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Chapter 2

Emission, Contamination and Exposure, Fate and Transport, and National Management Strategy of Persistent Organic Pollutants in South Korea

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Abstract

Public concern over persistent organic pollutants (POPs) re-emerged in the 1990s due to studies describing endocrine disrupting effects of some POPs. While monitoring data suggested that concentrations of contaminants were less in South Korea than in more industrialized countries, the public perception was that there were significant risks posed by POPs. This perception may have resulted from inaccurate and insufficient information about the status of POPs in South Korea. The South Korean government, as a signatory authority of the Stockholm Convention, is obliged to submit a national implementation plan to ban or minimize POPs emissions. To date, little has been known regarding the overall POPs status including inventories, usage patterns, sources, emission, fate, and distribution in South Korea. To assess the status of four emerging POPs as well as the 12 existing classical POPs in South Korea we have compiled and reviewed all the available literature published since the mid-1990s on POPs in South Korea. We present and discuss: (1) emission inventories of individual POPs; (2) concentrations of the various POPs in various compartments of the environment; (3) conducted ecosystem and human exposure assessments; (4) report a case study of fate and multi-media transport of POPs; and finally (5) propose an appropriate strategy to minimize the risks of POPs in South Korea. In brief, concentrations of POPs were found to be relatively small, compared to most other industrialized countries. In fact, concentrations of many of the classical POPs were less than the environmental quality criteria suggested by government agencies, except for a few 'hot spots'. However, due to a lack of sufficient information, the status and trends of PCDDs/DFs,

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DDTs, PBDEs and PFAs could not be assessed. Additional information from monitoring studies would be needed for certain locations. For instance, the emission of dioxin-like compounds, expressed on a per area basis, was estimated to be one of the greatest in the world, even though the recent governmental efforts to reduce the emission have resulted in decreased releases of several classes of POPs. Concentrations of some perfluoroalkylated compounds (PFAs), in some freshwater locations were found to be some of the greatest in the world. Identification of hot spots followed by remediation (for classical POPs) and nationwide monitoring surveys (for emerging POPs) would be strongly needed.

2.1. Introduction

The 'dirty dozen' or the 12 legacy organochlorine substances, consist of two byproducts (polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans (PCDDs/DFs)), one industrial product (polychlorinated biphenyls (PCBs)), and nine pesticides (DDTs, chlordanes, heptachlor, aldrin, endrin, dieldrin, toxaphene, mirex, and hexachlorobenzene (HCB)) (Stockholm Convention on Persistent Organic Pollutants, 2001). PCBs and HCB are also emitted as byproduct or impurities (UN-ECE/EMEP, 2002; Breivik et al., 2004). These dirty dozen persistent organic pollutants (POPs) were prioritized for global action by the Stockholm Convention on POPs, developed under the auspices of the United Nations Environment Program (UNEP). The criteria for prioritization included persistence, bioaccumulation potential, and toxic potency (PBT) in addition to their potential for long-range transport (LRT).

Since 1998, the United Nations/Economic Commission for Europe (UN/ECE, 1998) has included, in addition to the dirty dozen POPs, hexachlorocyclohexanes (HCHs), polycyclic aromatic hydrocarbons (PAHs), chlorodecone, and hexabromobiphenyl. The New Protocol on Persistent Organic Pollutants to the Convention on Long-Range Trans-boundary Air Pollutant (CLRT protocol) has served as the basis for these criteria. Besides the 12 existing POPs, the emerging POPs such as polybrominated diphenyl ethers (PBDEs) (Palm et al., 2002; Tanabe, 2004) and perfluoroalkyl acids (PFAs) (Giesy and Kannan, 2001; Renner, 2001a,b; Prevedouros et al., 2006) are also of increasing concern due to their extensive worldwide usage as well as possible PBT and LRT characteristics. Alkylphenols (APs) such as nonylphenol (NP) and octylphenol (OP) with endocrine disrupting properties are also stable in the environment with a half-life of approximately two months in water and in the order of years in sediments (Mackay et al., 2000; Ying et al., 2002).

National concern regarding POPs in South Korea was amplified in the mid-1990s when dioxins, carcinogenic, and endocrine disrupting compounds (EDCs), were reported to be emitted primarily from waste incinerators throughout South Korea. The total number of incinerators in South Korea reached a maximum of 16,000 in 1998. During the same period, it was reported that some of the POPs, including dioxins, could cause effects, such as breast cancer and reduced semen quality and potentially be 'environmental hormones'. The South Korean government classified them as priority pollutants for regulatory actions in South Korea. Based on a survey conducted in 2003, 91.4% of South Koreans responded that they had heard the term 'environmental hormone' at least once in their life (Kim, 2003). A survey of risk communication and perception in environmental problems (Kim et al., 2002a), found that over 82% of 574 people surveyed thought that the South Korean environment was polluted. Nevertheless, little was known in the public surveyed (i.e., <2.5 points of total 13 points) about the causes, pathways, toxic effects, and there was no personal action plan to decrease exposure to dioxins and other POPs. Most of the people responded that their knowledge regarding POPs was acquired from the mass media rather than from the scientific research papers. Recent nationwide monitoring studies have found that concentrations of POPs such as PCBs and organochlorine pesticides (OCPs) in the environment, are less than those in other industrialized countries such as the USA and Japan, and generally less than the established criteria, except for some hot spots (Kim et al., 2002b; Hong et al., 2006). Thus, in addition to having only limited information on the status and trends of contaminants and their potential toxic effects with which to conduct risk assessments and implement risk management, there was also a gap in risk communication. Thus, the public could not assess the real risks that POPs may pose to the health of the South Korean population and ecosystem.

In addition to public concern, international conventions to reduce or eliminate emissions of POPs in South Korea took effect in May 2004. The South Korean Government, as a signatory participant of the Stockholm Convention on POPs, plans to ratify the convention. Then the government is obliged to submit a National Implementation Plant (NIP) to minimize POPs emission and update the plan and provide a status report every two years from the time of ratification (Stockholm Convention on Persistent Organic Pollutants, 2001). Therefore, it is inevitable that a scientific assessment and review of the domestic status and history of the POPs pollution including emissions, contamination levels (or distribution), exposures, and risks will be needed. This information will be disseminated to the public and also used to develop efficient control

strategies to minimize exposure of humans and the wildlife to the POPs. Here, we facilitate this process by bringing together in one place, for the first time, a comprehensive compendium of information available on POPs in Korea.

Studies of POPs in South Korea can be arbitrarily categorized into 3 periods: from the 1960s to mid-1990s, mid-1990s to 2000, and 2001 and later. The first study of POPs in South Korea to appear in the literature was about OCPs residues in South Korean foodstuffs performed by the 'Rural Development Administration' in 1967 before the beginning of restriction of the use of OCPs in South Korea (Lee, 1982). Until the mid-1990s, most studies of POPs in South Korea had been conducted on OCP residues in agricultural products such as vegetables (Park and Yoo, 1972; Park et al., 1974; Kim et al., 1981), foods (Kim and Lee, 1980; Lee, 1982; Ryu et al., 1986), agricultural soils (Park and Ma, 1982; Choi et al., 1987), and marine food/sediment (Lee et al., 1976, 1977; You and Park, 1984; Suh et al., 1986). The most extensive collection of data during the 1960s to mid-1990s appeared on OCPs and specifically for calculation of daily intake rates (Lee, 1982) and on the investigation of OCP residues in 236 agricultural soils (Park and Ma, 1982). Due to these initiatives and some pioneering efforts, the uses of POPs have been restricted in South Korea since late 1960s. Because early analytical techniques involving packed GC columns were less accurate and sensitive than more modern methods, the accuracy of the historical analyses of OCPs is questionable. Since the mid-1990s, several studies by academic institutions applied state-of-the-art analytical techniques to quantify POPs. During this period, in 1996, standard analytical methods were established for quantifying dioxins in air. The development of sensitive and standard analytical techniques improved the accuracy of POPs quantification, which made international comparisons of concentrations possible. These data were available for robust meta-analyses. Governmental efforts began only in the last decade for through investigation and effective management of POPs. Since 1999, the South Korean government and the Ministry of Environment (KMOE) began nationwide monitoring of POPs in the environment and in humans. Since 2001, comprehensive projects have been undertaken to establish the national emission inventory of some priority POPs (dioxins in particular).

While comprehensive research and surveys were delayed, or lacking, no significant effort had been put toward the overview of the existing data of emissions, sources, distribution, exposure, and fate of POPs. Lee and Kim (1999) collected published data on concentrations of EDCs for almost all environmental media and human tissues and performed a meta-analysis to examine the relationship between the sperm quality of South Korean

males and exposure to EDCs. Environmental concentrations and rates of emission of POPs, such as PCBs and OCPs, have been reviewed by KMOE (KMOE, 2004a). These preliminary studies were limited to the collection of data on concentrations in various environmental compartments.

In an attempt to provide an overview and assessment of the POPs in South Korea, we have conducted a comprehensive literature search and compiled the existing POPs data (including the emerging POPs) from surveys conducted since the mid-1990s in South Korea. This chapter presents: (1) emission inventories of individual POPs; (2) concentrations in various environmental including humans; (3) exposure assessment in the ecosystem and humans; (4) a case study of fate and multi-media transport of POPs; and finally, (5) a proposal for a strategy to minimize releases and ultimately eliminate POPs in South Korea.

2.2. Methodology: Literature survey and target POPs

For this chapter, we searched the peer-reviewed, open, scientific literature by using the Science Citation Index (SCI) and 'ScienceDirect', 'SCIRUS', and 'SpringerLink'. Domestic articles that would not be listed in these citation services were collected using online websites of 33 South Korean science societies and 'Korean studies Information Service System (KISS)' of Korean Studies Information Corporation. Literature collected includes: (1) 91 articles published in SCI journals; (2) 57 domestic articles; (3) 40 research reports performed or funded by governmental agencies; and (4) 4 dissertations (Fig. 2.1). Reports quoted consisted of 25 regular nationwide monitoring studies (6 by National Institute of Environment Research (NIER), 16 by the South Korea Food and Drug Administration (KFDA), and 3 by the Ministry of Maritime Affairs and Fishery (MOMAF)), 12 emission inventories, and 3 exposures and meta-analyses. Most inventories of emissions were either performed or funded by the KMOE or NIER.

In the present study, 17 compounds including dirty dozen POPs (OCPs, PCDDs/DFs, PCBs), two emerging POPs (PBDEs and PFAs), and two potential POPs (APs and PAHs) in the South Korean environment were reviewed (Table 2.1 and 2.2). Among the 17 POPs, the most studied pollutant was determined to be chlorinated dioxins and dioxin-like compounds such as PCDDs/DFs and dioxin-like PCBs for which occurrence, distribution, contamination level, fate, exposure, and control techniques have been fairly thoroughly investigated. The literature on dioxin research included 42 SCI articles and 57 domestic articles. Because most of

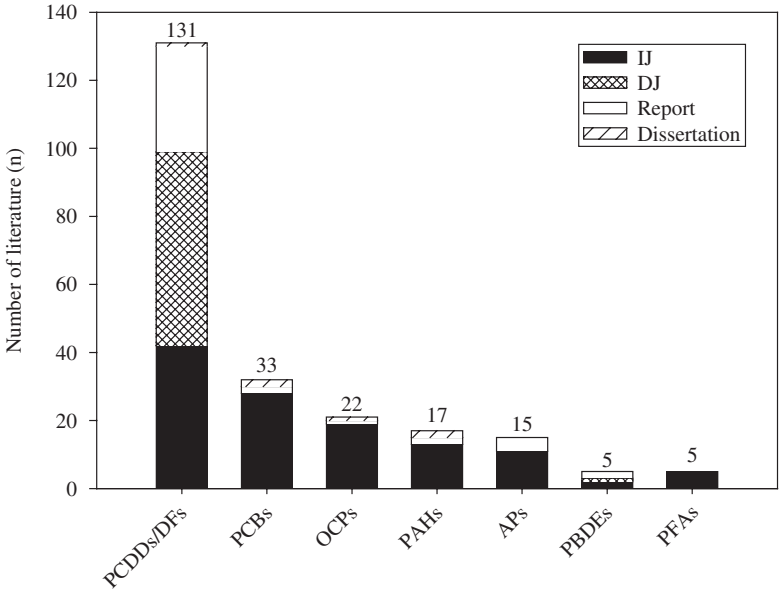

Fig. 2.1. Korean literatures collected for meta-analysis of target POPs substances: SCI article (IJ), Domestic article (DJ), Research report (Report), and PhD dissertation.

the data originated from 13 laboratories certified for dioxin analysis, the domestic articles were also included in this survey. Although 80 domestic articles were reviewed, information from only 33 SCI articles were used for the meta-analysis of PCBs, OCPs, and PAHs because they were considered to provide sufficient information to allow us to describe distribution and concentrations in the environment. Most of these articles reported concentrations in the environment as well as possible sources. In addition to legacy POPs, we also reviewed information on emerging contaminants of concern. Data on alkylphenols were extracted from 11 SCI articles and 4 reports; PBDEs from 2 SCI articles, 1 domestic article, and 2 reports; PFAs from 5 SCI articles. The number and volume of the literature indicates that the South Korean POP studies have primarily focused on legacy POPs (i.e., dioxins, PCBs, and OCPs) while little attention is given to emerging POPs (i.e., PBDEs and PFAs).

The physico-chemical properties of these POPs are illustrated in Fig. 2.2. APs and PFAs (hydrophilic substances) are relatively more water soluble than the legacy POPs, which suggests that these pollutants could easily reside in water column and then can be carried to remote areas by hydrospheric movement (Yamashita et al., 2005).

Table 2.1. Target POPs involved in this study and the regulatory action in Korea

Pollutant category	Major use in Korea	Regulatory action						
		ACMA[a]	TCCA[b]	ISHA[c]	EBA[d]	CACA[e]/ WQCA[f]/ SECA[g]	WCA[h]	Food residual level[i]
Existing POPs								
Polychlorinated biphenyls (PCBs)	Dielectic fluid etc.	–	Ban ('96) (> 50 ppm, '99)	Ban ('03)	Ban ('79)	Specific hazardous chemical	Hazardous waste (> 2 ppm)[k]	–
OCPs[j]								
DDTs	Insecticide	Ban ('69)	Ban (> 1%, '91)	Ban	–	–	–	<0.1 ppm
Chlordanes (CHLs)	Insecticide/additive agent	Ban ('69)	Ban (> 1%, '99)	Ban	–	–	–	<0.02 ppm
Hexachlorocyclohexane (HCHs)	Insecticide	Ban ('69)	Ban (> 1.5%, '91)	Ban	–	–	–	
Aldrin	Insecticide/additive agent	Ban ('69)	Ban (> 0.1%, '99)	Ban	–	–	–	<0.01 ppm
Endrin	Insecticide/additive agent	Ban ('69)	Ban (> 1%, '99)	Ban	–	–	–	<0.01 ppm
Dieldrin	Insecticide/additive agent	Ban ('70)	Ban (> 1%, '99)	Ban	–	–	–	<0.01 ppm
Heptachlor	Insecticide/additive agent	Ban ('79)	Ban (> 6%, '99)	Ban	–	–	–	<0.01 ppm
Toxaphene (Camphechlor)	Insecticide	Ban ('82)	Ban (> 1%, '91)	Ban	–	–	–	–
Endosulfan	Insecticide	–	Ban (> 1%, <'03)	Ban	–	–	–	–
Tetrachlorobenzene (TeCB)	Intermediate in pesticide	–	–	–	–	–	–	–
Pentachlorobenzene (PeCB)	Intermediate in pesticide	–	Ban (> 1%, <'03)	Ban	–	–	–	–
Hexachlorobenzene (HCB)	Domestic no use	–	–	–	–	–	–	–
Mirex	Domestic no use	–	–	–	–	–	–	–
Dioxins/furans (PCDDs/DFs)	By-products	–	–	–	–	–	Restricted release ('97)[l]	–
Coplanar PCBs (coPCBs)	By-products	–	–	–	–	–		–
Hexachlorobenzene (HCB)	By-products	–	–	–	–	–	–	–

Table 2.1. (*Continued*)

Pollutant category	Major use in Korea	Regulatory action						
		ACMA[a]	TCCA[b]	ISHA[c]	EBA[d]	CACA[e]/ WQCA[f]/ SECA[g]	WCA[h]	Food residual level[i]
Emerging POPs								
Polybrominated diphenyl ethers (PBDEs)	Flame retardant	–	–	–	–	–	–	
Perfluoroalkyl acids (PFAs)	Acids of fluoropolymer, rust/oil/ water retardant							
Others								
Alkylphenols (APs)		–	–	–	–	–	–	–
Polycyclic aromatic hydrocarbons (PAHs)		–	–	–	–	–	–	–

[a]ACMA (Agrochemical Management Act); the ban of registration of pesticides, indicating implicitly the ban of import, manufacture, and selling.
[b]TCCA (Toxic Chemical Control Act); the restricted use (until '90) and the ban (from '91) of manufacture, import, and use of non -food additive agent.
[c]ISHA (Industrial Safety and Health Act); the same legal action as TCCA was enacted in 2003.
[d]EBA (Electricity Business Act); the ban of use for electric heater of facilities used PCB -containing dielectic fluid.
[e]CACA (Clean Air Conservation Act); the specific air hazardous chemical.
[f]WQCA (Water Quality Conservation Act); the water quality criteria of PCBs as specific water hazardous chemicals.
[g]SECA (Soil Environment Conservation Act); the air quality criteria of PCBs as specific water hazardous chemicals.
[h]WCA (Waste Control Act); the restriction of waste treatment.
[i]Guideline of Korean Food and Drug Administration (KMOE, 2004).
[j]OCPs (Organochlorine Pesticides).
[k]PCBs as specific hazardous waste should be treated in separated and specific method and reported and confirmed by KMOE administrator.
[l]Release guideline of waste incinerators (see Table 3).

Table 2.2. Target compounds of persistent organic pollutants (POPs) reviewed; PCDDs/DFs, PCBs, OCPs, PAHs, APs, PDBEs, and PFAs, respectively

Abbreviation	Full name
PCDDs/DFs	Polychlorinated dibenzo-*p*-dioxins/-dibenzofurans
TCDD/DF	Tetrachlorinated dibenzo-*p*-dioxin/-dibenzofuran
PeCDD/DF	Pentachlorinated dibenzo-*p*-dioxin/-dibenzofuran
HxCDD/DF	Hexachlorinated dibenzo-*p*-dioxin/-dibenzofuran
HpCDD/DF	Heptachlorinated dibenzo-*p*-dioxin/-dibenzofuran
OCDD/DF	Octachlorinated dibenzo-*p*-dioxin/-dibenzofuran
PCBs	Polychlorinated biphenyls
di-CB	Dichlorinated biphenyl
tri-CB	Trichlorinated biphenyl
tetra-CB	Tetrachlorinated biphenyl
penta-CB	Pentachlorinated biphenyl
hexa-CB	Hexachlorinated biphenyl
hepta-CB	Heptachlorinated biphenyl
octa-CB	Octachlorinated biphenyl
nona-CB	Nonachlorinated biphenyl
deca-CB	Decachlorinated biphenyl
OCPs	Organochlorine pesticides
CBs	Chlorobenzenes
1,2,3,4-TCBZ	1,2,3,4-Tetrachlorobenzene
1,2,4,5-TCBZ	1,2,4,5-Tetrachlorobenzene
PCBZ	Pentachlorobenzene
HCB	Hexachlorobenzene
HCHs	α-, β-, γ-, δ-Hexachlorocyclohexane
α-HCH	α-Hexachlorocyclohexane
β-HCH	β-Hexachlorocyclohexane
γ-HCH	γ-Hexachlorocyclohexane (lindane)
δ-HCH	δ-Hexachlorocyclohexane
CHLs	Chlordanes
PentaCA	Pentachloroanisole
HeptaC	Heptachlor
HeptaCE	Heptachlor epoxide
OxyCHL	Oxychlordane
γ-CHL	γ-Chlordane (*trans*-Chlordane)
α-CHL	α-Chlordane (*cis*-Chlordane)
cis-nonaC	*cis*-Nonachlor
trans-nonaC	*trans*-Nonachlor
Drins	Drins (Aldrin, Dieldrin, Eldrin, Endrin sulfate)
Endosulfan	Endosulfan I, II, Endosulfan sulfate
DDTs	Dichlorodiphenyl trichloroethanes
o,p′-DDE	*o,p′*-Dichlorodiphenyl dichloroethane
p,p′-DDE	*p,p′*-Dichlorodiphenyl dichloroethane
o,p′-DDD	*o,p′*-Dichlorodiphenyl dichloroethylne
p,p′-DDD	*p,p′*-Dichlorodiphenyl dichloroethylne
o,p′-DDT	*o,p′*-Dichlorodiphenyl trichloroethane
p,p′-DDT	*p,p′*-Dichlorodiphenyl trichloroethane

Table 2.2. (*Continued*)

Abbreviation	Full name
PAHs	Polycyclic aromatic hydrocarbons
NAP	Naphthalene
2-MNAP	2-Methylnaphthalene
1-MNAP	1-Methylnaphthalene
BiP	Biphenyl
2,6-DiMNAP	2,6-Dimethylnaphthalene
ACY	Acenaphthylene
ACE	Acenaphthene
2,3,5-TriMNAP	2,3,5-Trimethylnaphthalene
FLU	Fluorene
PHE	Phenanthrene
ANT	Anthracene
1-MPHE	1-Methylphenanthrene
FLT	Fluoranthene
PYR	Pyrene
BaA	Benzo[a]anthracene
CHR	Chrysene
BbF	Benzo[b]fluoranthene
BkF	Benzo[k]fluoranthene
BeP	Benzo[e]pyrene
BaP	Benzo[a]pyrene
PER	Perylene
I123cdP	Indeno[1,2,3-cd]pyrene
DahA	Dibenzo[a,h]anthracene
BghiP	Benzo[ghi]perylene
APs	Alkylphenols
NP	Nonylphenol
OP	Octylphenol
BP	Butylphenol
BPA	Bisphenol A
PBDEs	Polybrominated diphenyl ethers
penta-BDE	pentabromodiphenyl ether
octa-BDE	octabromodiphenyl ether
deca-BDE	decabromodiphenyl ether
PFAs	Perfluorinated alkyl compounds
PFOS	Perfluorooctanesulfonate
PFHxS	Perfluorohexanesulfonate
PFBS	Perfluorobutanesulfonate
PFOSA	Perfluorooctanesulfonamide
PFDA	Perfluorodecanoate
PFNA	Perfluorononanoic acid
PFOA	Perfluorooctanoate
PFHpA	Perfluoroheptanoate
PFHxA	Perfluorohexanoate

Table 2.2. (*Continued*)

Abbreviation	Full name
Dioxin-like compounds	
PCDDs	2378-TCDD, 12378-PeCDD, 123478-HxCDD, 123678-HxCDD, 123789-HxCDD, 1234678-HpCDD, OCDD
PCDFs	2378-TCDF, 12378-PeCDF, 23478-PeCDF, 123478-HxCDF, 123678-HxCDF, 123789-HxCDF, 234678-HxCDF, 1234678-HpCDF, 1234789-HpCDF, OCDF
coPCBs	non-*ortho* PCBs (PCB 77, 81, 126, 169), mono-*ortho* PCBs (PCB 105, 114, 118, 123, 156, 157, 167, 189)

2.3. POPs issues and control strategy in South Korea

Although numerous books and research articles describing the geography, climate, weather, population, and other national statistics of South Korea have been published, we provide a brief overview of these geographical and demographic characteristics of South Korea because this information is pertinent to understand the POPs issue in South Korea. Furthermore, we have provided a historical overview of government policies and regulations regarding POPs in South Korea. This overview covers the history of POPs issue, national strategy for POPs control, and national surveillance and monitoring in South Korea.

2.3.1. Description of South Korea

2.3.1.1. Geography

The Korean Peninsula is located in the northeastern part of Asia, which lies between 33°N and 43°N parallels, 124°E and 132°E meridians bordered on the north by China and Russia. South Korea is about 1100 km long and 300 km wide with total land area of ~222,000 km^2, almost the same size as the United Kingdom. The geomorphology of South Korea is characterized by mountains and hills. Such hilly terrain, while occurring mostly in the eastern part of the Korean peninsula accounts for about 70% of its territory (Fig. 2.3). More than 3200 islands are scattered along or near the southern and southwestern coastlines. Due to the many islands that make up parts of Korea, the coastal length of the islands (~8600 km) is almost equal to that of main peninsula (~8700 km). Most of the rivers flow into the Yellow Sea and the South Sea after draining the western and southern slopes of the peninsula. POPs study as well as other regular monitoring programs have covered the far eastern island (i.e., Dok-do) in the East Sea and the southern Jeju Island.

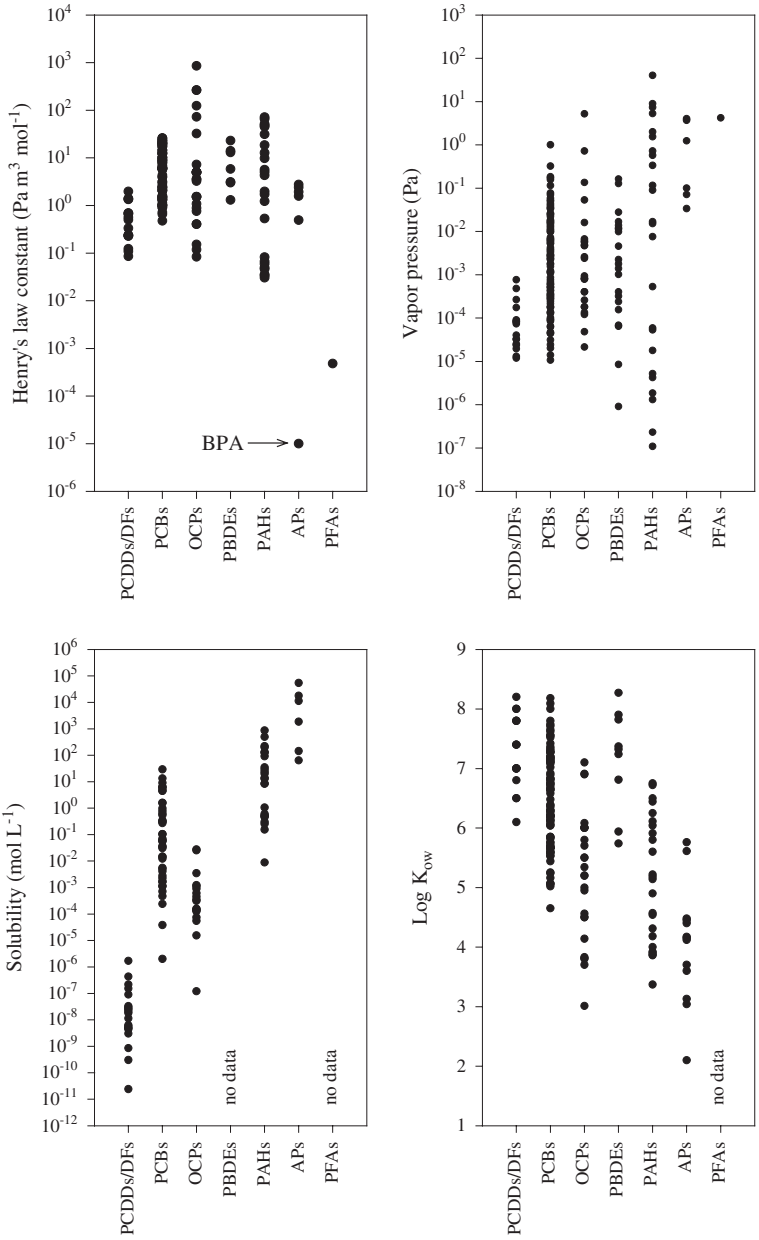

Fig. 2.2. Physicochemical properties (at 25°C) in literatures of target POPs for the present study; Mackay et al., 2000; Cetin and Odabasi, 2005; Lau et al., 2006; Bruner et al., 1990; Park et al., 2002a; Bamford et al., 1999; Paasivirta et al., 1999; Wong et al., 2001; Falconer and Bidleman, 1994; Braekevelt et al., 2003; Hawker and Connell, 1988.

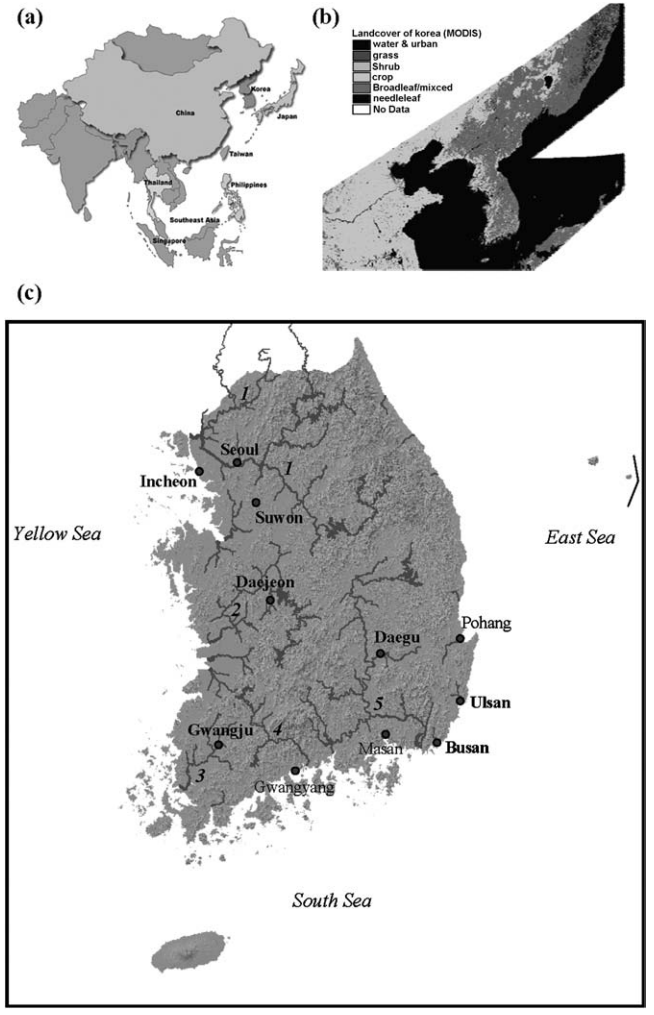

Fig. 2.3. Maps of Asia (a) and Korean peninsula: landscape (b) and digital elevation map (DEM, (c)): Major eight cities with bold character have population over one million. Iron steel mills located at Pohang, Gwangyang and Incheon, and big industrial complexes at Incheon, Ulsan, and Masan. Fiber industry complex has been developed in Daegu since 1950. Major rivers are magnified; Han River (1), Kum River (2), Youngsan River (3), Seomjin River (4), and Nakdong River (5).

2.3.1.2. Climate

The climate of South Korea is temperate monsoon, in general, but geographically and seasonally varied. The western coast open to continental Asia is vulnerable to the influence of the winter monsoon. In contrast, the eastern coast is sheltered from the winter monsoon by the Taebaeksan range, the backbone mountain of the Korean Peninsula. Winter is dry and cold (−6–7°C in January) and is influenced primarily by the Siberian air mass. Summer is hot and humid with temperatures ranging from 23 to 27°C. The climate is the result of the maritime Pacific high-pressure ridge. Temperatures in all seasons are somewhat less than those that occur at corresponding latitudes in other continents, such as North America or Western Europe. Annual precipitation is about 1500 mm in the central region. More than a half of the total rainfall is concentrated in summer, while precipitation of winter is less than 10% of the total.

2.3.1.3. Population

South Korea, the southern half of the Korean Peninsula, is composed of Seoul, seven major cities, and nine provinces with a total population of ca. 48 million (2005 Census). Seoul, the capital city of South Korea, is densly populated (11 million), accounting for about a quarter to the total population in South Korea. Other seven major cities with population of greater than 1 million are Busan, Daegu, Incheon, Gwangju, Daejeon, Ulsan, and Suwon. Since the early 1960s, South Korea has achieved a rapid economic growth, which has been termed the 'Miracle on the Han River'. Recently, the size of the economy of South Korea has grown up to be the 10th largest in the world, and the 3rd largest in Asia, next only to China and Japan.

Substantial urbanization has followed economic growth. South Korea is one of the countries where available area per capita is the least in the world (population density of South Korea is 473 km^{-2}). Futher, the concentration of population density and industries within its urban regions (viz. Seoul and seven major cities) has reached serious proportions, accounting for approximately 50% of the total population in only 5.5% of the land area and approximately 55% of the manufacturing industries in only 11.8% of the land area. In recent years, the government has sought to minimize the urbanization in the capital region, but the trend of urbanization continues to increase. Urbanization rate, the ratio of population in urban area to total population, has increased dramatically, from 28.3% in 1960 to 89.2% in 2004 (KMOE, 2005b). This implies that pollution is likely to be localized in urban and industrial areas.

Furthermore, urbanization means a significant increase in the surfaces without vegetation such as asphalt and building cover. Surface conditions could substantially influence the residence time and fate of POPs in a system. Agricultural activities, of which the dominant activity is rice cultivation, occur primarily in the western and southern areas of the South Korean peninsula. Thus, the water quality of the inland rivers is associated with OCP use history and agricultural products contamination in South Korea.

2.3.1.4. Economy and potential hot spots

Heavy industrialization together with urbanization brought by rapid economic growth has resulted in serious environmental problems in some areas. Now, more than 570 Industrial Complexes (ICs) (30 national ICs, more than 210 regional ICs, and more than 330 agricultural ICs) are in operation nationwide (http://www.kicox.or.kr). Majority of large scale ICs are located near large coastal cities such as Incheon, Busan, Ulsan, Masan, and Pohang. Since many of the ICs are situated near the larger cities, large amounts of industrial wastes together with municipal wastes from inland areas are discharged into the coastal regions. Organic contaminants could also be transported into nearby coastal areas by rivers and/or streams. Thus, greater environmental pollution with POPs and EDCs would be expected to occur along the coastal areas of highly industrialized regions.

2.3.2. POPs issues in South Korea

2.3.2.1. History of POPs issues

There have been accidental releases of pollutants that have cause effects on humans and wildlife, such as Yucheng in Taiwan, Yusho in Japan (Ikeda, 1996), PCB contamination of poultry in Belgium (Bernard et al., 1999), Seveso in Italy (Mocarelli, 2001), and dioxin contamination by redistribution and burial of contaminated soils at Love Canal in the USA (Vianna, 1983). However, despite intensive and rapid industrialization and urbanization, such massive POPs related cases have not occurred in South Korea. Since the 1990s, monitoring South Korea has rarely found POP concentrations as great as parts per million (ppm).

