

## ORGANICAL CHEMICALS AS CONTAMINANTS OF WATER BODIES AND DRINKING WATER

**Yoshiteru Tsuchiya**

*Department of Applied Chemistry, Faculty of Engineering, Kogakuin University, Japan*

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### Summary

Pollution by organic chemicals in the aquatic environment occurs by various mechanisms. Naturally occurring organic chemicals produced by aquatic microorganisms, such as 2-methylisoborneol and geosmin, which have an earthy-musty odor, and microcystin, which shows hepatotoxicity, contaminate surface water such as rivers, lakes and reservoirs. Industrial waste containing artificial chemicals flows into and contaminates many water areas. Volatile organic compounds (VOCs), pesticides, phenolic compounds, phthalates, and nitrogen-containing compounds, are often detected in polluted water. VOCs such as trichloroethylene, tetrachloroethylene and 1,1,1-trichloroethane, are most often found in groundwater. Dioxins and polynuclear aromatic hydrocarbons (PAHs), produced during combustion of organic materials, are also found in surface water. On the other hand, chlorination by-products such as trihalomethanes (THMs), haloacetic acids and MX, which are produced during the water purification process by reaction of chlorine and organic materials, are found in drinking water. In the WHO Guidelines for Drinking-Water Quality, levels are set for 28 organic constituents (i.e. microcystin-LR, chlorinated alkanes, chlorinated ethenes, aromatic hydrocarbons, chlorinated benzenes and miscellaneous), 33 pesticides, and 9 disinfectant by-products, due to their health effects on humans. In recent years, occurrence of PPCPs (pharmaceutical and personal care products) and perfluoroalkyl acids (PFAs, i.e. PFOS and PFOA) in aquatic environment has been recognized as emerging issues in environmental chemistry.

## 1. Introduction

Pollution of organic chemicals in the aquatic environment occurs from natural products of aquatic microorganisms and artificial contaminants from industrial chemicals or human wastes. However, the effects of organic pollutants in rivers, lakes and ponds, reservoirs, groundwater and drinking water, differ for each specific contaminant. In general, organic chemical pollutants affecting human health are regulated by legal limits such as WHO Guidelines for Drinking-Water Quality, EPA National Primary Drinking Water Regulations, water quality standard in waterworks law, environmental standards and wastewater standards. Estimated organic contaminants are as follows:

- Organic chemicals produced by algae or Actinomycetes which growing in the natural aquatic environment.
- Organic pollutants of industrial chemicals, from industrial activity;
- Water contamination by pesticides from agricultural or other uses;
- Organic chemicals from unintentionally produced contaminants during combustion or burning of organic materials such as polynuclear aromatic hydrocarbons (PAHs);
- Pollutants in waste from the human environment such as domestic effluent, and
- Newly produced materials during the water purification process of drinking water.

This chapter describes organic compounds suspected of contaminating the aquatic environment.

## 2. Contamination of metabolites produced by aquatic microorganisms

Water pollution from organic chemicals occurs under various condition and from various causes. Naturally occurring organic chemicals produced by aquatic microorganisms such as blue-green algae (Cyanobacteria) or *Streptomyces sp.* often cause water pollution. Such aquatic microorganisms can show remarkable increases in eutrophic water areas such as lakes, ponds, rivers, and reservoirs, and produce various metabolites. The eutrophication of water area occurs when nutrients such as N or P are increased by input of industrial effluent or domestic waste. Natural organic chemicals related to contamination of water are metabolites of blue-green algae and *Streptomyces sp.* which contain volatile odorous compounds as well as toxic compounds.

## 2.1. Volatile metabolites

Volatile metabolites produced by blue-green algae and streptomyces are often found in water areas inhabited by these organisms. The volatile metabolites, 2-methylisoborneol (2-MIB) and geosmin (trans-1,10-dimethyl-trans-9-decalol), both of which produce an earthy/musty odor at an extremely low level ( $\text{ng L}^{-1}$ ), may be contained in supplied drinking water. The problem of unpleasant drinking water has occurred in various countries such as USA, Canada, Brazil, Australia, Europe, South Africa, Japan and others. It is known that blue-green algae, *Oscillatoria geminata*, *Phormidium tenue* and others produce 2-MIB; *Oscillatoria splendida*, *Anabaena macrospora* and others produce geosmin, and *Streptomyces sp.* produce both compounds. They can cause a musty odor at concentrations in water of 5 to  $10\text{ng L}^{-1}$ . These compounds are chemically stable without a double bond in the chemical structure as the third grade alcohol terpenoid.

