## INTRODUCTION

Elemental carbon refers to carbonaceous aerosols in particulate matter (PM). As a type of elemental carbon, black carbon (BC) encompasses carbonaceous aerosols in PM defined by their optical properties and BC is usually produced by the incomplete combustion of fossil fuels. BC is important in climate change research because it can alter radiative forcing via light absorption (McMurry *et al.*, 2004), making BC one of the most notorious substances among air pollutants for its influence on global warming. Although global warming is considered to be mainly caused by greenhouse gases, research has suggested the possibility that warming of the Earth's atmosphere may be caused by BC (Andreae, 2001). Unlike greenhouse gases, BC is composed of solid particles that heat the atmosphere via direct absorption of solar radiation. Because it is only present in the atmosphere for short durations, BC is referred to as a short-lived climate forcer, and has been reported to influence local climate change due to the high variations in BC concentrations among industrialized cities and remote suburbs (Chameides and Bergin, 2002).

Several preliminary studies have assessed the factors affecting BC concentrations. Before industrialization, large-scale forest fires or volcanic activity were the main causes of increased BC concentrations. However, after the industrial revolution, particulate emissions from direct combustion of hydrocarbon fuels (e.g., coal and petroleum) increased drastically (McConnell *et al.*, 2007). In particular, PM is generated not only via anthropogenic processes (e.g., fuel

combustion, vehicle emissions, and chemical production processes), but also by condensation of $SO_2$ and volatile organic compounds via secondary processes (Volkamer *et al.*, 2006). As such, studies on the secondary generation of aerosols have been actively conducted in recent years. Overall, a great attention has been paid not only to primary (i.e., direct) production, but also to the secondary (i.e., indirect) production of atmospheric PM.

As typical products of primary atmospheric aerosols, we measured and monitored BC concentrations for eight months from September 2015 to April 2016 in Chungcheong Province, Korea, to clarify regional BC emissions. Chungcheong Province is located in central South Korea far from the Seoul Metropolitan Area; therefore, it is recognized as being less affected by pollutants from Seoul Metropolitan Area and other urban regions. Further, we assessed the variations in BC concentrations in atmospheric aerosols in relation to $PM_{10}$ and $PM_{2.5}$ concentrations, as well as wind direction and wind velocity. Using these data, we identified the main meteorological factors influencing the behavior of $PM_{10}$, $PM_{2.5}$, and BC.

## EXPERIMENTAL METHODS

We used a multi-angle absorption photometer (MAAP) for the measurement of BC. MAAPs employ a filter-based technique to measure the light absorption of BC, where atmospheric aerosols are deposited onto a filter substrate and a laser beam with a wavelength of 637 nm is shot toward the deposited filter. Two detectors located on the same side as the laser source

71 measure the light back-scattered by BC deposited on the filter. In addition, a detector located on
72 the opposite side of the laser source measures the light transmitted through the filter.
73 Conventional BC measurement instruments only have a detector on the opposite side of the laser
74 source and do not measure the scattering of light, which means that only transmitted light can be
75 detected. As a result of the detection of signal from back-scattering in MAAPs, the scattering
76 effect can be compensated for by correcting the signal from transmitted light with the signals of
77 the scattered light (Petzold and Schönlinner, 2004). The BC concentration derived from filter-
78 based techniques (e.g., aethalometers and MAAPs) is referred as equivalent BC (eBC). As such,
79 "BC concentration" hereafter represents the equivalent BC concentration.

80 In the present study, the BC concentration was measured in 1-min increments at the Korea
81 University of Technology and Education (KOREATECH) located in Byeongcheon-myeon,
82 Cheonan city, Chungcheongnam-do (Fig. 1), using a commercially available instrument (MAAP
83 5012; Thermo Scientific). BC concentrations were monitored from September 2015 to April 2016.
84 The $PM_{10}$ and $PM_{2.5}$ concentrations were obtained from the Air Korea database
85 (http://www.airkorea.or.kr) operated by the Korea Environment Corporation. According to Air
86 Korea, $PM_{10}$ and $PM_{2.5}$ concentrations are measured with automatic unmanned equipment using
87 the β-ray absorption method, where PM is collected on a filter for 1 h and β-rays pass through the
88 PM on the filter. Then, changes in absorption or extinction before and after the deposition of PM

89 for 1 h are measured and converted into mass concentration. The principle of the β-ray absorption
90 method is similar to the quantification of BC by filter-based instruments (e.g., aethalometers). In
91 this study, we used PM data from Ochang-eup, Cheongju city, Chungcheongbuk-do (Fig. 1),
92 from September 2015 to April 2016.

93 The seasonal mean BC, $PM_{10}$, and $PM_{2.5}$ concentrations were calculated from raw data for fall
94 2015 (September–November 2015), winter 2015 (December–February 2016), and spring 2016
95 (March–April 2016).

96 Wind direction and wind speed displayed once hourly were collected from the Korea
97 Meteorology Administration (KMA) database (http://www.kma.go.kr). Wind direction and wind
98 speed were measured at Shibang-dong, Cheonan, Chungcheongnam-do, which is located 14 km
99 from KOREATECH (Fig. 1). Windrose diagrams of wind direction and wind speed obtained
100 from September 2015 to April 2016 are shown in Fig. 2. It should be noted that these windrose
101 diagrams show only wind intensity and direction, not BC concentrations. In fall 2015, south-
102 easterly winds were equally dominant as westerly winds. However, in winter 2015, westerly
103 winds were dominant. In spring 2016, westerly winds were more dominant than easterly winds.
104 Overall, westerly winds were dominant during the measurement period from fall 2015 to spring
105 2016.
106

