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# Assessment of the clear-sky bias issue using continuous PM<sub>10</sub> data from two AERONET sites in Korea

Yongjoo Choi, Young Sung Ghim\*

Department of Environmental Science, Hankuk University of Foreign Studies, Yongin 449-791, Korea

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## ABSTRACT

A bias in clear-sky conditions that will be involved in estimating particulate matter (PM) concentration from aerosol optical depth (AOD) was examined using PM<sub>10</sub> from two Aerosol Robotic Network sites in Korea. The study periods were between 2004 and 2007 at Anmyon and between 2003 and 2011 at Gosan, when both PM<sub>10</sub> and AOD were available. Mean PM<sub>10</sub> when AOD was available (PM<sub>AOD</sub>) was higher than that from all PM<sub>10</sub> data (PM<sub>all</sub>) by 5.1 and 9.9 μg/m<sup>3</sup> at Anmyon and Gosan, which accounted for 11% and 26% of PM<sub>all</sub>, respectively. Because of a difference between mean PM<sub>10</sub> under daytime clear-sky conditions (PM<sub>clear</sub>) and PM<sub>AOD</sub>, the variations in ΔPM<sub>10</sub>, the difference of PM<sub>all</sub> from PM<sub>clear</sub> rather than from PM<sub>AOD</sub>, were investigated. Although monthly variations in ΔPM<sub>10</sub> at the two sites were different, they were positively correlated to those in ΔT, similarly defined as ΔPM<sub>10</sub> except for temperature, at both sites. ΔPM<sub>10</sub> at Anmyon decreased to a negative value in January due to an influence of the Siberian continental high-pressure system while ΔPM<sub>10</sub> at Gosan was high in winter due to an effect of photochemical production at higher temperatures than at Anmyon.

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## Introduction

Particulate matter (PM) is a major target of air quality management in many countries (McMurry et al., 2004; World Health Organization, 2006a), because high concentration of PM causes serious health problems in the cardiovascular system as well as the respiratory system (Laden et al., 2000; Mar et al., 2005). PM is also a main ingredient of smog causing visibility impairment by scattering and absorbing light (Watson, 2002; Pitchford et al., 2007). As a result, PM is monitored at many locations in the world with standardized methods in all weathers, independent of rainy and cloudy conditions. However, this monitoring generally targets populated areas and is scarce in remote areas even in developed countries. For PM in vast areas of the remaining regions,

information is limited because measurements have only been performed in the form of campaigns with special purposes (Bates et al., 1998; Ramanathan et al., 2001; Haywood et al., 2003).

Despite a high cost of installation and operation, monitoring stations in remote areas have been deployed to study long-range transport and/or background conditions of air pollutants. However, advancement in satellite and ground-based remote sensing of aerosols has provided a new area of research for monitoring not only regional PM distributions but the global one (Gupta et al., 2006; van Donkelaar et al., 2010). Aerosol optical depth (AOD), derived from satellite or ground-based observations, is directly related to particle mass loading, and its association with surface-level PM mass concentration gives a valuable tool to monitor PM (Wang

\* Corresponding author. E-mail: [ysghim@hufs.ac.kr](mailto:ysghim@hufs.ac.kr) (Young Sung Ghim).

and Christopher, 2003; Engel-Cox et al., 2006; Gupta and Christopher, 2008a). These remote sensing technologies have obvious advantages, such as ease of acquiring data and cost effectiveness of monitoring PM, where *in situ* measurements are scarce or lacking altogether. Despite many advantages for monitoring air quality, these techniques have a crucial restriction in that AOD is available only under daytime clear-sky conditions (Gupta and Christopher, 2008b; Christopher and Gupta, 2010).

The Aerosol Robotic Network (AERONET) is a representative network among ground-based aerosol remote sensing applications, which has been used to measure aerosol optical properties in key locations around the world (Holben et al., 1998, 2001; Dubovik et al., 2002; Omar et al., 2005). The sun photometer of AERONET measures the AOD of an entire column, as with the Moderate Resolution Imaging Spectroradiometer (MODIS). However, the measurement of the sun photometer has an advantage of high temporal resolution, 15 min intervals or every hour, depending on the measurement scenario, while MODIS measures only twice a day, on Terra in the morning and on Aqua in the afternoon. Although the ground-based networks were originally designed to validate the satellite observations, the results from these networks are regarded as ground truths in many research works, such as evaluation and improvement of air quality modeling (Carmichael et al., 2009; Chin et al., 2009).

Various studies have investigated the relationship between AOD from satellites and PM at the surface. The correlation coefficients between the two typically ranged between 0.5 and 0.7 (Hoff and Christopher, 2009), but could be higher than 0.90 when PM concentrations were divided into several bins and the bin-averaged concentrations were correlated with AOD (Wang and Christopher, 2003; Hutchison et al., 2005; Gupta et al., 2006). Only a few studies have paid attention to AOD from AERONET to obtain information on PM (Corbin et al., 2002; Schaap et al., 2009) despite a higher temporal resolution than that from satellite. This may be because the data from stationary AERONET sites are not well suited for obtaining information on regional-scale variations, compared with those from satellite. Research works examining the clear-sky restriction are scarce even with satellite data. Christopher and Gupta (2010) compared mean PM<sub>2.5</sub> for all days (ALLPM) and that for clear days (CLEARPM) from monitoring stations in the United States in 2006. The mean difference for each Environmental Protection Agency (EPA) region in each season was less than  $\pm 2.5 \mu\text{g}/\text{m}^3$ , and they concluded that cloud cover was not a major problem in determining monthly and yearly mean PM<sub>2.5</sub> from satellite data.

