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Characteristics of the Major Atmospheric Aromatic Hydrocarbons in the Yellow Sea

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ABSTRACT

We measured the concentrations of five aromatic hydrocarbons (benzene, toluene, ethylbenzene, m,p-xylene, and styrene) in the atmosphere during four seasonal campaigns at Deokjeok and Jeju Islands in the Yellow Sea from October 2005 to June 2006. Toluene was the most abundant aromatic hydrocarbon, with median of 0.24 ppb at Deokjeok and 0.20 ppb at Jeju, followed by benzene (0.21 ppb, 0.15 ppb) and m,p-xylene (0.06 ppb, 0.06 ppb). Aromatic hydrocarbon measurements exhibited the typical seasonality of the major emission sources, such as vehicle exhaust, solvent evaporation, and regional circulation patterns. The ratios of m,p-xylene/ethylbenzene of 1.57 at Deokjeok and 1.05 at Jeju reflected the degree of proximity to outflows of each source region, South Korea and China. The toluene/benzene ratios of 1.0 were consistently both on field observations and on the 3-D chemical model simulation, which is slightly higher than that in the Western Pacific area. It implied that the air over the Yellow Sea was influenced to a great extent by the surrounding areas. We confirmed that current emission inventories of aromatic hydrocarbons in Northeast Asia reasonably reproduced temporal and spatial variations of toluene and benzene over the Yellow Sea.

Key words: Aromatic hydrocarbons, Toluene, Benzene, Continental outflows, Yellow Sea

1. INTRODUCTION

A significant fraction of non-methane hydrocarbons (NMHCs) in the atmosphere exists as forms of aromatic compounds, such as benzene, toluene, ethylbenzene, m,p-xylene, o-xylene (BTEX), and styrene. In Northeast Asia, aromatic compounds have been continuously increasing over the last decades (Tang *et al.*, 2009). With growing industrial development, their emissions are expected to rise in the coming decades (Wang *et al.*, 2005a; Street *et al.*, 2003).

The concentration ratios among various hydrocarbons have been widely used as tracers for the history of air masses and as indicators of anthropogenic activities (Baker *et al.*, 2011; Barletta *et al.*, 2009; Iovino *et al.*, 2009; Liu *et al.*, 2008; Monod *et al.*, 2001; Brocco *et al.*, 1997; Clarkson *et al.*, 1996; Nelson and Quigley, 1983). The atmospheric lifetime for benzene (57 h) is four times longer than that of toluene (14 h) in the summer season (Rappenglück *et al.*, 1998). Thus, as air is transported away from a source region, the toluene/benzene ratio likely will decrease due to successive differential photochemical degradation. A change in this ratio with distance traveled has been observed in air masses transported from the Asian continent in a few cases (Wang *et al.*, 2005b; Kato *et al.*, 2004; Hsieh and Tsai, 2003).

Regional and national estimates of emissions of hydrocarbons, including aromatic compounds, over Asia are subject to a high degree of uncertainty (within the range of 35-59%) (Street *et al.*, 2003). Hence, the measurement of concentrations of major aromatic compounds in background air and chemical transport model comparisons are very important for evaluating uncertainties in their emission inventories. The objectives of this study are to determine the concentrations of major aromatic compounds and to investigate their temporal and spatial variations during different seasons at two islands in the northern and southern part of the Yellow Sea. We examined the concentrations and ratios of selected aromatic hydrocarbons to evaluate their photochemical characteristics during the transport process. In addition, we evaluated the current emission inventories of these chemical species by comparing observations with the results of 3-D chemical transport modeling.

