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INFLUENCES OF SECONDARY CHEMICAL GAS PROCESSES ON PM_1, $PM_{2.5}$ AND PM_{10} CONCENTRATIONS OVER THE KOREAN COAST

Hyo Choi[*,†] and Milton S. Speer[‡,§]
[†]*Department of Atmospheric Environmental Sciences*
Kangnung National University, Kangnung, Korea
[‡]*Climate and Environmental Dynamics Laboratory*
School of Mathematics and Statistics
The University of New South Wales, Sydney, NSW
Australia
[*]*du8392@hanmail.net*
[§]*mss@maths.unsw.edu.au*

An aerosol sampler was installed at Kangnung city in the east coast of Korea to measure mass concentrations of PM_{10}, $PM_{2.5}$, and PM_1 covering particle diameter sizes ranging from $300\,\eta$m to $20\,\mu$m from February 14 to 16, 2005. The concentrations of PM_{10}, $PM_{2.5}$, and PM_1 showed a high morning (09:00 LST) and afternoon (17:00 LST) concentration and a low concentration around midday (12:00 LST). First maximum concentrations of PM_{10}, $PM_{2.5}$, and PM_1 occurred at 20:00 LST. Secondary maximum concentrations were detected at 01:00 LST, February 15. The distribution of CO and NO_x concentrations showed a similar diurnal distribution to those of PM_{10}, $PM_{2.5}$, and PM_1, except for the morning of February 14. The first and secondary maximum concentrations of NO_x occurred at the same times as those of PM_{10}, $PM_{2.5}$, and PM_1. This implies that the increase in NO_x and CO emissions from road vehicles and combustion gases from boilers in residential areas can contribute substantially to the increase in PM concentration. As a result of a typical daytime westerly wind regime over Korea, the rotor action of a lee-side easterly breeze transports PM from Kangnung toward the mountains, which is then recycled toward Kangnung in the westerly airflow at night. Some PM from Wonju city upwind on the western side can also contribute significantly to the secondary maximum at Kangnang city on the eastern or lee side.

1. Introduction

Recently, there has been a research focus on fundamental aspects of aerosol science.[1–4] Gao and Anderson[5] investigated characteristics of Chinese aerosols determined by individual particle analysis, where most measurements of aerosol size were determined by laser technology. Adby and Demster[6] developed an algorithm as a dependent method while Hansen[7] and Hansen and O'Leary[8] utilized the independent L-curve approach. Goldberg,[9] Michelewicz[10] and Xu *et al.*[4] applied a genetic algorithm to treat the optimization problem of particle size distribution quite differently from data of multi-spectral extinction measurements.

During the Asian Dust season (Spring) in 2001, the factors of TSP, PM_{10} and $PM_{2.5}$ affecting the cycle of aerosols and their chemical properties and composition were investigated in the Seoul district of Korea.[1] Also, since 2001, comprehensive studies of particulates and gases at many measurement points in China, Korea and Japan have been carried out.[11,12] In Korea, routine measurement of aerosols has been established at Gosan, Jechu island and Taean peninsula on the western side of the Korean peninsula by the Korean Meteorological Administration, but the measurement focus has centered on the mass concentration of TSP and PM_{10} and their chemical composition, while notably excluding the critical PM_1 group. Furthermore, that research does not include particle size distribution of aerosols and the relationship between mass concentration and particle size distribution.

Thus, the objective of this study was to explain the diurnal variation of aerosol concentrations around Kangnung in eastern Korea of not only PM_{10} and $PM_{2.5}$, but also PM_1, which greatly influences the effects of local pollution on human health. The 2-day sampling period of February 14 and 15, 2005 includes dust transported downstream from China in a typical spring synoptic scale westerly wind regime. In Sec. 2, the instruments and experiment are outlined including a brief description of the study area topography. A synoptic overview of the meteorology of the 2-day period then follows in Sec. 3. The measurement results are presented and discussed in Sec. 4 followed by the conclusions in Sec. 5.

2. Methodology and Data

2.1. *Instruments and experiment*

GRIMM sequential mobility particle sizer and counters (Model 1108), was installed at the Gangwon Meteorological Administration office at Kangnung city (37°45′N, 128°54′E), which is located on the eastern side of the mountainous coastal region of the Korean peninsula. Aerosol particle size distributions ranging from $300\,\eta$m to $20\,\mu$m and collection of all three fractions of PM_{10}, $PM_{2.5}$, and PM_1 were measured by the two aerosol samplers from February 13 to 17, 2005, which included both dust in Korea emanating from dust storms further upstream in China, and also, non-duststorm conditions in Korea.

