig. 3.11**, temperature improvement under new model in the whole Yatsushiro is significant compared with previous model, surface temperature increases about 2 degrees in most areas. It is indicated that temperature difference of July 10th (**Fig. 3.11a**) is about 1-2 degrees smaller than that of July 15th (**Fig. 3.11b**) generally. Through inspecting the rainfall input data shown in **Fig. 3.6**, although rainfall at noon time of both days was 0 mm, precipitation lasted for a whole morning on July 10th, while rainfall has stopped one day before July 15th. It can be deduced that rainfall caused cooling of temperature as set in the sensible heat boundary condition and cloud cover stopped most part of solar radiation to ocean surface. Consequently, small scale cooling from precipitation module with rainfall input shown in the comparison results of surface temperature in **Fig. 3.8 (b)** is probably because the observation time is most during sunny days or long time after rainfall, high magnitude short wave radiation will rise up the surface temperature soon with sufficient sunlight in summer season. Precipitation effect revealed on the salinity different in **Fig. 3.12** is generally more visible, although salinity of some areas on southwest has a slight increase compared with previous model. Salinity decrease is significant on July 10th after total morning rain in contrast with sunny day on July 15th, the corresponding increased river discharge of Komenotsu River and Minamata River in model setting coupled with rainfall input reduces the surface salinity field in the whole south Yatsushiro Sea. New freshwater boundary conditions still affected the surface salinity field despite rain has stopped since July 14th.

Due to the slight fluctuation of temperature and salinity at middle and bottom layers, only surface layers’ temporal variation data are presented. **Fig. 3.13 (a)** shows the temporal variation of surface salinity and precipitation data during the simulation period. The salinity changing tendency of ST.1, 2 and 3 basically kept consistent due to the relatively close position, all the three station were located in the Minamata Bay. Although ST.3 is located in shallower water compared with other two stations, the salinity value keeps in a uniform level. In the mid-term simulation of ST.3, slight difference was shown and this may be caused by the discharge of Minamata River, ST.3 located near to the estuary boundary. Simulation results of ST.5 presented an apparent response to the discharge of Komenotsu River, the special river mouth
Fig. 3.11 Temperature difference on surface layer of Yatsushiro Sea between new model with precipitation and previous model at noon after: (a) 5 days, (b) 10 days

Fig. 3.12 Salinity difference on surface layer of Yatsushiro Sea between new model with precipitation and previous model at noon after: (a) 5 days, (b) 10 days
Fig. 3.13 Simulated temporal variation of surface data at five stations

(a) Salinity (PSU) and rainfall (mm/h)
(b) Temperature (°C) and average solar radiation (W/m²)

Fig. 3.14 Simulated surface flow field in the Yatsushiro Sea during flood tide and ebb tide

(a) Flood tide on July 8th
(b) Ebb tide on July 15th
(a) Flood tide on July\textsuperscript{8th}  

(b) Ebb tide on July\textsuperscript{15th}

Fig. 3.15 Simulated depth-averaged flow field in the Yatsushiro Sea during flood tide and ebb tide

location kept surface salinity in a low level. Due to the lack of B-class river discharge, discharge of Komenotsu river was treated as a calibration term due to the visible impact on surface salinity of ST.5. Five stations’ surface salinity increased on July 15\textsuperscript{th} because rainfall stops. Fig. 3.13 (b) shows the temporally changing surface temperature and averaged solar radiation reaching on ocean surface of five stations. In fact, simulated results of solar radiation are slightly different at five stations. Here an average solar radiation value of five stations was taken to present the comparison results. Compared with the constant setting of heat parameters, the new thermal radiation heat boundary condition could accurately reflect diurnal variation of surface ocean temperature. The increase and decrease of temperature roughly corresponded with the variation of radiation. The overall improved temperature is more consistent with actual temperature changes on ocean surface.

Fig. 3.14 shows the surface flow field in Yatsushiro Sea during flood tide on July\textsuperscript{8th} and ebb tide on July\textsuperscript{15th} under large elevation variation to present current dynamics with same reference velocity. It is evident that flow velocity is in a high magnitude on the southwest Nagashima channel compared with other areas, which indicates the Nagashima channel plays a dominant role in the ocean water exchange of the Yatsushiro Sea, and this simulation result is in accordance with the research of Murakami et al. (2004). The flow velocities in north part are
stronger than south part during flood tide, and strong currents from the Nagashima channel flow to north, which leave the southern sea in a stable level. Due to the relative closure environment in south area, currents from north would converge the south currents during ebb tide and flow out of Yatsushiro Sea through the Nagashima channel. **Fig. 3.15** shows the depth-averaged flow field during flood tide and ebb tide in the same simulation duration. Flow directions are basically consistent with surface flow field. However, magnitude of flow velocity becomes weaker compared with the flow field on surface layer, which indicates the surface current is dominant in the vertical current distribution.

### 3.4 Chapter Summary

With the development of computer technology, researches about ocean dynamics are changing from theoretical study, laboratory experiment and field observation to the combination with ocean circulation numerical models. The development of numerical model is constantly improving with more simulation functions for different detailed research objectives, more streamlined code structure, and more accurate simulation results. Residual mercury dynamic has been the research emphasis since mercury contamination was publicly recognized in Minamata Bay. In addition to the periodic field observation to get ocean water factors, a numerical model which provides a more effective approach for comprehending the ocean dynamics is necessary. In order to provide basic environment for the simulation of sediment and mercury transportation, a new precipitation module has been constructed and coupled with the hydrodynamic model POM with modified heat flux forcing which varied temporally and spatially.

The hydrodynamic simulation with original POM in Minamata Bay was presented firstly. Simulation results of July 2016 obtained from the previous model showed relatively large deviation both in temperature and salinity on surface layer compared with measurements, while the simulation results on lower layers showed acceptable agreement. This problem was caused by the constant setting of surface thermal radiation boundary conditions and frequent precipitation. Consequently, the hydrodynamic model was improved with a new precipitation module to adjust the rainfall impact in plum rain season, also with new thermal radiation boundary conditions which could change temporally and spatially. It is indicated that the accuracy of predicted temperature and salinity was improved to some extent in contrast with the previous simulation. Nevertheless, simulation results of new model under rainy and sunny conditions showed the cooling impact of precipitation was slight, through presenting the
surface temperature and salinity different between rainy and sunny days in whole Yatsushiro Sea, precipitation effect still exists although the precipitation effect on sensible fluxes is on lower magnitude in contrast with short wave radiation fluxes. Thus the slight cooling scale shown in comparison results is probably due to the selected observation time under sunny weather, and cooling effect of precipitation on ocean surface temperature may rise up soon after rain due to sufficient sunlight radiation in summer season. Because of the special location of ST.5, river discharge of B-class River was set to different values under the case of rainy and sunny day after calibration, while the discharge of Minamata River may has a slight effect on the simulation of ST.3. The simulation results of flow field indicate ocean water exchange in Yatsushiro Sea mainly happens in the Nagashima channel. Current magnitude in north is larger than south part and flow intensity on surface layer is higher than lower layers.

This study indicates that the effect of precipitation on rainy season’s numerical simulation, especially for the salinity field, shouldn’t be neglected. Precipitation effect is an important factor for ocean surface numerical simulation during rainy season. New thermal radiation boundary conditions provide more accurate simulation for the diurnal and regional temperature change. The precision of simulation will be higher if a more accurate river inflow boundary condition could be supplied. With more precise simulation of the hydrodynamic environment, the simulation of sediment and mercury transport will be optimized.
4. Cohesive Sediment transport module

4.1 Sediment researches in Minamata Bay

Sediment transport has a significant impact on the marine hydraulic and ecosystem environment with a complex process both in water column and seabed (Warner et al. 2008). Mercury dynamic study in Minamata Bay is invariably combined with sediment researches, mercury in dissolved form in water column could be adsorbed with suspended sediment and settled to the seabed with it, similarly, desorption of mercury could happen from suspended sediment. Erosion or deposition of bottom sediment will also result in the mercury cycle between water and seabed. After the outbreak of Minamata Disease, the Minamata Bay Pollution Prevention Project was carried out for dredging and retreatment of contaminated sludge, because large quantities of mercury were found in sedimentary sludge caused by the settlement of mercury contained sediment. This project decreased mercury concentration significantly through removing contaminated sediment. Therefore, sediment transport is an important factor of water quality dynamics in Minamata Bay. In order to figure out transport pattern of mercury, it is necessary to comprehend the sediment transport pattern first.

Sediment researches in Minamata Bay area are inseparable from field observation, sediment samples are in general obtained through different containers and sampling tools for water and bottom sediment, separately. Early in the 70s of last century, Rifardi et al. (1978) have conducted sediment sampling in order to analyze mercury content in seawater and suspended matter, however, this research put emphasis on mercury instead of sediment distribution. In 1996, Rifardi et al. (1998) carried out bottom sediment sampling at 74 stations in the southern part of the Yatsushiro Sea, distribution of bottom sediment in different diameters was analyzed. It is shown that general feature of bottom sediments in this area is characterized by fine to very fine sand and the distribution pattern of fine-grained sediments polluted by mercury was related to the current. Based on the research data, Yano et al. (2014) concluded the long term bottom sediment dynamics through simulation of cumulative deposition and erosion thickness distribution in Minamata Bay by using the numerical model Delft3d. They analyzed the relationship between bottom sediment dynamics and baroclinic structure, which indicated that the freshwater inflow also determined the transport of bottom sediment, same with this numerical model, simulation of layer thickness change of bottom sediment and the tidal flow in Yatsushiro Sea was conducted with finer grids and increased
layer numbers (FATHYA 2017). The Minamata Bay Pollution Prevention Project has changed the bottom topography in Minamata Bay, which may cause water environment variations. Flow current before and after the reclamation project has been analyzed by previous research (LOUSHA 2013), and simulation results showed flow field became weaker in western and southern parts after the project. The narrow strait formed by reclamation in north part caused a strong current which may lead to erosion of sediment. Another simulation regarding to the reclamation project showed velocity magnitude was decreased in bay area after dredging and slowed the sediment transport (FATHYA et al. 2016), and the project prevented the contaminated sediment transport to southern part of Yatsushiro Sea to some extent. Time series analysis of characteristic of the sediment was carried out aiming at the ocean environment change characteristic in Yatsushiro Sea (Masuda et al. 2011), the deterioration of sediment in inner part, near estuary and deep area in center part was found. Monthly water sampling was conducted for measurement of vertical profile of suspended solid grain size distribution in Minamata Bay (Yano et al. 2012), both in water columns and bottom seabed. Significant difference of sediment grain size distribution has been found between upper and lower layers in density stratification, fine grain size sediment took majority in bay area.