It is known that clear-sky conditions often result from high-pressure systems, associated with low wind speed, sinking air motion, reduced vertical motion, and enhanced photochemistry (National Research Council, 1991). These conditions lead to the accumulation of pollutants and higher PM<sub>2.5</sub> at the ground (Baumann et al., 2003; Cabada et al., 2004; Fine et al., 2008) and a positive bias of CLEARPM compared to ALLPM (Christopher and Gupta, 2010). However, Corbin and Denning (2006) indicated that higher mixing height and air temperature due to strong solar radiation lowered pollutant concentrations on clear days, particularly in the summer. Furthermore, in both Christopher and Gupta (2010) and Corbin

and Denning (2006), wintertime variations were different from those in summer.

In this study, we examined a bias in clear-sky conditions for estimating PM from AOD. Differing from previous studies using satellite data, we used data from AERONET sites, Anmyon and Gosan, in Korea (see Fig. 1 for their locations) where PM<sub>10</sub> concentrations were also available. Because both AOD and PM<sub>10</sub> concentration were provided at the same sites, we could separate the conditions when AOD was available more precisely than using the satellite data with which those conditions were distinguished only on an area and/or daily basis.

## 1. Methods

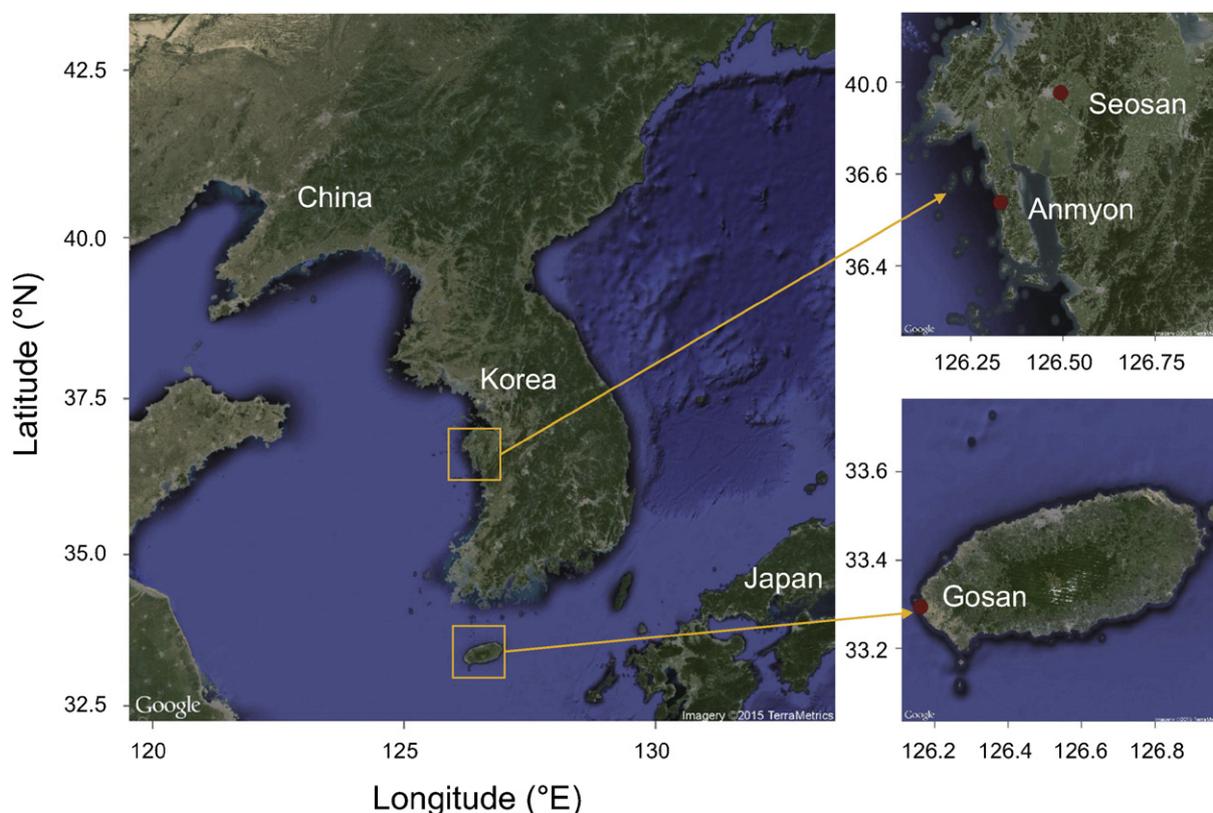
### 1.1. Sun/sky radiometer and PM<sub>10</sub> measurements

The radiometers at Anmyon and Gosan (CE 318, CIMEL Electronique) have a 1.2° full field of view and are mounted on preprogrammed trackers, enabling direct sun measurements as well as sky radiance (diffuse) measurements. A filter wheel allows measurements up to eight spectral bands (340, 380, 440, 500, 675, 870, 940, and 1020 nm). All of these spectral bands are used in the direct sun measurements, but only four of them (440, 675, 870, and 1020 nm) are used for the sky radiance measurements (Holben et al., 1998). The direct sun measurements require about 8 sec to scan all eight wavelengths (repeated 3 times within 1 min). The sky radiance measurements in almucantar geometry are made at optical air masses of 4, 3, 2, and 1.7 in the morning and afternoon and once per hour in between (Eck et al., 2010). The spectral AOD data are screened for clouds following the methodology of Smirnov et al. (2000). AODs are available from both direct sun measurements and inversion retrievals using diffuse measurements. We used level 2.0 (quality assured) AODs from direct sun measurements as well as those from inversion retrievals.

PM<sub>10</sub> concentration is measured using a FH 62 C14 Continuous Particulate Monitor (Thermo Scientific). It is operated by the principle of beta-ray attenuation through particles collected on a movable filter tape. The measuring position is continuously fed with a new section of the filter tape. Then, ambient air is pulled through the inlet and sample tube to induce the deposition of particles on the filter through which the beta beam passes. The intensity of the beta beam expressed as the count rate from the detector is inversely proportional to the particle mass loaded on the filter. The minimum detection limit for PM<sub>10</sub> is  $<1 \mu\text{g}/\text{m}^3$  with the precision of  $\pm 2 \mu\text{g}/\text{m}^3$  (Ahmed et al., 2015).

