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Case Report

Toxic Volatile Organic Compounds (VOCs) in the Atmospheric Environment: Regulatory Aspects and Monitoring in Japan and Korea

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Abstract: In the past decades, hazardous air pollutants (HAPs), so-called air toxics or toxic air pollutants, have been detected in the atmospheric air at low concentration levels, causing public concern about the adverse effect of long-term exposure to HAPs on human health. Most HAPs belong to volatile organic compounds (VOCs). More seriously, most of them are known carcinogens or probably carcinogenic to humans. The objectives of this paper were to report the regulatory aspects and environmental monitoring management of toxic VOCs designated by Japan and Korea under the Air Pollution Control Act, and the Clean Air Conservation Act, respectively. It can be found that the environmental quality standards and environmental monitoring of priority VOCs (i.e., benzene, trichloroethylene, tetrachloroethylene, and dichloromethane) have been set and taken by the state and local governments of Japan since the early 2000, but not completely established in Korea. On the other hand, the significant progress in reducing the emissions of some toxic VOCs, including acrylonitrile, benzene, 1,3-butadiene, 1,2-dichloroethane, dichloromethane, chloroform, tetrachloroethylene, and trichloroethylene in Japan was also described as a case study in the brief report paper.

Keywords: hazardous air pollutant; volatile organic compound; air quality monitoring; regulatory system; human carcinogen

1. Introduction

In the past decades, many studies have been focused on the non-carcinogenic health effects (e.g., respiratory disease or irritation) of non-criteria air pollutants in the atmospheric environment, which are mostly released from a variety of anthropogenic sources such as petrochemical facilities, motor vehicles, metal processing/finishing industries, gas stations, and energy sectors [1]. However, a very high incidence of leukemia and lung/liver cancers has been occurring in the urban environment between the developed and developing countries [2], showing a large number of carcinogenic air pollutants in the ambient air, including benzene, 1,3-butadiene, formaldehyde, vinyl chloride, perchloroethylene, and polycyclic aromatic hydrocarbons (PAHs). Among them, benzene may be the most notable environmental carcinogens because it has been classified by the International Agency for Research on Cancer (IARC) as the Group 1 carcinogen (confirmed as a human carcinogen) [3]. As a consequence, there is an additional control program to address human health concerns resulting from exposure to hazardous air pollutants (HAPs) other than the criteria air pollutants with ambient air quality standards since the late 1980s. Thereafter, the Clean Air Act Amendments of 1990 in the USA changed the focus from HAPs to the industry sectors emitting specific HAPs and the use of Maximum Achievable Control Technology (MACT) [4,5]. With these regulation changes, the control of HAPs has become much more effective in the USA. By contrast, the Air Quality Guidelines of the

World Health Organization (WHO), first set in 1987, were advised by the European Commission as a starting point for deriving limit values on HAPs [6,7].

Volatile organic compounds (VOCs) are generally referred to as the highly reactive and/or toxic organics emitted by both human-made and natural sources due to their high volatility at normal atmospheric conditions [8]. As a result, the definition varies among scientific organizations and official agencies in different countries, but they are characterized as organic compounds that have a relatively high vapor pressure. It should be noted that VOCs are known, or may be reasonably anticipated to pose a threat of adverse air quality and human health effects [9]. In the urban environment, toxic VOCs (e.g., gasoline) and solvents also easily react with nitrogen oxides (NO_x) in the presence of sunlight to form ozone (O_3) under a series of photochemical reactions, inducing a photochemical smog in the troposphere. More significantly, many VOCs are on the list of HAPs because they pose a threat of adverse human health effects, including cancer and respiratory illness [10]. As a consequence, some states in the USA have requested to develop their own HAPs programs on toxic VOCs (e.g., acrylonitrile, benzene, epichlorohydrin, ethylene dibromide, ethylene oxide, formaldehyde, and vinyl chloride), leading to ambient air levels (AALs) or ambient air guidelines [11].

In recent years, HAP management in the Western countries and the USA has been reviewed [6,12,13]. However, there has not been any literature addressing toxic VOC management in Asian countries. Therefore, this paper is a brief report about the regulatory aspects and environmental monitoring management of toxic VOCs designated as HAPs by Japan and Korea. Furthermore, this brief report will focus on the benzene, chlorinated VOCs, and other carcinogenic VOCs, in line with international concern about the carcinogenic risks of the VOCs in recent years. Moreover, the emission reduction of some designated VOCs in Japan is also described as a case study in this review paper.

