

Characterization of Sea Salt in PM₁₀ at Inland and Seashore in Busan, Korea

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Abstract

In an effort to characterize temporal and spatial variability of PM₁₀ and to quantitatively estimate contribution of sea salt aerosol to PM₁₀ mass in Busan area, twenty four-hour averaged concentration of PM₁₀ were measured in two distinct areas, Gwaebeopdong(inland) and Dongsamdong(seashore), Busan for summer and fall, 2007. It was found that sea salt accounted for 2.9% and 9.5% of PM₁₀ mass in Gwaebeopdong and Dongsamdong, respectively for the study period, indicating that contribution of sea salt to PM₁₀ mass and total ion concentration in seashore area were consistently higher by a factor of three compared to inland area. Temporal analysis suggested that sea salt contributions to PM₁₀ in Dongsamdong were higher in summer due to the southerly sea breeze while there was no significant fluctuation of sea salt contribution for the summer and fall months in Gwaebeopdong. Sea salt enrichment factors(EF_{sea}) of K⁺, Ca²⁺ and SO₄²⁻(>10) indicated major contributions from anthropogenic sources and EFs of Mg²⁺ and Cl⁻ exhibited strong association with oceanic origins for both areas.

Key Words : PM₁₀, Sea salt aerosol, Enrichment factor, Coastal urban area, Inland, Seashore

1. Introduction

Sea salt aerosol is the most portion of the suspended particle that originates from natural sources (Butler, 1979; Christian, 1963; O'Dowd et al., 1997). Sea salt aerosol exerts direct and indirect influences on radiation transfer and annual emissions of sea spray are known to be as high as $2,096 \times 10^9$ kg/yr (Green, 1972; Kim, 2006; Kim, 2000). Sea salt aerosol is involved in formation of suspended particles through physicochemical reactions with gas and other particles during the migration from ocean to coastal area. Also, sea salt particles control the radiative properties of the clean background atmosphere

by scattering incoming sunlight(Murphy et al., 1998). Therefore, understanding impact of sea salt aerosol on particle formation is of substantial importance in investigating characteristics of particulate matter within a coastal urban airshed(Tombach and Brewer, 2005; US EPA, 2005; Zhao and Hopke, 2004). Sea salt-rich particulate matters in coastal urban areas are responsible for not only adverse health effects but economic losses caused by malfunctions of indoor facilities and corrosions of urban infrastructures including electric cables, metals, and buildings(Kim et al., 1994). Many studies in the past have been engaged in evaluating impact of natural sources including sea salt aerosol on particulate matter concentrations in coastal urban airshed and have been utilized to determine national air quality standards.

Kang(1988) investigated contribution of sea salt aerosol observed in Seoul and identified that sea salt

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aerosol concentration ranged from $4.8 \mu\text{g}/\text{m}^3$ to $13.2 \mu\text{g}/\text{m}^3$ and is attributed to 3.5 to 10.9% of total suspended particles(TSP) and average contribution of sea salt aerosol to the TSP was 6.83%. Moon(1992) indicated that contributions of sea salt aerosol to particulate matter observed at Kangwha in west coastal areas and at Yangyang in east coastal areas in Korea were 32.8% and 36.3%, respectively. This demonstrated that sea salt aerosol's contribution in east coastal area was more pronounced than in west coastal area. Chun et al.(1994) evaluated temporal variability of sea salt aerosol contribution to TSP measured in Seoul, suggesting that sea salt aerosol accounted for 7.31%, 3.19%, 5.04%, and 5.41% of TSP in winter, spring, summer, and fall, respectively and was the lowest in spring and the greatest in winter. The study suggested that the lowest contribution of sea salt in spring resulted from dominancy of the continental air parcels with high level of TSP associated with the Asian Yellow Dust and scattering of soil particles caused by dry and strong winds. Kim(2006) characterized sea salt aerosol at ambient air in Jeju Island and found that level of sea salt aerosol in TSP and contribution to TSP was inversely correlated to distance from the seashore. Wai and Tanner(2004) described that contribution of sea salt aerosol to PM_{10} observed at a coastal area in Hong Kong decreased as the distance increased from seashore to inland.