### 1.2. Data use and analysis

In Korea, four AERONET sites have been operated since the early 2000s. Among them Anmyon (36.54°N, 126.33°E) and Gosan (33.29°N, 126.16°E) are the two sites with the longest operation periods (Kim et al., 2007, 2008, 2010). In addition, Anmyon is a GAW (Global Atmosphere Watch) station of the World Meteorology Organization, and Gosan was a supersite of many international campaigns including ACE (Aerosol



**Fig. 1** – Locations of the two Aerosol Robotic Network (AERONET) sites, Anmyon and Gosan in Korea. Scales of the two panels on the right-hand side are the same; the sizes are  $1^\circ \times 1^\circ$  in latitude and longitude. The Seosan weather station is located about 30.3 km northeast of Anmyon.

Characterization Experiments)-Asia (Huebert et al., 2003). Anmyon was operated from October 1999 to November 2007, and Gosan has been operated since April 2001.

At both sites,  $PM_{10}$  concentration has been measured since April 2003. Thus, the study periods for which both AOD and  $PM_{10}$  concentration were available were from April 2004 to November 2007 for Anmyon and April 2003 to June 2011 for Gosan. To determine the clear-sky bias that will be involved in estimating PM from AOD, we first investigated the difference between mean  $PM_{10}$  from  $PM_{10}$  data when AOD was available ( $PM_{AOD}$ ) and that from all  $PM_{10}$  data available ( $PM_{all}$ ). Next, we examined the difference between mean  $PM_{10}$  under daytime clear-sky conditions (regardless of AOD availability) ( $PM_{clear}$ ) and  $PM_{all}$ . This was because we could not fully address the clear-sky bias using  $PM_{AOD}$  since it was revealed that AOD was not provided for a number of clear-sky hours in the daytime, which will be discussed later.

Although both Anmyon and Gosan are operated by the Korea Meteorological Administration, meteorological data are available only at Gosan. Thus, meteorological data observed at Seosan, about 30 km to the northeast (Fig. 1), were used for Anmyon. Seosan is the nearest weather station to Anmyon, and the terrain is simple with a small bay in between. Wind speed, air temperature, and cloud amount (from 0 to 10, representing the amount of sky cover) were used in this study. While wind speed and air temperature data were

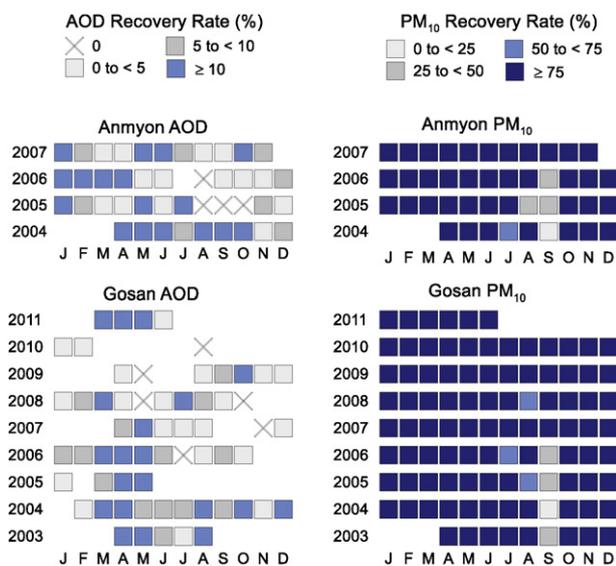
provided every hour, cloud amount data were provided at 3 hr intervals.

## 2. Results and discussion

### 2.1. Data recovery rates

To compare the characteristics of AOD data with those of  $PM_{10}$  data, the data recovery rate, which is defined as the ratio of the number of valid data divided by the total number of possible data during the study period, was calculated. Because  $PM_{10}$  concentration was measured hourly, the data recovery rate of AOD was also calculated on an hourly basis. Therefore, when two AODs were available on a certain day but within the same hour period, the recovery rate on that day was 1/24, not 2/24. Fig. 2 shows the recovery rates of AOD and  $PM_{10}$  concentration at Anmyon and Gosan by month during the study period. The recovery rate of  $PM_{10}$  concentration was more than 75%, except for September from 2004 to 2006 at Anmyon and from 2003 to 2006 at Gosan, which was less than 27%. Reasons for the particularly low recovery rates in the specific months at the two sites are unknown. The recovery rate of  $PM_{10}$  concentration at Anmyon was also similarly low in August 2005.

Compared with  $PM_{10}$ , the recovery rate of AOD was much lower and highly variable, especially at Gosan. Monthly



**Fig. 2 – Monthly recovery rates of AOD and  $PM_{10}$  at Anmyon and Gosan during the study period.**

recovery rates of AOD were less than 19% and 21% with mean values of 7.3% and 4.4% at Anmyon and Gosan, respectively. Furthermore, there were no AODs for 11% and 44% of total months at Anmyon and Gosan, respectively, during the study period. The reasons for the absence of AODs for an entire month could vary. One reason was that no measurements were made because the instrument was taken out of service for repair or calibration. In this case, there were no raw data for both direct sun and inversion products. Persistent precipitation throughout the entire month could yield the same results, but highly unlikely even in the monsoon season in Korea. We expressed as blanks in Fig. 2 for those cases when no measurements were made. At Gosan, there are many blanks during the study period since the instrument was frequently taken out of service for repairs.

