# Estimation of the optimal heated inlet air temperature for the beta-ray absorption method: analysis of the PM10 concentration difference by different methods in coastal areas

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**Abstract.** Based on the measurement data of the particulate matter with an aerodynamic diameter of less than or equal to a nominal 10  $\mu$ m (PM10) by the  $\beta$ -ray absorption method (BAM) equipped with an inlet heater and the gravimetric method (GMM) at two coastal sites in Korea, the optimal inlet heater temperature was estimated. By using a gas/particle equilibrium model, Simulating Composition of Atmospheric Particles at Equilibrium 2 (SCAPE2), water content in aerosols was estimated with varying temperature to find the optimal temperature increase to make the PM10 concentration by BAM comparable to that by GMM. It was estimated that the heated air temperature inside the BAM should be increased up to 35~45°C at both sites. At this temperature range, evaporation of volatile aerosol components was minor. Similar (30~50°C) temperature range was also obtained from the calculation based on the absolute humidity which changed with ambient absolute humidity and chemical composition of hygroscopic species.

**Keywords:** PM10;  $\beta$ -ray absorption method; gravimetric method; water content; gas-particle equilibrium; optimal inlet air temperature increase

## 1. Introduction

Airborne particulate matter causes adverse health effects including respiratory and cardiovascular diseases. Respiratory system depends on particle size, shape, and density. An important point is that PM10 can reach the human lungs, and, thus can affect human health significantly. Therefore, the Korean Ministry of Environment is setting PM10 as one of seven ambient air quality standard species.

Reliability of the measurement of ambient trace species is an important issue because measurement data is used to establish a policy. It is imperative to assess reliability of BAM which is the standard method for PM10 measurement in several countries. There are several measurement methods for the mass concentration of particulate matter in the ambient air. The GMM is a direct method in the determination of the mass concentrations, and, thus thought as the most reliable one. On the while, other methods such as the BAM, the tapered element oscillating microbalance (TEOM) system, the

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light scattering and light transmission methods are indirect by using physical characteristic of particles which can be related with the mass concentration. These indirect methods require less man-power than GMM and, thus, have been widely used.

BAM is the standard method for the PM10 measurement in Korea (NIER 2009). However, some studies pointed out reliability problem of measurement results. Thus, several researchers performed reliability assessment of BAM results, mainly in urban area. It is necessary to assess the reliability of BAM in coastal area. It is an instrument based on the assumption that absorption rate of beta-ray increases in proportion to the mass of the collected particles. Hourly PM10 concentration is automatically reported from the increase of absorption of the beta-rays due to particles collected on the filter. It has been known that water absorbed in the particle is not removed in BAM if incoming air is not de-humidified. We can say "positive error" when the mass concentration is overestimated. Thus, this absorbed water can be called "positive error" (Gobeli *et al.* 2008). Recently, a heater system in which the RH (Relative Humidity) of the sampled air is recued by heating the inlet air has been introduced to mitigate the problem.

GMM has been the reference method for the PM10 measurement in the USA (USEPA 1998). In this method, a filter is weighed before and after the sampling to determine the mass change due to the collected particles. The filter is equilibrated in a desiccator under the constant temperature and RH condition for at least 24 hours before and after the sampling to weigh the mass of dried particles.

Through comparing the PM10 mass concentration of dried particles by GMM with that by BAM, we can quantify the measurement error caused by water absorption in BAM. The sampling time of GMM is usually 24 hours, longer than BAM. Thus, volatile species collected on the filter at GMM may be lost on the filter. It is called "negative error" (Turpin *et al.* 2000).

We have described other potential factors that may lead to the PM10 mass concentration differences between the measurement methods in our previous study (Shin *et al.* 2011). These are the differences in (1) cut off diameters, (2) flow rates, (3) materials of the filters, (4) water content of aerosol, and (5) volatilization loss of volatile species of aerosols. Both methods have same standards about (1) and (2). The effect of (3) and (5) is associated with negative error of GMM which should be minor. The detailed explanations of these factors can be found in Shin *et al.* (2011). In this study, the effect of water content of aerosol was mainly discussed.

