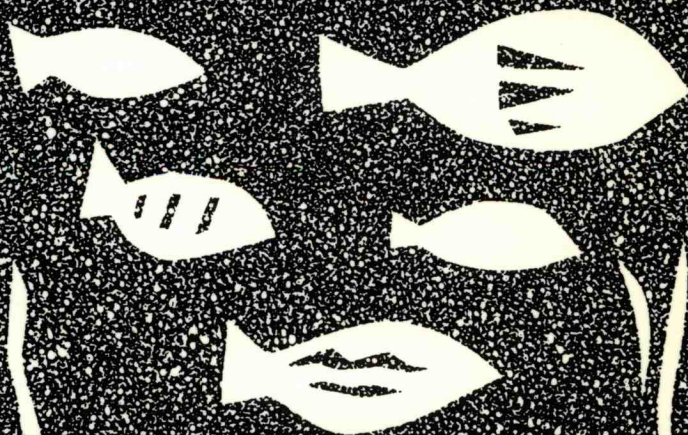
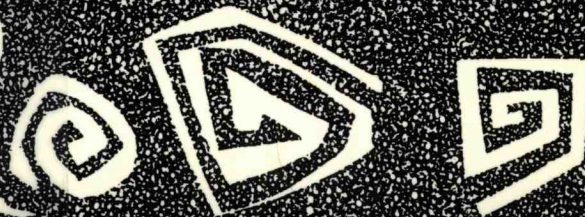


Marine Ecosystem Enclosed Experiments

Proceedings of a symposium held
in Beijing, People's Republic
of China, 9-14 May 1987



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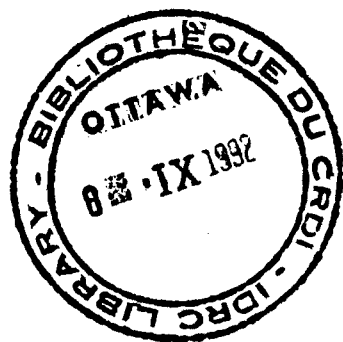


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Marine Ecosystem Enclosed Experiments

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Editor: C.S. Wong and P.J. Harrison



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Abstract

This symposium on marine ecosystem enclosed experiments (MEEE) consists of nine review papers that describe various types of ecosystem enclosures and a series of papers resulting from enclosure experiments in Xiamen, People's Republic of China, and Saanich Inlet, BC, Canada. The reviews on types of enclosures include benthic enclosures for rocky and sandy shores and the effects of pollutants (primarily hydrocarbons) on bacteria, macroalgae, and invertebrates. The pelagic enclosures were used to study the control of phytoplankton blooms, the uptake and release of dissolved organic substances, and the effects of pesticides on freshwater ecosystems.

Six enclosure experiments were conducted in China and Canada from 1986–87. Some of these experiments examined the effects of contaminated sediments, primarily heavy metals, on bacteria, phytoplankton, and zooplankton and the pathways and fates of these heavy metals in the seawater. Other experiments studied the chemistry and biological effects of chemically dispersed oil.

Résumé

Ce compte rendu du symposium sur les expériences faites en écosystèmes marins comprend neuf communications qui décrivent les écosystèmes retenus et les expériences faites à Xiamen en République populaire de Chine et à Saanich Inlet, C.-B., au Canada. Les communications portent, notamment, sur les écosystèmes benthiques des littoraux rocheux et sablonneux et sur les effets des polluants (surtout les hydrocarbures) sur les bactéries, les grandes algues et les invertébrés. Les expériences sur le contrôle des brutales pullulations ("blooms") du phytoplancton furent menées dans les écosystèmes pélagiques, ainsi que l'absorption et le dégagement des substances organiques dissoutes et les effets des pesticides sur les écosystèmes d'eau douce.

Six expériences ont été faites en Chine et au Canada entre 1983 et 1987. Certaines ont porté sur les effets des sédiments contaminés, principalement par des métaux lourds, sur les bactéries, le phytoplancton et le zooplancton et sur le cheminement et le sort de ces métaux lourds dans l'eau salée. D'autres expériences portaient sur la chimie et les effets biologiques du pétrole dispersé chimiquement.