Public concern over POPs (or EDC) was initiated by the dioxin issue, evoked by environmental groups and mass media in the mid-1990s, on waste incinerators. The public concern was further heightened following the reports of adverse effects of tributyl tin (TBT) on mollusks in South

Korean coastal waters (Shim et al., 2000). The issue of human health effects from exposure to POPs, was first investigated relative to releases of dioxins from incinerators to neighboring residents who claimed to have experienced great rates of cancer compared to the control population (CIES, 2002). Whether the 'increased rate of cancer' in Pyeongtak area has truly occurred was not clear in the following and more comprehensive study (Pyoungtak City, 2003). Even though there was no clear relationship between exposure and adverse outcomes, such as cancer and concentrations of PCDDs/DFs in blood, there was a relationship between proximity to incinerators and the concentrations and patterns of PCDDs/DFs in soils and blood. Elevated CYP 1B1 enzyme activity was observed in individuals living near incinerators. In contrast to acute inhalation exposure, due to the increased consumption rate of fatty food such as fishes and meat in South Korea, another recent concern has been chronic exposure to POPs through consumption of contaminated food. According to a recent food consumption survey, fish/shellfish and meat constitute 13% of the total daily intake of food, which is 1280 g d^{-1} $person^{-1}$. Particularly, the fish/shellfish consumption rate contributes as much as 7% (KREI, 2004) of the total diet. Intake of fish/shellfish and meat in 1980 represented only 1% and 4%, respectively (Lee, 1982). Cereal intake has decreased, while animal food consumption has increased; in which, seafood intake is gradually increasing. Consumption of fish/shellfish is thought to be a dominant factor in human exposure to POPs in South Korea.

2.3.2.2. National strategy for POPs control

Regulatory controls of POPs-containing raw materials or wastes in South Korea are based on eight individual acts (Table 2.1). In 2005, one POPs specific ordinance was presented for registration to effectively enact the use, distribution, emission/discharge, environmental residual levels, and treatment and management over the whole life cycle.

The early regulation was focused on classical POPs, namely, the OCPs (Table 2.1). Like many other developed countries, almost all OCPs were banned from use in South Korea, during the early stages of POPs control in the late 1960s. The AgroChemical Management Act (ACMA) banned the registration of OCPs in 1969, which implies the ban of import, manufacture, and sale. After the ban of other OCPs, heptachlor and toxaphene were widely used as alternatives (see later Fig. 2.9). However, in 1980, these two pesticides were also banned from use in South Korea. HCB and mirex were neither imported nor produced in South Korea as pesticide. Endosulfan, known to affect the central nervous system

(Paul and Subramaniam, 1997; Roberts et al., 2004), has not been banned and is still being used. It is know that OCPs were imported and used illegally for a few years even after the ban. Furthermore, some OCPs are known to have been used as a non-food additive agent. For example, chlordane was used as an adhesive for plywood and DDTs as an intermediate in the production of dicofol. It was not until the 1990s that regulation of OCPs in South Korea was mandated by the Toxic Chemical Control Act (TCCA).

PCBs were imported from abroad and their use was legally regulated for the first time in 1979. The Electricity Business Act (EBA) prohibited the use of electric heaters with PCB-containing dielectic fluids. However, existing PCB-containing articles were allowed to be used for other purposes until 1996 when production, import, and use of articles containing concentrations of PCBs greater than 50 ppm were banned by the TCCA. Waste containing concentrations of PCBs greater than 2 ppm was classified as specific hazardous wastes was treated by special methods listed in the law, and should be reported to and confirmed by the KMOE.

According to the Stockholm Convention, PCB-containing equipments such as capacitors, condensers, and transformers should be forbidden from use by 2025 and are recommended to be treated by environmentally sound methods by 2028 (Stockholm Convention on Persistent Organic Pollutants, 2001). The South Korea Ministry of Environment (KMOE), South Korea Electric Power Corporation (KEPCO) and six electric power companies, and three non-governmental organizations (NGO) reached a cooperative and administrative agreement in 2004 for the removal of PCBs in South Korea by 2015 (KMOE website). An investigation of the domestic status of PCB-containing articles is currently being conducted.

In spite of the regulations, government actions on POPs began practically only after a public concern on the dioxin release in waste incinerators in 1997. Since then, the emission of PCDDs/DFs was included in regulations. Recently, in response to the Stockholm Convention, periodic monitoring has been conducted in South Korea.

The total amount of waste generated in South Korea in 2003 was 300,000 ton day^{-1}. Construction waste, a dominant waste-type in South Korea, has increased from 16% in 1966 to 49% of the total amount of the waste currently. Almost 90% of this waste is recycled and just 1.5% is incinerated. Thus, the proportion of solid waste that is incinerated (6%) is rather small, relative to other countries in Asia (Fig. 2.4). However, the proportion of municipal and industrial waste that contributes to 17% and 34% of the waste that is incinerated has increased continuously. In 1993, the ratio of municipal solid waste (MSW) that was incinerated in South

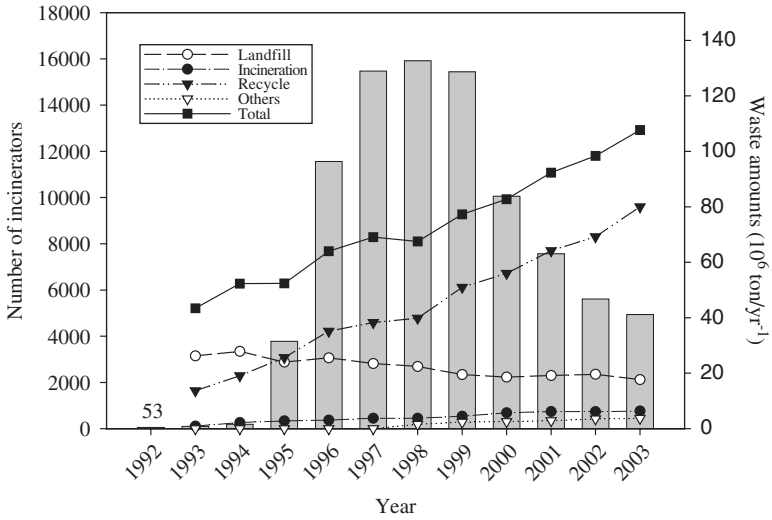

Fig. 2.4. Annual trend of number of all the incinerators in operation (bar plots from Kim et al., 2002d) and waste amounts treated by each method (symbol-line plots from KMOE & NIER, 1997, 1998, 1999, 2000, 2001, 2002, 2003) in Korea.

Korea was about 2.4%, which increased to 14.5% in 2003 (KMOE, 2005b). The proportion of industrial waste that is incinerated has increased from 1.9% to 7.8% during the same period. The proportion of waste that is incinerated is expected to increase in the future as land becomes more and more developed and lesser land is available for landfill.

Waste incineration is a known source of dioxins and other dioxin-like compounds. As a part of change in waste management policy since the early 1990s, many incinerators have been constructed. The number of incinerators in South Korea reached a maximum of about 16,000 in 1998 (from only 100 in 1993). However, it has decreased noticeably to ~5000 in 2003 since an introduction of stringent dioxin emission regulations for the incinerators in 1997 (Fig. 2.4). The majority of these incinerators (more than 90%) are small-scale facilities with a capacity of less than 0.2 ton hr^{-1}. Although the number of large-scale incinerators with capacities of greater than 4 ton hr^{-1} is only 0.2% of the total incinerators, the quantity of wastes treated by these large-scale incinerators is over 70% of the total incernation (Table 2.3). Thus in 1997, the government has started to require emission control of newly installed municipal incinerators with incineration capacities equal to or greater than 50 ton day^{-1} (Table 2.4). Since that time, large-scale incinerators have been required to restrict emission of dioxins to less than 0.1 ng I-TEQ Nm^{-3}. Subsequently,

Table 2.3. Temporal trend of the number of incinerators (unit; *n*) and waste amounts incinerated (unit; 10^3 ton yr^{-1})

	Incinerator size					References
	Year	Small	Medium	Large	Total	
Incinerator facility (*n*)[a]	2000	11,841	394	103	12,338	KMOE, 2001
	2001	7697	362	114	8173	KMOE, 2002a
	2002	5733	309	126	6168	KMOE website
Incineration amount	2000	328	646	2335	3309	KMOE, 2001
	2001	257	848	3481	4586	KMOE, 2002a
	2002	322	751	3899	4972	KMOE website

[a]Data were collected in June every year. Final values are less than these.

medium- and small-scaled incinerators have been included in these regulations (Table 2.4). Now, all the incinerators are required to reduce emissions of dioxins to less than 0.1–10 ng I-TEQ m^{-3}. Under the Waste Control Act (WCA), facilities that incinerate less than 0.025 ton hr^{-1} are no longer allowed (Kim, 2001). Another provision of the WCA is that all incinerators are obliged to conduct regular monitoring for dioxin-like compounds: semiannually, annually, and biannually for large, medium, and small scale, respectively. Since these regulations went into effect, the best available technologies (BAT) such as spray absorber/bag filter (SDA/BF) with lime and activated carbon mixture sprayed into the SDA have been applied to incinerators (Kim et al., 2001a) and the number of smaller, less economically feasible incinerators has greatly decreased.

In addition to regulations on waste incineration, discharge/emission guidelines for dioxins released from industrial sources, such as metallurgical plants, are expected to be established in 2006 (KMOE website). In 2005, an administrative agreement on reduction of dioxin emission through industrial processes was adopted by among KMOE, NGOs, and four industries, including the iron & non-iron metal industry, cement industry, and chemical industry. The groups agreed to reduce dioxin emission to 70% of that estimated for 2001 by 2008 and 50% by 2010. To achieve this goal best environmental practice (BEP) in addition to BAT will be adopted.

Establishment of emission and/or discharge guidelines for each source and environmental quality criteria for each compartment are legal regulations that have been established to reduce environmental exposure to POPs. South Korean guidelines and quality criteria are applied to dioxins from municipal waste incinerators, and waste and environmental quality guideline for PCBs (Table 2.1). The extensive guidelines will be applied following a ministerial ordinance entitled 'Specific Act on Management of

Table 2.4. Guideline of dioxins release from waste incinerators (ng I-TEQ m^{-3})

Waste type	Incinerator size		Guideline and application date			
			New incinerator		Existing incinerator	
Municipal solid waste	Category	Capacity/application	From Jul./97	To Jun./99	To Jun./03	From Jul./03
	Large	> 50 ton/day	0.1	0.5	0.5	0.1
Other all wastes	Category	Capacity/application	From Jan./01	To Dec./02	To Dec./05	From Jan./06
	Large	≥4 ton/hr	0.1	20	20	1
		2–4 ton/hr	1	40	40	5
	Medium/small	0.025–2 ton/hr	5	40	40	10
Hospital/medical waste	Category	Capacity/application	From Aug./04	To Dec./05	To Jun./06	From Jul./07
	Large	≥4 ton/hr	0.1	0.1[a] (20)[b]	0.1 (1)	0.1 (1)
		2–4 ton/hr	1	1 (40)	1 (5)	1 (5)
	Medium	1–2 ton/hr	1	5 (40)	5 (10)	1 (5)
		0.2–1 ton/hr	5	5 (40)	5 (10)	5 (5)
	Small	0.025–0.2 ton/hr	5	–	10 (10)	10 (10)

[a]For hospital/medical waste incinerators installed between Jan./01 and Jul./04.
[b]For hospital/medical waste incinerators installed before Jan./01.

Persistent Organic Pollutants including Dioxins (Specific Act for POPs control)' that was recently promulgated (KMOE website). This ordinance involves regulation and activities to reduce exposure to POPs including: (1) prohibition and restriction of production, distribution, and use of POPs; (2) establishment of environmental quality guidelines (EQGs) in each compartment, human tolerable daily intake (TDI), standard operation procedure (SOP), and regular monitoring system; (3) emission control including establishment of emission guidelines and emission inventories; (4) management of POPs containing waste; and 5) protection of POPs contamination of soils and restoration of POPs-contaminated soils.

2.3.2.3. National surveillance and monitoring

Beginning in July 1999, several governmental agencies have established 'A 10-year National Research Program for Endocrine Disruptors (1999–2008)' (Choi, 2002). As part of this program, it has been agreed that seven governmental ministries, including the Ministry of Environment (KMOE), Rural Development Administration (RDA), South Korea Food and Drug Administration (KFDA), Ministry of Maritime Affairs and Fisheries (MMAF) and the Ministry of Labor (MOL) should perform various kinds of research and surveillance of EDCs (or POPs). Risk assessments and regular monitoring for POPs exposure from foods and environmental media, and in humans should be conducted and emission inventories developed (Table 2.5).

Nationwide surveillance of POPs is now being performed by a number of governmental agencies. They are establishing emission inventories and regular monitoring of residual levels. National emission inventories are necessary for tracing the life cycle of pollutants, estimating their behavior and fate in environments, and managing and controlling POPs substances by use of BAT in an integrated strategy. Studies of the emissions and/or discharge have focused on unintentionally produced by-products including PCDDs/DFs, coPCBs, PAHs, and HCB because product POPs such as PCBs and OCPs, the uses of which were regulated previously. The earliest emission study was on atmospheric emission of PAHs through pyrolytic processes (MCIE, 1999). This study estimated emission rates by methods developed by the US EPA (US EPA, 1998a). Similar top-down approaches using emission rates and activities of individual sources were applied to estimate preliminary emission inventories for PCDDs/DFs (KMOE, 2001, 2002a) and byproduct PCBs, HCB, and PAHs (KMOE, 2003b) from point sources, and PCDDs/DFs from non-point sources (KMOE, 2004c). The estimates provided by these preliminary studies

Table 2.5. Governmental investigation projects for POPs in Korea (modified from KMOE website (http://www.me.go.kr/dev/board/)

Target medium	Investigation contents	Target POPs[a]	Year[b]	Governmental institution	References
Emission inventory					
Point source	Waste incinerators ($n = 1800$), metallurgical/chemical/energy/mineral production industries ($n = 288$)	PCDDs/DFs, coPCBs	2001–2005	KMOE[c]	KMOE website; NIER, 2000b; KMOE, 2001, 2002a, b, 2003a, 2004b, 2005a
Non-point source	Transport, uncontrolled/residential fuel combustion, fires etc.	PCDDs/DFs, coPCBs, HCB	2004–	KMOE	KMOE, 2004c, 2005a
Products	PCBs product and waste, and OCPs	PCBs, OCPs	2003–2005	KMOE	KMOE, 2004a
By-products	Unintentional PCBs and HCB, and PAHs in point and non-point sources	PCBs, HCB, PAHs	2004–2006	KMOE	MCIE, 1999; KMOE, 2003b; KMOE, 2005a
Monitoring					
Environmental media	Air, freshwater, river/lake sediment, soil, and fresh fishes	67 EDCs	1999–	NIER[d]/KMOE	NIER, 2000a; 2001; 2002; 2003; 2004; 2005a; KMOE website
	Coastal water, sediment, and sentinel organism (i.e., mussel-watch program)	PCDDs/DFs, coPCBs, PCBs, OCPs, PAHs etc.	1999–	MOMAF[e]	Kim et al., 2002b; Oh et al., 2003; Hong et al., 2006
	Agricultural farm land	PCDDs/DFs	–	RDA[f]	–

	Multi-media nearby incinerator (stack gas/ ambient air, discharged/ ambient water, soil, sediment etc.)	PCDDs/DFs	2003–2008	NIER	KMOE website, NIER, 2005b, Kim et al., 2005b
Food	Food items and tolerable daily intake (TDI)	PCDDs/DFs, PCBs, PAHs, PBDEs etc.	2000–	KFDA[g]	KFDA website
	Stock farm products	PCDDs/DFs	–	NVRQS[h]	–
	Seafood	PCDDs/DFs, coPCBs, PCBs, OCPs, PAHs etc.	2001–	MOMAF	Oh et al., 2005a, Yim et al., 2005, Moon and Ok, 2006
	Seafood for export	PCDDs/DFs, PCBs etc.	2005–	NFRQIS[i]	MOMAF website
	Agricultural farm products	PCDDs/DFs	–	RDA	–
Human	Human blood and milk	PCDDs/DFs	2000–	KFDA/NITR[j]	KFDA website
	Resident blood nearby waste incinerators	PCDDs/DFs	Intermittently		–

[a]PCDDs/DFs (Dioxins and Furans), coPCBs (coplanar PCBs), PCBs (polychlorinated biphenyls), HCB (hexachlorobenzene), OCPs (organochlorine pesticides; DDTs, Chlordanes, Dieldrin, Aldrin, Endrin, Heptachlor, Toxaphen, Hexachlorocyclohexane, endosulfan etc.), EDC (Endocrine Disrupting Compounds).
[b]The duration indicates the duration of measurement performances for emission and monitoring.
[c]KMOE (Korea Ministry of Environment).
[d]NIER (National Institute of Environmental Research).
[e]MOMAF (Ministry of Maritime Affairs and Fisheries).
[f]RDA (Rural Development Administration).
[g]KFDA (Korea Food and Drug Administration).
[h]NVRQS (National Veterinary Research Quarantine Service).
[i]NFRQIS (National Fisheries Products Quality Inspection Service).
[j]NITR (National Institute of Toxicological Research).

may have large uncertainties due to use of emission factors developed abroad. Since 2001, extensive measurements have been made to establish actual inventories of PCDDs/DFs, coPCBs, HCB, by-product PCBs, and PAHs (i.e., benzo[*a*]pyrene) for both point and non-point sources (Table 2.5). In 2005, the KMOE announced the first emission inventories with a reference year of 2001 for dioxin-like compounds (KMOE website). Domestic status on usage and waste management of product POPs such as PCBs and OCPs have also been investigated since 2003 (KMOE, 2004a). In particular, during 2003–2004, the NIER investigated the PCB contents of 1237 capacitors. They are now contemplating surveying an additional 3000 capacitors (KMOE website). KMOE plans to investigate PCB-containing capacitors of 130,000 electrical plants between 2005 and 2007 (KMOE website).

Several agencies are responsible for the nationwide monitoring of POPs. Long-term monitoring for environmental media includes inland, marine, and agricultural regions (Table 2.5). In this large-scale monitoring program, the NIER began with the investigation of 37 EDC substances in the inland environment including 43 water sampling stations, 11 river/lake sediment stations, 24 air sites, and 35 soil sites in 1999, The sampling campaign is continuing and the number of target substances and sites has been expanded every year. For example, since 2000, fishes and amphibians were included at 31 sites in five main rivers (Jeong et al., 2001a, b; Kim et al., 2004a). The regular monitoring involves PCDDs/DFs, PCBs, OCPs, and alkylphenols. Marine environments are also monitored annually by MOMAF for sea water, sediment, and bivalves along the coastal area, as mussel watch program (Kim et al., 2002b; Oh et al., 2003; Hong et al., 2006). Fishes from coastal environment and/or markets were also investigated for dioxin-like compounds (Oh et al., 2005a; Moon and Ok, 2006) and organochlorines including PCBs and OCPs (Yim et al., 2005). In addition to the long-term environmental monitoring described above, more targeted, site-specific monitoring is performed for dioxin-like compounds in multi-media (i.e., stack gas, discharged wastewater, and ambient air/water/soil) around 34 waste incinerators from 2003 to 2008. In addition to environmental monitoring, since 2000 foodstuff and human tissues have also been monitored by each responsible governmental institution for stock farm products, seafood, and agricultural farm products. For instance, the KFDA monitors PCDDs/DFs, PCBs, and PAHs in foodstuffs and PCDDs/DFs, PCBs, OCPs, bisphenol A, and PBDEs in human tissues (KFDA website). According to the 'Specific Act for POPs control' the monitoring data from these studies are expected to be integrated into assessments of environmental and human risk, and establish environmental quality criteria and tolerable daily intake on the basis of

contamination status and trends in South Korea. While extensive monitoring is being performed by governmental agencies, the reported detection limits are insufficient. Improvements in analytical methods applied in some of the monitoring programs will be necessary to provide information for meaningful risk and fate assessments.

2.4. POPs emission in South Korea

2.4.1. By-product POPs

POPs are generated as waste itself, or during waste treatment, and from synthesis and production of products. Measurements of by-product POP emission began with PCDDs/DFs for waste incinerators in the late 1990s and from 2001 to 2005 have been performed annually at many industrial sources and non-point sources. Other by-product POPs including HCB, BaP, and PCBs were added to these investigations in 2005. The South Korean government is planning to perform long-term measurement for 'emission inventories of by-product POPs including PCDDs/DFs, coPCBs, and HCB' from 2006 and to report the result biennially.

2.4.1.1. Dioxin-like compounds (PCDDs/DFs and coPCBs)

The emission inventory of dioxin-like compounds in South Korea was determined during two preliminary studies (KMOE, 2001, 2002a). For emission factors, the preliminary study (KMOE, 2001) adopted measured values for waste incinerators and the values of UNEP chemicals Toolkit (UNEP Chemicals, 2001) for the other sources. Estimated PCDDs/DFs emission in 1999 ranged from 1163 to 1595 g I-TEQ yr^{-1} due to uncertainties in emission factors and activities (Table 2.6). Besides the preliminary estimate, since the late 1990s extensive measurements of PCDDs/DFs have been performed at waste incinerators and the emission data by 2004 had been compiled for 1800 incinerators. Moreover, nationwide industrial sources have been investigated every year since 2001; 34 ferrous/non-ferrous metal production factories in 2001, 114 non-ferrous metal and mineral production factories in 2002, 73 chemical/energy/landfill factories and crematories in 2003, and 63 municipal wastewater treatment plants and 9 types of vehicles in 2004. By 2005, measurements of total dioxin emissions had been made on 288 industrial sources. Based on these measurements, KMOE made the first official estimate of PCDDs/DFs emission in South Korea. It has been estimated that the total PCDDs/DFs emission was 1021 g I-TEQ yr^{-1} in 2001 (KMOE website) (Table 2.6). This emission was approximately 62% of that

Table 2.6. Summary of annual inventories of PCDDs/Fs emission in Korea (g I-TEQ yr^{-1})

Source category[a]	1998[b]	1999[c]	2001[d]	Conc.[e]
Waste incineration				
Municipal waste (Large, L)	1.47	0.355	163.5	0.02
Municipal waste (Medium, M & Small, S)	105	327		7.15
Industrial furnace	733	429	728.2	25.3
Construction waste		152		
Hazardous waste	126	129–310		6.66
Medical/hospital waste		2.99–7.18		–
Sludge	–	0.398–3.98	–	–
Total (average)	965.5	1040–1230	891.6	–
Ferrous and non-ferrous metal production				
Iron ore sintering	–	9.37–125	96.4	0.043
Coke production	–	2.86		
Iron and steel production plants	–	53.8–173		
Foundries	–	0.049–1.62		
Copper production	–	0.034	15.0	4.234
Aluminum production (all secondary)	–	–		0.25
Lead production	–	0.025–4.00		0.15
Zinc production	–	–		0.036
Brass production	–	–		–
Magnesium production	–	–		–
Thermal non-ferrous metal production	–	–		–
Shredders	–	–		–
Thermal wire reclamation	–	–		–
Total	–	66.1–307	111.4	–
Power generation and heating				
Fossil fuel power plants	–	17.2	9.8	0.019
Biomass power plants	–	–		–
Landfill and biogas combustion	–	–		–
Household heating and cooking (biomass)	–	–	–	–
Domestic heating (fossil fuels)	–	8.39	–	0.074
Total	–	25.6	9.8	–
Production of mineral products				
Cement kilns	–	7.29	3.1	0.095
Lime	–	0.196		0.050
Glass	–	0.023		0.018
Ceramics	–	–		0.223
Asphalt mixing	–	0.190		–
Total	–	7.70	3.1	–
Transport				
4-Stroke engines	–	–	(0.057)[f]	0.138
2-Stroke engines	–	–	(0.187)[f]	
Diesel engines	–	0.591	(0.756)[f]	0.559

Table 2.6. (*Continued*)

Source category[a]	1998[b]	1999[c]	2001[d]	Conc.[e]
Heavy oil fired engines	–	15.2	(13.5)[f]	–
Total	–	15.8	(14.5)[f]	–
Uncontrolled combustion processes				
Fires/burnings (biomass)	–	0.114	–	–
Landfill/industrial fires, waste burning	–	7.32	–	–
Total	–	7.43	–	–
Production of chemicals or consumer goods				
Pulp and paper mills (boilers)	–	0.046	0.6	0.002
Chemical industry	–	–		1.538
Total	–	0.046	0.6	–
Miscellaneous				
Drying of biomass	–	–	–	–
Crematory	–	0.002–0.403	4.1	0.127
Smoke houses	–	0.070–0.704	–	–
Dry cleaning residues	–	–	–	–
Tobacco smoking (pg item^{-1})	–	0.009	–	–
Total	–	0.081–1.12	4.1	–
Total	–	1163–1595	1021 (1036)[f]	–

[a]UNEP Chemicals (2001).
[b]Modified from NIER (2000b) by considering gas amount emitted per unit ton for different incinerator type and size.
[c]KMOE (2001); Emission estimated from top-down approach. Ranges are estimated when varying incinerators' activities (i.e., emission gas amount per ton) or emission factor.
[d]Website of MOE (www.me.go.kr/dev/board/); First emission calculated from measurements for point sources (i.e., up to 2001 for waste incinerator and up to 2005 for others).
[e]Average concentration of stack gas measured at each source since 2000 (ng I-TEQ m^{-3}); Municipal waste (L) from KMOE (2004a), other waste incinerators from KMOE website, ferrous and nonferrous metal industry from Yu et al. (2006), transport from KMOE (2005a), and others from KMOE (2004b).
[f]KMOE (2004c); Results of PCDDs/DFs for non-point sources. Total value in parenthesis includes the non-point emission.

reported for 2001 in Japan, a country that has one of the largest releases worldwide (JMOE website).

A major contributor of PCDDs/DFs release to environments is waste incinerators (77–89% in preliminary study and 86% in follow-up studies) followed by 6–19% from ferrous/non-ferrous metal industry and $<5\%$ from others including power generation and heating, mineral production, chemical industry, and non-point sources. Non-point sources like transportation and uncontrolled burning processes showed a minimal contribution of $<2\%$.

All studies found that more than 70% of PCDDs/DFs released from waste incinerations were from industrial waste incinerations including

industrial waste furnace, construction waste incineration, hazardous and medical/hospital waste incineration. In particular, small- and/or medium-sized municipal waste incinerators (~30%) were one of the major contributors with industrial waste furnaces contributing approximately 35–40% of total dioxin emission. In 2002, the number of small incinerators comprised 93% of the total number of incinerators, although the amount of waste treated by smaller incinerators was only 6.5% (Table 2.3). Relatively great amounts of dioxins were emitted from small- and medium-sized incinerators and industrial incinerators, despite the lesser amount of waste treated. This could be due to poor operating conditions, lack of emission control equipments, and ill management practice. The UNEP estimated the emission of gas per unit ton of waste incinerated to be 5000 Nm^3 ton^{-1} when BAT, including activated carbon addition and SCR/DeNOx were applied, while the value was estimated to be 10000 Nm^3 ton^{-1} for incinerators with poor operating conditions (UNEP Chemicals, 2001). In 1999, the amounts of stack gas emissions from domestic incinerators was 3758–8401 (average = 5230 Nm^3 ton^{-1}) for large waste incinerators, 4400–15000 (average = 12000 Nm^3 ton^{-1}) for small/medium-incinerators, and 4400–15000 (average = 12000 Nm^3 ton^{-1}) for industrial furnaces (KMOE, 2001). Thus, improvements in operating conditions for industrial furnaces and small/medium-sized incinerators will be necessary to achieve further reductions in emissions of PCDDs/DFs. In 1998, the Environmental Management Corporation measured dioxins and furans released from selected incinerators, including various types of incinerators and reported an estimate of national emission as 446.5 g I-TEQ yr^{-1}. This value is certainly an underestimated one because of the use of such assumptions as best operating conditions for all incinerators (i.e., 5000 Nm^3 ton^{-1}) (NIER, 2000b). This actual emission rate, based on real operating conditions as in 1999, would be expected to be 965.5 g I-TEQ yr^{-1} (KMOE, 2001). Predicted contributions of individual incinerator types to dioxin emission were similar with that determined by other studies. Overall, the studies indicated that PCDDs/DFs emission from waste incinerators in South Korea was approximately 1000 g I-TEQ yr^{-1} (891.6–1230 g I-TEQ yr^{-1}) between the late 1990s and early 2000s (Table 2.6).

Because of the relatively great contribution of waste incineration to dioxin emission, since 1997 the South Korean government has regulated releases of dioxins from waste incinerators (Table 2.4). Almost all large municipal waste incinerators with BAT reduced the dioxin release to rates that are less than the emission guideline of 0.1 ng I-TEQ Nm^{-3} (Table 2.6). Guidelines have now been established for all types of incinerators. Furthermore, incineration in facilities with capacities below 0.025 ton hr^{-1} is prohibited. As a result of this regulation, the number

of incinerators, particularly smaller incinerators, has decreased. In 2003, the smaller incinerations comprise only about 31% as many as were operating in 1998 (see Table 2.3 and Fig. 2.4).

Some PCBs are dioxin-like compounds due to their ability to bind to the arylhydrocarbon receptor (Ah-R). In particular, these are the non- and mono-*ortho*-substituted congeners. Because of their ability to assume a somewhat planar configuration that can result in a molecular size and conformation that is similar to 2,3,7,8-TCDD these congeners are sometimes referred to as co-planar PCBs (coPCBs). The total release of dioxin-like compounds would be underestimated if coplanar PCB congeners were not included. KMOE also reported the emission of dioxin-like compounds including PCDDs, PCDFs, and coPCBs, to be 1247 g WHO-TEQ yr^{-1} for 2001. Here we have corrected the emissions of 1998 and 1999 using the correction factor (~10%) of I-TEQs to WHO-TEQs for PCDDs/DFs and the ratios of coPCBs to PCDDs/DFs for individual sources presented in the KMOE emission report for 2001 (Table 2.6). When this is done, the contribution of coPCBs to the WHO-TEQs was estimated to be approximately 10% for waste incinerators and 6–26% for industrial sources. Corrected emissions were determined to be 1322 and 1927 g WHO-TEQ yr^{-1}. The conversion factor of I-TEQs (NATO/CCMS, 1988) to WHO-TEQs (van den Berg et al., 1998) is frequently less than 10% for municipal waste incinerators in South Korea; 0.7–18% (average 7%) for gas, ash, and smelted slag (Kim et al., 2005a) and on average 4% for stack gas (Kim et al., 2001b) and fly ash (Shin and Chang, 1999). Thus, the corrected values seem to be a slightly greater than actual expected emission rates.

The South Korean government also reported annual emissions of dioxin-like compounds from waste incineration from 2001 to 2005. During this period the rates of emission of WHO-TEQ in 2004 decreased by 23.5% relative to that in 2001 (Fig. 2.5). Assuming a similar emission as in 2001 for non-waste incineration sources, total atmospheric release of dioxin-like compounds were predicted to decrease 67% between 2001 and 2004, to 410.6 g WHO-TEQ yr^{-1}. The release of dioxins in 2004 is comparable to 401.9 g WHO-TEQ yr^{-1} in Japan in 2003 (Fig. 2.5). The reduction in emissions of dioxin-like compounds from waste incineration seems to reflect the successful regulation for incinerators in South Korea. For instance, the emissions of dioxin-like compounds, expressed as I-TEQ from only 33 incinerators (9.8% of 336 investigated including 243 small- and medium-sized incinerators) exceeded the emission guidelines; so do 12 of 82 municipal incinerators and 21 of 254 industrial incinerators (KMOE website). Furthermore, the average release of TEQ from large municipal waste incinerators decreased from 5.97 ng I-TEQ Nm^{-3} in 1997 to 0.06 ng TEQ Nm^{-3} (Lee et al., 2002).

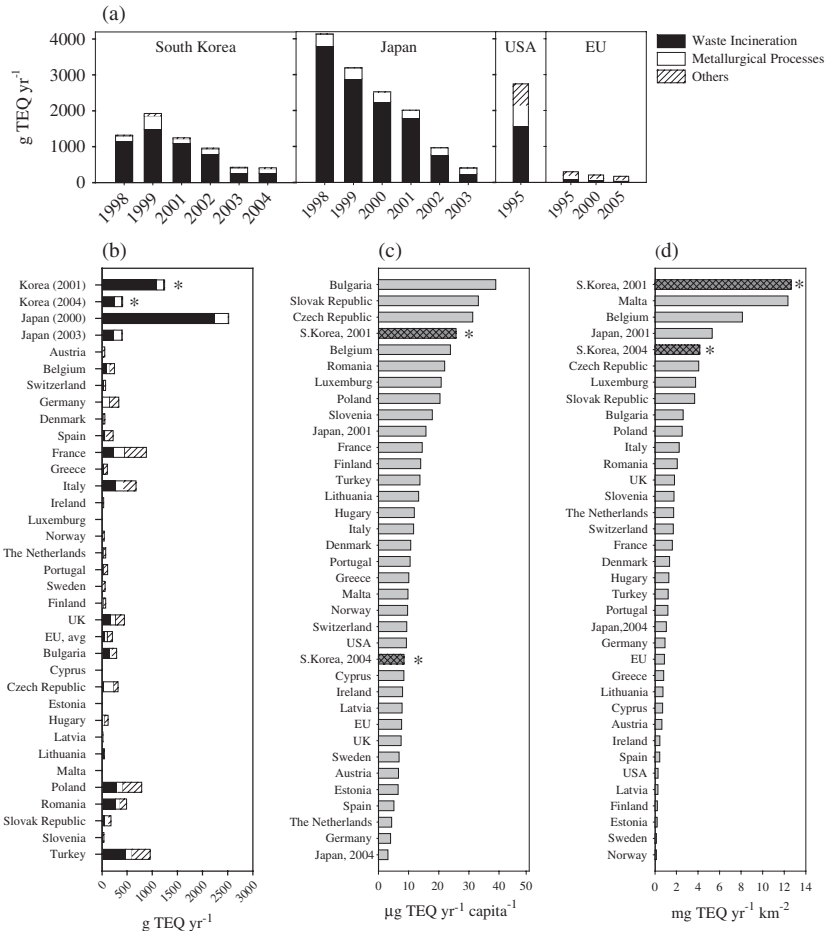

Fig. 2.5. Nationwide annual emission inventories to air of dioxin-like compounds in worldwide countries; (a) Average of emissions of EU countries were used for EU: Korea 1998 and 1999 (recalculated from NIER, 2000b and KMOE, 2001, respectively), Korea 2001–2004 (from KMOE website), Japan (1998–2003) (from (JMOE website), USA 1995 (from US EPA, 1998b), EU 1995 (UNEP, 1999), EU 2000 and 2005 (estimated for 2000 and projected for 2005 from Quaβ et al., 2000), (b) emission inventories of 2000 for EU (Quaβ et al., 2000) and central European countries (Pulles et al., 2005), (c) and (d) calculated from values of (b). The emission of South Korea (S. Korea) and Japan is on basis of WHO-TEQ including PCDDs/DFs and coPCBs, while those of other countries based on I-TEQ of PCDDs/DFs alone. Population and area of each country were quoted from Wikipedia website.