At Gosan, one of the national background sites in the Republic of Korea and located at seashore as shown in Fig. 1, it was found that PM10 concentration by BAM without an inlet heater was higher than GMM by about 69% and the correlation between them was low (r = 0.57) for the data between 2001 and 2008 (Shin *et al.* 2010). It was suggested that this discrepancy was mainly caused by water absorption in aerosols because there was no inlet heater at BAM and the ambient RH was high, the yearly average of 69% (Shin *et al.* 2010).

The effect of water absorption by high RH at Gosan is quantified by estimating the water content of PM10 based on the particle composition data by GMM using a gas/aerosol equilibrium model, SCAPE 2. The estimated water content was significant, 9.94  $\mu$ g m<sup>-3</sup> or about 50% of the mass concentration difference. However, there was still unaccounted mass concentration discrepancy and, even after accounting for the estimated water content, the correlation between them was not good (Shin *et al.* 2011).

Chang *et al.* (2001) showed that when the ambient RH was high, water absorption to aerosols might lead to higher PM10 concentration at BAM without an inlet heater compared to GMM. Chang and Tasi (2003) estimated water absorption to aerosols at BAM by applying a gas/particle equilibrium model, ISORROPIA, and the simulated GMM mass concentrations were in agreement



Fig. 1 The locations of the Padori (Site A) and Gangneung (Site B) sites

with the concentrations by BAM measured during November 1999 at four stations in Taiwan.

On the contrary, BAM with an inlet heater generally has been reported to have good correlation with GMM. Jung *et al.* (2007, 2009) reported that the PM10 concentration by BAM with an inlet heater was comparable with GMM within  $3\sim6\%$  of the measurement mass concentration and the correlation between them was high (r = 0.98) at a site in Incheon which was located about 10 km from seashore. Still, even with an inlet heater, BAM has shown inconsistent result. At Bakreungdo which is located at seashore, the correlation between BAM with inlet heater and GMM was generally high (r = 0.9). However, the correlation between them became lower (r = 0.8) in summer, and the PM10 concentration of BAM was higher than GMM at high RH (> 80%) cases (Kong *et al.* 2010).

In this study, (1) the PM10 concentrations by BAM with an inlet heater and GMM at two coastal sites with varying distance from seashore are compared, (2) based on comprehensive chemical composition data for PM10, water content at both sites are estimated, and (3) the optimal inlet heater condition is suggested for the accurate measurement for BAM.

### 2. Data

PM10 measurements were made at two coastal sites; Padori at the west coast and Gangneung at

the east coast as shown in Fig. 1. Padori site (Site A) is one of the national background and acid precipitation measurement sites, and located 200 m from seashore. Thus, it can be affected by seasalt particles and high RH air directly. There are few local air pollutants' emission sources nearby. Gangneung site (Site B) is located 4.5 km from seashore. This site is affected by the air mixed with natural and anthropogenic pollutants (Park *et al.* 2011). The sampling periods were between April and June (spring period), and July and August (summer period), 2009.

At both sites, the MetOne BAM1020 was used. The air entered the instrument through an impactor designed to have a 50% collection efficiency for PM10. On the filter in BAM, PM10 were collected for 1 hr and then a new clean filter surface was moved to the sampling area on which particles were collected.

The "Smart Heater," as called by the manufacturer was covering the inlet tube of BAM. The manufacturer suggested the RH of the inside of BAM be 45%. The RH sensor was located below the filter paper. Since ambient air samples were collected at a flow rate of 16.7 liter per minute (LPM), the radius of the inlet tube was 0.6 inch, and the length of the heater was 4 inch, the residence time through the Smart Heater was approximately 0.3 sec, enough for water being in equilibrium between the gas and particle phase (Kim *et al.* 1993a).