Resumen

Este simposio sobre Experimentos Marinos en Ecosistemas Cerrados (MEEE) consistió en nueve trabajos de análisis que describen varios tipos de enclaustramientos ecosistémicos y una serie de trabajos derivados de experimentos con estos enclaustramientos en Xiamen, República Popular de China, y en Saanich Inlet, Canadá. Los estudios incluyen enclaustramientos bentónicos para costas rocosas y arenosas, y los efectos de los contaminantes (fundamentalmente hidrocarburos) sobre bacterias, macroalgas e invertebrados. Los enclaustramientos pelágicos se utilizaron para estudiar el control de la reproducción del fitoplancton, la ingestión y expulsión de sustancias orgánicas disueltas y los efectos de pesticidas en los ecosistemas de agua dulce.

Se realizaron seis experimentos en ecosistemas cerrados en China y Canadá, de 1983 a 1987. Algunos de estos experimentos examinaron los efectos que ejercen los sedimentos contaminados, fundamentalmente los metales pesados, sobre bacterias, fitoplancton y zooplancton, y el ciclo y destino final de estos metales pesados en el agua de mar. Otros experimentos estudiaron los efectos químicos y biológicos de los aceites crudos dispersados por medios químicos.

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Biogeochemical Processes of Mercury in Marine Enclosures

Xu Kuncan, Wu Liqing, Zhen Changchun, and Zhan Xiumei

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This work is part of the enclosed ecosystem study carried out in Xiamen in 1985. Experimental results show that a strong affinity of Hg to particulate organic matter plays an important role in the removal of Hg in enclosed ecosystems. Distribution coefficients of Hg (K_d) in organic matter range from 2 to 15, with direct correlation with particulate organic carbon. Absorption of Hg on particulate organic matter can be described by Freundlich or linear isotherm absorption equations, or both, and the removal rate of Hg can be expressed by first-order equations. First-order removal constants in experimental bags M1, M2, and M3 were 0.050, 0.049, and 0.038; corresponding removal times were, 14, 14, and 18 d respectively.

Since the discovery of Minimata disease in Japan, the problem of Hg pollution in the ocean has received much attention. A wide range of programs have been developed to monitor and study marine Hg pollution. Many studies (Cranton and Buckley 1972; Fitzgerald and Lyons 1973; Wallace 1982; Wallace et al. 1982; Xu et al. 1985, 1986) indicate that the strong complexing of Hg to organic materials significantly affects its speciation, toxicity, and fate.

Experimental ecosystem enclosures have been used to study the biogeochemical processes of Hg (Santschi et al. 1980; Wallace 1982; Wallace et al. 1982). This project, as part of the Xiamen marine ecosystem enclosure experiment (MEEE) jointly sponsored by Canada and China, uses the same approach to study the effect of biogenic organic matter on the biogeochemical processes of Hg.

Methods

Enclosures and additions of Hg

The Hg experiment described in this paper was conducted in a large pool at the Third Institute of Oceanography, Xiamen (MEEE Group, this volume). Five experimental polyethylene bags, each 2 m in diameter \times 4.5 m long with a volume of about 10 m³, were filled with seawater pumped from 150 m offshore during high

tide. Nutrient supplements were added to each bag to produce a phosphate concentration of about $0.8 \mu\text{g}\cdot\text{L}^{-1}$ in the water column.

Two control bags were designated as C1 and C2. Another three bags M1, M2, and M3 were treated with HgCl_2 to produce a total Hg concentration of 185, 258, and $1\,060 \text{ ng}\cdot\text{L}^{-1}$ respectively. After the addition of Hg, the enclosed water was stirred thoroughly and sampling began 2 h later.

Sampling and analysis

Water samples were taken integrally using a peristaltic pump from the upper 3 m of the water column and placed into acid-cleaned polyethylene bags. Subsamples for Hg analyses were stored in acid-washed glass bottles. After each water sample was taken, settled materials were pumped from the bottom of the bags. Both the water and sediment samples were analyzed in the laboratory immediately.