2. METHODS

2. 1 Sampling

We measured aromatic hydrocarbon concentrations at two island sites in the northern and southern part of the Yellow sea: Deokjeok and Jeju Islands, respectively (Fig. 1). Deokjeok Island, which has an area of 20.9 km^2 and a population of about 900, is located 75 km west of the Korea Peninsula. Samples were collected on the roof of an elementary school on the southeastern coast of the island. The southwest to northeastern areas of Deokjeok Island are mostly pine forest, whereas the rest of the island faces the sea. No local emission sources were found near the sampling location. Jeju Island lies ~100 km to the south of the Korean Peninsula, ~250 km west of Kyushu, Japan, and ~500 km east-northeast of Shanghai, China. On Jeju Island, sampling was conducted at Gosan; it is considered an ideal location to monitor the regional background atmosphere of East Asia because no local industrial sources exist (Lim *et al.*, 2011; Chen *et al.*, 1997; Carmichael *et al.*, 1996). Gosan lies 75 m above sea level and is located on the western tip of Jeju Island. This observational station was designated as a supersite for the ACE-Asia (Aerosol Characterization Experiment-Asia) program in 2001 (Huebert *et al.*, 2003) and for the ABC-EAREX2007 (Atmospheric Brown Cloud-East Asia Regional Experiment 2007) program (Lee *et al.*, 2007).

Samples were collected at the same time on the same date at the two stations during four different seasons: October 15-25, 2005; January 5-20, 2006; April 1-16, 2006; and June 6-16, 2006. At the Jeju Island site, eight samples (each accumulated for 3 h) were collected each day using an automated tube sampler. At the Deokjeok site, samples were taken manually every 3 h from 6 am to 12 am and a 6-h integrated sample was collected from 12 am to 6 am. Air samples were collected in 12-cm long glass adsorption tubes (ID=6 mm) packed with Tenax TA (60/80 mesh) and Carbosieve SIII (60/80) that were purged with high purity helium at 330°C for 2 h and sealed prior to each campaign period (McClenny *et al.*, 2002; US EPA, 1999a; McClenny and Colón, 1998). Air samples passed through a Nafion Dryer filled with silica gel and then a crystalline KI trap at 100 ml min^{-1} (1 atm, 25°C) before reaching an adsorption tube to remove water vapor and oxidants, respectively. Adsorption tubes containing samples were brought to the laboratory and analyzed within 3 weeks of collection.

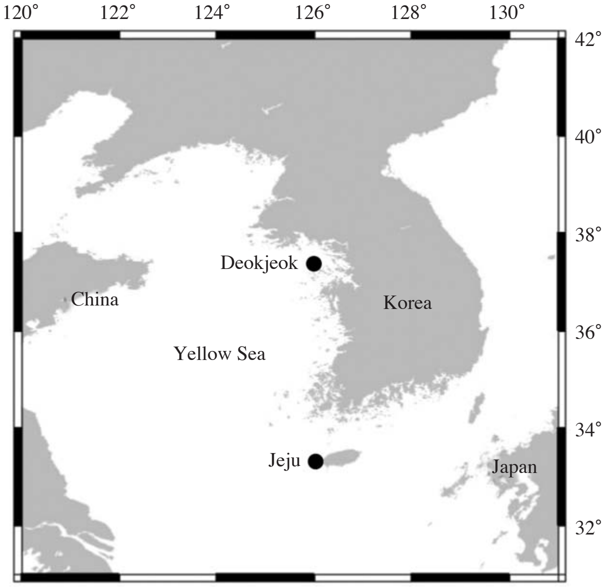

Fig. 1. Locations of the sampling sites in the Yellow Sea.

2. 2 Analytical Methods

Adsorption tubes containing samples were analyzed using a GC/FID (Donam 6200, Donam Instrument (Seong Nam, South Korea) equipped with a cryofocusing preconcentration unit (Aerotrap 6000, Tekmar, USA). Samples were desorbed using a homemade thermal desorption unit at 300°C for 5 min, transferred to a cryogenic pre-concentration trap at -120°C, then transferred to a cryofocusing unit at the tip of a GC column. The aromatic compounds were separated in a 60-m long, 0.32-mm ID, 1.0 μm film thickness DB-1 fused silica column (J&W Scientific, USA). The GC column oven temperature was programmed to start at 35°C for 5 min and then to increase at a rate of 5°C /min to 200°C, then stay at 200°C for 5 min. To calibrate the GC/FID, adsorption tubes were prepared in the same way as the samples, but diluted concentrations from a 100 ppb TO-14A calibration mix (Supelco, USA) and a dynamic dilutor (Entech, USA) were used. R-square values for standard calibration curves for the six investigated aromatic compounds were >0.995.

