

# CHLORINATED HYDROCARBONS IN THE SOUTHERN OCEAN

Shinsuke TANABE and Ryo TATSUKAWA

*Department of Environment Conservation, Ehime University, Tarumi 3-chome, Matsuyama 790*

**Abstract:** PCBs and chlorinated hydrocarbon pesticides such as DDT and HCH (BHC) were measured in air and water samples taken from the Antarctic, western Pacific and eastern Indian Oceans.

Chlorinated hydrocarbons are widely distributed in the open ocean environment over the northern and southern hemispheres. The most striking fact was their presence in the Antarctic Ocean.  $\Sigma$ DDT (sum of *p*, *p'*-DDE, *p*, *p'*-DDT and *o*, *p'*-DDT) and  $\Sigma$ HCH (sum of  $\alpha$ ,  $\beta$  and  $\gamma$  isomers) concentrations in air and surface water were found to be relatively high in the mid-latitudes of the northern hemisphere. In addition, fairly high concentrations of  $\Sigma$ DDT were observed in the tropical regions and in the mid-latitudes of the southern hemisphere, but  $\Sigma$ DDT levels between the two hemispheres were not much different. HCH isomer compositions found in the northern hemisphere were in the order of  $\alpha$ -HCH >  $\gamma$ -HCH >  $\beta$ -HCH, while in the southern hemisphere  $\gamma$ -HCH was apparently dominant. DDT compound compositions were rather uniform in all locations surveyed, where *p*, *p'*-DDT occupied more than 50%. These results indicate that the environmental pollution by these persistent chemicals spread all over the earth, and that the world situation regarding PCB and pesticide use and the atmospheric circulation seem to account for their global distribution.

Chlorinated hydrocarbons were also found in the subsurface water as deep as several thousand meters. The vertical profiles of PCBs and  $\Sigma$ DDT were rather uniform, while  $\Sigma$ HCH concentrations decreased systematically with depth. These profiles seem to be associated with the sinking particles in the water column. A rapid downward transport of chlorinated hydrocarbons is expected in the Antarctic Ocean where primary productivity is very high.

## 1. Introduction

During the last decade, most of industrialized countries have banned or restricted the use of persistent chlorinated hydrocarbons because of their bioaccumulative characteristics and harmful biological effects. On the contrary, in the developing countries especially in the southern hemisphere and tropical regions, the use of chlorinated hydrocarbon pesticides such as DDT and HCH (BHC) is considered to be indispensable for the agriculture and public health in order to sustain the food supply and to prevent the propagation of malaria (SNELSON, 1977; U.S. COUNCIL ON ENVIRONMENTAL QUALITY AND DEPARTMENT OF STATE, 1980). Considering from the world demand of chlorinated hydrocarbon pesticides, their consumption is expected to continue in the next decade at least, and the transport of these pesticides from land to ocean is unlikely to decline in the near future.

It has been predicted that the open ocean environment serves as a final sink for the

persistent man-made organics (NATIONAL ACADEMY OF SCIENCES, 1971, 1979). In order to make clear the global pollution and the ultimate fate of persistent chemicals, many researchers have reported the occurrence and distribution of chlorinated hydrocarbons in the open ocean air and water (HARVEY *et al.*, 1973, 1974; BIDLAMAN and OLNEY, 1974; JONAS and PFAENDER, 1976; HARVEY and STEINHAEUER, 1976a, b; DAWSON and RILEY, 1977; TANABE and TATSUKAWA, 1980; ATLAS and GIAM, 1981; BIDLAMAN *et al.*, 1981). However, the comprehensive description of their global extent seems to be not established yet, because most measurement data are confined to the northern hemisphere and little information is available in the southern hemisphere. NATIONAL ACADEMY OF SCIENCES (1978) recommended that the most urgent effort must be made to obtain the monitoring data of persistent chlorinated hydrocarbons in the southern hemisphere.

From November 1980 to March 1981, we surveyed the Antarctic, western Pacific and eastern Indian Oceans (Fig. 1), and measured PCBs and chlorinated hydrocarbon pesticides such as DDT compounds (including *p, p'*-DDE, *p, p'*-DDT and *o, p'*-DDT) and HCH isomers (hexachlorocyclohexane, including  $\alpha$ ,  $\beta$  and  $\gamma$  isomers) in air and water samples. This report describes the global distribution of chlorinated hydrocarbons in air and surface water and their vertical profiles in the deep sea water column. Furthermore, their characteristic distribution and vertical transport in the Southern Ocean are also discussed.

This study was supported as part of the BIOMASS Program (Biological Investigations of Marine Antarctic Systems and Stocks) carried out during the cruise of the T.S. UMITAKA MARU, Tokyo University of Fisheries and the JARE-22 Program (the 22nd

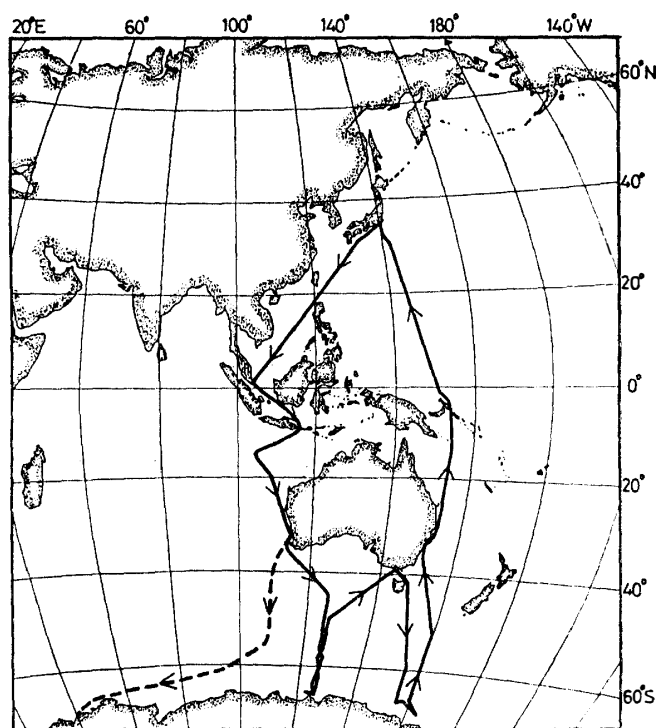


Fig. 1. Survey cruise routes. The solid and broken lines show the cruise routes of the T.S. UMITAKA MARU and icebreaker FUJI, respectively. Air and water samples were collected on the cruise routes.