Annual average values of PM_{10} in Busan have exceeded the Korea National Ambient Air Quality Standard of $50 \mu\text{g}/\text{m}^3$ and have ranked the second highest among the metropolitan cities in Korea from 2004 to 2006(Ministry of Environmental, 2008). Although spatial and temporal characteristics of fine particulate matter and its trace elements measured in Busan have been investigated by other studies(Jeon, 2003; Jeon and Hwang, 2007; Kim et al., 2006), evaluations of sea salt aerosol in particulate matters for inland and seashore areas in Busan have not been

performed up to present. Thus the concrete evaluations of sea salt aerosol and its contribution to PM_{10} formation can provide insightful information that can be of assistance in establishing air quality standards for air quality management of particulate matters in Busan. In this study, sea salt aerosols in PM_{10} were collected at monitoring sites located in inland(Gwaebeopdong) and seashore(Dongsamdong) areas in Busan to identify temporal and spatial characteristics of sea salt contribution to PM_{10} .

2. Measurements and Methods

2.1. Study sites and periods

As shown in Fig. 1, this study was conducted in Gwaebeopdong(Silla University) and Dongsamdong (Busan Southern High School), Busan.

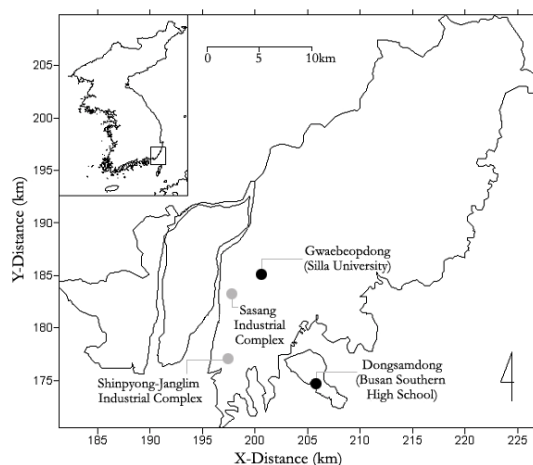


Fig. 1. Locations of the two sites involved in the study.

Gwaebeopdong monitoring site is located within a vicinity of residential and forest areas and is adjacent to industrial facilities. Gwaebeopdong site is distant approximately 10km from the south seashore. The Gwaebeopdong monitoring site is surrounded by the Bakyang mountain range spanning from northeast to southwest of the site and a small valley is situated southwest of the site. Sasang industrial complex used

to be positioned 3km southwest of the Gwaebeopdong monitoring site but many facilities from the industrial complex migrated to other areas, which resulted in remarkable reduction of point source emissions in Gwaebeopdong. Recently large-sized commodity distribution complexes, however, were replaced in Sasang industrial complex and subsequent increase of traffic volume augmented on-road emissions. Current point sources neighboring the Gwaebeopdong monitoring site are Shinpyong and Janglim industrial complexes located 8km south of the monitoring site. With southwesterly winds, emission plumes from the complex facilities may be transported to the monitoring site. Dongsamdong monitoring site is located very near to seashore line. This site can be influenced by natural sources such as sea salt aerosol due to its close distance to seashore and also by emissions from marine vessels that traveled in and out the Busan Port. The measurements were performed July to August(summer) and September to October (fall), 2007 for the days with no precipitation. Sampling duration was 24 hours starting at 0900LST and ending at 0900LST next day. Number of measurements was 21 in summer and 16 in fall, 37 in total, at the Gwaebeopdong site and 14 in summer and 15 in fall, 29 in total, at the Dongsamdong site.

2.2. Instrumentation

For measurements of PM₁₀, Mini Volume Air Sampler(U.S. Air Metrics Inc.) was used with 5.0 ℓ /min of inlet air flow and 47mm Quartz Fiber filter for 24 hours(Baldauf et al., 2001; Bogo et al., 2003). This sampler proceeds measurements with high degree of vacuum and air flow, provides convenient portability and consistent operation without pulse. It adopts the media injection method that separates particles less than 10 μm through inertia collision method. MINI PUMP(Model MP-603T) was utilized for filter sampling. This instrument has 5 ℓ /min of maximum air inlet and accordingly enables to adjust wide range

of air flow. It can be used to collect both toxic and normal gas. Filters used in this study are Quartz Fiber filter(QMA, 47 mm, No. 1851-047, Whatman Co.).