In some months, there were no AODs for an entire month although the presence of raw data for direct sun and inversion products indicated that the measurements were made. For this case, all AOD data from both direct sun measurements

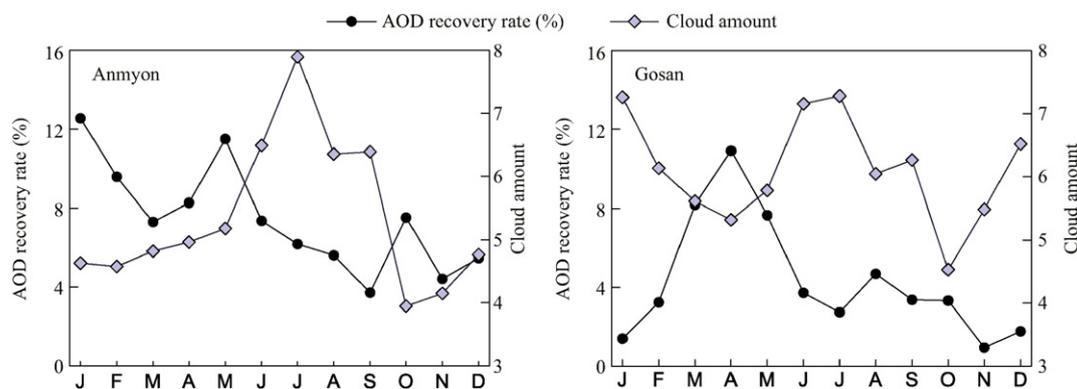
and inversion retrievals should be discarded during the quality assurance processes ([http://aeronet.gsfc.nasa.gov/new\\_web/Documents/Quality\\_Control\\_Checklist.pdf](http://aeronet.gsfc.nasa.gov/new_web/Documents/Quality_Control_Checklist.pdf)). Various instrumental malfunction and optical contamination may be presumed, but it is uncertain that no AODs were provided for an entire month or more due to these reasons.

Fig. 3 shows the variations in monthly mean recovery rate of AOD. The recovery rates at the two sites were quite different in January and February; they were high at Anmyon but low at Gosan. Monthly variations in cloud amount are also shown because AOD is available only under clear-sky conditions. We can interpret many variations in the recovery rate of AOD in terms of cloud amount although the correlation coefficients were not high,  $-0.30$  and  $-0.41$  for Anmyon and Gosan, respectively. To begin with, high recovery rates in January and February and low recovery rates in summer at Anmyon were due to low and high cloud amounts, respectively. Cloud amounts at Gosan were generally higher than those at Anmyon, and thus, the recovery rate at Gosan was lower. Different from Anmyon, the cloud amount at Gosan was higher in winter (similarly to that in summer), and the recovery rate in winter was lower than that in summer.

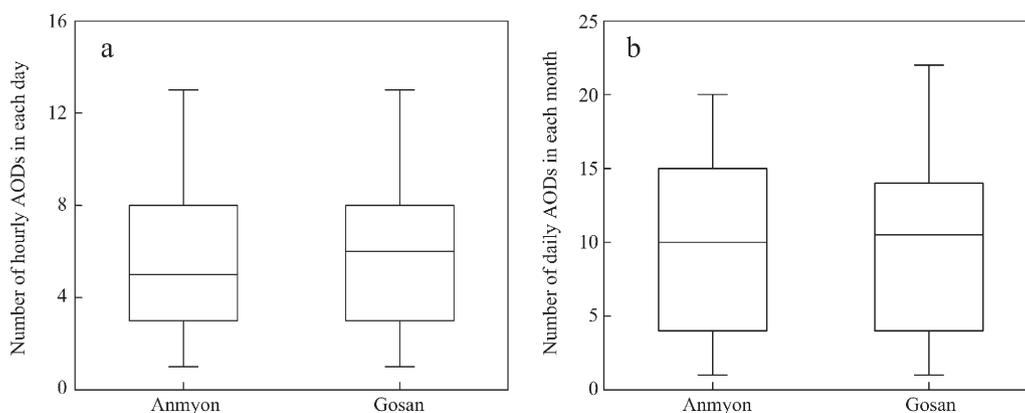
## 2.2. Differences between $PM_{AOD}$ and $PM_{all}$

The difference between  $PM_{AOD}$  and  $PM_{all}$  was investigated on a daily basis because short-term risk for PM has been assessed using daily averages and short-term standards for PM in most countries are set based on this average (World Health Organization, 2006a, 2006b). We calculated daily  $PM_{10}$  when at least 75% of hourly data were valid among 24 hr a day according to the statistical method adopted by the Ministry of Environment of Korea (National Institute of Environmental Research, 2011). This criterion of 75% was also used in the calculation of monthly mean  $PM_{10}$ , to be discussed later. However, the number of occurrences of clear-sky conditions was generally too small for this criterion. Fig. 4a shows the number of hourly data on each day when AODs were available. The median numbers of hourly data at Anmyon and Gosan were five and six, respectively.

It is noticeable that the median numbers of hourly data from inversion retrievals were only two at both sites (not shown). Nevertheless, the AERONET site (<http://aeronet.gsfc>.



**Fig. 3 – Monthly variations in AOD recovery rate and cloud amount.**



**Fig. 4 – Numbers of hourly AODs in each day and daily AODs in each month. Both direct sun and inversion product AODs were counted.**

nasa.gov/) separately provides daily AODs not only from direct sun measurements but also from inversion retrievals whenever AODs are available regardless of the number. It provides monthly AODs in a similar manner, calculated from daily means. The monthly AOD from ‘all points’ AODs is also provided but in the name of “weighted average”, probably because the weight of the day depends on the number of data on that day. It is apparent that the difference between simple and weighted average is large when the number of daily data is small and highly variable. Fig. 4b shows that the median numbers of daily data in each month were 10 and 10.5 at Anmyon and Gosan, which were 33% and 35% of the total number of days (assuming 30 days on average), respectively. We used simple average in calculating monthly and other means because the number of daily data using both direct sun and inversion product AODs was larger than that using inversion product AOD alone.

Table 1 shows the mean values of PM<sub>10</sub> at each site under various conditions. At both Anmyon and Gosan, PM<sub>AOD</sub> was higher than PM<sub>all</sub>, the difference was larger at Gosan. It is

difficult to directly compare the differences at Anmyon and Gosan with those reported in Christopher and Gupta (2010) because of differences in particle size and methods for obtaining mean values. However, it is apparent that the values of PM<sub>AOD</sub> – PM<sub>all</sub> given in Table 1, 5.1 μg/m<sup>3</sup> for Anmyon and 9.9 μg/m<sup>3</sup> for Gosan, are among the highest in comparison with the differences shown for the 10 EPA regions in Christopher and Gupta (2010).

