

Contents lists available at ScienceDirect

# **Environmental Research**



journal homepage: www.elsevier.com/locate/envres

# Spatial and seasonal variations of polychlorinated biphenyl (CB-153) in the Seto Inland Sea and its response to declines in atmospheric and riverine input

Yaxian Li<sup>a</sup>, Xinyu Guo<sup>b,\*</sup>, Qian Leng<sup>b</sup>, Min Yang<sup>c</sup>, Yu Bai<sup>a</sup>, Xueting Zhao<sup>a</sup>

<sup>a</sup> Graduate School of Science and Engineering, Ehime University, 2-5 Bunkyo-cho, Matsuyama, 790-8577, Ehime, Japan

<sup>b</sup> Center for Marine Environmental Studies, Ehime University, 2-5 Bunkyo-cho, Matsuyama, 790-8577, Ehime, Japan

c State Key Laboratory of Satellite Ocean Environment Dynamics, Second Institute of Oceanography, Ministry of Natural Resources, 36 Baochu Road, Hangzhou,

310012, China

#### ARTICLE INFO

Keywords: POPs ocean model Dissolved CB-153 Particulate CB-153 Spatial and seasonal variations Seto Inland Sea

### ABSTRACT

Polychlorinated biphenyls (PCBs) concentrations in rivers and the atmosphere have decreased in recent years, which raises a necessity for a quantitative analysis of the responses of PCBs concentration in coastal seas to this decline. A hydrodynamic-ecosystem-PCB coupled model was developed to investigate the spatial and seasonal variations of CB-153 in the Seto Inland Sea and its responses to the decline of CB-153 input from rivers and atmosphere. The model simulated dissolved and particulate (phytoplankton- and detritus-bound) CB-153 and their physical and biogeochemical behaviors. Model results indicated that the dissolved CB-153 concentration peaked in July, which was caused by atmospheric and river input. The particulate CB-153 peaked in April when the phytoplankton uptake was largest due to the spring phytoplankton bloom, and in July when the dissolved CB-153 concentration of 10.3 ng m<sup>-3</sup> while the other areas had an annual mean of 2.9 ng m<sup>-3</sup>. The CB-153 budget calculation revealed that the atmosphere input through air–sea diffusion process was the largest source (61%), while rivers discharges served as a secondary source (31%). Three scenarios were designed to explore the responses of the Seto Inland Sea to decline of CB-153 input from atmosphere and rivers. With the decline of CB-153 input from atmosphere, the CB-153 concentration decreases over the Seto Inland Sea except for Osaka Bay. On the other hand, Osaka Bay greatly responds to the decline of CB-153 input from rivers.

## 1. Introduction

Polychlorinated biphenyls (PCBs) are a traditional and ubiquitous group of persistent organic pollutants (POPs) that were extensively manufactured and utilized until they were banned in the 1970s. Because of their persistence and long-range transport, PCBs have long been stored in oceans (Jurado et al., 2004b), shelf sediments (Jönsson et al., 2003), and terrestrial soils (Meijer et al., 2003). Owing to their bio-accumulation, biomagnification, and high toxicity, PCBs are detrimental to the health of organisms, particularly advanced predators (Carpenter, 2006; Madgett et al., 2022).

The atmosphere is recognized as the primary source of PCBs in open oceans (Zhang and Lohmann, 2010). Wet and dry deposition can move PCBs from the atmosphere into the oceans, but air–sea diffusion of PCBs is the dominant depositional process in open oceans (Jurado et al., 2004a, 2005). Oceans are one of the most important reservoirs for PCBs (Iwata et al., 1993; Wania et al., 1998), with CB-153 concentration being 0.12 pg L<sup>-1</sup> in the Northern Hemisphere oceans and about 0.002 pg L<sup>-1</sup> in the Southern Hemisphere (Wagner et al., 2019). In addition, urban and industrial emissions of PCBs have a substantial impact on coastal and shelf seas through wastewater emissions and river runoff, with rivers being significant sources of coastal and shelf seas, resulting in CB-153 concentration in these seas with a range of 1–120 pg L<sup>-1</sup> that is much higher than those in open oceans (Lohmann and Dachs, 2019). Consequently, PCBs in some shelf seas and coastal water become supersaturated and volatilize to the atmosphere as re-emission sources (Berrojalbiz et al., 2014).

PCBs tend to be absorbed by planktons in seawater because they have

https://doi.org/10.1016/j.envres.2025.122000

Received 10 January 2025; Received in revised form 6 April 2025; Accepted 27 May 2025 Available online 28 May 2025 0013-9351/© 2025 Elsevier Inc. All rights are reserved, including those for text and data mining, AI training, and similar technologies.

<sup>\*</sup> Corresponding author. Center for Marine Environmental Studies, Ehime University, 2-5 Bunkyo-Cho, Matsuyama, 790-8577, Japan. *E-mail address:* guo.xinyu.mz@ehime-u.ac.jp (X. Guo).

a relatively high octanol-water partition coefficient ( $K_{ow}$ ), which indicates their lipophilicity (O'Driscoll et al., 2013). Phytoplankton affects the biogeochemical processes of PCBs in two aspects: as primary producers, they carry PCBs into the marine food chains; as biogenic particles, they influence the partition of PCBs in dissolved and particle phases, thereby affecting the air-sea diffusion flux and the downward flux to the sediment (Dachs et al., 2000, 2002; Gioia et al., 2012; Madgett et al., 2022; Nizzetto et al., 2012; Wang et al., 2024).

Since the production and use of PCBs were gradually banned on a global scale, PCBs concentrations have significantly decreased in the environment (Pavlova et al., 2014; Schuster et al., 2010). Atmospheric levels have declined due to the reduction of primary emissions (Breivik et al., 2007; Nizzetto et al., 2010). PCBs concentrations in coastal sediments follow the temporal trend of global PCBs emissions and respond rapidly to changes in global industrial usage (Sobek et al., 2015). The declines of PCBs in the North Sea were suggested as the result of declines in atmospheric and riverine PCBs concentrations (O'Driscoll et al., 2013).

This study focused on the Seto Inland Sea due to its early industrialization and dense population, which increase the likelihood of potential PCBs emissions through the rivers and atmosphere. Long-term monitoring by the Ministry of the Environment of Japan indicated that the Seto Inland Sea was one of the areas with relatively high concentrations of PCBs in both water and atmosphere. Previous studies have suggested that large amounts of PCBs residues in seawater, blue mussels, and coastal pellets in the Seto Inland Sea (Endo et al., 2005; Tsuno et al., 2007; Ueno et al., 2010; Isobe et al., 2011). Furthermore, PCBs were found to be more abundant than other POPs in the samples of high trophic-level organisms collected from the Seto Inland Sea (Tsuno et al., 2007; Ueno et al., 2010; Isobe et al., 2011). Our simulation targeted CB-153, which is one of the 209 congeners and easily accumulates in organisms. CB-153 concentrations in phytoplankton and higher trophic level organisms dominate all PCB congeners in the Seto Inland Sea (Matsumoto et al., 2016).