Despite this decrease, in 2004 the emission rates were greater than that of some European countries (Fig. 2.5a, b). Particularly, when emissions of TEQ were normalized by expressing them on a per capita or unit area which was 25.7 μg WHO-TEQ capita^{-1} yr^{-1} and 12.7 mg WHO-TEQ km^{-2} yr^{-1} in 2001 (Fig. 2.5c, d). These values are 1.6 and 2.4 times greater, respectively, than those of Japan in 2001. The emission per unit area remains the greatest in the world, even though the value in 2004 decreased to 4.2 μg WHO-TEQ km^{-2} yr^{-1} (Fig. 2.5d). The relatively great emission rate suggests that the exposure level at least during recent years of South Korean environments and humans has been greater than that for other countries.

In spite of the new introduction of emission regulations, waste incineration is still a major contributor to dioxin emission (greater than 60% of the total emission) in South Korea. In Europe, however, iron ore sintering is the predominant emission source followed by the previous 'No. 1', municipal waste incineration (Quaβ et al., 2004). The South Korean government also plans to regulate the release from industrial sources. For instance, government, NGO and industries have agreed to reduce the release of TEQ from industrial sources to 70% of the 2001 by 2008 and further to 50% by 2010 by establishing release guidelines for the industrial sources. Banning of small-scale incinerators and enforcing the release guideline of 0.1 ng Nm^{-3} for all the waste incinerators is expected to result in a considerable reduction of the emission. According to the emission reduction scenarios (KMOE, 2001), 92% and 68–83% of the emissions are expected to be reduced from waste incineration and from all sources, respectively. With further application of this release guideline to iron ore sinters and steel electric furnaces, total dioxin emission in South Korea are predicted to decrease between 91% and 94%.

Non-point sources are minor contributors (~4% of total emission for 2001, 33.96 g I-TEQ yr^{-1}) (KMOE, 2004c). Among non-point sources, it has been estimated that large ships, using heavy oils, contribute 45% of the total emissions from non-point sources. Major harbors could thus be affected by this source (Cooper, 2005). The releases from landfill leachate, municipal wastewater, and paper/pulp wastewater contribute 2.254 g WHO-TEQ yr^{-1} (KMOE, 2004c). This is comparable to Japanese emission for 2003 (http://www.env.go.jp/en/press/2004/).

2.4.1.2. PCBs

By-product PCB emission from all potential sources was estimated by the 'top–down' approach (KMOE, 2003b). This estimate was made for 2000 by using emission factors from UK National Atmospheric Emissions

Inventory (UK NAEI website) and the Co-operative Program for Monitoring and Evaluation of the Long Range Transmission of Air pollutants in Europe (EMEP) workshop (MSC-E, 2002) and an activity database of individual sources in South Korea. Estimated emission of by-product PCBs was 1085 kg yr^{-1} and is likely to be greater than that for many European countries in 2000 (Fig. 2.6). Considering the greater PCDDs/DFs emission in South Korea than that in European countries, and the similar emission sources of both PCDDs/DFs and PCBs, relatively greater PCBs emission might be reasonable, although the emission estimates are associated with large uncertainties. Among the sources, the ferrous metal industry was a major contributor, 93% (i.e., ~1000 kg yr^{-1}) to the total emissions, followed by fuel combustion (5.9%, 64.2 kg yr^{-1}), waste incineration (1.3%, 14.23 kg yr^{-1}), and others including non-ferrous/mineral production (<0.01 kg yr^{-1}). A total of 872 kg PCB yr^{-1} is estimated to be released from electric furnaces producing iron from scrap. This is different from the source profile of Europe where the fuel combustion is a major source. All ferrous steel mills are located in coastal areas in South Korea (see Fig. 2.3), suggesting the possibility of contamination of localized marine environments with by-product PCBs.

2.4.1.3. HCB

HCB was neither imported nor manufactured in South Korea. Domestic emission, therefore, should originate from process such as combustion of organic matters combined with chlorine and from impurities in the production of chemicals and pesticides. HCB emission for 2000 in South Korea was estimated in a preliminary study of emission inventory using 'top-down approach' (KMOE, 2003b). Emissions from some sources in 2005 were determined by measuring concentrations in archived samples originally collected for analysis of PCDDs/DFs in 2001–2003 (KMOE, 2005a).

Domestic HCB emissions have been estimated to be 245 kg yr^{-1} for 2000, which falls approximately in the mid-range of the values reported for EU countries (Fig. 2.6). However, the direct intercomparisons among the countries may be misleading due to the varying uncertainties in emission inventories of these countries. According to the preliminary study (KMOE, 2003b), the source contribution of HCB emission is as follows: waste incineration (62%, 152 kg yr^{-1}) > iron ore sinter (19%, 50 kg yr^{-1}) > fuel combustion (14%, 35 kg yr^{-1}) > non-ferrous metal/mineral production (3.6%, 8.7 kg yr^{-1}) > transport (1%, 2.4 kg yr^{-1}). Emission of the electric furnace for scrap iron was not included in the preliminary study. Thus, the contribution of the ferrous metal manufacturing would

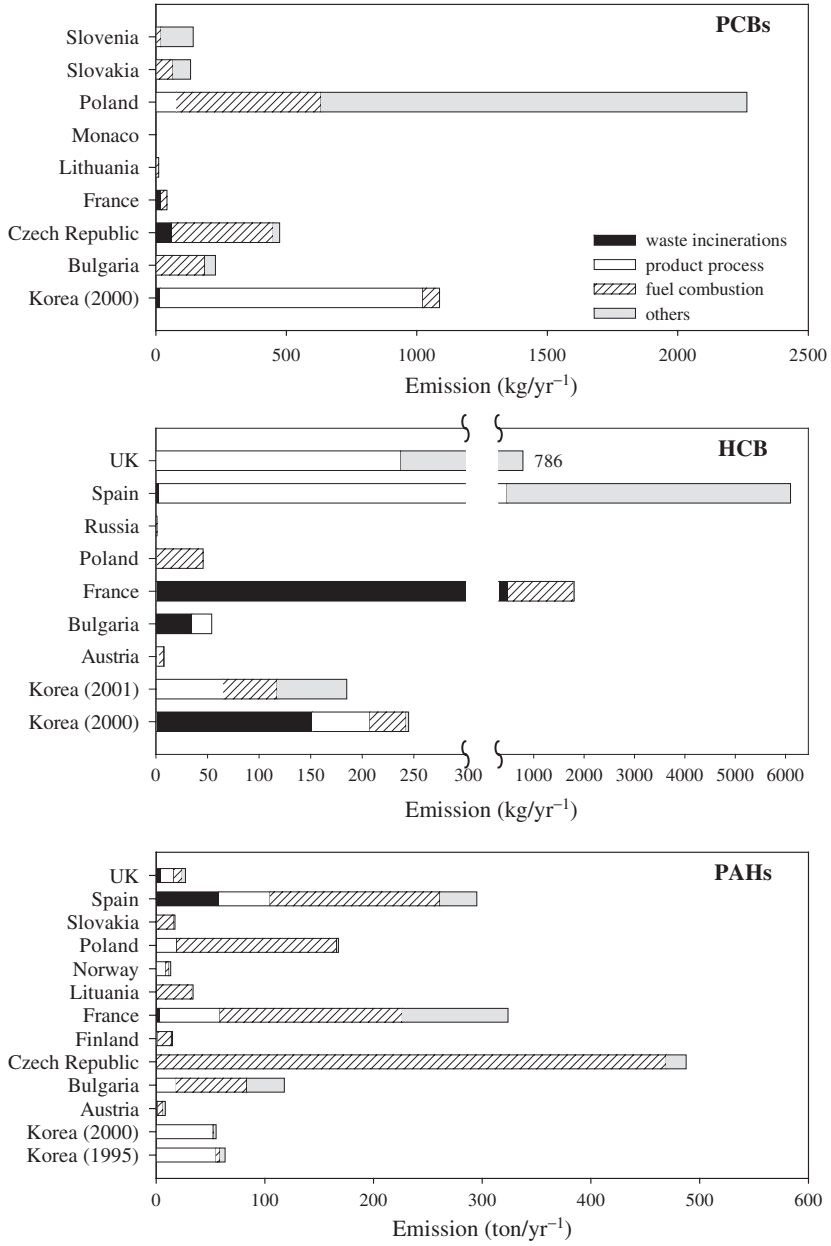

Fig. 2.6. Annual emission of by-product POPs (PCBs, HCB, and PAHs); Korea 1995 (from MCIE, 1999), Korea 2000 (from KMOE, 2003b); Korea 2001 (from KMOE, 2005a); Other countries 2000 (from UNECE/EMEP, 2002). PAHs emissions were based on 7 PAHs for Korea and 6 PAHs for other countries.

be greater if released from electric furnaces are included. KMOE (2005a) measured HCB emission in archived samples from ferrous/non-ferrous metal production, mineral production, chemical and energy production plants from 2001 to 2003, and some additional metal production factories, wastewater treatment plants, and non-point sources such as boiler and uncontrolled incineration in 2005. The measured emission rate was 185 kg yr^{-1} for 2001 as a reference year (Fig. 2.6). In contrast to the preliminary estimate, the individual source categories showed fairly even emission rates; that is, 34% from ferrous/non-ferrous metal production (63 kg yr^{-1}), 28% from fuel combustion (52 kg yr^{-1}), 36% from transportation (68 kg yr^{-1}). Iron ore sinter released four times less HCB than the estimated value for 2000, while the electric furnaces emitted 30 kg yr^{-1}. This discrepancy indicates that the uncertainty of preliminary estimates is still great and warrants further emission measurements. The later study of actual measurements did not include emission from waste incineration. Thus, the total HCB emission observed in 2001 would be greater if waste incineration were included as a source. For instance, total HCB emission would be 337 kg yr^{-1} if estimates of the preliminary study (ca. 152 kg yr^{-1}) are simply added. Based on an assembled list of potential sources for HCB, it was estimated that approximately 23000 kg HCB yr^{-1} (with a range of 12000–92000 kg yr^{-1}) was released in global environments during the mid-1990s (Bailey, 2001). No single major source of HCB was identified. Uniform contribution from pesticide application (28%), manufacturing (41%), and combustion (30%) were estimated. The contribution of South Korea to global HCB emissions was estimated to be approximately 1.3%.

2.4.1.4. PAHs

PAHs are not included in the list of 'dirty dozens POPs' of the Stockholm Convention. However, in addition to 12 existing POPs, UN/ECE (United Unions Economic Commission for Europe) considers PAHs, HCHs, chlorodecone, and hexabromobiphenyl as potential POPs to be restricted based on 'The New Protocol on Persistent Organic Pollutants to the Convention on Long-range Trans-boundary Air Pollution' adopted in 1998. PAHs may be listed as POP in the future Stockholm Convention.

PAHs are released to the environment from a number of sources: pyrogenic sources including fossil fuel combustion and pyrolytic processes of organic matter such as incineration and petrogenic sources such as oils spills. Direct oil spilled from stationary sources and accidents cause contamination of land. Oil spills at US Army bases in South Korea is reported as a source of soil and groundwater contamination to nearby area

and it has been suggested that remediation of these sites is required. Oil spills from ship accidents could be a major source of PAH to the marine environment. For example, the supertanker *Sea Prince* was stranded near Sori Island off the South Coast of South Korea. This accident alone released 4160 tons of crude oil and 880 tons of fuel oil into the marine environment (Yim et al., 2002).

Two preliminary studies of atmospheric releases of PAHs in South Korea have been conducted. The top–down approach was adopted to estimate the emission rate of each source by multiplying the emission factor reported by the US EPA (1998a) and the statistics of activities of the each source in South Korea (MCIE, 1999; KMOE, 2003b).

The total emissions of seven PAHs including BaA, CHR, BbF, BkF, BaP, DahA, I123cdP (see Table 2.2 for full names), identified by the International Agency for Research on Cancer (IARC) as animal carcinogens, were 63.4 ton yr^{-1} for 1995 and 55.3 ton yr^{-1} for 2000 (Fig. 2.6). A dominant contributor to PAH emissions was found to be coke oven process for the steel manufacturing, emitting over 60% of the total quantity. Chemical production processes and fuel combustion contributed approximately 10% and 5%, respectively. The total mass of the 16 priority or 'indicator' PAHs suggested by the US EPA, emitted in South Korea during 1995 was estimated to be 768 ton (MCIE, 1999) and 857 ton during 2000 (KMOE, 2003b). In the two preliminary studies, because many sources could not be included due to the lack of information, the total emissions were likely to be underestimated. In contrast to South Korea, fuel combustion was the predominant source of PAHs in EU countries. The discrepancy of national PAH emission profile among countries depends significantly on its industry patterns. One of the important reasons for the large contribution of coke oven process in Korea is the huge scale production of steel in Korea as compared to most European countries. The steel industry is not only a large emitter of an array of pollutants but an energy intensive one, and as a result, it is not blooming in most developed countries. This can be one reason that the inherent emission measurement based on its industrial pattern is required for each country.

2.4.1.5. Comparison of by-product POPs emission inventories

Measurements of emissions of by-product POPs, such as dioxin-like compounds and HCB have been performed and preliminary estimations exist for other POPs. However, due to uncertainty in estimations, comparisons among POPs should be made with caution. Most by-product POPs are likely to be produced from similar sources, principally pyrolytic

processes. According to the emission estimates, more than 60% of dioxin-like compounds and HCB were estimated to be emitted from waste incinerators, while substantial portions of PCBs and PAHs were released from metallurgical processes, including electrical furnaces and coke ovens, respectively (Fig. 2.7). These waste incineration and the metal production processes contributed over 80% to the total emission for each of

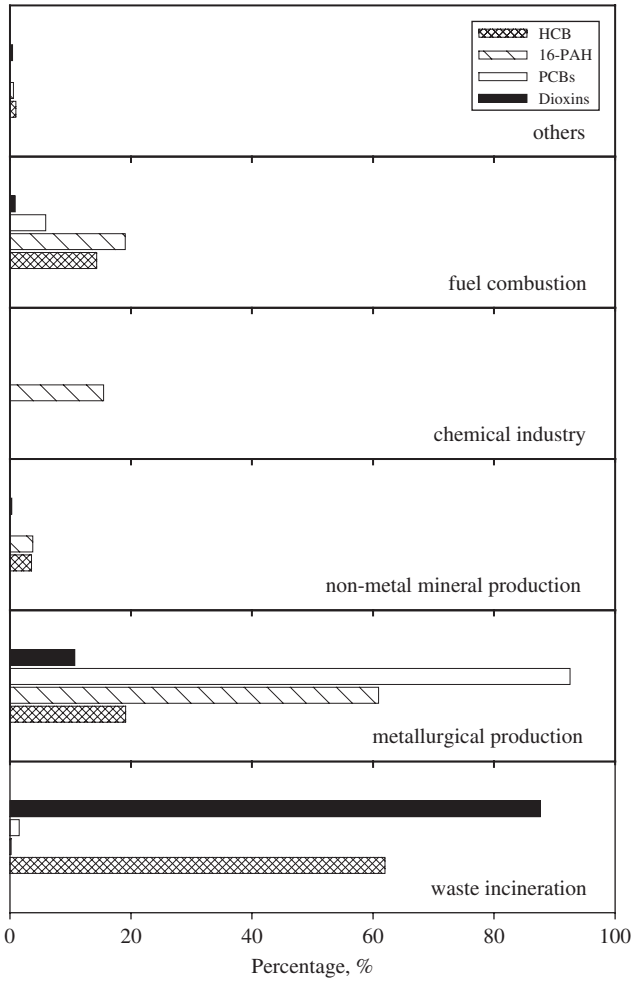

Fig. 2.7. Contribution of each emission source to total emission of by-product POPs in Korea. 'Dioxins' includes PCDDs, PCDFs and coPCBs measured in 2001 (see Table 2.6), and '16-PAHs' means the total emission of EPA priority 16 PAHs. PCBs, HCB, and PAHs emissions were quoted from the preliminary estimation study (KMOE, 2003b).

by-product POPs except for PAHs. For PAHs, the secondary contributors were fuel combustion (19%) and oil refineries (15%). This indicates that the release of by-product POPs in South Korea can be reduced efficiently by focusing the regulation of these two source categories of waste incineration and metallurgical processes.

2.4.2. Product POPs

The governmental works on the emission inventories of product POPs (i.e., PCBs, OCPs, and PBDEs) began only in 2003 as one of the strategies on Stockholm Convention. Preliminary studies (KMOE, 2003b; KMOE, 2004a) and investigation campaigns to determine the domestic status of PCB-containing facilities (KMOE website) are underway. The history of production and use of OCP products and intermediates was recently reviewed (KMOE, 2004a). Domestic use of PBDEs has been reviewed in a report on the use and import of brominated flame retardants (KMOE, 2005c). Distribution and emission of NP as one of APs has been investigated annually by KMOE. However, there is no information on the production and or use of PFAs in South Korea.

2.4.2.1. PCBs

The Stockholm Convention stipulates that, use of PCBs-containing articles should be forbidden after 2025, and it is recommended that PCB stocks (articles and wastes) be destroyed by 2028 by use of environmentally sound methods, and that national efforts for removal of PCBs should be submitted by signatory nations (Stockholm Convention on Persistent Organic Pollutants, 2001). Under an agreement among KMOE, electric power companies, and three NGO for removal of product PCBs in South Korea, the domestic use of PCB-containing transformers is being investigated nationwide and methods of establishing appropriate collection, storage, and treatment implemented.

A governmental investigation on PCB use is in process in South Korea. However, as no report is available yet, we reviewed the estimation of Breivik et al. (2002a), which provided the estimates of consumption of 22 major PCB congeners for 113 individual countries. These estimates are based on reports from PCB manufacturers and information on imports, exports, and consumption as well as restriction on production and imports, and the population and economic status. The total, historical global production of PCBs has been estimated to be approximately 1.3 million tons and Monsanto, the manufacturer of the Aroclors, produced about 50% of the total global production. The estimated cumulative

consumption of the 22 predominant PCB congeners in South Korea ranges from 3000 to 6500 tons (default consumption = 4300 tons), which ranks 16th out of 113 countries for which information is available (Fig. 2.8). This quantity is 2% and 20% of the consumption of the USA and Japan, respectively. Products containing PCBs such as Aroclor,

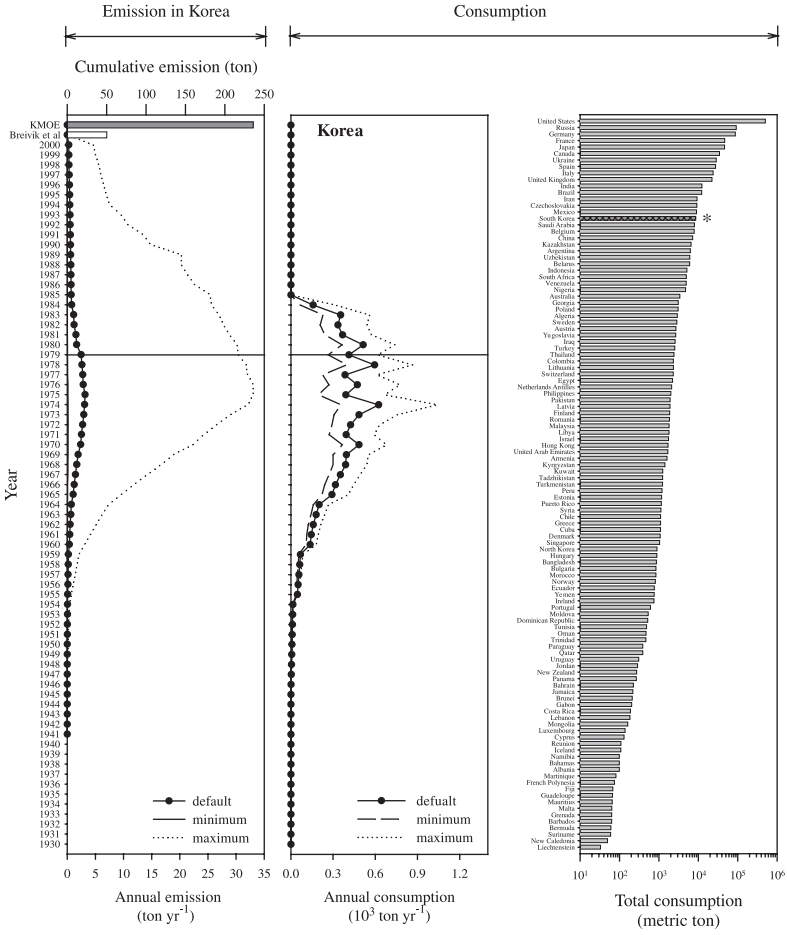

Fig. 2.8. Consumption (total PCBs's consumption modified 22 congener's consumption of Breivik et al., 2002a; *see* the text) and emission (sum of 22 congeners from Breivik et al., 2002b) of PCBs. Two bars in emission plot are the sum of emissions from 1945 to 2000 from Breivik et al., 2002b (white) and KMOE, 2003b (dark gray). Horizontal lines in the plots mean the first legal restriction point in Korea, 1979 (EBA; see Table 2.1). Total consumption of each country (far-right plot) was the sum of annual total PCBs consumption of each country (modified from the default 22 congeners' consumption of Breivik et al., 2002a).

Kaneclor, and Chlophen contain a wide range of congeners. Based on studies of sources of PCB contamination, the South Korean environment appears to be contaminated by a mixture of various products. The predominant product was found to be Aroclor 1254 in Incheon, Aroclor 1242 in Masan, Aroclor 1260 (or Kanechlor-600) in Ulsan, and Aroclor 1242 in the Han River (Kim et al., 2000a, 2002b; Kim, 2004; Hong et al., 2006). Breivik et al.'s 22 congeners comprise of only ~50% in Aroclor mixture (1242:1254:1260 = 1:1:1). Using the fact, we could estimate the total PCBs consumption in South Korea by multiplying a factor of 2 with the inventory for 22 congeners. Based on this estimation, total PCBs consumption in South Korea was estimated to be 6000–14000 tons (default, 9000 tons). When this 2-fold assumption was applied to other countries, the estimated total PCBs consumption was 462000–552000 (default, 500000 tons) for the USA and 30600–62200 (default, 47000 tons) for Japan. Assuming a total production of approximately 680000 tons in the USA (http://www.epa.gov/opptintr/pcb/) with essentially all of the production used domestically, our estimate matched well with the reported value. Furthermore, historical total consumption in Japan was reported to be 54000 tons (JEMCAA, 1997). This value is similar to the consumption estimated in the present study.

Emission of 22 PCB congeners, originating from unintentional use of PCBs-containing product such as accidental releases and disposals including landfill, open burning, waste incinerations, and destruction, has also been estimated (Breivik et al., 2002b). According to their estimation, the historical release of PCBs to the South Korea has been estimated, based on the 22 indicator congeners, to be 2.5–742 tons (default 53 tons). KMOE (2003b) also estimated the total release of product PCBs to the South Korean environment between 1945 and 2000 to be 235 tons by using an emission factor of 0.13 g capita^{-1} yr^{-1} as presented in EMEP/CORINAIR guidebook (EMEP/CORINAIR, 2000).

The primary use of PCBs, over 50%, was in dielectic fluids for transformer and capacitors (UNEP Chemicals, 1999). Moreover, leakage from transformers and capacitors is the major contributor of PCB to the environment (EMEP/CORINAIR, 2000). The South Korean government investigates PCBs-containing transformers nationwide and provides funds for development of PCBs waste treatment technologies. The South Korea Electric Power Corporation (KEPCO), practically the only electricity company in South Korea, has 1.7 million transformers and 540 capacitors (KMOE website). Recent voluntary investigations of electricity companies for 1237 transformers revealed that 22% (i.e., 272 transformers) contained PCBs at concentrations of greater than 2 ppm, which is the threshold to be categorized as a specific waste by the WCA

(KMOE website). The number of transformers containing more than 50 ppm PCBs, which is the guideline for the prohibition of use set by the TCCA, was 19 (i.e., 1.5%). In another study of 90 transformers (KMOE, 2004a), similar results were obtained. That is, 2.2% of transformers contained more than 50 ppm PCBs while 22% contained more than 2 ppm PCBs, while 78% of the transformers contained less than 2 ppm PCBs. The KEPCO reports that 13,610 kl of waste dielectic fluids and 394, 529 waste transformers occurred in South Korea and 643 tons of PCBs-containing waste was exported between 2000 and 2005.

2.4.2.2. OCPs

The classical OCPs have been restricted from use in South Korea since the late 1960s (Table 2.1). OCPs residues currently in the environment are unlikely to have originated from their recent use, they are rather residuals from historical applications. Historical production, distribution, and consumption of seven individual OCPs including DDTs, CHLs, heptachlor, aldrin, endrin, dieldrin, and toxaphene for pesticides and additive agents have been reviewed (KMOE, 2004a). Here, we have compiled the consumption data reported by KMOE for the OCPs and those of other studies for HCHs and Endosulfan (Lee, 1982; Jeong et al., 2001c). There is a large uncertainty in the database for OCPs usage in South Korea. DDTs and toxaphene consumption estimates from individual studies varied by a factor of 2 but those for aldrin and heptachlor estimated varied widely. For instance, heptachlor consumption was reported to be approximately 600 tons in one study (Lee, 1982), which is only 4% of the estimate of KMOE.

Total historical consumption of OCPs in South Korea was 31,000 tons with the following relative proportions: heptachlor (53%) > endosulfan (16%) > aldrin (10%) > DDTs (9%) > HCHs (6%) (Fig. 2.9). DDT, was the first OCP applied in South Korea and was widely used from the late 1940s to the early 1970s. This pesticide was replaced by other OCPs after the toxicity of DDTs to wildlife was known. Use of most OCPs was banned in the late 1960s, but less persistent compounds such as heptachlor and toxaphene were still used until the early 1980s. Endosulfan is still used in large quantities in many parts of the world. Endosulfan has been in use for more than three decades. It was introduced as a replacement for other more persistent OCPs when they were banned. For example, about 1000 tons were consumed in South Korea over five-year periods from 1971 to 1999 (Jeong et al. (2001c). According to KMOE's annual report for distribution and emission of industrial hazardous chemicals, 1144 kg of endosulfan was released to atmospheric from

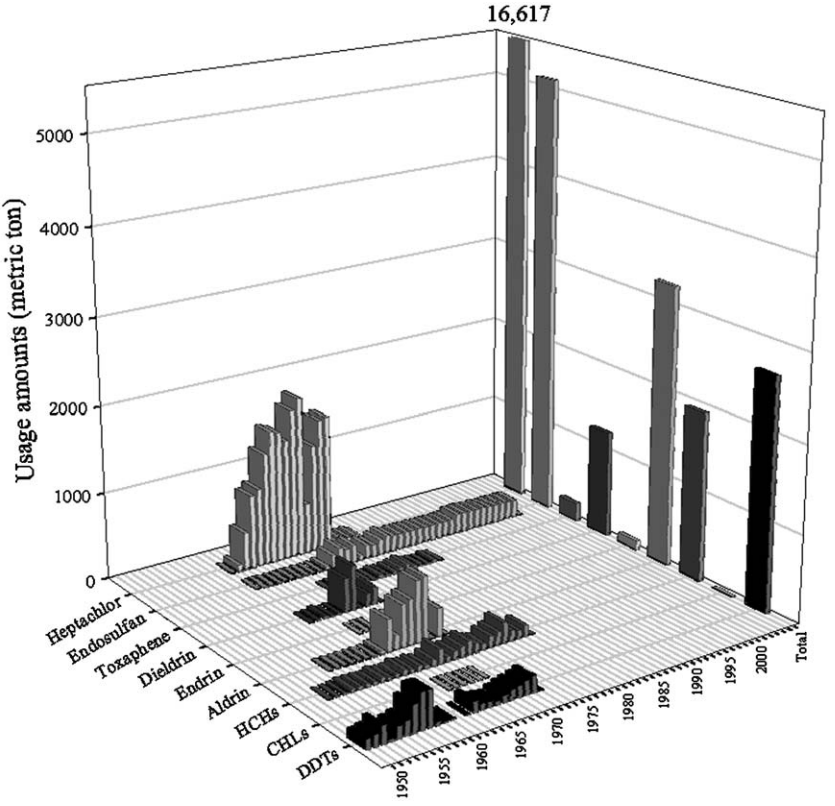

Fig. 2.9. Historical usage amounts of each organochlorine pesticides: DDTs, CHLs, Aldrin, Endrin, Toxaphene, and Heptachlor from KMOE (2004c), HCHs from Lee (1982), and Endosulfan from Lee (1982) by 1980 and modified from Jeong et al. (2001c) since 1981. Total endosulfan use of five years (from Jeong et al. (2001c)) was converted to annual average amount.

chemical processes and/or chemical manufacturing processes in South Korea. But, the amount released varies among years. For instance, 4 kg was estimated to be released in 1999 while 819 kg was estimated to have been released in 2003. These estimates are associated with great uncertainties. Furthermore, it is not certain whether the KMOE report includes releases of endosulfan from agricultural applications.

The order of contributions of OCPs to the total mass of OCPs measured in bivalves (Kim et al., 2002b) and sediment (Hong et al., 2006), was DDTs (63%) > HCHs (21%) > CHLs (11%) > dieldrin (1%). A possible explanation for this observation is the difference in chemical properties

among pesticides. The half-lives of DDTs, γ-HCH, and CHLs in soil and sediment are 3–10 times longer than those of heptachlor, aldrin, and endosulfan (Mackay et al., 2000; Ghadiri and Rose, 2001). Furthermore, the vapor pressures of heptachlor and aldrin are 10- to 1000-fold greater than that of DDT, chlordane, and β-HCH. Since the pesticides were applied on land, they may have been volatilized. More volatile pesticides have shorter residence times in soil and aquatic systems. DDTs and CHLs were used as additive agents even after the ban in use as pesticide. For instance, it has been reported that CHLs were used as an adhesive for plywood until 1995 (KMOE, 2004a). DDTs were formed as an impurity during the manufacturing of a pesticide, dicofol. Based on the consumption of dicofol from 1972 to 2002 with the DDT content of 0.1% in dicofol, additional releases of DDTs to the environment during the three decades was estimated to be 578 kg (KMOE, 2004a). This quantity is substantially smaller (0.02%) than its consumption as a pesticide until the early 1970s and does not affect the overall estimates of releases.

HCB is frequently detected in South Korean environments at concentrations that are less than DDTs > HCHs > CHLs. HCB was neither imported nor produced as a pesticide, but is known to be released into the environment at minimal concentrations as a trace contaminant in chlorothalonil. Estimated emission on the basis of HCB content (ca., 0.0219%) in chlorothalonil has been, on average, 77 kg yr^{-1} between 1970 and 2004 (KMOE, 2004a). This amount corresponds to between 20% and 30% of the emission as a by-product.

In the global estimation the usage of HCH in South Korea has been estimated to have been between 10,000 and 100,000 tons (Li, 1999). The great estimate was based on an assumption of very great application rates of 10–40 tons kha^{-1}. However, these estimates appear to be overestimated compared with approximately 2000 tons reported in other studies (Lee, 1982; Jeong et al., 2001c). This inconsistency could be due to false information of usage. Indeed, the concentrations of HCHs in South Korean environments were comparable or less than in many other countries. For instance, residual concentrations in coastal bivalves and sediment ranged from 0.2 to 6.6 (median = 0.1 ng g^{-1} wet wt.) and ND to 5.5 (median = 0.32 ng g^{-1} dry wt.), respectively.

The consumption of OCPs in South Korea can be compared with that of China, which is one of the largest consumers of pesticides in the world. China has used 200,000 ton yr^{-1} of total OCPs every year between 1970s and 1980s (Wang et al., 2005). Three major OCPs including HCHs, DDTs, and toxaphene, collectively accounted for 80.1% of total pesticide

use in 1970. The usage of HCHs and DDTs in 1980 corresponded to 70% and 5%, respectively, of gross pesticide usage.

2.4.2.3. APs

APs mainly originate from the degradation of alkylphenol polyethoxylates (APEOs), which are used as nonionic surfactants or detergents in industries and household products for over 40 years (Shang et al., 1999). APEOs are contained in cleaning products, paints, herbicides, and pesticides, and are used to facilitate processes in pulp and paper production and textile manufacturing (Field and Reed, 1996). APs, including NP and bisphenol A (BPA), have endocrine disrupting activity. In the past 40 years, APEOs production was estimated at $>300 \times 10^3$ ton yr^{-1} worldwide (White et al., 1994). Primary degradation of APEOs in wastewater treatment plants or in the environment generates more persistent and toxic shorter-chain APEOs and APs such as NP, OP, and butylphenol (BP) and AP mono- to triethoxylates than the parent substances (Giger et al., 1984).

Nonylphenol is a degradation product of nonylphenol polyethoxylates (NPEOs), which have been the most widely produced APEOs. NPEOs have a worldwide production of $\sim 700 \times 10^3$ tons annually with wide applications such as industrial and institutional (30%) or household (15%) cleaning agents (Ying et al., 2002; Knepper and Berna, 2003; Jonkers et al., 2005), and many other industrial applications including wetting agents, dispersants, emulsifiers, solublizers, foaming agents, and polymer stabilizers. NPEOs have been discharged into surface waters, mostly in wastewater or sewage. Many countries have banned the usage of APEOs surfactants: for instance, the European Union restricted the use of NP and APEOs (Cox and Drys, 2003). Neither inventory efforts nor regulatory action has been taken in South Korea.

BPA is used as an intermediate in the manufacture of epoxy resins and polycarbonate plastics. Global use of BPA is estimated to be more than a million tons per year with 347×10^3 tons yr^{-1} of estimated use in Western Europe in 1993 (BUA, 1997). Although BPA is expected to be released mainly via sewage and industrial wastewater system (Fürhacker et al., 2000), the significant emission is also estimated from combustion sources such as uncontrolled domestic waste burning with $\sim 75 \times 10^3$ kg yr^{-1} in the USA (Sidhu et al., 2005). KMOE reports the emission of hazardous chemicals including BPA annually. According to the report, atmospheric emission of BPA was 4446 kg yr^{-1} in 2002 and 1923 kg yr^{-1} in 2003 (KMOE website).