The sampler is specifically designed for PM_{10}, $PM_{2.5}$, and PM_1 ambient environmental air analysis using laser light scattering technology. It enables very precise diameter ranges to be determined for all three PM size classifications. The system allows the user to collect all three PM fractions simultaneously without changing sampling heads or weighing filters. Moreover, the 1107 sampler is the only PM instrument monitor to offer dual technology consisting of both optical and gravimetrical analysis. It 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 analyses on the collected residue. It was developed for particulate measurements via 900 laser light scattering and with multiple particle size passes through a flat laser beam produced by a precisely focused laser and several collimator lenses.

The scattered light is detected by a 15-channel pulse height analyzer which is used for size classification. The counts from each size classification are converted to mass by a well established equation. The complete system consists of 165 fiberglass housing, drying temperature control system, 1107 PM dust monitor, sensors for humidity and temperature and 170M sampling system. Real-time data displays of PM_1 and $PM_{2.5}$ and even PM_1 are sampled as often as every 6 s.

The sensitivity of the system is as low as $0.01\,\mu g/m^3$ and it can be operated directly as a stand-alone system in the field. The purpose of the 1108 sampler is to count the number of particles from 300 ηm to $20\,\mu$m and collect all three fractions of PM_{10}, $PM_{2.5}$, and PM_1. The Model 1108 different from the Model 1107 is able to count the number of particles from $300\,\eta$m to $20\,\mu$m and collect all three PM fraction of PM_{10}, $PM_{2.5}$, and PM_1.

2.2. *Topography in study area and model domain*

The study area is located in the mountainous coastal region of eastern Korea (Figs. 1 and 2). The two GRIMM aerosol samplers (Models 1107 and 1108) were installed at Gangwon Meteorological Administration Office in Kangnung city, which is situated adjacent to the East Sea.

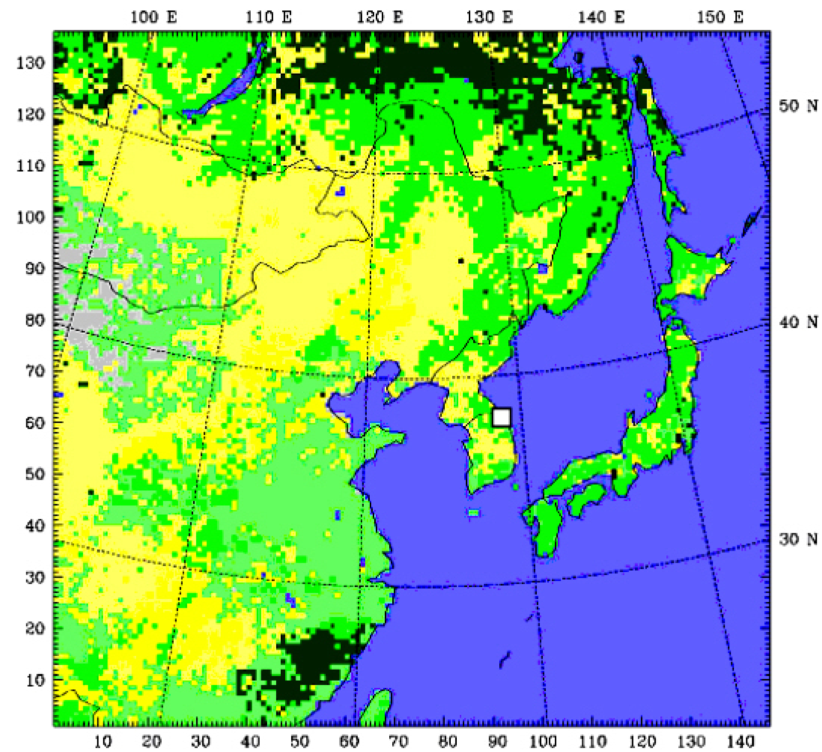


Fig. 1. Map of northeast Asia highlighting land-use data (horizontal resolution 27 km) and the inset box denotes the study area including Kangnung city, Korea.

A horizontal grid interval of 2.5° was used for the largest model domain and simulated streamline and other output obtained from a three-dimensional numerical model called MM5 and 3 km for the fine mesh domain.

3. Synoptic Overview

At 09:00 LST (00:00 UTC), February 14, 2005, high pressure with a central value of 1033 hPa was located over the northern part of the Korean peninsula and another high (1031 hPa) was located over central Japan. Between the two high pressure systems, a weak low-pressure (1028 hPa) was located to the north of Kangnung city over the East Sea (Fig. 3a).