Although there were some observations of PCBs in the Seto Inland Sea, they were not sufficient for a full understanding of the physical transport and biogeochemical processes of PCBs in the sea. As an alternative way, we will use a numerical simulation to quantitatively understand the CB-153 transport and budget in the Seto Inland Sea. Three-dimensional transport models have been applied to assess PCB levels in open oceans and shelf seas (Ilyina et al., 2006; Ono et al., 2012; Alekseenko et al., 2018; Wagner et al., 2019; Yang et al., 2022, 2024). In the region close to the Seto Inland Sea, a coupled model for PCBs in the Northwestern Pacific Ocean has been used to study the role of the Kuroshio in the air–sea exchange of PCBs (Yang et al., 2022). Although this model covers the Seto Inland Sea, its horizontal resolution ( $\sim$ 10 km) is too coarse to resolve the transport and budget of PCBs in the Seto Inland Sea.

In this study, we developed a high resolution three-dimensional hydrodynamic-ecosystem-POP coupled model to reproduce the fate and transport of CB-153, aiming to elucidate the spatial and seasonal variations and budget of CB-153 in the Seto Inland Sea. Because the decreasing trends of CB-153 have been observed by the Ministry of the Environment in the estuaries and atmosphere around the Seto Inland Sea, we also used this model to investigate the response of CB-153 in the Seto Inland Sea to the reduction from atmospheric and riverine sources.

# 2. Model description

The Seto Inland Sea is a semi-enclosed shelf sea in western Japan. It has a surface area of approximately 23,000 km<sup>2</sup> and an average depth of about 38 m. The sea is bordered by Honshu, Shikoku, and Kyushu, which are highly industrialized regions in Japan. A total of 21 first-order rivers and 644 second-order rivers empties this sea. It connects to the Pacific Ocean via the Bungo and Kii channels.

The model domain (130.98-135.5°E, 32.8-34.8°N) is shown in Fig. 1a. It features a horizontal resolution of approximately 1 km and 21 layers in the vertical direction. The simulation for PCBs includes hydrodynamic, ecosystem, and PCB modules. The hydrodynamic module is a three-dimensional ocean circulation model based on the Princeton Ocean Model (Blumberg and Mellor, 1987). When compared with in-situ observations from the Seto Inland Sea, the simulated results of the hydrodynamic module for current fields, water temperature, and salinity suggest that the model well reproduced the features of the physical field (Chang et al., 2009; Zhu et al., 2019; please refer to Appendix A for its details). The ecosystem module is a lower-trophic model that simulated the interactions among nutrients, phytoplankton, zooplankton, and detritus, providing the phytoplankton biomass and related biogeochemical parameters for the PCB module. The comparison of simulated phytoplankton biomass versus observations suggests that the ecosystem module works well in areas with low biomass ( $<250 \text{ mg C m}^{-3}$ ) but underestimates the high biomass in nearshore regions, and their linear correlation coefficient was 0.63 (Leng et al., 2023; please refer to Appendix A for its details).

#### 2.1. PCB module



The three states of CB-153 (dissolved, phytoplankton-bound, and detritus-bound) were used for simulations in the PCB module, which includes CB-153 uptake in the phytoplankton matrix and adsorption on

**Fig. 1.** (a) Bathymetry of the Seto Inland Sea. The blue dots and green dots represent the position of first-order and second-order river mouths. The solid black lines indicate the Bungo and Kii Channels, which are the southern boundaries of the model. (b) Conceptual scheme of the coupled model for CB-153 simulation.  $C_{a}$ ,  $C_{wp}$ , and  $C_{wd}$  are the CB-153 concentrations of the gaseous phase, dissolved, phytoplankton-bound, and detritus-bound, respectively.

the phytoplankton surface (Fig. 1b). The initial conditions for each state of CB-153 concentration were set to 0 ng m<sup>-3</sup>, and the control equations are given by Eqs. (1)–(4):

$$\frac{\partial C_w}{\partial t} + u \frac{\partial C_w}{\partial x} + v \frac{\partial C_w}{\partial y} + w \frac{\partial C_w}{\partial z} = \frac{\partial}{\partial z} \left( K_h \frac{\partial C_w}{\partial z} \right) + D(C_w) + Bio(C_w)$$
(1)

$$\frac{\partial C_{wpm}}{\partial t} + u \frac{\partial C_{wpm}}{\partial x} + v \frac{\partial C_{wpm}}{\partial y} + (w + w_{sp}) \frac{\partial C_{wpm}}{\partial z} = \frac{\partial}{\partial z} \left( K_h \frac{\partial C_{wpm}}{\partial z} \right) + D(C_{wpm}) + Bio(C_{wpm})$$
(2)

$$\frac{\partial C_{wps}}{\partial t} + u \frac{\partial C_{wps}}{\partial x} + v \frac{\partial C_{wps}}{\partial y} + (w + w_{sp}) \frac{\partial C_{wps}}{\partial z} = \frac{\partial}{\partial z} \left( K_h \frac{\partial C_{wps}}{\partial z} \right) + D(C_{wps})$$

$$+ Bio(C_{wps})$$
(3)

$$\frac{\partial C_{wd}}{\partial t} + u \frac{\partial C_{wd}}{\partial x} + v \frac{\partial C_{wd}}{\partial y} + (w + w_{sd}) \frac{\partial C_{wd}}{\partial z} = \frac{\partial}{\partial z} \left( K_h \frac{\partial C_{wd}}{\partial z} \right) + D(C_{wd}) + Bio(C_{wd})$$
(4)

where  $C_w$  (ng m<sup>-3</sup>) is the dissolved CB-153 concentration, and  $C_{wpm}$ ,  $C_{wps}$ , and  $C_{wd}$  are the concentrations (ng m<sup>-3</sup>) of CB-153 in the phytoplankton matrix, phytoplankton surface, and detritus, respectively. The sum of  $C_{wpm}$ ,  $C_{wps}$ , and  $C_{wd}$  is defined as particulate concentrations in this study; *t* is time (s); *x*, *y*, and *z* are the zonal, meridional, and vertical coordinates (m), respectively; *u*, *v*, and *w* are the velocity components (m s<sup>-1</sup>) in three directions;  $w_{sp}$  and  $w_{sd}$  are the sinking velocities of phytoplankton (0.1 m d<sup>-1</sup>) and detritus (1.0 m d<sup>-1</sup>), respectively; *K<sub>h</sub>* is the vertical eddy diffusion coefficient (m<sup>2</sup> s<sup>-1</sup>);  $D(C_w)$ ,  $D(C_{wpm})$ ,  $D(C_{wps})$ , and  $D(C_{wd})$  are the horizontal diffusion terms; and  $Bio(C_w)$ ,  $Bio(C_{wpm})$ ,  $Bio(C_{wps})$ , and  $Bio(C_{wd})$  are the biogeochemical terms for each compartment which will be described in section 2.2. The biogeochemical terms are described as Eqs. (S1)–(S8) in Appendix B.

The numerical scheme of the CB-153 calculation is like the state variables such as nutrient concentration and phytoplankton biomass in the ecosystem module. Eqs. (1)-(4) are solved in two steps, with the explicit scheme for the advection, horizontal diffusion, and biogeochemical terms, and the implicit scheme for vertical diffusion.

#### 2.2. Biogeochemical processes of CB-153

The biogeochemical processes of CB-153 (Fig. 1b) are expressed as the *Bio* term in Eqs. (1)–(4) for  $C_w$ ,  $C_{wpm}$ ,  $C_{wps}$ , and  $C_{wd}$ . In seawater, dissolved CB-153 is absorbed by the surface and matrix of phytoplankton and is also degraded through both photolytic and biological processes. Phytoplankton-bound CB-153 is affected by two primary processes: uptake and adsorption from dissolved CB-153, and the release of CB-153 from phytoplankton back into seawater through depuration and desorption (Del Vento and Dachs, 2002). Additionally, the natural mortality of phytoplankton transforms phytoplankton-bound into detritus-bound CB-153. Some of the detritus-bound CB-153 is transported downward to the seabed by sinking of detritus, while the others are returned to the dissolved phase through the remineralization of detritus.