Japanese Antarctic Research Expedition) of the Ministry of Education, Science and Culture during the cruise of the icebreaker FUJI.

## 2. Materials and Methods

### 2.1. Sample collection

Atmospheric chlorinated hydrocarbons were collected by the high-volume or low-volume air sampler with polyurethane foam plugs. About 1000 m<sup>3</sup> of air were drawn at flow rates of 550 to 650 l/min for PCBs and DDT compounds, and about 200 m<sup>3</sup> at 25 to 27 l/min for HCH isomers.

An all-metal bucket and a large-volume sampler were employed for the collection of surface and subsurface water samples, respectively. About 300 to 500 l of water were sampled and immediately passed through Amberlite XAD-2 resin columns. In several samples, about 500 to 1000 l of water were filtrated through glass fiber filters (Toyo GC-50, 142 mm  $\phi$ ) by a pressure filtration apparatus. The filtrated water was immediately passed through Amberlite XAD-2 resin columns. The glass fiber filters were stocked in a freezer ( $-18^{\circ}\text{C}$ ) until analysis.

The details of collection methods of air and water samples and collection efficiency of chemicals by these methods are given elsewhere (TANABE and TATSUKAWA, 1980, 1983; TANABE *et al.*, 1982).

### 2.2. Chemical analysis

Chlorinated hydrocarbons trapped on polyurethane foam and Amberlite XAD-2 resin were eluted with acetone and ethanol, respectively. The crude extracts from air and water samples were transferred to hexane and then concentrated to 10 ml. The concentrated hexane was cleaned up by shaking with 5% fuming sulfuric acid. The final hexane solution was further concentrated to 100  $\mu\text{l}$  with a microconcentrator under blowing of purified nitrogen gas (TATSUKAWA *et al.*, 1979).

The glass fiber filters were dipped in acetone, and chlorinated hydrocarbons were extracted by shaking for 2 hours. The acetone extracts were concentrated and then transferred to hexane. After clean-up with fuming sulfuric acid, hexane extracts were concentrated to 100  $\mu\text{l}$  and then subjected to the quantitative analysis.

The determination of chlorinated hydrocarbons was conducted on a gas chromatograph-mass spectrometer equipped with a selected-ion monitor (Shimadzu LKB-9000) and occasionally performed on an electron-capture gas chromatograph (Shimadzu GC-5A and GC-4BM). The details of determination procedures and measurement conditions were the same as those reported previously (TATSUKAWA *et al.*, 1979).

Contaminations from sampling equipments, glassware, adsorbents and solvents were checked preliminarily and found to be negligible. Overall procedural blanks of PCBs,  $\Sigma\text{DDT}$  (sum of *p,p'*-DDE, *p,p'*-DDT and *o,p'*-DDT) and  $\Sigma\text{HCH}$  (sum of  $\alpha$ ,  $\beta$  and  $\gamma$  isomers) were less than 2, 0.1 and 0.5 pg/m<sup>3</sup> for air samples respectively, and less than 3, 0.2 and 1 pg/l for water samples respectively.

### 3. Results and Discussion

#### 3.1. Global distribution and atmospheric transport

All chlorinated hydrocarbons examined were detected in open ocean air and surface water samples over both hemispheres (Figs. 2, 3 and 4). The most interesting finding was their presence in the Antarctic Ocean. There are several papers on the pollution of chlorinated hydrocarbons in Antarctica. These works, however, are confined mostly in biological samples (HIDAKA and TATSUKAWA, 1981). With an exception of PCB measurements in South Atlantic water reported by HARVEY and STEINHAUER (1976b), there are no other reports on the persistent chlorinated hydrocarbons in the atmosphere and water of the southern hemisphere. The results presented here indicate that the influence of human activities in terms of PCBs and chlorinated hydrocarbon pesticides extends all over the world.

$\Sigma$ HCH concentrations in the northern hemisphere were much higher than those in the southern hemisphere and the seas bordering the Asian Continent were found to have high concentrations in both air and water samples. A global distribution of HCH residues has been clarified in recent publications which reported their presence over the North Pacific and Indian Oceans (TANABE and TATSUKAWA, 1980) and the North Atlantic Ocean (BIDLEMAN *et al.*, 1981). HCH has been used in many countries. Among these, India, China and Japan have consumed a large amount of HCH for a long period (FOOD AND AGRICULTURE ORGANIZATION, 1979; WANG, 1981; TATSUKAWA *et al.*, 1972). Although reliable statistics on the world production and use of HCH are not yet available, it is safe to assume that the major consumption areas of HCH are still concentrated in the northern hemisphere, especially in the Asian Continent. The wide distribution and high concentration levels of HCH residues in the northern hemisphere depend largely on its extensive use in the Asian Continent.

Besides HCH distribution, high concentrations of  $\Sigma$ DDT were found in the seas near the Asian Continent, but  $\Sigma$ DDT levels between the two hemispheres were not much different (Fig. 3). DDT residues in open ocean air and water have been well investigated in the North Atlantic (BIDLEMAN and OLNEY, 1974; DAWSON and RILEY, 1977; BIDLEMAN *et al.*, 1981), North Pacific (TANABE and TATSUKAWA, 1980; ATLAS and GIAM, 1981) and Indian Oceans (TANABE and TATSUKAWA, 1980; BIDLEMAN and LEONARD, 1982). As compared with the results previously reported and presented in this study, DDT residue levels are not much different among the oceans. GOLDBERG (1975) predicted that the areas of DDT use would shift from the northward to the southward as called "the southward tilt", and he also estimated that about one million tons of DDT would be consumed in the world during the period from 1971 to 1981. Actually in India, 14000 metric tons of DDT were consumed for agriculture between 1975 and 1977 (FOOD AND AGRICULTURE ORGANIZATION, 1979), and 7500 metric tons were required for public health in 1978 to 1979 (JALEES and VEMURI, 1980). In recent years, fairly large amounts of DDT have been consumed also in the tropical and southern countries such as Mexico, Guatemala, El Salvador, Nicaragua, Argentina and so on (FOOD AND AGRICULTURE ORGANIZATION, 1979; CHAPIN and WASSERSTRÖM, 1981). Similar DDT levels between the two hemispheres found in this study may be due to the