The wind direction observed at the Gangwon Meteorological Administration office was southwesterly due to the influence of the low

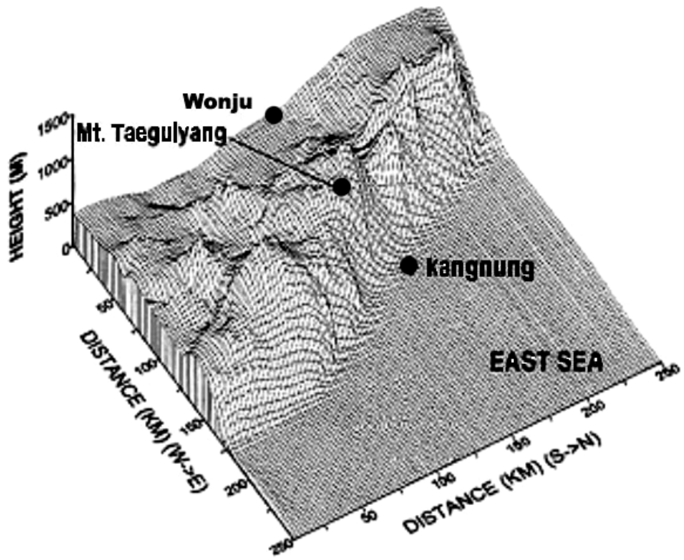
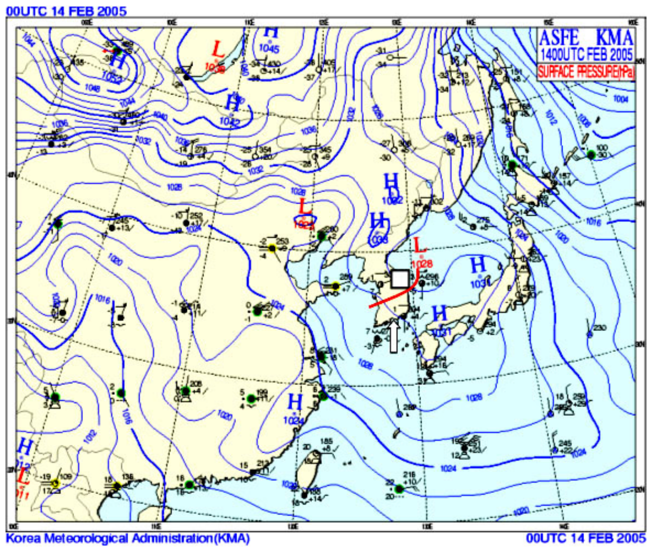


Fig. 2. Map of study area near Kangnung in eastern Korea highlighting topography surrounding Kangnung (horizontal resolution 5 km) and place names mentioned in the text. Kangnung city and Mt. Taegulyang are 20 and 860 m above mean sea level, respectively. Actual north is in 90° left from topographical north in figure.

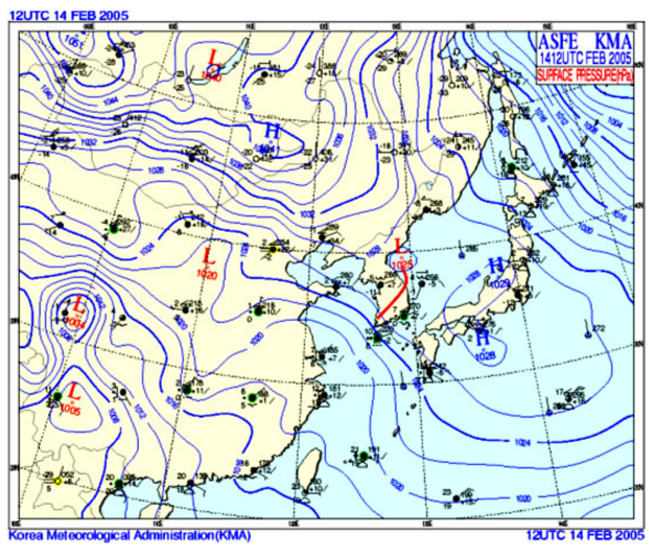
pressure system, although a northeasterly breeze was evident from 10:00 to 17:00 LST. At 21:00 LST (12:00 UTC), the low pressure centre had strengthened slightly to a value of 1025 hPa and the southern portion of a cold front had edged eastwards while the northern section of the cold front had rotated (anti-clockwise) slightly westwards. A southwesterly wind prevailed at Kangnung city until 09:00 LST, February 15 (Fig. 3b).

From that time, both a high pressure system (1038 hPa) located over Manchuria in China and northern Korea and another low pressure center (1013 hPa) located over the southeastern part of China controlled the synoptic weather pattern. The local observed wind direction until 14:00 LST was northeasterly, thereby providing moisture advection from the sea over inland areas. After 14:00 LST, the wind direction changed to southwesterly until 18:00 LST (Fig. 4a).