Seawater samples, both unfiltered (for total Hg) and filtered through a $1.2\text{-}\mu\text{m}$ membrane (for dissolved Hg), were digested with a mixture of H_2SO_4 and $\text{K}_2\text{S}_2\text{O}_8$ at room temperature overnight. Mercury analyses were carried out the next day by differential cold vapour atomic absorption (Xu et al. 1983), with a detection limit of $1 \text{ ng}\cdot\text{L}^{-1}$. The concentration of particulate Hg was the difference between total and dissolved Hg.

The concentration of Hg in sediments, dried at low temperature and digested with a solution of H_2SO_4 and KMnO_4 in a 100°C water bath for 2 h, was determined using the same technique as mentioned above, but following the procedure described in a State Oceanic Administration manual (SOA 1979).

Particulate organic carbon was determined using a CHN analyzer (Perkin-Elmer Model 240C). A Turner fluorometer was used to determinate amounts of chlorophyll *a*.

Results and discussion

Temporal variation of chlorophyll *a*

The growth periods of phytoplankton bags M1 and M2 were similar (Fig. 1). During the first 5 d before the phytoplankton bloom, the chlorophyll *a* concentration was low. During the phytoplankton bloom, from days 5 to 11, there was a rapid rise in chlorophyll *a*, then a decline from days 11 to 17. The peak concentration in bag M2 ($26.2 \mu\text{g}\cdot\text{L}^{-1}$) was more than twice that in bag M1 ($11.0 \mu\text{g}\cdot\text{L}^{-1}$). In bag M3, the peak of the phytoplankton bloom was delayed 6 d appearing only on day 17. The first 5 d were again the prebloom period; the bloom occurred from days 5 to 17, and then a decline occurred from days 17 to 20. Although this bloom lasted for about 12 d, the peak concentration of chlorophyll *a* was only $15.1 \mu\text{g}\cdot\text{L}^{-1}$ (see Qian et al., this volume, for further details).

Temporal variations in particulate organic carbon

Particulate organic carbon (POC) concentration in bags M1, M2, and M3 changed greatly during the experiment (Fig. 2) and followed a trend similar to that

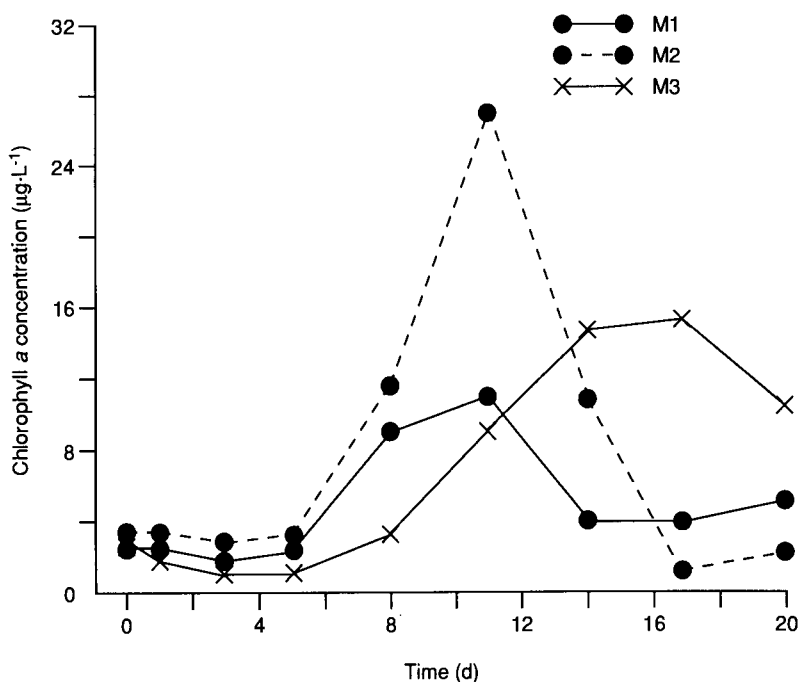


Fig. 1. Variations of chlorophyll *a* with time.

of chlorophyll *a*. Linear regressions of POC against chlorophyll *a* gave correlation coefficients of 0.95, 0.92, and 0.87 for bags M1, M2, and M3 respectively. These results suggest that living phytoplankton carbon accounted for the bulk of POC in the experiment.