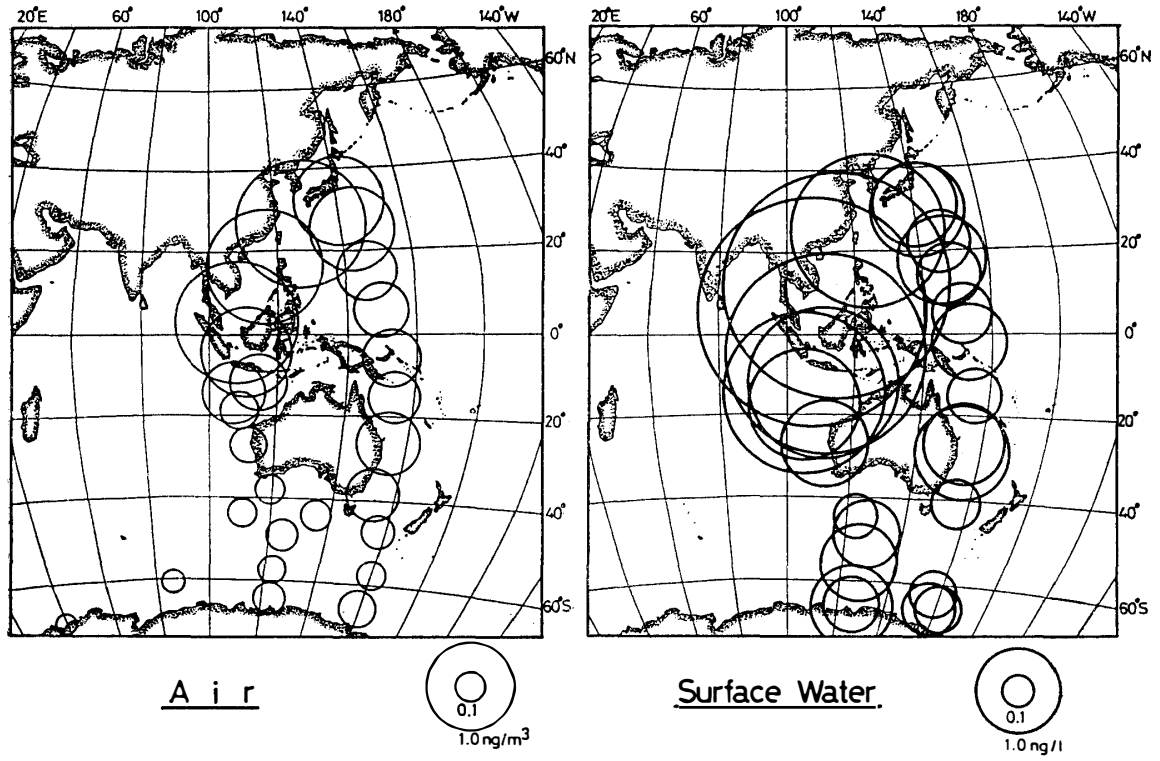


Fig. 2.  $\Sigma$ HCH distributions in air and surface water.

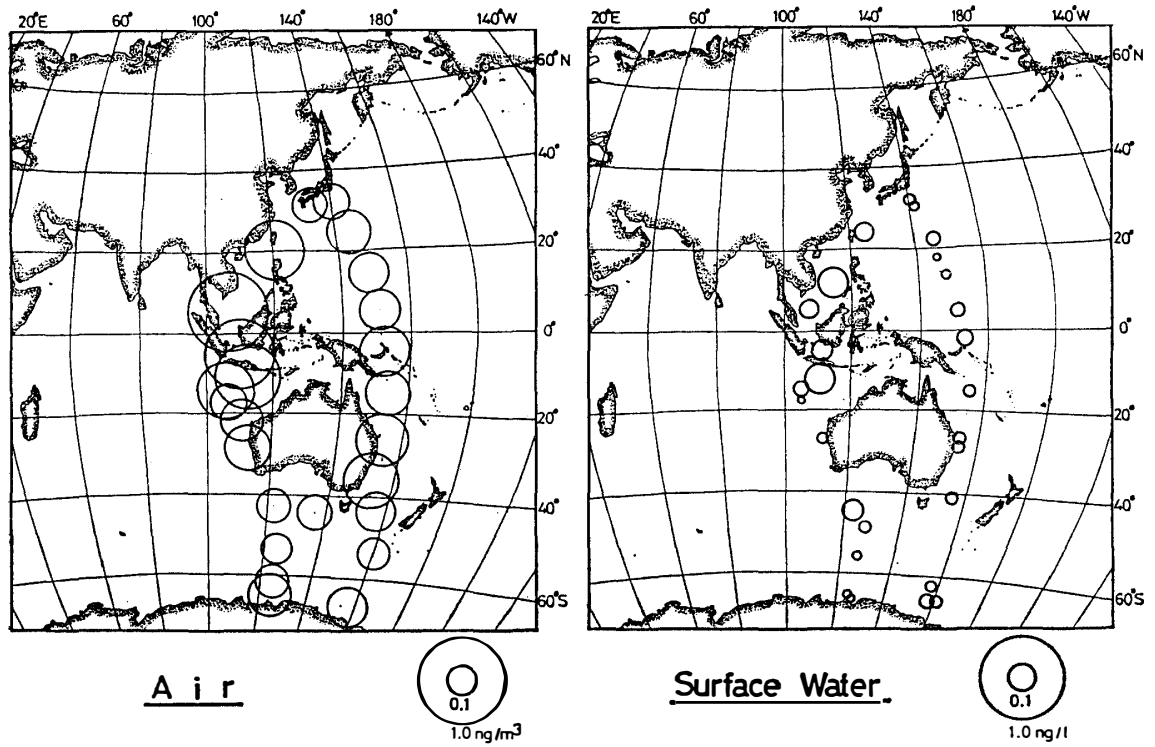


Fig. 3.  $\Sigma$ DDT distributions in air and water samples.