Table 1. Detection limits, precision, and accuracy of aromatic compound analysis using (A) GC/FID with an adsorption tube used in this study compared to (B) GC/MSD with canister sampling during the experiment.

Species	Precision %	Accuracy % (ppb)	Detection limit ppb	Comparison #1		Comparison #2		Comparison #3	
				A	B	A	B	A	B
Benzene	3.8	12 (0.06)	0.02	0.10	0.07	0.17	0.07	0.11	0.05
Toluene	6.8	12 (0.06)	0.06	0.17	0.17	1.76	1.19	0.14	0.09
Ethylbenzene	4.0	11 (0.06)	0.01	0.02	0.02	0.06	0.02	0.06	0.01
m,p-xylene	5.2	12 (0.06)	0.01	0.03	0.02	0.06	0.02	0.08	0.01
Styrene	5.7	13 (0.06)	0.01	0.03	0.03	0.05	0.01	0.02	0.01
o-xylene	4.4	11 (0.06)	0.01	0.03	0.01	0.06	0.01	0.06	0.01

The detection limits for the compounds were determined using average field blanks plus three folds of their standard deviations. The detection limits for benzene and toluene were 0.02 and 0.06 ppb, respectively (Table 1); the limits for ethylbenzene, m, p-xylene, styrene, and o-xylene were 0.01 ppb. The precision determined from the relative standard deviation of the lowest standard concentration (~0.6 ppb) ranged from 4% to 8%, and the analytical accuracy ranged from 11 % to 13% at the same lowest standard concentration.

2. 3 Chemical Transport Model

We used CAMx (Comprehensive Air Quality Model with Extensions) to model the regional distribution of aromatic compounds; CAMx is a Eulerian photochemical dispersion model that allows for an integrated “one-atmosphere” assessment of gaseous and particulate air pollution over many scales ranging from urban to regional (ENVIRON, 2004). One of important components of the CAMx probing tools, the Reactive Tracer (RTRAC) Source Apportionment for air toxics, is particularly suitable for simulating benzene and other individual aromatic compounds (Morris *et al.*, 2003). RTRAC has been widely used to simulate the photochemical conditions and distribution of air toxins over various regions of the world (Tesche *et al.*, 2006; Tanaka *et al.*, 2003; Dunker *et al.*, 2002). In our study the model domain covered North East Asia (centered at 40° N and 120° E, covering the eastern part of China, Korea, and Japan) with 97 × 90 horizontal grids by 60 km resolution. We used the MM5 model (NCAR/Penn State Mesoscale Model) for CAMx meteorological input fields. Initial and boundary conditions in MM5 were initialized from NCEP AVN final analysis. Emissions of NO_X, SO_2, CO, and volatile organic compounds (including benzene) were prepared from emission inventories compiled by Ohara *et al.* (2007). The CAMx model was run using the CB-IV chemical mechanisms scheme. We selected the spring as simulation period for the CAMx modeling analysis; at this time of year the long-range transport and active horizontal mixing processes prevailed.

3. RESULTS AND DISCUSSION

3. 1 Side-by-side Comparisons

Due to the complexity of the measurements, particularly when compounds are present at low concentrations, accurate determination of aromatic compounds is very challenging. Measurements using the adsorbent method are particularly vulnerable to various artifacts (Sanchez and Sacks, 2004; McClenny *et al.*, 2002). Thus, it is necessary to assess the quality of measurements before further data analysis. To evaluate the overall quality of sampling and analytical procedures using the adsorbent tube samplers, we collected three 6 L SUMMA canister samples at the Deokjeok site while the adsorption tube samples were being continuously taken during the campaign in June 2006 (US EPA, 1999b). These canister samples then were analyzed using a cryo pre-concentration unit (Entech, USA) and GC/MSD (Agilent, USA) in the laboratory.

