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Technical Information

The Potential Contribution of VOCs on Ambient Air Odor

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ABSTRACT In order to evaluate the potential contribution of VOCs on ambient air odor in roadside, urban, suburban and mountainous environments, the odor activity values (OAVs) of 66 VOCs were calculated using the environmental concentrations and the odor detection threshold values. The OAVs were less than one, but were not small, considering previous reports on odor concentrations in urban ambient air. The OAVs from roadside and urban areas were nearly identical, indicating that automobile exhaust gas does not have an important role in the potential odor. Acetaldehyde, which is directly emitted from anthropogenic sources and is also formed as a byproduct of photochemical reactions in the air, appears to be the most important substance contributing to the potential odor in each area. We also found that the contribution of biogenic VOCs was large in mountainous regions. In order to determine more precise OAVs, further work is required to generate more reliable odor detection threshold values.

KEY WORDS Environmental odor, Odor concentration, Odor activity value, Odor detection threshold, Acetaldehyde

1. INTRODUCTION

The ambient air from areas where there are no specific factories or industries has certain odors even though humans cannot perceive them clearly. Even if the odor concentration, (i.e. the number of dilutions with neutral air required to reach the odor detection threshold), is not measureable by normal dilution methods such as the triangle odor bag method (Iwasaki, 2003) or dynamic olfactometry (EN13725, 2003), it may still impact air quality.

The measurement of low odor concentrations in ambient air has been attempted in Japan. Tatsuichi *et al.* (1992) measured odor concentrations of 1.1–1.5 in residential suburbs and 7.4–9.8 in roadside areas utilizing the Japanese triangle odor bag method with preconcentration using three adsorbents and thermal desorption. These results demonstrated the contribution of vehicle exhaust gas to the odor concentrations. Tatsuichi and Ueno (1997) measured the odor concentrations in five districts in Tokyo. They also conducted a questionnaire survey of the residents, focused on the pleasantness of the air, which revealed that the residents found the air unpleasant if the odor concentration was above 2.5. Masuda *et al.* (2008, 2004) reported odor concentrations of 0.3–5.6 in Osaka city with higher odor concentra-

tions at roadside areas. They utilized cryogenic condensation conjunction with the triangle odor bag method to quantify the odor concentrations. Cryogenic condensation effectively traps more odorous substances compared to the trapping by the absorbents.

Specific VOCs may contribute to low odor concentrations. Masuda *et al.* (2008) suggested that acetaldehyde was the most significant odor substance in the urban atmosphere according to the VOC concentrations and the odor detection thresholds (Nagata, 2003). Seo *et al.* (2014) measured VOC concentrations in the ambient air from industrial and residential areas of South Korea. They found that specific aldehydes had higher odor impact, based on the values calculated by dividing the observed concentrations with the standard odor intensity concentrations.

In this study, we examined the potential contribution of VOCs to odor in urban, roadside, suburban and mountain air in Tokyo. This analysis was performed by evaluating the odor activity values (OAVs) calculated based on the VOC concentrations and the odor detection threshold values. Since the VOC concentrations comprise the 24-hr average, the ambient air odor discussed in this study correspond to the background odor level of the area.

2. MATERIALS AND METHODS

2.1 VOC Data

VOC concentrations in ambient air were obtained from the Bureau of the Environment, Tokyo Metropolitan Government (Bureau of the Environment, 2019, 2015). The data from Fiscal year (FY) 2013 and FY2017 were used, because the quality of the measurements, which depends on the analytical equipment, was consistent during these periods. VOC measurements with 24-hr sampling were performed once a month at 13 sites in Tokyo, including urban, suburban, roadside and mountainous areas (Fig. 1). Air samples were collected in a canister for most of the VOCs and analyzed by gas chromatography with flame ionization detection (GC-FID) for low volatile compounds (C_2-C_4 hydrocarbons), and gas chromatography-mass spectrometry (GC-MS) for other compounds. Aldehydes samples were collected with a 2, 4-dinitrophenyl hydrazine (DNPH) cartridge and extracted with acetonitrile followed by HPLC analysis. At each sampling point, air pollutants such as ozone and NO_x were continuously monitored.