# 2.3. Source of PCBs

The model has PCB input from 41 rivers (21 first-order rivers and 20 second-order rivers) whose positions are shown in Fig. 1a. The river input amount of CB-153 ( $F_{riv}$ , ng s<sup>-1</sup>) is calculated by,

$$F_{riv} = C_{riv}Q \tag{5}$$

where  $C_{riv}$  (ng m<sup>-3</sup>) is the dissolved CB-153 concentration in river water, which was monitored by the Ministry of the Environment, Japan; Q (m<sup>-3</sup> s) is river discharge whose daily values of 21 first-order rivers were

provided by the Ministry of Land, Infrastructure, Transport and Tourism, and those of 20 second-order rivers by the local governments. The annual mean river discharge, CB-153 concentrations, and flux of first-order rivers are given in Table S3. The sum of second-order river flux is small and is at an order of flux from one first-order river.

The air–sea interface considers two processes: air–sea diffusion between gaseous and dissolved CB-153, and wet deposition. Dry deposition flux was suggested lower than the air–sea diffusion flux at coast water by one order of magnitude (Totten et al., 2004). In addition, we also cannot find the data of dry deposition data around the Seto Inland Sea. Therefore, we did not include it in the PCB module. Consequently, the surface boundary condition for the CB-153 is expressed as:

$$\zeta_h \frac{\partial C_w}{\partial z} = F_{as} + F_{wp} \tag{6}$$

where  $F_{wp}$  (ng m<sup>-2</sup> s<sup>-1</sup>) is the wet deposition flux, which was obtained from the GEOS-Chem global atmospheric model (Friedman and Selin, 2016);  $F_{as}$  (ng m<sup>-2</sup> s<sup>-1</sup>) is the air-sea diffusion flux calculated by a two-film model (Bidleman and McConnell, 1995), whose formula is

$$F_{as} = k_{as}(C_a / H' - C_{ws}) \tag{7}$$

where  $k_{as}$  (m s<sup>-1</sup>) is the air-sea transfer velocity which is a function of wind speed and sea surface temperature.  $C_{ws}$  (ng m<sup>-3</sup>) is the dissolved CB-153 concentration in the sea surface;  $C_a$  (ng m<sup>-3</sup>) is the CB-153 gaseous concentration; and H' is the dimensionless Henry's law constant for CB-153, which depends mainly on the water temperature (Kames and Schurath, 1992; Cetin and Odabasi, 2005; please refer to Appendix C for its details).

Although the atmospheric concentrations of PCBs calculated by the GEOS-Chem model have been evaluated at some sites in the Arctic, Europe, and North America (Friedman and Selin, 2016), the model's performance over the Seto Inland Sea remains unknown. Therefore, we compared the atmospheric concentration of CB-153 from the GEOS-Chem model with the monitored data provided by the Ministry of the Environment (Fig. S1), and found that the GEOS-Chem model significantly underestimated the atmospheric concentration of CB-153 around the Seto Inland Sea. Consequently, based on the slopes of the linear fitted results from the observed data and model results (Fig. S1), we enhanced the GEOS-Chem gaseous CB-153 concentration by 30 times, and the seasonal variations were maintained.

Besides rivers and air–sea interface, Seto Inland Sea has two lateral boundaries (Bungo and Kii channels) connected to the Pacific Ocean, where the monthly averaged concentrations of dissolved and particulate CB-153 derived from the results of Yang et al. (2022) were specified.

#### 2.4. Model validation

ŀ

We collected some observed data near the coast in the Seto Inland Sea, which are the sum concentrations of dissolved and particulate CB-153 in the surface layer (Tsuno et al., 2007), to validate the model results. Fig. S2a shows the mean concentration along the shoreline in July and October. The concentration of CB-153 from the model results and observation are nearly identical along the coasts of Suo Nada, Iyo Nada, and Hiuchi Nada; however, the concentrations from model results are lower than those from observations along the coasts of Hiroshima Bay, the northern part of Hiuchi Nada, and Uwajima. Fig. S2b shows the mean concentrations from May to December, which generally agree with the observation on the northern coast of Harima Bay and the head of Osaka Bay, except for the west coast of Awaji Island, where the model results were lower than the observations. These underestimations may be caused by either the omission of the local source of CB-153 or the omission of non-biogenic particles in the model. Fig. S2c demonstrates a significant positive relation (p < 0.01) between the observations and model results, with a correlation coefficient of 0.6. The root mean squared error (RMSE) and mean absolute error (MAE) are 6.7 and 6.3 ng  $\rm m^{-3},$  respectively. Overall, the model results generally capture the spatial variations of CB-153 in the Seto Inland Sea.

## 3. Results and discussion

#### 3.1. Spatial and seasonal variations of CB-153 concentrations

The average concentrations of dissolved and particulate CB-153 over the entire model domain in three years of the simulation are shown in Fig. 2. The CB-153 concentrations in the second and third simulation years were almost identical, indicating that the calculations reached a stationary state in the third simulation year. In the following analysis, we used the results from the third simulation year to investigate the spatial and seasonal variations of CB-153 concentrations in the Seto Inland Sea.

Dissolved and particulate CB-153 concentrations exhibit a clear seasonal variation (Fig. 2a). The dissolved CB-153 concentration ranged from 1.3 to 2.8 ng m<sup>-3</sup>, with an average of 1.9 ng m<sup>-3</sup>, while the particulate CB-153 concentration ranged from 0.2 to 0.6 ng m<sup>-3</sup>, with an average of 0.4 ng m<sup>-3</sup>. Dissolved CB-153 concentration was highest in July, whereas particulate concentration peaked in April and July, respectively. Both phases of CB-153 concentrations were the lowest in January. Particulate CB-153 consists of phytoplankton- and detritusbound CB-153, both of which exhibited two peaks over a year (Fig. 2b).

We selected January, April, July, and October to represent the four seasons and examined the spatial distributions of CB-153 concentrations in these four months. There are apparent spatial differences in the horizontal distributions of the dissolved CB-153 concentration (Fig. 3a, b, c, &d). The range of dissolved CB-153 concentration is broad, with the highest concentrations of 35.0, 32.4, 76.6, and 29.1 ng m<sup>-3</sup> in Osaka Bay in these four months, primarily due to the input of dissolved CB-153 from Yodo River into Osaka Bay. The other regions with relatively high concentrations are the Suo Nada, Iyo Nada, Hiuchi Nada, Harima Nada, and Hiroshima Bay, where the concentration decrease from the nearshore to the offshore areas. The minimum dissolved CB-153 concentration in the offshore areas is approximately 0.2 ng m<sup>-3</sup>. The



**Fig. 2.** Time series of the averaged concentrations over the entire model domain for (a) dissolved and particulate CB-153, (b) phytoplankton- and detritus-bound CB-153. The particulate CB-153 concentration is the sum of phytoplankton- and detritus-bound CB-153 concentrations.

particulate CB-153 concentration also changes spatially in four seasons (Fig. 3e, f, g, &h). High concentration is observed in the estuary areas, especially in the nearshore area of Osaka Bay, due to the presence of the high concentration of dissolved CB-153 and high phytoplankton biomass there. In general, the concentration of dissolved CB-153 is higher than that of particulate CB-153 in the Seto Inland Sea except for the nearshore area of Osaka Bay where the two phases of CB-153 have the same concentrations.