At 21:00 LST, February 15, the high and low pressure systems moved further toward the east, with the high pressure center (1043 hPa) located near Vladivostok, Russia and the strengthened low pressure (1010 hPa) now centered over the Yellow Sea near the southwestern side of the Korean peninsula (Fig. 4b). This synoptic scale pressure pattern over the

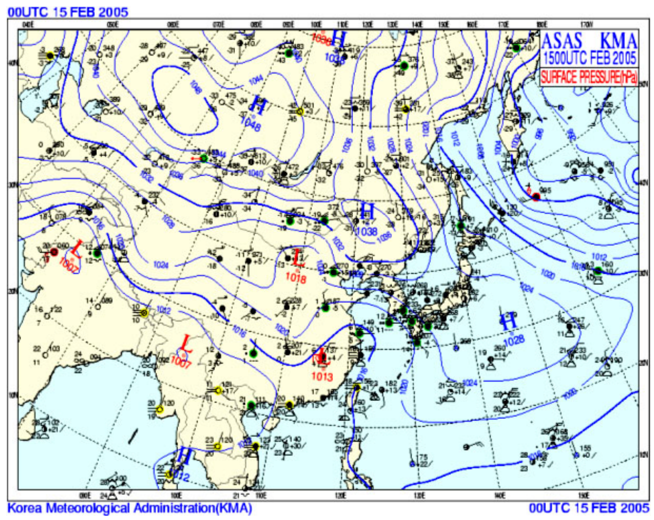


(a)

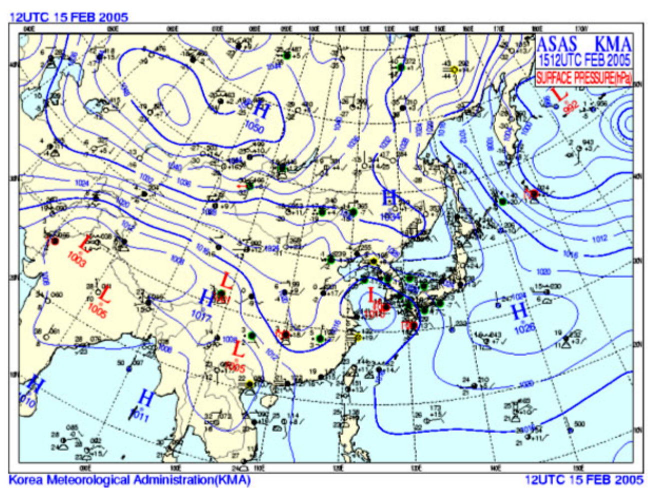


(b)

Fig. 3. Weather charts on February 14, 2005 for (a) 09:00 LST and (b) 21:00 LST. The arrow, square, and curved line below the square indicate the Korean peninsula, the study area including Kangnung city, and the cold front, respectively.



(a)



(b)

Fig. 4. (a) As in Fig. 2, except for 09:00 LST and (b) 2100 LST, February 15.

topography of the Korean peninsula strongly influences the development of a northeasterly rotor circulation over the coast near Kangnung in the lee of the mountains. Moisture advection from the East Sea, as a result of the northeasterly wind, and subsequent cloud formation, was responsible for a total of 6.5 mm of rainfall at Kangnung city.

4. Result and Discussion

4.1. *Aerosol mass concentration*

From the 1107 sampler, the distribution of 10-min averaged mass concentration of PM_{10}, $PM_{2.5}$ and PM_1 near the ground at the Kangnung site is shown in Fig. 5. From 00:00 LST February 14 until 00:00 LST, February 15, the mass concentration of PM_{10} was within the range 50.62–145.12 $\mu g/m^3$, with the maximum concentration of 145.12 $\mu g/m^3$ at 14:20 LST occurring after passage of the cold front.

The concentrations of $PM_{2.5}$ and PM_1 had similar variations to PM_{10}. Their concentrations were in the range 15.14–56.19 $\mu g/m^3$ for $PM_{2.5}$ and 10.73–44.71 $\mu g/m^3$ for PM_1. From 00:00 LST, February 14, the concentrations of PM_{10}, $PM_{2.5}$, and PM_1 remained almost unchanged until 17:00 LST, just before peak-hour traffic and near sunset.