2. Legislation on Toxic VOCs in Japan and Korea

Toxic VOCs have been detected in the ambient air at low concentration levels. People have become concerned about the effect of long-term exposure to such pollution on human health such as cancer and tumors. In contrast to the European countries and North America, there was a delayed response to HAPs in the Asian countries like Japan and Korea. Table 1 summarizes the regulations for HAPs in Japan and Korea under the Air Pollution Control Act and the Clean Air Conservation Act, respectively.

Table 1. Summaries of regulations for hazardous air pollutants (HAPs) in Korea and Japan.

Country	Japan	Korea
Central authority	Ministry of the Environment	Ministry of Environment
Relevant law/act	Air Pollution Control Act	Clean Air Conservation Act
Definition	Any substance that is likely to harm human health if ingested continuously and that is a source of air pollution.	Air pollutants that are feared to directly or indirectly inflict any harm or injury on the health and property of humans or on the birth and breeding of animal and plants.
List of HAPs	22 [a]	35 [b]
Relevant measures	- Enterprise shall take the necessary measure to determine the status of emission and discharge into the atmosphere of HAPs. - The State shall endeavor to implement studies in collaboration with local public entities in order to determine the status of air pollution by HAPs, and shall periodically make public the results of human health risk evaluation.	- The central authority shall install measuring networks and constantly measure the level of air pollution. - Permissible emission levels, reduction facility installation and operation, leakage monitoring, and maintenance standards will be applicable to each industry according to the facility management standards.

[a] They are required to take priority action. Among them, the environmental air quality standards for five HAPs (i.e., benzene, trichloroethylene, tetrachloroethylene, dichloromethane, and dioxins) have been established. [b] They were defined as specific hazardous air pollutants.

2.1. Japan

With the economic growth rate of over 10% during the period of 1960s, the Japanese experienced a strong energy demand until the oil crisis of 1973. In this regard, air pollution and degradation of environment were fired all around Japan during this period. This thus led to the promulgation of the Basic Law for Environmental Pollution Control in 1967. Thereafter, the Japanese government launched some legal enactments and revisions to enforce anti-pollution measures. Presently, the basic law governing air pollution from the emissions of soot, smoke, particulate, VOCs, hazardous air pollutants (see Table 1) and motor vehicle exhausts is the Air Pollution Control Act, which was initially passed in June 1968 and amended several times. Its main aim is to protect the health of citizens and the living environment from air pollution. Under the implementation of monitoring of the air pollution levels, it showed that air pollution is a serious environmental problem in Japan, particularly in urban areas and industrial zones [14]. Various carcinogenic air pollutants, such as benzene and chlorinated VOCs, have been detected in the ambient air in low concentrations. As a result, 22 substances are designated as priority HAPs for which measures should be taken with special action like emission monitoring and human health risk. Among them, there are 13 toxic VOCs as priority HAPs listed in Table 2.

Table 2. Summaries of toxic volatile organic compounds (VOCs) designated as HAPs in Korea and Japan. ● means that this VOC is desiganted as HAPs in Korea and/or Japan.

Toxic VOCs	Japan	Korea
Acetaldehyde	●	●
Acrylonitrile	●	●
Aniline		●
Benzene	●	●
1,3-Butadiene	●	●
Carbon tetrachloride		●
Chloroform	●	●
Chloromethyl methyl ether	●	
1,2-Dichloroethane	●	●
Dichloromethane	●	●
Dimethyl sulfide		●
Ethylbenzene		●
Ethylene oxide	●	●
Formaldehyde	●	●
Phenol		●
Propylene oxide		●
Styrene		●
Tetrachloroethylene	●	●
Trichloroethylene	●	●
Vinyl chloride	●	●

2.2. Korea

With rapid economic growth and urbanizations since the early 1970s, there was a great concern about the adverse effect of air pollutant on human health in Korea. The government began to set air quality standards for criteria pollutants as key policies under the authorization of the Clean Air Conservation Act. Air quality standards for sulfur dioxide (SO_2), carbon monoxide (CO)/nitrogen dioxide (NO_2)/total suspended particles (TSP)/ozone (O_3)/hydrocarbons, lead, benzene, and particulate matter ($PM_{2.5}$ and PM_{10}) were set in 1978, 1983, 1991, 1995, 2010, and 2011, respectively [15]. For example, benzene, known to be a human carcinogen (leukemia), exists as an ingredient in gasoline. Its air quality standard is set at 0.005 mg/m^3 based on annual average as shown in Table 3. It should be noted that benzene has been included into the National Emission Standards for Hazardous Air Pollutants (NESHAP) under the U.S. Clean Air Act Amendments of 1977.