The difference between PM<sub>AOD</sub> and PM<sub>all</sub> was due to the fact that PM<sub>AOD</sub> was only for daytime clear-sky conditions, excluding nighttime (PM<sub>night</sub>), and daytime with precipitation (PM<sub>wet</sub>) and clouds (PM<sub>cloud</sub>). Thus, we traced the changes in the variation of PM<sub>10</sub> concentration by excluding the specific conditions in Table 1. PM<sub>10</sub> concentrations in the daytime (PM<sub>day</sub>) and nighttime were calculated from hourly data in the daytime and nighttime, respectively, similarly to PM<sub>all</sub> with the 75% criterion as described earlier. PM<sub>10</sub> concentrations in the daytime without precipitation (PM<sub>dry</sub>) and under clear-sky conditions with cloud amount 0 (PM<sub>clear</sub>) were calculated similarly to PM<sub>clear</sub>.

**Table 1 – Mean values of PM<sub>10</sub> (μg/m<sup>3</sup>) under selected conditions.<sup>a</sup>**

	Mean (RSD) <sup>b</sup>		Mean (RSD)		Difference between the means <sup>c</sup>
<b>(a) Anmyon</b>					
PM <sub>all</sub>	47.8 (0.84)			PM <sub>AOD</sub> – PM <sub>all</sub>	5.1
PM <sub>day</sub>	47.4 (0.90)	PM <sub>night</sub>	48.1 (0.85)	PM <sub>day</sub> – PM <sub>all</sub>	–0.4
PM <sub>dry</sub>	48.0 (0.90)	PM <sub>wet</sub>	34.3 (1.12)	PM <sub>dry</sub> – PM <sub>day</sub>	0.6
PM <sub>clear</sub>	56.6 (1.01)	PM <sub>cloud</sub>	46.3 (0.93)	PM <sub>clear</sub> – PM <sub>dry</sub>	8.6**
PM <sub>AOD</sub>	52.8 (0.95)			PM <sub>AOD</sub> – PM <sub>clear</sub>	–3.8
<b>(b) Gosan</b>					
PM <sub>all</sub>	38.6 (0.84)			PM <sub>AOD</sub> – PM <sub>all</sub>	9.9**
PM <sub>day</sub>	37.7 (0.87)	PM <sub>night</sub>	39.5 (0.91)	PM <sub>day</sub> – PM <sub>all</sub>	–0.9
PM <sub>dry</sub>	39.0 (0.88)	PM <sub>wet</sub>	29.1 (1.02)	PM <sub>dry</sub> – PM <sub>day</sub>	1.3
PM <sub>clear</sub>	47.2 (0.95)	PM <sub>cloud</sub>	37.1 (0.87)	PM <sub>clear</sub> – PM <sub>dry</sub>	8.2**
PM <sub>AOD</sub>	48.5 (0.98)			PM <sub>AOD</sub> – PM <sub>clear</sub>	1.4

<sup>a</sup> PM<sub>all</sub> represents mean PM<sub>10</sub> from all PM<sub>10</sub> data, and PM<sub>AOD</sub>, mean PM<sub>10</sub> when AOD was available. Others are mean PM<sub>10</sub> in the daytime (PM<sub>day</sub>), nighttime (PM<sub>night</sub>), daytime without precipitation (PM<sub>dry</sub>), daytime with precipitation (PM<sub>wet</sub>), daytime under clear-sky conditions (PM<sub>clear</sub>), daytime with clouds (PM<sub>cloud</sub>).

<sup>b</sup> RSD: Relative standard deviation, defined by the standard deviation divided by the mean.

<sup>c</sup> Statistical significance was tested by the Student’s t-test: \*p < 0.05, \*\*p < 0.01.

Although AOD was measured under clear-sky conditions, the fraction of AODs measured at cloud amount 0 was only 40% and 43% at Anmyon and Gosan, respectively (Fig. 5). The fraction of AODs decreased with cloud amount, but was not negligible even at high cloud amounts above 7–8. This was probably because the cloud amount observed at 3 hr intervals was assumed to last for 3 hr, while the measurement of AOD was counted on an hourly basis. Therefore, if clouds observed at the time of observation were cleared within 3 hr, AOD could have been measured despite the existence of clouds on the record. However, it could also be attributable in part to the difference in cloud perception between AERONET and human observation, which is adopted at the weather stations in Korea, similar to that between human observation and electronic sensing (Duchon and O'Malley, 1999; Wacker et al., 2015).

As shown in Table 1,  $PM_{10}$  concentration approached  $PM_{AOD}$  as the condition was close to that for measuring AOD. Because  $PM_{all}$  was lower than  $PM_{AOD}$ ,  $PM_{10}$  concentration increased generally, except  $PM_{day}$  which was lower than  $PM_{night}$ .  $PM_{night}$  higher than  $PM_{day}$  was caused by lower mixing height and also by lower temperature and higher relative humidity, which are favorable for partitioning of semi-volatile components into the particulate phase (Seinfeld and Pandis, 1998). On the other hand,  $PM_{10}$  concentration increased by excluding lower  $PM_{wet}$  and  $PM_{cloud}$ . Among three categories,  $PM_{day}$ ,  $PM_{dry}$ , and  $PM_{clear}$ , the difference between the means was the highest and statistically significant ( $p < 0.01$ ) by excluding  $PM_{cloud}$ ,  $8.6 \mu\text{g}/\text{m}^3$  at Anmyon and  $8.2 \mu\text{g}/\text{m}^3$  at Gosan. This indicates that clouds in the daytime were a key to explaining the difference between  $PM_{AOD}$  and  $PM_{all}$ .