Table 1 shows the results for the aromatic compounds measured from the tube samples using GC/FID versus the canister samples using GC/MSD. The detection limits were low enough to verify inter-comparison of selected species using GC/FID and GC/MSD methods even in these background level conditions. The first set of comparisons shows the excellent agreement between the two methods for all measured species, even at very low concentration levels. In contrast, the other two comparisons clearly indicate that the tube method tends to yield higher concentrations than the canister method. Schmidbauer and Oehme (1988) reported that sufficiently low blanks in absorbents could be maintained for a storage period of 1 week. However, our samples had to be stored for up to 3 weeks before analysis. This relatively long storage period might have increased the possibility of adverse positive effects in adsorbents. It is noteworthy that the most significant difference in our comparisons occurred near the detection limits. In general, the calculated detection limits, precision, and accuracy for benzene, toluene, ethylbenzene, and m,p-xylene were adequate

Table 2. Statistical summary of major aromatic hydrocarbon measurements taken during the four sampling campaigns at the Deokjeok and Jeju Island sites.

	Deokjeok					Jeju				
	Mean	Median	Std*	Min*	Max*	Mean	Median	Std	Min	Max
Benzene	0.23	0.21	0.16	nd	1.03	0.17	0.15	0.12	nd	0.96
Toluene	0.66	0.24	1.16	nd	8.89	0.46	0.20	0.74	nd	8.01
Ethylbenzene	0.08	0.04	0.13	nd	1.09	0.05	0.03	0.06	nd	0.47
m,p-xylene	0.14	0.06	0.24	nd	2.37	0.08	0.06	0.08	nd	0.60
Styrene	0.02	0.01	0.07	nd	1.08	0.02	0.01	0.04	nd	0.44
o-xylene	0.06	0.04	0.08	nd	0.71	0.02	0.02	0.02	nd	0.23

*Std, min, and max stand for standard deviation, minimum, and maximum. All concentrations are in ppbv.

to assess their behaviors in this relatively clean air. Particularly, the observed variations of benzene, toluene, and m,p-xylene concentration compared to their analytical uncertainties were large enough to discuss further detail.

3. 2 General Characteristics

Table 2 lists the statistical summary of the major aromatic compound measurements taken during the four sampling campaigns. Among the target aromatic hydrocarbons, toluene was the most abundant at both sites. The means, medians, maxima, and standard deviations of all measured compounds at the Deokjeok site were higher than those at Jeju, which clearly illustrates that the Deokjeok site experiences more anthropogenic influences due to its proximity to a major industrial region in Korea. The benzene median (0.15 ppb) at the Jeju site was slightly lower than those (0.17-0.20 ppb) of benzene at the same latitude measured during the PEM-WEST B and TRACE-P studies, which were conducted mainly in the late winter and early spring of 1994 (Blake *et al.*, 2003; Blake *et al.*, 1997). If we consider only the spring (0.16 ppb) and winter (0.18 ppb) medians at the Jeju site, the benzene level over this region did not change much over the 1990s. In contrast, the toluene levels at the Jeju site during the spring and summer of this study were very high compared with those of other similar coastal background locations in the East Asia (Wang *et al.*, 2005b; Yang and Lo, 1998).