In addition to criteria air pollutants, specific hazardous air pollutants and monitored hazardous air pollutants, defined in the Clean Air Conservation Act (seen in Table 1), have been of great concern in Korea because they could be carcinogenic to human and harmful to environmental quality. Herein, a "specific HAP" is a monitored HAP that may be harmful in the event of long-term consumption or exposure, even at low concentrations, and is deemed by committee evaluation to require atmospheric emission control. A "monitored HAP" is an air pollutant that may be harmful to human health or animal and plant growth and development, and is deemed by committee evaluation to require continuous measurement, monitoring, or observation. Accordingly, air quality management policies are shifting toward health-oriented risk and taking priority for public health. Currently, 35 substances were designated as specific hazardous air pollutants for special control and prevention [16]. Among them, 19 toxic VOCs are designated as specific HAPs in Korea as listed in Table 2. In order to reduce the health risk of carcinogenic VOCs from their fugitive emissions, prevention and control management standards for HAP-emitting facilities were enacted under the amendment of the Clean Air Conservation Act, which has been effective as of 1 January 2015 [15]. These facility management standards include permissible emission levels, reduction facility installation and operation, leakage monitoring, and maintenance standards [17].

Table 3. Environmental air quality standards for toxic VOCs in Japan and Korea.

HAPs	Environmental Air Quality Standards (Based on Annual Average)	
	Japan	Korea
Benzene	0.003 mg/m^3	0.005 mg/m^3
Trichloroethylene	0.2 mg/m^3	–
Tetrachloroethylene	0.2 mg/m^3	–
Dichloromethane	0.15 mg/m^3	–

– Not available.

3. Environmental Monitoring & Management

3.1. Japan

The Amendment of Air Pollution Control Act in 1996 required the environmental quality standards and environmental monitoring of priority HAPs by the state and local governments. Based on the carcinogenicity, physicochemical property, use, consumption, and monitoring data, benzene, trichloroethylene, tetrachloroethylene, and dichloromethane were first designated as HAPs for promotion of countermeasures and defined in the environmental quality standards (see Table 3). In compliance with the Air Pollution Control Act, local governments have monitored these toxic VOCs in the atmosphere in the past decade according to "the guideline for hazardous air pollutants monitoring" and the "manual for monitoring method of hazardous air pollutants" published by the Ministry of the Environment (MOE). To control the designated substances including benzene and trichloroethylene, the Ministry of Environment and the Ministry of Economy, Trade and Industry in Japan established a "Guideline for the Promotion of Voluntary Control of Hazardous Air Pollutants by Business Entities". Under this guideline, each industry group developed a nationwide and voluntary reduction plan in 2003. Results of the monitoring survey have been compiled by the MOE [18,19]. As shown in Table 4, the concentration levels of four toxic VOCs basically indicated a decreasing trend during this period. However, it can be found that their concentrations in ambient air fluctuated and even increased in recent years, which may be attributed to the strength of the emission source, the monitoring conditions, and locations.

Table 4. Environmental monitoring levels for toxic VOCs in the atmosphere over the past 10 years (2003–2012) in Japan [a].

HAPs	Environmental Air Quality Levels (Unit: μg/m^3) [b]									
	2003	**2004**	**2005**	**2006**	**2007**	**2008**	**2009**	**2010**	**2011**	**2012**
Benzene	1.9	1.8	1.7	1.7	1.5	1.4	1.3	1.1	1.2	1.2
Trichloroethylene	0.92	0.93	0.75	0.90	0.76	0.62	0.53	0.44	0.53	0.50
Tetrachloroethylene	0.38	0.38	0.28	0.31	0.25	0.23	0.22	0.17	0.18	0.18
Dichloromethane	2.4	2.6	2.1	2.8	2.3	2.3	1.7	1.6	1.6	1.6
Acrylonitrile	–	0.11	0.10	0.11	0.10	0.093	0.079	0.073	0.088	0.080
Chloroform	–	0.26	0.33	0.23	0.21	0.22	0.21	0.19	0.21	0.20
1,2-Dichloroethane	–	0.13	0.13	0.15	0.15	0.16	0.17	0.16	0.18	0.17
1,3-Butadiene	–	0.26	0.22	0.23	0.19	0.18	0.16	0.14	0.15	0.14

[a] Sources [16,17]. [b] Based on annual average concentration. – Not available.