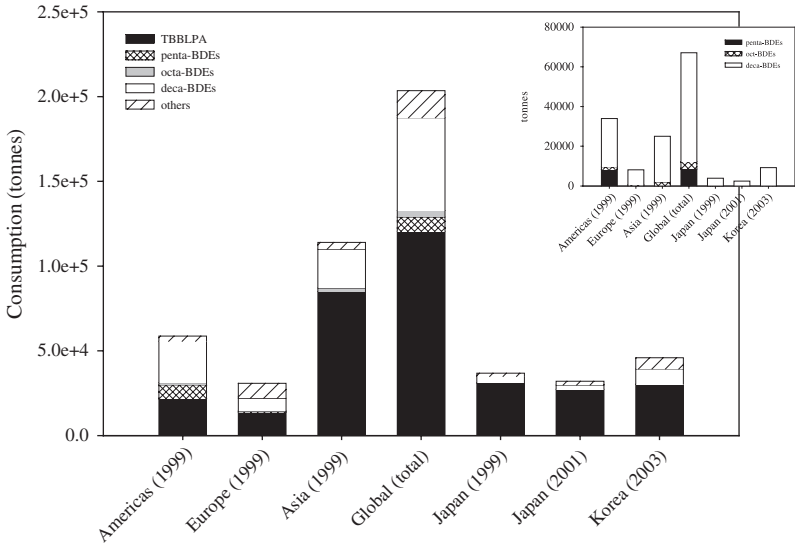

Fig. 2.10. Worldwide Brominated Flame Retardants (BFRs) consumption: Americas, Europe, Asia and Global consumption in 1999 (www.bsef.com), Japan in 1999 and 2001 (Watanabe and Sakai, 2003), and Korea in 2003 (KMOE, 2005c). Korea's values are not consumed but imported amounts. However, the consumption amount is estimated to be similar with this pattern (see the text).

2.4.2.4. PBDEs

The main types of brominated flame retardants (BFRs) are polybrominated biphenyls (PBBs), PBDEs, and tetrabromobisphenol A (TBBPA). PBDEs have replaced PBBs, which are the first brominated organic compounds to be used as flame retardants and were phased out because of environmental issues, but are now being replaced by TBBPA (Renner, 2000). The global consumption of BFRs is estimated to be 203425 tons in 1999 with PBDEs accounting for 33% (Fig. 2.10). Deca-BDEs, 82% of total PBDEs, are the most widely used product. Domestic BFR consumption in South Korea in 2002 was 49050 tons, of which 25% (i.e., 12408 tons) was PBDEs with deca-BDE accounting for 12324 tons and penta- and octa-BDEs accounting for 84 tons (KMOE, 2005c). Imported amounts of each product in 2003 showed a similar distribution to the consumption estimated in 2002 (Fig. 2.10). In BFR market of Western Europe, consumption of PBDEs has declined from 26% in 1996 to 11% in 1998 (DEPA, 1999). Penta-BDEs product has been restricted for over one decade and is now banned within Europe. Furthermore, there has

been a trend to substitute PBDEs with other flame retardants in northern Europe since 1996 (Renner, 2000). However, the market of BFRs in South Korea, sharing ~56% of flame retardant market, has increased annually by 13.5% on the average (KMOE, 2005c). In South Korea, PBDEs are regulated under the 'EcoLabelling policy'.

Possible major sources of BFRs are wastewater effluent and flue gases from BFR factories and other facilities processing BFRs (Watanabe and Sakai, 2003). Additives such as PBDEs may leach from the surface of products treated with these compounds, in contrast to TBBPA that are reactive and chemically bound within the material matrices (de Wit, 2002). Based on a model estimate, Danish EPA concluded that PBDE source is evaporation from products in use (DEPA, 1999). Thus, indirect releases from products should also be considered in emission inventories.

PBDEs, one of the major types of brominated flame-retardants, are used as additives in polymetric materials such as polystyrenes, plastics, paints, textiles, machines and household electric appliances such as computer and television, upholstery, and building materials. Commercial PBDE products consist predominantly of penta-BDEs, octa-BDEs, and deca-BDEs products. Each product is composed of several congeners. PBDEs is known to cause various adverse effects including neurobehavioral development (Darnerud, 2003), thyroid system (deBoer et al., 1999), and, some congeners elicit carcinogenicity (McDonald, 2002). Concentrations of PBDEs in environments have been rising in contrast to classical POPs whose levels have been declining markedly during the last 20 years (de Wit, 2002).

2.4.2.5. PFAs

Concern about per- and poly-fluoroalkyl acids (PFAs) including perfluorocarboxylates (PFCA) and perfluoroalkylsulfonates (PFAS) is increasing due to their persistence, bioaccumulation potential and potentially toxicity (i.e., PBT). Due to the presence of the high-energy carbon–fluorine bond, these compounds are resistant to hydrolysis, photolysis, microbial degradation, and metabolism by vertebrates. The PFAs are also bioaccumulated and biomagnified through food chains (Kannan et al., 2005; Sinclair et al., 2006). These anthropogenic compounds reportedly exert various toxic effects, including peroxisome proliferation and potential carcinogenicity (Renner, 2001a), accumulation of triglycerides in liver. These compounds have been routinely and globally measured in environmental matrices; water, wildlife, and human populations (Giesy and Kannan, 2001, Kannan et al., 2004; Yamashita et al., 2005).

The concentrations of PFAs in environments have increased sharply over time (Holmstrom et al., 2005). The sorption/accumulation and fate of PFAs differ from classical chlorinated and brominated POPs in that perfluorinated molecules have surfactant characteristics (i.e., having polar and nonpolar domains) and oleophobic properties. Thus, PFAs is likely to bind to serum protein rather than lipid in tissues (Jones et al., 2003).

Because of their chemical stability, surface tension lowering properties, and ability to create stable foams, PFAs are used in a wide variety of industrial and commercial applications and processes; metal plating and cleaning, coating formulations, fire-fighting foams, polyurethane production, inks, varnishes, vinyl polymerization, lubricants, pesticides, surfactants, gasoline and oil, and water repellents for leather, paper, and textiles (Holzapfel, 1966). Recent studies report that levels of PFAs precursors, degraded to more toxic and persistent metabolites, are greater in indoor than outdoor air due to their application to upholstery and carpets (Shoeib et al., 2004, 2005). The global distribution of PFAs has been hypothesized to be due to volatile neutral precursors of PFCA and PFAS that are distributed via atmosphere, and undergo long-range transport to remote areas, and degrade to yield the free acids (Martin et al., 2002; Renner, 2001b).

Due to their potential PBT properties, the major producer of polyfluorinated sulfonamidoalcohols in the USA and the only major company to use the electrochemical fluorination process, 3M, announced a phasing out of these products beginning in 2001. However, many companies including DuPont (USA), Clariant (Germany), Atofina (France), Asahi Glass (Japan), Daikin (Japan) still produce PFAs such as PFCAs and the products are still in use, worldwide.

The global historical industry-wide emissions of total PFCAs since 1950 are estimated to be 3200–7300 tons from direct (manufacture, use, consumer products) and indirect (PFCA impurities and/or precursors) sources with the majority (~80%) of fluoropolymer manufacture and use (Prevedouros et al., 2006). Total global production of sulfonated PFA is not known but global production by the major manufacturer as a raw material in 2002 was approximately 3700 tons (3M Company, 2000).

Little domestic emission inventory of PFAs has been estimated or investigated. However, according to the personal communication with 3M, total PFAs market in South Korea reached a maximum in 2000 since the stead introduction in the mid-1980s and dropped off recently. Most PFAs were imported from 3M, DuPont, Asahi Glass, Daikin, and others, of which over 90% was used as a water repellant for fiber and leather industries. However, the South Korean 3M stopped import of PFAs after 2004 following the official phase-out announcement of 3M in 2001.

PFCAs produced by telomerization process are likely to be the main PFAs consumed in South Korea. Since the 1950s, a number of textile factories have operated in Daegu and neighboring Kyeongsangbukdo. The 60–80% of PFAs imported is known to be consumed in these areas. Studies are needed for emission inventory and monitoring of PFAs in environments.

2.5. Levels, distribution, and exposure to POPs

2.5.1. Dioxin-like compounds (PCDDs, PCDFs, and coPCBs)

2.5.1.1. Environmental concentrations

Most studies of PCDDs/DFs conducted in South Korea have been focused on air quality (ca. 31%) or human exposure (ca. 28%; residues in human body tissues or food items). Concentrations of dioxin-like compounds in various environmental media are summarized in Table 2.7. We provide information stratified based on location as a source site, background site, or a nationwide indicator site. Concentrations of air ranged from ND to 9.946 pg I-TEQ m^{-3} with median value of 0.124 pg I-TEQ m^{-3}. The levels were in the order of nearby site of waste incinerator > industrial area > urban > rural. Compared with rural air (ND~0.235, median = 0.025 pg I-TEQ m^{-3}), concentrations in air at urban and source sites were, on average, 5 and 10 times greater, respectively. Larger cities tend to show greater contamination (Park and Kim, 2002). For example, in two larger cities with populations of over one million the concentration level was from 0.236 to 0.882 pg WHO-TEQ m^{-3} ($n = 6$, median = 0.882 pg WHO-TEQ m^{-3}) while that of a medium-sized city was 0.103–0.573 pg WHO-TEQ m^{-3} (median = 0.196 pg WHO-TEQ m^{-3}; $n = 8$). For three small-sized cities with populations of less than 0.3 million, the concentration level ranged from 0.017 to 0.18 pg WHO-TEQ m^{-3} ($n = 7$, median = 0.072 pg WHO-TEQ m^{-3}). Greater contaminations were also observed in pine needles from larger cities, where concentrations of PCDDs/DFs were 10-fold greater in the second largest city in South Korea, Busan, than that of Jeju, a remote island (Ok et al., 2002). Air of industrial cities ($n = 102$) was more contaminated (twice on average), relative to ambient air of eight major cities with a population over one million but with less industrialization ($n = 235$) (NIER, 2000a, 2001, 2002, 2003, 2004, 2005a). For instance concentrations of 0.006–8.450 pg I-TEQ m^{-3} (median = 0.255) were reported for more industrial cities while a range of 0.008–2.478 pg I-TEQ m^{-3} (median = 0.121) for cities with

Table 2.7. Concentrations of PCDDs/DFs in various ambient environmental media in Korea

Sampling			Total PCDDs/DFs				References[a]
Sample type	Year	*n*	Min	Max	Mean	Median	
Air			pg I-TEQ m^{-3}				
Near incinerators (or in industrial complex)	1999–2004	50	nd	9.95	0.56	0.29	(1), (2)
Urban area	1999–2004	408	nd	8.45	0.31	0.13	(3)–(8)
Rural area	1999–2004	42	nd	0.24	0.04	0.03	(2)
Nationwide (NIER)	1999–2004	386	nd	8.45	0.30	0.12	(2)
Soil			pg I-TEQ g^{-1}, dw				
Near incinerators (< 500 m)	2000–2004	162	0.01	130	13.6	5.37	(2), (9)–(12)
Near incinerators (> 500 m)	2000–2004	119	nd	56.45	3.72	1.48	(2), (9)–(12)
Industrial area	1999–2000	18	1.52	120	18.0	9.14	(8)
Nationwide (NIER)	1999–2004	251	nd	65.4	1.08	0.02	(7)
Water			pg I-TEQ L^{-1}				
Land-fill leachate	1998–2003	33	nd	175	18.4	4.02	(13)–(17)
Industrial/municipal wastewater	1999–2004	78	nd	50.5	1.60	0.006	(2), (18)–(20)
Liver/lake water	1998–2004	298	nd	3.35	0.066	0.007	(7), (15)
Coastal water	2002	5	0.127	0.996	0.377	0.207	(21)
Sediment			pg I-TEQ g^{-1}, dw				
Freshwater sediment							
Outlet of incinerator	2002–2004	9	nd	410	46.8	0.008	(2), (22)
River and lake	1998–2004	94	nd	15.1	0.222	0.009	(23), (24)
Marine sediment							
Masan bay	1992	11	0.554	73.7	11.1	2.92	(25)
Pohang bay	2000–2002	20	2.09	15.5	7.89	7.44	(26)
Ulsan bay	2000–2002	22	1.63	5.50	3.45	3.41	(26)
Busan bay	2000–2002	41	1.50	22.1	6.57	5.04	(26)
Jinhae bay	2000–2002	26	1.21	39.6	7.08	3.93	(26)

Gwangwang bay	2000–2002	35	0.569	2.12	1.37	1.38	(26)
Pohang bay (coPCBs)	2000–2002	20	0.324	2.67	0.615	0.463	(26)
Ulsan bay (coPCBs)	2000–2002	22	0.667	27.7	6.77	3.55	(26)
Busan bay (coPCBs)	2000–2002	41	0.273	24.0	2.99	1.71	(26)
Jinhae bay (coPCBs)	2000–2002	26	0.139	1.718	0.453	0.299	(26)
Gwangwang bay (coPCBs)	2000–2002	35	0.069	0.278	0.145	0.139	(26)
Organism			pg I-TEQ g^{-1}, ww				
Freshwater fishes							
Han River	1998–2001	20	nd	0.740	0.255	0.205	(15), (27)
Nakdong River	1998–2001	21	0.018	4.10	0.814	0.475	(15), (27)
Kum River	1998–2001	13	nd	0.740	0.152	0.100	(15), (27)
Youngsan River	1998–2001	15	nd	0.430	0.122	0.079	(15), (27)
Other river/stream/wetland	1998–2001	44	nd	3.405	0.514	0.283	(15), (23), (27)
Freshwater frog							
Han River	2001	6	0.150	0.615	0.327	0.273	(27)
Nakdong River	2001	6	0.180	1.26	0.682	0.705	(27)
Kum River	2001	4	0.030	0.375	0.201	0.200	(27)
Youngsan River	2001	3	0.220	0.760	0.427	0.300	(27)
Other river/stream/wetland	2001	8	0.010	0.680	0.279	0.205	(27)
Saltwater fish							
Masan Bay and Shihwa lake	1991–2000	8	0.027	0.637	0.302	0.348	(4), (28)
Saltwater Bivalvia							
Total coast	1996–2002	79	0.019	2.74	0.330	0.217	(26), (29), (30)
West coast	1999	9	0.043	0.760	0.175	0.077	(30)
South coast	1999	9	0.052	1.37	0.295	0.115	(30)
East coast	1999	9	0.026	2.74	0.648	0.239	(30)
Water birds			pg I-TEQ g^{-1}, fat				
Nakdong River estuary	1992–1994	42	15.8	899	–	–	(31)

[a](1) Oh et al., 2001, (2) NIER, 2005b, (3) Eem et al., 2003, (4) Im et al., 2004, (5) Park & Kim, 2002, (6) Kim et al., 2001a, (7) NIER, 2000a; 2001, 2002, 2003, 2004, 2005a, (8) Oh, 2001, (9) Kim et al., 2002c, (10) Park et al., 2004, (11) Kim et al., 2005b, (12) Kim et al., 2005d, (13) Lee et al., 2000, (14) Kim et al., 2001d, (15) Jeong et al., 2001a, (16) Choi et al., 2004, (17) KMOE, 2003a, (18) Shin & Jang, 2001, (19) Chung et al., 2001, (20) KMOE, 2005a, (21) MOMAF, 2003, (22) Pyoungtak City, 2003, (23) Kim & Hong, 2000, (24) Ryoo et al., 2005, (25) Im et al., 2002b, (26) Moon, 2003, (27) Kim et al., 2004a, (28) MOMAF, 2001, (29) Hashimoto et al., 1998, (30) Oh et al., 2003, (31) Choi et al., 2001a.

larger populations, but less industrialized. The population of South Korea tends to be concentrated in urban areas, with 90% of the people living in cities. Fifty percent of the total national population lives in eight major cities that occupy only 5.5% of the land area. That results in a population density of approximately 1875 people km^{-2} for these cities. The population density and industry of these largest cities generates a patchy distribution of contamination. Thus, human and ecological exposure may be more serious in these areas.

A concentration in soil, 3720 pg I-TEQ g^{-1} dw, was found in 1994 near an open burning site of industrial waste in the Masan Industrial area (Im et al., 2002a). Concentrations of PCDDs/DFs in 569 ambient soils measured since 1999 ranged from less than the limit of quantification to 130.4 pg I-TEQ g^{-1} (average = 5.84 and median = 0.61 pg I-TEQ g^{-1}). Recent monitoring of soils around waste incinerators has revealed that a strong source signature appeared only within a few hundred meters of incinerators when based on spatial distribution of the concentration (Park et al., 2004; Kim et al., 2005b; NIER, 2005b). Soils from 149 sites within 500 m from incinerators had concentrations of TEQ that were 5-fold and 14-fold greater than concentrations in soils from more distant locations; in that study, 124 soils from far sites between 500 m and 5 km from incinerators and 281 soils from random sites of non-incinerator measured nationwide were compared with those close to incinerators, respectively (Table 2.7). When concentrations in the areas near to incinerators were excluded, the eight major cities (0.045 pg I-TEQ g^{-1}) contained in soils average 4-fold greater concentration of PCDDs/DFs than other cities (median = 0.018 pg I-TEQ g^{-1}). The impact of incinerators on PCDDs/DFs soil contamination will be described in more detail (see part Section 2.6.1).

As mentioned earlier, direct release of PCDDs/DFs into aquatic systems (2.3 g WHO-TEQ yr^{-1}) is 0.6% of atmospheric release of 401.9 g WHO-TEQ yr^{-1} in 2004. The release of PCDDs/DFs into water occurs via landfill leachate or discharge of industrial and/or municipal wastewater, particularly paper-mill wastewater. PCDDs/DFs investigated in 413 samples ranged from ND to 175 pg I-TEQ L^{-1}. Although concentrations in some wastewaters discharged from paper-mills or waste incinerators exceeded 10 pg I-TEQ L^{-1} (Japanese standard for effluents), generally wastewaters discharged from incinerators or industrial and/or municipal waste treatment plants did not contain concentrations of PCDDs/DFs that were significantly greater than those of ambient water (two-tailed t-test, $p = 0.07$). Alternatively, leachates of waste landfill, contained significantly greater concentrations of PCDDs/DFs, relative to wastewater and ambient water (one-tailed t-test, $p < 0.01$). Particularly,

leachates contained concentrations of PCDDs/DFs that were more than 100-fold greater (median = 4.02 pg I-TEQ L^{-1}, $n = 33$) than those in ambient water (median = 0.007 pg I-TEQ L^{-1}, $n = 295$). This observation indicates that more studies should focus on the landfill leachates. No statistically significant difference was found among five major river systems (ANOVA, $p = 0.40$). However, it is important to note that the greatest concentrations were measured in the Han River and the Nakdong River, two of the longest rivers in South Korea. The river and sea water sampled at the Nakdong River estuary in 2003 contained the greatest concentrations measured in ambient waters (i.e., ~0.3 and 0.996 pg I-TEQ L^{-1}, respectively) (MOMAF, 2003). Additionally, samples collected at 31 national monitoring sites for freshwater showed a relative greater concentration in the Han River (median = 0.34 pg I-TEQ g^{-1} ww) and the Nakdong River (median = 0.43 pg I-TEQ g^{-1} ww) than other river (median = 0.19 pg I-TEQ g^{-1} ww) (Kim et al., 2004a).

Concentrations of PCDDs/DFs in sediments have been measured in three areas, including sediment at the wastewater discharge from waste incinerators, ambient freshwater sediments, and seawater sediments along the coast (Table 2.7). Sediments at the incinerators' wastewater discharge exhibited the greatest range of concentrations with concentrations ranging from below the limit of quantification to a maximum of 409.6 pg I-TEQ g^{-1} dw. Due to the small number of samples ($n = 9$), it is impossible to determine whether the concentrations were significantly greater in sediment at incinerator wastewater discharge. Approximately 77% of the concentrations of PCDDs/DFs in freshwater sediments ($n = 80$) were less than 0.1 pg I-TEQ g^{-1}. Relatively great concentrations were measured in inland sediment from waterways, into which wastewater from industrial areas is discharged. In particular, a few hundreds of pg I-TEQ g^{-1} were measured in sediment from the Hyungsan River in the vicinity of POSCO I.C. in Pohang city (Koh et al., 2004). This is the largest steel production facility in South Korea. Concentrations of PCDDs/DFs in marine sediments from major bays (Pohang, Ulsan, Busan, Jinhae, and Gwangyang) were greater than 1 pg I-TEQ g^{-1} (Moon, 2003). Concentrations of PCDDs/DFs in sediments from these five bays were: Pohang (7.24 pg I-TEQ g^{-1}) > Busan (6.55 pg I-TEQ g^{-1}) > Masan (3.88 pg I-TEQ g^{-1}) > Ulsan (3.18 pg I-TEQ g^{-1}) > Gwangyang (1.33 pg I-TEQ g^{-1}) on basis of median value (Moon, 2003). However, contamination of sediments from Ulsan Bay is as serious as Pohang and Busan with median concentrations of 7–8 pg I-TEQ g^{-1}, if coPCBs are included (Table 2.7). Co-planar PCBs accounted for about 55% and 27% of the total TEQs in surface sediments from Ulsan and Busan Bays, respectively. These relatively greater contributions of coPCBs, compared with <10% for the other bays, indicate

that PCDDs/DFs contamination may be caused by commercial PCBs formulations at these two bays. The average contamination level for marine sediment along the entire coastal area is likely to be lower because the five bays are sort of hot spots of POPs contamination in Korea.

coPCBs contributed >90% to total TEQs of dioxin-like compounds in most PCBs products such as Aroclor, Kanechlor, and Clophen (Wakimoto et al., 1988). Major 'hot spots' of PCDDs/DFs in coastal areas such as big harbor can be influenced by releases of PCBs products. This conclusion is based on the relative contribution of coPCBs to total TEQs or PCDDs/DFs pattern relative concentrations of congeners measured in the costal areas samples. Homologue compositions of PCDFs in the more contaminated sediments, such as those from Masan Bay were similar to that of commercial PCB products, KC-300 or KC-400 (Im et al., 2002b). Additionally, according to the results of the mussel watch programs from 1987 to 1999 (Oh et al., 2003), bivalves collected along the entire coastal area showed a clear difference in coPCB contribution to total TEQs between 1987–1990 and 1996–1999, and between the larger harbor areas and others. On average 82% of the TEQ was contributed by coPCBs in the earlier years' samples ($n = 13$) but only 32% in more recently collected sediments ($n = 38$). This indicates that concentrations of PCBs have decreased in these areas, instead of concentrations of PCDDs/DFs increasing over time. In more recently deposited sediments there is a greater contribution from coPCBs relative to PCDDs/DFs in larger harbors, such as Incheon, Ulsan, Masan, and Busan (see the map in Fig. 2.3). In contrast, mussels from the vicinity near the POSCO facility contained relatively less contribution (7%) by coPCBs to total TEQs. It has been suggested that PCB contamination in larger harbors could be ascribed to commercial formulations used in ship-painting (Hong et al. 2005). Consequently, contamination of larger harbors with dioxin-like compounds could be caused by commercial PCBs products, whereas impact of combustion sources seem to be greater in other areas.

There are relatively few reports of concentrations of PCDDs/DFs in wildlife from South Korea. The few available studies are comprised of monitoring for: (1) fishes and amphibian in inland rivers and lakes; (2) shellfishes and fishes in coastal region; and (3) birds and pine needles (Table 2.7). Little difference in concentrations of PCDDs/DFs was observed between inland fishes and marine shellfishes (two-tailed t-test, $p = 0.99$). Nevertheless, coastal environments are expected to be more heavily contaminated considering the possibility that fishes at higher tropic levels can bioaccumulate these residues to a greater extent. For instance, concentrations of PCBs in shellfishes for 1999 (Kim et al., 2002b) and fishes for 1997 (Yim et al., 2005) sampled along the entire

coastal area can be compared. Concentrations in fishes were, on average, two-fold greater than those in shellfishes, even though PCBs in fishes were measured for major 22 congeners while 104 congeners were analyzed in shellfishes. That is, 4.5–96 ng g^{-1} ww (average = 23 ng g^{-1} ww) for fishes vs. 0.27–84.6 ng g^{-1} ww (average = 12.6 ng g^{-1} ww) for shellfishes. Among coastal regions, concentrations of PCDDs/DFs greater than 0.5 pg I-TEQ g^{-1} ww was found in bivalves around larger industrial complexes such as Incheon, Yeochun, Ulsan, and Pohang (Oh et al., 2003). Together with these areas, bivalves from Masan and Busan exceeded 1 pg WHO-TEQ g^{-1} ww when including coPCBs TEQs (Oh et al., 2003). Concentrations of TEQs (i.e., PCDDs/DFs TEQs + coPCBs TEQs) in the other coastal regions were less than 0.5 pg WHO-TEQ g^{-1} ww in bivalves (Oh et al., 2003).

Monitoring of residues in fish and amphibians has been performed by the NIER at 31 sites along major Rivers (Jeong et al., 2001a; Kim et al., 2004a). In two extensive monitoring programs, concentrations of PCDDs/DFs were generally greater in rivers flowing through larger cities and industrial areas. The order of decreasing concentrations is: Nakdong River > Han River > other rivers and wetlands. Mountain frogs from the Nakdodng River contained the greatest concentrations among amphibians from rivers and wetlands investigated (Kim et al., 2004a). Furthermore, in the period from 1992 to 1994, concentrations of PCDDs/DFs as great as 100 pg WHO-TEQ g^{-1} fat were measured in the subcutaneous fat of water birds from Nakdong River estuary (Choi et al., 2001a).

There was little correlation between PCDDs/DFs (Jeong et al., 2001a) and total PCBs (Jeong et al., 2001b) in the fish samples from 31 inland sites. Based on the composition of coplanar PCBs congeners, it was proposed that contamination of the atmosphere with PCBs could be caused by waste incinerators (Kim et al., 2003b; Kim et al., 2005c). However, the contribution of waste incinerators to environmental release of byproduct PCBs is trivial. Net gaseous exchange of PCBs occurs from water to air and PCB profiles in river sediments resemble that of commercial PCBs products (Kim, 2004). Moreover, concentrations of PCB in South Korean air vary with temperature (Yeo et al., 2004a). Based on these observations, concentrations of PCB in South Korean air are unlikely to be controlled by atmospheric emission (see the part Section 2.6.2.1 for more discussion). Consequently, concentrations of PCBs in aquatic environments are less dependent on atmospheric concentrations, but more likely due to direct releases into water. In contrast to PCBs, the major source of PCDDs/DFs in South Korea is airborne releases, particularly from waste incinerators. Therefore, the discrepancy between PCBs and PCDDs/DFs residues in inland fishes could be due to the difference in their sources.

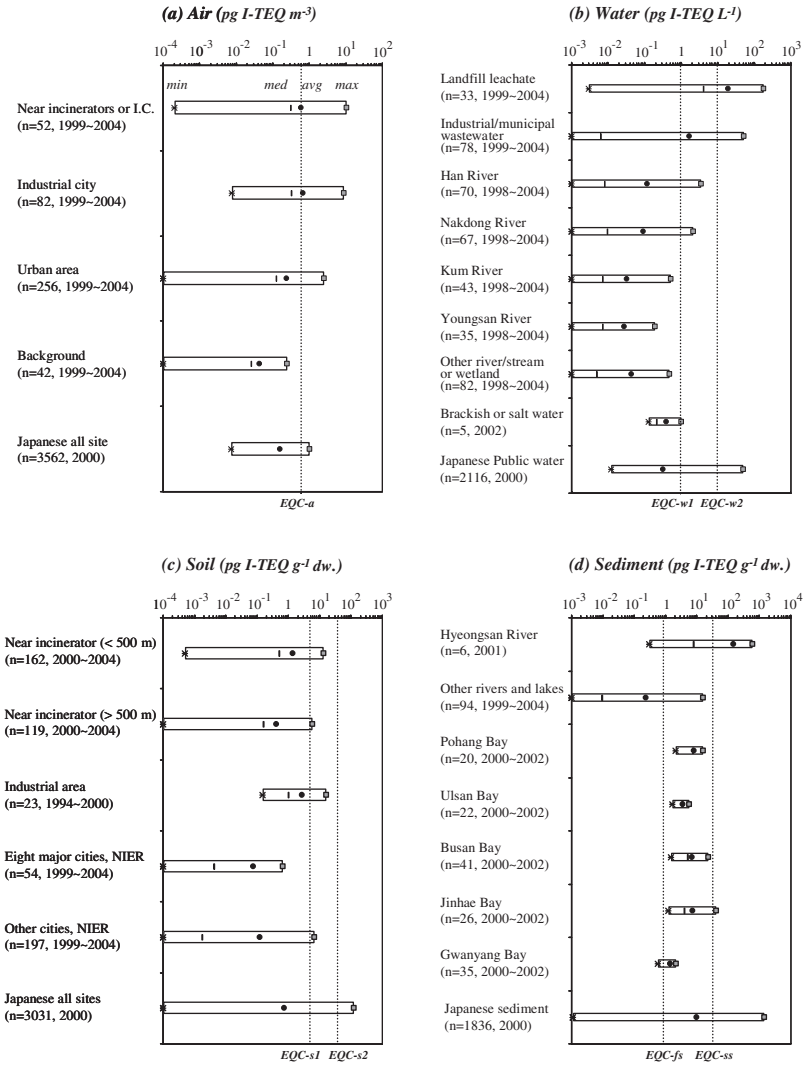

Fig. 2.11. Comparison of PCDDs/DFs levels in Korean environments with environmental quality criteria proposed worldwide. Range indicates minimum (min), maximum (max), median (med), and arithmetic mean (avg). Dotted lines indicate environmental quality criteria: 0.6 pg TEQ m^{-3} for ambient air (EQC-a), 1 pg TEQ L^{-1} for ambient water (EQC-w1) and 10 pg TEQ L^{-1} (EQC-w2) for wastewater, 5 (EQC-s1) and 40 (EQC-s2) pg TEQ g^{-1} dw for soil, and 0.85 (EQC-fs) and 21.5 (EQC-ss) pg TEQ g^{-1} dw for sediment. Japanese data is based on WHO-TEQs including PCDDs/DFs and coPCBs (JMOE, 2002).

2.5.1.2. Comparison of environmental levels with EQCs

Since the early 1990s, the Japanese government has conducted extensive surveys of both sources and environmental concentrations of dioxin-like compounds nationwide (JMOE, 2002). Comparison of concentrations of residues between South Korea and Japan suggests that concentrations in air are greater in South Korea than in Japan (Fig. 2.11). However, for other media, concentrations are similar or less in South Korea than in Japan. Intake rates of PCDDs/DFs, expressed as TEQ, by people in South Korea is similar to or less than those of people in other countries (see Section 2.5.1.5). As presented earlier, environmental emissions of dioxin-like compounds per unit area is greater in South Korea than other countries. Thus, relatively lesser concentrations in the South Korean environment except air seem to be inconsistent with emissions. One possible explanation for the inconsistency may be associated with the short history of waste incineration. The mass balance of PCDDs/DFs has not yet been thoroughly evaluated. However, according to multi-media modeling, it is evaluated that it takes more than a decade for POPs to reach steady state in soil or sediment (Lee et al., 2004). Waste incineration in South Korea became significant only after mid-1990s (Fig. 2.4). Furthermore, since 1997, releases from waste incinerators have been regulated. This suggests that their levels in soil or sediment may increase with time in the future unless emission level decreases significantly. Another reason could originate from the possibility that the number and location of the monitoring sites may not be representative.

In South Korea, no environmental quality criteria (EQCs) have been proposed to protect human health and wildlife from dioxin-like compounds. Thus, we compared concentrations of TEQ measured in individual media with EQCs or benchmarks for PCDDs/DFs proposed by other governments worldwide (Fig. 2.11). Japan proposes EQCs for ambient air and water of 0.6 pg WHO-TEQ m^{-3} for air, 1 pg WHO-TEQ L^{-1} for public water and 10 pg WHO-TEQ L^{-1} for wastewater (JMOE, 2002). Since 1999, concentrations of TEQ measured in air from within 1 km of waste incinerators and/or industrial area and sites in industrial cities, have exceeded the EQCs established by Japan (0.6 pg WHO-TEQ m-3) 22% and 29%, respectively. Alternatively, only 7% of air samples from urban and background sites exceeded the EQCs (Fig. 2.11a). This indicates that air quality is poorer in South Korea than in Japan where only 1.1% of 920 air samples collected in Japan during 2000 exceeded EQC (Shibata, 2002). The EQC values are applied differently, depending on the objective. Generally, landfill leachates exceeded the EQC (10 pg WHO-TEQ L^{-1}) more frequently (30%) than the wastewaters discharged

from treatment plants or industrial areas (3.8%). Only two samples of 302 fresh and salt waters measured contained concentrations of TEQ that exceeded the 1 pg WHO-TEQ L^{-1}, which is less than 3.9% of 2116 sites for Japanese waters were over the EQC. However, it should be noted that TEQs contributed by coPCBs were included for Japanese TEQ values but not for those from South Korea.

EQCs for soils vary among countries from 4 to 1000 pg TEQ g^{-1} dw. For instance, 1000 pg TEQ g^{-1} was suggested as the limit for residential soils in Japan and the USA but 500 pg TEQ g^{-1} has been proposed as the criterion in Finland, 10 pg TEQ g^{-1} in Sweden, and 4 pg TEQ g^{-1} in Canada (Schulz, 1993; JMOE, 2002; CCME website). Of the countries, Germany adopts specialized soil EQCs to protect the human health and the environment for different soil uses (Schulz, 1993). According to the EQCs, no restrictions are placed on use of soils containing less than 5 pg TEQ g^{-1} dw, precautious use and identification of PCDDs/DFs contamination source are required for soils containing between 5 and 40 pg TEQ g^{-1}, and, for soils containing more than 40 pg TEQ g^{-1}, use for cultivation of fruits, legumes, or forage plants is prohibited. Furthermore, soils containing more than 100 pg TEQ g^{-1} should be removed in playgrounds and, for soils over 1000 pg TEQ g^{-1}, measure should be taken to reduce contact with soil. Since 1999, 10 (ca. 4%) of 251 samples measured in non-source areas of South Korea exceeded the 5 pg TEQ g^{-1} criterion. Only two concentrations exceeded 40 pg TEQ g^{-1}. On the contrary, soils in the vicinity of sources like incinerators or industrial complex frequently exceeded 5 pg TEQ g^{-1} and, to a less degree, exceeded 40 pg TEQ g^{-1}. For instance, 49.4% of soils within 500 m of incinerators, 21% of soils more than 500 m from incinerators, and 69.6% of soils in industrial areas (as in Masan/Changwon I.C. in 1994 and Gwangyang/Yeocheon I.C. in 1999 and 2000) were greater than 5 pg TEQ g^{-1}. Soils with greater than 40 pg TEQ g^{-1} over which cultivation is prohibited account for 11.7% for within-500 m of waste incinerators and 13% for industrial origin as compared to only 0.8% of the soils beyond 500 m from the incinerators. Two samples were found to exceed 100 pg TEQ g^{-1} for both near-incinerator and industrial area. Only one sample near incinerator was measured to be 3720 pg TEQ g^{-1} in 1994. Based on this observation, ambient soils in non-source areas require no restrictions on use. For soils in the vicinity of sources such as waste incinerators and in industrial complexes, however further investigation and regulations are needed.