Each day during each sampling campaign, we sampled the air seven times at the Deokjeok site and eight times at the Jeju site to investigate the possible diurnal variation of aromatic compound concentrations. Although no statistically significant diurnal variations were found at either sampling sites, we did observe seasonal variations (Fig. 2). Benzene concentration was highest in winter and lowest in summer at both sites. This observed seasonal tendency of benzene is consistent with the general circulation pattern of the air in the study region, which facilitates the effective transport of continental outflows during winter and the inflow of relatively clean marine air during summer. Accordingly, several researchers have reported that in many Asian cities, high benzene concentrations correspond to increased traffic activity in winter (Morino *et al.*, 2011; Lee *et al.*, 2002; Na and Kim, 2001; Sin *et al.*, 2000). Solvent usage is known as the largest source of atmospheric toluene, ethylbenzene, and xylene (Brocco *et al.*, 1997). The observed summer and fall maxima of xylene and ethylbenzene at both sites can be explained by the evaporation rate and seasonal solvent sales figures in Korea (Na and Kim, 2001). However, spring maxima of toluene at both sites likely reflect the increased influences of local sources in the islands.

3. 3 Relationships between Aromatic Compounds

We found a good linear correlation between m,p-xylene and ethylbenzene at both sampling sites. Nelson and Quigley (1983) reported that the ratio of these two compounds is constant in the range of 3-4 for their various sources, such as vehicle exhaust, solvent petrol, and fuel evaporation. Monod *et al.* (2001) confirmed that these two compounds exhibited the strong linear relationship and that their ratios remained relatively constant at different source locations in Europe, Asia, and South America because they share common emission sources (Table 3). They also identified this ratio as a useful tool for estimating the photochemical age of an air mass. In this study, the ratios of m,p-xylene/ethylbenzene determined by the slope of the fitted line were 1.57 at the Deokjeok site and 1.05 at the Jeju site. The distinct and well-defined ratios at these two sites confirm that they can be useful to trace the photochemical age of air masses over this region. Correlations among species in our results, such as toluene, ethylbenzene, and m,p-xylene, were generally high, indicating that they were emitted from common source types (probably solvent usage).