2.3. Measurement methods

Filters were first dried for two days at minimum using an automatic dry/up desiccator(SIBATA DUV-12) with constant temperature(20℃) and humidity(50%) and then weighed through a electronic microbalance (Sartorius microbalance, Germany) with sensitivity up to 0.01 mg to measure mass concentration of PM₁₀ before and after sampling of particles.

To analyze trace elements and ions in the filter sample, all analysis procedures were in compliance with the Air Pollutant Analyzing Procedure Standard (Air Pollution Research Association, 2000). For analysis of trace elements, ultrasound wave extraction method was adopted to preprocess the filter before the elements analysis. The extraction was performed in a way that with 6 ml mixture of 1.3 M nitric acid and 2.23 M hydrochloric acid(1:1) is added to a beaker where a filter is contained and then an ultrasound wave extractor retrieve solution from the beaker for 2 hours with 28 KHz. After the ultrasound treatment, the beaker will be filled with distilled water up to 20 ml so the resulting concentration of nitric acid - hydrochloric can be 0.31M and 0.67 M (1:1) extracts. After the ultrasound wave preprocessing of the filter, the extracts pass through syringe filter(PVDF, Whatman Co. pore size 0.45 μm). Then using ICP/AES (ICP-IRIS, Thermo Jarrell Ash Co., USA), Al, Ca, Fe, Mg, K, Na, S, Si, Sr, Ti, V and Zn were analyzed. Using ICP/MS, Cd, Cr, Cu, Mn, Ni and Pb were quantified.

To analyze water-soluble ionic compositions in PM₁₀, sample filters were placed in 30 ml HDPE bottle(Nalgene Co.) with 20 ml(Direct Q Millipore, 18.2 MΩ ultrapure water and then the bottle was stored in a refrigerator to prevent change in quality. The stored samples were unfrozen and passed through

ultrasound wave preprocessing in a way that the bottle was placed in an ultrasound washer for an hour to extract sample solution. Syringe filter(PVDF, Whatman Co. pore size 0.45 μm) was used for filtering of the extracts. Then the filter extracts was analyzed using ICP/AES for Na^+ , K^+ , Ca^{2+} , Mg^{2+} and using Ion Chromatography with CS12A Column and CSRS-ULTR II suppressor for anions(NH_4^+) and with AS14 and ASRS-ULTR II suppressor for cations(NO_2^- , NO_3^- , SO_4^{2-} , Cl^-).

3. Results and discussion

3.1. Contribution of sea salt aerosol to PM_{10}

Table 1 summarizes levels of PM_{10} and sea salt aerosol as well as contribution of sea salt to composition of PM_{10} measured in Gwaebeopdong and Dongsamdong for summer and fall, 2007. Sea salt aerosols at ambient air comprise of Cl, Na, S, Mg, Ca, K that existed in seawater and Na element is an indicator of sea salt aerosol source(Willison et al., 1989). Since Na accounts for 30.6% of total mass in seawater, concentration of the airborne sea salt can be calculated based on the measured Na as the equation (1)(Chun et al., 1994).

$$\text{Seasalt aerosol}(\mu\text{g}/\text{m}^3) = \frac{100}{Na_{\text{seawater}}(\%)} \times Na_{\text{atm}}(\mu\text{g}/\text{m}^3) \quad (1)$$

Where, Na_{seawater} : Na percentage in sea salt aerosol (30.64%)