For this reason, decomposition or removal of these compounds by water treatments such as chlorination or other common methods in water purification plants, is difficult. In Japan, aquatic microorganisms occur in about 40% of water sources every year in summer, and an earthy-musty odor develops in the water. From investigations in the 1970s, tap water with an earthy-musty odor is supplied every year to more than ten million people, and the odor is troublesome. Recently, however, ozone/activated carbon treatments have been carried out in many purification plants for water control, and the supply of water with an earthy-musty odor has decreased, to about one and a half million people in an investigation in 2000. Earthy-musty odors were investigated for one water area in Japan from April to November. 2-MIB was detected in the water supply source in the range of 2 to  $1000\text{ ng L}^{-1}$  and geosmin at 1 to  $500\text{ ng L}^{-1}$ . However, 2-MIB was detected at a maximum level of  $12\text{ ng L}^{-1}$  and geosmin at  $36\text{ng L}^{-1}$  in the tap water.

Some cyanobacteria may multiply even when the water temperature is low. For example, the highest rate of multiplication is shown by *Aphanizomenon flos-aquae* which inhabits ponds; a concentration of  $7600\text{ ng L}^{-1}$  for geosmin was detected in the water when the water temperature was only  $3.5\text{ }^{\circ}\text{C}$  in winter. A comfortable guideline value for waterworks law in Japan recommends an extremely low concentration of  $10\text{-}20\text{ ng L}^{-1}$  for 2-MIB and geosmin. These cyanobacteria also produce various metabolites such as aliphatic hydrocarbons (e.g. n-heptadecane, n-pentadecane, 1-heptadecene), fatty acids, amines (e.g.. trimethylamine), aromatic compounds (e.g. ethylbenzene),

sesquiterpene alcohol and sulfur-containing compounds (e.g. methyl mercaptan, isopropyl disulfide).

## 2.2. Toxic substances produced by blue-green algae

Other species of blue-green algae containing toxic compounds produce blooms in eutrophic water bodies such as lakes, ponds and rivers. It has been found worldwide that certain animals (e.g. cow, horse, sheep, pig, chicken) can die after drinking water containing an algal bloom. The blue-green algae producing such toxic substances are as follows: *Anabaena flos-aquae*, *Aphanizomenon flos-aquae* produce anatoxin-a, anatoxin-a(s), and aphantoxin which shows neurotoxicity. *Microcystis aeruginosa*, *M. viridis*, and *Oscillatoria agardhii* produce microcystin which shows hepatotoxicity. The toxic substance, nodularin, is produced by *Nodularia spumigena*, and cylindrospermopsin is produced by *Cylindrospermopsis raciborskii* and *Umezakia natans*. Microcystin has more than 50 isomers, but microcystin-LR, -RR and -YR, are mostly found in water and in cells of algae. The typical chemical formula of microcystin is shown in Figure 1.