Furthermore, in response to over thousands of toxic substances commonly used in Japan, the government implemented state and local public entity policies and measures under the Chapter II-4 ("Promotion of Countermeasures of Hazardous Air Pollutants") of the Air Pollution Control Act, including environmental monitoring guideline, health risk evaluation, and guideline values defined for other HAPs. Currently, there are four toxic VOCs for which guideline values are specified as a guide to reduce health risks resulting from HAPs in the atmosphere, especially in industrial zones. Table 4 also shows the monitoring results of these toxic VOCs for which the guideline values established [18,19]. However, it can be seen that the concentrations of 1,2-dichloroethane slightly increased, possibly due to its extensive use in the industrial sector.

3.2. Korea

As described above, Korea has set air quality standards for key air pollutants as policy objectives on air quality control and has been making efforts to satisfy these standards. In order to understand the actual air quality trend in national levels, about 200 air quality monitoring stations have been installed by each environmental management office and each city and province in urban areas or nearby industrial complex [17]. During the past two decades, all sewage treatment facilities in Korea emit significant VOCs (e.g., toluene, chlorinated hydrocarbons) from the liquid surface [20,21]. In order to reduce the fugitive emissions of toxic air pollutants, facility management standards for VOC-emitting facilities (including laundry shops, printing, painting facilities and gas stations) were enacted and have been effective as of 1 January 2015.

In Korea, the environmental policy for air quality management has recently shifted to a focus on pollution prevention and health risk-oriented management [15]. Accordingly, the Clean Air Conservation Act will be tentatively improved to reclassify HAPs into monitored HAPs (97 compounds) and specific HAPs (38 compounds, as compared to 35 compounds in Table 1) according to the following indices: toxicity, environmental and physicochemical properties, impact on ecosystems, atmospheric emission volume, ambient concentration level, and international regulations. These classifications are required to be designated by an evaluation committee. To identify the contamination state by specific HAPs in urban areas or nearby industrial complex, several organic toxics, including 13 types of VOCs and 7 types of PAHs [16,17], have been measured by 31 stations in the HAPs monitoring network. The monitored VOCs are acetaldehyde, acrylonitrile, benzene, 1,3-butadiene, carbon tetrachloride, chloroform, ethylbenzene, ethyl dichloride, formaldehyde, propylene oxide, styrene, tetrachloroethylene, and trichloroethylene.

4. Conclusions

HAPs, also called air toxics, represent a designated classification for harmful substances of anthropogenic emission sources that exist in measurable quantities in the atmospheric air, and are

defined under the laws and acts of developed countries. In this paper, the legislation on toxic VOCs designated as HAPs by Japan and Korea were reviewed. It can be found that the environmental quality standards and environmental monitoring of priority VOCs (i.e., benzene, trichloroethylene, tetrachloroethylene, dichloromethane, acrylonitrile, chloroform, 1,2-dichloroethane, and 1,3-butadiene) have been set and taken by the state and local governments of Japan since the early 2000s, but not completely established in Korea. In line with the international concern about the carcinogenic risk, the results of the environmental monitoring of 8 designated VOCs in Japan were described and discussed as a case study. It was found that the monitoring data indicated a decreasing trend during this period. However, it can be found that their concentrations in ambient air fluctuated and have even increased in recent years.

In the past decades, cancer remains the most common cause of death in developed countries. A number of environmental factors have been implicated in the inductions of human cancer. However, environmental and occupational exposure to toxic VOCs (e.g., benzene, formaldehyde, and chlorinated hydrocarbons) and toxic metals (e.g., beryllium, cadmium, chromium, and nickel), especially those that are airborne, is indicative of the confirmed evidence of human carcinogenicity. As a result, the regulatory and voluntary actions to reduce stationary emissions and further study the relationship between human health risk and long-term exposure to their atmospheric concentrations should be performed. Furthermore, an advanced new concept, like the maximum available control technology (MACT) in the USA, will be able to lessen the emissions of fugitive HAPs from the petrochemical factories and refineries.

Conflicts of Interest: The author declares no conflict of interest.

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