Na_{atm} : Na concentration in airborne suspended particle

Average sea salt concentration in PM_{10} measured at Gwaebeopdong site was higher in summer(2.21 $\mu\text{g}/\text{m}^3$) than in fall(1.99 $\mu\text{g}/\text{m}^3$) and 2.12 $\mu\text{g}/\text{m}^3$ for all the sample periods. As for Dongsamdong, average sea salt concentration during summer(6.03 $\mu\text{g}/\text{m}^3$) and fall(6.23 $\mu\text{g}/\text{m}^3$) was 6.13 $\mu\text{g}/\text{m}^3$ which was three times higher than that of Gwaebeopdong. Especially, overall average sea salt contribution to total PM_{10} mass in summer and fall was 2.9% at the Gwaebeopdong site while that of Dongsamdong was 9.5%, approximately 3.3 times higher compared to Gwaebeopdong. This analysis indicates that Gwaebeopdong and Dongsamdong areas experienced substantially different contribution of sea salt aerosol to PM_{10} formation. In addition, percentage contributions of sea salt at the Dongsamdong site which is located in coastal area present higher in summer(10.4%) than in fall(8.9%). This is most probably due to the sea breezes that are related to southerly winds and occur more predominant in summer than in fall. On contrast, the Gwaebeopdong site which is located inland did not undergo a significant seasonal variability of sea salt contribution in summer(2.8%) and fall(3.1%). Kim(2006) presented 13.1% and 15.2% of sea salt aerosol contribution to PM_{10} observed in summer and fall, respectively, at an inland site in Jeju Island that is 2 km distant from the seashore. Those levels were much higher level than sea salt aerosol contributions identified in this study. Wai and Tanner(2004) estimated 1.2%~3.9% of sea salt aerosol contributions to PM_{10} measured at Hong Kong, which

Table 1. Mean sea salt concentration and contribution ratios of sea salt of PM_{10} in Busan

Station	Season	Mean PM_{10} concentration ($\mu\text{g}/\text{m}^3$)	Mean sea salt concentration ($\mu\text{g}/\text{m}^3$)	Contribution rate of sea salts (%)
Gwaebeopdong	Summer	79.5	2.21	2.8
	Fall	63.6	1.99	3.1
	Mean	72.7	2.12	2.9
Dongsamdong	Summer	58.1	6.03	10.4
	Fall	70.0	6.23	8.9
	Mean	64.3	6.13	9.5

were similar to those in Gwaebeopdong. This indicates that air quality management and policy making process for a coastal urban area such as Busan should take into consideration significant contributions of natural sources markedly linked with geographical configuration.

3.2. Contribution of sea salt to total ions composition

To estimate contribution of sea salt to total ions composition (Lee and Ho, 1999; Mason, 1966), the following equation was applied;

$$CR_{ss}(\%) = \frac{\sum_i C_{i,ss}}{\sum_i C_{i,t}} \times 100 \quad (2)$$

where, $C_{i,ss}$ is concentration of each ion based on the standard element Na

$C_{i,t}$ is concentration of each water-soluble ion.

$$\sum_i C_{i,ss} = [Na^+] + [SS - SO_4^{2-}] + [SS - Cl^-] + [SS - K^+] + [SS - Ca^{2+}] + [SS - Mg^{2+}]$$

$$\sum_i C_{i,t} = [Cl^-] + [NO_3^-] + [SO_4^{2-}] + [NH_4^+] + [Ca^{2+}] + [K^+] + [Mg^{2+}] + [Na^+]$$

where, sea salts(SS) and non sea salts(NSS) are calculated as below

$$Seasalts = 3.270 \times [Na^+]$$

$$[SS - Cl^-] = 1.798 \times [Na^+]$$

$$[SS - SO_4^{2-}] = 0.251 \times [Na^+]$$

$$[SS - Ca^{2+}] = 0.038 \times [Na^+]$$

$$[SS - K^+] = 0.036 \times [Na^+]$$

$$[SS - Mg^{2+}] = 0.1205 \times [Na^+]$$

$$[NSS - SO_4^{2-}] = [SO_4^{2-}] - 0.251 \times [Na^+]$$

$$[NSS - Ca^{2+}] = [Ca^{2+}] - 0.038 \times [Na^+]$$

Table 2 presents contributions of sea salts to total ions that constitute total PM₁₀ in seasons. For Gwaebeopdong, total ion concentration was higher in summer(30.24 $\mu\text{g}/\text{m}^3$) than in fall(23.81 $\mu\text{g}/\text{m}^3$) and average of 27.46 $\mu\text{g}/\text{m}^3$ for all the sample periods. The Dongsamdong site exhibited higher level in summer(33.41 $\mu\text{g}/\text{m}^3$) than in fall(27.86 $\mu\text{g}/\text{m}^3$) and the overall average of 30.54 $\mu\text{g}/\text{m}^3$, which was higher than that of Gwaebeopdong.