### 2.3. Additional considerations in the differences between $PM_{AOD}$ and $PM_{all}$

In the previous section, we compared mean  $PM_{10}$  concentrations between Anmyon and Gosan under various conditions. However, these values could be altered depending on the study period and/or exclusion of high concentration days because the sample data were changed. To check the sensitivity of the values shown in Table 1 due to the changes in the sample data, we examined their variations by excluding the days with mean  $PM_{10}$  concentrations exceeding the

highest 1%. Total numbers of days with valid daily means at Anmyon and Gosan were 1172 and 2707, respectively, and thus 11 and 27 days were excluded. Among them, Asian dust was observed on 8 and 22 days, which accounted for 73 and 81% of the days excluded at Anmyon and Gosan, respectively.

As high values were excluded,  $PM_{all}$  and  $PM_{AOD}$  became reduced to  $45.2$  and  $49.1 \mu\text{g}/\text{m}^3$  at Anmyon, and  $36.5$  and  $43.5 \mu\text{g}/\text{m}^3$  at Gosan, respectively. The values of  $PM_{AOD} - PM_{all}$  were also reduced to  $3.9$  and  $7.0 \mu\text{g}/\text{m}^3$  at Anmyon and Gosan, respectively, which still fall in the higher range compared with the differences shown in Christopher and Gupta (2010). The ratios of the values by excluding the highest 1% to the original values shown in Table 1 were between 69% and 101% with mean  $\pm$  standard deviation of  $90\% \pm 9\%$ . One exception was  $PM_{AOD} - PM_{clear}$  at Gosan, which was reduced to  $0.1 \mu\text{g}/\text{m}^3$ , 7% of the original value at  $1.4 \mu\text{g}/\text{m}^3$ . These results demonstrate that the values and their variations in Table 1 were mostly not much affected by the presence of high concentration days.

Although the difference between  $PM_{AOD}$  and  $PM_{clear}$ ,  $PM_{AOD} - PM_{clear}$ , was not large particularly at Gosan, Table 1 shows that  $PM_{AOD}$  was not the same as  $PM_{clear}$ . This was because no AODs were available on occasion despite daytime clear-sky conditions irrespective of measurements of solar radiation (Fig. 2). If AOD was not provided for a number of clear-sky hours in the daytime, it is difficult to fully assess the clear-sky bias of PM by examining the difference between  $PM_{AOD}$  and  $PM_{all}$ . Thus, we decided to compare  $PM_{all}$  with  $PM_{clear}$ . In Table 1, we can see that the  $PM_{clear}$  was also larger than  $PM_{all}$ , but the differences were  $8.8 \mu\text{g}/\text{m}^3$  at Anmyon and  $8.6 \mu\text{g}/\text{m}^3$  at Gosan ( $p < 0.01$  for both values). Interestingly, the differences of  $PM_{clear} - PM_{all}$  at the two sites were similar compared with those of  $PM_{AOD} - PM_{all}$ , as in the case of those of  $PM_{clear} - PM_{dry}$ .

### 2.4. Differences between $PM_{clear}$ and $PM_{all}$

Fig. 6 shows the monthly variations in  $PM_{clear}$  and  $PM_{all}$ . A bimodality of the variations was distinct, which was caused mainly by the lower concentrations in summer. Lower concentrations between June and September were due to scavenging by frequent precipitations and also to less influence of polluted air masses from the west (Ghim et al., 2001, 2015). On the other hand, higher concentrations in

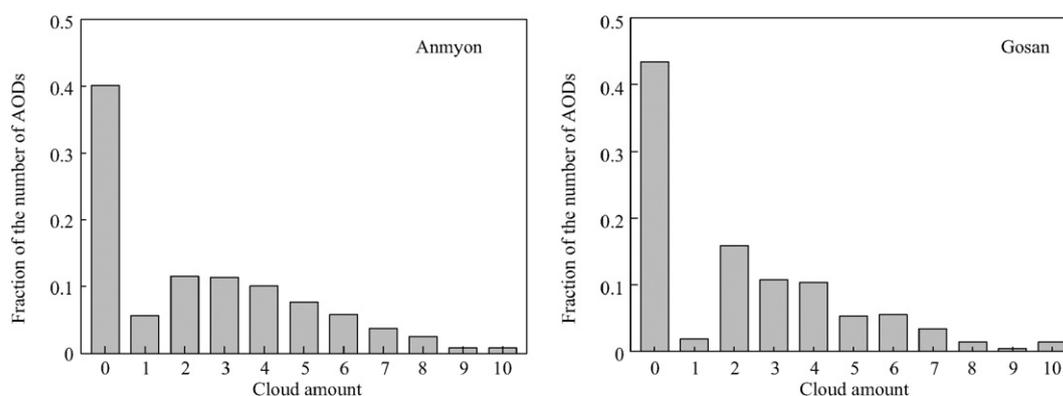
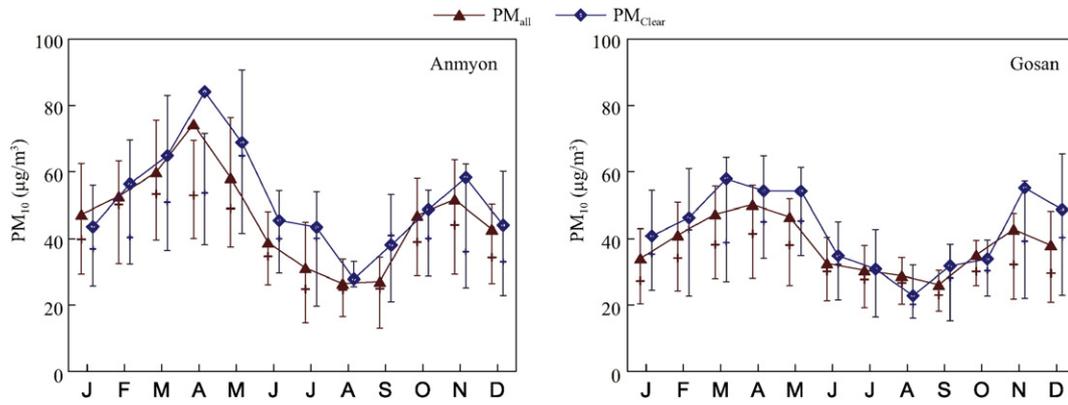


Fig. 5 – The fraction of the number of AODs at each cloud amount.