As mentioned before, the periodic joint observation has been carried out in Minamata Bay by our research group. In addition to the ocean indexes monitored by integrated electrode sensor, sediment concentration was measured in water columns through a measuring device called LISST-100X (made by Sequoia Co., Ltd.). Sediment diameters from 2.5 to 500 μm could be detected along the vertical water depth at each observation station (Yano and et 2013). Meantime, sediment concentration was also analyzed through the water samples for mercury concentration on water surface, 6 m, 10m, bottom+1 m and bottom +0.1 m by the National Institute for Minamata Disease. In addition to the analysis of sediment concentration in water column, sampling for ocean bed sediments was also conducted and the ore sediment of bottom sediment was sampled by a sediment sampler. However, core sediment sampling was infrequent during the field observation and the collected bottom sediment data was not complete. Due to the time and labor consumption of field monitoring, obtained data are eventually limited and difficult to present detailed regularities of sediment distribution in Minamata Bay and the Yatsushiro Sea. Consequently, numerical simulation of sediment transport is essential for further understanding sediment dynamic pattern. Combined with the field observation data of sediment and the hydrodynamic model, a current induced cohesive sediment transport module was developed under consideration of characteristic of sediment distribution in Minamata Bay.

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4.2 Description of Cohesive Sediment Transport Module

Compared with in-situ observation methods, numerical simulation is more effective for understanding distribution and transportation of sediments. As a main factor affecting the coastal and offshore areas, numerical simulation of sediment transport is getting more and more attention. Numerous open source or commercial numerical modules were developed for simulating the transport of sediment based on different hydrodynamic models, such as ECOMSED, FVCOM, Delft3D and MIKE 21, etc. Most of these numerical models are capable of simulating complete transport processes of different sediment form in estuarine and coastal ocean systems. Nevertheless, sediment dynamics simulation in Minamata area was barely conducted, Yano et al. (2014) and Fathya et al. (2016) simulated the cumulative deposition and erosion thickness of bottom sediment in Minamata Bay with the numerical model Delft3d and summarized the sediment dynamics based on the thickness changes. In order to further present the detailed transport pattern of sediments and transformation processes between sediments and mercury in Minamata Bay, a cohesive sediment transport module based on the new established hydrodynamic module was developed, which consisted of flocculation, deposition and erosion processes. Meantime, field observation data were chosen for the setting of initial conditions and verification of simulation accuracy.

4.2.1 Sediment Categories for Numerical Simulation

Sediment in ocean environment could be categorized into many types due to the large variety of particle diameters, from small size mud and silt to larger sand and gravel, diameter difference may be hundreds or even thousands of times. While numerical simulation could hardly specify to a certain size due to the limitation of computational capacity. For the calculation simplification, numerical simulation for sediment transport typically classifies sediment categories into cohesive sediment, non-cohesive sediment and coarser particles move as bed load transport based on different diameter range (Blumberg 2002), as shown in Table. 3.1. While the defined diameter range may be slightly different in various numerical models. Meanwhile, some sediment modules used in ocean circulation model didn’t consider the bed load transport of coarser particles, which only comprises a small fraction of bed and has a negligible effect in estuarine and ocean system.

Fine-grained sediment which is referred as cohesive sediment has been identified as an important carrier for heavy metal contaminant in coastal and offshore area (Ackermann et al.
Numerical simulation of cohesive sediment transport is essential for understanding dynamic pattern of pollutants, and the change of cohesive sediment concentration has a good response to the ecosystem. The basic processes of cohesive involve flocculation, deposition, erosion, etc. The settling process of cohesive sediment is more complex than non-cohesive sediment because of the existence of flocculation, which could be affected by sediment concentration, salinity, flow turbulence and so on (Floyd et al. 2016). Some researches specifically aimed at cohesive sediment were also conducted in recent decades. Johansen (1998) quantified the main transport processes of cohesive sediment through laboratory experiments. Normant (Normant 2000) proposed a 3D cohesive sediment model to reproduce the typical features of macro-tidal estuaries. Liu et al. (2007) developed a 2D numerical model to simulate cohesive transport in river estuarine. Lumborg and Pejrup (2005) analyzed net transport of cohesive sediment around tidal area based on MIKE 21 MT. Chao et al. (2008) developed a current and wind waves induced cohesive sediment transport model for shallow lake area. Wang et al. (2018) combined the cohesive sediment transport with adjoint data assimilation method. Non-cohesive sediment and coarser bedload particles comprise of a wide range of particle sizes, especially in the ocean bed, which means the sampling and survey analysis about ocean bed are important for simulation. Except for the transport model in water column, a bed model is also needed to reflect the complicated processes in bottom ocean layer and ocean bed for larger particles. Ocean bed will be also divided into some layers similar with the water column (Blumberg 2002; Warner et al. 2008), different bed layers show different characteristics like layer thickness, mean grain diameter, and density, mean shear stresses, or even the consolidation process.

Table 4.1 Primary sediment categories during numerical simulation

<table>
<thead>
<tr>
<th>Primary Categories of Sediment</th>
<th>Particle Diameters (μm)</th>
<th>Transport Processes</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cohesive Sediment</td>
<td>&lt; 75</td>
<td>Vertical deposition (flocculation) and resuspension</td>
</tr>
<tr>
<td>Non-cohesive Sediment</td>
<td>75 - 500</td>
<td>Vertical deposition and resuspension</td>
</tr>
<tr>
<td>Bed Load Coarse particle</td>
<td>&gt; 500</td>
<td>Horizontally transport (negligible in coastal area)</td>
</tr>
</tbody>
</table>
The reason for establishing a sediment transport module is to provide the simulation environment for mercury. A partial form of mercury dissolved in water column with the resuspension and transport of contaminated sediment. Because heavy metals could be easily absorbed by cohesive sediment and transported with it, many previous researches of mercury were concentrated in fine grain size sediment transport (Abi-Ghanem et al. 2011; Barghigiani et al. 1996; Carroll et al. 2000), less than 63μm (Miller et al. 1999) or even 20μm (Širca et al. 1999), which were all belong to the cohesive sediment category. Sediment sampling for analyzing suspended solid grain size distribution in Minamata Bay showed the average proportion of cohesive sediment was about up to seventy percent (Yano et al. 2012). Interaction between mercury and sediment in water column is mainly about absorption and desorption, higher settling velocity of large particle size will make non-cohesive sediment or larger particles sink faster which causes little reaction time. Meantime, due to the limitation of data collection, it is difficult to present the complicated changing processes of larger particles in ocean bed, so the transport of non-cohesive sediment and coarser particles were not taken into account in this study. In consideration of low magnitude measured sediment concentration, relative small flow velocity and wind speed in the bay area, a current induced cohesive sediment transport module coupled with the hydrodynamic module was developed under consideration of the characteristic of sediment distribution in Minamata Bay.

4.2.2 Establishment of Cohesive Sediment Transport Module

Governing equation for the calculation of cohesive sediment transport in most models are basically consistent. Based on the algorithmic features and code structures of POM, the three-dimensional governing equation for cohesive sediment transport in water column consists of advection and diffusion processes is presented as:

\[
\frac{\partial C}{\partial t} + \frac{\partial CU}{\partial x} + \frac{\partial CV}{\partial y} + \frac{\partial (\omega - \omega_s)C}{\partial z} = \frac{\partial}{\partial x}[A_H \frac{\partial C}{\partial x}] + \frac{\partial}{\partial y}[A_H \frac{\partial C}{\partial y}] + \frac{\partial}{\partial z}[K_H \frac{\partial C}{\partial z}] 
\]

(4.1)

where \( U, V, \omega \) represent the current velocity components along horizontal and vertical directions; \( \omega_s \) is settling velocity of cohesive sediment and \( C \) is cohesive sediment concentration; \( A_H \) and \( K_H \) are the horizontal diffusivity and vertical eddy diffusivity coefficients, respectively, which are the same with the temperature and salinity equations.

An important component of cohesive sediment dynamics is the settling process leading to the deposition of sediment. The settling process of cohesive sediment is complicated because of the existence of flocculation, which could be affected by sediment concentration, salinity, and
many other factors. The flocculation process is dynamic and complex, and suspended fine sediment particles are aggregated to produce flocs in water column through this process, meantime, breakup processes also happen. There is still no a uniform formula for the calculation of flocculation, most equations are empirical analysis based on numerous data collected from laboratory (Floyd et al. 2016). As a result, settling velocity formulas for cohesive adopted in different numerical models show great discrepancies. ECOMSED considers effect of concentration and water column shear stress and Delft3D considers effect of salinity (Qinghua 2006), while MIKE21 considers concentration and flocculation constant (Warren and Bach 1992). The formula of settling velocity adopted in this study for cohesive sediment considers the influence of sediment concentration and water column shear stress, which has the same format with the formula in ECOMSED:

$$\omega_s = a (CG)^b$$ (4.2)

$$G = \rho K_u \left[ \left( \frac{\partial U}{\partial Z} \right)^2 + \left( \frac{\partial V}{\partial Z} \right)^2 \right]^{1/2}$$ (4.3)

where $G$ is water column shear stress calculated by the hydrodynamic model and $\rho$ is water density; $C$ is cohesive sediment concentration; $a=0.0026$ and $b=0.28$ are experimental parameters obtained from experiment conclusion of Burban (1990) for seawater, which was based on regression analysis from extensive experimental data of sediment concentration, fluid

![Settling velocity comparison with different parameters](image)

**Fig. 4.1** Settling velocity comparison with different parameters
shear stress and particle diameters. Compared with the parameter used in ECOMSED, the velocity value with new parameters has a slight increase as shown in Fig. 4.1, and unit displayed is meters per day.

