Persistent Organic Pollution and Arsenic Contamination in Asia Pacific Water: Case Study of Emerging Environmental Problems in Vietnam

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Abstract : This paper provides a comprehensive overview of the present status of several environmental problems caused by emerging toxic substances such as persistent organic pollutants (POPs), endocrine disrupting chemicals (EDCs), and arsenic in various environmental media in Vietnam. Monitoring data reported during the 1990s demonstrated elevated contamination of DDTs in most of these compartments in Vietnam. Studies in frame of the Asia-Pacific Mussel Watch Program revealed that fish, mussels and resident birds from Vietnam contained higher concentrations of DDTs as compared to other countries in region, suggesting the role of Vietnamese environment as a significant emission source of DDT in the Southeast Asian region. The estimated dietary intakes of PCBs and DDTs for Vietnamese were relatively high among Asian developing countries, suggesting potential risk for humans posed by thesechemicals. Widespread contamination of some endocrine active compounds such as alkylphenols and phthalates was observed at various sites along the coasts of northern and middle Vietnam. The presence of significant source of bisphenol-A along Red River estuary was revealed with the concentrations comparable to those reported for developed nations. A case study on seasonal variation of alkylphenols and phthalates in surface water of river delta and estuary of north and middle Vietnam indicated the differences in distribution of these compounds between dry and rainy seasons. Higher concentrations of alkylphenols and phthalates were found in dry season in estuary; while the contrasting pattern was observed in the river delta, showing elevated residues in rainy season. This result suggests the different behavior of alkylphenols and phthalates in river delta and coastal environment. From ecotoxicological perspectives, concentrations of bis-phenol A and di(2-ethylhexyl)phthalates [DEHP] in surface water from some locations in Vietnam exceeded the guideline values for Ecotoxicological Effects and the Environmental Risk Limit, respectively, suggesting potential for toxic implications on aquatic wildlife.

Widespread and elevated arsenic contamination was discovered inour recent surveys in groundwater in a large area of suburban areas of Hanoi city, the capital of Vietnam. The most recent investigation in 4 villages showed about more than 50 % of groundwater samples contained As concentrations exceeding 50 g/L (the WHO and Vietnamese standard). In particular, in Son Dong villages, 58 % of samples analyzed contained As concentrations higher than 200 g/L. Good correlations were found in As concentrations in water and hair and urine of peoples in corresponding families, suggesting the chronic exposure to As by people living in As-contaminated ground water areas. In Son Dong village, As levels in hair (mean: 1.7 mg/kg dry wt) and urine (g/g creatinine) exceeding the reference values recommended by WHO, suggesting potential for human risk posed by long term accumulation of As in human body. Future studies should be focused on the time trends of POPs and EDCs in biota in Vietnam in order to predict future trend of contamination and to reveal new clues for understanding possible toxic impacts on aquatic organisms. The issues of arsenic contamination in groundwater and their chronic toxic implications on human health should be systematically investigated in the future.

Keywords : Arsenic, Bis-phenol A, DDTs EDCs, POPs

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Introduction

Environmental pollution caused by man made toxic chemicals has been of great concern over the last four decades in many industrialized nations. In tropical region, with rapid economic growth, environmental problems arising from the uncontrolled usageof toxic chemicals as pesticides, untreated domestic and industrial waste discharges has become common in developing countries. Vietnam is located at the center of the South East Asian region. There are two major river deltas: the Red river delta in the north and Mekong river delta in the south. These regions are inhabited by approximately 28 million people, and are among the most populous areas in the world.

Persistent Organic Pollutants (POPs) are chemicals that have high persistency in the environment, high bioaccumulation through the food chain and pose risk of causing adverse effects to humans and wildlife. Widespread contamination by POPs and their toxic effects on wildlife and humans have been major concerns and have received considerable attentionduring the past four decades. In developed nations, the environmental issues concerning POP pollution and their fate, behavior and toxic effects have been extensively investigated. On the other hand, natural contamination of groundwater by arsenic has become a crucial water problem, particularly in Bengal Delta region (Bangladesh and West Bengal, India) (Nickson *et al*., 1998). Given similar geological characteristics and groundwater composition of Red river delta and Bengal Delta, elevated contamination ofAs in groundwater is anticipated. In this context, monitoring data of POPs and arsenic in Vietnam play a key role in understanding their fate, transport and toxic potential to humans, wildlife and environmental quality. In this article, monitoring data reported for Vietnam - a tropical developing country in Southeast Asian region, during the last decade are reviewed. This paper briefly reviews the data of major compounds of concern such as DDT and its metabolites (DDTs), hexachlorocyclohexane (HCHs), polychlorinated biphenyls and some other contaminants such as polycyclic aromatic hydrocarbons (PAHs), alkylphenols, which were reported by various laboratories during the 1990s. In addition, groundwater contamination by As and their human exposure were also reviewed on the basis of the recent investigation conducted in our laboratory.

