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CONTENTS

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Characteristics of odorous carbonyl compounds in the ambient air around a fishery industrial complex of Yeosu, Korea

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Abstract

In this study, the amounts of odorous carbonyl compounds (OCCs) including acetaldehyde (Acet-A), propionaldehyde (Pron-A), butylaldehyde (Buty-A), iso-valeric aldehyde (Iso-Vale-A) and *n*-valeric aldehyde (N-Vale-A) emitted from a fishery industrial complex near the exhibition facilities of "Expo 2012 Yeosu Korea" were measured. Acet-A was found to be the most abundant OCC, and the total concentrations of the OCCs were the highest in the summer. However, due to vehicular exhaust and photochemical reactions, the concentrations of some of the OCCs presented their highest levels in the fall. A significant correlation between Acet-A and Buty-A was found at the major fishery facilities (r = 0.816, p = 1.87E-15, n = 60) and at the border areas (r = 0.809, p = 3.40E-12, n = 48) of this fishery industrial complex. The concentrations of OCCs at the border areas were not worse than those at the urban areas in other places, indicating that the concentrations of ambient OCCs at the border areas were not greatly influenced by manmade activities.

Key words: fishery industrial complex; odorant; carbonyl compounds; acetaldehyde; seasonal variation

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Introduction

Carbonyl compounds, including aldehydes and ketones in the atmosphere, are important in atmospheric chemistry (Atkinson, 2000; Bakeas et al., 2003; Huang et al., 2008). Their diffusion in the air causes adverse health effects such as eye and lung irritations (WHO, 2000; Weng et al., 2009; Báez et al., 1995) and can be the primary cause of odorant problems (Dincer and Muezzinoglu, 2006; Kim et al., 2008). Therefore, carbonyl compounds have been investigated in many places around the world in urban, rural, indoor, outdoor, industrial and non-pollution areas (Gallego et al., 2009; Guo et al., 2004; Pang and Mu, 2006; Wang et al., 2010; Pang and Lee, 2010; Santarsiero and Fuselli, 2008; Feng et al., 2010; Chung et al., 2011; Wang et al., 2007). Yeosu is a city in the southern coast of Korea. A great deal of carbonyl compounds has been emitted from a fishery industrial complex at Yeosu, and many complaints about odorants from this industrial complex have been reported (Seo, 2007). Yeosu, on the other hand, will host the World Exposition 2012 under the theme of 'The Living Ocean and Coast', so the ambient air quality in Yeosu has become one of the most sensitive issues, attracting much governmental and public concern. To comply with the social and environmental demands to resolve the odorant-related problems, the fishery industrial complex is trying to control the emissions of odorous carbonyl compounds (OCCs).

This work aims are: (1) to quantify the atmospheric concentrations of OCCs and their seasonal variations at the sampling sites; (2) to identify the major OCCs by using the odorant quotient (OQ); (3) to find the potential relationship among the OCCs, (4) to compare the concentrations of OCCs with those reported in other previous studies.

1 Materials and methods

1.1 Sampling sites and periods

Samples were collected from the major fishery facilities and the border areas in a fishery industrial complex during spring, summer and fall, twice in each season for a total of 6 times in 2007. The related meteorological conditions at the sampling sites are presented in our previous study (Seo et al., 2011). The fishery industrial complex is located in a northeastern area of Yeosu City near the exhibition facilities of "Expo 2012 Yeosu Korea", shown in Fig. 1. Prior to sampling, a field survey was carried out, and five

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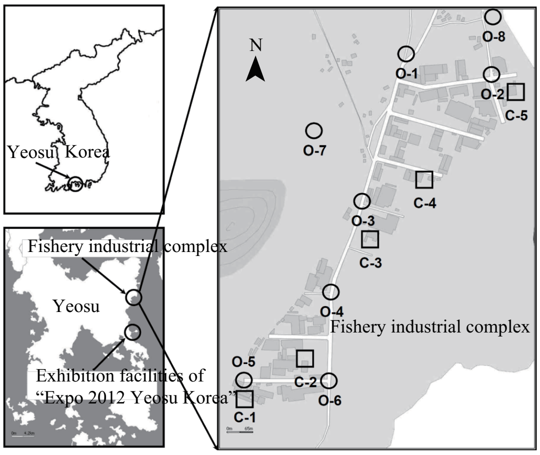

Fig. 1 Geographical location of the study area and sampling sites (C1–C5, O1–O8).

typical facilities (from C-1 to C-5) that showed higher odorant annoyance levels than the other facilities by the direct olfactory method were selected. At each major facility, two typical sites were also selected, for a total of 10 sampling sites at the five major fishery facilities. In the border areas, the sampling sites were divided into six border (from O-1 to O-6) sites and two complaint (O-7 and O-8) sites, which were the main crossroads and the main complaint sites in this industrial complex, respectively. The information on the general characteristics of the selected sampling sites is provided in Table 1.

