

DIURNAL VARIATIONS OF PARTICLE SIZE DISTRIBUTION AND MASS CONCENTRATION IN THE KOREAN COAST

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ABSTRACT

To investigate mass concentrations of PM-10, PM-2.5 and PM-1 and the distribution of particle size from 300 nanometer to 20 micrometer diameters, two aerosol samplers were equipped at Kangnung Meteorological Administration, Kangnung city in the coast from February 14 through 16, 2005. The concentrations of PM10, PM2.5 and PM1 showed a typical pattern such as high concentration near 0900 LST (beginning time of office hour) and 1700 LST (ending time of office hour) and their low concentration near noon. PM10, PM2.5 and PM1 had the first maximum concentrations of 145.12 $\mu\text{g}/\text{m}^3$ of PM10 and 88.93 $\mu\text{g}/\text{m}^3$ of PM2.5 and 44.71 $\mu\text{g}/\text{m}^3$ of PM1 at 2000 LST (three hours later after sunset), respectively.

The secondary maximum concentration is detected with a magnitude of 93.06 $\mu\text{g}/\text{m}^3$ at 0100 LST, February 15. The distribution of CO and NO_x concentrations showed a similar tendency of PM10, PM2.5 and PM1 concentrations, except for concentrations in the morning on February 14. When PM10, PM2.5 and PM1 had the first maximum concentrations at 2000 LST and their second maximum at 0100 LST, February 15, NO_x had the first and secondary maximum concentrations at the same times. It implies that the increases of emitted gases like NO_x and CO from vehicles on the street and combustion gases from boilers in the resident area for nighttime heating could make a great contribution to the increase of PM concentration. After sunset, much shrunken surface inversion layer than daytime convective atmospheric boundary layer could also increase the concentrations of particle matters, but after midnight, the concentration gradually decreased due to the reduction of vehicle number on the street. Under westerly wind, the particles transported from the city into the mountain side in the west for daytime returned again to the city and some amount of particles from an upwind side city toward the coastal city could also make a great contribution to the occurrence of secondary maximum. The number densities of particle size distributions, regardless of particle diameters were much lower in the early morning than both afternoon and night. Particle size distribution at 1800 LST to 2350 had bigger number densities in the diameters of 1.2 μm to 20 μm .

Key Words : Particle Size Distribution, Nanometer Particulate, PM10, PM2.5, PM1

1. INTRODUCTION

In recent years, many research papers relating to particle size distribution have been published as fundamental aspect of aerosol science (Xu et al., 2004, Kim et al, 2003). Gao and Anderson (2001) investigated characteristics of Chinese aerosols determined by individual particle analysis, where most of measurement of aerosol size is based upon laser sizing on the particles. Adby and Demster (1974) developed powell algorithm as a dependent method and Hansen (1992) and Hansen and

O'Leary (1993) utilized L-curve as an independent one. Goldberg (1989), Michelewicz (1996) and Xu et al. (2004) applied genetic algorithm to treat the optimization problem in a quite different way in the particle size distribution from data of multispectral extinction measurements.

During the Asian Dust period in 2001, the factors of TSP, PM₁₀ and PM_{2.5} affecting the cycle of aerosols and their chemical properties and compositions was investigated in the Seoul district, Korea (Kim and Kim; 2003) and comprehensive researches on particulates and gases at many measurement points in China, Korea and Japan from 2001 to nowadays have been carried out (Carmichael et al, 1997; Xuan and Soklik, 2002). In Korea, persistent measurement of aerosols have been established Gosan, Jechu island and Taean peninsula in the west of Korea by Korean Meteorological Administration, but their measurement have been focussed on mass concentration of total suspended particulate and PM₁₀ and their chemical component and heavy metals, except for PM₁. In addition, their researches do not include partial size distribution of aerosol and the relation between mass concentration and partial size distribution.

Thus, the objective of this study is to explain diurnal variation of aerosol concentration such as PM₁₀, PM_{2.5} and PM₁, which greatly influences upon local pollution state and particle size distribution on human health condition, considering especially time band of their high concentrations.

2. MEASUREMENT OF AEROSOL AND TOPOGRAPHY

2.1. Instrument and experiment

Two aerosol samplers called GRIMM aerosol samplers – Model 1107 and Model 1108 were fixed at Kangnung Meteorological Administration (KMA; 37°45'N, 128°54'E) in Kangnung coastal city, which is in the eastern mountainous coastal region of Korean peninsula. Particle size distribution of aerosol from 300 nano to 20um and collection of all three PM fractions of PM₁₀, PM_{2.5} and PM₁ were investigated by GRIMM aerosol samplers from February 13 through 17, 2005, which included dust storm and non-dust storm periods of China and Korea in spring.