Levels, patterns and toxic implications of POPs and EDCs in Vietnamese environment

During the recent decade, extensive monitoring surveys have been made to determine the residueconcentrations in various environmental compartments in Vietnamese environment such as air, water, sediments, soils and biota. The present paper briefly reviews the monitoring data from these laboratories and the results are summarized in Table 1. These observations indicate clearly that persistent organic compounds occurred in most of the environmental compartments in Vietnam at detectable levels. Survey conducted in the early 1990s in air water and sediments from the same locations in Hanoi, Hue and Hochiminh city made clear the distribution of organochlorine pesticides and PCBs along north and south of Vietnam (Iwata *et al*., 1994). Elevated concentrations of HCHs were observed in air and water, while higher concentrations of DDTs were noticed in biological samples. In Read River and Duong River, DDT concentrations

were found to be very high in a survey on 1998/99 (Hung and Thiemann 2002) as compared to that reported in 1995 (Iwata *et al*., 1994). Although the water samples might not be collected at the same specific locations along the Read River region, this phenomenon indicates the widespread and high contamination by DDTs in northern Vietnam. Similarly, DDTs and HCHs were also reported at higher levels in recent surveys (Viet *et al*., 2000, Nhan *et al*., 1999, 2000) compared to the previous examination (Iwata *et al*., 1994). Elevated DDT concentrations in water and sediments from Red River delta region in recent years suggests the fresh input of this pesticide into the watershed of Red River.

An extensive monitoring study in water samples conducted by Hung and Thiemann, (2002), examined various organochlorine pesticides in a wide range of the capital of Hanoi, including lakes, irrigation canals and Read River and Duong River, the two largest river flows through the northern delta zone of Vietnam. Elevated concentrations of DDTs and HCHs were found in various locations along the Read River, while residues in lakes from Hanoi were significantly lower. This result suggests the different behaviors of persistent semivolatile compounds in tropical lakes and rivers, showing shorter residence time of these compounds in lakes than in rivers. Higher contamination in coastal areas of the tropical region was also common in tropical developing belt in Asian − Pacific (Iwata *et al*., 1994; Tanabe *et al*., 1994). In the south of Vietnam, elevated concentrations of DDTs, HCHs, CHLs and PCBs were found in sediment from Hochiminh city and the downward gradient was observed from the upper to lower stream of the estuary (Iwata *et al.* 1994). Interestingly, this result was inconsistent with those observed in water samples from the Red River in the northern region as discussed above. This suggests that the evaporation of organochlorines from sediment, which is facilitated by high temperature in tropical southern region of Vietnam, could be key factor controlling the fate of semivolatile compounds. In this context, sediment estuary areas may be less contaminated as compared to the locations in the city.

In recent years, there is a growing concern over the toxic effects of organic pollutants to disrupt the endocrine functions of wildlife and various reproductive abnormalities has been linked to exposure to elevated levels of these contaminants (Colborn *et al*., 1993). Alkylphenolic compounds are among the groups of chemicals that are known to exhibit the endocrine disrupting effects. Our laboratory has recently conducted surveys to examine the occurrence of alkylphenols including bis-phenol A from different locations in the Read River delta and along the estuary and the Huong River, middle part of Vietnam (Viet *et al*., 2002). Our results demonstrated ubiquitous occurrence of alkylphenolic compounds in the Red River delta and along its estuary (Figure 1). Higher concentrations of alkyphenols were found in the water collected in Red River delta as compared to its estuary, suggesting the presence of the sources of these phenolic compounds in the river delta. Interestingly, concentrations of bis-phenol A in Read River water, north Vietnam were comparable to those reported for some developed countries such as Japan, United States and some western European countries (Viet *et al*., 2002). Elevated contamination of bis-phenol A in Vietnam suggests the need for further studies. To our knowledge, our results represent the first extensive study of alkylphenols in Vietnam.

Table 1. Residue concentrations of persistent organic pollutants (POPs) in biological samples from Vietnam reported during the recent decade.