Various sediment quality guidelines (SQGs) and benchmark values have been proposed by several states and federal agencies in the USA and other countries. However, none of these benchmarks have been adopted for widespread regulatory applications. Concentrations of TEQ in

sediments of South Korea measured since 1999 were compared with SQGs for PCDDs/DFs proposed by Canadian Council of Ministers of the Environment (CCME) to protect human health and wildlife (CCME, 1999). CCME proposed 0.85 pg TEQ g^{-1} dw as an interim guideline and 21.5 pg TEQ g^{-1} as probable effect level for both freshwater and saltwater sediment. Except for sediment from the Hyeongsan River (Koh et al., 2004), none of the concentrations of TEQ in 94 freshwater sediments exceeded the probable effect level, over which an incidence of adverse biological effects of 46% would be predicted. Concentrations of TEQ in only three samples were in the range of 0.85–21.5 pg TEQ g^{-1}, indicating 24% incidence of adverse biological effects. Most of the 144 marine sediments from industrialized or urbanized bays contained concentrations of TEQ that ranged from 0.85 to 21.5 pg TEQ g^{-1}. The proportion of TEQ contributed by coplanar PCB was not included for sedimentary TEQ values in the present study. While concentrations of PCDDs/DFs in South Korean environments are generally less than proposed quality criteria for ambient environments, but more control is needed for environments in the immediate vicinity of sources.

2.5.1.3. Source identification of PCDDs/DFs in environmental media

For appropriate and cautious use of soils, sources of PCDDs/DFs contamination and their impacts should be evaluated. One way to differentiate source from ambient sample is the fraction of total PCDDs are greater in ambient samples than in the source. The ratios of PCDDs are on average 0.59($\pm$0.20) and 0.80($\pm$0.19) for landfill leachate and ambient water, respectively. The ratios are 0.53 and 0.81($\pm$0.36) for sediment at incinerator wastewater discharge and ambient river/lake sediments, respectively. This trend was also observed for soils. The proportion of the PCDDs fraction of TEQ increases with increasing distance from incinerators with the following proportions: 0.58($\pm$0.16) for ashes; 0.58($\pm$0.17) for soils within 500 m from incinerator; 0.74($\pm$0.15) for soils beyond 500 m from incinerator; and 0.91($\pm$0.18) for ambient soils. The dramatic increase in the proportion of the TEQ attributed to PCDD in the ambient environment results from the phase out of PCDF congeners and persistence of OCDD (Fig. 2.12). For all environmental media, lesser chlorinated PCDDs/DF congeners are uniformly detected in source samples. However, the less chlorinated congeners are distinctly depleted in ambient samples while OCDD is enriched. The transition of the relative proportions of PCDDs/DFs from the initial source to remote areas will be discussed in detail (see Section 2.6.1).

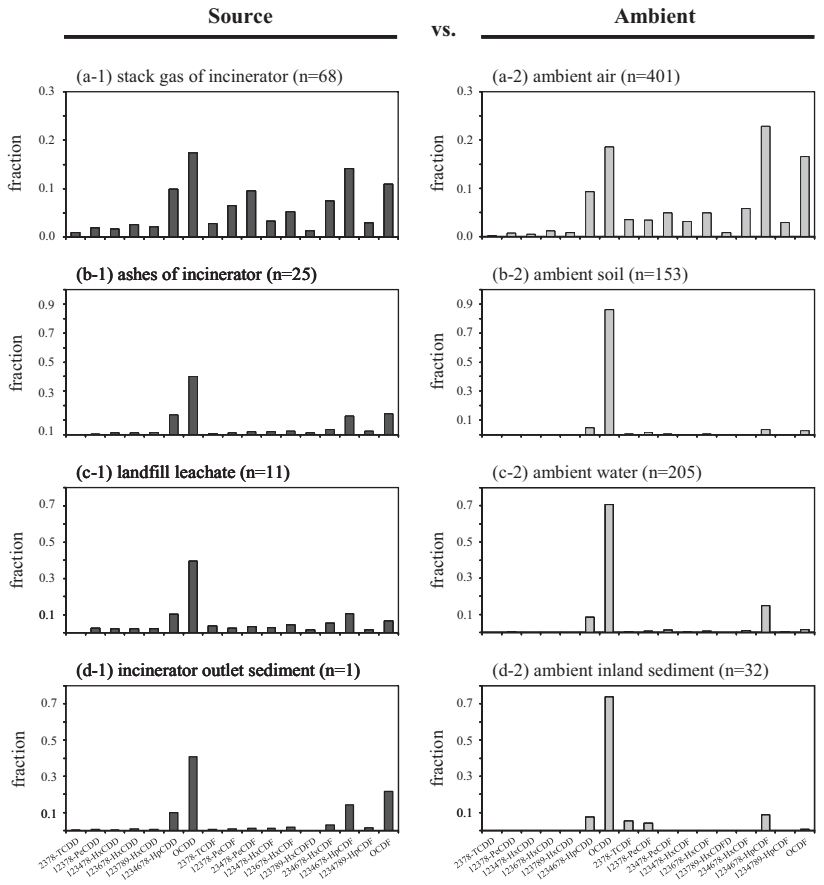

Fig. 2.12. Comparison of average fraction profiles in individual media between source and ambient environments; Data were collected from references in Table 2.7.

2.5.1.4. Temporal distribution

The decrease in concentrations of PCBs in coastal bivalves, between 1987–1990 and 1996–1999 was observed more clearly than that of PCDDs/DFs (Oh et al., 2003). However, it is not possible to assess temporal trend of PCDDs/DFs, because there is little information about PCDDs/DFs contamination level for environmental media before the late 1990s.

Emission of dioxin-like compounds has decreased progressively since the regulation with a 67% decrease between 2001 and 2004 (see Fig. 2.5).

Thus, it is important to assess whether the decline of emission has resulted in decreases in concentrations in the various environmental compartments. The NIER, a principal research organization for environmental residue monitoring of POPs in South Korea, has been annually conducting a regular monitoring at 130 sites for ambient media including air, water, sediment, and soil since 1999 (NIER, 2000a, 2001, 2002, 2003, 2004, 2005a). There is little available concentration data for temperal variation of other POPs (i.e., 'not detected' for lots of measurements reported or 'detected' in only a specific media) because the detection limits were relatively great. Meanwhile, enough monitoring data has been reported for PCDDs/DFs due to the use of standard operation procedure (SOP) with the state-of-the art analysis technique such as HRGC/HRMS and administrative notification for the Certified Laboratory for PCDDs/DFs analysis since the late 1990s.

In general, concentrations of PCDDs/DFs have decreased in all media (Fig. 2.13). Decrease in ambient air was significant with annual incremental decreases observed. For instance, the declines in all of arithmetic mean, median, and maximum concentrations were significantly correlated with those in annual emission of individual years presented in Fig. 2.5a ($r > 0.7$, $p < 0.05$). An eight-fold decrease was observed over a period of 6 yr. This decline in concentrations of PCDDs/DFs is related to regulation of emissions in South Korea. Because most of environmental release occurs primarily into air, residues in other media would also be expected to follow the same declining trend observed in air.

It seems, however, less clear if decreases in concentrations of PCDDs/DFs have occurred in soils or water. The responding point at which the concentration in soil begins to decrease is known to be delayed by a few years (typically ~3–5 years) depending on the emission scenario. According to the model performance (Lee, 2005), concentrations in soils continues to increase gradually even after regulation, then reaches a maximum in 3rd year after regulation and thereafter decreases. This may be because of overall atmospheric input (i.e., dry/wet deposition and sorption) to soil is still greater than total output from soil including volatilization, run-off, and degradation. It is not until the reversal phase occurs between input and output that residue in soil is expected to decrease. This means that atmospheric concentrations should be reduced up to the level that can result in a decrease in concentrations in the soil. Furthermore, it has been determined that concentrations in soil decrease slowly due to its strong sorption capacity. Thus, it takes decades for concentrations to decrease to the half of the original steady state level in realistic emission reduction conditions (Lee, 2005). It should also be

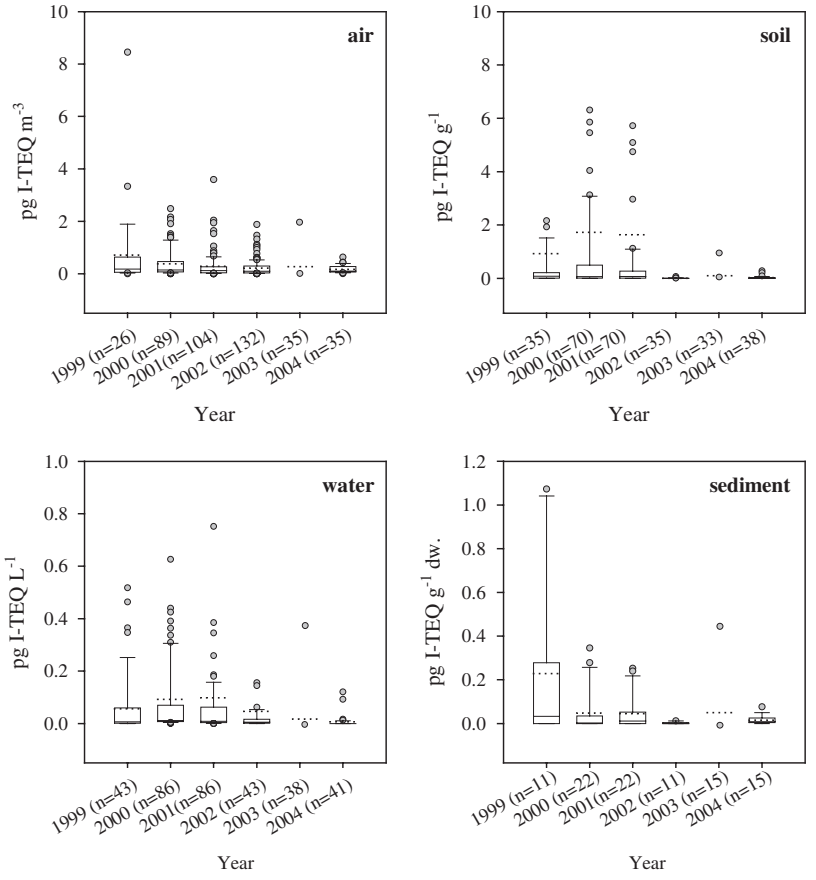

Fig. 2.13. Temporal trend of PCDDs/DFs concentration in individual environmental ambient media (NIER, 2000a, 2001, 2002, 2003, 2004, 2005a). The boundary of the box indicates the 25th and 75th percentile, and a line within the box marks the median. Whiskers above and below the box indicate the 90th and 10th percentiles. Dotted lines and points are the arithmetic mean and outlying points, respectively. Values in the parenthesis are the number of samples. Two points and a dotted line of individual media for 2003 indicate maximum (upper) and minimum (below), and arithmetic mean, respectively. Outliers in early years (i.e., four for water (1–4 pg I-TEQ L^{-1}) and five for soil (20–70 pg I-TEQ g^{-1})) were excluded.

noticed that concentrations of PCDDs/DFs in air cannot decrease dramatically, even after regulation because soil plays a secondary source for air contamination after regulation. Therefore, residues in both soil and air need to be monitored continuously. In brief, considering the temporal trend of the residues in the media, the distribution and fate of

PCDDs/DFs in South Korean environments seems to be strongly influenced by emissions to the atmosphere (see Section 2.6.2.1).

2.5.1.5. Human exposure

Concentrations of PCDDs/DFs in human blood, adipose and breast milk have been monitored in the South Korean population. Concentrations of PCDDs/DFs in tissues of the South Korean population ranged from 0.1 to 105 pg I-TEQ g^{-1} lipid. In spite of the great variation, the average exposure was 10–20 pg I-TEQ g^{-1} lipid (Fig. 2.14). This level is within the average human background range, 10–30 pg I-TEQ g^{-1}, as suggested by WHO in 1998 (WHO, 1998). The difference in the average exposure between workers and residents living near waste incinerators seems not clear in many cases (Fig. 2.14). However, the exposure may vary with the nature of the work within an incinerator. According to the study of Kim et al. (2005d), the body level was over 60 pg I-TEQ g^{-1} lipid for incinerator workers, handling fly ash, or operating and maintaining the incinerators for more than 6 years whereas that of new employee or office worker was about 10 pg I-TEQ g^{-1} lipid. This suggests that exposure via inhalation could be significant for incinerator workers. The residents in urban and industrial cities showed the body level 2–5 times higher than that of the residents in remote areas (~4 pg I-TEQ g^{-1} lipid) (Fig. 2.14). TEQ contributed by dioxin-like PCBs (coPCBs) in human blood accounted for ~70% of PCDDs/DFs TEQs for both workers and the general population (Kim et al., 2005e). Thus, when coPCBs are included, the total TEQ concentration could be approximately two-fold greater.

When excluding accidental or occupational exposure, over 90% of human background exposure to dioxin-like compounds is estimated to occur through the diet, with food of animal origin being the predominance source (US EPA, 2004). Residues of dioxin-like compounds in various food items collected from South Korean markets are summarized in Table 2.8. Fishes and shellfishes bore the greatest concentrations of TEQ, followed by meat/poultry > dairy product > other non-animal food-items. Considering the gradual increase in animal food consumption from 7% of the diet in the late 1970s to 20% in the 2000s, exposure of humans to these toxic compounds would be expected to be greater in more recent years. In contrast to those in other food-items, contribution of coPCBs to total TEQ in seafood appeared to be substantial, accounting for $71 \pm 26\%$ (average $\pm$ standard deviation). Thus, coPCBs need to be included to assess human exposure of dioxin-like compounds.

Average Daily Intake (ADI) via dietary exposure to dioxin-like compounds was estimated by multiplying residue concentrations and

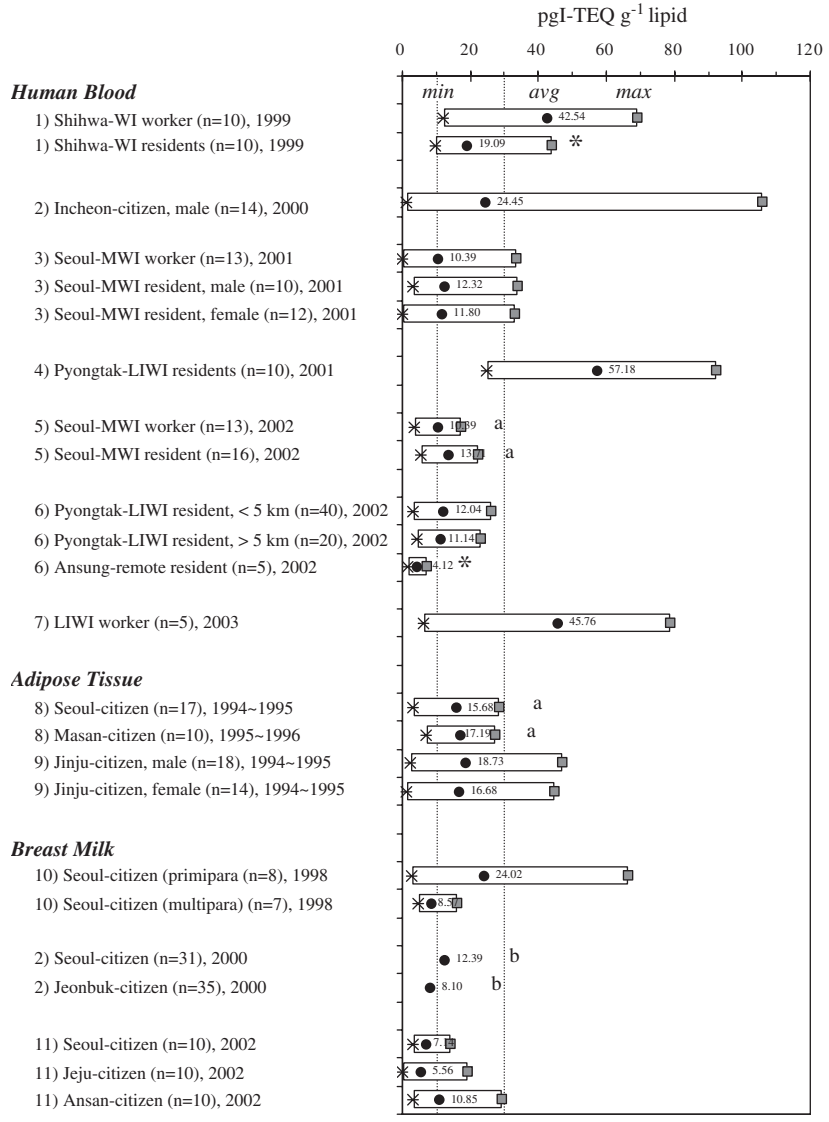

Fig. 2.14. PCDDs/DFs residues in Korean human body. The upper- and down-side of bars indicates minimum and maximum, respectively, and black circles in the center of bars indicate arithmetic means. Exceptionally, the upper- and down-side in 'a' bars indicate 'arithmetic mean $\pm$ 1 standard deviation' and the arithmetic means alone were presented for 'b'. '*' means the significant difference PCDDs/DFs residues in between residents around waste incinerator and incinerator workers (paired t-test, $p<0.01$). Two dotted lines indicate average human background level suggested by WHO. (1) Yang et al. (2001), (2) Kim et al. (2000b), (3) Kim et al. (2005e), (4) CIES (2002), (5) Leem et al. (2003), (6) Pyoungtak City, 2003, (7) Kim et al. (2005d), (8) Kang et al. (2002), (9) Kang et al. (1997), (10) Yang et al. (2002), (11) Kang et al. (2003).

Table 2.8. Residual levels of dioxin-like compounds in various food-items from South Korea since 1999

Food items	Sample number (*n*)	PCDDs/DFs (pg WHO-TEQ g^{-1}, ww)	coPCBs (pg WHO-TEQ g^{-1}, ww)	Total TEQs (pg WHO-TEQ g^{-1}, ww)	References[a]
Cereals	74	ND–0.358[b] (0.0123, 0.0005)[c]	ND–0.005 (0.0005, 0)	ND–0.358 (0.0141, 0.0025)	(4)–(6), (9)–(11)
Vegetables	23	ND–0.046 (0.0037, 0.0010)	ND–0.004 (0.0007, 0)	ND–0.006 (0.0020, 0.0010)	(6), (9)
Fruits	15	ND–0.008 (0.0042, 0.0048)	ND–0.002 (0.0004, 0)	ND–0.008 (0.0045, 0.0048)	(6), (9)
Meat/poultry	147	ND–0.454 (0.0375, 0.0060)	ND–1.065 (0.0325, 0.0035)	ND–1.519 (0.0696, 0.0155)	(2)–(11)
Fish/shellfish	276	ND–7.235 (0.2300, 0.0350)	ND–19.037 (0.7888, 0.2199)	ND–19.441 (0.9757, 0.2902)	(1)–(6), (8)–(12)
Dairy products	55	ND–0.080 (0.0142, 0.0050)	ND–0.195 (0.0280, 0.0070)	ND–0.239 (0.0351, 0.0120)	(2), (4)–(6), (8)–(11)

[a](1) Oh et al. (2005a), (2) Choi et al. (2002), (3) Kim et al. (2001e), (4) Chae et al. (2004), (5) Kim et al. (2004b), (6) Kim et al. (1999a), (7) Choi et al. (2001b), (8) Won et al. (2000), (9) Won et al. (2001), (10) Hong et al. (2002), (11) Hong et al. (2003a), (12) Moon and Ok (2006).
[b]Range (minimum–maximum).
[c]Average value (arithmetic mean, median).

Table 2.9. Average daily intakes (ADIs) investigated for market food-items

Food group assessed	Collected year	Percentage of food-items assessed (%)[a]		ADI (pg WHO-TEQ kg(bw)$^{-1}$ day^{-1})			References
		Of total	Of fish/shellfish	PCDDs/DFs	coPCBs	Total TEQs	
31 Items in 6 food-groups (cereals, vegetables, fruits, meat/poultry, dairy products, fishes/shellfishes)	1999	72.7	26.1	0.63[b]	–[c]	–[c]	Kim et al., 2000c
13 Items in 3 food-groups (cereals, meat/poultry, fishes/shellfishes)	1999[d]	28	20	0.26	0.39	0.64	Won et al., 2000
23 Items in 6 food-groups (cereals, vegetables, fruits, meat/poultry, dairy products, fishes/shellfishes)	2000[d]	52	23.4	0.08	0.23	0.31	Won et al., 2001
23 Items in 3 food-groups (meat/poultry, dairy products, fishes/shellfishes)	2001[d]	34	37.3	0.14	0.78	0.92	Hong et al., 2002
17 Items in 4 food-groups (cereals, meat/poultry, dairy products, fishes/shellfishes)	2002[d]	30	15.3	0.06	0.13	0.19	Hong et al., 2003a

40 Fishes/shellfishes	2002–2003	3.6	54.1	0.28	0.4	0.68	Moon & Ok, 2006
32 fishes/shellfishes	2003	5.2	76.7	0.26	0.67	0.93	Lee et al., 2007
14 Items in 4 imported food-groups (cereals, meat/poultry, dairy products, fishes/shellfishes)	2003[d]	25.3	10	0.04	–[c]	–[c]	Chae et al., 2004
17 Items in 4 food-groups (cereals, meat/poultry, dairy products, fishes/shellfishes)	2003[d]	29	20.2	0.12	0.29	0.41	Kim et al., 2004b
Average food-items (cereals, vegetables, fruits, meat/poultry, dairy products, fishes/shellfishes)	1999–2003	82.1	100	0.08–10.95 (0.42)	0.26–22.6 (0.96)	0.39–25.7 (1.32)	This study[e]

[a]Average daily consumption rate of total food and fishes/shellfishes are 1280 and 86 g capita^{-1} day^{-1}.
[b]pg I-TEQ kg(bw)$^{-1}$ day^{-1}.
[c]Not analyzed.
[d]Estimated year from 'published date minus one year' because of not presented.
[e]Minimum–maximum (arithmetic mean) which were estimated by assuming as representative residues of individual food groups median, maximum, and arithmetic mean, respectively, of residual levels in individual food groups presented in Table 2.7.

consumption rate of individual food items. The estimated ADI for South Korean people since 1999 ranged from 0.19 to 0.93 pg WHO-TEQ kg bw^{-1} day^{-1} (Table 2.9). There existed great uncertainties in these estimates because the assessments did not include all food items that South Koreans consume daily or did not represent average composition of individual items in food consumption. Most studies involved ~30% (~400 g $capita^{-1}$ day^{-1}) of average daily food consumption rate, ca. 1280 g $capita^{-1}$ day^{-1}. Fishes/shellfishes, containing the greatest concentrations of dioxin-like compounds among food-groups, were assessed for 20% (~17 g $capita^{-1}$ day^{-1}) of average daily consumption rate, ca. 86 g $capita^{-1}$ day^{-1}, in most studies (Table 2.9). Thus, true ADI is likely to be greater than previous estimates. For instance, Lee et al. (2007) estimated that ADI was 0.93 pg WHO-TEQ $kg(bw)^{-1}$ day^{-1} from fishes/shellfishes alone in which 77% of daily fishes/shellfishes consumption rate was included. This corresponds to 1.2 pg WHO-TEQ $kg(bw)^{-1}$ day^{-1} for all food items. We also estimated ADI using median, maximum, and arithmetic mean values observed for individual food groups. Assuming that a South Korean is exposed to the observed maximum levels for all individual food groups, ADI would be 25.7 pg WHO-TEQ $kg(bw)^{-1}$ day^{-1}. Although this exposure rate seems unrealistically great since it might represent a worst-case scenario. If it is assumed that individuals are exposed to the arithmetic mean concentration of each residue for all individual food groups, the ADI is estimated to be 1.32 pg WHO-TEQ $kg(bw)^{-1}$ day^{-1}. Since values less than the limit of quantification were included in the database as 'zero', ADI calculated using median residues would underestimate the actual ADI.

Fishes/shellfishes contributed 82% of the ADI, followed by cereals (6.2%), meat/poultry (6.1%), dairy products (3.9%), fruits (1.1%), and vegetables (0.7%). Again, coPCBs accounted for 70% of the total TEQs, implying the significant contribution by seafood. A similar contribution from fishes/shellfishes (>70% of total TEQs) and coPCBs (>60% of total TEQs) was also observed in ADI assessments of the Japanese population (Tsutsumi et al., 2001; Sasamoto et al., 2006). The contribution of seafood to total TEQs worldwide varies from a value of less than 6% for the USA (Charnley and Doull, 2005) and Egypt (Loutfy et al., 2006) to more than 70% for South Korea, Finland (Kiviranta et al., 2004), and Japan. Major contributors are determined depending on specific consumption style in individual nations. For instance, meats and dairy products are a major contributor in the diet of people in the USA and Egypt. In South Korea, it is notable that fishes/shellfishes, account for only 6.7% of daily food ingestion, but account for about 80% of total TEQs. Dried anchovy alone accounted for 0.62 pg WHO-TEQ $capita^{-1}$ day^{-1}

(Lee et al., 2007). To reduce exposure to dioxin-like compounds, therefore a change in food consumption pattern may be helpful, together with control of environmental release. In some previous studies (Kim et al., 2000b; Yang et al., 2001; Lee, 2002), the role of fishes/shellfishes was underestimated as 37–55% of total TEQs. This is because the co-planar PCBs were excluded from the analysis and many kinds of fishes/shellfishes were not included from their estimation.

Besides dietary exposure, the daily dioxins intake rate can be also predicted on the basis of its level in breast milk, blood, and adipose tissue as shown in Eq. (1) (WHO, 1998; US EPA, 2000).

$$\mathrm{EDI} = \frac{C_{\mathrm{tissue}} \times 0.693 \times f_2}{h_{12} \times f_1} \tag{1}$$

In this calculation the EDI is an average daily intake estimated (pg TEQ kg(bw)$^{-1}$ day^{-1}), $h_{1/2}$ is the half-life of dioxins in human body (day), f_1 is the fraction of dioxins absorbed (dimensionless), and f_2 is the proportion of body fat to weight (dimensionless). These constants were assumed to be 7 yrs, 0.9, and 0.3 for $h_{1/2}$, f_1, and f_2, respectively. We estimated EDIs from body residues presented in Fig. 2.14. These EDIs for the general population except incinerator workers were less than 4 pg WHO-TEQ kg(bw)$^{-1}$ day^{-1} (Fig. 2.15). In particular, average EDIs for individuals of general population ranged from 0.37 pg TEQ kg(bw)$^{-1}$ day^{-1} to 2.21 pg TEQ kg(bw)$^{-1}$ day^{-1} with a mean of 1.21 pg TEQ kg(bw)$^{-1}$ day^{-1}. These EDIs were predicted using PCDDs/DFs residues alone and I-TEQ basis. Considering that coPCB can contribute as much as 70% of the concentration of PCDDs/DFs observed in South Korean human tissues, average EDI is expected to be about 2 pg WHO-TEQ kg(bw)$^{-1}$ day^{-1}. This is greater than the average ADI (i.e., 1.32 pg WHO-TEQ kg(bw)$^{-1}$ day^{-1}) of dietary intake that had been estimated previously. However, if longer half-lives ($h_{1/2}$) are applied in the Eq. (1), the average EDI including coPCBs will would be less than 2 pg WHO-TEQ kg(bw)$^{-1}$ day^{-1}. Half-life of the TCDD (ca., 7 yr) was used in the present study but those of other congeners are generally 2- to 100-fold longer than TCDD (Geyer et al., 2002). Based on the ADIs and EDIs estimated here, the general South Korean population with an average body weight of 60 kg is expected to ingest dioxin-like compounds of between 60 and 120 pg WHO-TEQ per day which is equivalent to the WHO limit of 1–2 pg WHO-TEQ kg(bw)$^{-1}$ day^{-1}. This exposure rate is within the range of other countries (Fig. 2.15).

Although the contribution of coPCBs to the ADI was greater than of that of PCDDs/DFs, concentrations in humans were relatively small. The likely reasons for this include relatively rapid excretion of coPCBs and

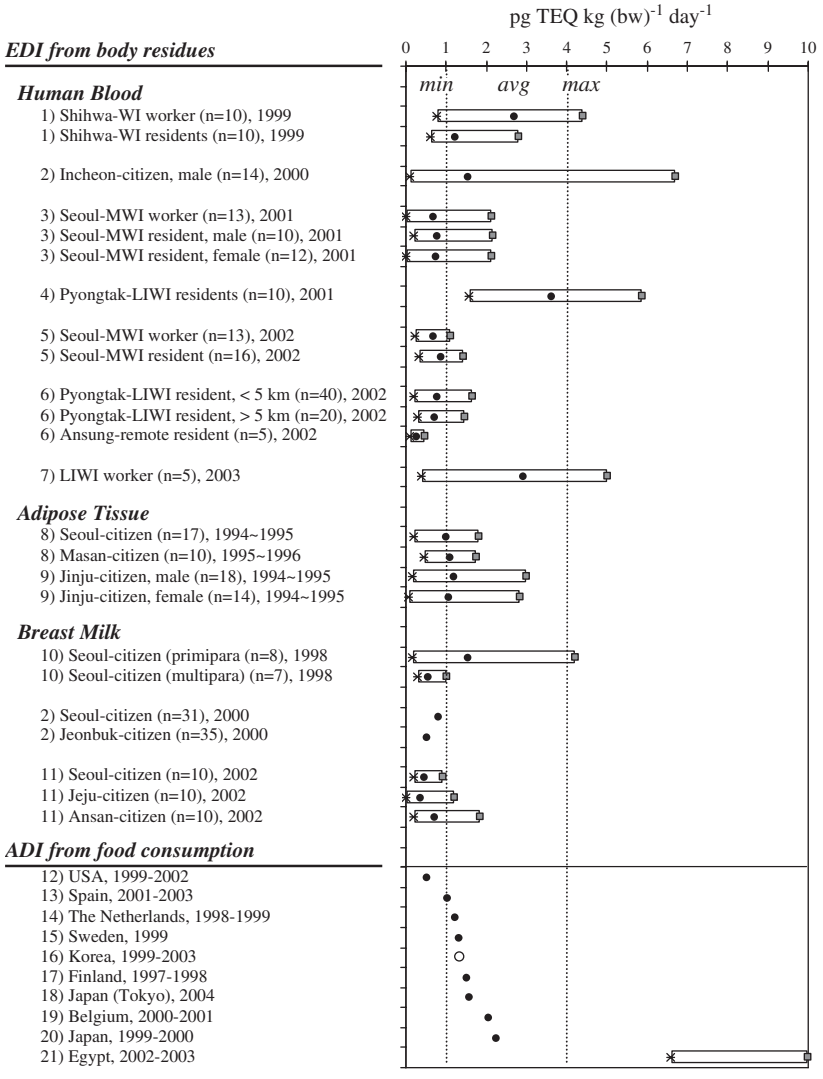

Fig. 2.15. EDI (Estimated Daily Intake) predicted from human body residues in Fig. 2.14 and ADI (Average Daily Intake) reported worldwide. ADI for Korean was predicted from average residues of individual food-groups in Table 2.7: (1)–(11) in Fig. 2.14, (12) Charnley and Doull (2005), (13) Bocio and Domingo (2005), (14) Baars et al. (2004), (15) Darnerud et al. (2006), (16) this study, (17) Kiviranta et al. (2004), (18) Sasamoto et al. (2006), (19) Focant et al. (2002), (20) Tsutsumi et al. (2001), (21) Loutfy et al. (2006). Two dotted lines indicate tolerable daily intake (TDI) guidelines of WHO.

change of chemical status during cooking. In 1997 the KFDA established 4 pg I-TEQ kg(bw)$^{-1}$ day^{-1} as a guideline for the TDI. In 2005, the committee for dioxin risk assessment, joined by five governmental ministries (Ministry of Environment, Ministry of Agriculture and Forestry, Ministry of Maritime Affairs and Fishery, Rural Development Administration, and South Korea Food and Drug Administration), set a plan to establish EQGs for individual environmental compartment and a new TDI on the basis of domestic contamination and exposure levels. Through this plan, uncertainties in exposure assessment are expected to decrease.

2.5.2. PCBs

Among the POPs reviewed, the PCBs were found to be one of the most intensively studied compounds following PCDDs/DFs. PCBs are one of the classical POPs and have been detected in various environmental media for over the last 30 years worldwide. In South Korea, despite the ban in 1970s on the use of PCBs, these compounds were detected in various environmental samples including air, sediment, and aquatic organisms (Table 2.10). In the earlier stages (viz. mid-1990s) of POPs study, efforts had been focused on the coastal contamination (mainly coastal sediments) (Khim et al., 1999b; Lee et al., 2001a). However, later studies have monitored air, water, freshwater-, brackish-, marine-sediments, and aquatic organisms.

2.5.2.1. Environmental levels

Compared to sediment and aquatic organisms, little is known about concentrations of PCBs in the atmosphere of South Korea. Concentrations of total PCBs (gas + particle phase) in the air of Ansung City ranged from 0.02 to 0.26 ng m^{-3}, with a mean of 0.07 ng m^{-3}. Gas phase PCBs accounted for approximately 90% of the total concentration of PCBs, showing that atmospheric PCBs predominantly existed in the gas phase.