# 3.2. CB-153 mass flow and balance

The annual mean masses of dissolved, phytoplankton-, and detritusbound CB-153 in the Seto Inland Sea are 1.71, 0.23, and 0.14 kg, respectively. To understand their fate and transformation, we calculated the mass flow with the biogeochemical processes and the mass balance with the physical processes (Fig. 4). The annually integrated fluxes ( $F_p$ , kg y<sup>-1</sup>) of the biogeochemical processes are calculated using Eq. (8),

$$F_p = \int_1^T \iiint F(x, y, z, t) dx dy dz dt$$
(8)

where F (kg m<sup>-3</sup> s<sup>-1</sup>) is the flux due to a biogeochemical process; dx, dy, and dz are the unit distance (m) in three directions; and T and dt are a year and unit time (s), respectively. Equation (8) was applied to all the grid points of the model domain and obtained the total mass flow corresponding to each biogeochemical process in the Seto Inland Sea.

Regarding the biogeochemical flux of dissolved CB-153, the net absorption (uptake-depuration) by phytoplankton is 11.9 kg y<sup>-1</sup>, the degradation is 1.0 kg y<sup>-1</sup>, and the decomposition of detritus is 9.5 kg y<sup>-1</sup>; their total effects is the loss of 3.4 kg y<sup>-1</sup>. Regarding the physical flux, the river input is 2.0 kg y<sup>-1</sup>, the atmosphere input is 4.0 kg y<sup>-1</sup>, and the output to the Pacific Ocean is 2.6 kg y<sup>-1</sup>; their total effects is the supply of 3.4 kg y<sup>-1</sup>, which is balanced with the loss of CB-153 due to biogeochemical processes.

Regarding the phytoplankton-bound CB-153, its uptake of dissolved CB-153 (11.9 kg y<sup>-1</sup>) is balanced by the mortality of phytoplankton that transformed phytoplankton-bound into the detritus-bound CB-153 (11.5 kg y<sup>-1</sup>), and the outflow of phytoplankton-bound CB-153 into the Pacific Ocean (0.4 kg y<sup>-1</sup>). Regarding the detritus-bound CB-153, in addition to its production by the mortality of phytoplankton (11.5 kg y<sup>-1</sup>) and its decomposition into dissolved phase (9.5 kg y<sup>-1</sup>), its sinking to the sea bottom is 1.7 kg y<sup>-1</sup> and its outflow to the Pacific Ocean is 0.3 kg y<sup>-1</sup>.

The ratios of each physical process and biogeochemical process to the total input or output are presented (Fig. 4b). At the air–sea interface, diffusion induces two directions of flux. One is from air to sea (3.9 kg y<sup>-1</sup>), while the other is from sea to air (0.4 kg y<sup>-1</sup>). The air–sea diffusion flux from air to sea accounts for approximately 61% of the total input of dissolved CB-153 (6.4 kg y<sup>-1</sup>). River inputs (2.0 kg y<sup>-1</sup>) account for approximately 31% of the total input of CB-153. As shown later, river input exhibits a strong regional dependence because the Yodo River contributes 75% of the total river input and is the primary source for Osaka Bay. Besides the primary input of air-sea diffusion flux and the secondary input from rivers, wet deposition from air is the third one, supporting 8% (0.5 kg y<sup>-1</sup>) of the total input of CB-153 into the Seto Inland Sea.

The outflow flux of CB-153 from the Seto Inland Sea into the Pacific Ocean is 3.3 kg y<sup>-1</sup>, with the fluxes through the Bungo and Kii channels are 1.1 and 2.2 kg y<sup>-1</sup>, respectively. Its proportion to the total sink is 51%, suggesting that lateral transport to the open ocean is the primary removal pathway for CB-153 in the Seto Inland Sea. The sinking of detritus-bound CB-153 to the sea bottom is 1.7 kg y<sup>-1</sup>, being the secondary sink (27%). The degradation of the dissolved CB-153 is 1.0 kg y<sup>-1</sup>, which corresponds to 16% of the total sink, consistent with the situation in the global ocean (Wagner et al., 2019). The air–sea diffusion flux from the Seto Inland Sea to the above air (0.4 kg y<sup>-1</sup>) has a



Fig. 3. Horizontal distributions of vertically averaged concentrations of (a-d) dissolved and (e-h) particulate CB-153 in four representative months for the season.



**Fig. 4.** (a) Annual mass flow and balance of CB-153. Each arrow denotes a physical or biogeochemical process with a unit of kg  $y^{-1}$ . The values in parentheses represent the current stock of each state of CB-153 with a unit of kg. "Chs." means the outflow to the Pacific Ocean. (b) The ratios of each source to total input (positive bars) and those of each sink to total loss of dissolved CB-153 (negative bars).

proportion of 6% of the total sink, being the fourth sink for CB-153.

# 3.3. The reasons for the seasonal variations of dissolved and particulate CB-153 concentrations

As shown in Fig. 2, the concentrations of dissolved and particulate CB-153 in the Seto Inland Sea exhibit apparent seasonal variations. The

concentrations of CB-153 in seawater are affected by the physical processes at several boundaries and by the physical and biogeochemical processes inside the ocean. We integrated the fluxes of these processes over the model domain and analyzed their effects on the seasonal variations in dissolved and particulate CB-153 concentrations, respectively.

The net flux ( $F_{net}$ , g d<sup>-1</sup>), net physical flux ( $F_{phy}$ , g d<sup>-1</sup>), and net biogeochemical flux ( $F_{bio}$ , g d<sup>-1</sup>) for dissolved (\* = dis) and particulate

(\* = par) CB-153 are defined in Eqs. (9)–(14).

Net flux of dissolved 
$$CB - 153 F_{dis\_net} = F_{dis\_phy} + F_{dis\_bio}$$
 (9)

Net flux of particulate 
$$CB - 153 F_{par_net} = F_{par_phy} + F_{par_bio}$$
 (10)

Net physical flux of dissolved CB - 153 
$$F_{dis_phy} = F_{as} + F_{riv} + F_{wp} - F_{dis_bd}$$
(11)

Net biogeochemical flux of dissolved CB – 153  $F_{dis\_bio} = -F_{nu} + F_{rem} - F_{deg}$ (12)

Net physical flux of particulate 
$$CB - 153 F_{par_phy} = -F_{sk} - F_{par}$$
 (13)

Net biogeochemical flux of particulate  $CB - 153 F_{par_{bio}} = F_{nu} - F_{rem}$  (14)

Here,  $F_{as}$ ,  $F_{riv}$ ,  $F_{wp}$ ,  $F_{bd}$ , and  $F_{sk}$  are the physical fluxes for net air–sea diffusion (sum of air to sea diffusion and sea to air diffusion), river input, wet deposition, lateral boundary (sum of the Bungo and Kii channels), and detritus sink, respectively. The  $F_{nub}$ ,  $F_{rem}$ , and  $F_{deg}$  are the biogeochemical fluxes corresponding to net adsorption by phytoplankton, detritus decomposition, and self-degradation of dissolved CB-153, respectively. The unit of these fluxes is g d<sup>-1</sup>.