Compound	Location	Survey year	Environmental compartment			Reference
			Mussel/Bivalve (ng/g lipid wt)	Fish (ng/g lipid wt)	Bird (ng/g lipid wt)	
DDTs	Hanoi	1990		1900(680-4000)		Kannan *et al*., 1995
	Red River delta	1995/96	(100-400)	13000		Nhan *et al*.,, 1998
	Red River estuary	1997		4200	6200(1100-13000)	Minh *et al*., 2002
	Red River estuary	1997			2900(750-6800)	Minh *et al*., 2002
	Red River estuary	1997	4000(250-31000			Monirith *et al*., 2000
	Northern coast of Vietnam	1997	(590-1200)			Nhan *et al*., 1999
	Phu Da	1990		1100(205-2700)		Kannan *et al*., 1995
	Hochiminh city	1990		1100(89-4100)		Kannan *et al*., 1995
HCHs	Hanoi	1990		121(47-210)		Kannan *et al*., 1995
	Red River estuary	1997		120	150(23-310)	Minh *et al*., 2002
	Red River estuary	1997			330(20-1700)	Minh *et al*., 2002
	Red River estuary	1997	5.4(3.6-9.1)			
	Northern coast of Vietnam	1997	(71-2400)			Nhan *et al*., 1999
	Phu Da	1990		48(32-74)		Kannan *et al*., 1995
	Hochiminh city	1990		105(33-200)		Kannan *et al*., 1995
PCBs	Hanoi	1990		580(270-950)		Kannan *et al*., 1995
	Red River estuary	1997		110	780(250-2400)	Minh *et al*., 2002
	Red River estuary	1997			530(82-1600)	Minh *et al*., 2002
	Red River estuary	1997	130(18-310)			
	Northern coast of Vietnam	1997	840			Nhan *et al*., 1999
	Phu Da	1990		630(160-1300)		Kannan *et al*., 1995
	Hochiminh city	1990		1400(190-3100)		Kannan *et al*., 1995

Values of concentrations were rounded to 2 significant digits for comparison among studies. Single values indicate mean concentrations. Figures in parentheses represent range concentrations.

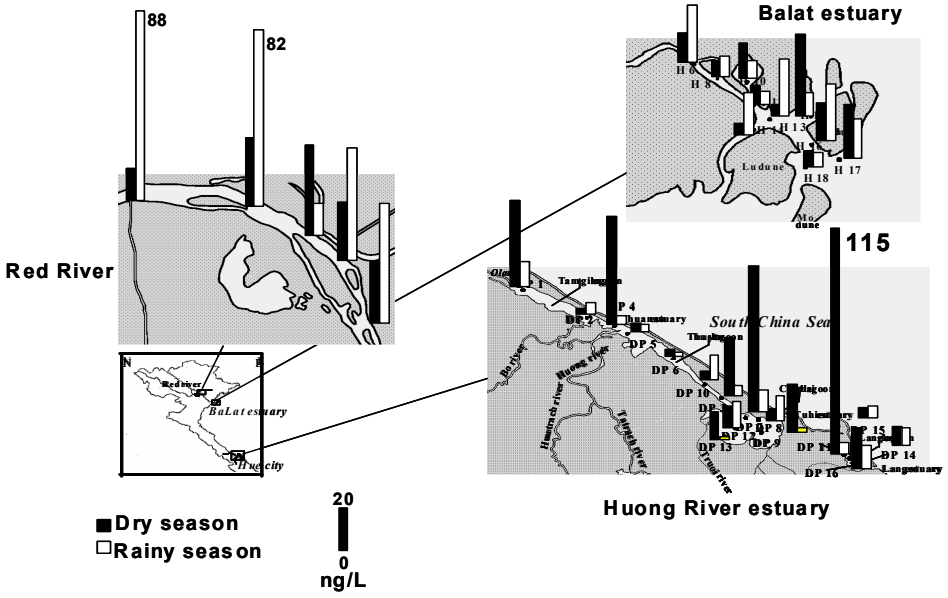

Figure 1. Widespread occurrence of alkylphenols in Red River delta and estuary (northern part) and Huong River estuary (middle part) of Vietnam