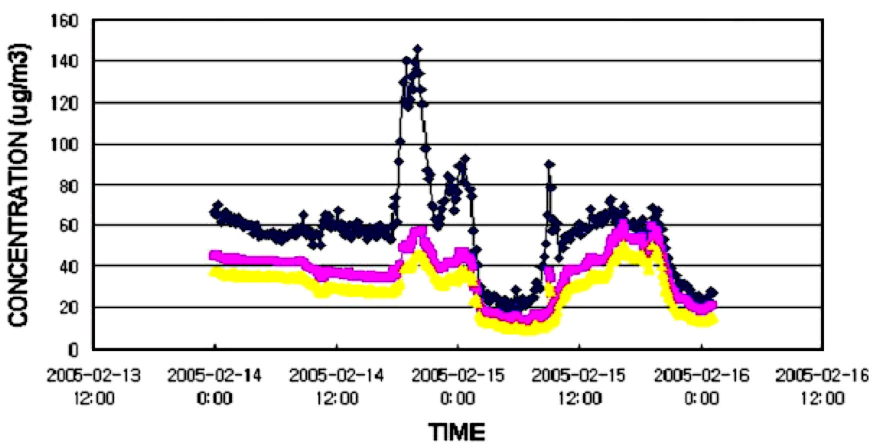


Fig. 5. Hourly concentrations ($\mu g/m^3$) of PM_{10} (blue), $PM_{2.5}$ (pink) and PM_1 (yellow) at Gangwon Meteorological Administration Office in Kangnung city from February 14 to 16, 2005.

After 18:00 LST with peak hour traffic increasing, the concentration of aerosols rapidly increased to 145.12 μg/m^3 for PM_{10}, 58.93 μg/m^3 for $PM_{2.5}$ and 44.71 μg/m^3 for PM_1 by 20:00 LST which is about 2 h after sunset. The PM_{10} concentration decreased to 59.78 μg/m^3 at 22:00 LST, February 14, and then increased again to 93.06 μg/m^3 at 01:00 LST, February 15. Thus, two maximum concentrations of PM_{10}, $PM_{2.5}$, and PM_1 were detected with their first maximum at 20:00 LST, February 14 and a secondary maximum at 01:0 LST February 15.

After 01:00 LST, the PM_{10} concentration continued to decrease to 19.32 μg/m^3 by 06:00 LST. A relatively moderate southwest wind in the range from 2.9 to 4.7 m/s occurred at Kangnung city until 10:00 LST. Then the wind speed decreased to less than 2 m/s and the wind direction changed to northeasterly between 11:00 and 17:00 LST, due to the passage of the cold front and easterly sea breeze directed toward Kangnung city. Relative humidity values remained low at below 38%.

The maximum concentration of PM at 20:00 LST, February 14 is most likely due to the increase in vehicle numbers on the roads after office hours. A secondary reason might be as a result of the nocturnal surface inversion layer (NSIL) forming due to the cooling of the surface at night, compared to the depth of the daytime convective boundary layer (CBL) over the city.[14]

Choi,[13] Choi *et al.*,[14] and Choi and Speer[15] indicated that in general, PM generated from industrial and vehicular emissions in the city of Kangnung drifts from the surface to the lower troposphere due to thermal convection and is transported from Kangnung city which is located near the coast, toward Mt. Taegulyang to the west under the influence of an easterly sea-breeze and valley wind until sunset. There, the PM is trapped under a temperature inversion close to the mountain slope.

After sunset, a strengthening downslope wind, reinforced by a katabatic wind and land breeze drives the PM back down the eastern slope of Mt. Taegulyang toward Kangnung city, resulting in a high concentration of particulates around midnight. With a reduction in vehicle numbers after peak hour traffic the PM concentrations also decrease during the night. Thus, a secondary maximum in PM concentration at night is most likely produced by the shallow NSIL and the recycling of some PM from the mountain slope toward the city.

After midnight, PM concentrations rapidly decrease throughout the morning until 09:00 LST, February 15 as most of the PM in Kangnung city has become dispersed over the East Sea. As a result, Kangnung city by this time is relatively free of PM with values less than 30 μg/m^3. The relatively high concentration of PM during the morning of February 14 can most likely be attributed to the transport of some PM from the upwind side of the mountains, where Wonju city is located. The maximum concentration of PM_{10} at 20:00 LST on February 14 also coincided with the maximum concentrations of both $PM_{2.5}$ and PM_1.

It is postulated that the gas phases of $PM_{2.5}$ and PM_1 can contribute significantly to the increase in PM_{10} concentration. Therefore, it is necessary to compare the gas concentrations of CO, NO_x $(= NO_2 + NO)$ and $O_x(= O_3 + NO_2)$ at the Kangnung Environmental Monitoring Site, established by Institute of Health and Environment, from February 14 to 16, 2005 (Fig. 6). The CO concentration closely matches that of PM_{10}, $PM_{2.5}$ and PM_1. The distribution of NO_x concentration is similar to that of CO, except for the morning of February 14.