Fig. 5a shows the seasonal variations of net flux, net physical flux, and net biogeochemical flux of dissolved CB-153 in the Seto Inland Sea. For dissolved CB-153, the physical processes are the source, while biogeochemical processes are the sink.  $F_{dis_net}$  is positive until August, indicating an increase in dissolved CB-153 concentration in the sea. During this period,  $F_{dis_nph}$  has a larger magnitude than  $F_{dis_bio}$ . As shown in Fig. S3a, air–sea diffusion flux is a primary contributor to  $F_{dis_nph}$ , and it displays two positive peaks in March and July, respectively, which is controlled by  $K_{as}$ , gaseous CB-153 concentration, and H', as shown in Eq. (7). The river input flux of CB-153 is the secondary contributor to  $F_{dis_nph}$ , exhibiting its highest value in July (Fig. S3a).

After August, with the decrease of  $F_{dis,phy}$ ,  $F_{dis,net}$  becomes negative values, indicating a decrease of dissolved CB-153 concentration.  $F_{dis,phy}$  approaches zero after August, indicating a state of dynamic equilibrium. As shown in Fig. S3a, such equilibrium is archived by the input of CB-153 from the atmosphere and rivers and the output from lateral boundaries. The lateral boundary flux has significantly increased since August, particularly in the Kii Channel (Fig. S3a). Interestingly, the peak flux in the Kii Channel occurs with a time lag of nearly 1 month as compared to the peak of river input flux, which is consistent with the age

of Yodo River water at the Kii Channel (=30 days, Wang et al., 2019). Therefore, a major part of CB-153 through the Kii Channel likely comes from the Yodo River. The air–sea diffusion flux input decreases significantly after August (Fig. S3a). At the end of the year, the Seto Inland Sea even plays a role in emissions of CB-153 into the atmosphere. Notably, the air–sea diffusion flux exhibits upward values throughout the year in the northeastern of Osaka Bay (Fig. S5). Yodo River carries a large amount of dissolved CB-153 into Osaka Bay, resulting in a supersaturation of dissolved CB-153 at the surface around the estuary. Consequently, Yodo estuary acts as a continuous source of CB-153 released to the atmosphere throughout the year. As a sink,  $F_{dis, bio}$  is always negative throughout the year, and the phytoplankton adsorption is the major reason for its seasonal variation (Fig. S3b).

Fig. 5b shows the seasonal variations of net flux, net physical flux, and net biogeochemical flux of particulate CB-153 in the Seto Inland Sea. For particulate CB-153, physical processes act as a sink, while biogeochemical processes act as a source. Fpar, net is positive from January to April, making the concentration of particulate CB-153 peaks around April (Fig. 2). During this period, the phytoplankton bloom increases the phytoplankton uptake flux (Fig. S4b, Fig. S6). From April to June, the concentration of particulate CB-153 is declining because the sinking flux keeps a high value, resulting in  $F_{par_net}$  becoming negative. Phytoplankton mortality transformed phytoplankton-bound into detritusbound CB-153, which sinks to the sea bottom. The peak in particulate CB-153 concentration around July is attributed to a brief positive value of Fparnet. During this period, a large amount of dissolved CB-153 is adsorbed to the phytoplankton (Fig. 2, Fig. S4b), and subsequently, phytoplankton-bound CB-153 is converted to detritus-bound through the mortality process. After August, the negative value of  $F_{par net}$  makes the particulate CB-153 concentration decline. During this period, Fpar.phy is mainly influenced by the sink flux. Meanwhile, the outflux through the lateral boundaries also begins increasing, especially in the Kii Channel (Fig. S4a).

# 3.4. Response to atmospheric and riverine CB-153 concentrations

Air–sea diffusion and river input are the two primary inputs that bring over 90% of CB-153 into the Seto Inland Sea. They are affected by atmospheric and riverine CB-153 concentrations. According to the monitoring by the Ministry of the Environment, Japan, both riverine and atmospheric concentrations of CB-153 around the Seto Inland Sea have declined by approximately 50% over the past decades. Therefore, we



Fig. 5. Time series of (a) dissolved and (b) particulate CB-153 for net flux, net physical flux, and net biogeochemical flux in the Seto Inland Sea. Positive fluxes increase CB-153 concentration, while negative ones decrease CB-153 concentration.

aim to examine how the Seto Inland Sea responds to the decline of riverine and atmospheric CB-153 concentrations by conducting sensitivity experiments. We designed three experiments in each of the scenarios for the atmospheric concentration, riverine concentration, and their combination. In three experiments, we reduced the atmospheric CB-153 concentration, the riverine CB-153 concentration, and both by 25%, 50%, and 75%, respectively, but maintained the same seasonal variations as them in the original calculation (hereafter, refer as to control experiment).

# 3.4.1. Response of dissolved and particulate CB-153 concentrations

We calculated the reduction ratio, i.e.,  $(C_{control-exp} - C_{scen-exp})/C_{control-exp}$  to assess the responses of dissolved and particulate CB-153 concentrations in the Seto Inland Sea. Here,  $C_{control-exp}$  represents the CB-153 concentration in the control experiment, and  $C_{scen-exp}$  represents the CB-153 concentration in the experiments of three scenarios.

Because the seasonal variations of dissolved CB-153 for the three scenarios exhibited similar patterns to those given in the control experiment (Fig. S7), we present the annual mean concentration and its reduction for examining the responses (Table 1). The annual mean concentrations of total CB-153 concentrations in the Seto Inland Sea are 1.9, 1.4, and 1.0 ng m<sup>-3</sup> for experiments A-25, A-50, and A-75, respectively; are 2.3, 2.2, and 2.1 ng m<sup>-3</sup> for experiments R-25, R-50, and R-75 respectively; are 1.8, 1.3, and 0.7 ng m<sup>-3</sup> for experiments AR-25, AR-50, and AR-75, respectively. The CB-153 decreased more in the atmospheric scenario than in the riverine scenario, which is due to the larger CB-153 input from the atmosphere than from rivers in the control experiment (Fig. 4b). Furthermore, the reduction of the combination scenario is almost the sum of the reduction ratios of the atmospheric and riverine scenarios (Table 1).

Fig. 6 shows the horizontal distribution of the reduction ratio for annual mean concentration of total CB-153. In the atmospheric scenario, total CB-153 concentration shows a significant reduction in most regions of the Seto Inland Sea except for Osaka Bay (Fig. 6a, b, &c), indicating that the air–sea diffusion input dominates these areas. In the riverine scenario, the total CB-153 concentration shows decreased apparent in Osaka Bay (Fig. 6d, e, &f), indicating a strong control of river input on the CB-153 concentration there. In the combination scenario, there is little spatial variations in the reduction ratio (Fig. 6g, h, &i). The comparison of different scenarios but with the same reduction ratio in Table 1 tells us that the atmospheric scenario explains about 80% of the reduction in the combination scenario, and the riverine scenario explains the other 20%.

#### 3.4.2. Response of air-sea diffusion flux of CB-153

In the control experiment, the concentration of dissolved CB-153 in the western part of the Seto Inland Sea is always lower than  $C_a/H'$ , resulting air–sea diffusion flux from air to sea (Fig. S6). In the atmospheric scenario, when the atmospheric CB-153 concentration decreases while the riverine input is kept, the downward air–sea diffusion flux

# Table 1

Reduction ratio of annual mean dissolved, particulate, and total CB-153 in three experiments for the atmospheric scenario, riverine scenario, and their combination.