2.2 Odor Detection Threshold

The odor detection threshold values used in this study were measured using the triangle odor bag method (Iwasaki, 2003). The following three Japanese institutes quantified the odor detection threshold values; the Japan Environmental Sanitation Center (Nagata, 2003), the Tokyo Metropolitan Research Institute for Environmental Protection (Iwasaki, 2004) and the Hiroshima Prefectural Research Center for Environmental Science (Matsusita *et al.*, 1984; Ito *et al.*, 1982). The odor detection threshold value depends on the sensitivity of the assessors who participated in the olfactory measurements. The three institutes used six different assessors. To reduce odor detection threshold value variability, we used the geometric mean in case of multiple data for each compound.

2.3 OAV

The OAV is defined as the ratio of a compound's concentration to the odor detection threshold. The OAV has been widely used to evaluate the relative contribution of

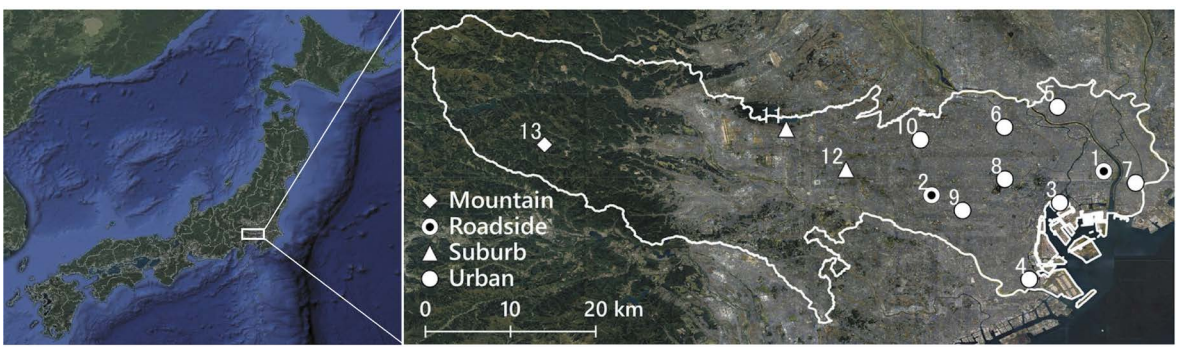

Fig. 1. Location of sampling sites in Tokyo, Japan.

odorous compounds to odor concentration (Rincón *et al.*, 2019; Capelli *et al.*, 2008). When only a single substance is present in the air, the OAV corresponds to the odor concentration. In the case of gas mixtures, the sum of individual OAVs (OAV_{sum}) and the maximum OAV (OAV_{max}) can be used for odor assessment.

In this study, the OAVs of 66 VOC compounds (alkane and cycloalkane: 24, alkene: 3, biogenic: 5, aromatic: 15, aldehyde: 2, ketone: 3, oxygenate: 7, halide: 5, others: 2), which have the odor threshold values, were calculated. The compounds and the corresponding odor detection threshold values are listed in Table 1.

3. RESULTS AND DISCUSSION

3.1 OAV

The OAVs (calculated from the annual average VOC concentrations) and NO_x concentrations at each site as well as the corresponding mean values from the different areas are shown in Table 2. Both OAV_{max} and OAV_{sum} values are less than 1, but not significantly small. Although there are no recent reports of ambient air odor concentration values, a previous study (Masuda *et al.*, 2008) measured the odor concentration of ambient air using the triangle odor bag method with cryogenic preconcentration. They reported that the odor concentrations at 26 points in Osaka, Japan varied from 0.6 to 3.8. This report also found that the odor concentrations tended to be higher at roadside areas and lower at suburban regions. Osaka is a densely populated city, comparable to Tokyo, Japan. Considering the data, the OAVs values in Table 2 imply that the VOC odor is not clearly perspective by humans, but may contribute to the odor of ambient air.

Roadside area OAVs are nearly identical to the urban area OAVs as shown in Table 2. However, NO_x concentrations from roadside regions are higher than those from urban areas, indicating that VOCs from automobiles have small direct effect on the OAVs. Previous studies (Masuda *et al.*, 2008; Tatsuichi and Ueno, 1997) showed high odor concentrations at roadside areas. This suggests that the recent improvement of treatment systems for vehicle exhaust gas has effectively reduced the emission of odorous substances. On the other hand, the OAVs and NO_x concentrations are the highest in urban regions followed by suburban areas and are the lowest in mountainous locations. This implies that higher total anthropogenic activity results in higher OAVs, although more data is required to substantiate this hypothesis.