The Model 1107 is specifically designed for PM-10, PM-2.5 and PM-1 environmental ambient air analysis using the laser light scattering technology. This technology enables the Model 1107 to make very precise “cut points” for all three PM size classifications and this system allows the user to collect all three PM fractions simultaneously without changing sampling heads or weighing filters. However, the Model 1107 is the only PM monitor to offer dual technology consisting of both optical and gravimetric analysis. The Model incorporates a removable 47 mm PTFE filter, which allows the user to verify the optical analysis gravimetrically, as well as providing the option for other chemical analysis on the collected residue.

The 1107 model is devised for particulate measurements via 900 laser light scattering and air with multiply particle sizes passes through a flat laser beam produced by an precisely focused laser and several collimator lenses. The scattered light are detected by a 15-channel pulse height analyzer for size classification. The counts from each size classification are converted to mass by a well established equation. The data are presented in the P.S. EPA conventions for PM-10, PM-2.5 and PM-1. Its complete

system consists of 165 fiberglass housing, drying temperature control system, 1107 PM dust monitor, sensors for humidity and temperature and 170M sampling system. Displays real time data of PM-1 and PM-2.5 and even PM-1 is detected as quickly as every six seconds. Sensitivity is from 0.1 microgram/m³ upwards and stand-alone system is able to be operated directly all times out in the field. The Model 1108 different from the Model 1107 is able to count the number of particles from 300 nm to 20 μm and collect all three PM fraction of PM10, PM2.5 and PM1.

2.2. Topography in study area

The study area is located in the eastern mountainous coastal region of Korean peninsula (Fig. 1). Two GRIMM aerosol samplers of Model 1107 and Model 1108 were equipped at Kangnung Meteorological Administration (KMA), Kangnung city adjacent to the East Sea in the east. 2.5° degree interval terrain data was used for the largest domain and then 1km interval data was used for fine mesh domain, respectively.

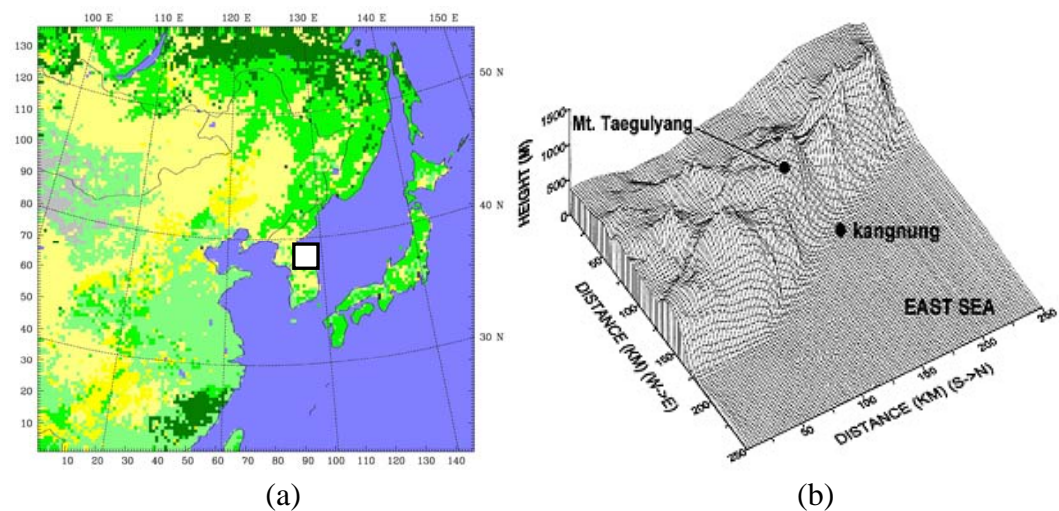


Figure 1. (a) Land-use data for a coarse domain of a horizontal grid size 27 km. Box denotes the adjacent area of Kangnung city (Korea). (b) 3D (right) topography near Kangnung city (20m above mean sea level, 10 km width; box area shown in Fig. 1a) with 90° turning into the right with a horizontal grid size 5 km, and Mt. Taegulyung (860 m), Korea.

3. RESULT AND DISCUSSION

3.2 Weather condition

At 0900 LST (0000 UTC), February 14, 2005, the center of a high pressure of 1033 mb was located in the northern part of Korean peninsula and another high pressure of 1031 mb was located in the mid of Japan. On the other hand, between two high pressure systems, a low-pressure of 1028 mb is located in the west of a coastal city called Kangnung city of Korean peninsula, that is, the East Sea (Fig. 2a) and most of winds observed at Kangnung Meteorological Administration were south-westerly

under the influence of low pressure system, except for north-easterly during 1000 LST to 1700 LST.