Once the heater system began to operate, the air temperature after the inlet tube rose and RH got lower. This lowered RH made the water vapor in the aerosols to evaporate. When the ambient RH was high, the RH difference might be not high enough for water in aerosols would not evaporate sufficiently. Another thing to note is that if we assume that the amount of water evaporated from particles be negligible compared to the amount of water vapor in the air, which is usually true, the absolute humidity in the air is still the same irrespective of the air temperature change.

The GMM consisted of a PM10 impactor (R&P, PM10 inlet 57-00596, USA) with a cut size 10  $\mu$ m at a flow rate of 16.7 LPM, a Teflon filter holder for 47 mm filters (Zefluor, PTFE 47 mm, 2  $\mu$ m pore size, Pall Co., USA). Calibration of the flow rate was conducted once a week during the measurement. Clean filters were equilibrated in a desiccator at 40 ± 5% RH and 20 ± 3°C for at least 24 hrs and, then weighed on a microbalance (Satorius CP2P-F). After the 24 hr collection period, the particle laden filter samples were retrieved, equilibrated, and reweighed.

Eight ions (Cl<sup>-</sup>, NO<sub>3</sub><sup>-</sup>, SO<sub>4</sub><sup>2-</sup>, Na<sup>+</sup>, NH<sub>4</sub><sup>+</sup>, K<sup>+</sup>, Ca<sup>2+</sup>, and Mg<sup>2+</sup>) were analyzed by an ion chromatograph (Metrohm 861). Details on the sampling and analysis were given in Park *et al.* (2011). To ensure the quality of the data, ion balance was used to check the validity of the data. The data with the ratio of the sum of the cation concentrations to the anion concentrations being within 30% were used for further data analysis (Park *et al.* 2004). The number of daily data of GMM reduced from 62 to 55 at Site A, from 59 to 47 at Site B after the quality control process. Information on the sampling periods is given in Table 1 along with the chemical composition data, average ambient temperature and RH data.

Hourly continuous automatic measurement data of BAM were converted into the daily mean data from 9 AM to 8 AM next day which was the sampling schedule of GMM.

Meteorological parameters at both sites were measured by conventional instruments which were calibrated and checked by the Korea Meteorological Administration once every year.

### 3. Model

Details of the gas/particle equilibrium model used, SCAPE2, are given elsewhere (Kim et al.

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(a) Padori (Site A)												
Average	GMM	BAM	$Na^+$	$\mathrm{NH_4}^+$	$K^+$	Ca <sup>2+</sup>	$Mg^{2+}$	Cl	NO <sub>3</sub> -	SO <sub>4</sub> <sup>2-</sup>	Temp.	RH
4/23-6/10(n=34)	51.09	63.92	1.21	4.81	0.63	0.55	0.24	0.86	3.93	12.57	14.48	88.72
7/25-8/30(n=21)	24.52	37.97	0.67	1.87	0.18	0.19	0.13	0.22	0.81	5.83	23.77	93.15
Total(n=55)	40.94	54.01	1.00	3.69	0.46	0.41	0.20	0.62	2.74	10.00	18.03	90.41
(b) Gangneung	(Site B)											
Average	GMM	BAM	$Na^+$	$\mathrm{NH_4}^+$	$K^+$	Ca <sup>2+</sup>	$Mg^{2+}$	Cl	NO <sub>3</sub> -	SO <sub>4</sub> <sup>2-</sup>	Temp.	RH
4/23-6/10(n=20)	47.49	49.29	0.53	2.89	0.30	1.18	0.16	0.48	3.25	7.45	16.91	54.99
7/25-8/30(n=27)	24.31	25.33	0.40	1.33	0.14	0.59	0.09	0.24	1.22	4.64	23.17	77.50
Total(n=47)	34.17	35.53	0.46	1.99	0.21	0.84	0.12	0.34	2.08	5.84	20.51	67.92

Table 1 Average PM10 mass and ionic concentrations, temperature and relative humidity (RH) at (a) Padori and (b) Gangneung (Park *et al.* 2011)

1993a, b, Kim and Seinfeld 1995, Meng *et al.* 1998, Choi and Kim 2010). For a closed multiphase system in chemical equilibrium at constant temperature T and pressure, the total Gibbs free energy of the system should be at a minimum.