Except for the settling velocity for the sediment movement in water column, the boundary conditions are also important to solve the three-dimensional cohesive sediment transport equation. Vertical sediment input at free surface and lateral sediment input for estuary are not considered and set to zero flux. At the bottom sediment-water interface, bottom boundary conditions for the mass transport equation are shown in Eq. 4.4. The well-known Partheniades-Krone formulas were adopted to calculate the erosion and deposition fluxes (Partheniades 1965), as shown from Eq. 4.5 to Eq. 4.7, while fraction of bottom cohesive sediment was added into the erosion flux equation.

\[
K_H \frac{\partial C}{\partial z} = E_b - D_b, \quad z \to H
\]  

\[
E_b = \begin{cases} 
0, & \tau < \tau_{ce} \\
E_o f_c \left( \frac{\tau}{\tau_{ce}} - 1 \right) (1 - P_o), & \tau > \tau_{ce}
\end{cases}
\]  

\[
D_b = \omega_b C P
\]

\[
P = \begin{cases} 
(l - \frac{\tau}{\tau_{cd}}), & \tau < \tau_{cd} \\
0, & \tau \geq \tau_{cd}
\end{cases}
\]

where \( E_b \) and \( D_b \) are the erosion and deposition fluxes at bottom, respectively. \( \tau \) is bed shear stress calculated by the hydrodynamic model. \( E_o \) is erosion parameter, \( f_c \) is fraction of cohesive sediment in bottom, \( P_o \) is bottom porosity. \( \tau_{ce} \) is critical erosion shear stress and \( \tau_{cd} \) is critical deposition shear stress.

Some sediment models only use one critical shear stress to judge erosion or deposition, which means erosion or deposition happens all the time. Here this process is separated into three parts, erosion occurs when bed shear stress is greater than critical erosion shear stress and deposition occurs when bed shear stress is less than critical deposition shear stress, when shear stress is less than critical erosion shear stress and larger than critical deposition shear stress, no sediment exchange at bottom, as shown in Fig.3.2. Some complete cohesive sediment transport models also have modules for bed dynamic to simulate the effects of sequential deposition and erosion realistically. Sediment bed is represented as discrete layers, and different bed layers show different characteristics like porosity, layer thickness, shear stresses and some other.
Fig. 4.2 Erosion and deposition processes at the bottom sediment-water interface

Factors (Hayter and Mehta 1986). Due to limitation of bed data collection, ocean bed was simplified to one layer in this study and assumed enough fine-grained sediment for erosion. To avoid unbalance sediment mass in water columns caused by excessive erosion, an active layer shown in Eq. 4.8 was adopted to limit the erosion thickness, the depth of the active layer is assumed to be proportional to excess shear stress (Harris and Wiberg 2001). The change of bed thickness was calculated in Eq. 4.9, changing thickness value couldn’t exceed the active layer when erosion happened.

\[
Z_a = \max \left[ 8(\tau - \tau_e), 0 \right] + 6D_{50} \tag{4.8}
\]

\[
\frac{\partial Z_b}{\partial t} = \frac{1}{\rho_b} (D_b - E_b) \tag{4.9}
\]

where \(Z_a\) and \(Z_b\) represent active layer and bed layer thickness, respectively. \(D_{50}\) is the median grain size, set to an average value of 30 μm for cohesive sediment. \(\rho_b\) is dry bed density.

4.2.3 Module Calibration and Computational Setting

4.2.3.1 Sensitivity Tests for Cohesive Sediment Transport Module

Both resuspension and deposition mechanisms depend upon the bottom shear stress induced at the sediment-water interface. Erosion and deposition processes were controlled by the comparison of bed shear stress and critical shear stresses when they are performed through
programmed codes. Model calibration is primarily focused on the bed shear stress and critical shear stresses before the start of simulation, which have a direct and significant impact on simulation results. Values of critical shear stress were in a wide range depending on different research environments (Chao et al. 2008). Without considering wave model in this case, the calculated bed shear stress was in a low level compared with wave combined model. Sensitivity tests are necessary to determine the value of cortical erosion and deposition shear stresses during the model calibration stage, and also other factors that could affect the change of bottom shear stress.

Analytical sediment data during field observation has much missing and discontinuous information. In order to obtain a relatively complete comparison with simulation results, total simulation duration was selected from July 6\textsuperscript{th} to July 21\textsuperscript{st}, 2015, centralized field observation was carried out 5 times and sediment concentration data at three stations (ST.1, ST.2, and ST.3) are available. Measured data of ST.1 on bottom layer was selected for the comparison during the stage of sensitivity tests. In order to maintain consistency with the mercury simulation, wind driving conditions were changed to the measured data of Minamata meteorological observation station, correspondingly, flow field in the whole simulation domain has been changed. Meantime, sensitivity tests for critical shear stresses have incorporated the freshwater inflows and all the sensitivity tests were conducted again, some parameters have also been adjusted for the new simulation environment. Consequently, simulation results of sediment transport are different from the study results in the previously published study. However, the study conclusions are generally consistent.

Fig. 4.3(a) shows the sensitivity test results of critical deposition shear stress, and simulated time serious of sediment concentrations at ST.1 under different scale of critical deposition shear stresses are presented. Value of critical erosion shear stress was fixed to 0.02 Pa, and all simulations were carried out with same initial, boundary conditions and external forcing setting under the consideration of river and wind input. Simulation was conducted five times with different critical shear deposition stress range from 0.005 Pa to 0.018 Pa. It is indicated that smaller deposition magnitude led to concentration increase at bottom layer. Through the comparison of simulated concentration and measurements, the critical shear stress for deposition value was set to 0.005 Pa which is displayed with a bold straight line in the figure. In addition to the deviation shown in second observation data, agreement of simulated results under corresponding critical deposition shear stress is acceptable. Fig. 4.3(b) presents the sensitivity test results of critical erosion shear stress, value of critical deposition shear stress was fixed to 0.005 Pa during five simulations. It is evident that the flux magnitude of erosion is
(a) Comparison of Simulated sediment concentration on bottom layer under different critical deposition shear stresses and measured data at ST.1

(b) Comparison of Simulated sediment concentration on bottom layer under different critical erosion shear stresses and measured data at ST.1

Fig. 4.3 Simulated time series of sediment concentration by sensitivity tests
larger than deposition and the effect of scale change is significant when larger than 0.02 Pa. Numerical divergence appears when the critical shear stress for erosion is large due to the low flow velocity and wind speed in bay area. As a result, the critical erosion shear stress value is set to 0.02 Pa as shown in bold straight line.

Wind forcing and river discharge are also important factors related to the bottom shear stress through the impact on current velocity. The day averaged wind data in July, 2015 is presented in Fig. 4.4 with day averaged wind speed and most prevailing direction of every day at the Minamata meteorological observation station. Although wind speed during research time was in a low level, with an average value about 1.3 m/s, the indirect wind effect to bottom shear stress was still analyzed, as shown in Fig. 4.5. Sensitivity test of wind forcing was carried out with the reference critical shear stresses under no consideration of river discharge. Compared

![Fig. 4.4 Wind velocity and direction of Minamata meteorological observation station](image1)

![Fig. 4.5 Wind effect on bottom shear stress at ST.1](image2)
with the simulation results without wind, overall influence on bottom shear stress at ST.1 is implicit, however, the effect is visible at some certain time, especially around July 13th. Meantime, through the comparison with the previous simulation, the significant deviation of second observation data with simulation results shown in Fig. 4.3(a) was probably caused by the impact of wind.

Sensitivity test for river discharge was presented in Fig. 4.6. Three rivers exist in the simulation domain and two of them are B-class rivers without discharge records, therefore B-class river’s discharge was set to a constant value of 80 m$^3$ s$^{-1}$ which was little higher than the simulation of precipitation module. Three pairs of contrast were conducted under the consideration of wind force to present obvious comparisons. It is indicated that the Kuma River which locates in the north part of the Yatsushiro Sea has almost no effect on the bottom shear stress at ST.1 because of long distance. Thus, the simulation result with Kuma River was treated as the circumstance under no river discharge for comparing the other two rivers’ effect. Compared with the Komenotsu River, effect of the Minamata River on bottom shear stress value is also in a low level, because most of flow from north will bypass the narrow north bay mouth during ebb tide, while transport to north during flood tide. Discharge from Komenotsu

![Fig. 4.6 Sensitivity test of river discharge effect at ST.1](image-url)
River effect coupling with wind is relatively remarkable, while previous research has revealed the river discharge effect without wind input is only visible during the late period of simulation, which indicates the coupling effect of wind and discharge of Komenotsu River is significant. However, the influence of Komenotsu River is still larger than other rivers, it can be deduced that discharge of Komenotsu River combined with wind impact changed the flow field around Minamata Bay from west bay mouth and inflow of fresh water varied ocean density, density flow was formed and caused variation of bottom shear stress. Due to the effect of river discharge is not so pronounced on bottom layers, and rainfall impact is concentrated on surface layers where sediment concentrations are in a lower magnitude with small changing range as shown in Fig. 4.7, the sensitivity tests of precipitation are not conducted.

4.2.3.2 Computational Setting

Time interval of cohesive transport model was set to 5 seconds which has the same value with internal mode of hydrodynamic model. Total simulation duration was from July 6th to July 21st, 2015 with the same horizontal grid arrangement. The prediction method mentioned in the hydrodynamic model was applied to obtain the harmonic constants of four tidal constituents (M2, S2, K1, O1) on July 6th, 2015, and the predicted harmonic constants were interpolated on two open boundaries using inverse distance weighted method as tidal forcing for surface elevation. Critical deposition and erosion shear stresses were set to 0.005 Pa and 0.02 Pa after the sensitivity tests, separately. Real-time wind velocity, Kuma River discharge, and factors for the calculation of fresh water and thermal radiation boundary conditions were interpolated into each internal time step by a linear interpolation method. Discharge of two B-class rivers
was set to a constant value of 80 m³ s⁻¹. Six times’ field observations on July 6th, 8th, 10th, 12th, 14th and 18th have been conducted during this period, observation data on July 6th was treated as initial conditions and interpolated into the whole simulation domain, and following data were selected for comparing the simulated results.

As shown in Fig. 4.7, measured data of ST.1 shows great distinction between bottom and upper layers. Sediment concentration on surface and middle layers are in a low and stable level, while concentration values on bottom layer are quite high, even three times larger than upper layers at some certain time. During simulation, numerical effect from high concentration on bottom layer to upper layers is evident with the previous 10 layers’ distribution in vertical σ-coordinate system, as displayed in Fig. 4.8(a). Numerical calculation positions in source code algorithm are expressed with black circle, the actual simulated bottom data is on the ninth layer and only three layers’ intervals exist between middle and bottom layers. Closer layer distance and larger concentration difference may cause abnormal higher simulation results on surface and middle layers, and the influence is more significant during the simulation of particulate mercury which was introduced in the next chapter. Therefore, a new vertical σ-coordinate with refined layer distribution was established to reduce the influences as shown in Fig. 4.8(b). Vertical coordinate was divided into 20 layers and 6 logarithmic layers on bottom with higher resolution. The detailed layer distribution and intervals of new and previous σ-coordinate systems are shown in Table. 4.2. The increase of layer number and addition of logarithmic layers effectively reduced the numerical influence caused by large concentration difference.