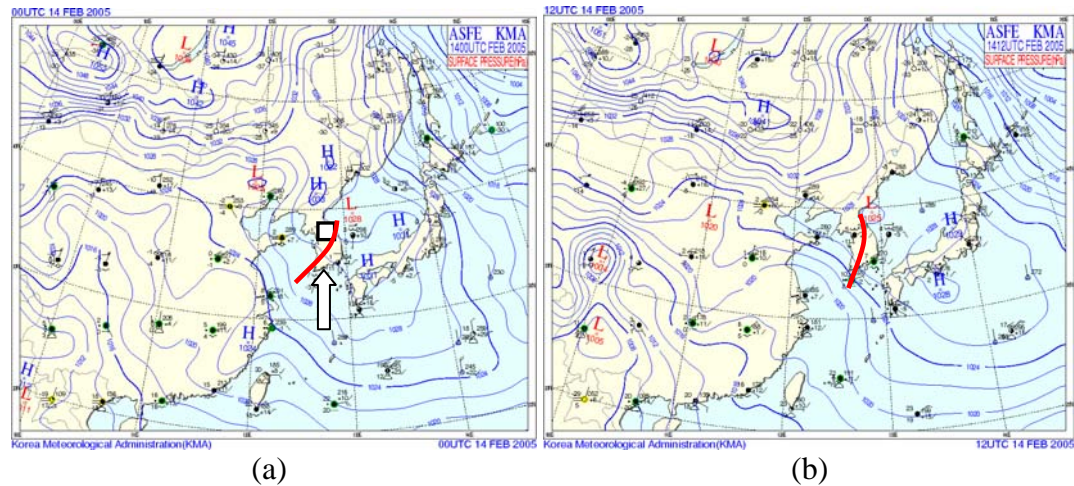


Figure 2. (a) Weather map at 0900 LST and (b) 2100 LST, February 14, 2005 near Korean peninsula. Arrow, box and red line indicate Korean peninsula, the area near Kangnung city and cold front, respectively.

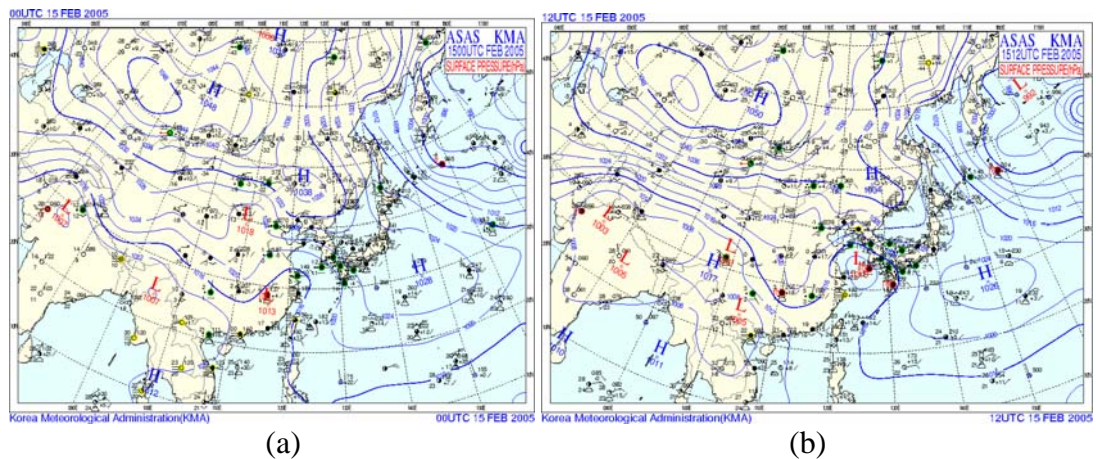


Figure 3. As shown in Fig. 2, except for (a) 0900 LST and (b) 2100 LST, February 15.

At 2100 LST (1200 UTC), the low pressure center became slightly enforced with a magnitude of 1025 mb and cold front moved anti-clockwisely, gradually showing its eastward movement. South-westerly wind still prevailed in the city until 0900 LST, the next day (Fig. 2b). From 0900 LST, February 15, both a high pressure system of 1038 mb located in Manchuria in China over the northern Korea and another low pressure center of 1013 mb located in the south-eastern part of China controlled synoptic weather pattern and local observed wind fields until 1400 LST were north-easterly, inducing moisture advection from the sea into the inland area. After the time, winds were changed into south-westerly until 1800 LST (Fig. 3a).