$$\sum v_{ij}\mu_i = 0 \tag{1}$$

Where  $v_{ij}$  is the stoichiometric coefficient of the *i*th species in the *j*th reaction,  $\mu_i$  is the chemical potential of species *i* given in Eq. (2)

$$\mu_i = \mu_i'(T) + RT \ln a_i \tag{2}$$

Where  $\mu_i^{\circ}(T)$  is the standard chemical potential for the *i*th species at temperature T in Kelvin, and  $a_i$  is the activity of the species *i* given in Eq. (3)

$$a_i = \gamma_i m_i \tag{3}$$

Where  $\gamma_i$  is its activity coefficient and  $m_i$  is the molality of species *i*. By substituting Eq. (2) and (3) into (1), Eq. (4) is obtained

$$\exp\left[-\frac{1}{RT}\sum v_{ij}\mu_i\right] = \prod a_i^{vij} \equiv K_j \equiv \Pi_i (\gamma_i m_i)^{vi}$$
(4)

Where  $K_j$  is the equilibrium constant of the *j*th reaction. Therefore, estimating accurate activity coefficient is essential to obtain accurate equilibrium concentration of the species with the given equilibrium constants. In SCAPE 2, three methods are available and the Kusik and Meissner (K-M) method is selected for calculating activity coefficient in this study.

For water content, the Stokes, Robinson, and Zdanovskii (ZSR) method is used in SCAPE2 because of simplicity.

$$W = \sum_{i} \frac{M_{i}}{m_{io}(a_{w})}$$
(5)

Where  $a_w$  is water activity, equal to the RH expressed as a fraction. And  $m_{io}(a_w)$  is the molality of the binary solution at the desired water activity  $a_w$  of the multi-component solution.  $M_i$  is the molar concentration of species *i* in the air (mol m<sup>-3</sup> air) and W is the mass concentration of water in the aerosol (kg water m<sup>-3</sup> air).

In this mode the input data for SCAPE2 are ammonium (particulate  $NH_4^+$ ), nitrate (particulate  $NO_3^-$ ), chloride (particulate  $CI^-$ ), sulfate ( $SO_4^{2-}$ ), carbonate ( $H_2CO_3$ ), calcium ( $Ca^{2+}$ ), magnesium ( $Mg^{2+}$ ), potassium ( $K^+$ ), sodium ( $Na^+$ ), RH, and ambient temperature (*T*). The output results are the equilibrium gas phase concentrations of HCl, HNO<sub>3</sub>, and NH<sub>3</sub>, and particulate water content and acidity. Because we did not know gaseous species concentrations, SCAPE2 model was run in "aerosol phase only" mode.

It is suggested that organic hygroscopic species might play a role. However, organic species are not accounted for at SCAPE2. Since water soluble organic carbon (WSOC) fraction in PM10 could be high (Zappoli *et al.* 1999, Yu *et al.* 2002, Lee *et al.* 2008), it is possible that the total water content in aerosol could be underestimated by not considering WSOC.

### 4. Results

### 4.1 General characteristics

As shown in Table 1, the mass and ionic concentrations at Site A were higher than Site B at both sampling periods. Note that the RH at Site A was also higher than Site B reflecting the distances from seashore. Especially in summer, the mean RH at Site A was very high, more than 90%. Though the area has been frequently covered by fog, this RH value was rather high. Since the instruments were checked by the Korean Meteorological Administration based on their quality control process, we have used the meteorological parameter values at both sites without any correction. As will be discussed in section 4.3, we did carry out a sensitivity analysis of the modeling results on the variation of RH at Site A.