4.3 Simulation Results of Cohesive Sediment Transport Module

4.3.1 Elevation Verification

Similar to the precipitation module, module verification is presented between the simulated tidal elevation and measurements. Japan Meteorological Agency predicted elevation (minus datum value) and measured data of the Kuma River from Japan Water Information System were adopted for elevation verification at ST.3 and the Kuma River. As shown in Fig. 4.9, in addition to slight deviations at the later stage of simulation, agreement of simulated results with measurements is generally acceptable. The predicted harmonic constants based on historical measured data of four tidal constituents for simulation are basically consistent with actual data, which could be applied for the tidal forcing conditions at open boundaries.
Fig. 4.8 Improvement of vertical layer distribution in $\sigma$-coordinate system

### Table 4.2 Layer distribution of previous and new $\sigma$-coordinate systems

<p>| Layer number | Layer distribution ($\sigma$) | Layer interval ($|\Delta\sigma|$) |
|--------------|------------------------------|----------------------------------|
|              | Previous                      | New (Approximately)              | Previous | New (Approximately) |
| 1            | 0                             | 0                                | 0.0625   | 0.03846             |
| 2            | -0.0625                       | -0.03846                         | 0.125    | 0.03847             |</p>
<table>
<thead>
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<tr>
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<td>0.125</td>
<td>0.07692</td>
</tr>
<tr>
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<td>-0.15385</td>
<td>0.125</td>
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<tr>
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<td>-0.23077</td>
<td>0.125</td>
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</tr>
<tr>
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<td>-0.30769</td>
<td>0.125</td>
<td>0.07692</td>
</tr>
<tr>
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<tr>
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<td>-0.53846</td>
<td>0.125</td>
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</tr>
<tr>
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</tr>
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<td>14</td>
<td></td>
<td>-0.92308</td>
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<td>0.03846</td>
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<tr>
<td>15</td>
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<tr>
<td>20</td>
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<td></td>
<td>-1</td>
<td>0</td>
</tr>
</tbody>
</table>

**Fig. 4.9 Verification of elevation at station 3 and Kuma River**
4.3.2 Simulation Results

Sediment concentration at bottom layer is more sensible to the erosion and deposition progresses and other external forcing input due to the high concentration values. Both measured and simulated results show concentration change of middle or surface layer is not obvious, therefore bottom layer’s comparison data of three stations are presented and analyzed firstly. Simulated cohesive sediment concentration and corresponding bottom shear stress are shown in Fig. 4.10.

Simulated results of ST.1 and ST.2 at Fig. 4.10 (a) and (b) have large range variation during the later period of simulation, which was caused by the intense change of bottom shear stress shown in the upper part of each figure, correspondingly, erosion and deposition happened frequently. Due to the high magnitude of erosion fluxes, erosion process will cause a sharp increase of simulated concentration, while the effect of deposition generally needs a relatively long period to produce concentration decrease. Relatively close change tendency of simulated bottom shear stresses at ST.1 and ST.2 may indicate that erosion and deposition level of west and middle area in Minamata Bay is in similar order of magnitude based on the station location shown in Fig. 3.1. Agreements of simulation and measurements at two stations are acceptable except the comparison on July 10th at ST.1, which was also appeared during the sensitivity tests of critical shear stress. In the previous study with different wind velocity data in other weather station, comparison result at this point was in a good agreement. It could be presumed that the sharp increase of concentration was caused by wind forcing, while the application of new wind condition interpolated to the whole simulation domain was not able to reproduce this change.

Contrast with ST.1 and ST.2, simulated concentration at ST.3 shows great difference and maintains in a low value all along, measured data of ST.3 also reveals that concentrations are about half value compared with other two stations. ST.3 which locates in the north part of Minamata Bay is closer to the coastal shore and water depth is shallower. Special geographical location leads to lower magnitude of bottom shear stresses which is also displayed in the time series of simulated bottom shear stresses, hence large scale erosion seldom occurs. The change of sediment concentration mainly depends on the advection and diffusion processes with current in water column.

Fig. 4.11 shows the comparison of simulated time series results at surface and middle layers with measurements at three stations. In contrast with concentration on bottom layer, concentration change of middle and surface layer is not obvious, and concentration values generally keep in a stable level both in simulation and measurements. Sediment concentrations
Fig. 4.10 Comparison of measured and simulated bottom layer’s data

(a) Surface and middle layers at ST.1

(c) ST.3
(b) Surface and middle layers at ST.2

(c) Surface and middle layers at ST.3

Fig. 4.11 Comparison of measured and simulated data of surface and middle layers
on both surface and middle layers were in a range of 5 to 10 mg/L, and the impact of erosion and deposition processes on upper layers is not less visible than bottom layer. Slight fluctuation exists on surface layers compared with concentration on middle layers. Simulation of three stations on upper layers obtained from the numerical model show generally good agreements with measurements.

In order to present the detailed flow dynamics inside bay, a non-uniform grid with high resolution in bay area was adopted, correspondingly, with smaller time interval and longer running time. Total resolution of the non-uniform grid is $153 \times 164$ and the comparison of two grid arrangement types is shown in Fig. 4.12. It is indicated that not only the Minamata Bay but the outward areas in horizontal and vertical direction are refined, which could cause the increase of calculation time with smaller time steps. As shown in Table 4.3, the CFL condition was used to keep the computational stability during numerical simulation and time interval of non-uniform grid was decreased to one-third values. Correspondingly, total code running time increases dramatically, due to the excessive longer running time, simulation with non-uniform grid was only conducted for presenting the detailed dynamics of bay area.

Fig. 4.13 (a), (b) and (c) have shown the simulated cohesive sediment concentration and current field after 5 days, 10 days and 15 days on bottom layer to further perform the bottom sediment concentration dynamics. Due to trends of concentration change on surface and middle layers are not pronounced, only simulated concentration fields on bottom layer are presented which have sensible response to the bottom boundary conditions. Meantime, the
### Table 4.3 Grid setting of uniform and non-uniform grids

<table>
<thead>
<tr>
<th></th>
<th>Uniform Grid</th>
<th>Non-uniform Grid</th>
</tr>
</thead>
<tbody>
<tr>
<td>Grid number</td>
<td>119×119</td>
<td>153×164</td>
</tr>
<tr>
<td>Time step (External)/(Internal)</td>
<td>1s/5s</td>
<td>0.3s/1.5s</td>
</tr>
<tr>
<td>Total running time under different grids</td>
<td>about 4 hours (Hydrodynamic)</td>
<td>about 35 hours (Hydrodynamic)</td>
</tr>
<tr>
<td></td>
<td>about 18 hours (Sediment )</td>
<td>about 130 hours (Sediment )</td>
</tr>
</tbody>
</table>

Fig. 4.13 Simulated bottom layer concentration and current field after: (a) 5 days, (b) 10 days, (c) 15 days
interpolated initial conditions with inverse distance weighted method and boundary conditions in the outside area of Minamata Bay may exist deviations with actual data, only simulated results of bay area was treated as reference for displaying the changing intensity of erosion and deposition. It is evident that flow field on bottom layer is in a relatively low magnitude compared with the surface flow field as shown in Fig. 3.14 during the simulation of hydrodynamic model. Simulated result after 5 days shown in Fig. 4.13(a) is during flood tide and flow direction is from south to north. Current velocity magnitudes of ST.1 and ST.2 are very close, which lead to roughly similar erosion or deposition levels and sediment concentration. Although narrow strait on northern bay mouth causes high velocity and sediment concentration when the current flows out of bay, velocity around ST.3 is still in a low level and the location with shallower water depth results in a more stable concentration change. Simulated result after 10 days shown in Fig. 4.13(b) is during an ebb tide, fast current velocities outside the bay area cause frequent erosion and sediment concentration is in a high level in the north part. It is indicated that fast current bypasses the north strait and flows to west, which ensures the stability of sediment concentration of northern bay area, and concentration at ST.3 is not influenced by the fast current blocked by the narrow strait and special shoreline distribution. However, a high concentration area appears in the west part of bay. Ebb tide causes confluence of bay and outside ocean currents and high bottom shear stresses appear in this area, large amounts of sediment from ocean bed are eroded into water column and result in the high concentration level. Nevertheless, location of ST.1 is not in the affected area and the sediment concentration value basically keeps consistent with ST.2. Simulated result after 15 days shown in Fig. 4.13(c) is during slack water after flood tide and flow directions are diverse in bay area. Flow influxes from west and south change the flow direction in southwest bay area, and high bottom shear stresses lead to high sediment concentration. However, sediment concentrations at three observation stations maintain in a low level during this period.

4.4 Chapter Summary

Cohesive sediment has been identified as an important carrier for heavy metal transport in coastal and offshore area. To further understand the distribution and transport of mercury contaminant in Minamata Bay, a sediment transport is necessary to reproduce the transformation processes between different mercury forms and supply basic simulation environment for mercury research. Consequently, a current induced cohesive sediment transport model was developed, considering the process of resuspension, deposition, settling,
etc. Hydrodynamic environment was provided by the POM with a new precipitation module.