## RESULTS AND DISCUSSION

### *Concentrations of BC, $PM_{10}$, and $PM_{2.5}$*

Table 1 presents the monthly average BC, $PM_{10}$ and $PM_{2.5}$ concentrations from September 2015 to April 2016. In fall 2015, winter 2015, and spring 2016, the average BC concentrations were 1.39 μg $m^{-3}$, 1.57 μg $m^{-3}$, and 2.30 μg $m^{-3}$, the average $PM_{10}$ concentrations were 36.7 μg $m^{-3}$, 48.8 μg $m^{-3}$, and 66.4 μg $m^{-3}$, and the average $PM_{2.5}$ concentrations were 30.9 μg $m^{-3}$, 37.7 μg $m^{-3}$ and 41.9 μg $m^{-3}$, respectively. Upon initial inspection, the trends in BC concentrations appeared to be similar to those of $PM_{10}$ and $PM_{2.5}$ concentrations. For instance, the highest and lowest average concentrations of BC, $PM_{10}$, and $PM_{2.5}$ were observed in spring and fall, respectively. As shown in Fig. 2, westerly winds were dominant in spring. Thus, the high concentrations in spring were likely related to the influence of Asian Dust transported by westerly winds. In addition, the higher concentrations of BC, $PM_{10}$, and $PM_{2.5}$ in winter and spring were possibly related to the transport of primary air pollutants produced via combustion processes from surrounding areas via westerly winds.

Meanwhile, BC, $PM_{10}$, and $PM_{2.5}$ concentrations in fall were slightly lower than those in winter. The wind direction pattern in fall 2015 differed from that in winter 2015 (Fig. 2), with greater wind direction variations in fall 2015. These conditions could support turbulent mixing, reducing overall air pollutant concentrations. By contrast, the relatively consistent wind direction in winter

126 2015 could support the formation of atmospheric laminar flow mainly from the west, enabling the
127 formation of a stable and stagnant air mass over the Korean Peninsula. In addition, the low BC,
128 $PM_{10}$, and $PM_{2.5}$ concentrations in fall may have been caused by decreased production of fine PM,
129 although further studies are necessary to confirm these trends and mechanisms.

130 Figure 3 shows the 24-h running average of the data collected during the eight-month
131 measurement period. The discontinuity of BC concentrations in Fig. 3 reflects the fact that the
132 measurement instrument was temporarily stopped for maintenance. Although the seasonal BC
133 concentrations initially appeared to follow the same trends as $PM_{10}$ and $PM_{2.5}$ concentrations, a
134 closer examination of the data in Fig. 3 revealed that BC concentrations did not follow the same
135 trends as $PM_{10}$ and $PM_{2.5}$ concentrations. This can be explained by the fact that PM consists not
136 only of BC, but also other substances (e.g., sulfates, nitrates, minerals, etc.). For example, in
137 December 2015, BC concentrations decreased, whereas $PM_{10}$ and $PM_{2.5}$ concentrations increased
138 (Fig. 3). Meanwhile, from January 2016 to mid-February 2016, BC concentrations increased
139 markedly, while $PM_{10}$ and $PM_{2.5}$ concentrations decreased slightly. Overall, the relationship
140 between BC concentrations and PM concentrations may vary spatially or temporally depending
141 on their sources or transport patterns.

142 Figure 4 shows a bar chart of the mass concentration composition of BC, $PM_{10}$, and $PM_{2.5}$.
143 Notably, the proportion of $PM_{2.5}$ was high throughout the study period, indicating that coarser

144 particles (i.e., $PM_{10}$) were relatively rare. We speculated that newly formed $PM_{2.5}$ through
145 secondary process were dominant in this area, although it was beyond the scope of this study
146 determine the mechanism driving the high proportions of $PM_{2.5}$.

147

148 ***Linear regression analysis of BC, $PM_{10}$, and $PM_{2.5}$ concentrations***

149 We analyzed the correlations among BC, $PM_{10}$, and $PM_{2.5}$ with linear regression, where the
150 coefficient of determination ($R^2$) was obtained from the linear regression of BC versus $PM_{2.5}$ or
151 $PM_{10}$ concentrations and $PM_{2.5}$ versus $PM_{10}$ concentrations.

152 Table 2 shows the $R^2$ values of the linear regressions among BC, $PM_{10}$, and $PM_{2.5}$
153 concentrations. Except for the $R^2$ between $PM_{10}$ and $PM_{2.5}$ in winter 2015, the $R^2$ values were
154 generally low, indicating that BC was not strongly correlated with $PM_{10}$ and $PM_{2.5}$. Although the
155 results in Fig. 3 suggested that the BC concentrations appeared to generally follow $PM_{10}$ and
156 $PM_{2.5}$ concentrations on a seasonal scale, the seasonal average BC concentrations were less
157 correlated with $PM_{10}$ and $PM_{2.5}$ concentrations based on linear regression.

158

159 ***Correlation between BC and $PM_{10}$***

160 We performed a linear regression analysis of BC and $PM_{10}$ concentrations during the three
161 seasons in the study period. Although BC concentrations appeared to be directly proportional to
162 $PM_{10}$ concentrations, the calculated $R^2$ was low (Fig. 5). $PM_{10}$ mostly consists of soil-derived