Table 2 shows that site No.4 has the highest OAV, even though the NO_x concentration is not especially high. In this area, there are many small factories which use organic solvents. This may explain the high OAV at the site No. 4 compared to other urban areas.

OAVs from 2017 are smaller than those from 2013 in all of the tested regions due to decreased VOC concentrations in ambient air. This was due to the combined efforts of various governments and industries whose goal was to reduce VOC and NO_x concentrations in order to reduce ozone and particulate matter in the air. These efforts also contributed to the reduction in potential odor at all the sites shown in Table 2.

3.2 VOC Compounds

The top ten VOC compounds from ambient air samples surveyed in 2017 were ranked according to their OAV and are shown in Fig. 2. Acetaldehyde was the largest contributor to the potential odor, comprising approximately 75% of the OAV_{sum} in each area. The compounds composition in roadside and urban areas was nearly identical. Aside from acetaldehyde, there were butyl alcohols, butyl acetate and aromatic compounds such as toluene. These are common organic solvents that are widely used in many industries. Formaldehyde was observed as well as acetaldehyde. Several biogenic VOCs including α-pinene and isoprene were observed. The α-pinene OAV was larger in suburban and mountainous locations. The data indicate that biogenic VOCs potentially contribute to odor emissions in areas with less human activity, and may play a role in air quality.

The high acetaldehyde OAVs shown in Fig. 2 are due to their relatively high concentrations (2–4 ppb) and low odor detection threshold values [6.4 ppb as a geometric mean of 1.5 ppb (Nagata, 2003) and 27 ppb (Iwasaki, 2004)]. Masuda *et al.* (2008) also found that acetaldehyde had the largest OAV using the smaller threshold of 1.5 ppb. They suggested that the high acetaldehyde OAV was due to automobile exhaust gas because the odor concentrations at roadside areas were high and the maximum emission source of acetaldehyde was estimated to be automobile exhaust gas (MOE, 2005). According to the Pollutant Release and Transfer Register (PRTR), in 2017 (MOE, 2017) the estimated emission of acetaldehyde from mobile sources in Tokyo was 113 t/year, while that from households was 36 t/year and that from stationary sources was only 2.9 t/year. However, in our

Table 1. VOCs and the odor detection threshold values used in this study.

Compound	Threshold[1] (ppm)		Compound	Threshold[1] (ppm)	
Alkane, Cycloalkane					
Propane	1500	a	Ethylbenzene	0.17	a
n-Butane	1200	a	m,p-Xylene[2]	0.37	a,b
n-Pentane	4.1	a,b	o-Xylene	0.34	a,b
iso-Pentane	1.4	a,b	Styrene	0.035	a
n-Hexane	5.0	a,b	m,p-Ethyltoluene	0.015	a,b
2,2-Dimethylbutane	45	a,b	1,3,5-Trimethylbenzene	0.31	a,b
2-Methylpentane	12	a,b	1,2,4-Trimethylbenzene	0.12	a
2,3-Dimethylbutane	0.42	a	1,2,3-Trimethylbenzene	0.032	b
3-Methylpentane	8.9	a	Isopropylbenzene	0.0078	a,b
2,4-Dimethylpentane	1.1	a,b	n-Propylbenzene	0.0061	a,b
Methylcyclopentane	3.1	a,	o-Ethyltoluene	0.086	a,b
2-Methylhexane	0.50	a,b	m-Diethylbenzene	0.033	a,b
2,3-Dimethylpentane	2.4	a,b	p-Diethylbenzene	0.00068	a,b
3-Methylhexane	0.84	a	Aldehyde		
Cyclohexane	8.7	a,b	Formaldehyde	0.50	a
2,2,4-Trimethylpentane	0.67	a	Acetaldehyde	0.0064	a,b
n-Heptane	4.4	a,b,c	Ketone		
Methylcyclohexane	0.87	a,b	Acetone	42	a
2-Methylheptane	0.36	a,b	Methyl ethyl ketone	0.44	a
3-Methylheptane	3.9	a,b	Methyl isobutyl ketone	0.17	a
n-Octane	3.0	a,b	Oxygenate		
n-Nonane	2.0	a,b	Isopropyl alcohol	26	a
n-Decane	1.6	a,b	Methylacetate	1.7	a
n-Undecane	0.62	a	n-Propylalcohol	0.094	a
Alkene			Ethylacetate	0.87	a
1,3-Butadiene	0.23	a	Isobutylalcohol	0.011	a
1-Butene	0.36	a	n-Buthylalcohol	0.038	a
1-Pentene	0.1	a	Butylacetate	0.016	a
Biogenic			Halide		
Isoprene	0.048	a	Dichloromethane	160	a
α-pinene	0.017	a,c	Chloroform	3.8	a
Camphene	0.88	c	Carbon tetrachloride	4.6	a
β-pinene	0.035	a,c	Trichloroethylene	3.9	a
Limonene	0.057	a,c	Tetrachloroethylene	0.77	a
Aromatic			Others		
Benzene	1.9	a,b	Propylene	17	a,c
Toluene	0.33	a	Acrylonitorile	15	a,c