When PM_{10}, $PM_{2.5}$ and PM_1 had their first and secondary maximum concentrations, NO_x also had its first and secondary maximum concentrations, namely, at 20:00 LST on February 14 and 01:00 LST on February 15. This means that NO_x and CO gases resulting from vehicle emissions due to fuel combustion and from boilers in resident areas can contribute significantly to the increase in PM_{10}. O_x is composed with $O_3 + NO_2$ and is assumed as permanent capacity of O_3. O_x concentrations are generally within the range 39–62 ppb, except for 30 ppb on February 15,

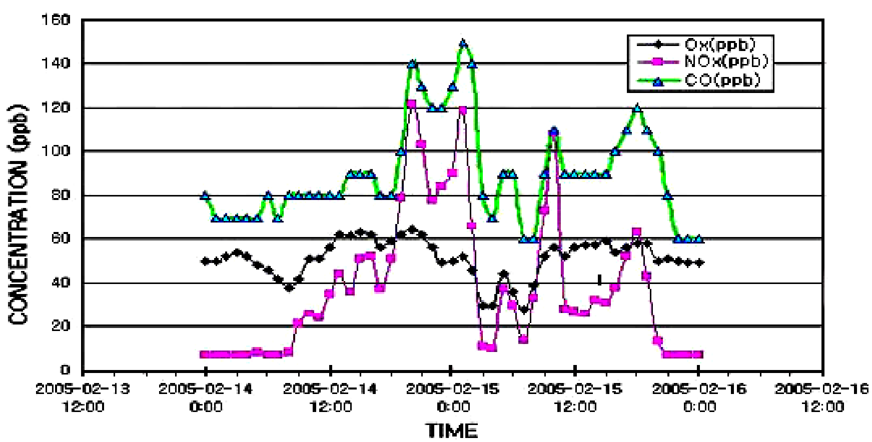


Fig. 6. As in Fig. 5, except for hourly concentrations (ppb) of CO, NO_x (from NO_2 + NO) and O_x (from $O_3 + NO_2$).

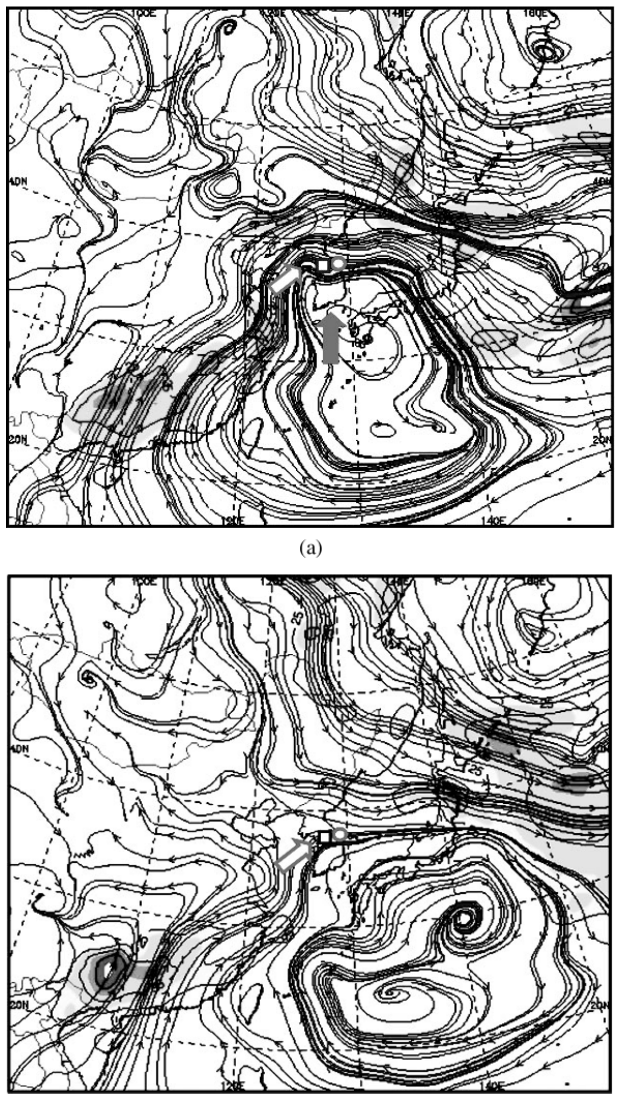
when NO_x concentrations were very low. The distribution of O_x did not reflect the distribution characteristics of CO or NO_x.

4.2. *Effect of upwind PM transport*

Further consideration was given to the transportation of particulate matter and gases from Wonju city, which is located in the west of Kangnung city (Fig. 2). Figure 7a, b shows streamline output simulated by MM5 (version 3.7), using a horizontal grid spacing of 2.5° at 12 h intervals from 09:00 LST, February 14 to 21:00 LST, February 15. Mass transport occurred from Wonju city downwind toward Kangnung city at 09:00 LST and 21:00 LST, February 14. However, on February 15, Kangnung city was no longer on the downwind side (i.c, upwind side) according to the streamline output, as revealed in Fig. 8a, b.