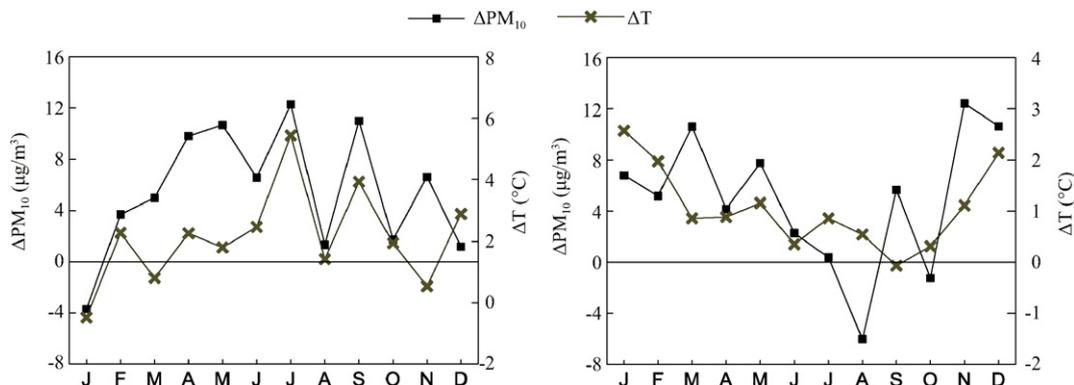
**Fig. 6 – Monthly variations in  $PM_{all}$  and  $PM_{clear}$ .** Solid triangles and open diamonds denote monthly means. Error bars with pluses at the center represent 25th, 50th (median), and 75th percentiles.

spring were due to the effects of fugitive dust including Asian dust (Choi et al., 2014). The means were generally higher than the medians because of a number of much higher concentrations than the median. At Anmyon in April, the means were even higher than the 75th percentiles due to several high concentrations on Asian dust days. It was mentioned that the mean value of  $PM_{clear}$  was higher than that of  $PM_{all}$  during the study period at both Anmyon and Gosan. However, Fig. 6 shows that  $PM_{clear}$  was lower than  $PM_{all}$  sometimes, such as in January at Anmyon and in August and October at Gosan. Christopher and Gupta (2010) also observed a negative value of  $PM_{clear} - PM_{all}$  during colder months.

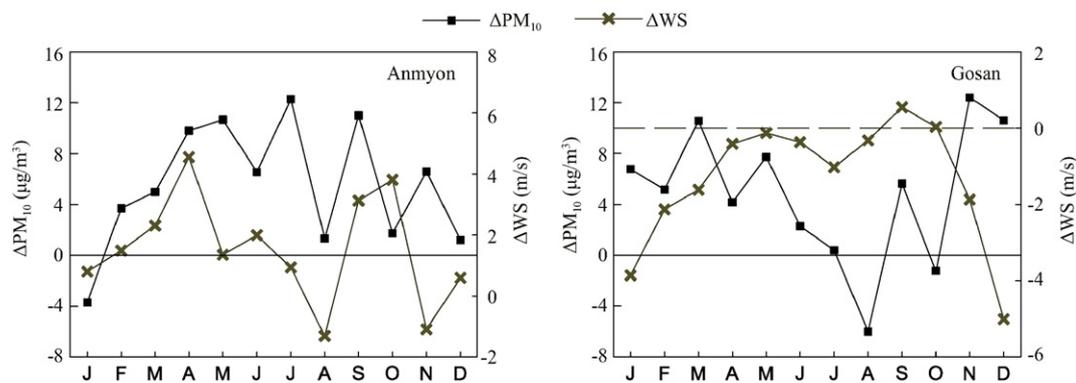
To look into the variations in  $PM_{clear} - PM_{all}$ ,  $\Delta PM_{10}$ , more closely, we present its monthly variations in Fig. 7. Monthly mean  $\Delta PM_{10}$  at both sites was highly variable and peaked at  $12 \mu g/m^3$  (much higher than the mean values estimated from the data shown in Table 1, 8.8 and  $8.6 \mu g/m^3$  at Anmyon and Gosan, respectively). The variations in  $\Delta PM_{10}$  at the two sites were completely different;  $\Delta PM_{10}$  was generally high in summer at Anmyon while the reverse was true at Gosan despite month-to-month variation. These variations were hardly expected because not only variations in  $PM_{10}$  in Fig. 6 were similar but those in cloud

amount in Fig. 3 and other meteorological variables were not much different between the two sites. Fig. 7 also shows the variations in  $\Delta T$ , the difference in air temperature between daytime clear-sky and all conditions ( $T_{clear} - T_{all}$ ). The variations in  $\Delta T$  at Anmyon and Gosan were similarly different to those in  $\Delta PM_{10}$ . As a result, the correlation coefficients between  $\Delta PM_{10}$  and  $\Delta T$  were both positive at Anmyon and Gosan, although the value at Anmyon was higher and statistically significant.

Higher  $\Delta PM_{10}$ , associated with higher  $\Delta T$  at Anmyon during the warm season (April to September) could easily be understood because higher temperatures were favorable for PM production (Fine et al., 2008; Christopher and Gupta, 2010; Ghim et al., 2015). However, there is also a positive correlation between  $\Delta PM_{10}$  and  $\Delta WS$ , particularly between April and September (correlation coefficient of 0.61 in comparison with 0.30 for the entire year) (Fig. 8). If PM increases with both temperature and wind speed, the effects of fugitive dust are likely to be of importance (Choi et al., 2014; Ghim et al., 2015). However, because the range of  $\Delta WS$  is too small to see the effects of fugitive dust (Harrison et al., 2001; Barmpadimos et al., 2012), it is reasonable that the variation in  $\Delta PM_{10}$  at Anmyon during the warm season was mostly associated with



**Fig. 7 – Monthly variations in  $\Delta PM_{10}$ ,  $PM_{clear} - PM_{all}$  and  $\Delta T$ ,  $T_{clear} - T_{all}$ .** Horizontal lines denote zero values. The correlation coefficients between  $\Delta PM_{10}$  and  $\Delta T$  are 0.65 (t-test,  $p < 0.05$ ) for Anmyon and 0.45 for Gosan.