[1]Geometric mean if there are more than one value. a: Nagata, 2003; b: Iwasaki, 2004; c: Ito *et al.*, 1982 and Matsusita *et al.*, 1984.
[2]Average of m-Xylene and p-Xylene.

studies, the OAVs at roadside locations were not significantly higher than those from urban residential areas (Table 2). Moreover, NO_x concentrations at roadside locations were significantly higher than those at urban areas (Table 2). Therefore, these OAVs may not only be due to automobiles, but may also originate from the following (1) industrial activity (which is not estimated in the PRTR); (2) secondary formation of acetaldehyde in the air (Seo *et al.*, 2014) or (3) unreliable odor detection threshold values (Masuda *et al.*, 2008).

Ishii *et al.* (2014) suggested that formaldehyde emission in Tokyo originated from combustion-related and non-combustion-related facilities such as boiler or wood factories. Acetaldehyde may also be emitted from unin-

Table 2. Annual average OAVs and NO_x concentrations at each site as well as the area corresponding mean values from the different areas.

	Site No.	2013			2017		
		OAV_{max}	OAV_{sum}	NO_x (ppb)	OAV_{max}	OAV_{sum}	NO_x (ppb)
	mean	0.29	0.48	48	0.25	0.33	37
Roadside	1	0.28	0.46	37	0.23	0.31	32
	2	0.31	0.50	59	0.27	0.35	42
	mean	0.29	0.48	26	0.24	0.31	24
	3	0.27	0.38	30	0.25	0.31	28
	4	0.35	0.55	30	0.33	0.42	27
	5	0.30	0.50	28	0.26	0.34	25
Urban	6	0.28	0.55	28	0.21	0.29	25
	7	0.28	0.39	25	0.23	0.30	24
	8	0.27	0.37	25	0.22	0.29	21
	9	0.30	0.50	21	0.27	0.34	19
	10	0.25	0.44	21	0.19	0.25	19
	mean	0.19	0.38	17	0.20	0.27	15
Suburb	11	0.19	0.37	17	0.20	0.27	16
	12	0.19	0.39	17	0.21	0.27	14
Mountain	13	0.15	0.22	4	0.14	0.19	4

Site No. are indicated in Fig. 1.