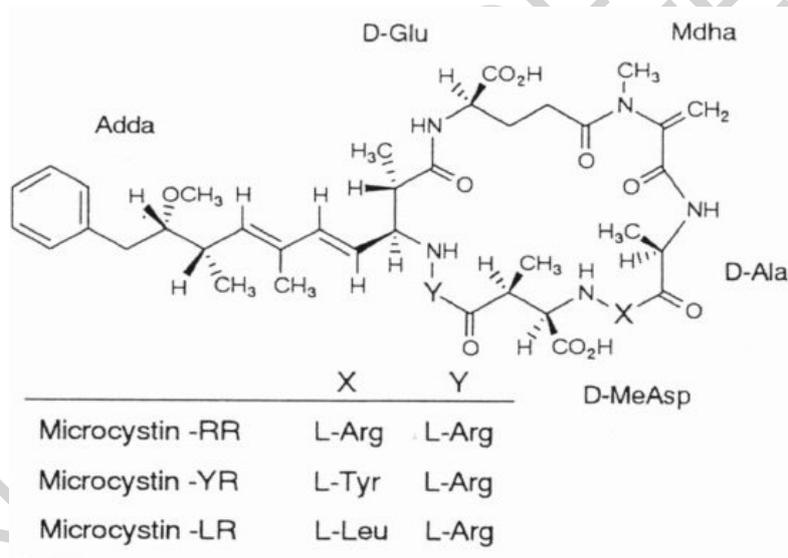


Figure 1. Chemical formula of microcystin

The first documented lethality to humans from cyanobacterial hepatotoxin (microcystin-LR), occurred via the intravenous route, in a dialysis clinic during 1996, when 76 deaths were reported. Microcystins are partly eluted to water by the environmental conditions, though they are generally contained in the cell body. Microcystins in the cells of cyanobacteria and in the water collected from water-blooms in eight water areas were analyzed. As a result, microcystin-RR was detected at a range of 0.03 to 8.8  $\mu\text{g L}^{-1}$  in the water samples, and 0.15 to 205  $\mu\text{g L}^{-1}$  in the algae body. Microcystin-YR was detected at 0.02 to 0.3  $\mu\text{g L}^{-1}$  in the water sample and 0.09 to 9.7  $\mu\text{g L}^{-1}$  in the algae body. Microcystin-LR was detected at 0.02 to 1.5  $\mu\text{g L}^{-1}$  in the water sample and 0.07 to 390  $\mu\text{g L}^{-1}$  in the algae body. About 1 to 5% of microcystins in the cell body are eluted to the water.

Microcystins in water decompose during reaction with chlorine, which is added for disinfection. For example, when  $10 \mu\text{g L}^{-1}$  of each microcystin was treated with  $1 \text{ mg L}^{-1}$  of free chlorine, microcystin-RR and -LR decomposed in about 60 minutes, and microcystin-YR decomposed in about 20 minutes. However, chlorination by-products change based on the kind of substances involved in the interaction and the toxicity of these by-products is not clear. However, these three compounds showed a high degree of heat-resistance and could not be decomposed even by 30 minutes of boiling. In the WHO Guidelines for Drinking-Water Quality, microcystin-LR is recommended at  $1 \mu\text{g L}^{-1}$  as a proposed guideline value. It is important to know whether the water supply source includes areas where there may be algal blooms.

### 3. Contamination by industrial chemicals

As a result of industrial activities, production of artificial chemicals and use of chemical products, various chemicals can flow into the environment and contaminate water bodies. Small amounts of organic chemicals are often detected in water. Among these contaminants, WHO guidelines for drinking-water quality has proposed recommended guidelines for limiting carcinogenic compounds and harmful compounds such as solvents, pesticides, raw materials from plastic products and organotin.

In Japan, about 2000 harmful substances were selected as priority research subjects. Field surveys of these chemicals in rivers, lakes, seas and their bottom sediments, and fish in the surrounding water area, have been carried out since 1974 by the Japanese Ministry of the Environment. The chemical substances were chosen according to the following priorities. Persistence in the environment, high bioaccumulation in the environment, chemical substances showing chronic toxicity, high consumption in the market. About 19% of the measured substances were detected in water as a result of investigating about 750 compounds at 10 to 400 sites between 1974 and 1998. Of the compounds detected in water, there were 22 volatile organic compounds (VOC), 6 phenolic compounds, 15 nitrogen-containing organic compounds, 5 pesticides, 5 PAHs (polynuclear aromatic hydrocarbons), 3 phthalates, 4 organotin compounds and 15 other substances. Of the VOCs detected, trichloroethylene, tetrachloroethylene and 1,1,1-trichloroethane were often found in water bodies at high concentrations. Other detected compounds were as follows: carbon tetrachloride, 1,2-dichloroethane, cis-1,2-dichloroethylene, trichloromethane, toluene, and others. Of the phenols, though bisphenol A which is an endocrine-disrupting chemical was not detected (0/60) (detected number/sample number) in 1976, it was detected in 27.7% (41/148) of water areas in the range of 0.01 to  $0.268 \text{ ng L}^{-1}$  in 1996. Phenol and p-butylbenzoic acid were also detected. Of the nitrogen-containing substances, amines and amides such as acrylamide, aniline, and cyclohexylamine were detected in river and lake water. Though aniline was detected at 58.5% (40/60) in the range of 0.02 to  $28 \mu\text{g L}^{-1}$  by the field survey in 1976, it was only detected in one sample among 141, at  $0.07 \mu\text{g L}^{-1}$ , during the investigation in 1998, indicating a clear decrease in water pollution. However, though 2-methylpyridine was not found (0/30) in 1986, an investigation in 1995 detected it at a range of 0.1 to  $2.4 \mu\text{g L}^{-1}$  in 11.7% of samples. PAHs such as acenaphthylene, acenaphthene, benzo[a,h]anthracene, pyrene and benzo[ghi]perylene were detected in 1 to 4 samples at the  $0.01$  to  $1 \mu\text{g L}^{-1}$  level.