At 2100 LST, February 15, the high and low pressure systems further moved toward the east, showing the locations of high pressure of 1043 mb in Vladivostok, Russia and the more strengthened low pressure of 1010 mb in the southern Yellow Sea near south-western side of Korean peninsula (Fig. 3b). This pressure pattern could strongly induce north-easterly wind and a great amount of moisture advection from the East Sea into the Kangnung city, showing 6.5 mm precipitation amount.

3.2 Mass concentration of aerosol

From Model 1107, the distribution of 10 minutes averaged mass concentration of PM₁₀, PM_{2.5} and PM₁ near the ground surface at KMA of Kangnung coastal city was given by Fig. 4. From 0000 LST to until 0000 LST, February 14, the mass concentration of PM₁₀ was in the range of 50.62 $\mu\text{g}/\text{m}^3$ ~ 145.12 $\mu\text{g}/\text{m}^3$, especially showing its maximum concentration at 1420 LST, after passage of cold front. The concentrations of PM_{2.5} and PM₁ had also similar patterns to the concentration of PM₁₀. Their concentrations were in the range of 15.14 $\mu\text{g}/\text{m}^3$ ~ 56.19 $\mu\text{g}/\text{m}^3$ for PM_{2.5} and 10.73 $\mu\text{g}/\text{m}^3$ ~ 44.71 $\mu\text{g}/\text{m}^3$. From 0000 LST, February 14, the concentrations of PM₁₀, PM_{2.5} and PM₁ were in the almost steady until 1700 LST, just before the end of office hour and near sunset.

After 1800 LST near the ending time of office hour, the concentration of aerosol rapidly increased up to 145.12 $\mu\text{g}/\text{m}^3$ of PM₁₀ and 88.93 $\mu\text{g}/\text{m}^3$ of PM_{2.5} and 44.71 $\mu\text{g}/\text{m}^3$ of PM₁ at 2000 LST (two hours later after sunset), respectively.

Then the concentrations decreased to 59.78 $\mu\text{g}/\text{m}^3$ at 2200 LST and increased again up to 93.06 $\mu\text{g}/\text{m}^3$ at 0100 LST, February 15. Thus two maximum concentrations of PM₁₀, PM_{2.5} and PM₁ were detected with their first maximum at 2000 LST and secondarily maximum at 0100 LST. After 0100 LST, the concentrations continuously decreased to 19.32 $\mu\text{g}/\text{m}^3$ at 0600 LST. Relatively moderate winds with speeds in the range of 2.9 m/s ~ 4.7 m/s were south-westerly at Kangnung city, until 1000 LST. Then winds became weak with speeds less than 2 m/s and wind directions were also changed into north-easterly from 1100 LST to 1700 LST, due to the turning of cold front anti-clockwisely and easterly sea breeze toward Kangnung city in the eastern coast of Korean peninsula. Relative humidity were still low less than 38 %.

The maximum concentration of aerosol at 2000 LST might be due to the increase of vehicle number on the street near the ending time of office hour and resulted in the increase of aerosol concentrations. Another reason might be the much shrunken of atmospheric boundary layer (nocturnal surface inversion layer; NSIV) under the cooling of ground surface at night, comparing to the depth of daytime convective boundary layer at the city (Choi et al, 2004). Choi (2004) and Choi et al (2004) indicated that in general, particulate matters generated from the ground surface of the downtown were drifted under thermal convection from the surface into the lower atmosphere and transported from Kangnung city near the coast toward Mt. Taeguallung in the west of the city (downwind side) under easterly sea-breeze and valley wind until sunset, staying near the mountain side.

After sunset, downslope wind like mountain wind and land-breeze drove the daytime transported particulate matters in mountain side along the eastern slope of Mt. Taeguallung and land-breeze further induced those particulate matters toward

the city, resulting in a high concentration of particulates near mid night. As nighttime went on, after the ending time of office hour, the number vehicles continuously reduced and PM concentrations also had to decrease. Thus secondary maximum of aerosol concentration could be generated by the shrunken atmospheric boundary layer (NSIV) and the returning of some amount of transported particulate matters from the mountain side toward the city at night.