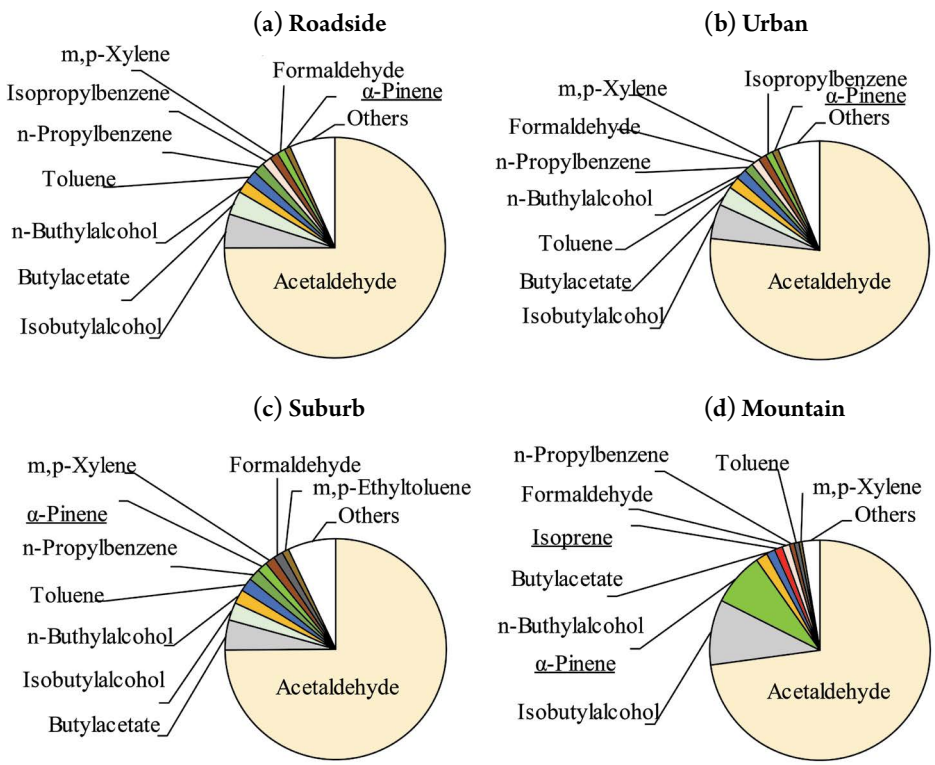

Fig. 2. Top ten VOC compounds from an ambient air survey conducted in 2017 that are ranked according to their OAV. Underlined substances indicate biogenic VOCs.

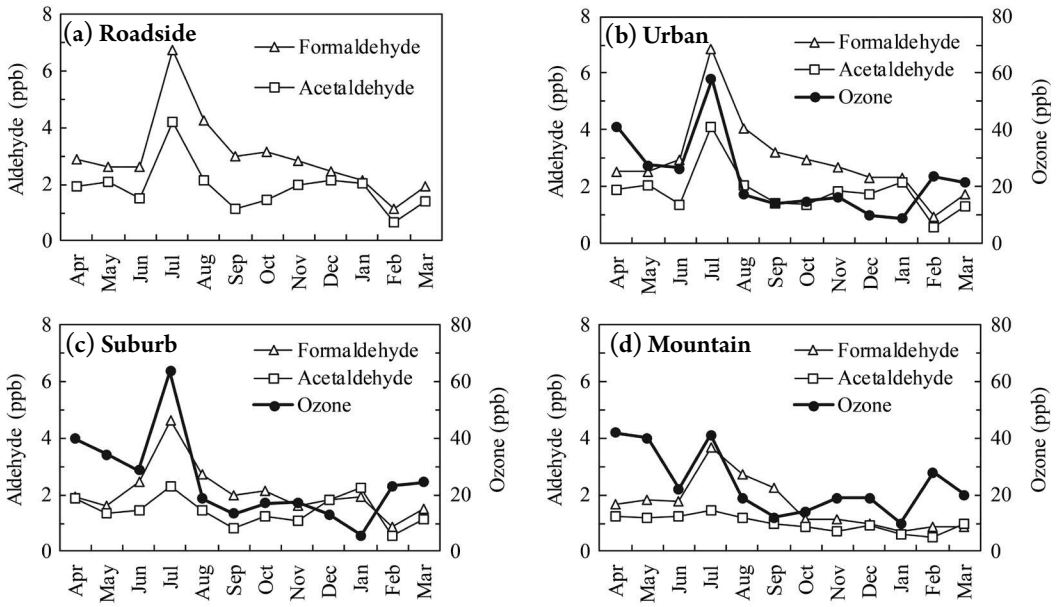

Fig. 3. Monthly data of formaldehyde, acetaldehyde and ozone concentrations in fiscal year 2013. Ozone was not observed at roadside sites.

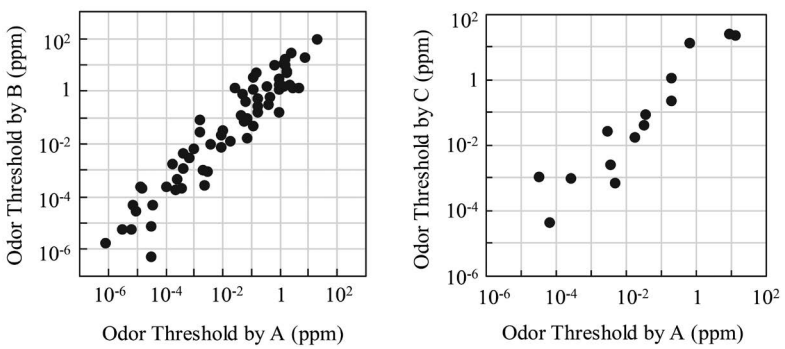

Fig. 4. Comparison of odor detection threshold values measured by three institutes. A: Japan Environmental Sanitation Center (Nagata, 2003); B: Tokyo Metropolitan Research Institute for Environmental Protection (Iwasaki, 2004) and C: Hiroshima Prefectural Research Center for Environmental Science(Matsusita *et al.*, 1984; Ito *et al.*, 1982).