Scenario	Reduction ratio (%)	Experiment name	Reduction ratio (%)		
			Dissolved	Particulate	Total
Atmospheric scenario	25	A-25	19.1	16.7	18.6
	50	A-50	38.1	33.3	37.1
	75	A-75	57.1	49.9	55.6
Riverine scenario	25	R-25	4.2	7.0	4.8
	50	R-50	8.3	13.9	9.5
	75	R-75	12.5	20.8	14.3
Combination scenario	25	AR-25	23.2	23.6	23.4
	50	AR-50	46.5	47.2	46.7
	75	AR-75	69.7	70.8	70.0

presents an apparent reduction in the western areas of the Seto Inland Sea (Fig. 7a, b, &c), indicating that the replenishment of the atmosphere is decreasing. Therefore, dissolved CB-153 concentration is reduced apparently in these areas (Fig. S8a, b, &c). In the riverine scenario, since  $C_a$  is not changed,  $C_{ws}$  will decrease with the reduction of CB-153 concentration in rivers, the term of  $(C_a/H - C_{ws})$  in eq. (7) increases and leads to an increase in air–sea diffusion flux from air to sea (Fig. 7d, e, &f). The increased air to sea flux of CB-153 can partly offset the reduction of CB-153 in the sea due to the reduction of riverine input of CB-153 in the western part of the Seto Inland Sea, where the surface dissolved CB-153 concentration in the river scenario as high as that in the control experiment in these areas (Fig. S8d, e, &f).

In Osaka Bay, the high river input flux making  $C_{ws}$  in Osaka Bay exceeds the  $C_a/H$ , inducing a negative air–sea diffusion flux of CB-153 in the control experiment (Fig. S6). In the atmospheric scenario, the upward air–sea diffusion flux in Osaka Bay extends to the Harima-Nada and Kii Channel (Fig. 7a, b, &c) with the reduction of atmospheric concentration of CB-153, where the surface dissolved CB-153 concentration exhibits a small reduction (Fig. S8a, b, &c). In the riverine scenario, the surface concentration of CB-153 in Osaka Bay decreases significantly (Fig. S8d, e, &f), because of the reduction of river input flux of CB-153 and the area with negative air–sea diffusion fluxes (Fig. 7d, e, &f).

# 3.4.3. Different reductions of dissolved and particulate CB-153 concentrations

The dissolved and particulate CB-153 concentrations show different reductions in these scenarios (Table 1). In the atmospheric scenario, the particulate CB-153 presents a smaller reduction than dissolved CB-153; in the riverine scenario, the particulate CB-153 shows a larger reduction than the dissolved CB-153. To understand this difference, we define a transformation efficiency (E) between the dissolved and particulate CB-153.

$$E = \frac{C_{wp} + C_{wd}}{C_w} \tag{15}$$

The *E* of control experiment is greater than one around the Yodo River estuary and decreases gradually in the offshore direction, and *E* of other areas of the Seto Inland Sea lower than one (Fig. S10a). The spatial of *E* is mainly caused by the phytoplankton uptake flux (Fig. S11). *E* displays a similar distribution in the experiments for the atmospheric and riverine scenarios with the control experiment (Fig. S10). The low value of *E* in the Seto Inland Sea, except for Osaka Bay, determines that the concentration of particulate CB-153 did not decrease as significantly as the concentration of dissolved CB-153 in the atmospheric scenario (Fig. S9a, b, &c). The high value of *E* in Osaka Bay relies on the high phytoplankton uptake flux (Fig. S11), which determines that the concentration of dissolved CB-153 in the riverine scenario (Fig. S9d, e, &f).

We calculated the difference (*Diff*) of *E* between the experiments for the atmospheric and riverine scenarios and the control experiment to evaluate *E*. *Diff* for all experiments in the atmospheric and riverine scenarios exhibit larger values in Osaka Bay than other areas in the Seto Inland Sea (Fig. 8, Fig. S12). Positive *Diff* in the atmospheric scenario indicates the increases in the transformation efficiency from dissolved to particulate CB-153 as compared to the control experiment (Fig. 8). Conversely, *Diff* in the river scenario is negative, which means that the decreases in the transformation efficiency from dissolved to particulate CB-153 as compared to the control experiment (Fig. 8). Summarily, reductions in CB-153 concentrations originating from atmospheric or riverine sources influence dissolved CB-153 through air–sea diffusion and river input flux, thereby regulating the partitioning efficiency of dissolved and particulate CB-153 in the Seto Inland Sea.



Fig. 6. Horizontal distributions of reduction ratio for annual mean concentration of total CB-153 in (a–c) atmospheric scenario, (d–f) riverine scenario, and of (g–i) their combination.



Fig. 7. Horizontal distributions of air-sea diffusion flux for the (a-c) atmospheric scenario and (d-f) riverine scenario. Positive values mean the input of CB-153 from air to sea, while negative values mean an emission of CB-153 from sea to air.

# 4. Conclusions

This study established a three-dimensional hydrodynamicecosystem-POP coupled model in the Seto Inland Sea to understand the spatial and seasonal variations of CB-153 and related physical and biogeochemical processes. The concentration of dissolved CB-153 peaked in July, while the particulate CB-153 concentration peaked in both April and July. The maximum CB-153 concentration appeared in Osaka Bay with a value of 76.6 ng m<sup>-3</sup> which is at the same order as those found in the Mediterranean Sea, North Sea, Baltic Sea, and South China Sea.

Over a time scale of one year, the model results indicates that the CB-153 in the Seto Inland Sea is supplied by the air–sea diffusion (61%), river inputs (31%), and wet deposition (8%). 51% of CB-153 in the Seto Inland Sea outflows to the Pacific Ocean, 27% accumulates at the sea bottom, and 16% of CB-153 is self-degraded. The seasonal variations in air–sea diffusion flux and river input of CB-153 strongly influence the seasonal variation of dissolved CB-153 in the sea. The air–sea diffusion flux is a major source of CB-153 in many areas except for Osaka Bay where Yodo River delivers a large amount of CB-153 into the bay and Osaka Bay becomes an area releasing CB-153 to the atmosphere. Biogeochemical processes influence the seasonal variation of particulate CB-153, which peaks in April due to the increase of phytoplankton biomass, and another one in July resulting from the high concentration of dissolved CB-153 in the sea.

The sensitivity experiments demonstrated that the air–sea diffusion process dominates the input of CB-153 in most regions of the Seto Inland Sea except for Osaka Bay, where river input of CB-153 is the dominant factor. Dissolved and particulate CB-153 concentrations exhibited different responses to the reduction in the atmospheric and riverine concentrations of CB-153. In particular, dissolved CB-153 is more sensitive than particulate CB-153 to the decreases of atmospheric CB-153 concentration, but particulate CB-153 is more sensitive than dissolved CB-153 to the decreases of riverine CB-153 concentration. This difference is attributed to the more efficient transformation of CB-153 from dissolved phase to particulate phase for CB-153 from the atmosphere than that from rivers.

The present study focused on the spatial and seasonal variations in climatological CB-153 concentrations, which is a foundational step for understanding the transport and cycle of CB-153 in the shallow sea. However, the behaviors of suspended particulate matter such as resuspension from sediments are likely an important to the transport and cycle of CB-153 in shallow seas. Our future modeling will incorporate the behavior of suspended particulate matter and its interaction with

#### Y. Li et al.



**Fig. 8.** Change of mean value of *Diff* over entire the Seto Inland Sea under two scenarios. The x-axis represents the reduction ratio of atmospheric or riverine concentrations.