163 dust particles, which are generated in large amounts from various types of emission sources and
164 has a short atmospheric residence time due to their larger size, thereby contributing less to
165 increases in BC concentrations. By contrast, BC consists of carbonaceous particles smaller than 1
166 μm generated from incomplete combustion of hydrocarbon fuel. In particular, in the study area,
167 there are some unpaved roads in small cities with relatively few mobile pollution sources.
168 Therefore, the low correlation between BC and $PM_{10}$ could be explained by the differences in
169 $PM_{10}$ and BC sources. The $R^2$ between BC and $PM_{10}$ was lower in winter 2015 than in the other
170 seasons (Table 2), indicating that fine dust in winter was weakly related to BC. In other words,
171 the regression analysis indicated that BC and $PM_{10}$ had different compositions and emission
172 sources.
173
174 ***Correlation between BC and $PM_{2.5}$***
175 Next, we performed a linear regression analysis of BC and $PM_{2.5}$ concentrations during the
176 three seasons in the study period. The correlations between BC with $PM_{2.5}$ concentrations were
177 similar in fall 2015 ($R^2$ = 0.47), winter 2015 ($R^2$ = 0.45), and spring 2016 ($R^2$ = 0.58) (Fig. 5).
178 The $R^2$ values were higher than those between BC and $PM_{10}$, but still relatively low, suggesting
179 that the $PM_{2.5}$ did not contain a large proportion of BC, and that BC concentrations were not
180 highly correlated with $PM_{2.5}$ concentrations in the study area. Mobile pollution (e.g., diesel

vehicle exhaust) is the main source of BC, whereas $PM_{2.5}$ typically originates from secondary formation from atmospheric industrial plant emissions. Therefore, the contribution of BC to $PM_{2.5}$ concentrations was likely low given that the measurements were performed in a small city, where mobile sources did not appear to influence the concentrations.

***Correlation between $PM_{10}$ and $PM_{2.5}$***

Finally, we performed a linear regression analysis of $PM_{10}$ and $PM_{2.5}$ concentrations during the three seasons in the study period. $PM_{10}$ and $PM_{2.5}$ concentrations were positively correlated in fall 2015 ($R^2 = 0.53$), winter 2015 ($R^2 = 0.90$), and spring 2016 ($R^2 = 0.61$) (Fig. 5). The particularly high correlation in winter 2015 may have been driven by the transport of primary pollutants generated from increased fossil fuel use for heating in continental Asia to the Korean Peninsula via westerly winds (Choi, 2008). In contrast, the lowest correlation was observed in fall 2015. Fossil fuel use for heating purposes was likely lower in fall than in winter. In addition, south-easterly and easterly winds were equally dominant as westerly winds in fall 2015, whereas westerly winds were dominant in winter 2015 (Fig. 2). Therefore, $PM_{2.5}$ was retardly entrained to the Korean Peninsula, resulted in the relatively low correlation between $PM_{2.5}$ and $PM_{10}$ in fall.

***Analysis on event days***

Periods with particularly high $PM_{10}$, $PM_{2.5}$, and BC concentrations can be observed in Fig. 3. We attempted to examine the cause of such events by referring to monthly reports generated by the KMA. In particular, $PM_{10}$, $PM_{2.5}$, and BC concentrations increased from mid-October to early November in 2015 (Fig. 6). Based on the monthly KMA reports, Asian Dust may have been one of the reasons for these increases. From the KMA report, Asian Dust originating from Inner Mongolia on October 26$^{th}$ passed over the Yellow Sea, and was observed over the Korean Peninsula on October 26$^{th}$. Asian Dust events are rare in October, although other fall events have been observed in 2009 and 2014, suggesting that Asian Dust transport may not occur exclusively in spring, but also occasionally in fall. As mentioned previously, the discontinuity in the BC measurements in Fig. 6 was due to regular maintenance of the measurement instrument.

High $PM_{10}$ and $PM_{2.5}$ concentrations were observed in March 2016 (Fig. 6). From the monthly KMA report, an Asian Dust even occurred at the end of March, where Asian Dust originating from Mongolia, the Inner Mongolian Plateau, and northern China was transported over the Korean Peninsula via north-westerly winds, increasing PM concentrations.

## CONCLUSIONS

We analyzed the concentrations of BC, $PM_{10}$, and $PM_{2.5}$ from September 2015 to April 2016 in central Korea, and compared them with various meteorological parameters. The highest and lowest $PM_{10}$, $PM_{2.5}$, and BC concentrations were observed in spring and fall, respectively. $PM_{10}$

219 concentrations were high in spring due to Asian Dust transported via westerly winds. In winter,
220 incomplete combustion of fossil fuels used for heating in continental Asia increased, resulting in
221 the transport of emissions over the Korean Peninsula via westerly winds, increasing PM
222 concentrations. Finally, the low PM concentrations in fall were likely due to increased wind
223 variations driving turbulent mixing.

224 The BC concentrations generally showed low correlations with $PM_{10}$ and $PM_{2.5}$ concentrations
225 in the three studied seasons. Sometimes, BC exhibited direct proportional relationships with both
226 $PM_{10}$ and $PM_{2.5}$ concentrations, albeit with low $R^2$ values. The low correlations were possibly due
227 to differences in the sources of BC (i.e., vehicle), $PM_{2.5}$ (i.e., secondary pollution from industrial
228 plant emissions), and $PM_{10}$ (i.e., soil-derived dust). In particular, the low contribution of BC to
229 $PM_{2.5}$ may have been due to the fact that the study site was located in a small city with little
230 influence from mobile pollution sources.

231 $PM_{10}$ and $PM_{2.5}$ concentrations showed a direct proportional relationship, with $R^2$ values as
232 high as 0.90 in winter 2015. This was likely due to the use of fossil fuels for heating in
233 continental Asia, as well as high concentrations of pollutants from industrial plants, where the
234 primary pollutants generated via incomplete combustion of heating fuel were introduced over the
235 Korean Peninsula via westerly winds from the west coast of Korea. By contrast, the relatively

low correlation ($R^2$ = 0.53) in fall was likely caused by south-easterly winds, blocking the influx of Asian Dust over the Korean Peninsula by westerly winds.