As for sediment, various sites from the inland rivers, lakes, and bays have been investigated since the mid-1990s (Table 2.10). Total PCBs concentrations measured in freshwater sediments ($n = 64$) ranged from less than 1.0 to 141 ng g^{-1} dw, with a mean range of between 10.5 and 38.5 ng g^{-1} dw for individual regions. Brackish sediment collected from Lake Shihwa contained detectable concentration of PCBs in the survey of 1996 (mean = 12.5 ng g^{-1}, dw; $n = 4$) and 1998 (mean = 4.37 ng g^{-1}, dw; $n = 11$). Lake Shihwa is an artificial lake created by the construction of 12.7 km of sea dike in 1994. In contrast to the limited reports on PCBs in

Table 2.10. Concentrations of total PCBs in the various environmental media in South Korea

Sampling				No. of isomers	Total PCBs			References
Samples	From	Year	*n*		Min	Max	Mean	
Air						ng m^{-3}		
Gas/particles								
Inland (gas)	Ansung (rural)	2001–2002	28	24	0.02	0.23	0.06	Yeo et al., 2003
Inland (particle)	Ansung (rural)	2001–2002	28	24	0.0004	0.03	0.01	Yeo et al., 2003
Soil						ng g^{-1}, dw		
Urban soil	Seoul	2001	6	20	6.5	12.4	9.8	Kim, 2004
Water						ng L^{-1}		
Freshwater	Han River, Seoul	2001–2003	15	105	0.427	3.27	1.353	Kim, 2004
	Wastewater, Incheon	1998	1	105	–	–	27.18	Kim et al., 2000a
	Saltwater, Incheon	1998	3	105	0.89	11.11	6.16	Kim et al., 2000a
Sediment						ng g^{-1}, dw		
Freshwater sediment								
Westward	2 Rivers, Shihwa	2000	8	98	9.00	122	30.3	Koh et al., 2005
	1 River, Han river	2001–2002	25	104	1.76	68	17.3	Kim, 2004
Southward	3 Rivers, Masan	2000	8	98	8.49	92.0	38.5	Koh et al., 2005
	1 River, Busan	1999	18	43	1.10	141	18.7	Jeong et al., 2001d
Eastward	2 Rivers, Onsan	1999	8	98	1.00	56.2	15.2	Koh et al., 2002
	3 Rivers, Ulsan	1999	14	98	0.56	52.2	10.5	Khim et al., 2001
	1 River, Pohang	2000	8	98	1.00	141	31.7	Koh et al., 2004
Brackish sediment								
West coast	Shihwa lake	1996	4	101	2.71	27.9	12.5	Lee et al., 2001a
	Shihwa lake	1998	11	98	1.00	12.3	4.37	Khim et al., 1999a

Marine sediment								
West coast	Incheon Harbor	1995	7	101	10.7	580	146	Lee et al., 2001a
	Incheon Harbor	1998	21	95	0.35	1093	133	Kim et al., 2000a
	Gyeonggi Bay	1995	47	101	0.39	10.2	3.12	Lee et al., 2001a
	Namyang Bay	1996	5	101	0.99	2.45	1.45	Lee et al., 2001a
South coast	Masan Bay	1997	20	22	1.24	41.4	15.0	Hong et al., 2003b
	Masan Bay	1998	28	98	6.94	148	36.0	Khim et al., 1999b
	Gwangyang Bay	2001	11	98	1.00	4.53	1.32	Koh et al., 2005
East coast	Onsan Bay	1999	14	98	1.00	13.0	3.31	Koh et al., 2002
	Ulsan Bay	1999	16	98	1.00	76.7	17.6	Khim et al., 2001
	Yeongil Bay	2000	26	98	1.00	32.3	3.93	Koh et al., 2006
Korean coasts	3 Coastal bays	1996–1998	6	134	10.0	140	40.8	Oh et al., 2003
	27 Coastal areas	2000	138	22	0.09	199	9.17	Hong et al., 2006
Open sea	Yellow Sea	2000	23	21	0.17	1.37	0.69	Oh et al., 2005b
Organism					ng g^{-1}, dw			
Freshwater fish								
Korean coasts	10 Rivers	2002	20	28	0.37	20.0	5.84	Moon et al., 2006
Saltwater fish								
Korean coasts	19 Coastal areas	1997	33	22	31.0	630	85.2	Yim et al., 2005
Bivalvia								
West coast	Incheon harbor	1998	12	95	49.0	422	228	Kim et al., 2002e
	10 Coastal areas	1990–1999	12	134	7.78	394	56.5	Oh et al., 2003
	17 Coastal areas	1997–1999	34	104	4.40	422	94.1	Kim et al., 2002b
South coast	Gwangyang bay	1998	9	98	32.3	162	72.7	Khim et al., 2000
	Masan bay	1998	9	98	64.2	313	134	Khim et al., 2000
	Busan bay	1998	5	98	63.6	407	185	Khim et al., 2000
	7 Coastal areas	1987–1999	21	134	2.83	389	93.9	Oh et al., 2003
	10 Coastal areas	1999	19	104	7.30	184	67.5	Kim et al., 2002b
East coast	Ulsan bay	1998	11	98	60.6	547	176	Khim et al., 2000
	6 Coastal areas	1999	13	104	5.30	170	54.2	Kim et al., 2002b
	10 Coastal areas	1990–1999	24	134	3.67	456	79.1	Oh et al., 2003

freshwater ($n = 64$) and brackish sediments ($n = 15$), a relatively large number of samples of coastal marine sediment ($n = 362$) have been analyzed. The maximum concentration of total PCBs in marine sediment was detected in Incheon Harbor, which had a concentration of 1090 ng g^{-1} dw in 1998 (Kim et al., 2000a). A recent study by Hong et al. (2006) reported total PCBs concentrations at 138 sites along the South Korean coasts to range from 0.09 to 199 ng g^{-1} dw (mean = 9.17). Overall, mean concentrations of total PCBs in coastal marine sediments in South Korea ranged from several to several tens of ng g^{-1} dw. Considering the detectable concentrations of total PCBs (0.17–1.37 ng g^{-1} dw) at the offshore sites in the Yellow Sea, widespread contamination is evident even in open seas.

Several studies have reported concentrations of PCBs in aquatic organisms such as freshwater fish ($n = 20$), saltwater fish ($n = 33$), and bivalves ($n = 169$) (Table 2.10). Total PCBs concentrations in aquatic organisms were generally within the range of those in sediments, with a mean of 5.84–228 ng g^{-1} dw. Among the biota samples analyzed from various sites, Incheon Harbor contained the greatest total PCBs (228ng g^{-1} dw), where a highest level of PCBs was also detected in the sediment in 1998 (Kim et al., 2000a). Overall, total PCBs concentrations in aquatic organisms in South Korea were comparable to or greater than those (mostly several ng g^{-1} dw levels) reported for developing countries in Asia (Monirith et al., 2003).

2.5.2.2. Sources and distribution

The homologue pattern of PCBs found in sediment from the various sites in South Korea suggests the source is technical mixtures such as Aroclors 1016, 1242, 1254, and 1260 or corresponding Kanechlor products. Lesser chlorinated CBs such as tri-, tetra-, and penta-CBs were the major PCB congeners found in freshwater sediments ($n = 64$) accounting for about 21%, 24%, and 20% of total PCBs, on average, respectively (Fig. 2.16). The predominant CBs shifted to more chlorinated congeners in marine sediments ($n = 195$); 27% penta-, 22% tetra- and 20% hexa-CBs (Fig. 2.16). This indicates that different sources could influence freshwater and marine environments. Based on a study of sources of PCBs, that the South Korean environment is contaminated by mixtures of products with various chlorine contents dominated by Aroclor 1254 in Incheon, Aroclor 1242 in Masan, Aroclor 1260 (or Kanechlor-600) in Ulsan, and Aroclor 1242 in the Han River (Kim et al., 2000a, 2002b; Kim, 2004; Hong et al., 2006). Thus, the type of PCBs that contaminated the marine environments is likely to depend on specific activities in the individual

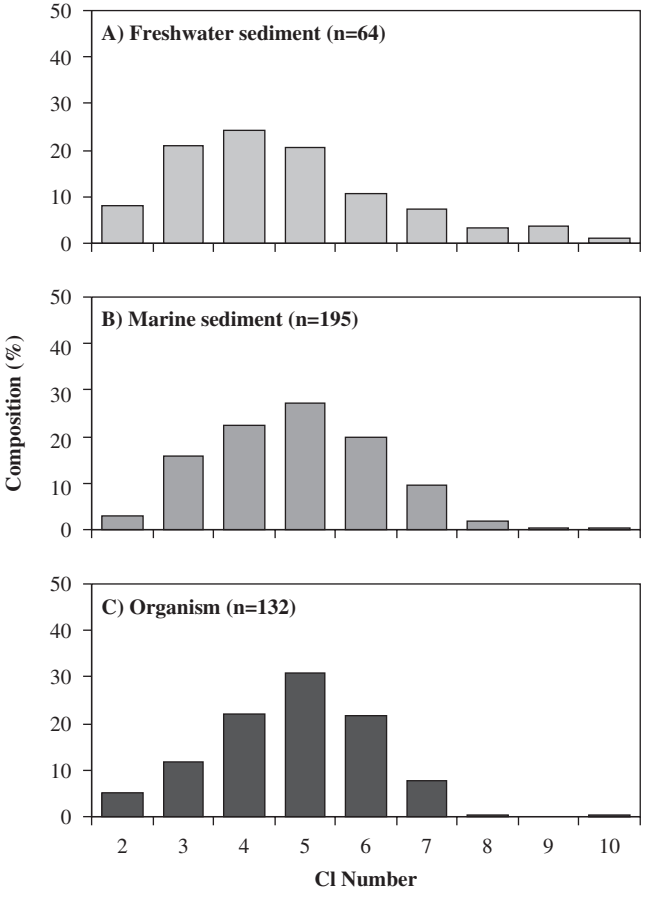

Fig. 2.16. Homolog composition (mean%) of polychlorinated biphenyls (PCBs) in (A) freshwater sediment ($n = 64$), (B) marine sediment ($n = 195$), and (C) organism ($n = 132$), from South Korea.

coastal area. CBs homologue of biota samples showed very strong correlation ($r^2 = 0.96$, $p < 0.05$) with that of sediment, reflecting bioaccumulation of sedimentary PCBs into aquatic organisms (Fig. 2.16).

Among 21 published reports of concentrations of PCBs in sediments (Table 2.10), Masan inland (mean = 38.5 ng g^{-1} dw, $n = 8$, 2000) and Incheon Harbor (mean = 146 ng g^{-1} dw, $n = 7$, 1995) were identified as the most contaminated areas for freshwater and marine sediments, respectively. PCBs in freshwater sediments from six areas exceeded ERL for total PCBs (22.7 ng g^{-1}, dw). However, the concentrations of sediment

PCBs from any of the stations ($n = 65$) did not exceed the ERM (180 ng g^{-1}, dw). PCBs were reported to be mostly less than ERL for total PCBs in the brackish sediments from the Shihwa lake in 1996 ($n = 4$) and 1998 ($n = 11$). PCBs in marine sediments reported from the South Korean coasts showed relatively great variations in terms of concentration and congener distribution (Fig. 2.17). Along the South Korean coasts, several hot spots were found in which concentrations were greater than the ERM for total PCBs. This was particularly true in Incheon Harbor, near the capital city of Seoul. However, the contamination level in several bays including the Onsan, Gyeonggi, Gwangyang, Namyang bays, and Yellow Sea did not contained concentrations that exceeded ERL for total PCBs. Overall, the concentrations of sedimentary PCBs in the inland rivers and bay areas were comparable. Their spatial distribution indicated the presence of the localized zones of extremely great concentrations (hot spots).

2.5.3. OC pesticides

During the past several decades, contamination of the environment by OCPs has been of great concern worldwide, due to their persistent, ubiquitous, and biologically harmful properties. Although their production and use had mostly been banned since the 1970s (Fig. 2.9), OCPs are still found in various environmental samples and widely distributed along the coasts of South Korea (Table 2.11). Studies investigating the environmental concentrations and distribution of OCPs have increased in the last 10 years, for both inland areas and coastal environment. In these studies, the environmental concentrations of OCPs (HCB, HCHs, CHLs, and DDTs) were reported for various environmental samples including air ($n = 36$), soil ($n = 10$), sediment ($n = 397$), and aquatic organism ($n = 133$) in South Korea.

2.5.3.1. Environmental concentrations

Little is known about concentrations of OCPs in the atmosphere over South Korea. Two studies reported concentrations of HCHs, CHLs, and DDTs in air over the cities of Seoul ($n = 19$) and Ansung ($n = 17$) in 1990–2000 (Yeo et al., 2004b). The mean concentration of total OCPs in air from Ansung (2.00 ng m^{-3}) was approximately three times greater than that in Seoul (0.71 ng m^{-3}). Among the atmospheric OCPs, HCHs were the greatest level, accounting for $>80\%$ of the total OCPs.

Total OCPs detected in soil ($n = 10$) from Jeonbuk province ranged from 1.57 to 52.9 ng g^{-1} dw, with a mean concentration of 27.8 ng g^{-1} dw (Kim and Smith, 2001). OCPs in sediments were reported for various sites

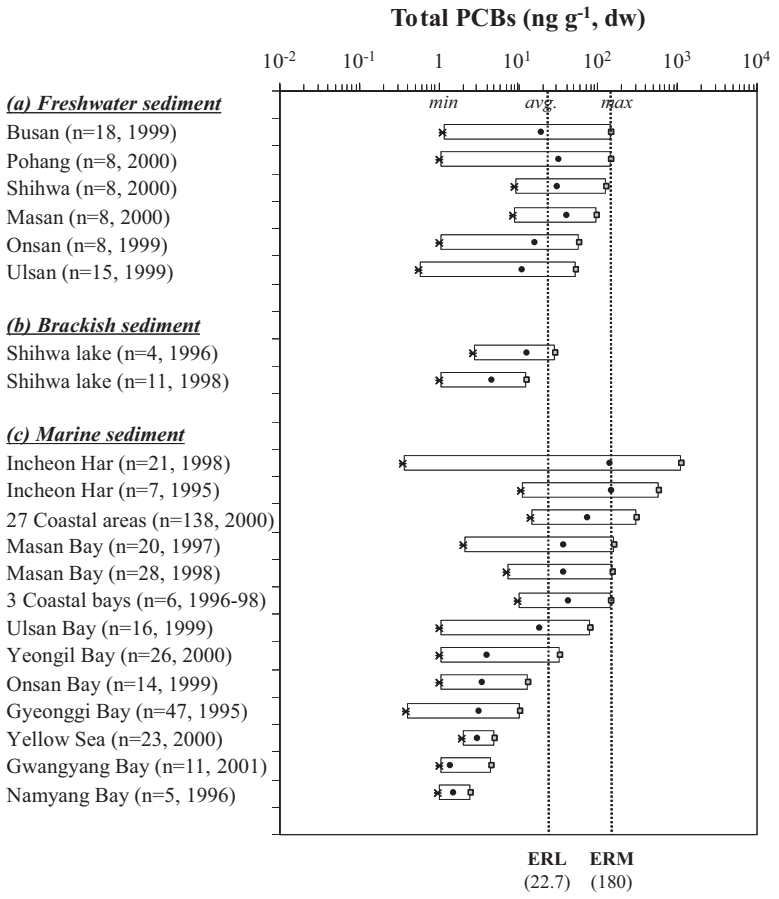

Fig. 2.17. Comparison of measured levels and sediment quality guidelines (SQGs) for total polychlorinated biphenyls (PCBs) in sediments from the various sites, in South Korea; (a) freshwater ($n = 64$), (b) brackish ($n = 15$), and (c) marine sediments ($n = 362$). Range indicates minimum (min), maximum (max), and arithmetic mean (avg.). Dotted lines represent SQGs of effect range low (ERL) and effect range median (ERM) for total PCBs (Long et al., 1995).

in inland ($n = 47$), lake ($n = 15$), and coastal bay ($n = 335$) areas since 1995 (Table 2.11). Total OCPs concentrations measured in freshwater sediments ($n = 47$) ranged from 0.04 to 47.0 ng g^{-1} dw, with a regional mean of 1.24–12.7 ng g^{-1} dw. Concentrations of OCPs such as HCB, HCHs, CHLs, and DDTs in 1996 ($n = 4$) and 1998 ($n = 11$) were measured in 15 sediment samples from Lake Shihwa. Mean concentrations of total OCPs in Lake Shihwa were comparable between 1996

Table 2.11. Concentrations of organochlorine pesticides (OCPs) in various environmental media in Korea

Sampling				HCB	HCHs	CHLs	DDTs	Total OCPs			References
Samples	From	Year	*n*	Mean				Min	Max	Mean	
Air				ng m^{-3}				ng m^{-3}			
Gas/particles											
Inland (*G*+*P*)	Seoul (urban)	1999–2000	19	–	0.27	0.01	0.04	0.05	4.14	0.71	Yeo et al., 2004b
Inland (*G*+*P*)	Ansung (rural)	1999–2000	17	–	0.22	0.04	0.06	0.04	15.7	2.00	Yeo et al., 2004b
Soil and sediment				ng g^{-1}, dw				ng g^{-1}, dw			
Soil											
Inland	Jeonbuk	1996	10	–	1.49	25.6	0.09	1.57	52.9	27.8	Kim and Smith, 2001
Freshwater sediment											
Westward	2 Rivers, Shihwa	2000	8	1.13	2.74	0.06	0.55	0.89	16.6	4.49	Koh et al., 2005
Southward	3 Rivers, Masan	2000	8	0.30	11.2	0.09	1.09	0.36	46.9	12.7	Koh et al., 2005
Eastward	2 Rivers, Onsan	1999	8	1.00	0.62	0.17	1.61	0.09	6.44	3.28	Koh et al., 2002
	3 Rivers, Ulsan	1999	15	0.23	0.28	0.17	0.21	0.04	5.81	1.24	Khim et al., 2001
	1 River, Pohang	2000	8	0.50	6.36	0.04	0.43	0.35	47.0	7.32	Koh et al., 2004
Brackish sediment											
West coast	Shihwa lake	1996	4	0.18	1.07	0.37	1.34	0.88	6.55	2.96	Lee et al., 2001a
	Shihwa lake	1998	11	1.31	0.18	0.02	1.28	1.04	5.47	2.80	Khim et al., 1999a

Marine sediment											
West coast	Incheon harbor	1995	7	1.52	0.67	29.6	12.4	3.68	167	44.2	Lee et al., 2001b
	Gyeonggi bay	1995	47	0.05	0.44	0.25	0.82	0.10	6.75	1.56	Lee et al., 2001b
	Namyang bay	1996	5	0.02	0.46	0.25	0.34	0.14	1.72	1.07	Lee et al., 2001b
South coast	Gwangyang bay	2001	11	0.08	0.25	0.01	0.07	0.10	0.99	0.42	Koh et al., 2005
	Masan bay	1997	20	0.13	0.26	0.65	16.0	0.84	97.9	18.1	Hong et al., 2003b
	Masan bay	1998	28	0.62	0.30	0.09	3.45	1.14	14.6	4.46	Khim et al., 1999b
East coast	Onsan bay	1999	14	0.15	0.21	0.02	0.43	0.07	2.81	0.78	Koh et al., 2002
	Ulsan bay	1999	16	0.05	0.35	0.08	2.99	0.14	20.2	4.21	Khim et al., 2001
	Yeongil bay	2000	26	0.03	1.07	0.08	0.52	0.05	13.8	1.70	Koh et al., 2006
Korean coasts	27 Coastal areas	2000	138	0.14	0.65	0.37	6.64	0.06	161	8.31	Hong et al., 2006
Open sea	Yellow Sea	2000	23	0.16	1.17	0.06	0.48	0.38	7.75	1.97	Oh et al., 2005
Organism				ng g^{-1}, dw				ng g^{-1}, dw			
Saltwater fish											
Korean coasts	19 Coastal areas	1997–2001	33	1.19	3.48	4.07	33.2	6.96	129	41.9	Yim et al., 2005
Bivalvia											
West coast	17 Coastal areas	1997–1999	34	–	7.09	8.49	33.0	9.59	128	48.6	Kim et al., 2002b
South coast	Yeosu	1998	9	0.46	1.80	0.31	23.5	10.0	64.3	26.0	Khim et al., 2000
	Masan	1998	9	1.44	1.99	1.69	46.0	16.7	114	51.1	Khim et al., 2000
	Pusan	1998	5	0.77	2.71	1.91	59.4	26.9	112	64.8	Khim et al., 2000
	10 Coastal areas	1999	19	–	5.29	2.52	19.1	8.81	61.8	26.9	Kim et al., 2002b
East coast	Ulsan	1998	11	3.28	2.83	1.69	23.5	12.0	101	31.4	Khim et al., 2000
	6 Coastal areas	1999	13	–	5.56	5.70	29.3	12.8	97.4	40.6	Kim et al., 2002b

(2.96 ng g^{-1} dw) and 1998 (2.80 ng g^{-1} dw), suggesting persistence of OCPs in environments. In contrast to the small number of reports on OCPs in inland and lake areas ($n = 62$), since 1995 there has been a large number of sites that have been investigated for concentrations of OCPs in sediments of bays ($n = 335$) along the South Korean coasts as well as the open sea. Concentrations of total OCPs varied greatly among locations, depending on sampling stations and/or areas ranging from 0.42 to 44.2 ng g^{-1} dw of regional mean concentrations. Total OCPs concentrations in marine sediments were comparable to those detected in freshwater and brackish sediments, however, the composition of HCB, HCHs, CHLs, and DDTs to total OCPs varied greatly among samples.

Several studies have reported OCPs concentrations in aquatic organisms such as saltwater fishes ($n = 33$) and bivalves ($n = 100$) along the South Korean coasts since 1997 (Table 2.11). Total OCPs concentrations in aquatic organisms were found to range from 26.0 to 64.8 ng g^{-1} dw, and the mean concentrations of OCPs were generally 10-fold greater than those in sediments. In sediments, DDTs represented the greatest portion (52%) of the total OCPs in sediment. Similarly, in aquatic organisms the DDTs were the prevalent measured. Overall, total OCPs detected in aquatic organisms in South Korea were within the range of OCPs reported from the developing countries in Asia (Monirith et al., 2003).

2.5.3.2. Sources and distribution

The contribution of individual OCPs to the total OCPs varied among environmental media (air, soil, sediment, and organism) (Fig. 2.18). The relative abundance of OCPs in air samples was found to be in the order of endosulfan (77%) > HCHs (18%) > DDTs (3.7%) > CHLs (1.6%). The large abundance of endosulfan in air samples could be explained by its usage, duration used in the past and chemical properties (Yeo et al., 2004b). The composition of OCPs in soil, freshwater, and coastal marine sediment samples were predominated by CHLs (92%), HCHs (72%), and DDTs (> 50%), respectively. The OCPs composition of soil was significantly different from that of sediment. The disparity indicates that OCPs contamination in marine environments is likely to be contaminated by potential coastal sources themselves rather than by transport of contaminants from inland sources. It should be noted that the relative composition of OCPs in aquatic organisms was similar to that found in marine sediment, which indicates the accumulation of contaminants in the body of organisms and sediment from water column via similar mechanisms (Fig. 2.18).

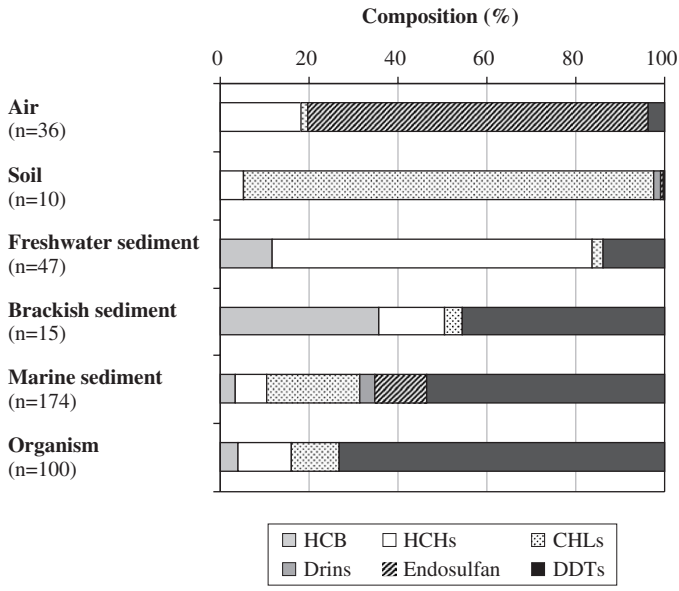

Fig. 2.18. Composition (mean%) of individual organochlorine pesticides (OCPs; HCB, HCHs, CHLs, Drins, Endosulfan, and DDTs) relative to total OCPs, detected in the various environmental media (air, soil, freshwater, brackish, marine sediments, and organism; no. of samples are in parenthesis) from the South Korea.

Inland soil collected from Jeonbuk province showed the OCPs concentrations as great as 52.9 ng g^{-1}, with a mean of 27.8 ng g^{-1}. The mean concentration was significantly greater than that in sediment (6.76 ng g^{-1}). Among 18 datasets concentrations of OCPs in sediments of South Korea (Table 2.11), Masan inland (mean = 12.7 ng g^{-1} dw, $n = 8$, 2000) and Incheon Harbor (mean = 44.2 ng g^{-1} dw, $n = 7$, 1995) were identified as the most contaminated areas with total OCPs for freshwater and marine sediments, respectively. These areas were found to be the most contaminated areas by PCBs as well, indicating a close relationship between PCBs and OCPs sources in Masan and Incheon areas. A correlation of the spatial distribution was frequently observed between the concentrations of PCBs and major OCPs such as DDTs and CHLs (Kim et al., 2002b; Hong et al., 2006). Intensive industrial and municipal activities near the cities of Masan and Incheon would cause the level of contamination by classical POPs such as PCBs and OCPs. Relatively low OCPs contamination has been found in the areas of Gwangyang (steel manufacturing, mean = 0.42 ng g^{-1}, $n = 11$, 2001) and Onsan Bay (non-ferrous metal manufacturing, mean = 0.78 ng g^{-1}, $n = 14$, 1999). The degree

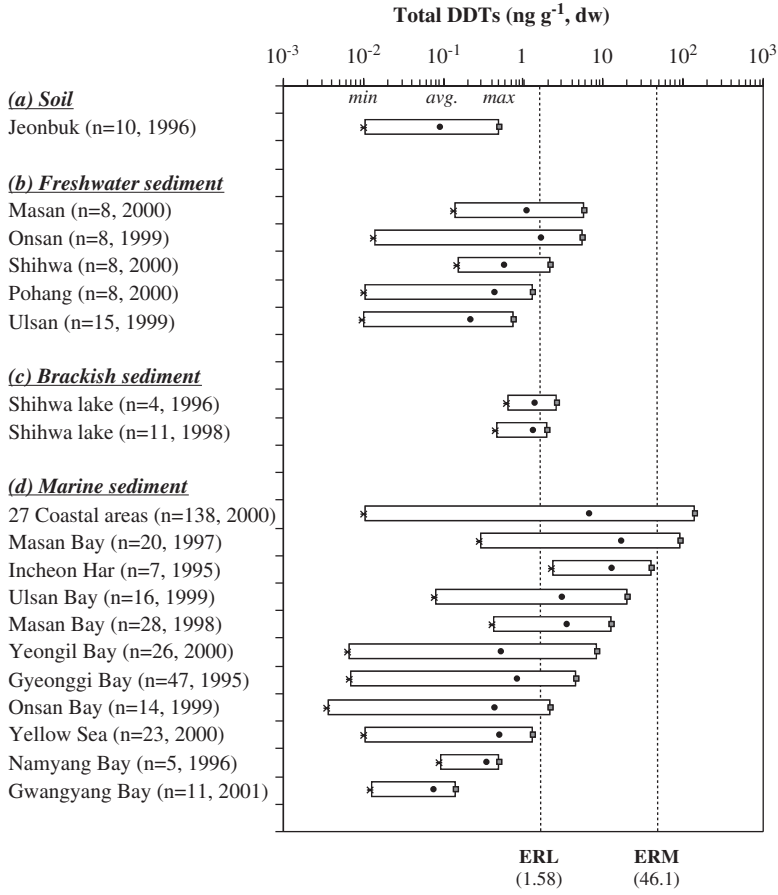

Fig. 2.19. Comparison of measured levels and sediment quality guidelines (SQGs) for total DDTs in soil and sediments from the various sites, in South Korea; (a) soil ($n = 10$), (b) freshwater ($n = 47$), (c) brackish ($n = 15$), and (d) marine sediments ($n = 335$). Range indicates minimum (min), maximum (max), and arithmetic mean (avg.). Dotted lines represent SQGs of effect range low (ERL) and effect range median (ERM) for total DDTs (Long et al., 1995).

of contamination of the South Korean coastal environment showed an existence of the clear but localized sources and distribution.

Among the OCPs, DDTs, CHLs, and γ-HCH (lindane) were found to be the most contaminated OCPs present in Korean sediments (Fig. 2.19). Out of 18 datasets examined ($n = 407$), 13, 10, and 9 of the total sites contained the concentration that exceeded the corresponding threshold effect concentrations such as ERL or TEL for total DDTs, total CHLs,

and γ-HCH, respectively. DDTs were found to be widespread OCPs contaminants across the inland and coastal-marine areas, accounting for $>50\%$ of the total OCPs both in sediment and aquatic organisms. Inland soils and sediments contained relatively low total DDTs over ERL (1.58 ng g^{-1}) at some stations but mostly below ERM (46.1 ng g^{-1}). Contrarily, in most coastal bay areas (8 out of 11 areas), the contamination level exceeded the ERL for total DDTs and even the ERM was exceeded at some sites. In spite of lower level than DDTs, CHLs showed the similar distribution spatially with DDTs. By contrast, γ-HCH was the most contaminated OCPs in the inland areas and distributed relatively evenly in marine areas. None of the site exceeded the SQGs of low apparent effects threshold (22 ng/g, dw) for HCB. Overall, the concentration and distribution of OCPs in Korean sediments reflected the widespread distribution with some localized hot spots of considerably great concentrations.

2.5.4. PAHs

Many of PAHs are known to be mutagenic and/or carcinogenic (Neff, 1979). Coke production, combustion of fossil fuels, chemical production, and oil spills are the potential sources of PAHs. In recent studies on PAHs, these compounds were found to be one of the prevalent organic contaminants in South Korean sediments (Kim et al., 1999b; Koh et al., 2002). However, compared to the reports on the classical POPs such as PCBs and OCPs, little is known about the degree of PAHs contamination in the South Korean environment, except for soil and sediments (Table 2.12). A total of 28 datasets reporting total concentrations of PAHs in South Korean samples, by sampling area, were found in the literature where total PAHs in air ($n = 70$), soil ($n = 232$), water ($n = 25$), and sediment ($n = 281$) were reported.

2.5.4.1. Environmental levels

A few studies reported total PAHs (gas + particle phase) in air in Seoul City during 1999–2002 and found concentrations between 9.10 and 350 ng m^{-3}, with a regional mean of 35.6–89.3 ng m^{-3} (Park et al., 2002b; Kim, 2004). Relatively great concentrations of atmospheric PAHs during winter were ascribed to increased fuel consumption, the lower mixing height and low air temperature.

A comprehensive study on total PAHs in soil from inland areas have been reported by Nam et al. (2003) through a survey of 226 sites in South Korea. Mean concentrations of total PAHs in soil from 6-areas were

Table 2.12. Concentrations of total PAHs detected in the various environmental media in South Korea

Sampling				No. of PAHs	Total PAHs			References
Samples	From	Year	*n*		Min	Max	Mean	
Air					ng m^{-3}			
Gas/particles								
Inland (*G*+*P*)	Seoul (urban)	1999	24	12	11.3	350	89.3	Park et al., 2002b
Inland (*G*+*P*)	Seoul (urban)	2001–02	5	24	11.4	81.0	44.3	Kim, 2004
Inland (*G*+*P*)	Seoul (suburban)	2001–02	5	24	9.10	70.3	35.6	Kim, 2004
Inland (P alone)	Seoul	1992–94	36	20	7.74	265	61.1	Panther et al., 1999
Water					ng g^{-1}, dw			
Freshwater								
Inland	Han River, Seoul	2001–03	15	15	13.44	75.73	43.03	Kim, 2004
Soil and sediment					ng g^{-1}, dw			
Soil								
Inland	Seoul (urban)	2001	3	24	791	1729	1175	Kim, 2004
	Seoul (suburban)	2001	3	24	874	1073	999	Kim, 2004
	Seoul-Gyeonggi	<2002	61	16	32.0	1057	257	Nam et al., 2003

	Gwangwon	<2002	50	16	43.0	1852	271	Nam et al., 2003
	Gyeongbuk	<2002	24	16	43.0	623	199	Nam et al., 2003
	Gyeongnam	<2002	31	16	41.0	336	171	Nam et al., 2003
	Jeolla	<2002	45	16	23.0	303	118	Nam et al., 2003
	Pohang	<2002	15	16	105	2833	578	Nam et al., 2003
Freshwater sediment								
Westward	2 Rivers, Shihwa	2000	8	16	8.61	643	224	Koh et al., 2005
	1 River, Seoul	2001–02	25	24	173	1721	858	Kim, 2004
Southward	3 Rivers, Masan	2000	8	16	16.3	481	212	Koh et al., 2005
Eastward	2 Rivers, Onsan	1999	8	16	1.00	573	128	Koh et al., 2002
	3 Rivers, Ulsan	1999	14	16	9.46	1373	265	Khim et al., 2001
	1 River, Pohang	2000	8	16	5.88	7685	1282	Koh et al., 2004
Brackish sediment								
West coast	Shihwa lake	1998	11	16	<1.00	31.4	11.4	Khim et al., 1999a
Marine sediment								
West coast	Han River mouth	1995	16	24	28.8	229	86.5	Kim et al., 1999b
	Incheon Harbor	1995	19	24	13.1	1430	272	Kim et al., 1999b
	Gyeonggi Bay	1995	31	24	9.54	167	53.1	Kim et al., 1999b
South coast	Gwangyang Bay	2001	11	16	10.5	105	36.8	Koh et al., 2005
	Masan Bay	1998	28	16	41.5	1102	353	Khim et al., 1999b
East coast	Onsan Bay	1999	12	16	1.00	50.6	21.6	Koh et al., 2002
	Ulsan Bay	1999	16	16	19.7	3095	409	Khim et al., 2001
	Yeongil Bay	2000	26	16	1.00	1872	227	Koh et al., 2006

found to be 257 (Seoul-Gyeonggi, $n = 61$), 271 (Gwangwon, $n = 50$), 199 (Gyeongbuk, $n = 24$), 171 (Gyeongnam, $n = 31$), 118 (Jeolla, $n = 45$), and 578 ng g^{-1} dw (Pohang, $n = 15$), respectively. The greatest concentration (2833 ng g^{-1} dw) of total PAHs in soil was found in the Pohang area near a large scale steel manufacturing plant. This spatial distribution is consistent with information of emission sources of PAHs described earlier that a major source of PAHs is coke oven from metal industries. The distribution of total PAHs found in soil samples were primarily of pyrogenic origins such as heavy industry emission and motor vehicle exhaust in South Korea.

Since 1995, concentrations of PAHs have been reported for various sites along the South Korean coasts. Concentrations of total PAHs measured in freshwater sediment ($n = 71$) varied greatly with sampling station, ranging from 1.00 to 7685 ng g^{-1} dw, with regional means of 128–1282 ng g^{-1} dw at the sites. Relatively small concentrations have been found in Lake Shihwa sediment with a mean of 11 ng g^{-1} dw of total PAHs. Many studies reported the sedimentary PAHs in coastal bay areas, and their total PAHs (mean = 266 ng g^{-1}, dw, $n = 226$) were similar to those measured in soil samples (mean = 255 ng g^{-1}, dw, $n = 216$). The maximum concentration (3095 ng g^{-1} dw) of total PAHs in marine sediments was found in Ulsan Bay near a shipbuilding area.