Fig. 2 Comparison of the PM10 concentrations between BAM and GMM at (a) Padori and (b) Gangneung

The correlation of the PM10 concentrations by two methods were good at both sites (r = 0.95 at Site A and r = 0.99 for Site B) as shown in Fig. 2. It was different from the result with low correlation coefficient (r = 0.57) at Gosan where no inlet heater was installed in BAM. The slopes of the scattergrams at both sites were almost 1. At Site B, the PM10 mean concentration by BAM was 35.53  $\mu$ g m<sup>-3</sup> while that by GMM was 34.17  $\mu$ g m<sup>-3</sup>. The difference between the PM10 mean concentration. However, at Site A, the mean PM10 concentration by BAM was 54.01  $\mu$ g m<sup>-3</sup> while that by GMM was 40.94  $\mu$ g m<sup>-3</sup>, with the difference of 13.07  $\mu$ g m<sup>-3</sup> or 32% of the mean GMM concentration. It suggests that the BAM measurement at Site A might contain positive error.

# 4.2 Analysis of the factors causing the mass concentration difference between two methods

To find the reason(s) of the large difference between BAM and GMM at Site A, we classified the PM10 concentrations between two methods according to RH since the previous study identified water absorbed in aerosols as the major reason for the difference (Shin *et al.* 2011). The RH value of 80% has been widely used to distinguish high RH case (Tsai and Cheng 1996, Kong *et al.* 2010). Thus, we chose 80% RH as a criterion.

As shown in Fig. 3, at Site B, the mean PM10 concentration under 80% RH by BAM was higher than that of GMM and the average relative error (ARE) between them was 6%. Meanwhile, over 80% RH, it showed the opposite trend and ARE was 1%. At Site A, when RH was higher than 80%, the PM10 concentration by BAM was also higher than that of GMM but ARE was high, 33%. Meanwhile, under 80% RH, it also showed the opposite trend but ARE was 26%. At Site B, the correlation coefficient (r) between them was 0.96 for the cases over 80% RH while the r value was 0.99 for the cases under 80% RH. Also at Site A, that for the cases the r value (0.95) for the cases over 80% RH was lower than that for the cases under the 80% RH (0.97).

At Site B, the difference between the PM10 mean concentrations of two methods over 80% RH was lower than that under 80% RH. Since Site B is far from seashore compared to Site A, it can be considered that Site B has the characteristics of urban site. On the other hand, Site A which is near to seashore showed that the PM10 concentration difference and relationship between BAM and GMM became larger and worse at high RH (> 80%). Thus, absorbed water on the aerosols, called "positive error" might be a major reason for the concentration difference even for a BAM with an inlet heater.

To identify whether the air mass trajectory causes the PM10 mass concentration difference between two methods, backward trajectory analysis was carried out. Backward trajectory was estimated by using the Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model (Draxler and Rolph 2003). The trajectories starting at Site A and Site B at the altitude of 1,500 m were estimated for 72 hours. It turned out that the pathways of air masses at two sites were almost same.

If humidity in the air be the major factor for the PM10 concentration difference between two methods, that difference would be more prominent for the air mass passing through sea. Thus, the trajectories were divided in four cases. Case 1 was the trajectories started from continent 72 hours ago and not passing through sea, Case 2 was the trajectories from continent with passing through sea, Case 3 was from the North Pacific without passing over land area, and Case 4 was from Japan or Korea with patterns of local circulation.



Fig. 3 Comparison of the PM10 concentrations between BAM and GMM according to RH (< 80% and > 80%)

At Site B, there was no distinct concentration difference or correlation coefficient change among the cases. But, at Site A as shown in Fig. 4, The correlation between two methods for Case 1 was the best (r = 0.97) and ARE was 18%. The correlation between two methods in Case 3 from the North Pacific was the lowest (r = 0.85) and ARE was 51%. Thus, this result also supports the hypothesis of the absorbed water in aerosols being not sufficiently removed with the inlet heater for the high RH cases.

# 4.3 Estimation of the optimal heated air temperature in BAM to minimize the effect of water absorption

Although an inlet heater has been used to minimize the error by water absorption at both sites, it is likely that the ambient RH at Site A was too high to be effectively controlled by the inlet heater. The water content in aerosols is determined by two major factors; relative humidity in the air and chemical composition and concentrations of aerosols (Kim *et al.* 1993a, b, Meng *et al.* 1995).