Surveys conducted during the last decade comprising the data for POPs in biota samples in Vietnam was within the Asia-Pacific Mussel Watch Program (Kannan *et al*., 1995, Monirith *et al*., 2000; Minh *et al.*, 2002, Sudaryanto *et al*., 2002). Surveys in fish carried out in 1990s revealed the significant contamination by DDTs and PCBs in fish from Vietnam as compared to other countries in Asia-Pacific (Kannan *et al*., 1995). Some species of resident birds collected from Red River estuary contained elevated DDT residues, suggesting again the extensive usage of this insecticide in the past and very recently in Vietnam. Data for fish and mussels also revealed higher contamination of DDTs in Vietnam as compared to other countries in region such as Cambodia, Indonesia, Malaysia, Thailand and India. Statistical data indicate the quantity of DDT used in Vietnam for malaria control from the period of 1957-1990 was 24,042 tons, in which 800 tons of DDTs were used during 1986-1990. In addition, DDTs was still used until 1995 in Vietnam with the amount of 445 tons during 1992-1995 (Sinh *et al*., 1999). Considering these data, elevated levels of DDT found in various environmental compartments in Vietnam suggests that Vietnam is a potential source of DDT in the Southeast Asian region.

Despite extensive studies have been made in recent years to understand the distribution and behavior of persistent organochlorines in Vietnam, no systematic research has been conducted on temporal trend of contamination. In frame of the "Environmental Monitoring and Analysis in East Asian Region: Technology Transfer and Environmental Governance Program"funded by United Nation University, our laboratory has started preliminary survey on temporal trend of contamination by DDTs and HCHs in water and sediments collected from Red River Delta, North Vietnam during 1995-2001 (Figure 2). DDT residues in water have declined relatively rapidly during 1995-1998 and remained constant until recent years at the levels below 20 ng/L (Figure 2). Concentrations of DDTs in sediments also exhibited declining trend but to a lesser extent. From the period of 1997–2001, concentrations of DDTs in sediments declined slowly. Considering the rapid decline

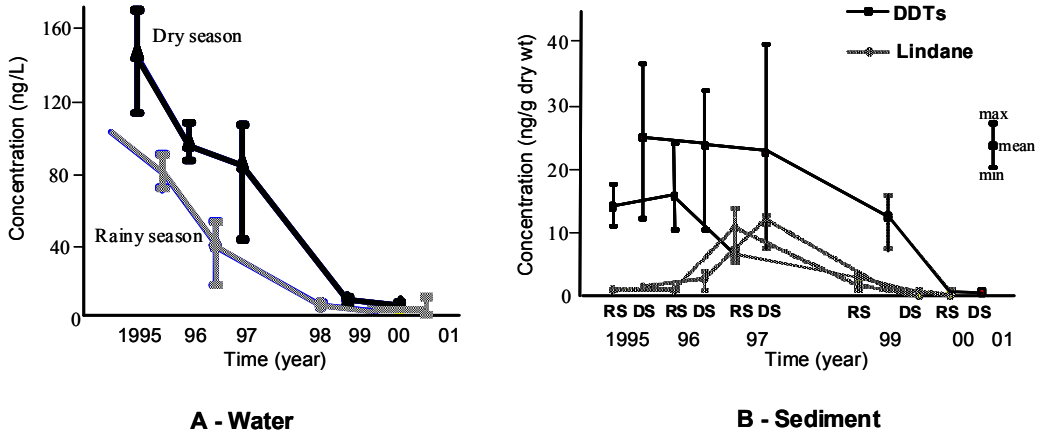

Fig. 2. Temporal trends of DDTs and HCHs in water and sediments from Red River delta during 1995–2001. A, water; B, sediments. RS and DS; rainy and dry season, respectively

of DDTs found in many countries in the world, result observed in Vietnam as well as other tropical developing countries such as India (Senthilkumar *et al*., 1999) indicate slow or event increasing trend. In this context, further studies would be necessary to trace the future trend of contamination in Vietnam as well as other tropical developing countries in region.

In the respect of environmental and human health implications, elevated residue levels of some endocrine active compounds such as bis-phenol A and di(2-ethylhexyl)phthalates [DEHP] observed in surface water in some locations in Vietnam is of concern. Concentrations of bis-phenol A in water samples from Red river delta and the river estuary (ranged from < 0.5 −1330 ng/L) were comparable to higher than those in surface water of many other location in the world, suggesting the presence of significant source of this compound along the Red river estuary (Viet *et al*., 2002). The concentrations in some water samples exceeded the ecotoxicological effect value of 1000 ng/L (predicted no-effect concentration in water − $PNEC_{water}$). Likewise, water from Huong River estuary, middle of Vietnam contained elevated concentrations of DEHP, and in some locations the residues were beyond the environmental risk limit value of 190 ng/L (Viet *et al*., 2002). This result suggests potential for toxic implications for aquatic wildlife and warrants further systematic monitoring and toxicological studies.