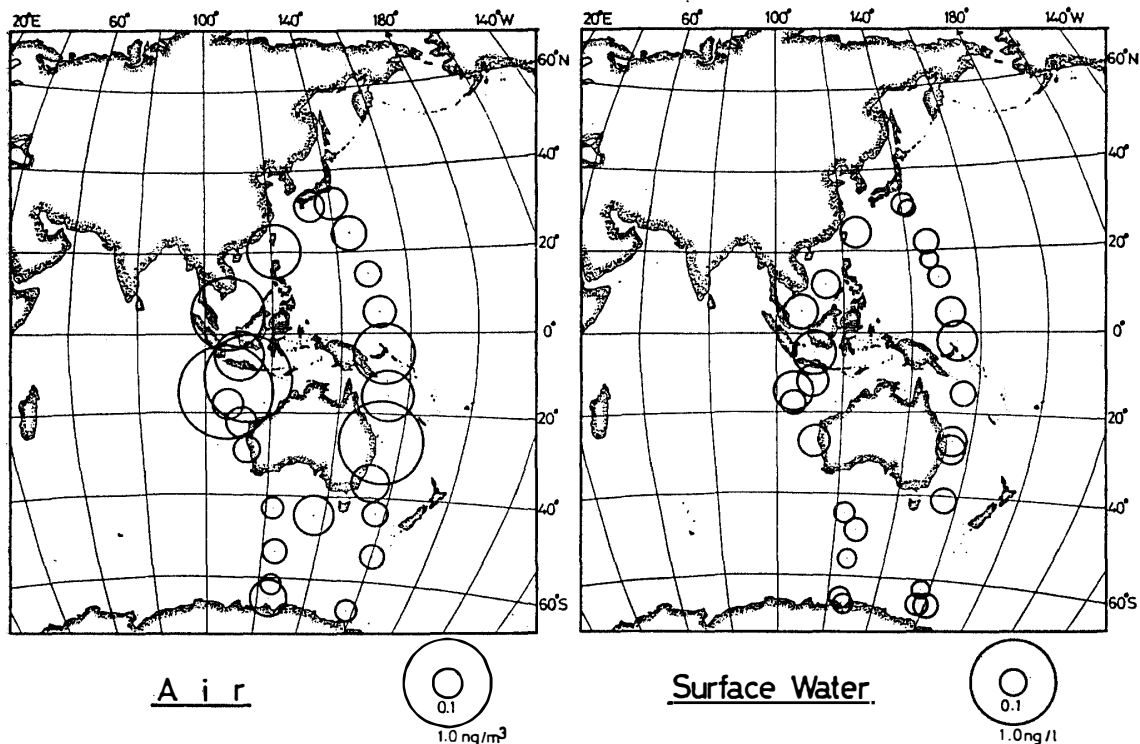


Fig. 4. PCB distributions in air and water samples.

continuous use of DDT in the tropical and southern countries, and due to the decrease of its discharge into open ocean environment from the developed countries in the northern hemisphere.

PCB distributions in air and surface water are given in Fig. 4. Higher concentrations were found in the coastal regions of the tropical and subtropical zones. PCB fluxes into open ocean environment from the developed countries seem to be decreasing due to the ban or restriction of its production and use. Although there is little information on the current use of PCBs in the world, the PCB use is most likely continued in tropical and subtropical countries without any restriction. A recent rice bran oil incident in Taiwan (CHEN *et al.*, 1980) supports the current use of PCBs in low-latitude countries. High PCB levels observed in low latitudes may be explained by its uncontrolled use in tropical and subtropical countries. However, in general, PCB concentrations in open ocean surface water found in this study were lower than those in the Atlantic Ocean (HARVEY *et al.*, 1973, 1974; BIDLAMAN and OLNEY, 1974; HARVEY and STEINHAUER, 1976b; JONAS and PFAENDER, 1976). This may reflect the smaller amounts of PCB use in Asian and Oceanic countries as compared with the United States and European countries.

DDT compound compositions were rather uniform in all oceans surveyed, where *p,p'*-DDT occupied more than 50% (Fig. 5), and that was nearly close to the DDT formulation used for agriculture and malaria control. In contrast, an apparent difference was found in the HCH isomer compositions between the two hemispheres (Fig. 6). In the northern hemisphere HCH isomer compositions were in the order of  $\alpha$ -HCH >  $\gamma$ -HCH >  $\beta$ -HCH, while in the southern hemisphere  $\gamma$ -HCH was dominant. There

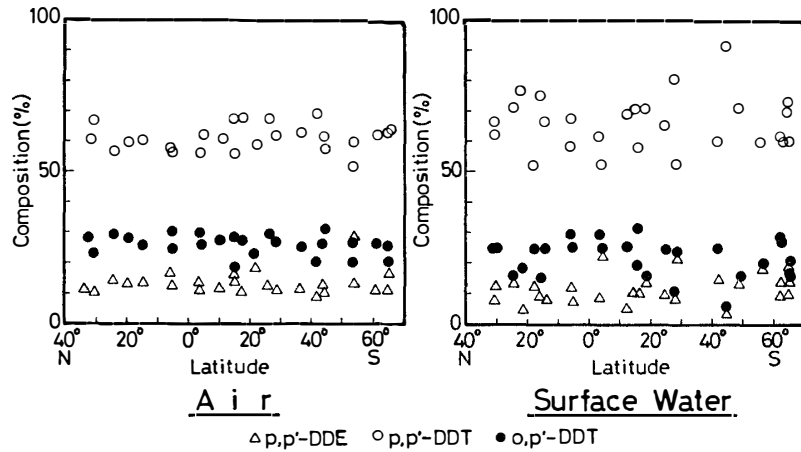


Fig. 5. DDT compound compositions in air and surface water.

