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学位論文要旨
Dissertation Summary

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論　文　名：Simulations of PCBs in the Northwestern Pacific Ocean with a Three-Dimensional High-Resolution Hydrodynamic-Ecosystem-PCB Coupled Mode
(Dissertation Title)

With the reduction of PCBs emission over land, the ocean, which has been a reservoir of PCBs for several decades, is likely to be a secondary source in recent years, PCBs in the surface ocean may re-enter the atmosphere by volatilization and further enter atmospheric transportation. Field studies have reported the degassing of PCBs from the Northeastern Pacific Ocean and the Atlantic Ocean. The main source of PCBs in the open ocean is atmospheric deposition which includes dry deposition, wet deposition, and air-sea diffusion flux, among which air-sea diffusion was the dominant one. The oceanic biological pump, current advection, and eddy diffusion affect the distribution of PCBs in the surface ocean and thus affect the air-sea diffusion flux. The ocean current brings the water with different concentrations of PCBs from a remote area and therefore changes the local concentration of PCBs; the eddy diffusion acts to reduce the spatial gradient of concentration of PCBs.

Western boundary currents, with a width of 100-200 km, flow close to the land. For example, the Gulf Stream flows along the east coast of North America; the Brazil current

flows along the east coast of South America; the Kuroshio flows along the south coast of Japan. Because of its close distance to the land, the high emission over land results in a high gaseous concentration of PCBs over these western boundary currents. Consequently, these currents may have a high dissolved concentration of PCBs and gradually release PCBs into the air as they flow far away from the land. Therefore, these western boundary currents may play an important role in the global air-sea exchange of PCBs.

The ocean is not only a dominant sink of PCBs but also a place demonstrating the intense bioaccumulation of PCBs in the food web. High PCBs concentrations have been detected in deep-sea organisms in global oceans owing to their lipophilicity and the potential of easily bioaccumulation. The occurrence of PCBs in deep water masses may be of concern in the future, as these may eventually rise to the surface as part of the global circulation belt. Clarifying the accumulation of PCBs in deep water and the relative influence of the microbial and food webs on the cycling of PCBs is therefore important for improving the estimates of the oceanic sink of PCBs, probably associated to the settling of dissolved phase PCBs.

The Asia-Pacific region has the largest human population and the largest manufacturing factories, making it an area of highly using of PCBs. There is about 30% of the total atmospheric deposition took place in the North Pacific. In addition, there is a semi-closed marginal sea in the NWPO, the Japan Sea. A bowl-shaped bathymetry of the Japan Sea confines the effect of throughflows on the upper 200m and separates the intermediate and deep waters from the adjacent seas. This closed feature or independency in the deep layer of the Japan Sea compared to the open ocean makes it appropriate to investigate the vertical accumulation process of PCBs in the deep ocean. To describe and understand the complex three-dimensional dynamics of PCBs in ocean and to predict fate of PCBs in ocean, besides field measurement, a numerical model is also a useful tool. Numerical modeling studies suggest that not only horizontal advection and diffusion by current and turbulence, but also vertical transport of PCBs absorbed into phytoplankton are important processes.

In this work, a three-dimensional hydrodynamic-ecosystem-PCB coupled model was developed for the northwestern Pacific Ocean (NWPO) to simulate the transport and biological processes of four PCB congeners. In this model, a high-resolution hydrodynamic model (1/12°) was used to reproduce a realistic western boundary current, and this module provides daily water temperature and current velocity in three directions, and the horizontal and vertical eddy diffusivity coefficients; a full coupling ecosystem module was used to present the biological pump effect on PCBs, this module provides daily concentrations of phytoplankton and related biogeochemical parameters; an atmospheric model results of PCBs were used as the surface boundary condition to analyze the spatial and temporal variations in the oceanic PCBs concentration.

The model results demonstrated the seasonal variation and spatial distribution of the dissolved and particulate PCBs concentration in the NWPO, which are compared with all the available observation data. The concentrations of dissolved PCBs increased from the subtropical region to the subarctic region, from marginal seas to the open ocean. The particulate PCBs concentrations were much lower than the dissolved phase, concentrated in the subarctic region. The seasonal variations of dissolved PCBs showed latitudinal diversity. The concentrations of dissolved PCBs were higher in winter, and lower in summer in most areas of the model domain, but this situation is reversed around 40N. The dissolved concentration of CB28 was 2-3 folds higher than that of CB153 due to the larger atmospheric deposition.

With the model results, firstly, the air-sea diffusion flux of four selected PCB congeners and the regulatory effect of ocean currents on the air-sea diffusion flux was elucidated. Then the accumulation process of PCBs in deep water and the corresponding dynamic mechanism were discussed.

The model revealed a fine structure in the air-sea flux that is sensitive to the presence of ocean currents, especially the Kuroshio, a western boundary current with a high surface speed. Intense downward and upward diffusion fluxes can be found in the Kuroshio region south of Japan and the Kuroshio Extension east of Japan, respectively. The air-sea flux of Σ_4PCBs ranged between -23.6 and 44.75 $ng\ m^{-2}\ d^{-1}$ in these regions, which is remarkable compared with that in weak current regions. The sensitivity experiments indicated that it takes ~4 days in the strong western boundary current region and 1~3 days in the weak current regions for the surface dissolved PCBs concentration to reach an equilibrium in the scenarios where only air-sea exchange is considered. On the other hand, the equilibrium time of surface dissolved PCBs concentration becomes ~1 day in the strong western boundary current region and 3~12 days in the weak current regions in the scenario where only ocean advection is considered. Therefore, the surface dissolved PCBs concentration in weak current regions is mainly controlled by the air-sea exchange because the air-sea exchange has a shorter equilibrium time than ocean advection does. For the same reason, the surface dissolved PCBs concentration in the western boundary current region is controlled by the ocean advection, indicating that dissolved PCBs from upstream carried by the Kuroshio can easily change the dissolved concentration in its downstream by disrupting the equilibrium with the original air-sea exchange and induce a new air-sea flux there. This is the cause for the appearance of downward and upward fluxes of PCBs along the Kuroshio south of Japan and Kuroshio extension, respectively. Therefore, ocean currents, especially the strong western boundary currents, should be correctly considered in future atmospheric transport models for PCBs.

The vertical distribution of CB153 concentration in the Japan Sea showed a three-layer structure in our model results: low concentration in the surface layer and deep layer but high concentration (~0.7 $pg\ L^{-1}$) in the intermediate layer. In addition, the high concentration in the intermediate layer showed little seasonal variation. It should be noted that the PCBs below the surface water were still being continuously accumulated because the standing stock of CB153 in the intermediate and deep layers increased by 3.35 and 16.56 $kg\ year^{-1}$, respectively. The different magnitude in the two layers means that the concentration of CB153 in our model will increase more in the deep water than in the intermediate layer if we extend the model integration time in the future.

Air-sea exchange and inflow from the Tsushima warm current were two sources of CB153 in the Japan Sea. Sensitivity experiments suggested that the biological process contributed 25% to the variation of CB153 mass in the surface layer, while the combination of air-sea flux, advection, and diffusion provide the residual (75%). In the intermediate and deep layers, the contribution of biological processes increased to 40%, which was owing to the decomposition of detritus bound CB153 below the mixed layer in the central Japan Sea. Through advection and diffusion, the Tsushima warm current contributed 10% while the air-sea exchange contributed 50% to the PCBs in the intermediate and deep layers.