The toluene/benzene ratio also can be used to trace

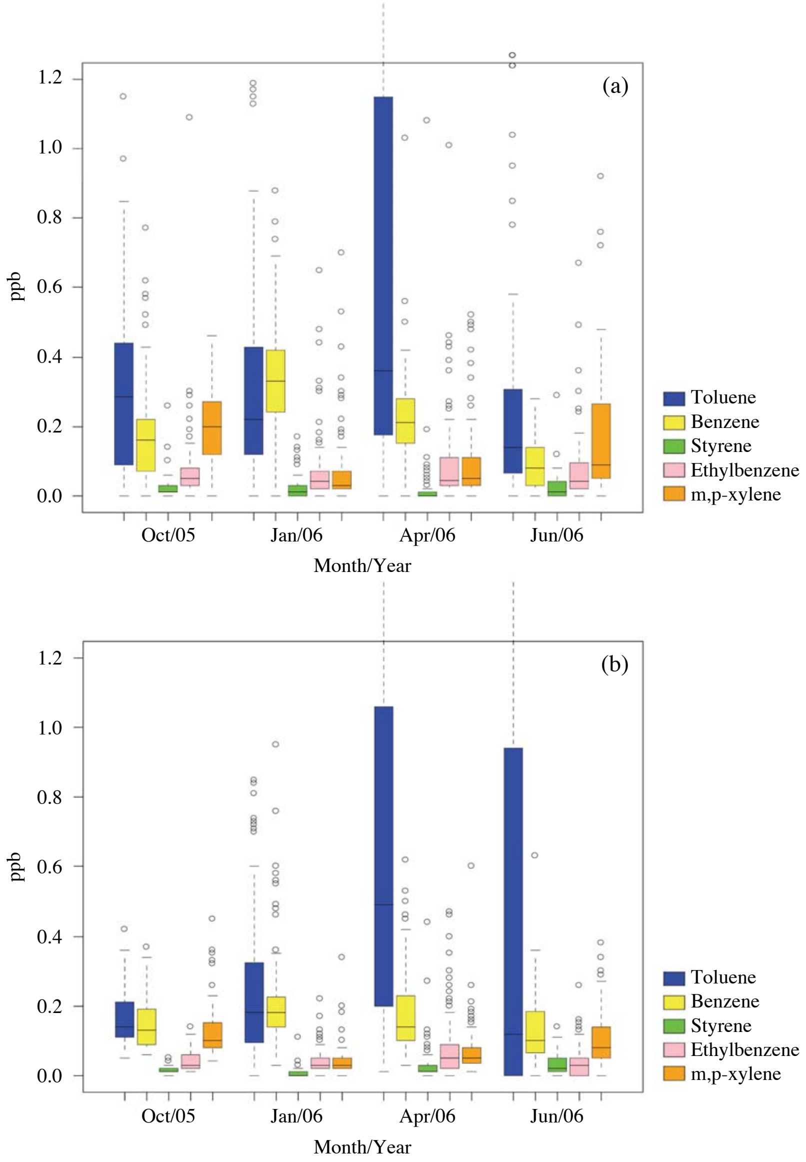

Fig. 2. Statistical distributions of measured species during the sampling periods at the (a) Deokjeok and (b) Jeju sites.

Table 3. Comparison of BTEX ratios at various sites over the North Pacific region.

	Deokjeok*	Jeju*	East Asia$^{1)}$	Southern China$^{2)}$	Eastern China$^{3)}$	HongKong$^{4)}$	Taiwan$^{5)}$	Seoul$^{6)}$
m,p-xylene/ethylbenzene	1.57	1.05				2.0		3.3
Toluene/benzene$^{7)}$	3.21	2.25	0.7	5.4	1.8	6.7	2.8-6.0	6.4
Toluene/benzene$^{8)}$	1.40	0.96						

*This study; $^{1)}$Kato *et al.*, 2001; $^{2)}$Wang *et al.*, 2005b; $^{3)}$Guo *et al.*, 2004; $^{4)}$Sin *et al.*, 2000; $^{5)}$Chang *et al.*, 2004; $^{6)}$Na and Kim, 2001; $^{7)}$This study with all data; $^{8)}$This study with the local effect

air mass age. The changes in its ratio with distance and transport time from Asian source regions have been consistently observed and characterized by other studies (Wang *et al.*, 2005b; Kato *et al.*, 2004; Hsieh and Tsai, 2003). However, meaningful correlations between toluene and benzene concentrations are not visible in our results.