1.2 OCCs sampling and analysis

In this study, OCCs, including acetaldehyde (Acet-A), propionaldehyde (Pron-A), butylaldehyde (Buty-A), iso-valeric aldehyde (Iso-Vale-A) and *n*-valeric aldehyde (N-Vale-A), which are Offensive Odorant Substances designated by the Korean Ministry of Environment (KMOE, 2007), were selected for analysis. For sample collection, a Personal Air Sampler (Gilian, USA) and an Electronic Flow Meter (Aalborg, USA) were used. The samples were flowed at 2 L/min for 10 min, and varied by less than 5% for all samplings. We used a 2,4-DNPH cartridge (S10, Supelco, USA), which is a 4-cm polypropylene tube coated with highly pure 2,4-DNPH. An ozone scrubber (Waters, USA) filled with KI was connected to the front part of 2,4-DNPH cartridge to prevent ozone interference. When the samples were collected, they were individually stored in containers, which were coated inside with aluminum, and stored below 4°C until solvent extraction. All the experimental apparatuses used for sample extraction and analysis were washed with the extraction solvent, HPLC-grade acetonitrile (J.T. Baker, USA). All the experimental apparatuses were dried at 60°C for more than 30 min, and purged with highly pure N_2 (99.999%). A vacuum elution rack (Supelco, USA) was used as the solvent extraction device and fixed with a 2,4-DNPH cartridge inside. The samples were extracted at a very low speed (1 mL/min) using 5 mL of acetonitrile, which had been filtered more than three times with a fat soluble filter (Φ47 mm, 0.45 μm, PTFE). Finally, the samples were analyzed by HPLC (Younglin, Korea) coupled to a UV detector, whose detection wavelength was 360 nm. All of the methods are recommended by the Korean Odorant Analysis Standards (NIER, 2007). In this study, two types of indices are used to evaluate the relationships among the OCCs: (1) the sum of odorant concentrations (SOCs), which depends on the concentration of each odorant (ppb) as measured by instrumental analysis and (2) the sum of odorant quotients (SOQs), which depends on the quotient of the concentration of each odorant (ppb) and the threshold limit value of each odorant (ppb).

1.3 Quality assurance (QA)

The basic quality assurance (QA) of our experimental method was performed in terms of method detection limit (MDL) and relative standard error (RSE) for all of the OCCs. MDL values were estimated by multiplying the standard deviation (SD) by a factor of 3.14, and RSE values were computed based on the seven repeated analyses of standards, as discussed in our previous study (Seo et al., 2011). In this study, the MDL values ranged from 0.06 ppb (Iso-Vale-A and N-Vale-A) to 0.15 ppb (Acet-A) while the RSE values ranged from 0.28% (Acet-A) to 0.86% (Buty-A).

Table 1 General characteristics of the selected sampling sites in this study

Index	Sampling sites	Locations of the sampling sites
Major fishery facilities	C-1: feed manufacturing	C-1-1: fresh fish treatment process
		C-1-2: crush, product depository
	C-2: fish oil manufacturing	C-2-1: maintenance room
		C-2-2: feed store room
	C-3: fish meat packing industry	C-3-1: wrapping room
		C-3-2: frozen food workshop
	C-4: fish meat packing industry	C-4-1: wrapping room
		C-4-2: materials workshop
	C-5: wastewater treatment plant	C-5-1: flow regulation set
		C-5-2: raw water tank
Border areas	O-1: crossroad-1	O-1: the main entrance of this fishery industrial complex
	O-2: crossroad-2	O-2: the crossroad near C-5
	O-3: crossroad-3	O-3: the crossroad near C-3
	O-4: crossroad-4	O-4: the intermediate zone of this industrial complex
	O-5: crossroad-5	O-5: the crossroad near C-1
	O-6: crossroad-6	O-6: the crossroad near C-2
	O-7: complaint site-1	O-7: the main complaint site located at the northern of this fishery industrial complex
	O-8: complaint site-2	O-8: the main complaint site located at the western of this fishery industrial complex