For human health point of view, surveys in foodstuffs in early 1992 revealed relatively high intakes of DDTs and PCBs by Vietnamese population (Kannan *et al*., 1992). DDT intakes estimated from foodstuffs for Vietnamese were higher than some other developed nations such as Japan, USA and Germany (Kannan *et al*., 1992). Subsequent survey in fish continuously indicated elevated intakes from fish by Vietnamese population (Monirith *et al*., 2000). The magnitude of contamination by DDTs in Vietnam is of concern and required studies. Elevated human exposure may continue to be still high in future in Vietnam. The lack of information and knowledge of toxic contaminant residues in foods is one of the most causative factors of chemical intoxication of humans in developing countries including Vietnam. Research and cooperation in suitable technology and safety equipment are critically necessary in these countries.

Arsenic contamination in groundwater and its implications on human health

During the last 5 years, our laboratory has been conducting extensive surveys to examine the status of contamination by arsenic in groundwater from several suburb areas around Hanoi city, the capital of Vietnam. Initial survey conducted during September 1999 −July 2000 in 68 private tube-wells from upper aquifers in rural areas of 4 districts: Dong Anh, Tu Lien, Gia Lam and Thanh Tri showed great variability of As levels in groundwater, ranging from 1 − 3050 g/L (mean: 159 g/L) (Berg *et al*., 2001). In highly affected area (Thanh Tri district), 89 % of the arsenic concentrations exceeded the US EPA and Vietnamese standards of 50 g/L; and elevated As concentrations were noticed on both sides of the Red River, indicating the source of contamination is widely distributed over a large area. In Thanh Tri district, very high concentrations of arsenic was observed (mean 432 g/L, n = 45). We suggest that groundwater abstraction from peat-rich aquifers may enhance dissolution of arsenic-rich iron oxyhydroxides and consequently lead to an increase in arsenicconcentrations in the upper

aquifers (Berg *et al*., 2001).

On the basis of the preliminary survey as discussed above, in the next stage we have extended our research on the "hot spot"sites in Thanh Tri district and nearby areas; and examined arsenic residue concentrations in hair and urine of humans living in these areas (Table 2). There was a good correlation between arsenic levels in groundwater and in humans samples of the corresponding families. In highly affected site (Son Dong), elevated concentrations of arsenic in groundwater, hair and urine were noticed, and these levels were apparently higher than those in reference site (Thuong Cat). In Son Dong, 70 % arsenic concentrations in hair exceeded 1 mg/kg, particularly; concentrations reached the level of 10 mg/kg. This is comparable to those reported in hair of people living in long term and highly affected area in West Bengal and Bangladesh (Anawar *et al*., 2002; Chakraborti *et al*., 2003) (Table 3). Average arsenic concentration in hair (1.71 mg/

Table 2. Arsenic concentrations in groundwater, hair and urine of peoples living in reference sites (Thuong Cat), As affected (Van Phuc) and highly affected (Son Dong) areas.

Location	Groundwater (mg /L)	Hair (mg/kg)	Urine (mg /g creatinine)
Thuong Cat			
n	20	23	10
Mean	9.6	0.27	77
Range	< 1.0 - 141	0.04 - 0.54	23 - 141
Van Phuc			
n	21	29	
Mean	132	0.5	
Range	< 1.0 - 357	0.01-2.75	
Son Dong			
n	54	200	50
Mean	252	1.71	146
Range	4.7 - 434	0.16 - 10.4	30 - 394

Table 3. Arsenic contamination in groundwater, hair and urine of people from some highly affected areas in Vietnam and other regions in the world

Location	Groundwater (mg /L)	Hair (mg/kg)	Urine (mg/g reatinine)	Reference
Son Dong, Vietnam	252 (5 - 434)* 39 % of samples > 300	1.71 (0.16 - 10.4)	146 (30 - 394)	present study
Bihar, West Bengal, India	20 % of samples > 300	4.37 (1.2 - 13.3)	598 (279 - 1474)	Chakraborti *et al*., 2003
Bangladesh	10 - 9000	1.1 - 19.9	50 - 9420	Anawar *et al*.., 2002
WHO reference value	10	0.2	< 100	

* Mean (Range)

kg) and urine (146 g/g creatinine) exceeded the reference values recommended by WHO (0.2 mg/kg for hair and < 100 g/g creatinine for urine, respectively). This result suggests chronic exposure to As by people living in Son Dong area, and the source of As is from groundwater consumption.