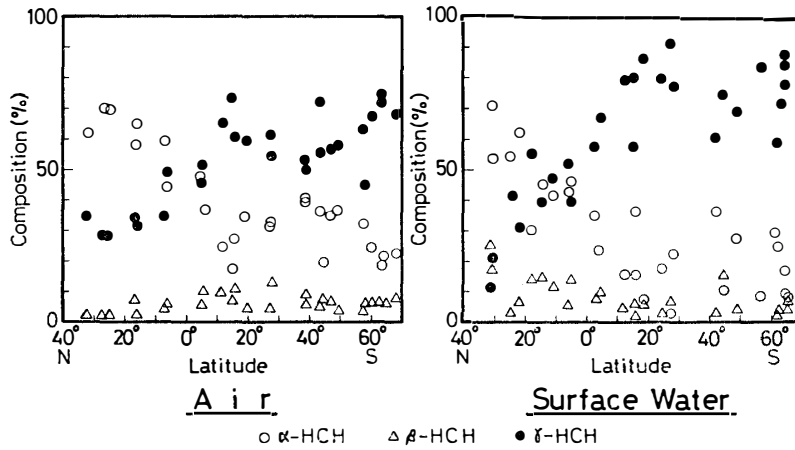


Fig. 6. HCH isomer compositions in air and surface water.

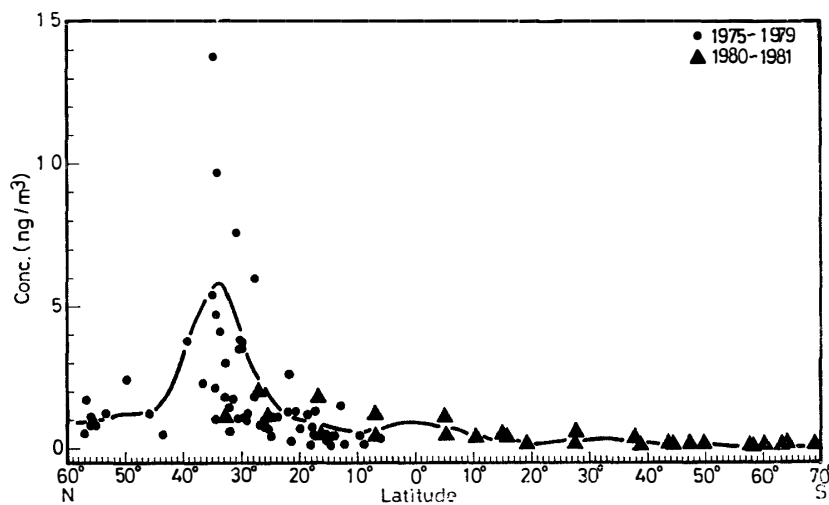


Fig. 7. Latitudinal distribution of  $\Sigma$ HCH in air surveyed from 1975 to 1979 ( $\bullet$ ) and from 1980 to 1981 ( $\blacktriangle$ ).

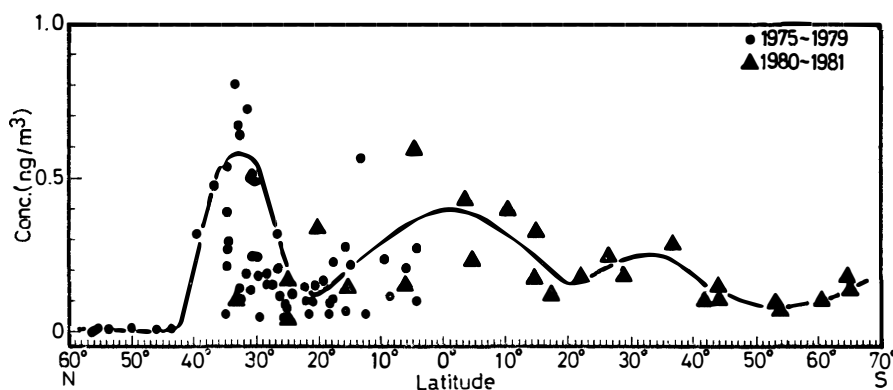


Fig. 8. Latitudinal distribution of  $\Sigma$ DDT in air surveyed from 1975 to 1979 (●) and from 1980 to 1981 (▲).  $\Sigma$ DDT presented in this figure includes only *p*, *p'*-DDT and *p*, *p'*-DDE because of the lack of *o*, *p'*-DDT data from 1975 to 1979.

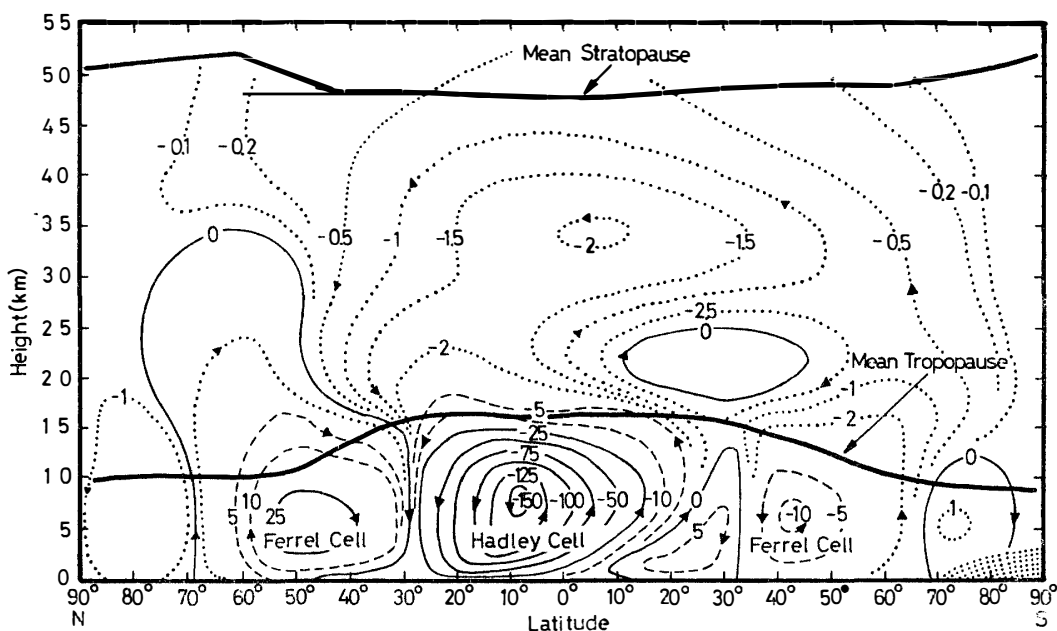


Fig. 9. Mean meridional circulation of atmosphere on December to February reported by LOUIS (1975). The figures in this diagram indicate the mass flow in units of  $10^9$  kg/s.