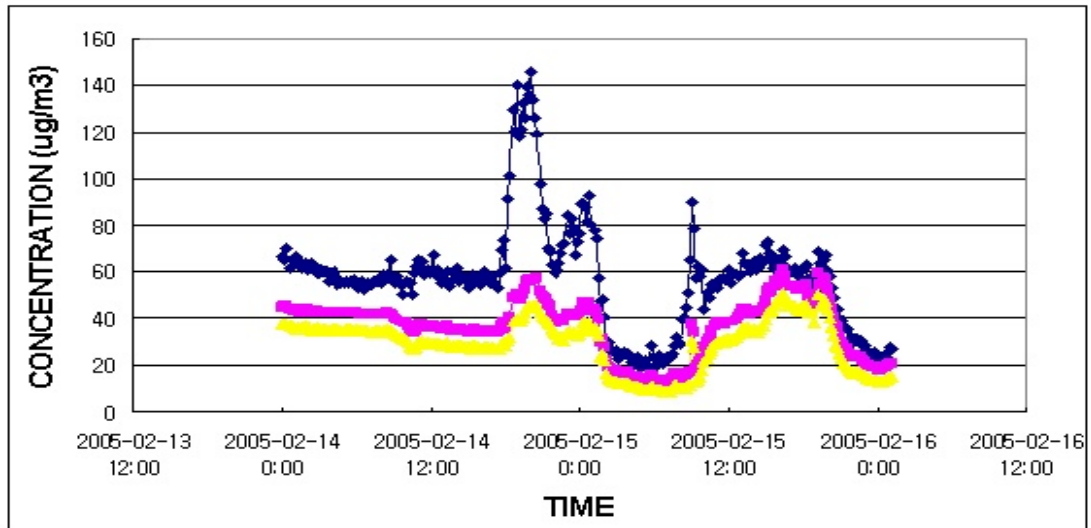


Figure 4. Hourly based concentration ($\mu\text{g}/\text{m}^3$) of PM10, PM2.5 and PM1 at an aerosol sampling point of Kangnung Meteorological Administration from February 14 to 16, 2005.

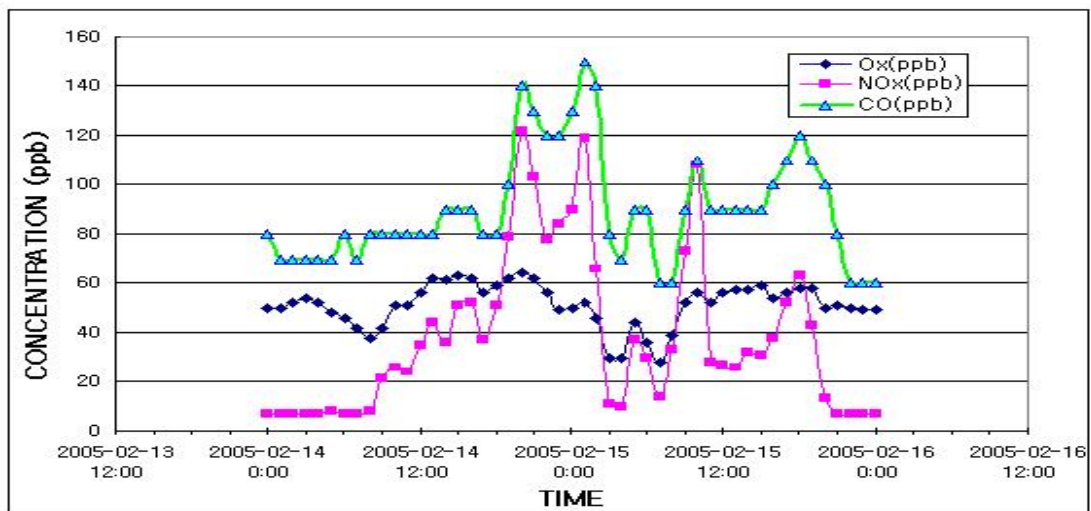


Figure 5. Hourly concentrations (ppb) of CO, NO_x(= NO₂ + NO), O_x(= O₃ + NO₂) at Kangnung Environmental Monitoring Site, Ministry of Environment from February 14 to 16.

After mid night, PM concentrations rapidly decrease until 0900 LST, the next day morning, as most of particulate matters in Kangnung city were dispersed into the East

Sea. Then Kangnung city was in a very clean atmospheric environmental condition less than $30 \mu\text{g}/\text{m}^3$. The relatively high concentration of PM in the morning on February 14 might be attributed to the transportation of some amount of particulate matters from upwind side area (Wonju city) toward Kangnung city. Under the maximum concentration of PM10 at 2000 LST on February 14, PM2.5 and PM1 also had maximum concentrations.