BC concentrations were high from later October to early November in 2015. According to the monthly KMA report, an Asian Dust event originating from Inner Mongolia occurred during this period. In addition, in March 2016, Asian Dust originating from Mongolia was transported over the Korean Peninsula. As such, it is necessary to further study the correlation between BC and Asian Dust in greater detail to clarify the contribution of BC to PM concentrations. Given these results, the belief held by some researchers that BC is only correlated with $PM_{2.5}$ and $PM_{10}$ may not be accurate and warrants further investigation. Overall, the results of this study help clarify the characteristics in BC concentrations over the Korean Peninsula. We believe that the BC concentrations shown in this study can be used as urban background values in the Korean Peninsula.

## ACKNOWLEDGMENTS

This research was supported by Basic Science Research Program through the National Research Foundation of Korea (NRF) funded by the Ministry of Education (2016R1D1A1B03931654). Some part of research was also supported by Korea Ministry of Environment as a Converging Technology Project (2013001650004).

255

256 **REFERENCES**

257

258 Andreae, M.O. (2001). The dark side of aerosols. *Nature* 409: 671-672.

259 Chameides, W.L. and Bergin, M. (2002). Soot takes center stage. *Science* 297: 2214-2215.

260 Choi, H. (2008). Comparison of $PM_1$, $PM_{2.5}$ and $PM_{10}$ concentrations in a mountainous coastal
261 city, Gangneung before and after the Yellow Dust event in spring (written in Korean with
262 English abstract). *J. Environ. Sci. Int.* 17: 633-645.

263 Kim, Y.S. (2012). Black carbon monitoring for managing hazardous air pollutants in the
264 metropolitan area (written in Korean with English abstract). *Seoul Studies* 13: 175-186.

265 McMurry, P., Shepherd, M. and Vickery J. (2004). *Particulate matter assessment for policy*
266 *maker: A NARSTO assessment, 1st Ed*, Cambridge University Press, Cambridge.

267 McConnell, J.R., Edwards, R., Kok, G.L., Flanner, M.G., Zender, C.S., Saltzman, E.S., Banta,
268 J.R., Pasteris, D.R., Carter, M.M. and Kahl, J.D.W. (2007). 20th-century industrial black
269 carbon emissions altered arctic climate forcing. *Science* 317: 1381-1384.

270 Petzold, A. and Schönlinner, M. (2004). Multi-angle absorption photometry—A new method for
271 the measurement of aerosol light absorption and atmospheric black carbon, *J. Aerosol Sci.* 35:
272 421-441.

273 Volkamer, R., Jimenez, J.L., San Martini, F., Dzepina, K., Zhang, Q., Salcedo, D., Molina, L.T.,
274 Worsnop, D.R. and Molina, M.J. (2006). Secondary organic aerosol formation from
275 anthropogenic air pollution: Rapid and higher than expected. *Geophys. Res. Lett.* 33: L17811,
276 doi:10.1029/2006GL026899.
277

## Table Captions

**Table 1.** Monthly average for BC, $PM_{10}$ and $PM_{2.5}$ concentrations (unit: μg $m^{-3}$).

**Table 2.** Correlation coefficients ($R^2$) between $PM_{10}$ and BC, $PM_{2.5}$ and BC, $PM_{10}$ and $PM_{2.5}$ concentrations.

284 **Table 1.** Monthly average for BC, $PM_{10}$ and $PM_{2.5}$ concentrations (unit: μg $m^{-3}$).

| Month | Sep. | Oct. | Nov. | Dec. | Jan. | Feb. | Mar. | Apr. |
|---|---|---|---|---|---|---|---|---|
| $PM_{10}$ | 31.6 | 43.6 | 34.9 | 52.9 | 48.6 | 44.8 | 63.8 | 69.0 |
| $PM_{2.5}$ | 21.2 | 37.0 | 34.6 | 40.5 | 38.9 | 33.7 | 44.1 | 39.6 |
| eBC in $PM_{2.5}$ | 0.79 | 1.71 | 1.67 | 1.41 | 1.21 | 2.08 | 2.15 | 2.44 |

285

286 **Table 2.** Correlation coefficients ($R^2$) between $PM_{10}$ and BC, $PM_{2.5}$ and BC, $PM_{10}$ and $PM_{2.5}$
287 concentrations.

| | 2015 Fall | 2015 Winter | 2016 Spring |
|---|---|---|---|
| eBC-$PM_{10}$ | 0.52 | 0.31 | 0.40 |
| eBC-$PM_{2.5}$ | 0.47 | 0.45 | 0.58 |
| $PM_{10}$-$PM_{2.5}$ | 0.53 | 0.90 | 0.61 |

288

289

## Figure Captions

**Fig. 1.** Location of monitoring site.

**Fig. 2.** Windrose diagrams for 3 seasons.

**Fig. 3.** BC, $PM_{10}$ and $PM_{2.5}$ concentrations monitored for 8 months.

**Fig. 4.** Monthly composition of mass concentration for eBC in $PM_{2.5}$, $PM_{10}$ and $PM_{2.5}$

**Fig. 5.** Correlation graphs for (a) $PM_{10}$ concentration vs. BC concentration, (b) $PM_{2.5}$ concentration vs. BC concentration, (c) $PM_{10}$ concentration vs. $PM_{2.5}$ concentration.