are two pesticide formulations of HCH consumed in the world. One is the technical HCH that consists roughly of 68 to 78%  $\alpha$ -HCH, 9%  $\beta$ -HCH, 13 to 15%  $\gamma$ -HCH and 8%  $\delta$ -HCH, and another is lindane of the purified  $\gamma$ -HCH (TATSUKAWA *et al.*, 1972). The difference of HCH isomer compositions between the two hemispheres implies the major use of technical HCH in the northern countries and the predominant use of lindane in the southern countries.

In order to understand the global distribution and transport of persistent man-made organics,  $\Sigma$ HCH and  $\Sigma$ DDT concentrations in open ocean air determined in a previous survey (TANABE and TATSUKAWA, 1980) and in this study are plotted against the latitude and given in Figs. 7 and 8, respectively. Higher concentrations of both pesticides were found in the mid-latitudes of the northern hemisphere. Furthermore,



relatively high concentrations of  $\Sigma$ DDT were also measured in the tropical region and in the mid-latitudes of the southern hemisphere. Figure 9 shows the meridional circulation of atmosphere described by LOUIS (1975). There are three meridional cells in the troposphere called the Hadley cell existing over the tropical zone and the Ferrel cell existing over the mid-latitudinal zone in both hemispheres. The regions with high concentrations of pesticides approximately correspond to the areas of the Hadley cell and Ferrel cells. These facts indicate that the meridional circulation of the atmosphere also contributes to the global transport of chlorinated hydrocarbons, besides their use situations in the world. CZEPLAK and JUNGE (1974) indicated that the interhemispheric exchange of the atmosphere was fairly slow because of the impeding action of the Hadley cell. We found the apparent variations of HCH isomer compositions and their concentration levels in air between the two hemispheres (Figs. 6 and 7). These observations may reflect the slow interhemispheric exchange of the atmosphere.

### 3.2. Vertical transport and residence time in the water column

Chlorinated hydrocarbons were found also in the subsurface water even at the depth of several thousand meters (Fig. 10). The presence of chlorinated hydrocarbons

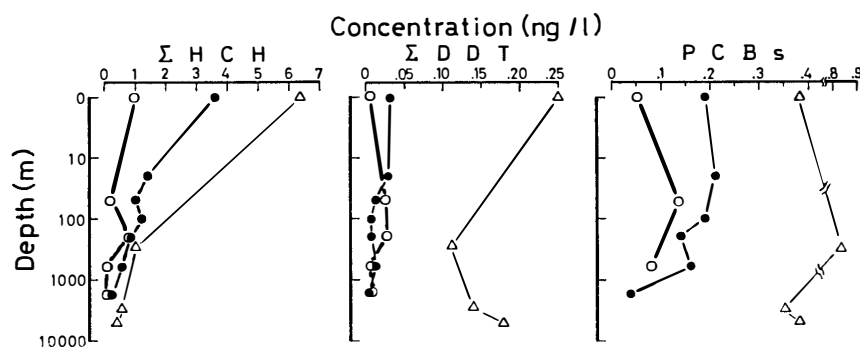


Fig. 10. Vertical profiles of chlorinated hydrocarbons in the open ocean water column surveyed in the Pacific ( $\Delta$ :  $29^{\circ}59'N$ ,  $145^{\circ}45'E$ ), Indian ( $\bullet$ :  $15^{\circ}04'S$ ,  $104^{\circ}49'E$ ) and Antarctic Oceans ( $\square$ :  $64^{\circ}55'S$ ,  $124^{\circ}40'E$ ). The data of Pacific Ocean are based on our previous survey (TANABE and TATSUKAWA, 1983).

in the deep water of open ocean has been suggested by their detection in deep sea fish (BARBER and WARLEN, 1977) and sediments (HARVEY and STEINHAEUER, 1976a). Several workers also measured the PCBs and DDT compounds in the subsurface water and reported the uniform distribution of these chemicals in the water column (HARVEY *et al.*, 1974; SCURA and MUCLURE, 1975; HARVEY and STEINHAEUER, 1976b; JONAS and PFAENDER, 1976). In this study, PCBs and DDT compounds were found to be rather uniform throughout the water column as reported previously, whereas HCH isomer concentrations decreased systematically with depth. The presence of chlorinated hydrocarbons in deep water cannot be explained by the convective and diffusional mixing of water, because it is no more than half a century since their production and use started. The vertical transport of chlorinated hydrocarbons seems to be strongly associated with sinking particles in the water column.

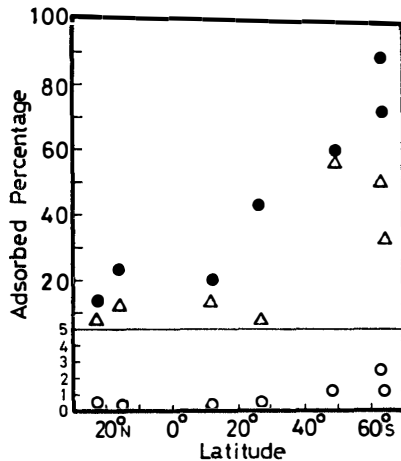


Fig. 11. Latitudinal variation of the percentage of PCBs ( $\Delta$ ),  $\Sigma$ DDT ( $\bullet$ ) and  $\Sigma$ HCH ( $\circ$ ) adsorbed on suspended solids in surface water.