#### CB-153.

#### CRediT authorship contribution statement

Yaxian Li: Writing – review & editing, Writing – original draft, Validation, Methodology, Formal analysis, Data curation, Conceptualization. Xinyu Guo: Writing – review & editing, Supervision, Methodology, Funding acquisition, Formal analysis, Conceptualization. Qian Leng: Data curation. Min Yang: Data curation. Yu Bai: Writing – review & editing, Xueting Zhao: Writing – review & editing.

#### Declaration of competing interest

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

### Acknowledgements

This research was supported by the Moonshot Research and Development Program (Grant no. JPNP18016), and the New Energy and Industrial Technology Development Organization (NEDO).

# Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envres.2025.122000.

## Data availability

Data will be made available on request.

# References

- Alekseenko, E., Thouvenin, B., Tronczyński, J., Carlotti, F., Garreau, P., Tixier, C., Baklouti, M., 2018. Modeling of PCB trophic transfer in the Gulf of Lions; 3D coupled model application. Mar. Pollut. Bull. 128, 140–155. https://doi.org/10.1016/j. marpolbul.2018.01.008.
- Berrojalbiz, N., Castro-Jiménez, J., Mariani, G., Wollgast, J., Hanke, G., Dachs, J., 2014. Atmospheric occurrence, transport and deposition of polychlorinated biphenyls and

hexachlorobenzene in the Mediterranean and Black seas. Atmos. Chem. Phys. 14, 8947–8959. https://doi.org/10.5194/acp-14-8947-2014.

- Bidleman, T.F., McConnell, L.L., 1995. A review of field experiments to determine airwater gas exchange of persistent organic pollutants. Sci. Total Environ. 159, 101–117. https://doi.org/10.1016/0048-9697(95)04255-Y.
- Blumberg, A.F., Mellor, G.L., 1987. A description of a three-dimensional coastal ocean circulation model. three-dimensional Coast. Ocean Model. (Oxf.) 4, 1–16. https:// doi.org/10.1029/CO004p0001.
- Breivik, K., Sweetman, A., Pacyna, J.M., Jones, K.C., 2007. Towards a global historical emission inventory for selected PCB congeners — a mass balance approach: 3. An update. Sci. Total Environ. 377, 296–307. https://doi.org/10.1016/j. scitotenv.2007.02.026.
- Carpenter, D.O., 2006. Polychlorinated biphenyls (PCBs): routes of exposure and effects on human health. Rev. Environ. Health 21, 1–24. https://doi.org/10.1515/ REVEH.2006.21.1.1.
- Cetin, B., Odabasi, M., 2005. Measurement of Henry's law constants of seven polybrominated diphenyl ether (PBDE) congeners as a function of temperature. Atmos. Environ. 39, 5273–5280. https://doi.org/10.1016/j.atmosenv.2005.05.029.
- Chang, P.-H., Guo, X., Takeoka, H., 2009. A numerical study of the seasonal circulation in the Seto Inland Sea, Japan. J. Oceanogr. 65, 721–736. https://doi.org/10.1007/ s10872-009-0062-4.
- Dachs, J., Eisenreich, S.J., Hoff, R.M., 2000. Influence of eutrophication on air–water exchange, vertical fluxes, and phytoplankton concentrations of persistent organic pollutants. Environ. Sci. Technol. 34, 1095–1102. https://doi.org/10.1021/ es900759e.
- Dachs, J., Lohmann, R., Ockenden, W.A., Méjanelle, L., Eisenreich, S.J., Jones, K.C., 2002. Oceanic biogeochemical controls on global dynamics of persistent organic pollutants. Environ. Sci. Technol. 36, 4229–4237. https://doi.org/10.1021/ es025724k.
- Del Vento, S., Dachs, J., 2002. Prediction of uptake dynamics of persistent organic pollutants by bacteria and phytoplankton. Environ. Toxicol. Chem. 21, 2099–2107. https://doi.org/10.1002/etc.5620211013.
- Endo, S., Takizawa, R., Okuda, K., Takada, H., Chiba, K., Kanehiro, H., Ogi, H., Yamashita, R., Date, T., 2005. Concentration of polychlorinated biphenyls (PCBs) in beached resin pellets: variability among individual particles and regional differences. Mar. Pollut. Bull. 50, 1103–1114. https://doi.org/10.1016/j. marpolbul.2005.04.030.
- Friedman, C.L., Selin, N.E., 2016. PCBs in the arctic atmosphere: determining important driving forces using a global atmospheric transport model. Atmos. Chem. Phys. 16, 3433–3448. https://doi.org/10.5194/acp-16-3433-2016.
- Gioia, R., Li, J., Schuster, J., Zhang, Y., Zhang, G., Li, X., Spiro, B., Bhatia, R.S., Dachs, J., Jones, K.C., 2012. Factors affecting the occurrence and transport of atmospheric organochlorines in the China Sea and the Northern Indian and South East Atlantic Oceans. Environ. Sci. Technol. 46, 10012–10021. https://doi.org/10.1021/ es302037t. 2012.
- Ilyina, T., Pohlmann, T., Lammel, G., Sündermann, J., 2006. A fate and transport ocean model for persistent organic pollutants and its application to the North Sea. J. Mar. Syst. 63, 1–19. https://doi.org/10.1016/j.jmarsys.2006.04.007.
- Isobe, T., Oshihoi, T., Hamada, H., Nakayama, K., Yamada, T.K., Tajima, Y., Amano, M., Tanabe, S., 2011. Contamination status of POPs and BFRs and relationship with parasitic infection in finless porpoises (Neophocaena phocaenoides) from Seto Inland Sea and Omura Bay, Japan. Mar. Pollut. Bull. 63, 564–571. https://doi.org/ 10.1016/j.marpoibul.2011.01.014.
- Iwata, H., Tanabe, S., Sakai, N., Tatsukawa, R., 1993. Distribution of persistent organochlorines in the oceanic air and surface seawater and the role of ocean on their global transport and fate. Environ. Sci. Technol. 27, 1080–1098. https://doi. org/10.1021/es00043a007.
- Jönsson, A., Gustafsson, Ö., Axelman, J., Sundberg, H., 2003. Global accounting of PCBs in the continental shelf sediments. Environ. Sci. Technol. 37, 245–255. https://doi. org/10.1021/es0201404.
- Jurado, E., Jaward, F., Lohmann, R., Jones, K.C., Simó, R., Dachs, J., 2005. Wet deposition of persistent organic pollutants to the Global Oceans. Environ. Sci. Technol. 39, 2426–2435. https://doi.org/10.1021/es048599g.
- Jurado, E., Jaward, F., Lohmann, R., Jones, K.C., Simó, R., Dachs, J., 2004a. Atmospheric dry deposition of persistent organic pollutants to the Atlantic and inferences for the Global Oceans. Environ. Sci. Technol. 38, 5505–5513. https://doi.org/10.1021/ es049240v.
- Jurado, E., Lohmann, R., Meijer, S., Jones, K.C., Dachs, J., 2004b. Latitudinal and seasonal capacity of the surface oceans as a reservoir of polychlorinated biphenyls. Environ. Pollut. 128, 149–162. https://doi.org/10.1016/j.envpol.2003.08.039.
- Kames, J., Schurath, U., 1992. Alkyl nitrates and bifunctional nitrates of atmospheric interes: Henry's law constants and their temperature dependencies. J. Atmos. Chem. 15, 79–95. https://doi.org/10.1007/BF00053611.
- Leng, Q., Guo, X., Zhu, J., Morimoto, A., 2023. Contribution of the open ocean to the nutrient and phytoplankton inventory in a semi-enclosed coastal sea. Biogeosciences 20, 4323–4338. https://doi.org/10.5194/bg-20-4323-2023.
- Lohmann, R., Dachs, J., 2019. Chapter 15 polychlorinated biphenyls in the global Ocean. In: Sheppard, C. (Ed.), World Seas: an Environmental Evaluation, second ed. Academic Press, pp. 269–282. https://doi.org/10.1016/B978-0-12-805052-1.00017-6.
- Madgett, A.S., Yates, K., Webster, L., McKenzie, C., Brownlow, A., Moffat, C.F., 2022. The concentration and biomagnification of PCBs and PBDEs across four trophic levels in a marine food web. Environ. Pollut. 309, 119752. https://doi.org/10.1016/j. envpol.2022.119752.
- Matsumoto, R., Tu, N.P.C., Haruta, S., Kawano, M., Takeuchi, I., 2016. Analysis of all 209 polychlorinated biphenyl (PCB) congeners (with special reference to dioxin-like PCB