Contribution of sea salts to total ion composition in Gwaebeopdong was higher in fall(10.43%) than in summer(8.18%) and average of 9.15% throughout the sample periods. As for Dongsamdong, contribution of sea salt in fall(25.44%) was higher than in summer(20.93%) and the overall average was 23.26%, which was about 2.5 times higher than Gwaebeopdong.

3.3. Sea salt enrichment factor(EF_{sea}) and mass fraction of sea salt

To calculate sea salt enrichment factor(EF_{sea}) of ions in PM₁₀ observed in Busan area, the equation (3) was adopted as below;

$$EF_{sea} = \frac{(X_i/Na)_{sample}}{(X_i/Na)_{sea}} \quad (3)$$

(X_i/Na) is concentration rate of each ion to the Na concentration in crustal component of PM₁₀. In order

Table 2. Contribution ratios of sea salt of total ion concentration in Busan

Station	Season	Total ion concentration($\mu\text{g}/\text{m}^3$)	Contribution rate of sea salts(%)
Gwaebeopdong	Summer	30.24	8.18
	Fall	23.81	10.43
	Mean	27.46	9.15
Dongsamdong	Summer	33.41	20.93
	Fall	27.86	25.44
	Mean	30.54	23.26

to evaluate origin of water-soluble ions of PM_{10} measured in Busan, enrichment factors were calculated for the monitoring sites, Gwaebeopdong and Dongsamdong, referring Mason(1966)'s average concentration of seawater. The results of enrichment factor analysis are listed in Table 3. The higher EF_{sea} values indicate the higher probability of particulate matter formation from anthropogenic origins(Guo et al., 2004; Zhang and Iwasaka, 1999). EF_{sea} values greater than 1.0 suggest contributions from other sources apart from the seawater and EF_{sea} values less than 10 are considered as indications that origins can not be specifically defined due to the difference of chemical compositions between sea salt and Mason's compilation(Kaya and Tuncel, 1997). In this study, Na^+ is used for reference element and K^+ , Ca^{2+} and SO_4^{2-} ions showed EF_{sea} values greater than 10, which denotes higher contribution of anthropogenic origin. Especially, EF_{sea} of Ca^{2+} was observed higher in fall than in summer, and higher in Gwaebeopdong than in Dongsamdong which is neighboring the seashore. As for EF_{sea} of K^+ , its origins related to non sea salt in Gwaebeopdong were pronounced while those were less significant in Dongsamdong. EF_{sea} of SO_4^{2-} observed high in both Gwaebeopdong and Dongsamdong suggested influences of the emissions from marine vessels activities at the surrounding ports. EF_{sea} of Mg^{2+} and Cl^- , two major components of the sea salt in particulate matter, suggested the origin

of seawater as well as geographical difference of Gwaebeopdong and Dongsamdong. EF_{sea} of NO_3^- and NH_4^+ were not evaluated since those ions originate from anthropogenic processes rather than seawater influences.

3.4. Changes in Cl^-/Na^+ ratio

The ratio Cl^-/Na^+ in airborne particulate matters can be calculated and compared with the seawater Cl^-/Na^+ ratio, 1.8, to investigate oceanic origin or anthropogenic source. As the ratio Cl^-/Na^+ is close to 1.8, the origin is considered to be seawater while the ratio becomes apart from 1.8 as the anthropogenic sources prevail (Mats and Gustafsson, 2000). Most of Na in airborne suspended particle is most likely attributed to seawater and thus is used for indicator of seawater origin. Therefore, anthropogenic sources are associated with chloride compounds and in that case the Cl^-/Na^+ ratio is supposed to be greater than 1.8(Chun et al., 1994; Kang, 1988). Cl^-/Na^+ ratio of sea salt aerosol is known to be 1.6 and hence in case where the ratio is close to 1.6, the origin tends to be oceanic source. Since most of anthropogenic sources are chloride compounds related, the ratio is greater than 1.6. When deficiency of chloride substances exists, the ratio is less than 1.6(Kang, 1988; Kim, 2006; Peter, 1988).