Only PM_{10} concentration could be measured at Wonju city, due to official PM_{10} measurement and the values ranged from 92 to 136 $\mu g/m^3$ between 00:00 LST and 19:00 LST (Fig. 9).[16] The PM_{10} concentration at Kangnung city, about 200 km east of Wonju city was approximately 60 $\mu g/m^3$, or about half the concentration showing at Wonju city. Since the two cities were experiencing westerly and southwesterly winds over this period on February 14, as indicated by the streamlines, the occurrence of high PM concentrations downwind at Kangnung city should be influenced by the transport of some PM from Wonju city, resulting in the PM_{10} concentration of approximately 60 $\mu g/m^3$.

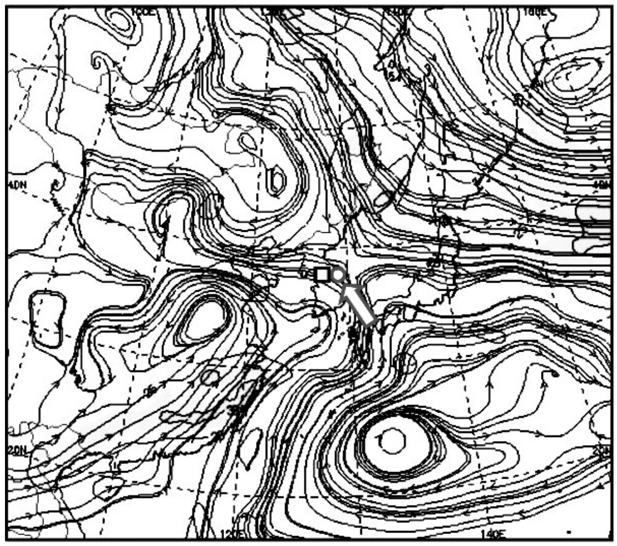
The concentrations of CO, NO_x and O_x at Wonju are shown in Fig. 10. The NO_x concentration at Wonju city (located on the upwind side on February 14), can influence the amount of NO_x concentration transported to Kangnung city and was generally higher than at Kangnung city. At Kangnung, the NO_x concentration rapidly increased to a value of 108 ppb and then to 163 ppb at 21:00 LST. Even though transport of NO_x from Wonju toward Kangnung over this period could be expected, the amount of NO_x actually transported did not contribute significantly to the occurrence of a maximum concentration of NO_x downwind at Kangnung at 20:00 LST on February 14. However, the transported NO_x might partially contribute to the occurrence of the secondary NO_x maximum at 01:00 LST, February 15. Generally, NO_x concentrations at Kangnung city are known to be very low, but when transport occurs under the influence of a strong westerly wind regime, its concentration increases as noted previously.[13]



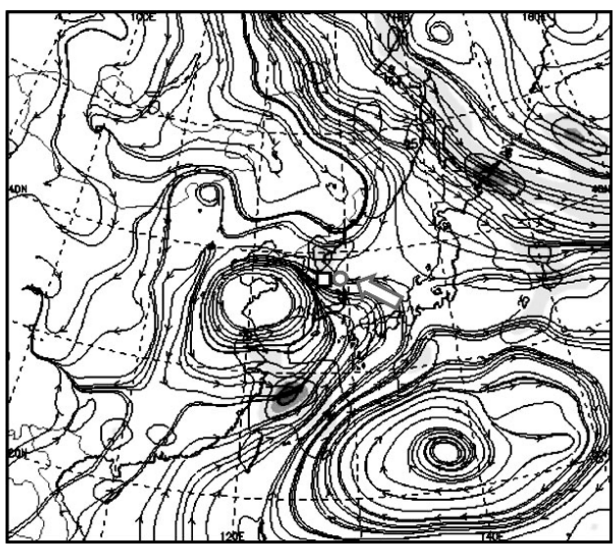
(a)

(b)

Fig. 7. Streamlines in northeast Asia simulated by MMS on February 14 at (a) 09:00 LST; big gray arrow (solid) and small black square (open) in the central part of map denotes Korean peninsula and Wonju city; gray circle (open) denotes Kangnung city and gray arrow (open) denotes upwind side. (b) At 21:00 LST.



(a)



(b)

Fig. 8. As in Fig.7, except for (a) 09:00 LST on February 15 and (b) 21:00 LST. Gray arrow (open) denotes upwind side, which has opposite direction to February 14.