2 Results and discussion

2.1 Concentrations of OCCs in ambient air

Seasonal distributions and statistical summaries of the five OCCs are listed in Table 2. Because formaldehyde is not designated as an Offensive Odorant Substance by KMOE (KMOE, 2007) and JMOE (JMOE, 2005), it does not appear as an analysis item in this study. Excluding formaldehyde, Acet-A was the most abundant OCC in ambient air with mean concentrations of 12.0 and 5.93 ppb at the major fishery facilities and at the border areas, respectively. This finding was in good agreement with the results of other previous studies of industrial areas in Korea (Kim and Park, 2008) and urban areas in the high-air-pollution days in China (Huang et al., 2008), which recorded Acet-A concentrations of 130 and 10.6 ppb, respectively. At the major fishery facilities, all of the OCCs were present in all sampling periods, but Iso-Vale-A in summer and fall and N-Vale-A in spring were not detected at the border areas.

Most of the OCCs concentrations in the summer were higher than those of any other season. However, the mean concentration of Acet-A at the border areas in the summer was lower than that in the fall. Pal et al. (2008) reported the concentration of Acet-A as 29.4 ppb in the fall, and as 18.5 ppb in the summer around a highly industrialized area. Similar Acet-A concentrations were also reported by Pang and Mu (2006), who found that the concentrations of Acet-A were 7.83 and 8.99 ppb in the summer and fall, respectively. Meanwhile, the concentrations of OCCs at O-1–O-6, which are the crossroads of high traffic density, were mainly the result of photochemical reaction in the summer and vehicular exhaust in the fall.

2.2 Identification the major OCCs by SOQs

Figure 2 shows the comparisons of SOQs of the OCCs at the major fishery facilities and border areas, respectively. The mean SOQs values were 31.5 and 8.36 at these two areas, respectively. For the major fishery facilities, the OQ of Acet-A (25.4%) was the highest, followed by those of Iso-Vale-A (22.9%), Buty-A (18.6%), N-Vale-A (17.5%) and Pron-A (15.6%), and at the border areas, the OQ of Acet-A (47.2%) was the highest, followed by those of N-Vale-A (22.8%), Buty-A (15.6%), Iso-Vale-A (8.4%), and Pron-A (6.0%). In addition, Acet-A was found in the highest proportion at each of the sampling sites, indicating that Acet-A was the major OCC at this fishery industrial complex.

The SOQs of site C-1-1 was 99.8, thus site C-1-1 was the major site of OCCs emission. However, site O-5, which is the sampling site in the border area nearest to site C-1-1, showed the lowest SOQs of 4.8. These results suggest that the OCCs emitted from the major fishery facilities do not have a big effect on the OCCs concentrations of the surrounding environment. Particularly, among the sites in the border areas, site O-7, which was a complaint site, showed the highest SOQs of 15.0; this is even higher than the SOQs of sites C-3 and C-4. This result indicates that photochemical reactions under strong solar radiation (Pal et al., 2008) or meteorological conditions (temperature, wind speed, and rainfall) (Andreini et al., 2000) can influence the concentrations of OCCs.

Table 2 Concentration (ppb) of OCCs in the ambient air of the fishery industrial complex

Compound	Spring (Ari. mean ± SD)	Summer (Ari. mean ± SD)	Fall (Ari. mean ± SD)	Min	Max	Ari. mean
Major fishery facilities						
Acet-A	7.34 ± 20.2	17.9 ± 23.3	10.9 ± 9.74	N.D.	88.0	12.0
Pron-A	3.75 ± 4.48	8.90 ± 11.0	2.08 ± 2.07	N.D.	35.7	4.91
Buty-A	1.42 ± 3.02	7.90 ± 13.7	2.45 ± 1.62	N.D.	50.8	3.92
Iso-Vale-A	0.14 ± 0.51	1.25 ± 2.29	0.77 ± 1.89	N.D.	8.20	0.72
N-Vale-A	1.01 ± 1.80	4.70 ± 8.96	1.06 ± 1.36	N.D.	33.1	2.26
Border areas						
Acet-A	1.43 ± 0.76	7.19 ± 3.21	9.17 ± 5.79	0.21	18.0	5.93
Pron-A	0.59 ± 0.88	0.82 ± 0.34	0.08 ± 0.23	N.D.	2.50	0.50
Buty-A	0.53 ± 0.41	1.08 ± 0.52	1.00 ± 0.57	N.D.	1.98	0.87
Iso-Vale-A	0.21 ± 0.58	N.D.	N.D.	N.D.	1.88	0.07
N-Vale-A	N.D.	2.10 ± 0.88	0.24 ± 0.38	N.D.	4.06	0.78