Temporal variations in Hg

During the experiment, the total Hg concentration in control bags, C1 and C2, ranged from 1.4 to 4.3 ng·L⁻¹ (Table 1). Temporal variations in total, particulate, and dissolved Hg concentrations in the three spiked bags are shown in Fig. 3. The total Hg concentration in enclosures M1, M2, and M3 decreased throughout the 20-d experiment by 68, 62, and 57% respectively (Fig. 3).

Mercury in seawater might be removed by three possible pathways: adsorption on the bag wall, evaporative loss, and settling through adsorption on particles. Losses through adsorption on the bag wall and evaporation were not determined in this study as these losses were considered to be minor. Adsorption of Hg on sinking particles, on the other hand, was regarded as the most important pathway for Hg removal in the water column (Wallace et al. 1982). This was supported by the results of mass-balance calculations.

Comparing the amounts of Hg added with those amounts in the sediments and in the water, it was found that the amounts of Hg in sediments and in water were 85, 127, and 88%, i.e., a mean of $100 \pm 18\%$, of the known amounts added to bags M1, M2, and M3 respectively. It can be estimated that losses through adsorption on the

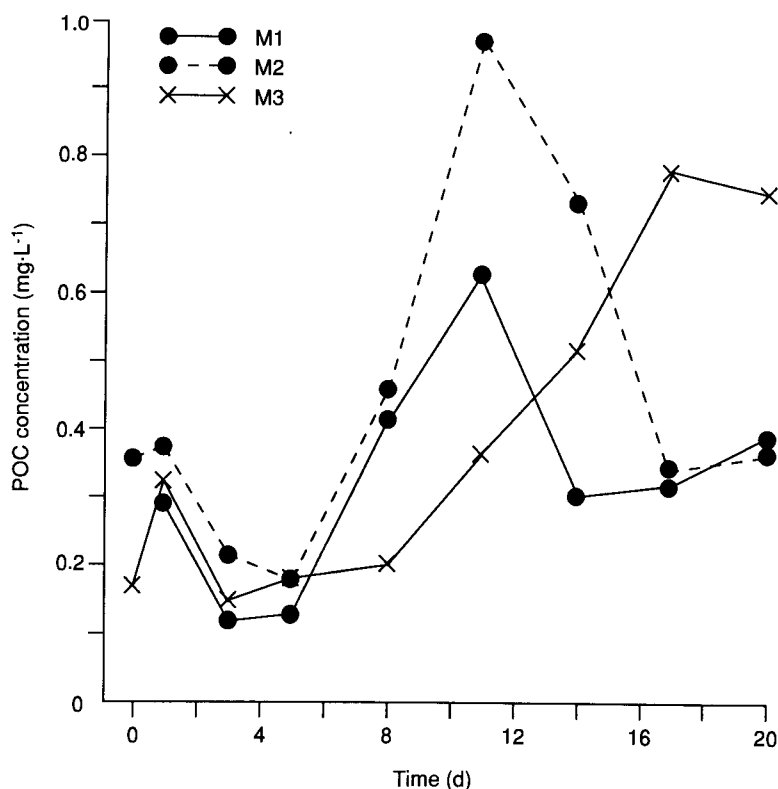


Fig. 2. Temporal variations of total, particulate, and dissolved Hg in (a) bag M1, (b) bag M2, and (c) bag M3.

bag wall and evaporation were less than 18%, although some analytical errors might exist in the results. Based on these results and the strong affinity of Hg to particulate matter, it can also be concluded that the settling of Hg through absorption on particles is the most important pathway for Hg removal from the water column.

Particulate forms of Hg always made up the major fraction of total Hg and the dissolved Hg fraction was minor (Fig. 3). In this experiment, 65% of the total Hg in the water column was readily adsorbed by particles 2 h after the addition of Hg. During the experiment, about 60–80% of the total Hg in the treated bags was in particulate form. The average percentage of 68% was slightly higher than the 53% reported by Wallace et al. (1982), providing evidence for the great affinity of settling particles to Hg.