**Fig. 6.** BC, $PM_{2.5}$ and $PM_{10}$ concentrations on two event days.

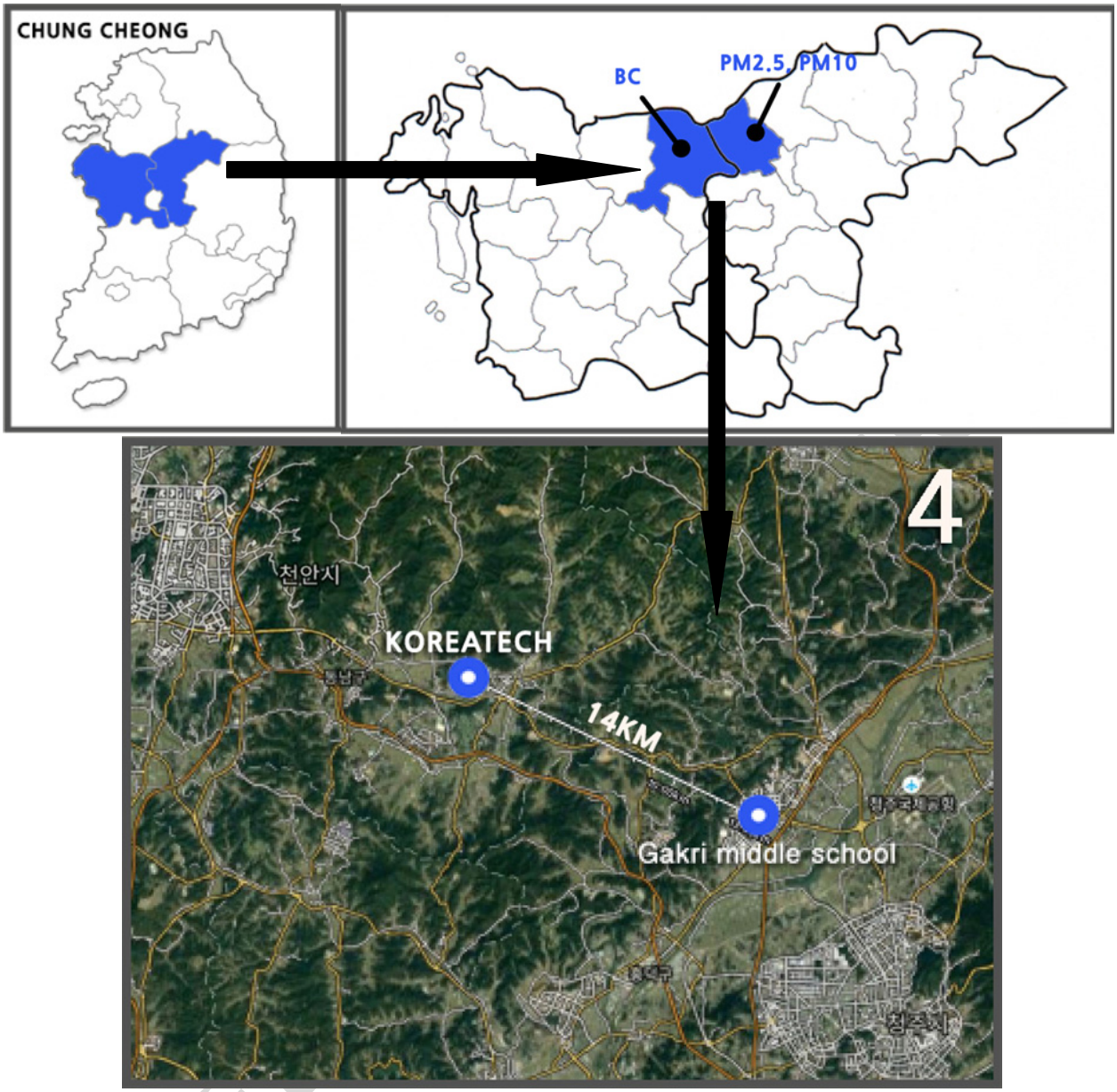
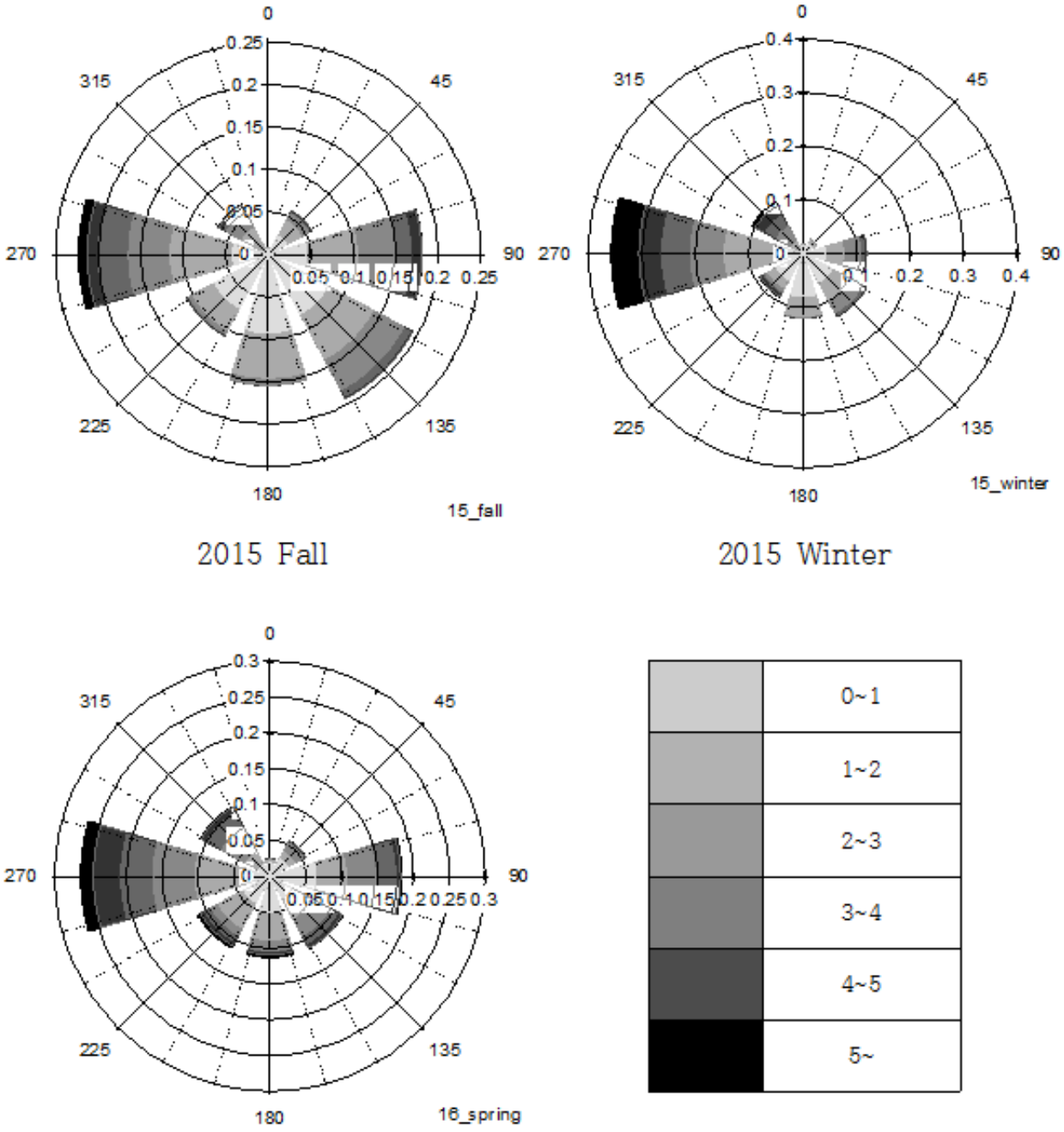