**Fig. 8** – Monthly variations in  $\Delta PM_{10}$ ,  $PM_{clear} - PM_{all}$  and  $\Delta WS$ ,  $WS_{clear} - WS_{all}$ . Horizontal lines denote zero values. The correlation coefficients between  $\Delta PM_{10}$  and  $\Delta WS$  are 0.30 for Anmyon and  $-0.52$  for Gosan.

that in  $\Delta T$ , and presumed to be more affected by photochemical production than the generation of fugitive dust.

A fall in  $\Delta PM_{10}$  in August was common at both sites, and was attributable to lower  $\Delta T$  and  $|\Delta WS|$  (absolute value of  $\Delta WS$  because  $\Delta WS$  was mostly positive at Anmyon and negative at Gosan). This was particularly true at Gosan, where  $\Delta PM_{10}$  was lowered with decreasing  $\Delta T$  and increasing  $\Delta WS$  during the warm season. By the same token, higher  $\Delta PM_{10}$  at Gosan during the cold season was caused by higher  $\Delta T$  and lower  $\Delta WS$ ; that is,  $PM_{10}$  under clear-sky conditions became higher in association with higher temperature and lower wind speed, indicating enhanced photochemical production under favorable meteorological conditions. The effect of photochemical production was pronounced at Gosan, because of the higher temperature than that at Anmyon ( $15.7\text{ }^{\circ}\text{C}$  vs.  $12.1\text{ }^{\circ}\text{C}$  for the entire year,  $8.7\text{ }^{\circ}\text{C}$  vs.  $1.8\text{ }^{\circ}\text{C}$  during the cold season, between November and February), which was manifested accompanying a large decrease in wind speed during the cold season.

For a reason for the negative  $\Delta PM_{10}$  in January at Anmyon, we could estimate an influence of the Siberian continental high-pressure system, which resides quasi-permanently over the Eurasian continent in winter (Jhun and Lee, 2004; Ryoo et al., 2004). Although sky was clear under a high-pressure system, temperature was low as well as  $PM_{10}$  because air masses were rapidly introduced from the north, passing through relatively clean areas (Kim et al., 2005).  $\Delta PM_{10}$  was also negative in August and October at Gosan, but a detailed mechanism is difficult to explain.

### 3. Summary and conclusions

A bias in clear-sky conditions for estimating PM from AOD was examined using  $PM_{10}$  between 2004 and 2007 at Anmyon and between 2003 and 2011 at Gosan, when both  $PM_{10}$  and AOD data were available. Monthly recovery rate of AOD was much lower and highly variable ( $7.3\% \pm 6.2\%$  and  $4.4\% \pm 5.7\%$ , Anmyon and Gosan, respectively) compared with that of  $PM_{10}$  ( $89\% \pm 23\%$  and  $91\% \pm 16\%$ ). Furthermore, there were no AODs for 11% and 44% of total months at Anmyon and Gosan,

respectively, during the study period. The variations in monthly mean recovery rate of AOD were largely proportional to cloud amount at both Anmyon and Gosan.

The difference in  $PM_{10}$  between  $PM_{AOD}$  and  $PM_{all}$  was assessed by examining the variation in  $PM_{10}$ , excluding specific conditions such as nighttime and daytime with precipitation and clouds.  $PM_{AOD}$  was significantly higher than  $PM_{all}$ , by  $5.1\text{ }\mu\text{g}/\text{m}^3$  and  $9.9\text{ }\mu\text{g}/\text{m}^3$ , at Anmyon and Gosan, respectively. These differences were not only absolutely but also relatively large, accounting for 11% and 26% of  $PM_{all}$ . However, there was still a difference between  $PM_{AOD}$  and  $PM_{clear}$ ,  $-3.8\text{ }\mu\text{g}/\text{m}^3$  and  $1.4\text{ }\mu\text{g}/\text{m}^3$  at Anmyon and Gosan, respectively, because no AODs were provided on occasion despite daytime clear-sky conditions.

As a result, we looked into  $\Delta PM_{10}$ , the difference of  $PM_{all}$  from  $PM_{clear}$  (regardless of AOD availability), rather than from  $PM_{AOD}$ . Monthly mean  $\Delta PM_{10}$  increased to more than  $12\text{ }\mu\text{g}/\text{m}^3$  in July at Anmyon and in November at Gosan. Monthly variations in  $\Delta PM_{10}$  at the two sites were completely different;  $\Delta PM_{10}$  was generally higher in summer at Anmyon while it was higher in winter at Gosan.  $\Delta PM_{10}$  at Anmyon even dropped to a negative value in January due to the effect of air masses from the north passing through relatively clean areas under clear-sky conditions with low temperatures, influenced by the Siberian continental high-pressure system. On the other hand,  $\Delta PM_{10}$  at Gosan was high in winter due to an effect of photochemical production at higher temperatures than that at Anmyon, which was pronounced in association with a decrease in wind speed.

Despite using the measurements from two sites, this study demonstrated that a clear-sky bias for estimating PM from AOD was significant. This may be manifested because we could distinguish the effect of clouds, which was a key to explaining the difference between  $PM_{AOD}$  (or  $PM_{clear}$ ) and  $PM_{all}$ , more clearly using hourly data from AERONET than using daily data from satellites. It also showed that seasonal variations in  $\Delta PM_{10}$  at the two sites were different. Although many of the variations at the study sites were explained in terms of those in temperature and wind speed, research efforts based on extensive data from a large number of sites are warranted.

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