Table 1. Total mercury concentration in the control bags.

Bag	Day							
	1	3	5	8	11	14	17	20
C1	3.1	2.6	3.3	1.4	1.7	1.8	2.7	2.5
C2	4.0	3.6	2.4	4.3	1.7	1.8	3.0	2.9

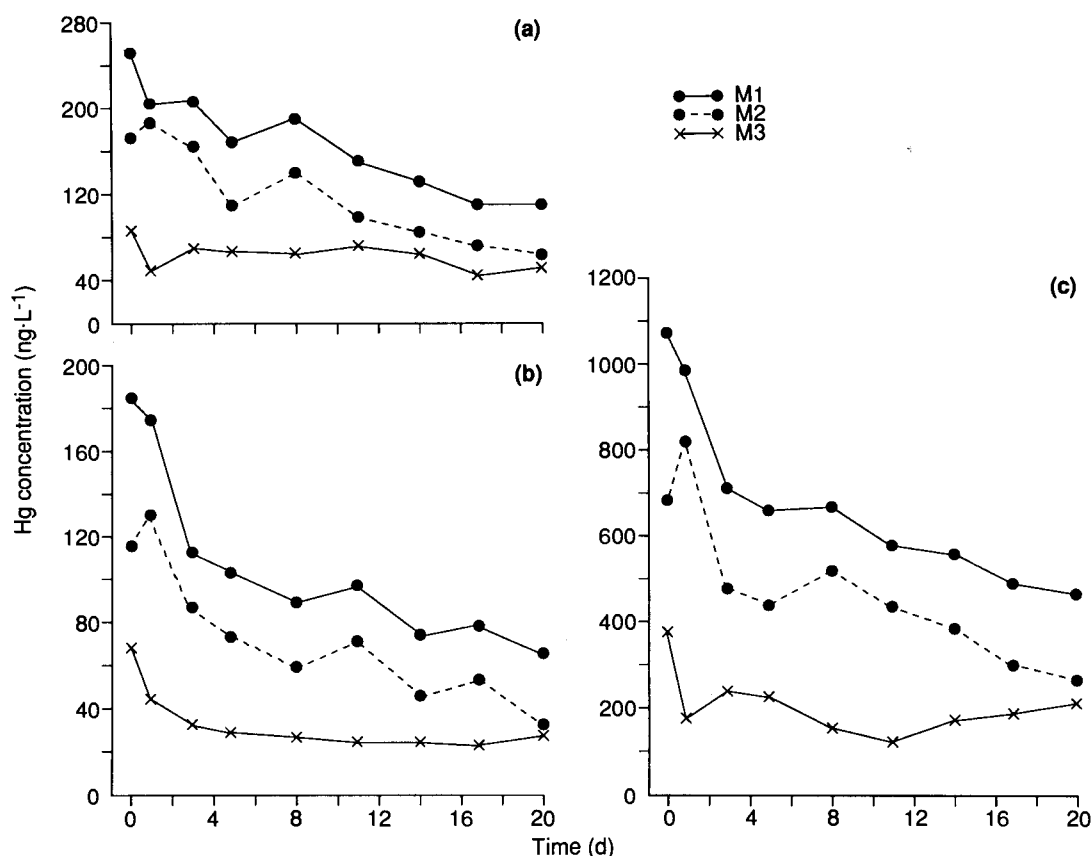


Fig. 3. Temporal variations of particulate (a), dissolved (b), and total (c) Hg in bags M1, M2, and M3.

Distribution coefficients and adsorption isotherms

The partition coefficient of Hg on particles is an important parameter used to gauge the bonding strength of particles with Hg. Olsen et al. (1982) pointed out that organic matter content in particles was a major controlling factor in the adsorption of metals, including Hg on particles. Wallace et al. (1982) used adsorption isotherms to describe the affinity of Hg for POC by normalizing particulate Hg concentrations to POC concentrations and determining the Hg distribution coefficients. The Hg distribution coefficients, K_d , can be represented by the following equation normalized to the concentration of POC:

$$[1] \quad K_d = (\text{Hg})_p \cdot (\text{POC})^{-1} \cdot (\text{Hg})_d^{-1}$$

where $(\text{Hg})_p$ is the concentration of particulate Hg ($\text{ng} \cdot \text{L}^{-1}$), POC, is the concentration particulate organic carbon ($\text{mg} \cdot \text{L}^{-1}$), and $(\text{Hg})_d$ is the concentration of dissolved Hg ($\text{ng} \cdot \text{L}^{-1}$).