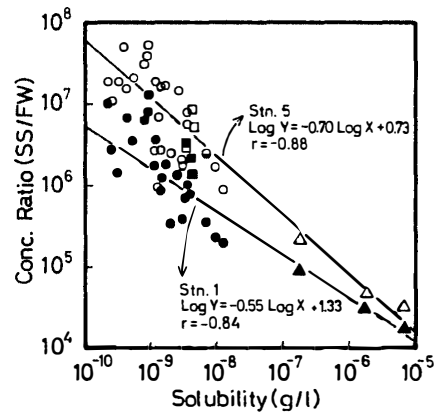


Fig. 12. Relationship between the water solubilities of PCBs ( $\circ$ ), DDT compounds ( $\square$ ) and HCH isomers ( $\Delta$ ) and their concentration ratios of adsorbed to dissolved fractions of suspended solids to filtered water.

So, we operationally separated the chlorinated hydrocarbons into the dissolved and adsorbed fractions by the filtration method. In consequence, it became clear that a large portion of DDT compounds and PCBs in surface water was retained on the suspended solids, while most part of HCH isomers were present in the dissolved fraction (Fig. 11). It is also noted that apparent negative correlations existed in the relationship between the water solubilities of chlorinated hydrocarbons and their concentration ratios of adsorbed to dissolved fractions (Fig. 12). Furthermore, the percentage of chlorinated hydrocarbons adsorbed on suspended solids in surface water increased towards the high latitudinal locations (Fig. 11), and this trend seems to be proportional to the concentrations of suspended solids in surface water. These facts indicate that HCH isomers have been slowly scavenged from surface to deeper layers through the water column, while PCBs and DDT compounds have been rapidly and abundantly

Table 1. Estimated residence time of chlorinated hydrocarbons in the open ocean euphotic zone by 100 m depth.

Oceans	$C_{\text{prod.}}^*$ ( $\text{g}/\text{m}^2/\text{yr}$ )	Residence time		
		$\Sigma$ HCH (year)	$\Sigma$ DDT (day)	PCBs (day)
Oligotrophic				
Pacific Ocean 22°05'N, 145°02'E	50-100	5.1-10	69-140	130-280
Mesotrophic				
Antarctic Ocean 48°59'S, 127°31'E	100-150	4.3-6.4	29-44	30-44
Eutrophic				
Antarctic Ocean 64°42'S, 124°15'E	150-250	2.0-3.4	11-19	26-44

\* KOBLENTZ-MISHKE *et al.* (1970).

transported by the sinking particles. It can be assumed that the transport velocity of chlorinated hydrocarbons greatly depends on their water solubilities and the primary productivities in the oceans.

Based on the measurement data obtained from this study and an equation on the relationship between the organic carbon flux data and the primary production rates in the world ocean reported by SUESS (1980), the residence time of chlorinated hydrocarbons in the euphotic zone, the top 100 m of the water column, was estimated (Table 1). The details of this estimation are given elsewhere (TANABE and TATSUKAWA, 1983). In general, the longer residence time of chlorinated hydrocarbons was obtained in more oligotrophic oceans.  $\Sigma$ HCH revealed the long residence time as more than 2 years in all locations, in which the longest residence time was found in the central pacific defined as the oligotrophic ocean. While, PCBs and  $\Sigma$ DDT showed rather short residence time less than one year, in which  $\Sigma$ DDT residence time in the Antarctic Ocean was extremely short as only for 11 to 19 days. FOWLER and SMALL (1972) pointed out that the sinking rate of euphausiid fecal pellets reached several hundred meters per day. It can be safely considered that the less water-soluble chlorinated hydrocarbons are rapidly removed from the euphotic zone by the particles with high sinking rates, particularly in the eutrophic oceans.

#### 4. Conclusions

The behavior and fate of chlorinated hydrocarbons in open ocean air and water are closely linked with their accumulation in marine organisms. If the production and use of these chemicals are stopped in the world, the concentrations of PCBs and DDT compounds in surface water will rapidly decrease due to their short residence time in water column and those in marine organisms will follow this with some temporal gaps. However, in the case of HCH isomers with long residence time, their concentration levels in surface water will slowly decline. Though HCH isomers are less bioaccumulative as compared with PCBs and DDT compounds, their biological pollution will continue for a long time. From the geographical viewpoint, the comparatively rapid decrease of chlorinated hydrocarbons is expected in the eutrophic ocean, notably in the Antarctic Ocean.

However, it is unlikely that the use of persistent pesticides will be stopped in the near future. The developing countries in the southern hemisphere and tropical regions are counting on the continuous use of DDT and HCH for the agriculture and public health at least until the end of this century. PCBs are also dragging on their spill into the environment from various PCB containing equipments that are now in service, although its chemical manufacture has now ceased in many developed countries. Thus, the inflows of chlorinated hydrocarbons into open ocean environment especially in the southern hemisphere will continue and their concentration levels in marine organisms as well as air and water do not seem to decrease for the present. The comprehensive survey on the bioaccumulation of these chemicals should be made in the southern hemisphere.

### Acknowledgments

The authors are grateful to Prof. M. MURANO, Tokyo University of Fisheries, and Dr. Y. YOSHIDA, National Institute of Polar Research, for arranging the cruises for sampling. We would like to thank the officers and crew of the T.S. UMITAKA MARU and icebreaker FUJI.