**Fig. 1.** Location of monitoring site.

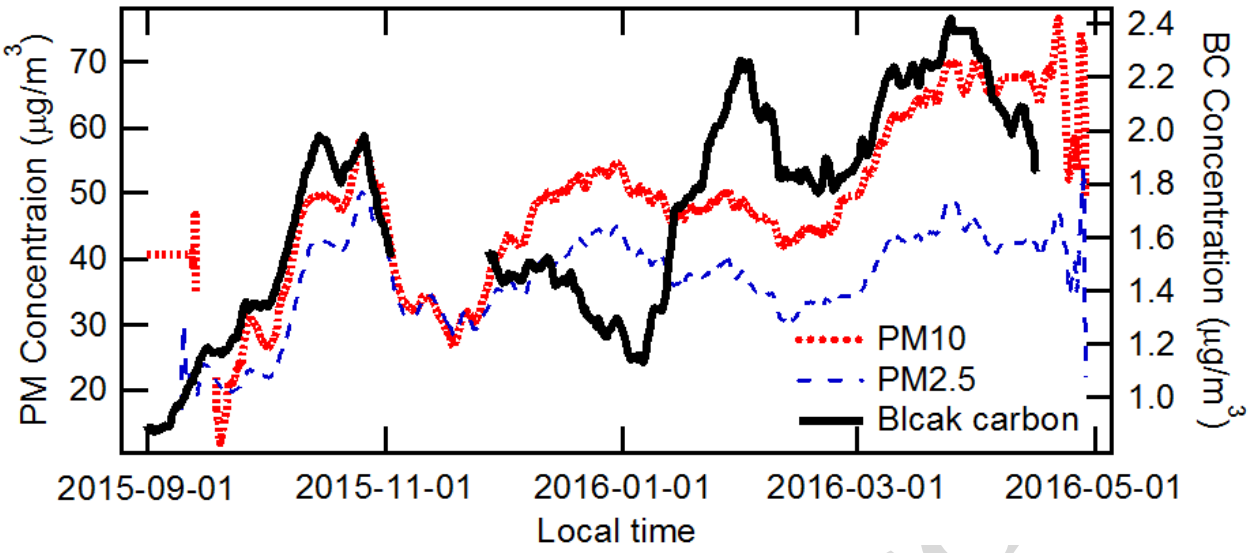



311

312

313 **Fig. 2.** Windrose diagrams for 3 seasons.