Table 4 listed Cl^-/Na^+ ratio in PM_{10} at the Gwaebeopdong and Dongsamdong sites for summer and fall. The Cl^-/Na^+ ratio in Dongsamdong were 1.31 and 1.59 in summer and fall, respectively and overall

Table 3. Regional enrichment factor(EF_{sea}) of water soluble ions in PM_{10} relative to non sea salts composition in Busan

Contents	Seawater		Gwaebeopdong			Dongsamdong		
	Abundance(mg/kg)	(X)/Na	Summer	Fall	Mean	Summer	Fall	Mean
Na^+	10773	1.0000	1.00	1.00	1.00	1.00	1.00	1.00
K^+	399	0.0370	23.59	26.56	24.80	5.49	10.56	8.15
Mg^{2+}	1294	0.1201	1.29	2.00	1.58	0.48	0.63	0.56
Ca^{2+}	412	0.0382	38.22	50.91	43.39	12.69	13.39	13.06
NH_4^+		0.0000	-	-	-	-	-	-
Cl^-	19344	1.7956	1.57	1.94	1.72	0.73	0.89	0.81
NO_3^-		0.0000	-	-	-	-	-	-
SO_4^{2-}	2712	0.2517	94.25	75.02	86.41	40.73	28.97	34.56

average was 1.46, which indicates oceanic source. However, the Cl⁻/Na⁺ ratio in Gwaebeopdong were as high as 2.79 and 3.49 in summer and fall, respectively and overall average was 3.08. Probable reasons for the high level of ratio could be not only oceanic source but also anthropogenic sources including Methyl Chloride produced from coal-combustion, wood-ombustion, Polyvinyl Chloride (PVC) combustion, Carbon Tetrachloride of cleaning agents, and Chloroform of chemical solvents(Kang, 1988).

Table 4. Cl⁻/Na⁺ ratios of PM₁₀ in Busan

Station	Season	Cl ⁻ ($\mu\text{g}/\text{m}^3$)	Na ⁺ ($\mu\text{g}/\text{m}^3$)	Cl ⁻ /Na ⁺
Gwaebeopdong	Summer	1.90	0.68	2.79
	Fall	2.13	0.61	3.49
	Mean	2.00	0.65	3.08
Dongsamdong	Summer	2.43	1.85	1.31
	Fall	3.03	1.90	1.59
	Mean	2.74	1.88	1.46

4. Conclusions

24-hr PM₁₀ filter-based sampling were conducted for summer and fall, 2007 in Gwaebeopdong(inland) and Dongsamdong(seashore), Busan and sea salts of the PM₁₀ components at the two sites were characterized in this study.

Sea salt concentrations in PM₁₀ were higher in Dongsamdong(seashore) than in Gwaebeopdong(inland) and likewise contribution ratio of sea salt in Dongsamdong was greater by a factor of three than in Gwaebeopdong. Contribution ratio of sea salt to PM₁₀ in Busan exhibited higher Dongsamdong(9.5%) than in Gwaebeopdong(2.9%) and overall average was 6.2%. In the case of contribution ratio of sea salt to total ion components, Dongsamdong, coastal area, showed higher level compared to Gwaebeopdong, inland area, for summer and fall months and overall contribution of sea salt was also 2.5 times higher in Dongsamdong than in Gwaebeopdong. In terms of

seasonal variability, total ion concentrations were higher in summer than in fall while contribution ratios of sea salt were observed to be higher in fall than in summer.

EF_{sea} of K⁺, Ca²⁺ and SO₄²⁻ were greater than 10, which is indicative of high contribution from anthropogenic sources. EF_{sea} of Mg²⁺ and Cl⁻ composing sea salts denoted strong association with oceanic sources. The analysis on Cl⁻/Na⁺ ratio in PM₁₀ suggested that Gwaebeopdong, an inland site, presented 2.79 and 3.49 in summer and fall, respectively substantiating large contribution of anthropogenic origins. The Cl⁻/Na⁺ ratio in Dongsamdong, coastal area, were 1.31 and 1.59 in summer and fall respectively, indicating effect of oceanic origin on PM₁₀.

The analyses in this study suggested that air quality management and decision making process in Busan, a coastal urban area, must have large implications for sea salts since geographical configuration and natural sources such as sea salt are considered as major determinants in particulate matter formation.

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