2.5.4.2. Sources and distribution

The relative contributions of the 16 individual indicator PAHs to the total concentration of PAHs in air samples significantly differed from soil or sediment samples (Fig. 2.20). Low molecular weight PAHs (L-PAHs) such as 2-3 ring aromatic hydrocarbons (NAP, ACY, ACE, FLU, PHE, and ANT) were predominant in air samples, accounting for $>50\%$ of total PAHs. This is attributable to the contribution of gaseous L-PAHs of greater volatilities. Particle-associated PAH composition is similar to those of soil and sediment. This is not only because their low volatility but dry/wet deposition of particulate PAHs plays a significant role as source to underlying soils. H-PAHs such as 4-5 ring aromatic hydrocarbons (FLT, PYR, BaA, CHR, BbK, BkF, BaP, and DahA) accounted for ca. 70% of total PAHs detected in soil and sediment samples. A significant relationship in composition of PAHs was found between soil and river- ($r^2 = 0.65$, $p < 0.05$) or marine-sediments ($r^2 = 0.71$, $p < 0.05$), indicating their sources are closely related with each other. Furthermore, the strong correlation ($r^2 = 0.97$, $p < 0.05$) between river and marine sediments

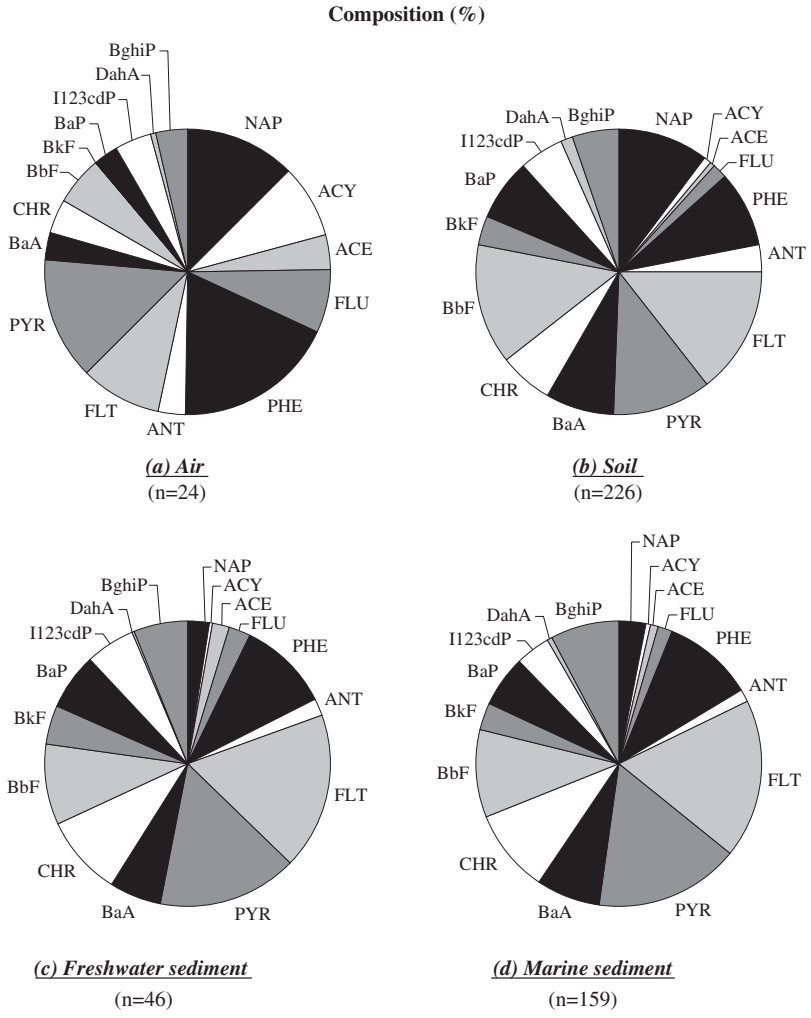

Fig. 2.20. Composition (mean%) of 16 individual polycyclic aromatic hydrocarbons (PAHs) to total PAHs detected in various environmental media; in (a) air ($n = 24$), (b) soil ($n = 226$), (c) freshwater ($n = 46$), and (d) marine sediment ($n = 159$), from the South Korea. Naphthalene; NAP, Acenaphthylene; ACY, Acenaphthene; ACE, Fluorine; FLU, Phenanthrene; PHE, Anthracene; ANT, Fluoranthene; FLT, Pyrene; PYR, Benz[*a*]anthracene; BaA, Chrysene; CHR, Benzo[*b*]fluoranthene; BbF, Benzo[*k*]fluoranthene; BkF, Benzo[*a*]pyrene; BaP, Indeno[1,2,3,*c*,*d*]pyrene; I123cdP, Dibenz[*a*,*h*]anthracene; DahA, Benzo[*g*,*h*,*i*]perylene; BghiP.

reflected their widespread distribution from similar origins across the inland and coastal bay areas.

Because PAHs can originate from pyrogenic sources (combustion) as well as petrogenic origins (petroleum), the concentrations, distribution, and fate of PAHs in environment would vary with their sources. Several molecular ratios of specific hydrocarbons such as PHE/ANT, FLU/PYR, FLU/(FLU + PYR), CHR/BaA, and L-/H-PAHs have been used to characterize sources and origins (Budzinski et al., 1997; Baumard et al., 1998; Dickhut et al., 2000; Yunker et al., 2002). Analysis of such ratios for air, soil, and sediment samples in South Korea suggested that airborne and soil PAHs were mainly pyrogenic while the sources for sediment were from both pyrogenic and petrogenic origins. It is also noteworthy that the sites near urban and industrial areas received more pyrolytic inputs whereas the bay areas or downstream of rivers close to harbor places and/or shipping yards could be influenced by more of petrogenic inputs (not presented).

The distribution of total PAHs in soil and sediment samples was dependent on the land use types (Fig. 2.21). Among the six areas investigated for total PAHs in soil from South Korea, the industrial area such as that at Pohang showed the greatest concentrations of PAHs and the west plain area in Jeolla province where agricultural activities are dominant tended to show lower concentrations of PAHs. The greater concentrations of PAHs in freshwater sediment have been found in the inland area near Pohang, further supporting the conclusion that there are direct sources of PAHs input to this area. Only in the Pohang area existed a site of contamination exceeding the ERL for total PAHs (4022 ng g^{-1} dw). In no stations of other five areas, Seoul, Ulsan, Shihwa, Onsan, and Masan did exceed the corresponding ERL. Meanwhile, total PAHs in brackish sediment measured from Lake Shihwa contained the least concentrations of PAHs in South Korea. Although the contamination of PAHs in marine sediments showed a wide spatial variability, at none of the stations exceeded the ERL for total PAHs. Overall, distribution for PAHs in soil (266 ng g^{-1}, dw, $n = 226$), freshwater (514 ng g^{-1}, dw, $n = 81$), and marine sediment (182 ng g^{-1}, dw, $n = 159$) suggested the widespread contamination of PAHs across the inland and coastal bay areas in South Korea.

2.5.5. APs and BPA

Distributions of APs and BPA in South Korea have been studied since the late 1990s. Concentrations of APs and BPA have been measured in

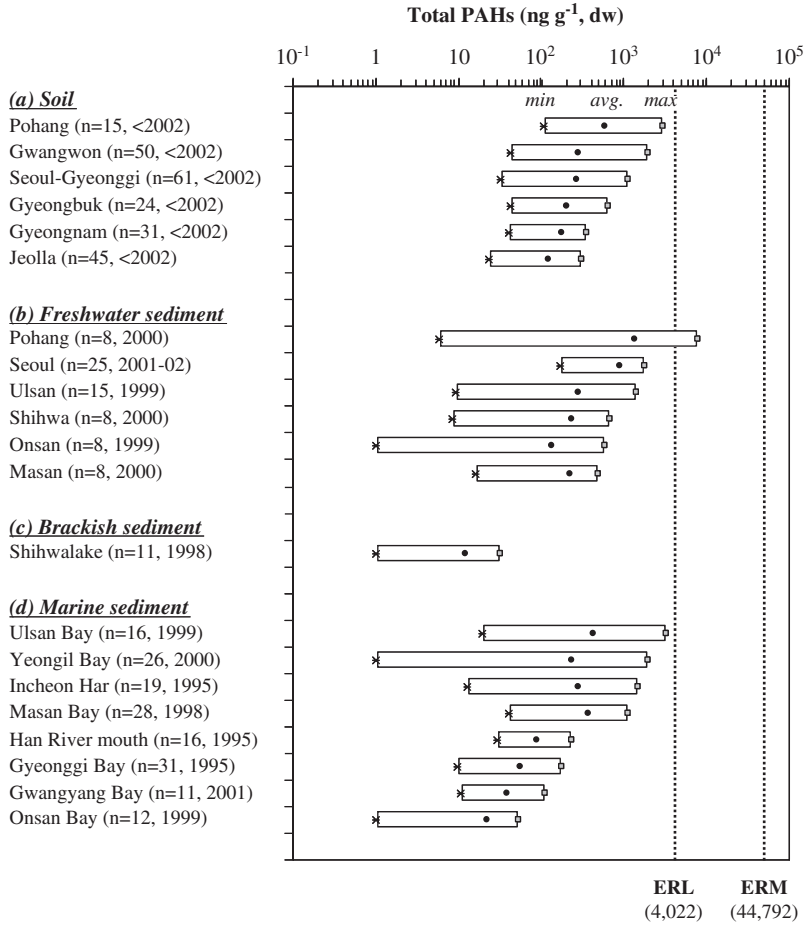

Fig. 2.21. Comparison of measured levels and sediment quality guidelines (SQGs) for total polycyclic aromatic hydrocarbons (PAHs) in soil and sediments from the various sites, in South Korea; (a) soil ($n = 226$), (b) freshwater ($n = 81$), (c) brackish ($n = 11$), and (d) marine sediments ($n = 159$). Range indicates minimum (min), maximum (max), and arithmetic mean (avg.). Dotted lines represent SQGs of effect range low (ERL) and effect range median (ERM) for total PAHs (Long et al., 1995).

water, suspended solids, and sediments in South Korea (Khim et al., 1999a, b, 2001; NIER, 2000a, 2001, 2002, 2003; Koh et al., 2002, 2005; Li et al., 2004a–c, 2005). NP was frequently detected and well reported compared to OP, BP, and BPA. Data available for these compounds are summarized in Table 2.13.

Table 2.13. Concentrations of alkylphenols (APs) and bisphenol A (BPA) in various environmental media in Korea

Sampling				Compounds measured	Total APs and BPA			References
Samples	From	Year	*n*		Min	Max	Mean	
Water					ng L^{-1}			
Freshwater								
Westward	Han river	1999	10	NP + OP + HP + PP + BPA	116	984	429	NIER, 2000a
		2000	10	NP + OP + BPA	30	1266	294	NIER, 2001
		2001	10	NP + BPA	< 10	1203	298	NIER, 2002
		2002	13	NP + BPA	< 10	432	96	NIER, 2003
Southward	Nakdong river	1999	10	NP + OP + HP + PP + BPA	190	874	367	NIER, 2000a
		2000	10	NP + OP + BPA	30	488	211	NIER, 2001
		2001	10	NP + BPA	< 10	871	528	NIER, 2002
		2002	9	NP + BPA	< 10	298	73	NIER, 2003
Westward	Geum river	1999	12	NP + OP + HP + PP + BPA	212	646	342	NIER, 2000a
		2000	6	NP + OP + BPA	38	87	67	NIER, 2001
		2001	6	NP + BPA	10	818	422	NIER, 2002
	.	2002	6	NP + BPA	< 10	380	94	NIER, 2003
Southward	Yeongsan river	2000	6	NP + OP + BPA	20	377	187	NIER, 2001
		2001	6	NP + BPA	47	687	392	NIER, 2002
		2002	4	NP + BPA	< 10	110	51	NIER, 2003
	Other rivers of lakes	1999	11	NP + OP + HP + PP + BPA	55	6340	1133	NIER, 2000a
		2000	11	NP + OP + BPA	20	1169	300	NIER, 2001
		2001	11	NP + BPA	< 10	1742	511	NIER, 2002
		2002	11	NP + BPA	< 10	432	104	NIER, 2003
Westward	Han river	Aug–2001	10	NP	71	188	103	Li et al., 2004(a)
	Han river	Oct–2001	10	NP	42	84	60	Li et al., 2004(a)
	Han river	Dec–2001	10	NP	17	72	43	Li et al., 2004(a)
	6 Creeks, Shihwa	Apr–2000	18	NP	46	41,300	3580	Li et al., 2004(b)
	6 Creeks, Shihwa	Aug–2002	6	NP	90	15,821	3010	Li et al., 2004(c)
	6 Creeks, Shihwa	Oct–2002	6	NP	118	4324	2309	Li et al., 2004(c)
	6 Creeks, Shihwa	Dec–2002	6	NP	131	5239	1339	Li et al., 2004(c)

Brackish								
West coast	Shihwa lake	Apr–2000	12	NP	37	770	183	Li et al., 2004(b)
	Shihwa lake	Aug–2002	10	NP	35	1533	280	Li et al., 2004(c)
	Shihwa lake	Oct–2002	10	NP	17	140	92	Li et al., 2004(c)
	Shihwa lake	Dec–2002	10	NP	31	87	61	Li et al., 2004(c)
Saltwater								
West coast	Saemangeum Bay	May–2002	18	NP	4.5	52	20	Li et al., 2005
	Saemangeum Bay	Aug–2002	18	NP	23	48	30	Li et al., 2005
	Saemangeum Bay	Nov–2002	18	NP	3.1	22	12	Li et al., 2005
	Saemangeum Bay	Feb–2002	18	NP	7.1	79	21	Li et al., 2005
	Saemangeum Bay	May–2003	18	NP	17	129	34	Li et al., 2005
	Saemangeum Bay	Aug–2003	18	NP	28	247	76	Li et al., 2005
Suspended solid						ng g^{-1}, dw		
Freshwater								
Westward	Han river	Aug–2001	10	NP	3.8	17	8.7	Li et al., 2004(a)
	Han river	Oct–2001	10	NP	1.1	9.3	3.6	Li et al., 2004(a)
	Han river	Feb–2001	10	NP	1.0	6.9	3.1	Li et al., 2004(a)
	6 Creeks, Shihwa	Apr–2000	18	NP	600	116,700	14,400	Li et al., 2004(b)
	6 Creeks, Shihwa	Aug–2002	6	NP	69	10,183	2136	Li et al., 2004(c)
	6 Creeks, Shihwa	Oct–2002	6	NP	120	3836	1323	Li et al., 2004(c)
	6 Creeks, Shihwa	Dec–2002	6	NP	8.3	3487	742	Li et al., 2004(c)
Brackish								
West coast	Shihwa lake	Apr–2000	12	NP	93	859	445	Li et al., 2004(b)
	Shihwa lake	Aug–2002	10	NP	31	87	61	Li et al., 2004(c)
	Shihwa lake	Oct–2002	10	NP	19	831	149	Li et al., 2004(c)
	Shihwa lake	Dec–2002	10	NP	15	38	26	Li et al., 2004(c)
Soil						ng g^{-1}, dw		
	Country wide	1999	35	BPA	<0.5	54	7	NIER, 2000a
	Country wide	2000	35	NP + OP + BPA	<0.5	14	3	NIER, 2001
	Country wide	2001	35	NP + BPA	<0.5	8	2	NIER, 2002
	Country wide	2002	35	NP + BPA	<0.5	6	2	NIER, 2003

Table 2.13. (*Continued*)

Sampling				Compounds measured	Total APs and BPA			References
Samples	From	Year	*n*		Min	Max	Mean	
Sediment					ng g^{-1}, dw			
Freshwater								
Westward	Han river	1999	5	NP + OP + HP + PP + BPA	7	61	26	NIER, 2000a
		2001	5	NP + BPA	2	5	3	NIER, 2002
		2002	4	NP + BPA	1	2	1	NIER, 2003
Southward	Nakdong river	1999	3	NP + OP + HP + PP + BPA	88	124	101	NIER, 2000a
		2000	1	NP + OP + BPA	28	28	28	NIER, 2001
		2001	3	NP + BPA	< 0.5	5	3	NIER, 2002
		2002	2	NP + BPA	1	2	1	NIER, 2003
Westward	Geum river	1999	1	NP + OP + HP + PP + BPA	104	104	104	NIER, 2000a
		2000	1	NP + OP + BPA	31	31	31	NIER, 2001
		2001	1	NP + BPA	6	6	6	NIER, 2002
		2002	3	NP + BPA	< 0.5	2	1	NIER, 2003
Southward	Yeongsan river	1999	2	NP + OP + HP + PP + BPA	31	80	55	NIER, 2000a
		2001	2	NP + BPA	< 0.5	2	1	NIER, 2002
		2002	1	NP + BPA	1	1	1	NIER, 2003
	Other rivers or lakes	2000	9	NP + OP + BPA	6	38	15	NIER, 2001
		2002	1	NP + BPA	1	1	1	NIER, 2003
Westward	2 Creeks, Shihwa	Oct–2000	8	NP + OP + BP + BPA	308	5124	1736	Koh et al., 2005
	6 Creeks, Shihwa	Apr–2000	18	NP	1.0	31,700	3008	Li et al., 2004(b)
	Han river	Aug–2001	10	NP	11	624	251	Li et al., 2004(a)
	Han river	Oct–2001	10	NP	56	357	157	Li et al., 2004(a)
	Han river	Dec–2001	10	NP	35	550	192	Li et al., 2004(a)

Southward	3 Rivers, Masan	Nov–2000	8	NP + OP + BP + BPA	158	1114	479	Koh et al., 2005
Eastward	2 Rivers, Onsan	Apr–1999	13	NP + OP + BPA	3.0	1074	168	Koh et al., 2002
	3 Rivers, Ulsan	Apr–1999	15	NP + OP + BPA	3.0	1161	180	Khim et al., 2001
Brackish								
West coast	Shihwa lake	Jan–1998	11	NP + OP + BP	37	1783	637	Khim et al., 1999a
	Shihwa lake	Apr–2000	12	NP	11	624	235	Li et al., 2004b
	Shihwa lake	Oct–2002	12	NP	10	5054	826	Li et al., 2004c
	Shihwa lake	Dec–2002	12	NP	16	2513	772	Li et al., 2004c
Marine								
South coast	Masan Bay	May–1998	28	NP + OP + BP + BPA	129	4168	547	Koh et al., 2002
	Gwangyang Bay	Feb–2001	11	NP + OP + BP + BPA	13	43	24	Koh et al., 2005
East coast	Onsan Bay	Apr–1999	13	NP + OP + BPA	3.0	6.3	3.9	Koh et al., 2002
	Ulsan Bay	Apr–1999	15	NP + OP + BPA	3.0	22	7.4	Khim et al., 2001
	Yeongil Bay	Mar–2000	26	NP + OP + BP + BPA	4.0	3199	194	Koh et al., 2006

2.5.5.1. Environmental levels

Based on the data from 1999 through 2003, concentrations of APs and BPA were relatively greater in creeks in industrial areas than in rivers and lakes (Table 2.13). Among APs and BPA, NP was the predominant chemical species in water and sediment both from inland and coastal areas. Most sites were contaminated with NP where other APs and BPA were generally low.

In a case study of the Shihwa-Banweol, creeks in this area were highly contaminated with NP (10- to 1000-fold greater than those in the Han River and Saemangeum Bay). The maximum concentration of NP observed was as great as 41300 ng L^{-1} in creeks with the average concentrations of from 1339 to 3580, depending upon sampling years. Concentrations of NP in Lake Shihwa ranged from 61 to 280 ng L^{-1}. These concentrations were similar to those of the Han River with 43–103 ng L^{-1} whereas, NP in coastal seawater from Saemangeum Bay were in the range of 12–76 ng L^{-1}.

The AP concentrations in sediments were also greater in creeks than in the brackish and marine environment. The average concentrations of NP, OP, and BP in sediment from Lake Shihwa were 609, 18, and 10 ng g^{-1} dw, respectively (Khim et al., 1999a). The NP concentration was detected up to 116700 ng g^{-1} dw in suspended solids, and 32000 n g^{-1} dw in sediments. The concentrations of APs and BPA in soil samples from the national monitoring sites of NIER were at a few ppb levels, implying that little APs and BPA were introduced into soil.

2.5.5.2. Temporal and spatial distribution

Concentrations of APs and BPA in South Korea appear to decrease with time. Based on the national monitoring data from NIER, the average concentrations of NP and BPA were 446 and 52 ng L^{-1} in 1999, 171 and 60 ng L^{-1} in 2000, 416 and 116 ng L^{-1} in 2001, and 79 and 67 ng L^{-1} in 2002. In the Shihwa-Banweol area, the concentration of NP might be decreasing with time. The greatest concentration of NP (41300 ng L^{-1}) measured in the tributaries to Lake Shihwa in April 2000 was reduced to almost one-third (15821 ng L^{-1}) in August 2002, and continuously decreased to 4000–5000 ng/L in October and December 2002. Concentrations of NP in suspended solids in the Lake Shihwa area was also observed to decrease. The average NP concentration in suspended solids reduced from 4744 to 151 ng g^{-1} dw during the period from 2000 to 2002. This is presumably due to the expansion of wastewater treatment plants (WWTP) and solid treatment plants (STP) in the Shihwa-Banweol

industrial area. However, concentrations of NP in sediments from Lake Shihwa did not show such a rapid reduction, indicating the persistence of NP in sediment. Half-life of NP and OP is known to be in the order of years in sediments (Ying et al., 2002).

2.5.5.3. Comparison with EQCs

To evaluate the risks by water-borne NP exposure, the concentrations of NP in water and sediments were compared to acute and chronic water

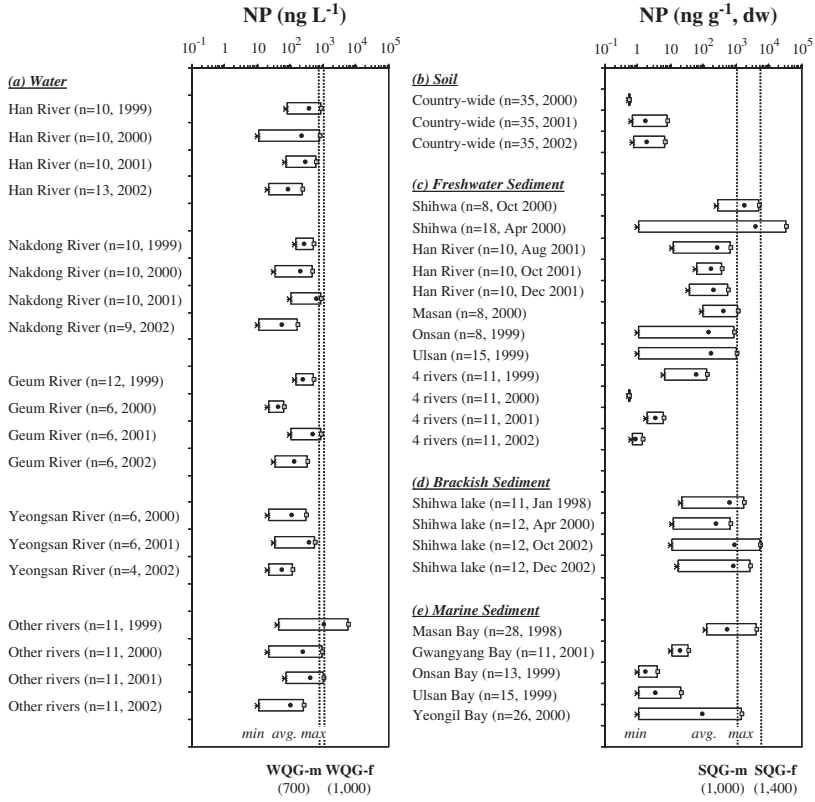

Fig. 2.22. Comparison of measured levels and environmental quality criteria (EQCs) for nonylphenol (NP) in water, soil and sediments from the various sites, in South Korea; (a) water ($n = 172$), (b) soil ($n = 105$), (c) freshwater sediment ($n = 131$), (d) brakish sediment ($n = 47$), (e) marine sediment ($n = 93$). Range indicates minimum (min), maximum (max), and arithmetic mean (avg.). Dotted lines represent Canadian EQCs proposed for marine water (WQC-m) and freshwater (WQC-f), and marine sediment (SQG-m) and freshwater sediment (SQG-f) (see the text).

quality criteria (WQC) and sediment quality guideline (SQG). In this evaluation, we used a final WQC and SQG for NP suggested by the USA and Canada (Environment Canada, 2001; US EPA, 2005). The acute criterion of 28000 ng L^{-1} and chronic criterion of 6600 ng L^{-1} were proposed for freshwater, while acute criterion of 7000 ng L^{-1} and chronic criterion of 1700 ng L^{-1} were proposed for salt water. These values are the concentrations of NP in water at which aquatic life is protected from acute and chronic adverse effects. In only 0.5% of the freshwater samples from South Korea the concentration was over the EPA's proposed chronic WQC of 6600 ng L^{-1} (Fig. 2.22). This is similar to the report that 99% of the detectable levels of APs/APEs and their biodegradation intermediates in US surface waters are below the chronic WQC (only four data among database exceeded chronic WQC). Canadian water quality guidelines (WQGs) for NP and its ethoxylates were proposed in 2002 and the guidelines were lower than USA chronic WQG. Based on the Canadian WQG for NP (1000 ng L^{-1}), 5.1% of water samples exceeded the WQG whereas 1.3% of salt water samples were over the guideline in salt water (700 ng L^{-1}). The fractions of the samples exceeding the Canadian sediment quality guidelines for NP in fresh water (1400 ng g^{-1}) and salt water (1000 ng g^{-1}) were 3.6% and 6.6%, respectively (Fig. 2.22). The percentages of sediment samples over SQG are larger than in water sample, indicating NP contamination in sediment might be more serious than that in water.

United States and Canada have water quality guidelines and European countries banned the use of alkylphenol polyethoxylates. To date, there is no standard available for regulating APs in the environmental media of South Korea. Most of the sites investigated throughout the South Korean peninsula are not expected to be sufficiently contaminated with APs or BPA to cause adverse effects. However, NP concentrations in some waters and sediments were over the guidelines of the USA and Canada. This means that aquatic organisms can be adversely affected by NP exposure in some aquatic systems. Therefore, actions should be taken to implement the regulations of NP to protect the aquatic ecosystems in South Korea.

2.5.6. PBDEs

Although the use of PBBs in South Korea was banned in 1999 the total use of BFRs including PBDEs has increased during the last decade (Park and Jeong, 2001). Concentrations of PBDEs in South Korea have only recently been measured. There are three peer-reviewed papers available, which reported the concentrations of PBDEs in air, sediments and bivalves. One report was available focusing on the PBDEs in human

blood. A few reports on environmental policy of PBDE management were also published (Park and Jeong, 2001; Kim et al., 2002f; KMOE, 2005c).

2.5.6.1. Environmental levels

PBDEs have been detected in air at the sites of Busan, Masan, and Jinhae cities (Moon et al., 2007a), in sediment and bivalve from coastal regions (Moon et al., 2001, 2007b), and in human blood samples from workers and general population in Seoul (Kim et al., 2005e). The predominant PBDE congener in all atmospheric deposition samples was deca-BDE (BDE 209), which accounted for >93% of the total PBDE concentrations, indicating that deca-BDE was mainly used for industrial applications in South Korea. This observation is consistent with domestic consumption history of PBDEs in South Korea. The depositional fluxes of PBDEs from air varied from 10.1 to 89.0 μg m^{-2} $year^{-1}$. The total PBDE concentrations ranged from 0.45 to 494 ng g^{-1} dw (average 27.8 ng g^{-1} dw) in sediments, and from 0.38 to 9.19 ng g^{-1} ww (average 2.94 ng g^{-1} ww) in bivalves. The predominant PBDE congener in sediments and bivalves was deca-BDE, which accounted for >90% and >60% of the total PBDE concentrations in sediment and bivalves, respectively. The relatively great contribution of deca-BDE in sediments may be due to large quantity of industrial use and chemically stable property of deca-BDE. However, relatively great accumulation of deca-BDE in organisms was not consistent with other studies reporting that penta-BDE was more persistent and accumulative in organisms than octa-BDE and deca-BDE, or could be formed from debromination process of heavier molecules (de Wit, 2002).

Specific accumulation of PBDEs in organisms may vary with the source and the organisms' diets and metabolic capacities. Laboratory studies of PBDEs using aquatic organisms showed high potential of bioaccumulation up to the bioconcentration factor of 1,400,000 (Gustafsson et al., 1999). In the Baltic Sea, strong biomagnification in different tropic levels was observed (Martin et al., 2004). As PBDE contamination appear to be an emerging problem, further studies on the distribution, fate and bioaccumulation of PBDEs should be conducted in future in South Korea.

2.5.7. PFAs

Only a few reports of concentrations of PFAs in South Korea have been published (Kannan et al., 2002; So et al., 2004; Rostkowski et al., 2006). The distribution, fate and exposure level of PFAs should be intensively

investigated in South Korea because the very great concentrations of PFAs from water bodies and human blood in South Korea have been reported and the single greatest concentration of perfluorooctanesulfonate (PFOS) was reported for water in Lake Shihwa.

2.5.7.1. Environmental levels

PFAs concentrations in 11 seawater samples collected from west and south coasts of South Korea in 2003 were measured (So et al., 2004). Concentrations of PFOS ranged from 40 to 730000 pg L^{-1} and the average concentrations of west and south coasts were 147000 and 748 pg L^{-1}, respectively. The greatest concentration, 730000 pg L^{-1}, was observed in the vicinity of Shihwa-Banweol industrial complex of west coast, which was thousands of times greater than those reported in open oceans such as the Pacific Ocean, Atlantic Ocean, Zulu Sea, South China Sea, and two orders of magnitude greater than those reported in coastal regions of Japan, Hong Kong and China. The average concentration of PFOA for overall South Korean waters was similar to that in Tokyo Bay, and 3–50 times lower than in Ai River from Japan, but 1–100 times greater than those reported for the open ocean (Table 2.14).

A detailed survey was conducted in the vicinity of the Shihwa-Banweol industrial complex in 2004 (Rostkowski et al., 2006). Concentrations in seawater in 2004 were 100-fold less than what had been in 2003. However, the average (range) PFOS concentration in water from the inland areas near Shihwa was as great as 89000 (8000–651000) pg L^{-1}. The concentration decreased to 12900 (7330–18300) pg L^{-1} in Shihwa Lake, and to 5210 (2240–8260) pg L^{-1} in coastal regions with increasing distance from the source area. The distribution of PFOA in this region was similar to PFOS with an average concentration of 19220 pg L^{-1} in inland area, 6140 pg L^{-1} in Shihwa Lake, and 2200 pg L^{-1} in bay area. The distribution pattern of PFNA in this region was similar but the concentrations of PFHxS, PFBS, PFOSA, PFDA, PFNA, and PFHxAha were one order of magnitude lower than those of PFOS and PFOA. This means that there is a continuous input of PFOS from Shihwa-Banweol inland area.

The concentrations of PFAs were reported for 43 individuals of 10 water bird species collected from Hongdo in the vicinity of Nakdong River, and Nando, west coast of South Korea, between 1993 and 1994 (Kannan et al., 2002). PFOS, PFOSA, PFOA and PFHS were analyzed in this study, but only PFOS was found in bird samples. The greatest and average concentration of PFOS in these bird liver samples from South Korea were 500 and 80 ng g^{-1} ww, respectively, while those in Japan were 650 and 215 ng g^{-1} ww, respectively. PFOS was detected in all bird

Table 2.14. Concentrations (pg L^{-1}) of PFAs in water from various regions

Sampling			PFOS			PFOA			PFHS			PFNA			References
Area	Year	*n*	Min	Max	Mean	Min	Max	Mean	Min	Max	Mean	Min	Max	Mean	
Korea															
River (Shihwa-Banweol)	12–04	17	8030	651,350	89,110	5210	61,690	19,220	1940	84,610	10,700	830	6950	3260	Rostkowski et al., 2006
Lake Shihwa	12–04	5	7330	18,320	12,850	1670	10,860	6140	630	2040	1310	630	2100	1320	Rostkowski et al., 2006
Gyeonggi bay	12–04	5	2240	8260	5210	940	3290	2200	550	990	720	510	830	680	Rostkowski et al., 2006
South coast of Korea	01–03	6	40	2300	748	240	11,000	4840	30	460	240	40	590	242	So et al., 2004
West coast of Korea	01–03	5	620	730,000	147,028	1300	320,000	65,680	160	52,000	10,898	50	13,000	2662	So et al., 2004
Japan															
Ai river	03–04	5	2900	12,300	46,300	16,700	87,100,000	236,017	–	–	–	–	–	–	Morikawa et al., 2006
Tokyo bay	02–04	8	338	57,700	–	1800	192,000	–	17	5600	–	163	71,000	–	Yamashita et al., 2005
Offshore of Japan	02–04	4	40	75	–	137	1060	–	3	6.1	–	–	–	–	Yamashita et al., 2005
China															
Coastal area of Hong Kong	02–04	12	70	2600	–	673	5450	–	5	311	–	22	207	–	Yamashita et al., 2005
Coastal area of China	02–04	14	23	9680	–	243	15,300	–	5	1360	–	2	692	–	Yamashita et al., 2005
Open ocean															
Sulu sea	02–04	3	17	109	–	88	510	–	0.2	0.2	–	–	–	–	Yamashita et al., 2005
South China sea	02–04	2	8	113	–	160	420	–	0.2	0.2	–	–	–	–	Yamashita et al., 2005
Western Pacific ocean	02–04	2	54	78	–	136	142	–	2.2	2.8	–	–	–	–	Yamashita et al., 2005
Central-Eastern Pacific ocean	02–04	12	1.1	20	–	15	62	–	0.1	1.6	–	1	16	–	Yamashita et al., 2005
North Atlantic ocean	02–04	9	8.6	36	–	160	338	–	4.1	6.1	–	15	36	–	Yamashita et al., 2005
Mid Atlantic ocean	02–04	7	37	73	–	100	439	–	2.6	12	–	–	–	–	Yamashita et al., 2005

samples collected from South Korea and Japan, indicating that it is one of the most persistent and accumulative PFAs. PFOS has a great potential for bioaccumulation relative to other PFAs including PFOA. The persistence and bioaccumulative potential of PFOS in organisms have been reported in other studies (Martin et al., 2003; Morikawa et al., 2006). The average concentration of PFOS in human blood in Daegu, South Korea, was similar to that in Japan, 5–7 times greater than that in Indian and Italian samples, and two times less than in American and Polish samples (Kannan et al., 2004). The average PFOA concentration in human blood was 62 ng ml^{-1} in South Korea. The level is three times greater than in American and Polish, six times greater than in Japanese and Malaysians, and over 10 times greater than in other general populations in other countries. It is noteworthy that: (1) most of imported PFAs might be used as water-repellant for textile and leather industries; (2) PFCAs produced by telomerization process are likely to be the main PFAs source in South Korea after phase out for perflurosulforamide products of 3 M; and (3) over 50% of PFAs imported from abroad might have been used in Daegu and neighboring Kyeongsangbukdo where a large number of textile plants have been in operation since the 1950s.