314




315

316

317

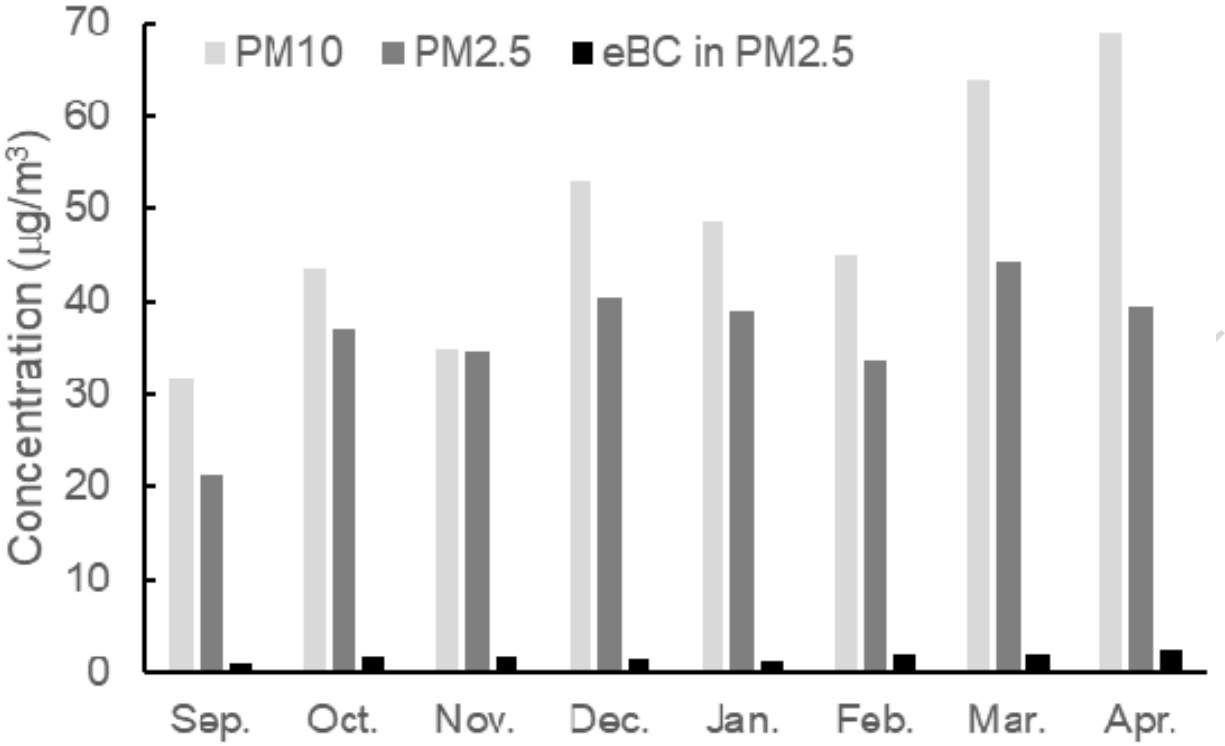
318

319

320

321 **Fig. 3.** BC, $PM_{10}$ and $PM_{2.5}$ concentrations monitored for 8 months.

322




323

324

325

326

327

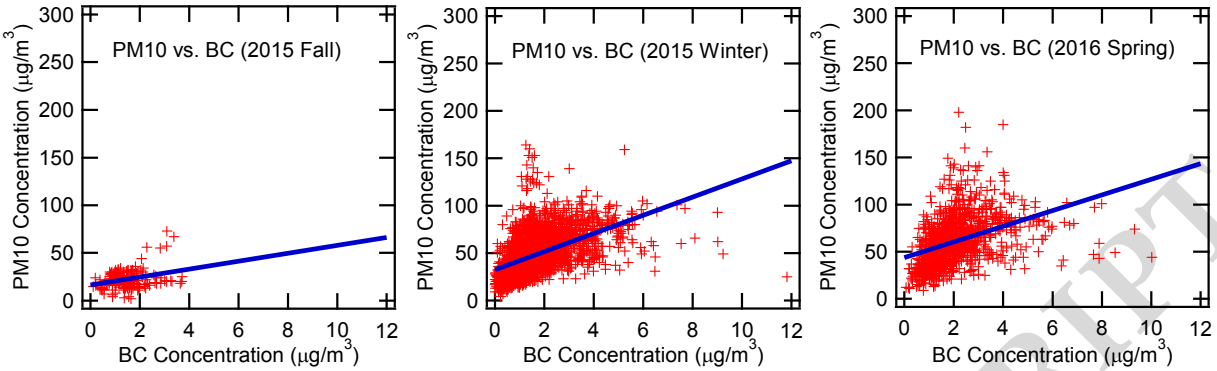
328

329

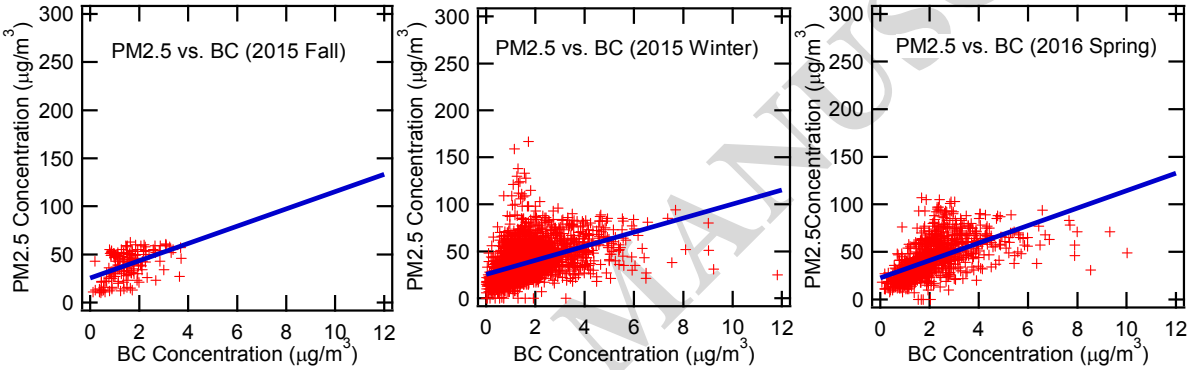
330 **Fig. 4.** Monthly composition of mass concentration for eBC in $PM_{2.5}$, $PM_{10}$ and $PM_{2.5}$

331

(a)




(b)




(c)