Fig. 4 Comparison of the PM10 concentrations between BAM and GMM according to the backward trajectories (Padori)

Thus, we quantified the water content in aerosols with the variations of RH and heated air temperature by using a gas/particle equilibrium model, SCAPE2 (Kim *et al.* 1993a, b, Kim and Seinfeld 1995, Meng *et al.* 1998). Based on the calculated aerosol water content, the effect of ambient humidity during measurement by BAM was quantified.

If the heated air temperature after the inlet heater be known either by measurement or by specification from the manufacturer, we can estimate the remaining water content in aerosols on the filter in BAM. However, at present, the heated inlet air temperature is not known. The manufacturer only suggests that the filter air temperature be higher than the ambient temperature by more than  $5^{\circ}$ C (BAM1020 Training manual, Met One Instruments).

Thus, we first carried out a reverse estimation on the heated air temperature by comparing the measured PM10 concentration by BAM and the sum of the PM10 concentration by GMM and the estimated water content on the chemical composition data by GMM (defined as simulate GMM) by varying the modeling input of the ambient temperature.

Before further analysis, two things should be considered. First, it has been reported that the water content in aerosols by GMM after equilibration of the filter under dry conditions is not zero (Kajino



Fig. 5 Estimation of the optimal filter air temperatures in BAM based on the comparable water contents between BAM and GMM

*et al.* 2006). They estimated that the water content due to various reasons accounted for  $1\sim5$  wt% of the PM10 concentration by GMM (Kajino *et al.* 2006). Considering it, we estimated the heated air temperature with the assumption that  $1\sim5\%$  of the PM10 concentration by GMM was water which was  $0.40\sim2.00 \ \mu\text{g} \text{ m}^{-3}$  and  $0.34\sim1.71 \ \mu\text{g} \text{ m}^{-3}$  at Site A and B, respectively (shown as gray area in Fig. 5).

Second, evaporation of volatile species in aerosols should be considered since we were heating the air surrounding the filter on which particles were collected.

Since we did not measure the gaseous species concentrations, we did not know the degree of evaporation of volatile aerosol component in BAM quantitatively. However, we carried out a gas/

particle equilibrium model based on the measured particle phase concentrations of volatile species. It turned out that between 25 and 70°C, the estimated gas phase concentrations of volatile species were almost same (Site A: 0.0007  $\mu$ g m<sup>-3</sup>, Site B: 1.16  $\mu$ g m<sup>-3</sup>) suggesting that no significant evaporation of volatile species in that temperature range.

The mean ambient temperatures for the whole sampling period were 18.0 and 20.5°C at Site A and B, respectively. With above considerations, the estimated heated air temperatures for which the PM10 concentration by BAM and the simulate GMM PM10 concentration being comparable were 25 and 30°C at Site A and B, respectively. At those temperatures, the estimated water contents at Site A and B were 13.10 and 2.58  $\mu$ g m<sup>-3</sup>, respectively. This large difference between two sites was mainly caused by the higher RH and concentrations of hygroscopic ions at Site A as shown in Table 1.

This result demonstrates that with the same inlet heater, the degrees of the water evaporation were different depending on RH and aerosol chemical composition. So, we estimated the optimal heated air or filter air temperature surrounding the filter in BAM in which the water content in aerosols of BAM becomes comparable that in PM10 by GMM.

Fig. 5 shows the water content in aerosols according to the surrounding air temperature. With the allowance of the water content in PM10 by GMM (shown as gray areas in Fig. 5), the optimal heated air temperature at both sites was estimated as 35~45°C range. In other words, to minimize the PM10 concentration measurement difference by water absorption in aerosols between BAM and GMM, the inlet heater should be controlled to maintain a filter temperature of 35~45°C at both sites irrespective of ambient temperature and aerosol composition.

Since the mean RH at Site A was very high, especially in summer, we carried out a sensitivity analysis by using the ambient RH data from Anmyondo site which was a monitoring site operated by Korean Meteorological Administration and about 30 km south of Site A. The same optimal heated air temperature range was obtained for that simulation.