For these particles in bags M1, M2, and M3, K_d values ranged between 15 early in the experiment to 2 by the end of the experiment, providing evidence for the great affinity of Hg to organic matter in settling particles. In general, values

Table 2. Distribution coefficients (K_d).

Bag	Day		
	1	11	20
M1	15	2.2	3.1
M2	7.6	2.8	3.5
M3	13	8.6	1.9

Table 3. Isothermal adsorption equations.

Bag	Type	Equation	r	n
M1	Linear	$Y = 2.75X - 41$	0.98	8
	Freundlich	$\log Y = 1.10 \log X + 0.14$	0.93	8
M2	Linear	$Y = 2.14X - 0.44$	0.91	9
	Freundlich	$\log Y = 1.20 \log X - 0.14$	0.92	9
M3	Linear	$Y = 1.74X + 496$	0.91	9
	Freundlich	$\log Y = 1.04 \log X + 0.24$	0.91	9

observed before the bloom were higher than those observed during the bloom, and K_d values correlated reciprocally with the concentration of POC (Table 2). Because POC correlates closely with chlorophyll a , the decrease in K_d values with increasing POC concentrations might be caused by the "biological dilution effect" of phytoplankton (Ibragim and Patin 1976; Xu et al. 1986).

Assuming that POC was used as the solid adsorbent and dissolved Hg as the solute, and that the adsorption of Hg by POC in the enclosures was in equilibrium under constant temperature ($19 \pm 2^\circ\text{C}$), isothermal adsorption equations could be established. Because POC concentrations varied during the experiment, both linear [2] and Freundlich [3] isotherm adsorption equations in the following forms were used to describe the adsorption of Hg by particulate organic matter (POM):

$$[2] \quad Y = aX + b$$

$$[3] \quad \log(Y) = \log(X)/n + k$$

in which $Y = (\text{Hg})_p/(\text{POC})$, and $X = (\text{Hg})_d/(\text{POC})$, where $(\text{Hg})_p$, $(\text{Hg})_d$, and POC are as defined for equation [1]. Separate equations for bags M1, M2, and M3 are given in Table 3.

Removal of Hg

Removal of Hg from the water body depends mainly on the adsorption of Hg on the particles and its subsequent sinking. Hence, the removal of Hg can be described by a first-order kinetic equation (Hesslein et al. 1980; Santschi et al. 1980; Wallace 1982; Wallace et al. 1982). The first-order removal rate is obtained using the equation:

$$[4] \quad \ln(\text{Hg}) = bt + a$$

where t is the time (d), (Hg) is the total concentration Hg (ng/L), and b is the total removal rate (d^{-1}).

Table 4. First-order removal rates and half-removal times of mercury.

Bag	Removal rate (b) (d^{-1})	$T_{0.5}$ (d)	r	n
M1	0.050	14	0.86	9
M2	0.049	14	0.95	9
M3	0.038	14	0.87	9

The half-removal time ($T_{0.5}$) is obtained using the equation:

$$[5] \quad T_{0.5} = \ln(2/b)$$

Table 4 gives the removal rates and half-removal times of Hg in bags M1, M2, and M3.

Conclusions

The high affinity of Hg to POM was observed and high distribution coefficients for Hg, ranging between 2 and 15, were determined. It was found that the concentration of POC in the water column has an effect on the distribution coefficients of Hg. Both linear and Freundlich adsorption isotherms could be used to describe the adsorption of Hg on POM.

Removal of Hg from the water column was mainly a result of the adsorption of Hg on POM and its subsequent sinking. Thus, the removal of Hg can be described by first-order removal equations.

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