One may postulate that gases phases of PM 2.5 and PM1 made a great contribution to the increase of PM10 concentration. Therefore, it is necessary to compare the concentration of gases of CO, NO_x ($= \text{NO}_2 + \text{NO}$), O_x ($= \text{O}_3 + \text{NO}_2$) at Kangnung Environmental Monitoring Site, Ministry of Environment from February 14 to 16, 2005. Tendency of CO concentration well matched the distribution of PM10, PM2.5 and PM1. The distribution of NO_x concentration also showed a similar, except for concentrations in the morning on February 14. When PM10, PM2.5 and PM1 had the first and secondary maximum concentrations, NO_x concentration had the first and secondary maximum concentrations at 2000 LST on February 14 and 0100 LST on February 15. It implies that gases of NO_x and CO emitted from vehicles on the road due to fuel combustion and boilers in the resident area can make a great contribution to the increase of PM10 concentration. O_x is composed with $\text{O}_3 + \text{NO}_2$ and is assumed as permanent capacity of O_3 . O_x concentrations were generally in the range from 39 ppb to 62 ppb, except for 30 ppb, when NO_x concentrations were very low on February 15. O_x distribution did not reflect the characteristics of CO and NO_x .

3.3 Effect of upwind side transportation

Further consideration was given to the transportation of particulate matter and gases from Wonju city in the upwind side of Kangnung. Fig. 6 showed streamlines simulated by MM5 model, version 3.5, with horizontal interval of 2.5° from 0900 LST, February 14, 2005 to 2100 LST, February 15. Mass transportation was expected from Wonju city (box) toward Kangnung city (red circle) in the downwind side at 0900 LST and 2100 LST, February 14 (Fig. 6a and b), but it might be difficult to be on February 15, because Kangnung city was not in the downwind side on the streamline (Fig. 6c and d). PM10 concentration (here, only possible measurement of PM10 concentration) at Environmental Monitoring Site, Wonju city was in the range of $92 \mu\text{g}/\text{m}^3 \sim 146 \mu\text{g}/\text{m}^3$ from 0000 LST to 1900 LST, and the concentration at Kangnung city, about 200 km away from Wonju city was in the range more or less than $60 \mu\text{g}/\text{m}^3$, showing a half concentration of Wonju city. As two cities were in westerly and southwesterly wind fields for this time period and on the streamline, the occurrence of high PM concentrations at Kangnung city in the downwind side should be affected by the transportation of some amount of particulate matters from Wonju city, resulting in more or less than $60 \mu\text{g}/\text{m}^3$.

Further consideration was given to the contribution of CO, NO_x and O_x concentrations at Wonju Environmental Monitoring Site (Fig. 8). NO_x concentration at Wonju city in the upwind side on February 14, which could influence upon NO_x concentration at Kangnung city was generally higher than ones at Kangnung city. At this city, NO_x concentration rapidly increase with a magnitude of 108 ppb and up to 163 ppb at 2100 LST. Even though the transportation of NO_x from Wonju toward Kangnung for this period could be expected, the transported amount of NO_x could not make a great

contribution to the occurrence of a maximum concentration of NO_x at the downwind side city at 2000 LST on February 14.