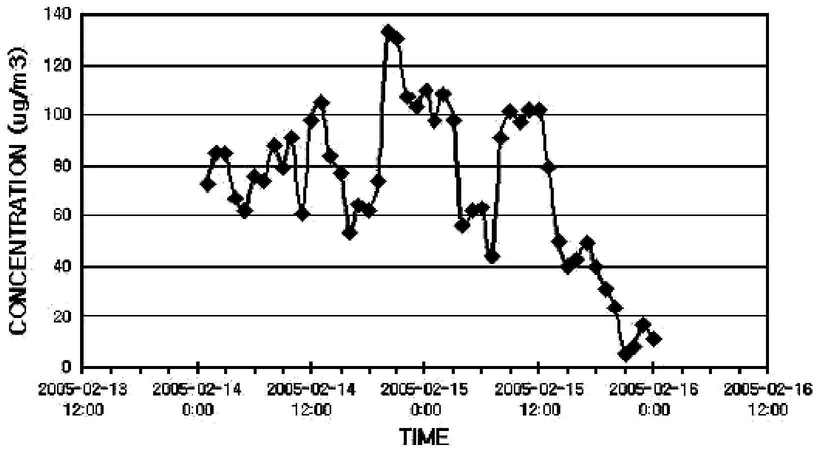


Fig. 9. Hourly concentrations ($\mu g/m^3$) of PM_{10} at environmental site, Wonju city on the upwind side of Kangnung city on February 14, 2005, but the downwind side on 15.

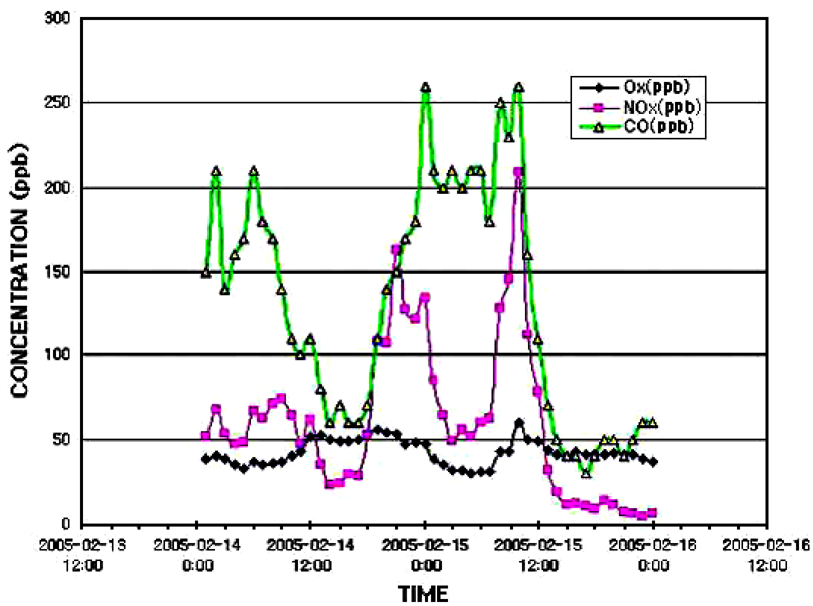


Fig. 10. (a) As in Fig. 9, except Wonju city in the west of Kangnung city.

5. Conclusions

At Kangnung city on the east coast of Korea, diurnal concentrations of PM_{10}, $PM_{2.5}$, and PM_1 measured over a 4-day period were presented for the two days, February 14 and 15, 2005. The concentrations showed variations consistent with high values around 09:00 LST (within morning peak hour traffic period) and 17:00 LST (within evening peak hour traffic period) and low values near midday. PM_{10}, $PM_{2.5}$ and PM_1 all had their primary maximum concentration at 20:00 LST (about 3 h after sunset) and a secondary maximum concentration at 01:00 LST (just after midnight). When PM_{10}, $PM_{2.5}$ and PM_1 had both their primary and secondary maximum concentrations, NO_x also had a primary and secondary maximum concentration.

The increase in NO_x and CO concentrations resulting from vehicle emissions and combustion gases from residential boilers at night can contribute significantly to the increase in PM concentration. Furthermore, a much shallower NSIL than the daytime CBL can also contribute to an increase in PM concentration. PM concentration decreases after midnight due to the reduction in vehicle numbers on the roads. However, under the influence of a westerly wind regime and resulting easterly rotor effect through Kangnung, the PM that is transported in the return easterly flow from Kangnung along the eastern slope toward Mt Taegulyang is recycled toward Kangnung again in the evening.

Also, PM directed from Wonju located upwind toward Kangnung contributes significantly to the occurrence of the secondary PM maximum. The densities of the particle size distributions, regardless of the particle diameter, were generally much lower in the early morning than in the afternoon or at night.

Acknowledgments

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