tentional sources. For example, in painting facilities, acetaldehyde is emitted during the drying process. Some amount of acetaldehyde may not be fully estimated by the PRTR.

Aldehydes are known to be byproducts of photochemical reactions in the air (Sakamoto *et al.*, 1993). Fig. 3 shows monthly data of formaldehyde, acetaldehyde and ozone concentrations from FY2013 during a typical summer peak of ozone was observed. In July, aldehydes and ozone concentrations were high. Ozone is produced by photochemical reactions, providing further evidence that aldehydes are also byproducts of photochemical reactions. According to the data from suburban and mountainous areas, formaldehyde is more likely to be formed than acetaldehyde. This tendency is consistent with a previous study (Ling *et al.*, 2016). Taken together, these results imply that acetaldehyde is directly emitted from industrial sources and is also formed as a byproduct of photochemical reactions.

3.3 Reliability of the Odor Detection Threshold

The OAV calculation is based on the assumption that the odor detection threshold is reliable. There are several compilations of odor detection thresholds. Ruth (1986)

and U.S. EPA (1992) reported acetaldehyde odor detection thresholds of 0.1–2200 ppb and 6.7–390 ppb, respectively. It is difficult to use these numbers for the assessment of odorous substances because of the significant variation of three or five orders of magnitude. The discrepancy is due to the variability in both the measurement methods and human olfactory responses (Cain and Schmidt, 2009). Different institutes measured the odor threshold values in this study but they all used the same method. Fig. 4 shows the correlations of odor detection threshold values. There are good correlations in the double logarithmic plots. However, there are one order of magnitude differences in some compounds. The odor detection thresholds of acetaldehyde are 1.5 ppb determined by the Japan Environmental Sanitation Center (A) and 27 ppb determined by the Tokyo Metropolitan Research Institute for Environmental Protection (B). In this study, we utilized the geometric mean calculated from the measurements of the acetaldehyde odor detection threshold by A and B, which was found to be 6.4 ppb. If 27 ppb is used for the calculation of the OAV, the ratio of approximately 75% for acetaldehyde shown in Fig. 2 changed to approximately 40%, suggesting that acetaldehyde is still the most important substance contributing to potential odor in ambient air.

These odor detection threshold values were mainly measured in 1980s. After the 1990s, quality control of both sensory and instrumental analyses has been improved through amendments of the Japanese Offensive Odor Control Law and the Japanese Air Pollution Control Act. Thus, the reliability of the odor detection threshold values should be ensured by acquiring the data from quality-certified analytical institutes.

3.4 Other Compounds

There may be other compounds that potentially contribute to the odor of ambient air. Sulphur compounds are possible candidates, due to their low odor detection threshold values. High concentrations of hydrogen sulfide were reported in a few urban environments in Greece (Kourtidis *et al.*, 2008). Masuda *et al.* (2008) measured sulphur compounds at 26 urban locations in Osaka city. They reported that the OAVs were not high compared to the acetaldehyde OAVs except for a few areas, one of which was close to a sewage treatment plant. The effects of sulphur compounds on potential odor are not so high in Tokyo because environment is similar to Osaka. However, compounds with low odor detection thresholds should be taken into account in this kind of research.

4. CONCLUSIONS

VOC OAVs calculated by dividing the environmental concentrations with odor detection thresholds imply that VOCs might contribute to the odor of ambient air. Acetaldehyde, which is not only emitted from anthropogenic sources but also produced secondarily in the air, appeared to be the most important substance potentially contributing to odor of ambient air. In addition, biogenic VOCs may have important roles in air quality in mountainous areas. To assess these issues more precisely, accumulation of odor detection threshold values measured by quality-certified analytical institutes is necessary.

ACKNOWLEDGEMENT

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