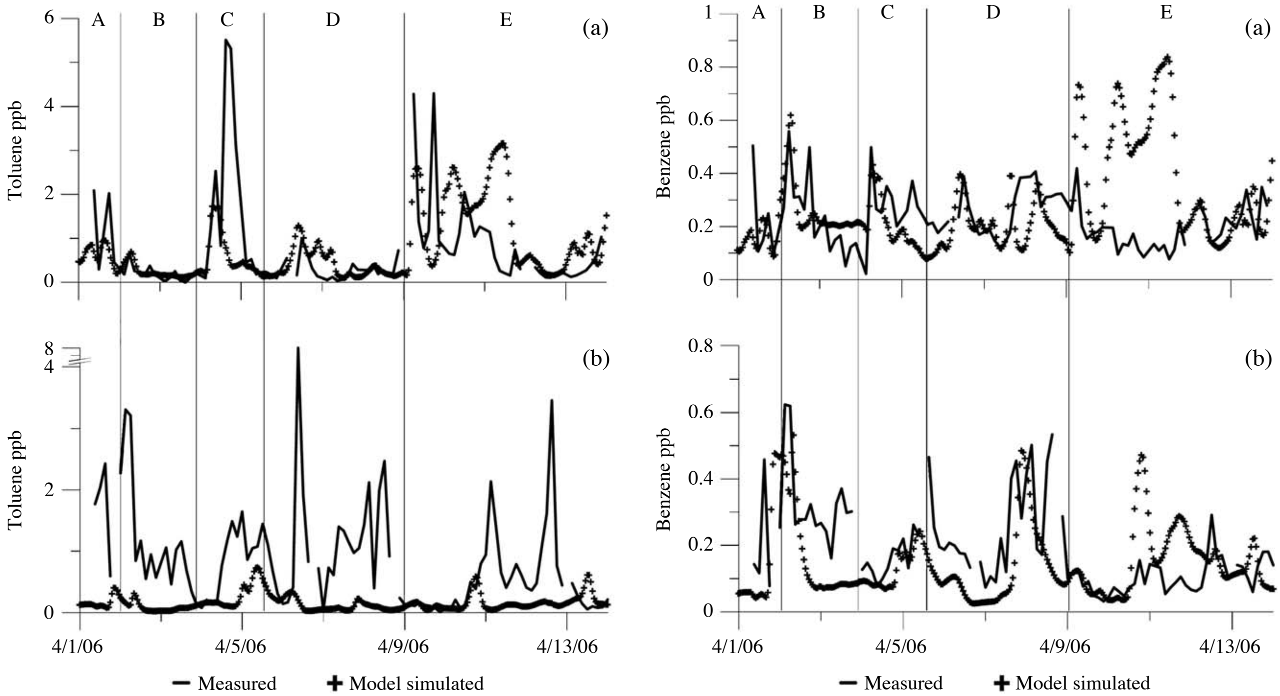

Fig. 3. Comparisons of observed and simulated toluene and benzene concentrations at the (a) Deokjeok and (b) Jeju sites.

3. 4 Model Comparison

The most effective way to characterize the regional distribution of trace species is to compare observations with mesoscale model calculations. With model results, it is often possible to see details of air mass history and transport paths. In our study, the selected simulation period for comparison covers 1-13 April, 2006 with a 2-day spin-up from March 29. Fig. 3 shows the comparison of simulated toluene and benzene concentrations with the observed data from the Deokjeok and Jeju sites. Overall, the model simulated the general trends and levels of both toluene and benzene at the Deokjeok site and benzene at the Jeju site. Based on the regional weather changes that occurred over the Yellow Sea during the spring campaign, we divided the data into five consecutive periods (A to E in Fig. 3). Periods A, C, and E were characterized by a prevailing low pressure system with precipitation over the region. At the Deokjeok site, enhanced toluene/benzene ratios are conspicuously visible in the measured data during these low pressure conditions. However, the observed toluene/benzene ratios at the Jeju site do not show any systematic variations with changing weather patterns. Observed toluene concentrations in Jeju site were almost always higher than simulated ones. The Jeju site was constantly influenced by a local toluene source presumably from construction activities that occurred at the same time as our sampling campaigns.

The toluene concentration field simulated by the CAMx model revealed that the Deokjeok and Jeju sites were under the direct influence of Korea during the C and E periods. In particular on 5 April, a strong pollution plume that originated from the Korean Peninsula stretched out from Deokjeok to Jeju site. Concurrent increases of observed toluene, benzene both at the Deokjeok and Jeju sites characterize well this Korean outflow. On two occasions (periods B and D), continental outflow from eastern China reached the Yellow Sea. These were periods without any precipitation or local contamination. During these two periods, the continental outflow signals in the model were very well captured at the Jeju site. However, local toluene contamination at the Jeju prevents us from verifying its influences using the data that were measured. At the Deokjeok site, continental outflow was evident only during period B; it was not clear during period D, as the marine air over the Deokjeok site was stagnant for several days.