Sediment researches in Minamata Bay were discussed first and the current induced cohesive sediment transport module was presented based on the regularities of sediment distribution and flow characteristics. Sensitivity tests were carried out for critical erosion and deposition shear stresses, wind and river discharge during the module calibration stage. High-value magnitude of critical erosion shear stress could cause more significant concentration change than the critical deposition shear stress. Discharge of Kuma and Minamata River had a slight impact on the bottom shear stresses, while the density flow caused by the coupling effect of Komenotsu River discharge and wind forcing had a relative visible impact on sediment transport in Minamata Bay. With the calibrated parameters obtained from the sensitivity tests, the current induced cohesive sediment transport simulation was carried out with a newly established vertical coordinate system. Simulation results on bottom ocean layers showed acceptable agreement with measurements at three observation stations, and concentration change trends basically kept consistent with the variation of bottom shear stresses. Simulated sediment concentrations on upper layers were in a good agreement with measured data due to the slight concentration fluctuation. Relationship between simulated flow field and concentration field of bottom cohesive sediment was also discussed with higher resolution grids. Sediment concentrations outside Minamata Bay were in a high magnitude, high current velocity caused more sediment eroded from ocean bed. Due to the special coast shape and narrow strait, most strong current bypassed the north entrance during ebb tide and ensured bay water in a stable level. While flow magnitude could be amplified when outflow on northern narrow strait happened during flood tide. Mass exchange of sediment may exist in different bay mouth under different tidal conditions. With the verification of simulated sediment concentration and measurements, the current induced cohesive sediment transport module combined with POM was generally able to present basic simulation environment for the simulation of mercury dynamics.
5. Numerical Simulation of Residual Mercury Variation

The speciation and transformation processes of different chemical mercury forms in aquatic environment are complicated. Major mercury species for research are known as elemental mercury, inorganic mercury, and methyl-mercury (Lessard 2012). For most numerical mercury cycling models, all inorganic mercury species were classified into a single species known as divalent mercury \((\text{Hg}^{II})\) (Massoudieh et al. 2010). The numerical simulation of residual mercury variation in Minamata Bay presented in this study also categorized three mercury species: element mercury \((\text{Hg}0)\), divalent mercury \((\text{Hg}^{II})\) and methyl-mercury \((\text{MethHg})\). Each species had two physical forms: dissolved form in ocean water \((\text{Diss})\) and particulate form transport with sediment particles \((\text{Part})\). Three kinds of mercury: dissolved total mercury \((\text{Diss-THg})\), dissolved methyl-mercury \((\text{Diss-MethHg})\) and particulate total mercury \((\text{Part-THg})\) were obtained during in-situ measurements and used to verify simulation. The primary category of mercury in this study is shown in Table 5.1. To reproduce the residual mercury dynamics in Minamata Bay, an integrated three-dimensional numerical model was developed and numerical simulation was conducted with total eight kinds of mercury.

5.1 In-Situ Measurement of Mercury Behavior in Minamata Bay

Ever since the Minamata Disease caused by polluted water discharge with mercury contamination was officially acknowledged, the dynamic behavior of residual mercury has been the research emphasis in Minamata area. Although the Minamata Bay Pollution Prevention Project significantly decreased the mercury concentration, the residual mercury was

<table>
<thead>
<tr>
<th>Mercury form</th>
<th>Mercury species</th>
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<tbody>
<tr>
<td>Dissolved mercury (Diss-THg)</td>
<td>Element mercury (Diss-Hg0, Part-Hg0)</td>
</tr>
<tr>
<td>Particulate mercury(Part-Thg)</td>
<td>Divalent mercury (Diss-HgII, Part-HgII)</td>
</tr>
<tr>
<td></td>
<td>Methyl-mercury (Diss-MethHg, Part-MethHg)</td>
</tr>
</tbody>
</table>
still in a high level compared with the background concentration value due to the long term discharge of mercury byproducts. The speciation and transformation processes of different chemical mercury forms in aquatic environment are complicated. Researches about the distribution and transport of mercury in Minamata Bay and the Yatsushiro Sea mainly focused on in-situ measurement based on field monitoring and observation. Early before the remediation project, Kumagai et al. (1978) conducted field sampling to analyze the horizontal and vertical distribution of mercury in Minamata Bay and Yatsushiro Sea, and the research showed particulate mercury was mainly from the bottom sediment and had transported from bay area to the Yatsushiro Sea with dispersing in seawater. Almost twenty years later, Tomiyasu et al. (2000) carried out another investigation of mercury distribution in sediment and suggested that the deposited bottom sediment were still under transportation. Their following field observation has indicated that MethHg concentration in sediment of Fukuro Bay was higher than Minamata Bay (Tomiyasu et al. 2006) and MethHg may be the predominant mercury species released from the sediment into water column (Tomiyasu et al. 2008). Matsuyama et al. (2010) concluded the average concentration of different mercury forms and suspended solids through a 2-year period observation, and suggested that bottom sediment was not the only source of MethHg which may be also produced from HgII in water column. Yano et al. (2013) evaluated annual transport value of Part-THg and Diss-MethHg from Minamata Bay to the Yatsushiro Sea by highly-frequent water sampling, and investigation results indicated the source of mercury in bottom and surface layers may be different. Matsuyama et al. (2014) re-evaluated the mercury distribution pattern with detailed field surveys after 25 years since the dredging project. Balogh et al. (2015) used stable Hg isotope to track mercury fate in fish and bottom sediment of Minamata Bay, and suggested the MethHg may be produced in bottom sediment pore-water or sediment-water interface and entered the marine food chain.

Monthly observation for mercury concentration has been conducted by the joint research group in Minamata Bay since 2006. In-Situ Measurement of Mercury in Minamata Bay was carried out on water surface, 6 m, 10m, bottom+1 m, and bottom +0.1 m during the periodic joint observation. Ocean water samples in deep depth were collected through a pump and a long polyvinyl chloride pipe. On the bottom +0.1 m position, a water sampler device made of acrylic boards was used to prevent sediment resuspension caused by flow disturbance during pumping (Yano and et 2013). The boards were sent from ocean surface to bottom, sampling started after the sediment concentration was confirmed to be stable. The seawater samples were pumped directly into amber glass bottles to avoid exposure to sunlight and these bottles were cleaned by a strong acid mixture and rinsed with distilled water for at least five times before sampling.
Water samples for analysis of mercury and sediment concentrations were kept in these glass bottles and conserved with cooling bags on the boat. The collected samples would be sent to the laboratory within 20 minutes and filtered with a membrane filter. The particulate mercury concentrations were measured by the method proposed by Akagi and Nishimura (1991), and modified by Akagi et al. (1995). The accuracy and precision of the method have been repeatedly verified by laboratory calibration exercises (Malm et al. 1995). The dissolved mercury concentrations were measured by the method of Akagi and Nishimura (1991) and modified by Matsuyama et al. (2010). The relevant analysis and measurement of mercury concentrations have been presented by Matsuyama et al. (2010), Akito et al. (2014) and Balogh et al. (2015). After analysis from the National Institute for Minamata Disease, concentrations of three mercury kinds: dissolved total mercury (Diss-THg), dissolved methyl-mercury (Diss-MethHg) and particulate total mercury (Part-THg) were obtained at each observation position. Simulation results of the mercury transport model were compared with these three kinds of measured mercury concentration. Due to the lack of continuous data of Part-THg, measured data of Diss-THg and Diss-MethHg concentration were presented with annual averaged value on three layers at three stations from 2006 to 2017. As shown in Fig. 5.1, the annual averaged concentration change of Diss-THg is not obvious during ten years, and concentration may have a slight increase in recent years. All three stations’ data indicate the averaged Diss-THg concentrations on middle layer are lower than surface and bottom layers. Fig. 5.2 presents the concentration change trends of Diss-MethHg, an overall downward tendency is displayed. In consideration of the slight raise of Diss-THg, the decrease of Diss-MethHg may indicate the concentration increase of other two kinds of dissolved mercury.
Fig. 5.1 Annual mean value of measured Diss-THg concentration on three layers of three stations from 2006 to 2017.
5.2 Integrated Numerical Model for Mercury Transport

Numerical simulation for mercury transport was barely presented for coastal and marine systems due to the completed cycling processes and unavailable parameters. Most of the existing mercury cycling models belong to steady-state mass balance models built for freshwater systems, which are derived from the understanding of mercury transformation processes in different environment. The steady-state mass balance models separate cycling processes into different segments and calculate fluxes of each process respectively based on massive statistical data and parameters. In Minamata Bay, the non-steady state three-dimensional model PCFLOW3D (Rajar et al. 2004a) has been used to simulate the mercury exchange, and Lou Sha (LOUSHA 2013) used a pollutant transport model to simulate the concentration field in bay area, nevertheless, the details of mercury distribution and transport processes were not presented during these simulations. To further understand the mercury transport pattern in Minamata Bay, an integrated three-dimensional numerical model for mercury dynamics simulation was developed and the mercury transport module was optimized and promoted based on the previous module framework (LOUSHA 2013).

5.2.1 Description of the Integrated Numerical Model for Mercury Transport

The integrated model of mercury transportation consists of three modules: (a) Hydrodynamic module is based on the Princeton Ocean Model for supplying basic hydrodynamic environment. In order to adapt to the drippy climate during summer simulation, a precipitation module was built and coupled with the POM as presented in chapter 3, also with a new thermal radiation boundary which could change temporally and spatially. Simulated
temperature and salinity field by the upgraded model showed good agreement with measured data during plum rain season. (b) Cohesive sediment transport module included flocculation, deposition and erosion processes and simulated results showed a satisfactory agreement as shown in chapter 4. The current induced cohesive sediment transport module was established based on the characteristic of sediment distribution, wind and current conditions in Minamata area. Critical bottom shear stresses were used to calculate the resuspension and deposition processes at the water and seabed interface. (c) Mercury transport module could describe the diffusion and advection process with ocean water, adsorption and desorption process with sediment of different mercury forms, including oxidation, reduction, and other chemical reaction processes among different mercury species.

The integrated mercury cycling model takes into account mercury exchange with atmosphere caused by surface deposition and evasion, mercury exchange with bottom pore water, and mercury exchange with bottom sediment caused by sediment resuspension and deposition. Fig. 5.3 shows the basic transformation processes of three mercury species in two forms during numerical simulation: the Dissolved total mercury (Diss-THg) consists of dissolved element mercury (Diss-Hg0), dissolved divalent mercury (Diss-HgII) and dissolved

Fig. 5.3 Major transformation processes of mercury during simulation
methyl-mercury (Diss-MethHg). Dissolved mercury transformation in water column includes oxidation and reduction between Hg0 and HgII, methylation and demethylation between HgII and MethHg. Particulate total mercury (Part-THg) contains particulate element mercury (Part-Hg0), particulate divalent mercury (Part-HgII), and particulate methyl-mercury (Part-MethHg). Through adsorption and desorption processes with suspended sediment, transformation between dissolved and particulate mercury happens.