Although classical POPs such as PCBs, DDTs and CHLs in environmental media from South Korea were relatively less compared to other industrialized countries, the emerging pollutants including PBDEs, and PFAs in South Korea were relatively great. Great concentrations of PFOS and PFOA in human serum were also reported in recent studies in some areas of South Korea. Therefore, it is necessary that the source, distribution, and fate of emerging pollutants in environmental media should be investigated nationwide and that the contamination levels and the exposure routes of pollutants to human and wild life should be assessed. Additionally, establishment of toxic release inventory, source control and management, and ecological/human risk assessment should be performed. Especially, human exposure from the consumption of fishes and shellfishes in South Korea should be assessed for these emerging POPs.

2.6. Fate and transport of POPs in South Korea

2.6.1. Sources of PCDDs/DFs in environmental media

Relatively great concentrations of PCDDs/DFs ranging from 22.15 to 92.99 pg I-TEQ g^{-1} fat wt (with an average concentration of 53.42 pg I-TEQ g^{-1} (fat wt)) were reported in the blood of residents near a large

scale industrial waste incinerator (CIES, 2002). Furthermore, more incidences of glycosuria and cancer were reported in the population near the incinerator facility than those in the distant areas. However, more epidemiological investigation would be required to accurately assess the effects of PCDDs/DFs. Extensive measurements of residues in soils, sediment at the wastewater discharging sites, agricultural products (including poultry), and human blood were performed from 2002 to 2003 (Pyoungtak City, 2003). These efforts were focused on revealing the health effects from exposure to point sources (i.e., waste incinerator) of PCDDs/DFs in the environment. Although no clear difference was found in the extent of contamination of human blood or environmental media between sites near and away from incinerators, the influence of incinerator on dioxin exposure was clearly shown by the congener patterns of PCDDs/DFs that changed with distance from the incinerator in soil and human blood (Park et al., 2004). Furthermore, concentrations of PCDDs/DFs in soils decreased as a function of distance from the incinerator over the first few hundred meters. However, no clear trend was found beyond 200 m (for instance, ~60 pg WHO-TEQ g^{-1} near incinerator; ~15 pg WHO-TEQ g^{-1} for 20 m; and ~3 pg WHO-TEQ g^{-1} for most samples over 200 m). Soils 200 m away from the incinerators had similar TEQs with an average of 2.32 pg WHO-TEQ g^{-1} (geomean, $n = 3$) detected at 7 km away from the incinerator. Thus, there is a characteristic spatial distribution of concentration with a clear decrease near the source followed by a fairly even distribution.

In contrast to the total concentration, a gradual transition was found for the relative composition of PCDDs/DFs congeners with distance from the incinerator. The proportion of PCDFs, particularly 1,2,3,4,6,7,8-HpCDF and OCDF, which were the dominant congeners in soils near sources such as stack gas and fly ash decreased progressively with distance while OCDD enriched with distance. A similar trend in patterns of relative concentrations of congeners was observed in blood of residents in the area. The composition of PCDF congeners among 2,3,7,8-substituted PCDDs/DFs was significantly greater in blood of residents living near incinerators than in blood of those living farther away from incinerators. Based on the congener composition together with concentration gradient, the exposure and effect zones were determined to be within 1–1.5 km of the incinerator (Park et al., 2004). Several other studies have also reported point sources of dioxins and their effects, i.e., the impact of sources to the surrounding environment. In the late 1990s, Kim et al. (2002c) and Oh (2001) compared concentrations and composition profiles in soils near incinerators used to combust solid wastes from paper mills or municipal solid waste incinerators with those of stack gas. In order to obtain

information on the emissions of industrial waste incinerators (IWI) to their surrounding environment, Kim et al. (2005d) determined the concentrations of PCDDs/DFs in various samples such as stack gas, fly ash, ambient air, soil, pine needles, rice plants, IWI worker blood collected from different distances in the vicinity of IWI. Recently, Kim et al. (2005b) and NIER (2005b) reported the results of a five year project conducted between 2003 and 2008 to measure concentrations of PCDDs/DFs in stack gas, wastewater, sediment, and soil around incinerators. Based on a meta-analysis of the data collected from these studies, it was concluded that there was a common composition pattern which changes gradually from the source while the concentrations decreased more rapidly before reaching a relatively uniform pattern (Fig. 2.23). At the same distance from incinerator, similar concentrations were detected in rice fields, upland crop fields, and forests, whereas 30-fold greater concentrations were measured in roadside soil (NIER, 2005b). Residual PCDDs/DFs concentrations in soil decreased dramatically to concentrations less than 10 pg I-TEQ g^{-1} within 750 m of the incinerator (Fig. 2.23a-1). Gaussian modeling using ISC (Industrial Source Complex) and ISCLT (Industrial Source Complex Long Term), showed maximum concentrations occurred at a distance of approximately 250–500 m from the incinerator (NIER, 2005b). Meanwhile, absolute and relative concentrations of PCDF and PCDD congeners, particularly 1,2,3,4,7,8,9-HpCDF and 1,2,3,4,6,7,8,9-OCDF decreased while the concentration of 1,2,3,4,6,7,8,9-OCDD increased progressively from source to distant locations beyond 1 km (Fig. 2.23a-2–a-4). It is noteworthy that the trend of transition in the congener composition was clear, although the meta-analysis did not consider wind direction and land-use pattern, when the roadside samples that were collected within 500 m from an incinerator were excluded. Thus, incinerators have a significant impact only within a few kilometers.

The distance-dependent transition in relative congener composition was also observed in aged soils that had been affected by a forest fire (Fig. 2.23b-1) (Kim et al., 2003a). Five months after burning, concentrations decreased to be the same in unburned soil at a remote area. However, the weathering of congeners seems to progress more slowly (Fig. 2.23b-2–b-4). The predominance of OCDD composition in aged or remote soils results from: (1) its intrinsic fate property such as low vapor pressure, water solubility, biodegradability, and reaction with hydroxyl radicals; (2) release from use of pentachlorophenol (PCP); and (3) long environmental residence due to strong air-particle association and its deposition to underlying surface. The analysis of congener composition is found to be useful to identify the sources.

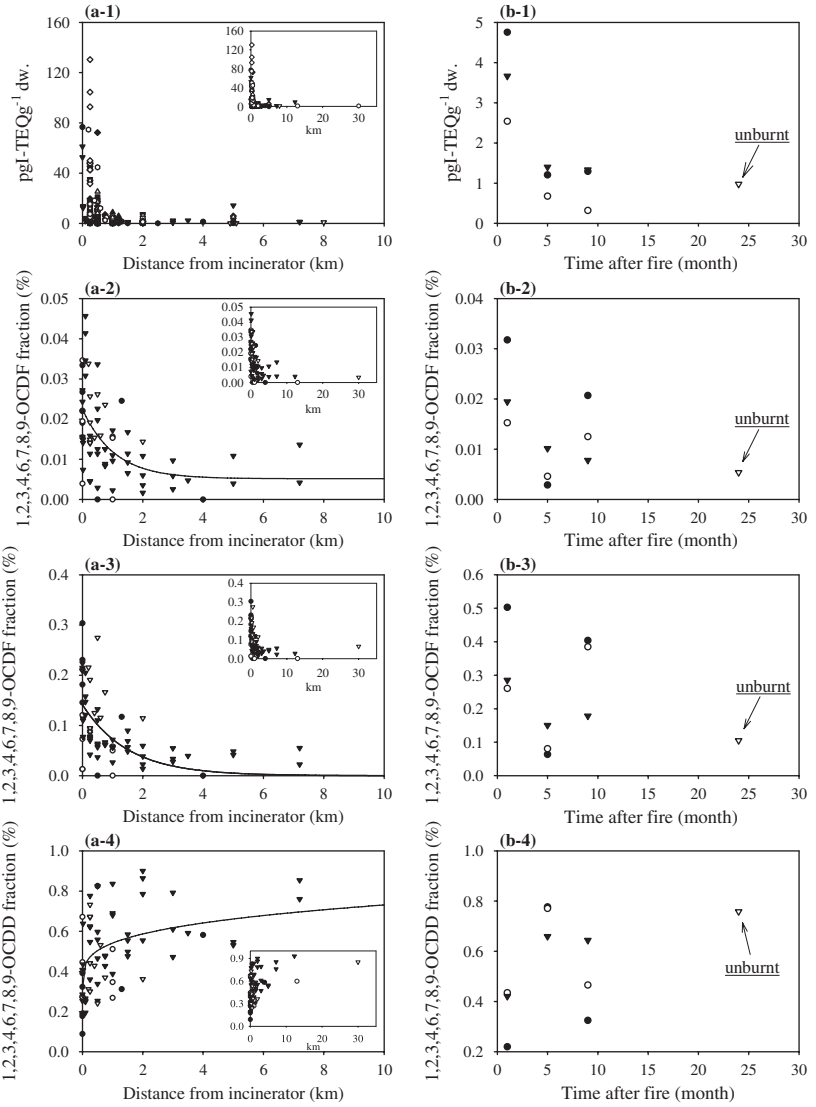

Fig. 2.23. Variation in total PCDDs/DFs concentration and congener fractions measured for soils with distance from a source ('a' group, Kim et al., 2002c; Park et al., 2004; Kim et al., 2005d; Oh et al., 2006; NIER, 2005b) and with time after forest fire ('b' group, Kim et al., 2003a). An unburned remote soil sample was assigned to 24th month after fire.

2.6.2. Case study of multi-media fate of POPs (PAHs vs. PCBs)

Classical organochlorine POPs are hydrophobic and semi-volatile. They distribute among environmental media via inter-media transfer, partitioning, volatilization, and dry/wet deposition. The direction and magnitude of movement across media are dependent on media and concentration at which they were emitted, chemical properties including volatility, solubility, hydrophobicity, and persistency, meteological/morphological/ecological conditions of the system. These factors vary with time and space. For determining the direction and magnitude of movement among media, it is inevitable to simultaneously measure chemical residues and environmental factors in each of the media. One can infer the transfer (or transport) among media through thermodynamic index such as fugacity ratio and environmental processes such as dry/wet or deposition flux.

The multi-media fate of PAHs and PCBs in Seoul Metropolitan city with a population of over 10 million has been investigated by use of concurrent measurements in each season for multiple environmental media including air (gas and particle phases), water (dissolved and suspended solid phases), sediment, terrestrial soil and plant leaf, and aquatic plank tonic organisms (Kim, 2004). To our knowledge, this is the only study to describe multimedia fate and distribution of hydrophobic organic compounds (HOCs) in South Korea that has been conducted based on field-measured data. The study revealed that knowledge of the interrelationships among different media is critical to elucidating distribution and behavior of HOCs in each medium. In the study, the differences in overall fate such as distribution, behavior, and transport pathway were observed between PAHs and PCBs. We describe below some of the highlights of the study of multimedia fate and transport of POPs in South Korea.

2.6.2.1. Air–water exchange

'Fugacity', representing 'escaping tendency' of a chemical in a medium, has been widely applied in assessing the status of fate and transport of a chemical in and among environmental media (Mackay, 1991). If one knows chemical concentration and fugacity capacity in each medium, the fugacity difference can be calculated as in the Eq. (2)

$$\frac{f_i}{f_j} = \frac{C_i Z_i}{C_j Z_j} \tag{2}$$

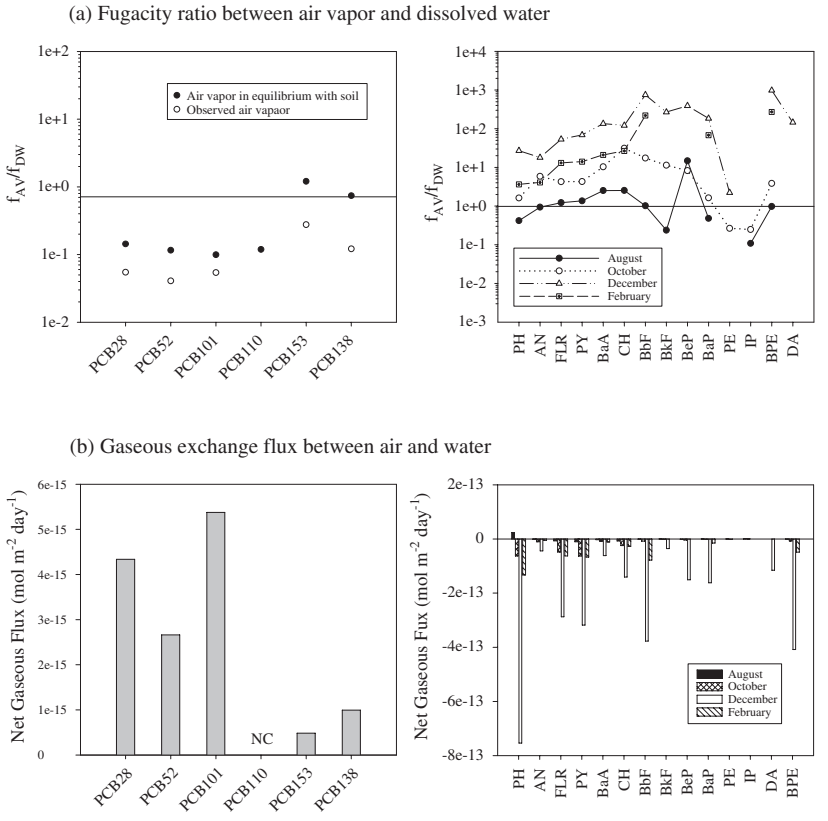

Fig. 2.24. Fugacity ratios (a) and gaseous exchange flux (b) between air and river water for PAHs and PCBs from Seoul Metropolitan city in 2001–2002 (quoted from Kim (2004)); fAV and fDW is fugacity in air vapor and dissolved water, respectively. Flux of PCBs was calculated on basis of observed vapor PCBs and 'NC' indicate 'not calculated'.

where, $C_{i,j}$, $Z_{i,j}$, and $f_{i,j}$, are chemical concentrations (mol m^{-3}), fugacity capacity (mol m^{-3} Pa^{-1}), and fugacity (Pa) in medium i and j, respectively. The ratio of i–j fugacity, f_i/f_j is a measure of departure from equilibrium. Values greater than unity indicate that there is net-diffusion from the medium i to medium j. Conversely, values less than unity indicate net diffusion to the medium i. For a system in equilibrium, f_i/f_j is approximately equal to 1. Based on these fugacity ratios among media, one can determine the moving direction of a chemical between two contacting media. For example, air–water fugacity ratios measured for PAHs in Seoul city area were over 1 in all seasons but summer, indicating the diffusive movement from air-to-water (Fig. 2.24a). Contrarily, the

air–water fugacity ratios of PCBs were below unity, which suggests evaporation from water to air (Fig. 2.24a).

In addition to qualitative assessment of direction of movement of a chemical using fugacity ratio, quantitative assessment is determined by calculating net gaseous diffusion across air–water interface (Mackay et al. (1986); Baker and Eisenreich, 1990; Achman et al., 1993). The two-film theory as shown in Eq. (3) has been frequently used to determine the gaseous diffusion rate of chemicals (Whitman, 1923; Liss and Slater, 1974).

$$F = k_{\mathrm{ol}}\left(C_{\mathrm{DW}} - \frac{C_{\mathrm{AV}}}{\mathrm{H}'}\right) \tag{3}$$

where, F is the flux (mol m^2 day^{-1}), and k_{ol} (m day^{-1}) is the overall mass transfer coefficient for movement across air–water interface. C_{DW}, C_{AV}, and H' are dissolved concentrations in bulk water and the air vapor concentration in bulk air of HOCs, and the dimensionless Henry's law constant, respectively.

The air-to-water flux of PAHs and PCBs in the Han River was distinctive (Fig. 2.24b). The overall gaseous transfer of PCB congeners occurred from water to air with greater transfer rates for lower chlorinated congeners; for instance, 4.34×10^{-5} mol m^{-2} day^{-1} for PCB 28 and 4.84×10^{-6} mol m^{-2} day^{-1} for PCB 153. Obviously, the mass transfer coefficient is greater for lighter substances. The transfer of PAHs occurred from air to water with higher values in winter than in summer. This observation indicates that the burden and fate of PAHs in the Han River aquatic system can be strongly influenced by the variation in airborne concentration (i.e., atmospheric emission) whereas the distribution of PCBs could be controlled by the burden discharged into the water body (e.g., desorption from historically contaminated sediments or direct release into water) rather than atmospheric input.

The difference in environmental release of PAHs and PCBs is further supported by their differences in seasonality of atmospheric concentration. PAH concentrations in air increased gradually in winter. This seasonality was also observed in more extensive measurements in Seoul city's air (Panther et al., 1999). The seasonality in concentrations of PAHs in urban areas often results from greater combustion of fuel for heating in wintertime. To the contrary of PAHs, the potential emission sources of PCBs such as waste incinerators or metallurgical processes do not show significant seasonality. Atmospheric concentrations of PCBs were greater in summer than in winter (Yeo et al., 2003). In particular, a strong negative linear regression between partial pressure (i.e., ln p; concentration) of atmospheric PCBs and the reciprocal of temperature (i.e., 1 T^{-1}) was observed in urban air (Yeo et al., 2004a). Negative slopes of greater

magnitude derived from the Clausius-Clapeyron equation (Eq. 4) indicate temperature-dependent distributions of PCBs in the air. PCBs in air may not originate from direct release but by secondary source such as volatilization from terrestrial surface.

$$\ln p = \frac{\mathrm{m}}{T} + \mathrm{b} \tag{4}$$

where, p is partial pressure (Pa), T is ambient air temperature (K), and m and b are constant.

The major source of by-product PCB emission (~93%) was estimated to be ferrous metal industry (see Section 2.4.1.1). All metal production factories are located in coastal areas far from the Seoul metropolitan area. A possible emission source in Seoul is waste incinerators. However, the contribution from waste incinerators is generally small (see Fig. 2.6). Thus, direct atmospheric release is not likely to influence the distribution of PCBs in the air over South Korea. On the other hand, the leakage or spillage from capacitors and transformers may be a potential emission source of PCBs. This emission process is categorized as a major contributor of PCBs into the environment (EMEP/CORINAIR, 2000). Although regulation of the use of PCBs began in 1979, existing PCB-containing articles were in use for other purposes, except electric heater, until 1996. Therefore, PCBs are likely to have been introduced into aquatic system through leakage or spills from the use and/or storage of the articles. PCB concentrations were greater at the sites in the tributaries of the Han River flowing through the urban and industrial areas (Kim, 2004).

OCPs such as DDTs, HCHs, CHLs, and endosulfan showed a similar seasonal variation of atmospheric levels as PCBs with greatest levels in summer (Yeo et al., 2004a). This seasonal distribution is due to limited environmental release of OCPs following ban of OCPs in South Korea.

In contrast to PCBs and OCPs, the seasonal trend of atmospheric PCDDs/DFs in South Korea is not likely to be influenced by temperature, i.e., evaporation from surface. Oh et al. (2006) measured seasonal trend in ambient air around a large-scale municipal waste incinerator from August 1999 to November 2000. PCDDs/DFs concentrations ranged from 0.22 to 1.15 pg I-TEQ m^{-3} ($n = 10$, geomean = 0.64 pg I-TEQ m^{-3}). The great concentrations, including the greatest value, were detected in winter even though there was a weak relationship between temperature and concentration ($r^2 = 0.247$). This seasonal trend was also observed in monthly bulk atmospheric deposition samples at Busan and its surroundings (Moon et al., 2005). PCDDs/DFs bulk deposition flux was lowest in the summer season when temperature and precipitation

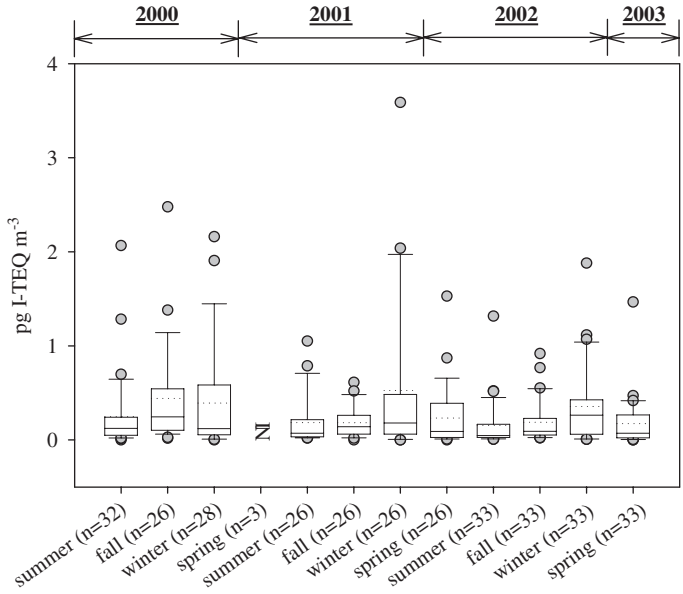

Fig. 2.25. Seasonal variation of atmospheric PCDDs/DFs concentration in Korea (from NIER, 2001, 2002, 2003). The boundary of the box indicates the 25th and 75th percentile, and a line within the box marks the median. Whiskers above and below the box indicate the 90th and 10th percentiles. Points and dotted lines are the arithmetic mean and outlying points. Values in the parenthesis are the number of samples. Three points from spring in 2001 ('NI') were not included.

were highest, whereas the flux was high in winter and moderate in spring and fall. Wet deposition by precipitation together with dry deposition is known to play a major role to scavenge PCDDs/DFs air to underlying surface. Thus, the lesser deposition flux that occurs in summer implies a minimum concentration would occur in air during the warm season. Extensive seasonal monitoring of ambient air was performed from 2000 to 2002 by NIER (2001, 2002, 2003). Atmospheric distribution of PCDDs/DFs was not expected to show any seasonality because waste incinerators and industrial facilities, two major emission sources of PCDDs/DFs, may emit PCDDs/DFs at a fairly constant rate throughout the year. However, as shown in Fig. 2.25, the concentrations in winter were significantly greater than those in other seasons in 2001 and 2002 (ANOVA test, $p < 0.05$). This might be a result of lower mixing heights and reduced photochemical degradation of PCDDs/DFs in winter. The little temperature dependency of atmospheric PCDDs/DFs concentration, is different from those of PCBs and OCPs, and indicates the ongoing release of

PCDDs/DFs into air. Thus, it is expected that the levels in other media, soil in particular, may also increase or only decrease slowly even though emission of PCDDs/DFs was dramatically reduced (Fig. 2.25). Therefore, further regulations on PCDDs/DFs emission is needed in South Korea.

The third piece of evidence for differences in the sources and fate of PAHs and PCBs is the fingerprint of isomers (or congeners) of each chemical group. Based on the differences in thermodynamic stability among PAH compounds, PAHs molecular indices have been widely used to identify potential PAH sources. For instances, PHE:ANT ratio <10 and FLT:PY ratio >1 tends to suggest that PAH contamination arises from pyrogenic origin (Yunker et al., 2002). Additionally, ratios such as BaA:CHR and I123cdP:BghiP have been also suggested as useful indices to identify various combustion processes (Dickhut et al., 2000; Yunker et al., 2002). Molecular indices from most sediment and suspended solid samples in the Han River suggested petroleum combustion as the major source of PAHs (Kim, 2004). Thus, it may be inferred that atmospheric release from petroleum combustion mainly contributed to contamination of the aquatic systems in Seoul. Alternatively, PCBs in sediment of the Han River appears to have originated from leakage of PCBs mixtures based on their congener pattern that is similar to that of Aroclor mixtures (ca., Aroclor 1242:1254:1260 = 2:1:1) (Kim, 2004). This pattern from sediment was significantly different with those of terrestrial soils and settled air particles (Kim, 2004).

2.6.2.2. Control factors for multimedia fate

Many factors influence in complicated manners the magnitude and direction of transport and the distribution of chemicals in the environment. The factors include both the chemical properties and environmental variables such as emission, metrological and morphological conditions. Therefore, it is necessary to identify dominant factors to understand the fate and distribution of POPs in a system and to establish system-specific control strategies for POPs.

The difference in the emission histories of PAHs and PCBs was the predominant factor for their fate, transport, and distribution in Seoul. Besides transfer directions among media such as air–water partitioning, the relative residue levels in individual medium may be determined based on sources of release. PCB 52 and BaA have similar hydrophobicities (i.e., log $K_{ow} = 5.84$ vs. 5.55 for PCB 52 and BaA, respectively) but Henry's law constant for PCB 52 is 100-fold higher. Thus, if both the compounds are assumed to be released to the same media, BaA would show higher partitioning to aqueous media in an air–soil–water-sediment

system as compared with PCB 52. However, the residual mass of PCB 52 in the aquatic system was 0.41% in the water column, 31% in sediment, and 68% in soil whereas almost all BaA of over 90% resided in soil, 0.01% was in the water column and 8.12% in sediment (Kim, 2004). This difference in relative residues in environmental media is associated with the difference in the main release media, i.e., into water via air for PAHs and into directly water for PCBs. The effect of release media on relative distribution of POPs among the media can be conveniently investigated by multi-media modeling simulation with different emission scenarios.

The size distribution of air particles not only influences the distribution and partitioning dynamics of POPs, but also determines dry and wet deposition flux of POPs. An interesting phenomenon was observed for relationship among atmospheric PAHs, particle size distribution, and the levels of PAHs in soil (Kim, 2004). For urban sites, the composition pattern and absolute concentrations of PAHs in soil were well correlated with those in air where the atmospheric particles' size was distributed evenly among seasons with predominant amount of fine particles $<3\,\mu m$. Dry deposition flux of PAHs followed seasonal variation in atmospheric concentration in urban site. However, at a suburban site with large seasonal variation in particle size distribution, dry deposition flux and soil residue did not reflect the seasonal variation of atmospheric PAHs. From this result, site-specificity in atmospheric particle distribution may also influence the distribution and residues in the underlying soil.

The behavior and phase distribution of POPs within an aquatic system is expected to be largely controlled by the affinity to particles according to their hydrophobicity and sorbent properties. In the Han River, which is frequently eutrophied, the partitioning between water and particles and distribution of PCBs were well explained by the conventional concept that their levels are determined by their hydrophobicity and the fractional organic carbon contents of particles. However, the K_{oc} values of PAHs obtained in the field were typically 10- to 100-fold greater than those predicted from the two-phase equilibrium partitioning model using the chemicals' hydrophobicity (i.e., K_{ow}) and the particles' fractional organic carbon (i.e., f_{oc}) (Fig. 2.26). In eutrophic seasons with algal blooms, the partition coefficients of PAHs were greater than those calculated by the combination of chemicals' hydrophobicity, K_{ow}, and organic carbon content, f_{oc}.

The same phenomenon was also observed in sediments (*not presented*). Gustafsson et al. (1997) explained the greater partition coefficients by using the expanded model, $K_d = f_{oc}K_{oc} + f_{sc}K_{sc}$, which accounts the role of soot-like particle with greater sorption capacity rather than organic carbon alone. Sorption capacities were extensively assessed in various

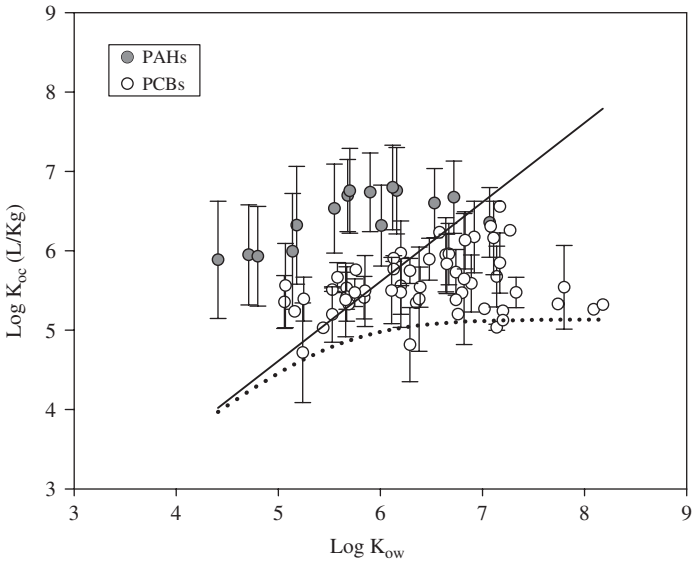

Fig. 2.26. Organic carbon-normalized partitioning coefficients (log K_{oc}) in the Han river water column for PAHs and PCBs (quoted from Kim (2004)); Linear solid line indicates the estimates from two-phase partitioning model (Karickhoff, 1981) and dotted line denotes those from three-phase partitioning model involving colloidal phase (Baker et al., 1986), where $K_{colloid} = K_{oc}$ was assumed and average DOC was used.

particles (Cornelissen et al., 2005) or other POPs (Persson et al., 2002). This observation supports that the fate and distribution of PAHs in locations with intensive combustion activity could be strongly influenced by external particles (e.g., soot-like particles) rather than organic carbon alone. Organic carbon-based chemical concentrations in sediment have been used to derive sediment quality guideline (SQG) to protect aquatic benthic organisms (DiToro et al., 1991; Swartz, 1999; Rogers, 2002). The equilibrium partitioning (EqP) method is currently supported by the US EPA for the development of sediment quality criteria for non-ionic organic chemicals. The EqP approach assumes that partitioning of sediment-bound chemicals between pore water and sediment organic carbon is governed by the organic carbon partition coefficient (K_{oc}) under steady-state conditions. These results indicates that further studies of sediment quality criteria needs to be conducted on soot carbon-bound concentration but not organic carbon-based concentration alone.

In contrast to PAHs, PCB congeners did not reach the equilibrium between suspended particles and dissolved water (Fig. 2.26). The more chlorinated congeners are, the greater is algal bloom, the greater was the

deviation in partition coefficients from the equilibrium, based on the $K_{oc}-K_{ow}$ plot (Kim, 2004). This indicates that PCBs are still in the process of sorption from dissolved water to suspended particles.

2.6.3. Long-range transport (LRT)

One of issues of POPs is their Long Range Transport (LRT) passing the national boundaries. UNECE/EMEP (United Nations Economic Commission for Europe/Co-operative program for monitoring and evaluation of long range transmission of air pollutants in Europe) adopted 'CLRT protocol' in 1998 to control trans-boundary movement of 16 potential POPs (http://webdab.emep.int/). In 2004, Stockholm Convention for international cooperation against POPs entered into force. In addition, the project, funded by the GEF (Global Environment Facility) and implemented by the UNEP, was performed to make regional assessment of the damages and threats posed by Persistent Toxic Substances (PTS; www.chem.unep.ch/pts). As a result of GEF-UNEP project, countries involved in Region VII (i.e., Central and North East Asia) submitted the regional report in which dioxins and PCBs were assigned as priority PTS to manage in the region in 2002. In East Asia, there is another international effort to control trans-boundary movement of POPs such as 'Establishment of POPs Information Warehouse in East Asia', through which countries in the region share POPs monitoring data and cooperate for POPs control.

LRT of atmospheric pollutants has become a concern in South Korea partly because release of numbers of pollutants in eastern China may be a potential source of air pollution in South Korea (Kwon et al., 2002). Studies of LRT, however, have focused on suspended particulate matter such as PM_{10} or $PM_{2.5}$ (Lee et al., 2006b), particle-associated heavy metals (Han et al., 2002), or acidic air pollutants (Park and In, 2002). Since Gosan in Jeju Island is used as a reference site for ACE-Asia (Asian-Pacific Regional Aerosol Characterization Experiment) project in 2001, many studies are performed for LRT of pollutants from East Asia. Ghim et al. (2003) reported the variation in atmospheric concentrations of POPs including PCDDs/DFs, coPCBs, OCPs, and PAHs in Asian dust at the reference site, Gosan. Little increase in atmospheric concentrations of POPs was observed during the Yellow Sand season except for PAHs. Another study measured atmospheric bulk deposition of PCDDs/DFs in urban and suburban areas in South Korea during one year including periods of Asian dust. Elevated levels were not also observed in the presence of Asian dust (Moon et al., 2005). Instead, they illustrated the effect of Asian dust on South Korean air by using the specific congener

patterns observed during the period, clearly differing from those of other seasons. These studies were, however, restricted to a short term. Thus, further long-term studies are needed on trans-boundary LRT of POPs in this region.

LRT dynamics of POPs can be predicted and evaluated by environmental multimedia modeling (MMM). The models with different spatial scale such as POPsME, EDCSeoul, and KoEFT-PBTs have been developed to predict the fate and transport of classical POPs or VOCs in multimedia environments (Lee et al., 2004; NIER, 2001, 2002, 2003; Lee, 2005). These models are considered to serve as a basis for the future development of LRT models of the north-east Asian region.

2.7. Conclusions

Due to their persistent, bioaccumulative, and toxic properties, POPs are worldwide categorized as priority pollutant group to be prohibited in their use and distribution, or removed from environments. Long-range transportation of POPs has result in relevant international efforts such as Stockholm Convention. South Korean government has followed actively the needs of the convention; 1) regulation in use, distribution, and treatment of POPs substances itself and/or POPs-containing facilities and wastes, 2) development and application of BAT and BEP to reduce POPs release, and 3) monitoring and investigation on POPs contamination nationwide. Nevertheless, the results of investigation and research efforts were not assessed through integrated overview and subsequently the POPs status in South Korean environments is not still evaluated until now.

The present study exhibits the meta-analysis results of data collected from total 228 references published since mid-1990s. Emission, contamination and exposure levels in environments and human for both classical and emerging POPs were firstly and thoroughly reviewed together with historical governmental management strategy and plans. Moreover, the environmental fate and transport natures of POPs in South Korean environments were analyzed, from which POPs behavior status were evaluated to suggest the efficient management strategy.

Most studies were focused on the classical POPs such as organochlorinated pollutants and little for emerging POPs such as PBDEs and PFAs. Preliminary emission inventory were estimated by use of the top-down approach for by-product POPs. Recently, an emission inventory, based on measurements, was established for dioxins. Over 80% of the total emission for all by-products was determined to be from two major sources, viz. waste incineration and metal production processes, among

the sources, suggesting the necessity of control focused on corresponding sources.

Based on the seasonal atmospheric levels and their environmental fate, existing emissions are still influencing significantly the distribution of dioxins and PAHs but not for PCBs and OCPs. Due to governmental regulatory action, during the most recent five years, emissions of dioxins have been significantly reduced. However, the concentration levels in environments are likely to have decreased but the reduction extent was less clear, compared with that of emission. In particular, annual emission rate per unit area was still in great, relative to other countries. Consequently, environmental concentrations and human exposure levels in the source areas frequently exceeded criteria suggested to protect environments and human. Thus, to decrease human and ecological exposure, more reductions in emissions should be required. Besides dioxins, contaminations of other classical POPs (including PCBs and OCPs) were localized in hot spots, such as coastal industrial area and/or big harbors and waste incineration areas. In some of these areas, concentrations of residues exceeded corresponding quality criteria. If these hot spots are excluded from the analysis, general populations and ambient environments did not contain concentrations exceeding established criteria. In contrast to classical POPs, South Korean environments were observed to be relatively more contaminated with emerging POPs, than were other countries. This is likely due to the recent dramatic economic growth of South Korea. However, to evaluate emission rates status and trends of contamination of South Korea further monitoring efforts should be performed, especially for the emerging pollutants.

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