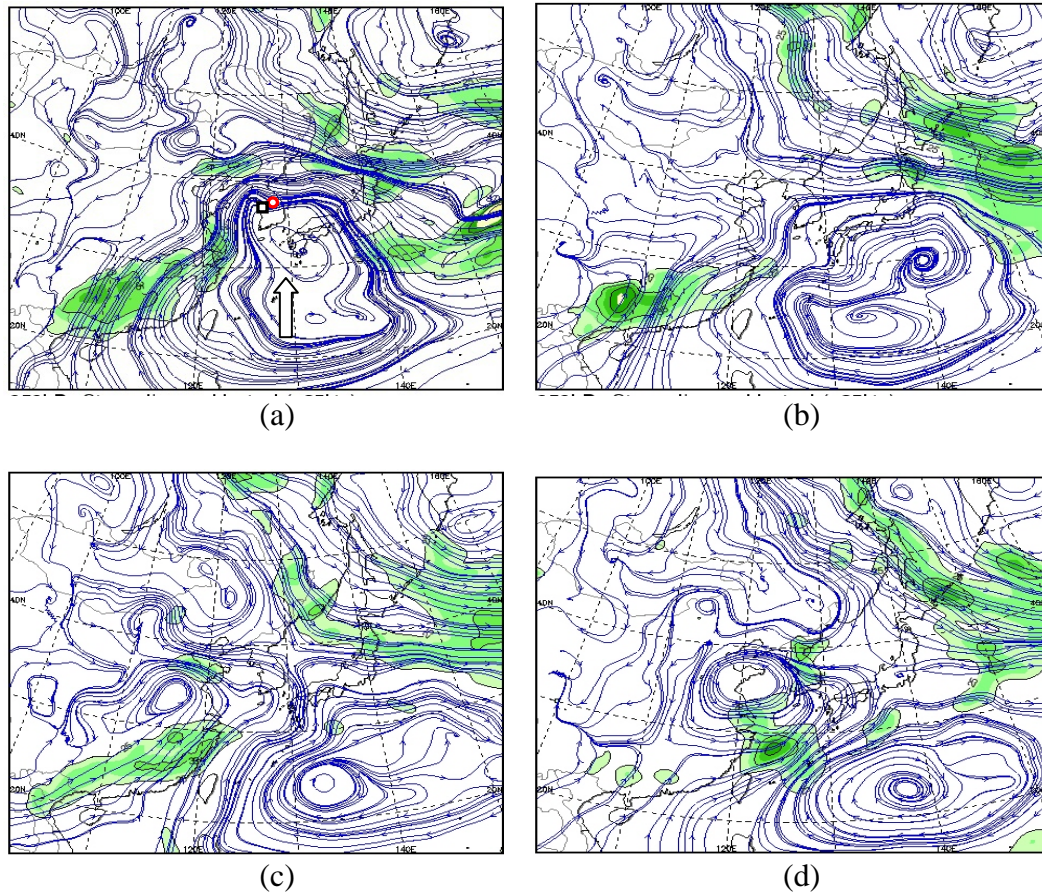


Figure 6. (a) Streamline simulated by MM5 model at 0900 LST, (b) 2100 LST, February 14, and (c) 0900 LST and (d) 2100 LST, February 15 in northeastern Asia. Arrow, box and red circle indicate Korean peninsula, Wonju city (upwind side) and Kangnung city, respectively.

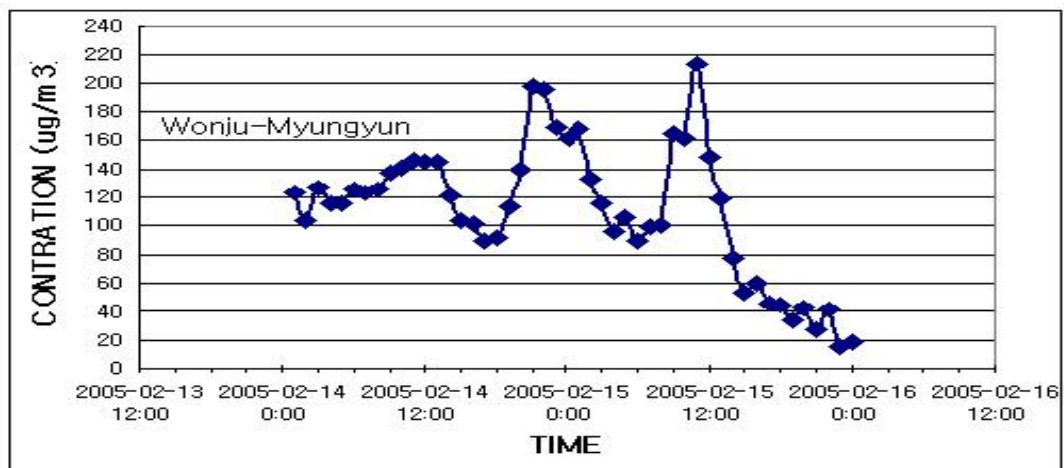
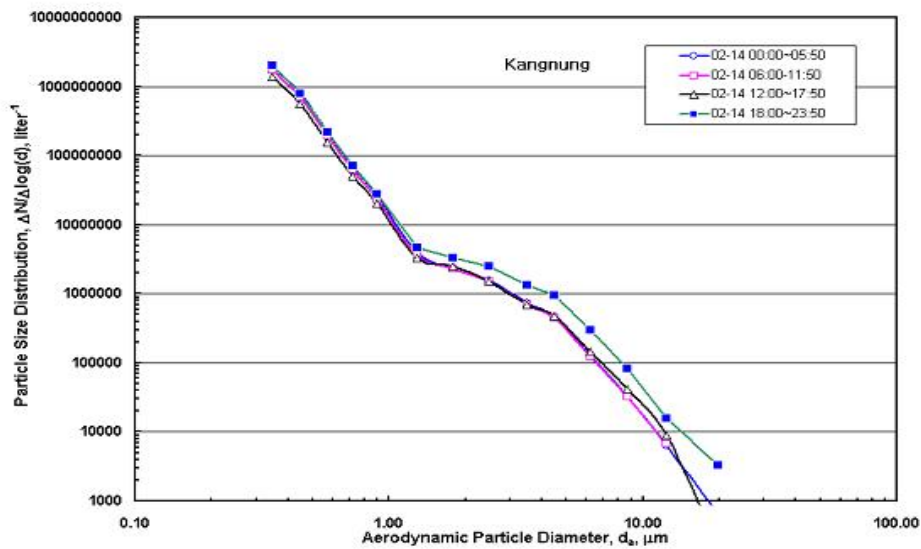