Governing equations for two mercury forms, dissolved mercury and particulate mercury, are described as:

\[
\frac{\partial C_{dHg(i)}}{\partial t} + \frac{\partial C_{dHg(i)} U}{\partial x} + \frac{\partial C_{dHg(i)} V}{\partial y} + \frac{\partial C_{dHg(i)} \omega}{\partial \sigma} = \frac{\partial}{\partial x} \left[ H_{dHg} \frac{\partial C_{dHg(i)}}{\partial x} \right] \\
+ \frac{\partial}{\partial y} \left[ H_{dHg} \frac{\partial C_{dHg(i)}}{\partial y} \right] + \frac{\partial}{\partial \sigma} \left[ K_{dHg} \frac{\partial C_{dHg(i)}}{\partial \sigma} \right] + \text{Flux}_{dHg(i)}
\]

\[
\frac{\partial C_{PHg(i)}}{\partial t} + \frac{\partial C_{PHg(i)} U}{\partial x} + \frac{\partial C_{PHg(i)} V}{\partial y} + \frac{\partial C_{PHg(i)} \omega}{\partial \sigma} = \frac{\partial}{\partial x} \left[ H_{PHg} \frac{\partial C_{PHg(i)}}{\partial x} \right] \\
+ \frac{\partial}{\partial y} \left[ H_{PHg} \frac{\partial C_{PHg(i)}}{\partial y} \right] + \frac{\partial}{\partial \sigma} \left[ K_{PHg} \frac{\partial C_{PHg(i)}}{\partial \sigma} \right] + \text{Flux}_{PHg(i)}
\]

where index \(i\) represents three mercury species: Hg0, HgII, and MethHg. \(C_{dHg(i)}\) is dissolved mercury concentration in water column; \(C_{PHg(i)}\) is particulate mercury concentration in sediment particles which is dimensionless dry weight; \(C_{as}\) is cohesive sediment concentration and \(\omega_{s}\) is settling velocity of cohesive sediment calculated in the sediment transport module; \(D\) is water depth, sum of average depth \(H\) and free surface elevation; \(U, V, \omega\) are the current velocity components along \(x, y\) and \(\sigma\) directions calculated in the hydrodynamic module; \(A_{H}\) and \(K_{H}\) are the horizontal and vertical diffusivity coefficients, respectively, which have the same values with salinity, temperature, and cohesive sediment transport equations; \(\text{Flux}_{dHg(i)}\) and \(\text{Flux}_{PHg(i)}\) are source and sink term of two mercury forms, representing the transformation processes of different mercury species.

(1) Mercury exchange at atmosphere-ocean interface

As shown in Fig. 5.3, mercury exchange between atmosphere and ocean consists of atmospheric deposition and oceanic evasion as the surface boundary conditions of mercury transport module, and the calculation of surface boundary conditions is presented in Eq. 5.3. Atmospheric mercury is able to enter the aquatic environment through both wet and dry deposition processes. The wet deposition processes are mainly caused by rainfall and snow, while the dry deposition processes happen due to the diffusion of gaseous mercury and gravitational settling of particulate mercury. Both wet and dry deposition contain dissolved and
particulate mercury, however, exact proportion of two mercury forms is unknown. In this study we treated wet deposition as dissolved mercury input approximately, while dry deposition as particulate mercury input. Wet deposition flux is obtained from the study of Marumoto and Matsuyama (2014), and dry deposition flux is calculated by a regression formula described by Marumoto and Imai (2015), as shown in Eq. 5.4. Dissolved gaseous mercury causes evasion loss of mercury on ocean surface and the main component is Diss-Hg0. Therefore, evasion flux is used as surface boundary condition of Diss-Hg0 output and calculated by the research data of Marumoto and Imai (2015) with Eq. 5.5.

\[ \text{Flux}_{\text{surface}} = \text{Flux}_{\text{wet}} + \text{Flux}_{\text{dry}} - \text{Flux}_{\text{evasion}} \]  
\[ \text{Flux}_{\text{dry}} = 0.387 \text{Flux}_{\text{wet}} + 2.59 \]  
\[ \text{Flux}_{\text{evasion}} = K_w \left( C_w - C_{\text{air}} / H' \right) \]

where \( K_w \) is the gas exchange velocity calculated from wind speed ten meters above sea surface and Schmidt number; \( C_w \) is the dissolved gaseous mercury concentration and \( C_{\text{air}} \) is the total gaseous mercury concentration in air; \( H' \) is the dimensionless Henry’s law coefficient.

(2) Mercury exchange in water column

The movement of dissolved mercury follows advection and diffusion of ocean flow simulated by the hydrodynamic module, while the adsorbed particulate mercury moves with the transport of suspended sediments simulated by the sediment transport module in water column. As shown in Fig. 5.3, dissolved mercury transformation in water column includes oxidation, reduction, methylation and demethylation, and the mercury fluxes of three dissolved mercury species are calculated by:

\[
 F_{D-Hg0} = (K_{\text{red}} C_{d-Hg0} - K_{\text{oxi}} C_{d-Hg0})D - K_{dp} (K_{d-Hg0} C_{d-Hg0} C_{s} - P_{p-Hg0} C_{s})D 
\]

\[
 F_{D-HgL} = \left[ K_{\text{red}} C_{d-HgL} + K_{d-L} C_{d-MeHg} - (K_{\text{red}} C_{d-HgL} + K_{d-L} C_{d-MeHg}) \right]D - K_{dp} (K'_{d-L} C_{d-HgL} C_{s} - P_{p-HgL} C_{s})D
\]

\[
 F_{D-MeHg} = (K_{\text{me}} C_{d-MeHg} - K_{d-L} C_{d-MeHg})D - K_{dp} (K'_{d-MeHg} C_{d-MeHg} C_{s} - P_{p-MeHg} C_{s})D
\]

where \( F_{D-Hg0} \) represents the flux of three kinds of dissolved mercury in water column. \( K_{\text{red}}, K_{\text{oxi}}, K_{\text{dm}}, K_{\text{me}} \) are reaction rates for oxidation, deduction, methylation, and demethylation; \( K_{dp} \) is exchange coefficient of dissolved and particulate mercury; \( K'_{d-Hg0} \) represents ratio coefficient of three mercury species between particulate mercury concentration in sediment and dissolved mercury concentration in water column. First term on the right side of each formula indicates
the transformation among different dissolved mercury species and second term is the change fluxes with particulate mercury.

The transformation and reaction rates among different mercury species are insufficiently understood and difficult to obtain from observation or laboratory experiments. A simple model based on the first order kinetics equation was used to calculate these reaction rates (Avramescu et al. 2011; Hintelmann et al. 2000), as shown in Eq. 5.9 and Eq. 5.10. To simplify the calculation of reaction rates, concentration change caused by particulate mercury is neglected during computation. The exchange coefficient \( K_{dp} \) and ratio coefficients \( K_{Hg(i)c} \) are also calculated based on field observation data, and assume these coefficients keep consistent between every twice calculations. However, statistical calculation is still unable to fully reflect the real situation of mercury exchange. Therefore all these parameters are treated as calibration terms with extensive sensitivity tests before simulation. Equations for the calculation of parameters are described as:

\[
\frac{dC_{Hg0}}{dt} = K_{red} C_{Hg0} - K_{em} C_{Hg0} (5.9)
\]

\[
\frac{dC_{Hg}}{dt} = K_{red} C_{Hg} - K_{em} C_{Hg} (5.10)
\]

\[
\frac{dC_{T-Hg}}{dt} = K_{dp} \Delta(C_{Hg} P_{Hg}) (5.11)
\]

\[
K_{Hg(i)} = \frac{P_{Hg(i)}}{C_{Hg(i)}} (5.12)
\]

where \( dt \) is the observation interval; \( C_{T-Hg} \) and \( P_{Hg} \) are the total dissolved and total particulate mercury concentrations, separately.

Transformation process of particulate mercury in water column is supposed to be adsorption and desorption with suspended sediments from dissolved mercury. Particulate mercury fluxes of each species equal to the second term on the right side of dissolved mercury flux equations as shown from Eq. 5.6 to Eq. 5.8:

\[
F_{P-Hg(i)} = K_{dp} (K_{Hg(i)} C_{Hg(i)} C_{Hg} - P_{Hg(i)} C_{Hg})D (5.13)
\]

where \( F_{P-Hg(i)} \) represents flux of three kinds of particulate mercury in water column. \( Hg(i) \) represents three mercury species and other parameters have the same meaning explained in the calculation of dissolved mercury fluxes.

(3) Mercury exchange between bottom water and ocean bed

Many studies have shown that bottom sediments are the main source of mercury derivation due to the deposition of mercury pollutants (Akito et al. 2014; Balogh et al. 2015; Kumagai and
Nishimura 1978; Tomiyasu et al. 2008). The speciation of MetHg is not only in water column transformed from HgII but also produced in bottom sediments and entered the marine food chain, this process probably happens in pore water or the sediment-water interface. Detailed reaction methylation or demethylation processes in ocean bed are difficult to reproduce with numerical simulation, and these processes are incorporated to the calculation of bottom boundary conditions. Bottom boundary condition of dissolved mercury is mass exchange with pore water. Dissolved mercury in pore water is assumed to be a linear gradient distribution and exchange flux is determined by using Fick’s first law as shown in Eq. 5.14 (Covelli et al. 1999; Ullman and Aller 1982). Mercury concentration was obtained from the analytical data of upper layer pore water by Matsuyama et al. (2018). Bottom condition for dissolved mercury is:

$$\frac{K_H}{D} \frac{\partial C_{at(i)}}{\partial \sigma} = \Phi D_u \left( \frac{\partial C_{at(i)}}{\partial z_p} \right) \quad z \rightarrow H$$

(5.14)

where $\Phi$ is sediment porosity; $D_u$ is diffusion coefficient of mercury in water; $z_p$ is depth of pore water. Particulate mercury exchange at bottom layer is resuspension and deposition with bottom sediments, the erosion and deposition processes are determined by bottom critical shear stresses which calculated in the sediment transport module. Bottom boundary conditions of particulate mercury are presented as:

$$\frac{K_H}{D} \frac{\partial C_{at(i)}}{\partial \sigma} = \begin{cases} E_b P_{at(i)}, & \text{erosion} \\ D_b P_{at(i)}, & \text{deposition} \end{cases} \quad z \rightarrow H$$

(5.15)

where $E_b, D_b$ are erosion and deposition fluxes of bottom sediments.

5.2.2 Numerical Setup of Integrated Model

For the mercury transport module, some parameters were given as an approximate value based on previous studies due to the limitation of data collection in Minamata Bay. The first is the proportion of Hg0 for setting the initial condition of Hg0 and HgII, and the observation data only included the concentration of THg and MethHG. Mercury researches about lake and river systems showed the concentration ratio of Hg0 was around 10% or less (Ethier et al. 2008; Lessard 2012; O’Driscoll et al. 2003), however, for marine system Hg0 proportion varied in a wide range (Laurier et al. 2003; Sunderland and Mason 2007) and the overall mean value is basically less than 30%. Approximately, initial proportion of Hg0 was set to 20% and HgII concentration was obtained after subtracting other two mercury species. Another parameter is porosity of bottom ocean bed, the porosity value was tentatively calculated through the sediment data after centrifuging from the research of Matsuyama et al. (2018). However, this value was too small compared with the typical range between 0.4 and 0.99 for ocean benthic
porosity (Randall 2006), and the sediment porosity in this study was approximately given as 0.4. Total simulation duration of mercury simulation was the same with the sediment transport module from July 6th to July 21st, 2015 and time interval was set to 5 seconds. Open boundaries and external forcings like wind, precipitation and cloud fraction also have the same setting with sediment simulation. Measured mercury concentration on July 6th was interpolated as initial conditions for mercury with the assumed proportion. Concentrations of three mercury kinds: Diss-THg, Diss-MethHg, and Part-THg at the depth of 0, 6, 10, bottom plus 1 and bottom plus 0.1 meters obtained at three observation stations were selected for the comparison with simulated results.