As for other detected compounds in this field survey, ABS was detected in 17.6% of samples in the range of 280 to 29 000  $\mu\text{g L}^{-1}$ , and phosphate substances (i.e. tributyl phosphate, tributoxyethyl phosphate, and tris-2-chloroethyl phosphate) were detected in 7.2 to 51.4% of samples in the range of 0.01 to 2.8  $\mu\text{g L}^{-1}$ . It is clear that various chemical substances used cannot be detected in environment waters. As for organotin compounds, investigations have been carried out by the Ministry of the Environment since 1991. Though organotin compounds were used as agricultural agents, tributyltin(TBT) and triphenyltin(TPT) were specially used as antifouling agents to paint the bottoms of ships and fish nets and cages used in culture. Recently, organotin compounds have been shown to be endocrine-disrupting chemicals. The result of a field survey between 1991 and 2000 based on 100 samples of seawater from 37 sites collected from estuaries, ports and bays is shown in Table 1.

Tributyltin was detected in about 68% of samples in 1991, but decreased gradually thereafter, being detected in 8.8% of samples in 2000. Though the detected concentration was 0.08  $\mu\text{g L}^{-1}$  in 1991, it decreased to a concentration of 0.005 $\mu\text{g}^{-1}$  in 2000. However, the detection frequency of triphenyltin was lower, in 6 to 10% of samples, on the initial survey, and hardly detected after 1995. The concentration of detected TPT showed 0.04  $\mu\text{g L}^{-1}$  in the beginning, but has been less than 0.01  $\mu\text{g L}^{-1}$  since 1995. In 1990, the Japanese government specified TBT as a special chemical compound, and restricted its use. It seems that this regulation has decreased the sea area affected by organotin compounds.

	Tributyltin(TBT)		Triphenyltin(TPT)	
	Detection rate (%)	Detection range $\mu\text{g L}^{-1}$	Detection rate (%)	Detection range $\mu\text{g L}^{-1}$
1991	60/93 (64.5%)	Nd-0.067	5/87 (5.7%)	Nd-0.014
1992	52/99 (52.5)	Nd-0.084	10/90(11.1)	Nd-0.044
1993	42/99 (42.4)	Nd-0.049	2/90 (2.2)	Nd-0.011
1994	35/99 (35.4)	Nd-0.03	4/92 (4.3)	Nd <sup>^</sup> 0.01
1995	31/105(29.5)	Nd-0.042	0/87	
1996	27/105(25.7)	Nd-0.014	0/108	
1997	21/107(19.6)	Nd-0.009	0/108	
1998	20/76 (27.6)	Nd-0.008	4/102(3.9)	Nd-0.0015
1999	16/105(15.2)	0.003-0.01	3/105(2.9)	0.001-0.004
2000	9/102 (8.8)	0.003-0.005	0/102	

Table 1. Concentration of TBT and TPT in seawater

### 3.1. Volatile organic compounds (VOCs)

Volatile organic compounds (VOCs) were often found in the environment because of their high volatility; they diffuse into environment when solvents are used. The diffused chemicals in the environment contaminate both the atmosphere and water bodies. Many volatile organic compounds are found in various water areas, e.g. benzene, toluene, xylene, dichloroethane (used as solvents), trichloroethylene, tetrachloroethylene and 1,1,1-trichloroethane (used in dry cleaning) and carbon tetrachloride (used as a raw material for chlorofluorocarbons). Dichlorobenzene, trichlorobenzene styrene and vinyl chloride are found in the water after using the raw materials of industrial products.

Trichloroethylene, tetrachloroethylene and 1,1,1-trichloroethane, used for metal washing and dry cleaning, were often the cause of groundwater contamination. Characteristics of these VOCs include poor solubility in water, heavy specific gravity, low surface tension, and low viscosity. Therefore, released VOC in the environment can readily percolate into groundwater. The Japanese Ministry of the Environment is monitoring 3500 to 4500 water samples targeting 11 compounds listed in the environmental standard of 1999. These 11 compounds were all detected in samples investigated in 1999, showing a 2.5 to 4.6% detection rate, exceeding the standard value for trichloroethylene, tetrachloroethylene and 1,1,1-trichloroethane. Fifteen samples of trichloroethylene demonstrated a maximal value of 0.99 mg L<sup>-1</sup>, and 23 samples of tetrachloroethylene demonstrated a maximum value of 0.19 mg L<sup>-1</sup>, exceeding the standard values. Other compounds exceeding the standard value were carbon tetrachloride, detected at a maximal value of 0.003 mg L<sup>-1</sup>, 1,2-dichloroethane detected at a maximal value of 0.35mg L<sup>-1</sup> and 1,1-dichloroethylene detected at a maximal value of 0.031 mg L<sup>-1</sup>.