Ari. mean: Arithmetic mean; SD: standard deviation; N.D.: not detected.

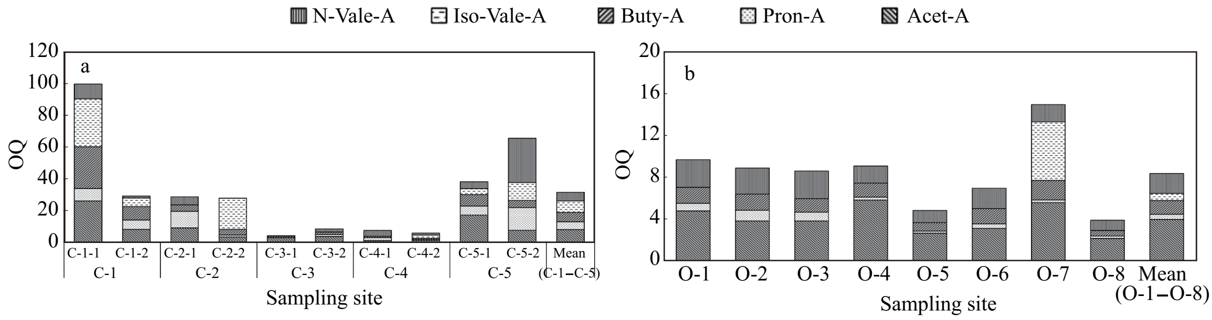

Fig. 2 Comparison of the sum of odorant quotients (SOQs) of the OCCs at the major fishery facilities (a) and border areas (b).

Table 3 Correlation of the different analysis items at the major fishery facilities and at the border areas

	Acet-A	Pron-A	Buty-A	Iso-Vale-A	N-Vale-A	SOCs	SOQs
At the major fishery facilities (n = 60)							
Acet-A	1.000						
Pron-A	0.597**	1.000					
Buty-A	0.816**	0.464**	1.000				
Iso-Vale-A	0.617**	0.424**	0.744**	1.000			
N-Vale-A	0.384*	0.808**	0.353	0.467**	1.000		
SOCs	0.947**	0.761**	0.853**	0.695**	0.614**	1.000	
SOQs	0.839**	0.757**	0.856**	0.856**	0.715**	0.945**	1.000
At the border areas (n = 48)							
Acet-A	1.000						
Pron-A	0.020	1.000					
Buty-A	0.809**	0.327	1.000				
Iso-Vale-A	–0.170	–0.165	–0.021	1.000			
N-Vale-A	0.404*	0.376	0.451*	–0.149	1.000		
SOCs	0.966**	0.167	0.862**	–0.142	0.533**	1.000	
SOQs	0.725**	0.226	0.780**	0.373	0.659**	0.799**	1.000

* Values are significant at $p < 0.05$; ** values are highly significant $p < 0.01$.

Table 4 Comparison of the concentrations of OCCs between this and other previous studies