5.2.3 Simulation Results of Mercury Transport Module

Model verification has been made by the cohesive sediment transport module as presented in Fig. 4.9 due to the same open boundary setting of elevation forcing. The comparison of simulation results is divided into three parts: Diss-THg, Part-THg, and Diss-MethHg. Each part presents the vertical distribution profiles of simulated and measured mercury concentration on five observation days. Simulation results on vertical 20 layers are connected with black straight lines and measured data on five depth positions are connected with dotted lines. Because five observation depth points can barely present the continuous vertical variation in whole water depth, measured data in vertical direction were smoothed through drawing software.

The first part is the vertical profile of simulated and measured Diss-THg concentrations of three stations as shown from Fig. 5.4 to Fig. 5.6. Water depth of station 1 is deeper than 20 meters and detailed distribution between 10 meters and bottom is not under measurements. The measured data displayed in this section was connected with straight dotted lines which may be different from the actual situation. However, the agreement of simulation results in vertical distribution is reasonable with measured data in all three stations. The vertical changing trends of Diss-THg concentration obtained from the numerical model were generally consistent with measured data. Vertical distribution of dissolved mercury is substantially uniform even before the remediation project (Kumagai and Nishimura 1978), the annual mean value of Diss-THg concentration shown in Fig. 5.1 also revealed the slight fluctuation of concentration change both in time series and vertical distribution. In addition to some certain points with a sudden increase of measurements which almost have double concentration change, simulation results of Diss-THg are basically satisfactory.
The second part is the vertical profile of simulated and measured Part-THg concentrations of three stations as shown from Fig. 5.7 to Fig. 5.9. Compared with simulation results on upper layers, large deviations appear at bottom layers during simulation. Sharp rise of measured particulate mercury concentration shows up from bottom plus 1 meters to bottom plus 0.1 meters and numeric differences are dozens of times, the difference even reached to ten times on July 10th at ST.1. We may reasonably deduce that some large particles existed or mixed into the
measurement samples, through flow disturbance caused by seabed flow or sampling process even with the newly established water sampler device, while this part of sediments did not participate in the vertical diffusion or advection to upper layers with ocean flow. The transport of these bottom sediments can’t satisfy the governing equations and is hardly achieved during numerical simulation. Meantime, the setting of large bottom conditions also causes disagreements near bottom layers. In addition to the vertical large difference of measured data

![Graphs showing simulated and measured Diss-THg vertical profiles at ST.2](image)

**Fig. 5.5** Comparison of simulated and measured Diss-THg vertical profiles at ST.2
near bottom layer, the time series in this area also change in a wide range, sudden times increase or decrease are difficult to simulate which could lead to numerical divergence during simulation, especially for the simulation of dissolved mercury which stays in steady level all the time. Due to the large setting of bottom boundary conditions numerous sensitivity tests have been carried out to adjust the exchange coefficient of dissolved and particulate mercury on bottom layers in order to prevent the numerical diffusion. The bottom concentration deviation
of ST.3 which locates in shallower area is not obvious compared with other stations, which is probably due to relatively weaker bottom flow and fewer erosion processes. During some periods which concentration fluctuation is not significant at ST.3, simulated vertical profiles basically keep consistent with measurement.

The third part is the vertical profile of simulated and measured Diss-MethHg concentrations of three stations as shown from Fig. 5.10 to Fig. 5.12. In contrast to Diss-THg

![Fig. 5.7 Comparison of simulated and measured Part-THg vertical profiles at ST.1](image)
Fig. 5.8 Comparison of simulated and measured Part-THg vertical profiles at ST.2

concentration, vertical variation of measured Diss-MethHg concentration is obvious as a result of smaller order of magnitude, the concentration of Diss-MethHg is almost an order of magnitude less than the concentration of Diss-THg. Consequently, concentration change in vertical direction shows larger fluctuations. Vertical distribution of measured Diss-MethHg differs from two total mercury forms where the surface layer’s concentrations are higher than bottom layer’s in most cases, which may indicate that the methylation processes on surface
layer are more frequent than bottom layer, or the demethylation processes are less frequent. Furthermore, time-varying concentration at the same depth is relatively frequent, such as ten meters of the last two observation days at ST.1 and six meters of the beginning three days at ST.2, the concentration exchange is two or three times larger. Even with extensive sensitivity tests for calibrating reaction and transformation parameters between Diss-THg and Diss-MethHg, the constant setting of different coefficients is still difficult to present a wide range of concentration fluctuations with greater accuracy. However, the agreement of simulated

Fig. 5.9 Comparison of simulated and measured Part-THg vertical profiles at ST.3
Diss-MethHg concentration is acceptable except for some period with large concentration variation. The variation trends of simulated vertical Diss-MethHg distribution can generally reflect the change of measurements.

Through the vertical profile comparisons of three mercury kinds between simulation results and measurements, simulated concentration trends can vary considerably in addition to
some points with large concentration fluctuations. Although concentrations of Part-THg on bottom layers show large deviations, through the calibration of exchange and ratio coefficients the stable simulation results of upper layers and other two mercury kinds are generally guaranteed. Reference to the comparison results of mercury concentration simulated with the PCFLOW3D model in Mediterranean Sea by Zagar et al. (Žagar et al. 2007), simulation results by the integrated mercury transport model are basically acceptable.

Fig. 5.11 Comparison of simulated and measured Diss-MethHg vertical profiles at ST.2
5.3 Simulation of Mercury Transport Pattern in Yatsushiro Sea

After verification of simulation results by the integrated numerical model for mercury transport, the concentration field in whole Yatsushiro Sea is presented as a reference for mercury transport. This simulation was carried out with modified mercury transport conditions.
in order to further understand the transport pattern of mercury. Initial conditions of mercury concentration were only given in the Minamata Bay and the concentrations were set to larger values collected before the remediation project (Kumagai and Nishimura 1978) for a more obvious display of mercury transport regularities. Distribution of initial Diss-THg was assumed to be uniformed with an averaged value of 60 ng/L on all three layers. The initial concentration conditions of Part-THg were set to 70 ng/L on surface layer, 140 ng/L on middle layer and

Fig. 5.13 Simulated concentration field of Diss-THg on three layers in Yatsushiro Sea

Fig. 5.14 Simulated concentration field of Part-THg on three layers in Yatsushiro Sea
500ng/L on bottom layer. As a principal source of mercury transmission, mercury contents of bottom sediments in Minamata Bay area were assumed to be enough high values for erosion. For the rest regions, erosion values were limited by the cumulative mercury concentration on ocean bed deposited from bottom water column.

Fig. 5.13 and Fig. 5.14 show the distribution of simulated Diss-THg and Part-THg concentration fields in the whole Yatsushiro Sea after 5 days, 10 days and 15 days, respectively. Simulation results are presented at surface, middle and bottom layers of water column at each presentation day. It is revealed that maximum simulated concentrations of two mercury kinds on bottom layers reduce to nearly half values of initial concentration after 5 days with largest concentrations around 30 ng/L and 250 ng/L compared with initial setting of 60 ng/L and 500ng/L, respectively. Meantime, mercury distribution on day 5 is almost concentrated around the bay area, it is evidently indicated that a substantial portion of mercury deposited to the ocean bed. The deposited mercury on ocean bed became the main source of mercury transmission, which was confirmed by various previous researches. It is noteworthy that both mercury forms show different spreading trends on surface layer with a southwest transport, and the differences of Diss-THg are more significant after 10 and 15 days. Compared with surface mercury, only limited mercury transport to west and south area from Minamata Bay on middle and bottom layers, while most part of these mercury spreads northward and high mercury concentrations appear in the northeast corner of Yatsushiro Sea. Even with a wider bay mouth in the west part as shown in Fig. 3.1, west transportation of mercury output from Minamata Bay mostly happens at upper layers. While the high mercury concentrations assembled in the northeast corner during simulation may be caused by the close boundary setting of the northwest part of the Yatsushiro Sea, as presented in Fig. 3.1, some narrow straits exist in the northwest and slight ocean water exchange may occur through these straits, however, this area was set to close boundaries for calculation simplification, To figure out this phenomenon, as a major factor affecting the transport of sediment and mercury, flow field at bottom layers in Minamata Bay was simulated with a high resolution grid arrangement in bay area as shown in Fig. 4.12 and simulation results are displayed in Fig. 5.15. Three kinds of tidal conditions on cbottom layer are presented on different simulation days, flood and ebb tides on July 10th and July 20th, slack tide after flood and ebb tides on July 15th. During flood tide on July 10th and 20th, bottom bay water which moves out through the west bay mouth confluences with outside torrent and flows to north direction. Generally, west and south
Fig. 5.15 Flow field on bottom layer in bay area
direction of flow transport caused by ebb tide will lead to the southwest transport of mercury from Minamata Bay through the western bay mouth. Nevertheless, as shown in the ebb tide situation on July 10th and 20th, outside currents flow into bay area during ebb tide and oppose the west outflow of bay water. Bay water exchange flows out west bay mouth is able to happen during slack tide after the ebb tide as shown in 9:00 on July 15th, while flow velocity during this period is in a low level which means the southwest transport of mercury on bottom layer is limited. The comparison of flow magnitude with same reference velocity is shown in Fig. 5.16, which is during ebb tide with north wind forcing. It is indicated that flow velocity at surface layers is higher than lower layers’, meantime, wind effect is more significant. The simulation results of flow field between surface and depth-averaged as shown in Fig. 3.14 and Fig. 3.15.
during hydrodynamic simulation, and the bottom flow field on bay area in Fig. 4.13 during sediment simulation also indicate a larger magnitude of flow velocity on surface layer. Higher velocity on surface layers will cause frequent water exchange at west bay mouth during ebb tide and the distribution of mercury at surface layers is uncertain due to the influence of wind. The southwest transport of mercury on bottom layers during slack tide after ebb tide is not significant due to the small magnitude of current velocity. It is evident that flow characteristic determines different propagation tendencies of mercury transport, which is a main factor affecting the residual mercury variation in the Minamata Bay and the Yatsushiro Sea.