If the air in Deokjeok during period B originated from a continental area and was free of local contamination, it provides an excellent opportunity to assess current toluene and benzene emission inventories in China. Fig. 4 depicts the toluene and benzene relationship in the simulated and observed data at the Deokjeok site during period B. The ratio was 1.0, and this

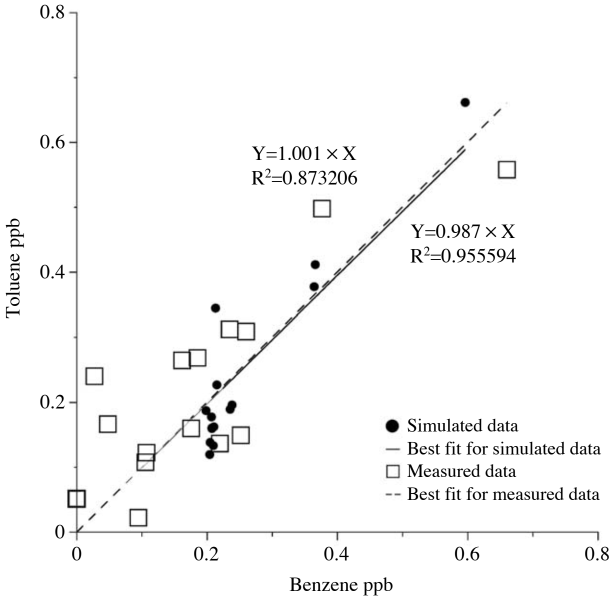

Fig. 4. Correlations between toluene and benzene with linear regression lines for the observed and the calculated for the period of continental outflow influences at the Deokjeok site.

was consistently derived for both measured and calculated values. Our toluene/benzene ratios are slightly higher than those from Okinawa Island, which ranged from 0.6 to 1.1 with an average of 0.7 (Kato *et al.*, 2004), which is reasonable considering the difference in air mass ages from the source regions.

With these results, we believe that the toluene and benzene emission inventories in China compiled by Ohara *et al.* (2007) correctly reflect the spatial and temporal distribution of these species over the Yellow Sea. A simple calculation with toluene and benzene concentration can be used to as a marker to trace the air mass history over this region if local influences are properly excluded. However, it should be stressed that this result was derived from a very limited data set. Additional characterizations using other chemical tracers for more extended periods of observation, such as hydrocarbons with a longer lifetime, are necessary to properly assess the air mass history and emission inventories in future studies.

4. CONCLUSIONS

Concentrations of five aromatic hydrocarbons (benzene, toluene, ethylbenzene, m,p-xylene, and styrene) were successfully determined during four different seasons at Deokjeok and Jeju Islands in the Yellow Sea. Among the measured aromatic hydrocarbons, toluene was the most abundant, followed by benzene and m,p-xylene, at both measuring sites. While benzene levels (median of 0.15 ppb) were comparable to those reported in other studies performed in the same region, we found toluene levels to be sporadically high due to local influences at both sites, with more severe effects at the Jeju site. Benzene showed a maximum in winter, which reflects the increased traffic emissions that occur during that same season. m,p-xylene and ethylbenzene showed rather broad maxima over fall and summer, which reflects the combined effect of elevated solvent evaporation and solvent usage.

The good linear correlations between m,p-xylene and ethylbenzene had a slope of 1.57 at the Deokjeok site and 1.05 at the Jeju site. The larger slope and better correlation at Deokjeok compared to Jeju indicate that the Deokjeok site has been consistently influenced by less diverse sources in close proximity. It is known that the Jeju site is a suitable sampling location for regional background air with various air mass histories over the Northeast Asia. However, this was not true for toluene due to local contamination from construction activity at the Jeju site, particularly during the spring and summer campaigns 2006.

Using the 3-dimensional chemical transport model CAMx coupled with current emission inventories of toluene and benzene over Asia, we were able to confirm that current emission inventories of aromatic hydrocarbons in Northeast Asia reasonable reproduced temporal and spatial variations of toluene and benzene over the Yellow Sea. We found toluene/benzene ratios to be 1.0 at the Deokjeok site in the continental outflow cases for both measurement and simulation. This value was slightly higher than the 0.7 found at Okinawa Island, which implied that the air over the Yellow Sea was relatively fresh compared to that in the Western Pacific area.

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