Location	Acet-A (ppb)	Pron-A (ppb)	Buty-A (ppb)	Iso-Vale-A (ppb)	N-Vale-A (ppb)	C_2/C_3	Sampling period	Site characteristics	Reference
Yeosu, Korea	12.0	4.91	3.92	0.72	2.26	2.45	Jun–Nov, 2007	Industrial area (Major fishery facilities)	This study
Yeosu, Korea	5.93	0.50	0.87	0.07	0.78	11.8	Jun–Nov, 2007	Industrial area (Border areas)	This study
AnSan, ShiHung, Korea	19.5	19.0	13.0	0.93	0.82	1.02	Aug, 2004–Sep, 2005	Industrial area	Pal et al., 2008
AnSan, ShiHung, Korea	299	34.4	186	11.9	3.31	8.69	Jun, 2004–Oct, 2005	Industrial area (Emission source)	Kim et al., 2008
Guangzhou, China	4.10	0.60	0.30	0.08	0.64	6.83	Jul–Sep, 2003	Industrial area	Feng et al., 2005*
Beirut, Lebanon	1.90	0.90	–	–	–	2.11	Jul and Dec, 2003	Urban	Moussa et al., 2006
Hong Kong, China	1.00	0.20	0.20	0.03	0.06	5.00	Oct, 1997–Sep, 2000	Urban	Sin et al., 2001*
Guangzhou, China	4.60	0.50	1.80	0.02	0.62	9.20	Apr–Nov, 2005	Urban	Lü et al., 2010*
Athens, Greece	8.30	0.80	2.50	–	–	10.4	Jun–Dec, 2000	Urban	Bakeas et al., 2003*
Guangzhou, China	6.70	0.80	0.80	0.16	1.50	8.40	Nov–Dec 2005	Urban	Lü et al., 2009*
Xalapa, Mexico	9.80	1.80	2.20	–	–	5.40	Oct, 1996–Mar, 1998	Urban	Báez et al., 2001*
Shanghai, China	9.90	1.10	0.20	0.15	0.77	9.00	Jan–Oct, 2007	Urban	Huang et al., 2008*

* The conversion equation is based on 298 K and 1 atmosphere: ppb = (μg/m^3) × 24.45/molecular weight.

2.3 Correlation of OCCs

The correlations among the OCCs at the major fishery facilities and the border areas are shown in Table 3. Acet-A and Buty-A showed excellent correlations at both the major fishery facilities (r = 0.816, p = 1.87E-15, n = 60) and the border areas (r = 0.809, p = 3.40E-12, n = 48), which corroborate the results reported by other studies (Lü et al., 2009; Feng et al., 2005). Moreover, Acet-A and Buty-A had higher correlations with respect to SOQs than the other OCCs did, suggesting Acet-A and Buty-A might have similar sources at all of the sampling sites. In addition, Acet-A had a strong correlation with the SOCs at the major fishery facilities (r = 0.947, p = 3.51E-30, n = 60) and the border areas (r = 0.966, p = 1.21E-28, n = 48). The analysis showed that the concentration of Acet-A was significantly higher than that of any other OCC.

2.4 Comparison of OCCs results with other previous studies

The comparisons of OCCs concentrations and Acet-A/Pron-A (C_2/C_3) ratios between this study and other previous studies in the literature are listed in Table 4. The OCCs concentrations at the major fishery facilities were much lower than those at AnSan, ShiHung (Kim et al., 2008), but were higher than those at Guangzhou (Feng et al., 2005). For the border areas, the OCCs concentrations were similar to those of Guangzhou (Lü et al., 2010), Shanghai (Huang et al., 2008) and Athens (Bakeas et al., 2003). This result suggests that although the border areas are located at the industrial area, the OCCs concentrations are not worse than those at the urban areas in other places.

C_2/C_3 ratios should be used as indicators of anthropogenic origin for ambient OCCs, since Pron-A is believed to be associated only with anthropogenic emissions (Feng et al. 2005). C_2/C_3 would be high in a clean area and low in a polluted area. In other studies, C_2/C_3 ratios range from 1.02 (Pal et al., 2008) to 8.69 (Kim et al., 2008) in industrial areas and from 2.11 (Moussa et al., 2006) to 10.4 (Bakeas et al., 2003) in urban areas. In our study, C_2/C_3 ratios were 2.45 and 11.8 at the major fishery facilities and the border areas, respectively. In particular, the C_2/C_3 ratios at the border areas were higher than those at the urban areas investigated in other previous studies. In other words, anthropogenic source had little or no influence on the ambient OCCs concentrations at the border areas.

3 Conclusions

This study investigated 5 OCCs in a fishery industrial complex in Yeosu, Korea. The concentrations of Acet-

A were greater than those of other OCCs, making it the major OCC at both the major fishery facilities and the border areas. In general, the highest OCCs concentrations were observed in the summer. Furthermore, non-industrial pollution sources like vehicular exhaust and photochemical reactions had a large impact on the concentration variations of OCCs. The results of the correlation analyses suggest that Acet-A and Buty-A might have similar pollution sources in this fishery industrial complex. The air quality affected by OCCs at the border areas was generally very similar to those at the urban areas of other studies.

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