Y. Li et al.

congeners) in Japanese seabass and related species by high-resolution gas chromatography/high-resolution mass spectrometry (HRGC/HRMS). Reg. Stud. Mar. Sci. 3, 119–130. https://doi.org/10.1016/j.rsma.2015.06.004.

- Meijer, S.N., Ockenden, W.A., Sweetman, A., Breivik, K., Grimalt, J.O., Jones, K.C., 2003. Global distribution and budget of PCBs and HCB in background surface soils: implications for sources and environmental processes. Environ. Sci. Technol. 37, 667–672. https://doi.org/10.1021/es0258091.
- Nizzetto, L., Gioia, R., Li, J., Borgå, K., Pomati, F., Bettinetti, R., Dachs, J., Jones, K.C., 2012. Biological pump control of the fate and distribution of hydrophobic organic pollutants in water and plankton. Environ. Sci. Technol. 46, 3204–3211. https://doi. org/10.1021/es2041760.
- Nizzetto, L., Macleod, M., Dorgå, K., Cabrerizo, A., Dachs, J., Guardo, A.D., Ghirardello, D., Hansen, K.M., Jarvis, A., Lindroth, A., Ludwig, B., 2010. Past, present, and future controls on levels of persistent organic pollutants in the global environment. Environ. Sci. Technol. 44, 6526–6531. https://doi.org/10.1021/ es100178f.
- O'Driscoll, K., Mayer, B., Ilyina, T., Pohlmann, T., 2013. Modelling the cycling of persistent organic pollutants (POPs) in the North Sea system: fluxes, loading, seasonality, trends. J. Mar. Syst. 111–112, 69–82. https://doi.org/10.1016/j. jmarsys.2012.09.011.
- Ono, J., Takahashi, D., Guo, X., Takahashi, S., Takeoka, H., 2012. A numerical study on the seasonal variability of polychlorinated biphenyls from the atmosphere in the East China Sea. Chemosphere 89, 389–397. https://doi.org/10.1016/j. chemosphere.2012.05.049.
- Pavlova, P.A., Schmid, P., Bogdal, C., Steinlin, C., Jenk, T.M., Schwikowski, M., 2014. Polychlorinated biphenyls in glaciers. 1. Deposition history from an alpine ice core. Environ. Sci. Technol. 48, 7842–7848. https://doi.org/10.1021/es5017922.
- Schuster, J.K., Gioia, R., Breivik, K., Steinnes, E., Scheringer, M., Jones, K.C., 2010. Trends in European background air reflect reductions in primary emissions of PCBs and PBDEs. Environ. Sci. Technol. 44, 6760–6766. https://doi.org/10.1021/ es070870h.
- Sobek, A., Sundqvist, K.L., Assefa, A.T., Wiberg, K., 2015. Baltic Sea sediment records: unlikely near-future declines in PCBs and HCB. Sci. Total Environ. 518–519, 8–15. https://doi.org/10.1016/j.scitotenv.2015.02.093.
- Totten, L., Gigliotti, C., Van, D., Offenberg, J., Nelson, E., Dachs, J., Reinfelder, J., Eisenreich, S., 2004. Atmospheric Concentrations and Deposition of Polychorinated

Biphenyls to the Hudson River Estuary. Environ. Sci. Technol. 38, 2568–2573. https://doi.org/10.1021/es034878c.

- Tsuno, H., Shinkai, T., Nakano, T., Nagare, H., 2007. Composition properties and origin presumption of PCBs in Seto Inland Sea. J. Jpn. Soc. Water Environ. 30, 457–462. https://doi.org/10.2965/jswe.30.457.
- Ueno, D., Isobe, T., Ramu, K., Tanabe, S., Alaee, M., Marvin, C., Inoue, K., Someya, T., Miyajima, T., Kodama, H., Nakata, H., 2010. Spatial distribution of hexabromocyclododecanes (HBCDs), polybrominated diphenyl ethers (PBDEs) and organochlorines in bivalves from Japanese coastal waters. Chemosphere 78, 1213–1219. https://doi.org/10.1016/j.chemosphere.2009.12.058.
- Wagner, C.C., Amos, H.M., Thackray, C.P., Zhang, Y., Lundgren, E.W., Forget, G., Friedman, C.L., Selin, N.E., Lohmann, R., Sunderland, E.M., 2019. A global 3-D ocean model for PCBs: benchmark compounds for understanding the impacts of global change on neutral persistent organic pollutants. Glob. Biogeochem. Cycles 33, 469–481. https://doi.org/10.1029/2018GB006018.
- Wang, A., Guo, X., Ding, X., Shi, J., Tang, J., 2024. Effect of hydrodynamic and ecosystem conditions on persistent organic pollutant temporal-spatial variations in the Yellow Sea. J. Hazard. Mater. 469, 134051. https://doi.org/10.1016/j. jhazmat.2024.134051.
- Wang, H., Guo, X., Liu, Z., 2019. The age of Yodo River water in the Seto Inland Sea. J. Mar. Syst. 191, 24–37. https://doi.org/10.1016/j.jmarsys.2018.12.001.
- Wania, F., Axelman, J., Broman, D., 1998. A review of processes involved in the exchange of persistent organic pollutants across the air-sea interface. Environ. Pollut. 102, 3–23. https://doi.org/10.1016/S0269-7491(98)00072-4.
- Yang, M., Guo, X., Ishizu, M., Miyazawa, Y., 2022. The Kuroshio regulates the air-sea exchange of PCBs in the Northwestern Pacific Ocean. Environ. Sci. Technol. 56, 12307–12314. https://doi.org/10.1021/acs.est.2c03459.
- Yang, M., Guo, X., Zheng, J., Miyazawa, Y., 2024. North-south discrepancy in the contributors to CB153 accumulation in the deep water of the Sea of Japan. Total Environ. 939, 173599. https://doi.org/10.1016/j.scitotenv.2024.173599.
- Zhang, L., Lohmann, R., 2010. Cycling of PCBs and HCB in the surface ocean-lower atmosphere of the open Pacific. Environ. Sci. Technol. 44, 3832–3838. https://doi. org/10.1021/es9039852.
- Zhu, J., Guo, X., Shi, J., Gao, H., 2019. Dilution characteristics of riverine input contaminants in the Seto Inland Sea. Mar. Pollut. Bull. 141, 91–103. https://doi.org/ 10.1016/j.marpolbul.2019.02.029.