As for the results of the monitoring investigation of groundwater pollution, 46.7% of samples contained tetrachloroethylene (maximal value of 60 mg/L), 29% trichloroethylene (maximal value of 110 mg L<sup>-1</sup>), and 20% cis-1,2-dichloroethylene (maximal value of 48 mg/L). VOCs contaminating groundwater were decomposed by microorganisms living in soil and water. Tetrachloroethylene decomposes into cis- and trans-dichloroethylene through trichloroethylene decomposition, and 1,1,1-trichloroethane decomposes into 1,1-dichloroethylene and 1,1-dichloroethane. For this reason, dichloroethylene or dichloroethane was sometime detected in the field survey of VOC polluted groundwater performed by the Japanese Ministry of the Environment, often at a low concentration. These compounds have guideline values for drinking water recommended by WHO, in view of their health effects on humans.

Compounds	Guideline value (µg L <sup>-1</sup> )	Remarks
chlorinated alkanes		
carbon tetrachloride	2	
dichloromethane	20	
1,2-dichloroethane	30	for excess risk of 10 <sup>-5</sup>
1,1,1-trichloroethane	2000(p)	
chlorinated ethenes		
vinyl chloride	5	for excess risk of 10 <sup>-5</sup>
1,1-dichloroethene	30	
1,2-dichloroethene	50	
trichloroethene	70(p)	
tetrachloroethene	40	
aromatic hydrocarbons		
benzene	10	
toluene	700	
xylenes	500	
ethylbenzene	300	
styrene	20	
benzo(a)pyrene	0.7	for excess risk of 10 <sup>-5</sup>
chlorinated benzenes		
monochlorobenzene	300	

1,2-dichlorobenzene	1000	
1,4-dichlorobenzene	300	
trichlorobenzenes(total)	20	
epichlorohydrin	0.4(p)	

(p): Provisional guideline value

Table 2. WHO Guidelines for Drinking-Water Quality for VOC

### 3.2. Raw materials of plastics

Raw materials, additives, plasticizers and hardeners of plastics often contaminate water during the manufacturing process, or through the use of plastic products. The plasticizer of PVC (polyvinyl chloride), di (2-ethylhexyl) phthalate (DEHP) and di (2-ethylhexyl) adipate (DEHA) are detectable in waste water and surface water at ng to  $\mu\text{g L}^{-1}$  levels. The WHO Guidelines for Drinking-Water Quality recommend a value of  $8 \mu\text{g L}^{-1}$  for DEHP,  $80 \mu\text{g L}^{-1}$  for DEHA and a proposed value of  $0.4 \mu\text{g L}^{-1}$  for epichlorohydrin. Bisphenol A, which is a raw material for polycarbonate plastics, and phthalates are suspected to be endocrine-disrupting chemicals. Monitoring the presence of these compounds in Japanese water area, bisphenol A, DEHP and DEHA were detected in 100% of wastewater samples and more than 60% of those from other areas such as rivers, lakes and ponds. WHO also recommended guideline values for other substances such as acrylamide of  $0.5 \mu\text{g/L}$ , hexachlorobutadiene of  $0.6 \mu\text{g L}^{-1}$ , EDTA of  $200 \mu\text{g L}^{-1}$ , nitrilotriacetic acid (NTA) of  $200 \mu\text{g L}^{-1}$  and tributyltin oxide of  $2 \mu\text{g L}^{-1}$ .

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### **Biographical Sketch**

**Yoshiteru Tsuchiya** is a Lecturer in the Faculty of Engineering of Kogakuin University, where he has been in his present post since 2000. He obtained a Bachelor Degree in Meiji Pharmaceutical College in 1964. He worked for Department of Environmental Health in Tokyo Metropolitan Research Laboratory of Public Health until 2000. In the meantime, he obtained a Ph.D in Pharmaceutical Sciences from Tokyo University. From 2002 to 2003, he worked for the Yokohama National University Cooperative Research and Development Center as Visiting Professor.

He has written and edited books on risk assessment and management of waters. He has been the author or co-author of approximately 70 research articles. He is a member of Japan Society on Water Environment, Pharmaceutical Society of Japan, Japan Society for Environmental Chemistry, Japan Society of Endocrine Disrupters Research, and International Water Association (IWA).