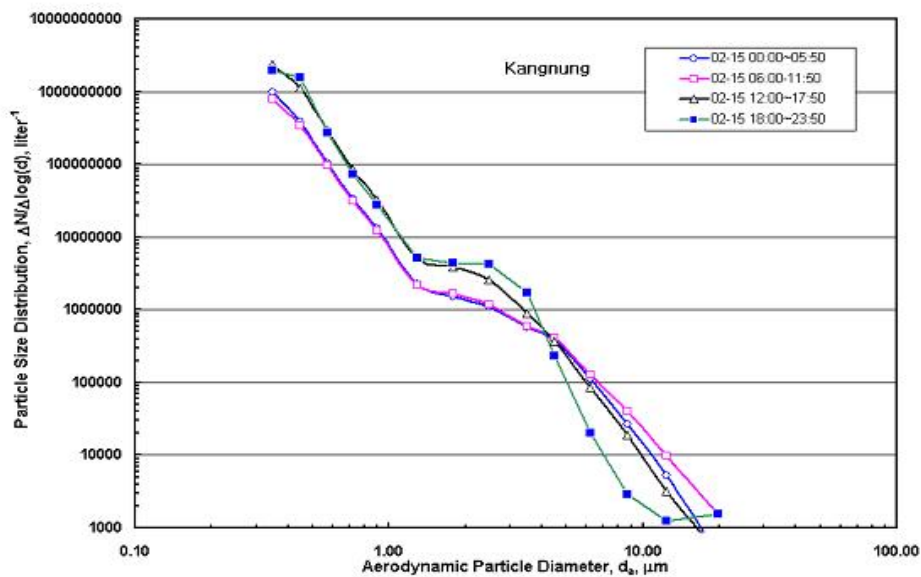
Figure 7. Hourly concentrations ($\mu\text{g}/\text{m}^3$) of PM10 at Environmental Monitoring Site from February 14 to 16, 2005.

3.3 Particle size distribution

The number densities of particle size distributions from 300 nm to 10 μm of particle diameters were given in Fig. 8. The number densities of diameter smaller than 1.2 μm rapidly increased, as the particle diameter decreased (Fig. 8a). On the other hand, the number densities of diameter greater than 1.2 μm to 20 μm gradually increased the diameter decreased.



(a)



(b)

Figure 8. Particle size distribution from 300 nm to 20 μm at KMA aerosol sampling point on February 14 and 15, 2005.

Especially particle size distribution at 1800 LST to 2350 had a different pattern from the others, showing slightly bigger number densities in the diameters of 1.2 μm to 20 μm . This result indicates that large size particles make better contribution to the occurrence of maximum concentrations of PM at 2000 LST and 0100 LST in the previous Fig. 4. On February 15, the number densities of particle size distributions from 300 nm to 4 μm were bigger from 1200 LST to 2350 LST than those from 0000 LST to 1150 LST, as the diameter decreased. The number densities of particle size greater than 4 μm to 13 μm were still increased, but they were much lower than the others. However, the number densities of particle size greater than 13 μm to 20 μm decreased, as particle size decreased.

4. CONCLUSION

The concentrations of PM₁₀, PM_{2.5} and PM₁ showed a typical pattern such as high concentration near 0900 LST (beginning time of office hour) and 1700 LST (ending time of office hour) and their low concentration near noon. PM₁₀, PM_{2.5} and PM₁ had the first maximum concentrations at 2000 LST (three hours later after sunset) and the secondary maximum concentration was detected at 0100 LST, near mid night. When PM₁₀, PM_{2.5} and PM₁ had the first and second maximum concentrations, NO_x had the first and secondary maximum concentrations at the same times.

The increases of NO_x and CO concentrations emitted from vehicles on the street and combustion gases from boilers in the resident area for nighttime could make a great contribution to the increase of PM concentration. Furthermore much shallower nocturnal surface inversion layer than daytime convective atmospheric boundary layer could also increase the concentrations of particle matters. In general, after midnight, the concentration particulate matters decreased due to the reduction of vehicle number, but under westerly wind, the particles transported from the city into the mountain side in the west for daytime returned again to the city and some amount of particles from an upwind side city toward the coastal city could also make a great contribution to the occurrence of secondary maximum. The number densities of particle size distributions, regardless of particle diameters, in general, were much lower in the early morning than in the afternoon and at night.

5. ACKNOWLEDGEMENTS

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