5.4 Chapter Summary

With the improvement and establishment of POM and sediment module, an integrated three-dimensional numerical model for mercury transport was presented for reproducing the residual mercury dynamics in Minamata Bay. The mercury transport module in the integrated model was modified and improved based on the framework of a pollutant transport module. This model was coupled with the POM and new precipitation module for providing hydrodynamic environment and cohesive sediment transport module for reproducing the mercury cycling between water column and sediments, incorporating oxidation, methylation and other reaction processes among different mercury species.

The in-situ measurements of residual mercury behavior in Minamata Bay were discussed first. Based on the previous researches of mercury distribution, the concept of numerical simulation for mercury transport was presented. Taking into account various reaction and transformation processes of different mercury species, an integrated three-dimensional numerical model for residual mercury dynamic simulation was described. Simulated Diss-THg and Diss-MethHg showed acceptable agreements and change trends with measurements. Simulation results of bottom Part-THg displayed notable deviations on bottom layer which were probably caused by the existence of abundant coarser particles, while mercury adsorbed by these sediments could barely suspend to upper layers. To further understanding the transport pattern of mercury, mercury transport and distribution in the Yatsushiro Sea were simulated with mercury source from Minamata Bay. Study results suggested that substantial mercury deposition happened during preliminary stage of simulation. It is indicated that different transport trend appeared between surface layer and lower layers. Analysis of flow field around bay area showed that outside flow at bottom layers opposed the bay outflow at west bay mouth and mercury mass exchange to west on bottom layers mainly occurs during slack tide after ebb
tide, which was a small current velocity magnitude compared with surface layers. The high mercury concentrations assembled in the northeast corner may be caused by the close boundary setting. Surface mercury transport is sensible to wind effect and higher velocity magnitude causes the west and south transport of mercury and the transport trends were different from lower layers. In general, it is feasible to present the temporal variations and spatial distributions of residual mercury dynamics with this integrated numerical model. If sufficient data are available, bedload transport module should be coupled in the mercury model to perform specific particulate mercury distribution at ocean bed, similarly, biological module considering mercury consumption and transformation caused by fish and plankton.
6. Conclusions and Recommendations for Further Studies

6.1 Conclusions

Since the Minamata Disease caused by wastewater discharge with mercury contaminant was discovered in Minamata Bay in 1956, numerous researches have been carried out in the surrounding areas. Study emphasis mainly focuses on mercury distribution and transport, which could be affected by various factors such as wind stress, tidal and flow dynamics, sediment movement and so on. Consequently, field observation and numerical simulation were conducted aiming at different study objectives. Field observations included water sampling for the analysis of water quality behavior and sediment transport in water column, current velocity monitoring for the study of flow dynamic and ocean bottom sampling for the distribution regularities of deposited mercury on bottom sediment. Field observation can truly reflect the distribution and variation of mercury and other ocean factors, however, data obtained from in-situ measurement was limited. Numerical simulation was a more effective approach for ocean environment study combined with field investigation. Based on the sampling data from period joint field observation, an integrated three-dimensional model for mercury transport was established coupled with the hydrodynamic module and sediment transport module. The study on water on water quality dynamics and residual mercury variation in Minamata Bay was presented in this thesis. The research achievements could be concluded as follows.

(1) A pioneering numerical model in ocean research called Princeton Ocean Model was selected to supply basic hydrodynamic environment for further simulation in Minamata Bay. Simulation results of salinity and temperature in July 2006 showed good agreements with measurements at each observation station on middle and bottom ocean layers. Nevertheless, large deviations appeared during the simulation on surface layer which was caused by the frequent precipitation during plum rain season and constant setting of surface thermal radiation boundary conditions. Accordingly, a new precipitation module was constructed with new thermal radiation boundary conditions which could change temporally and spatially and integrated into POM. The increase of simulated temperature and decrease of simulated salinity were significant with new precipitation module and thermal radiation boundary conditions.

The cooling impact on temperature was not obvious during several comparisons with measurements, while the presented surface temperature field indicated the precipitation effect still existed although precipitation effect on sensible fluxes was in lower magnitude in contrast
with short wave radiation fluxes. It could be deduced that slight cooling scale shown in
comparison results is probably caused by the selected observation time under sunny weather,
and temperature on surface layer reduced by precipitation will recover soon after rain due to the
sufficient sunlight radiation in summer season. The discharge of Minamata River might have a
slight effect on temperature and salinity of ST.3 and simulation of ST.5 was sensible to the
discharge of Komenotsu River. Simulation of flow field indicated current intensity in northern
Yatsushiro Sea was higher than southern part and flow velocity on ocean surface was dominant.
Ocean water exchange of Yatsushiro Sea mainly occurred in the south Nagashima channel.

(2) As an import factor affecting the mercury distribution in coastal and offshore areas, the
study on sediment transport was inseparable for mercury research. Combining the
characteristics of sediment distribution and flow magnitude in Minamata Bay, a current induced
cohesive sediment transport module was established based on hydrodynamic model to
reproduce the transformation processes between different mercury forms and supply basic
simulation environment for mercury transport. After the construction of basic code structures,
sensitivity tests were carried out during calibration period to determine the values of critical
erosion and deposition shear stresses and analyze the influence of wind and river discharge. It is
revealed that impact of critical erosion stress on bottom sediment concentration was higher than
critical deposition shear stress. Compared with Kuma River and Minamata River, the density
flow caused by the coupling effect of Komenotsu River discharge and wind forcing had a more
visible impact on sediment transport in Minamata Bay.

The simulation of current induced cohesive sediment transport was conducted with the
calibrated coefficients obtained from the sensitivity analysis under a newly established \( \sigma \)
coordinate system. The agreements between simulated concentrations and measurements were
acceptable on bottom layer and satisfactory on upper layers due to slight concentration
fluctuations, the trends of sediment concentration were generally consistent with the variation
of bottom shear stresses. The display of flow and sediment concentration fields in bay area
indicated most strong current bypassed the northern strait during ebb tide and ensured the
stability of bay water, and current magnitude could be amplified when flowed out through the
northern narrow strait during flood tide.

(3) After the construction and verification of hydrodynamic and sediment modules, the
integrated mercury transport model was presented to reproduce the residual mercury variation
in Minamata Bay. Mercury transport module in the integrated model was modified and
promoted from a pollutant transport module, and code structures have been optimized and
various external conditions based on the researches in recent years were applied. The integrated
three-dimensional mercury transport model considered comprehensive transformation processes of different mercury species in water column and with sediments.

Three mercury kinds obtained from in-situ measurements were selected for the comparison of simulation results. Simulated concentrations of Diss-THg and Diss-MethHg showed acceptable agreements with measurements, and changing trends of two kinds of mercury presented with vertical profiles generally kept consistent with field observation data. Notable deviations occurred in the simulation of Part-THg on bottom layer and measurements near bottom also showed great concentration difference, which were probably caused by the existence of abundant coarser particles. This part of sediments did not participate in the vertical diffusion or advection to upper layers with ocean flow and could barely be achieved during numerical simulation. The transport pattern of mercury was presented over the large Yatsushiro Yea considering mercury source from Minamata Bay. It is revealed that large amounts of mercury deposited into the ocean bed of bay area in the preliminary period of simulation. Meantime, mercury transport between surface layer and lower layers showed different tendencies with west on surface and north on lower layers. The flow field under different tidal conditions in bay area indicated the surface current was stronger than lower layers and sensible with wind forcing. The southwest transport of bottom mercury mainly happened during slack tide, while the magnitude was smaller compared with surface layer. High mercury concentration shown in the northeast corner of Yatsushiro Sea might be caused by the close boundary setting for computational simplification.

6.2 Recommendations for Further Studies

With the establishment and improvement of hydrodynamic, sediment and mercury modules, the integrated numerical model presented in this study was generally feasible to reproduce the temporal and spatial variation of residual mercury and other ocean indexes. However, some problems have been encountered during the research process and enhancements of module construction and field observation are necessary to optimize the simulation accuracy. Recommendations for further studies are listed as follows.

1) The optimization of pre-processing and post-processing. Most open source programs need complicated pre-processing effort before the start of simulation, and this problem is especially obvious on POM without interfaces to manipulate. Except for the re-compilation of source code setting, numerous input files are needed to specify the computational grids, topography, initial and external conditions. The number of input files in the mercury simulation
reaches up to twenty. Moreover, output of simulation results needs code programming depending on different research objectives. The design of user interfaces for optimizing the pre-processing and post-processing is recommended to simplify the simulation processes.

(2) The wave module. In consideration of low flow magnitude and limitation of wave data collection in Minamata area, this study didn’t consider the wave module and bottom shear stress was assumed to be consequence induced by current only. However, wave module is necessary for the improvement of simulation precision in the larger Yatsushiro Sea, and detailed flow dynamics could be presented with wave module.

(3) Grid nesting and parallel computation. The present uniform grid couldn’t display the detailed dynamics in small scale region, while the non-uniformed grid would increase the running time several times. The grid nesting technology is an effective approach to solve this problem with local refinement of research area without sacrificing computation resources. Parallel computation also contributes to save calculation time.

(4) The ocean bed module and more field observation information. Due to the restrictions of bed data collection, ocean bed was simplified to one layer and the transport of coarser particles was not considered in this study. The dynamic of ocean bed also plays an important role in ocean simulation, especially for bottom non-cohesive sediments and particulate mercury, which has been displayed during the mercury simulation. Sufficient field observation data are necessary to establish the bed module like bed layer thickness, sediment fraction, bulk density and porosity and so on. With enough field observation data, the river boundary conditions and the calculation of various coefficients will be optimized.

(5) Biological module. Except for the mercury transport in ocean and sediment environments, the biological module is also important to present the mercury cycling through biological processes. The mercury absorbed or consumed by marine creatures should be considered in the integrated mercury transport model with a biological module if sufficient field observation data are available in future research.

(6) Data assimilation. Data assimilation has been successfully applied in some ocean circulation numerical models like ROMS and FVCOM. Data assimilation is able to update and correct simulation results with input of newly observed data, this process will filter errors exist in simulation and measurements. This technology will seek the optimal combination of simulation and measurements to help predicted results closer to actual data.
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