Household sand filtration: a preliminary low cost measure to reduce risk of chronic arsenic exposure

The above results demonstrate clearly that arsenic contamination in groundwater in the upper aquifers of Red River Delta region has been widespread over a large area with considerable seasonal and spatial variations. Our result also suggests chronic exposure of As by people living in highly contaminated areas. In fact, people using both rain and groundwater for drinking in rainy season. In dry season, however, they have to use groundwater directly pumped from tubewells and thus, they are consuming highly arsenic contaminated water.

With effort to reduce the risk of such an elevated exposure, our laboratory in cooperation with Swiss Federal Institute for Environmental Science and Technology (EAWAG) has carried out a research to evaluate an existing simple, low cost method which is widely used in the rural areas in Vietnam to reduce the concentrations of arsenic in raw groundwater which is directly pumped from the upper aquifers of some highly affected areas such as Van Phuc and Son Dong villages (Luzi *et al*., 2004). This isthe sand filter, which is easily constructed in almost every family in the arenic contaminated areas. The sand filters comprise two superimposed containers: the upper container is filled with sand and the lower one serves to store the filtered water. Raw groundwater is pumped directly from the tubewell into the upper containers, trickles through the sand into the lower storage tank. Arsenic removal mechanism is governed by the precipitation of Fe hydroxides which form a coating on the sand surfaces. As adsorbs to the Fe hydroxides and remains immobilized under oxic conditions (Luzi *et al*., 2004). This method is very simple, because these sand filters can be prepared using locally available sand, operated without adding chemicals, is able to treat a reasonable amount of raw groundwater within a short time, and can be easily operated by the affected communities with little effort of education. Figure 3 shows results of the status of groundwater quality before and after the sand filtration process. In most of the household sand filters, arsenic concentrations were reduced drastically, and in many samples the levels were below the former Vietnamese standard (50 g/L). At least about 90 % of tested sand filters in affected areas were able to lower As concentrations to the level of 50 g/L. Subsequent quantitative analyses in the laboratory showed that sand filtrated groundwater contained an average concentration of 23 g/L in Van Phuc and 83 g/L in Son Dong. While sand filtration is able to reduce As concentrations effectively, many groundwater samples still contained higher As concentrations exceeding WHO (10 g/L) and even the former Vietnamese Standard (50 g/L) in highly affected area such as Son Dong village. In this context, ratio of Fe/As in groundwater and phosphate (PO_4^{3-}) concentrations could be important factors influencing the effectiveness of sand filters. It is estimated that in order to reduce the As levels below 50 g/L, the Fe/As ratio should be at least 50 and phosphate concentrations should be as low as < 2.0 mg/L.

Sand filtration technique is proved to be very

effective tool and is viable option for arsenic mitigation of iron-rich groundwater in Vietnam. Therefore, such a simple and economic technique should be further developed and applied in a large scale of highly affected areas in Vietnam to prevent the long term health effects caused by chronic elevated arsenic exposure.

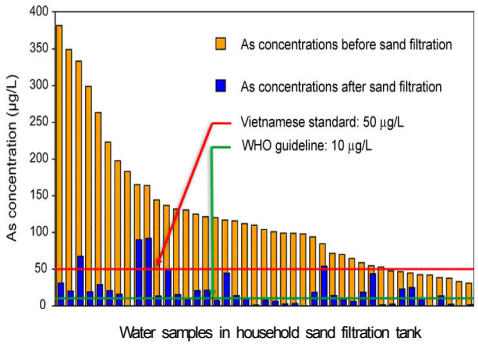

Fig. 3. Status of arsenic contamination in groundwater after and before sand filtration

Conclusions

Existing information regarding occurrence of POPs in air, water, sediments, soils and biota suggest elevated contamination in DDTs in most of these compartments. On the other hand, arsenic contamination in groundwater may become a key environmental problem in Vietnam in the 21st century. Comprehensive monitoring surveys along the coasts from north to south of Vietnam are required to provide further insights into the distribution and fate of POPs in recent years. The role of Vietnamese environment as potential emission sources of DDT in the Southeast Asian region is of concern and deserves future studies on temporal trend of contamination. A large scale study on arsenic contamination and its possible toxic effects on human health are urgently needed. Atthe same time, low cost, effective technologies to lowering arsenic concentrations in groundwater are critically important to reduce the risk of chronic arsenic poisoning of millions of people in Vietnam.

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