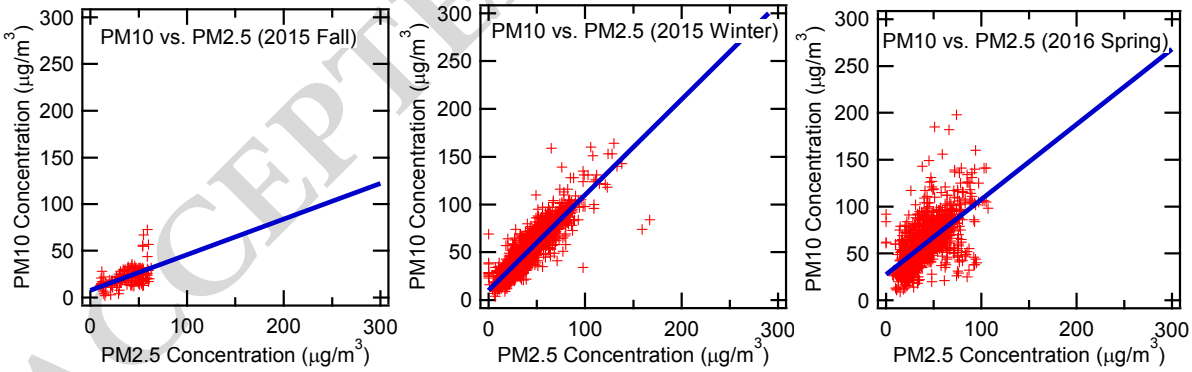
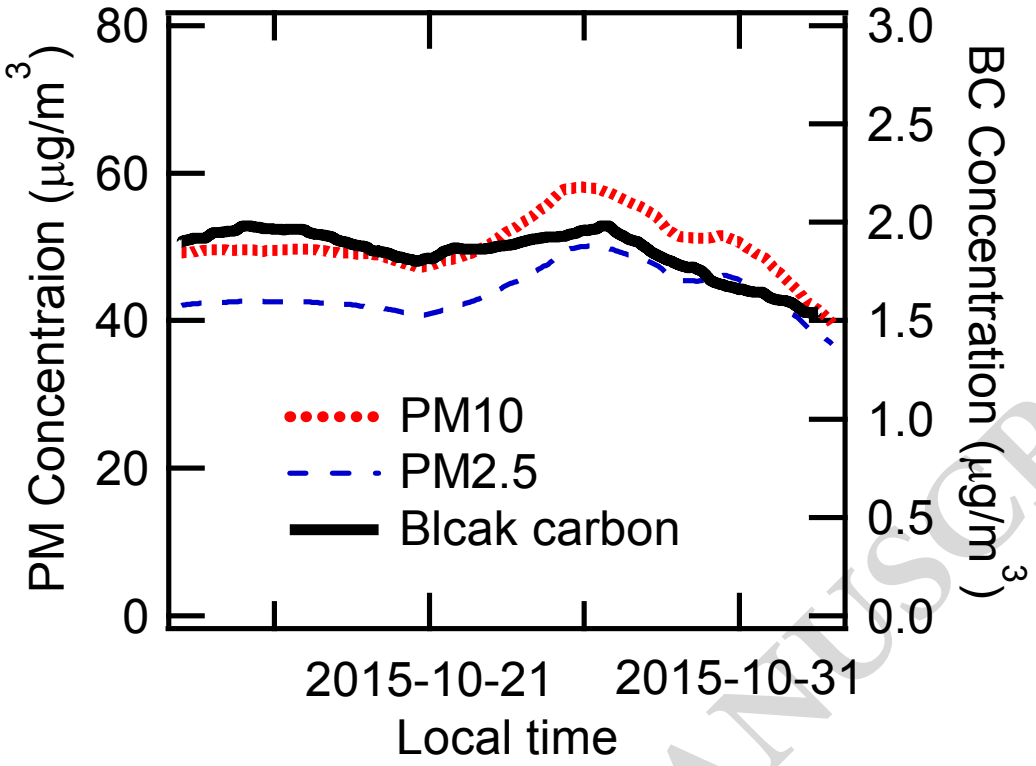
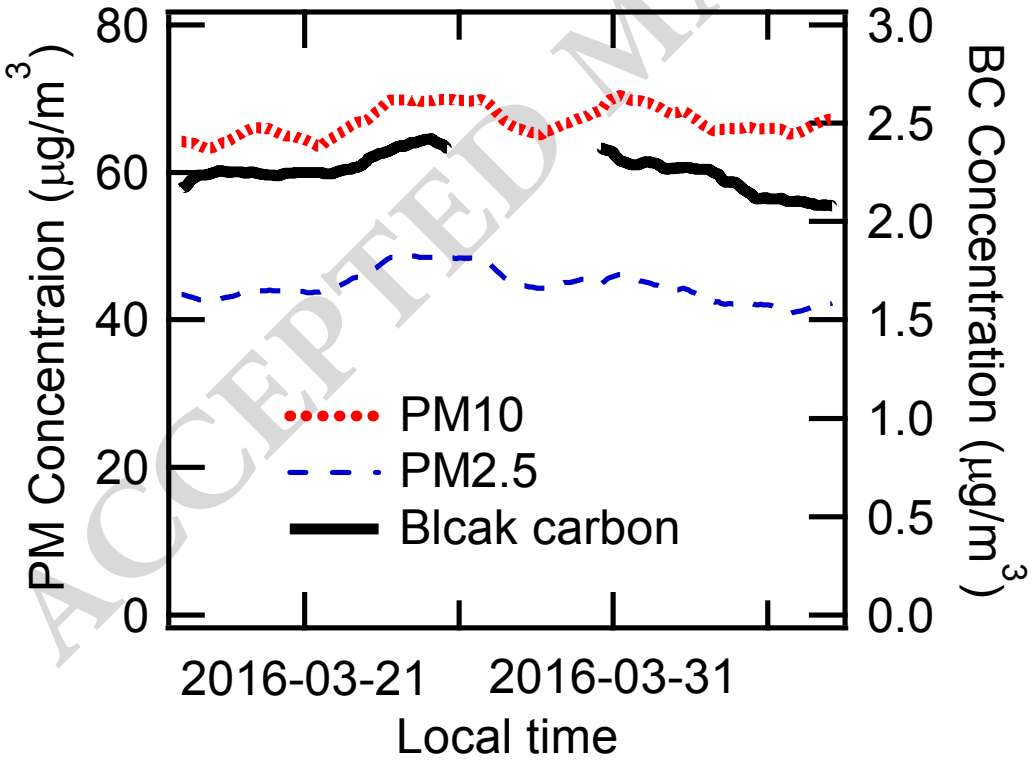





**Fig. 5.** Correlation graphs for (a) $PM_{10}$ concentration vs. BC concentration, (b) $PM_{2.5}$ concentration vs. BC concentration, (c) $PM_{10}$ concentration vs. $PM_{2.5}$ concentration.




342




343

344 **Fig. 6.** BC, $PM_{2.5}$ and $PM_{10}$ concentrations on two event days.