### References

- ATLAS, E. and GIAM, C. S. (1981): Global transport of organic pollutants; Ambient concentrations in the remote marine atmosphere. *Science*, **211**, 163–165.
- BARBER, R. T. and WARLEN, S. M. (1977): Organochlorine insecticide residues in deep sea fish from 2500 m in the Atlantic Ocean. *Environ. Sci. Technol.*, **13**, 1146–1148.
- BIDLEMAN, T. F. and OLNEY, C. E. (1974): Chlorinated hydrocarbons in the Sargasso Sea atmosphere and surface water. *Science*, **183**, 516–518.
- BIDLEMAN, T. F. and LEONARD, R. (1982): Aerial transport of pesticides over the Northern Indian Ocean and adjacent seas. *Atmos. Environ.*, **16**, 1099–1107.
- BIDLEMAN, T. F., CHRISTENSEN, E. J., BILLINGS, W. N. and LEONARD, R. (1981): Atmospheric transport of organochlorines in the North Atlantic Gyre. *J. Mar. Res.*, **39**, 443–469.
- CHAPIN, G. and WASSERSTRÖM, R. (1981): Agricultural production and malaria resurgence in Central America and India. *Nature*, **293**, 181–185.
- CHEN, P. H., GAW, J. M., WONG, C. K. and CHEN, C. J. (1980): Levels and gas chromatographic patterns of polychlorinated biphenyls in the blood of patients after PCB poisoning in Taiwan. *Bull. Environ. Contam. Toxicol.*, **25**, 325–329.
- CZEPLAK, G. and JUNGE, C. (1974): Studies of interhemispheric exchange in the troposphere by a diffusion model. *Adv. Geophys.*, **18B**, 57–72.
- DAWSON, R. and RILEY, J. P. (1977): Chlorine-containing pesticides and polychlorinated biphenyls in British coastal waters. *Estuarine Coastal Mar. Sci.*, **4**, 55–69.
- FOOD AND AGRICULTURE ORGANIZATION (1979): *FAO Production Year Book*, Vol. 32. Rome, FAO, 245 p.
- FOWLER, S. W. and SMALL, L. F. (1972): Sinking rates of euphausiid fecal pellets. *Limnol. Oceanogr.*, **17**, 293–296.
- GOLDBERG, E. D. (1975): Synthetic organohalides in the sea. *Proc. R. Soc. London, Ser. B*, **189**, 277–289.
- HARVEY, G. R. and STEINHAEUER, W. G. (1976a): Biochemistry of PCB and DDT in the North Atlantic. *Environmental Biochemistry*, Vol. 1, ed. by J. O. NRIAGU. Michigan, Ann Arbor Sci., 203–221.
- HARVEY, G. R. and STEINHAEUER, W. G. (1976b): Transport pathways of polychlorinated biphenyls in Atlantic water. *J. Mar. Res.*, **34**, 561–575.
- HARVEY, G. R., STEINHAEUER, W. G. and TEAL, J. M. (1973): Polychlorinated biphenyls in North Atlantic Ocean water. *Science*, **180**, 643–644.
- HARVEY, G. R., STEINHAEUER, W. G. and MIKLAS, H. P. (1974): Decline of polychlorinated biphenyl concentrations in North Atlantic surface water. *Nature*, **252**, 387–388.
- HIDAKA, H. and TATSUKAWA, R. (1981): Sôsetsu; Nankyoku ni okeru yûki enso kagô-butsu no kenkyû (Review; Environmental pollution by chlorinated hydrocarbons in the Antarctic). *Nankyoku Shiryô (Antarct. Rec.)*, **71**, 151–164.
- JALEES, K. and VEMURI, R. (1980): Pesticide pollution in India. *Int. J. Environ. Stud.*, **15**, 49–54.
- JONAS, R. B. and PFAENDER, F. K. (1976): Chlorinated hydrocarbon pesticides in Western North Atlantic Ocean. *Environ. Sci. Technol.*, **10**, 770–773.
- KOBLENTZ-MISHKE, O. J., VOLKOVINSKY, V. V. and KOBANOBA, J. G. (1970): Plankton primary production of the world ocean. *Scientific Exploration of the South Pacific*, ed. by National Academy of Sciences. Washington, D. C., 183–193.

- LOUIS, J. F. (1975): Mean meridional circulation. The natural stratosphere of 1974. CIAP Monograph 1, Washington, D. C., Dept. of Transportation, 6.23-6.31.
- NATIONAL ACADEMY OF SCIENCES (1971): Chlorinated Hydrocarbons in the Marine Environment. Washington, D. C., 42 p.
- NATIONAL ACADEMY OF SCIENCES (1978): Halogenated hydrocarbons. The Tropospheric Transport of Pollutants and Other Substances to the Oceans. Washington, D. C., 146-168.
- NATIONAL ACADEMY OF SCIENCES (1979): PCB transport throughout the environment. Polychlorinated Biphenyls. Washington, D. C., 11-76.
- SCURA, E. D. and MUCLURE, V. E. (1975): Chlorinated hydrocarbons in sea water; Analytical method and levels in the northeastern Pacific. *Mar. Chem.*, **3**, 337-346.
- SNELSON, J. T. (1977): The importance of chlorinated hydrocarbons in the world agriculture. *Ecotoxic. Environ. Saf.*, **1**, 17-30.
- Suess, E. (1980): Particle organic carbon flux in the Oceans; Surface productivity and oxygen utilization. *Nature*, **288**, 260-263.
- TANABE, S. and TATSUKAWA, R. (1980): Chlorinated hydrocarbons in the North Pacific and Indian Oceans. *J. Oceanogr. Soc. Jpn.*, **36**, 217-226.
- TANABE, S. and TATSUKAWA, R. (1983): Vertical transport and residence time of chlorinated hydrocarbons in the open ocean water column. *J. Oceanogr. Soc. Jpn.*, **39**, 46-55.
- TANABE, S., KAWANO, M. and TATSUKAWA, R. (1982): Chlorinated hydrocarbons in the Antarctic, western Pacific and eastern Indian Oceans. *Trans. Tokyo Univ. Fish.*, **5**, 97-109.
- TATSUKAWA, R., WAKIMOTO, T. and OGAWA, T. (1972): BHC residues in the environment. *Environmental Toxicology of Pesticides*, ed. by F. MATSUMURA *et al.* New York, Academic Press, 229-238.
- TATSUKAWA, R., TANABE, S. and YOSHIDA, T. (1979): Jinkô yûki kagô-butsumu (Man-made organics). *Kaiyô Kankyô Chôsaishô*, ed. by The Oceanogr. Soc. Japan. Tokyo, Kôseisha Kôseikaku, 232-269.
- U. S. COUNCIL ON ENVIRONMENTAL QUALITY AND DEPARTMENT OF STATE (1980): The food and agriculture projections and the environment; Ecological effects of fertilizers and pesticides. The Global 2000 Report to the President, **2**, Technical Report. Washington, Government Printing Office, 283-288.
- WANG, D. (1981): The status of pesticides in China and their future. *J. Pestic. Sci.*, **6**, 397-399.

(Received November 16, 1982; Revised manuscript received February 11, 1983)