While the result shown in Fig. 5 is essential for the effective BAM operation, it depends on several variables. For example, RH is varying with ambient temperature. On the contrary, as pointed out in Data section, absolute humidity is constant. Thus, we estimated the optimal heated air temperature according to the variation of the absolute humidity estimated from the ambient



Fig. 6 Estimation of the optimal filter air temperatures according to absolute humidity (g m<sup>-3</sup>) in BAM at (a) Padori and (b) Gangneung

temperature and RH data at each sampling day.

Fig. 6 shows the optimal heated air temperature according to the absolute humidity interval. With increasing absolute humidity, the optimal heated air temperature increased at both sites. However, as the absolute humidity increased, with the same absolute humidity value, the optimal temperature at Site A became higher  $(30 - 50^{\circ}C)$  than that at Site B  $(30 - 45^{\circ}C)$  reflecting higher concentrations of hygroscopic ionic species at Site A.

### 5. Conclusions

Reliable measurement of air pollutants' concentrations is a first step for the establishment of air quality control policy. It was found that the PM10 mass concentration by the BAM without inlet heater system has been higher than that by GMM at Gosan (Shin *et al.* 2010). Some previous studies mentioned that BAM has shown inconsistent result due to high ambient humidity.

To further understand the effect of ambient humidity to the PM10 mass concentration, the PM10 concentrations by BAM and GMM at two coastal sites, Padori (Site A) and Gangneung (Site B) were measured between April and June, and between July and August 2009 along with chemical composition of PM10.

Though the correlations between them were high at both sites, it was found that the PM10 concentration by BAM was higher than GMM by 13.07  $\mu$ g m<sup>-3</sup> (32%) at Site A. On the other hand, at Site B, the difference was only 1.36  $\mu$ g m<sup>-3</sup> (5%). Based on the first principle of equilibrium, it is postulated when the ambient RH was high, the raised temperature by inlet heater might be not enough to reduce the water absorbed to aerosols in BAM sufficiently. In other words, the degree of the temperature increase by the inlet heater was insufficient driving force to reduce water content in the aerosols in BAM at Site A at the high RH cases.

Other analyses were carried out to further confirm whether water absorbed in aerosols by BAM was the main reason for the difference. These are the classification of the PM10 concentrations between two methods according to RH and backward trajectories. These results also suggested that ambient humidity played a significant role in the PM10 concentration difference between two methods, especially at Site A where the RH values and the concentrations of hygroscopic species were higher than Site B.

To estimate the optimal heated air temperature after passing through the inlet heater, the present heated air temperature was estimated based on the assumption that the PM10 concentration difference was solely due to water content difference between two methods at both sites. A gas/particle equilibrium model, Simulating Composition of Atmospheric Particles at Equilibrium 2 (SCAPE2) was used to estimate aerosol water content. It was found that the inlet heater increased the air temperature from 18.0°C to 25°C at Site A and from 20.5°C to 30°C at Site B. With the same heat source, this different temperature increase was due to different humidifies and chemical compositions at two sites.

Therefore, we estimated the optimal heated air temperature surrounding the filter in BAM in which the water content in aerosols becomes comparable that in PM10 by GMM. The optimal heated air temperature at both sites was estimated as  $35{\sim}45^{\circ}$ C range. However, based on the absolute humidity concept, the estimated optimal temperature at Site A became higher ( $30{\sim}50^{\circ}$ C) than that at Site B ( $30{\sim}45^{\circ}$ C) reflecting higher RH and concentrations of hygroscopic species at Site A.

This result suggests that the inlet heater system in BAM should be controlled to account for both

ambient humidity and chemical composition to get comparable PM10 concentration by GMM. This point becomes more prominent since BAM is considered as one of the equivalent methods for PM2.5 in several countries. Since hygroscopic ion fraction in PM2.5 is higher than PM10, the relative difference between BAM and GMM would be higher in PM2.5 measurement than PM10. Thus, it is imperative to control the inlet heater system in BAM more accurately.

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