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Observational Study of Surface Ozone in Jeju Island

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Ozone measurements made from 4 sites in Jeju Island have been analyzed, including those from two urban and two rural locales. The data were analyzed in terms of the seasonal and diurnal trends. It should be clear that the surface ozone levels in Jeju area would be relatively sensitive to the external ozone supply originated from the region of Northeast Asia. It seems to be that due to the reactions of ozone with NOx and CO, the average ozone level in Jeju City appears lower than that in Seogwipo City although Jeju City is the largest city in Jeju Island.

Key Words : Surface ozone, Jeju Island, Long range transport, Urban and rural areas

1. Introduction

Ozone is one of the most important species in the atmosphere and surface ozone has been monitored sporadically at various sites around the globe for the past several decades. Most of the global surface ozone measurement stations in the Northern Hemisphere report a spring peak in their data[1,2]. It has been also reported the highest concentrations during the summer months in several sites in the southeastern United States[3].

Surface ozone is generally the combined result of different production and destruction processes, such as transport from the free troposphere, injection from the stratosphere to the troposphere and subsequent transport to the surface, photochemical production, photolysis, dry deposition and chemical destruction[4].

Recently emissions from anthropogenic sources in northeastern Asia have been increasing drastically. The air pollutant emitted from China can be transported to Korea, Japan and the North Pacific area. Hence there are the growing concerns over the pollution of the coast and shelf of the Yellow Sea. Jeju Island is regarded as the suitable site for the monitoring the long range transport of air pollutants. Several studies based on the data monitored at Gosan station have been reported. However, those studies are mostly focused on aerosol behavior and the characteristics of air quality variations[5,6]. No intensive attempt has been performed to investigate the characteristics of ozone behavior and the urban effects on surface ozone concentrations throughout Jeju Island.

In this study, surface ozone data integrated at four stations in Jeju area were analyzed in terms of temporal variations. The aim of this study is to characterize the ozone behavior in Jeju Island, where the averaged ozone concentrations are usually revealed high in comparison to other areas in Korea. This study will also shed light on the potential transport of regional pollution to Jeju Island.

2. Methods

2.1. Monitoring sites

Figure 1 shows geographical locations of ozone monitoring sites in this work. Jeju Island is a volcanic island with peak elevation of 1950 m and located in the East China Sea; about 500 ㎞ east-northeast of Shanghai, China; about 250 ㎞ west of Kyushu, Japan; about 100 ㎞ south of the Korean peninsular. It is well known that Jeju Island is one of the cleanest places in Korea.

Gosan station is built by the National Institute for Environmental Studies in 1995. The Ido and Donghong stations are managed by the local government of Jeju Province. Gosan and Ido stations have been operated from 1995 and Donghong station from 2002. The measurement at Cheona station starting from 2003 is

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only complementary observation to the investigation of ozone behavior in Jeju Island.

Ido station is located at the downtown of Jeju City with about 300,000 inhabitants and Jeju City is the largest city in Jeju Island. The Donghong station is located at the residential area of Seogwipo City with the population of 84,000. Gosan Station located at the western edge of Jeju Island is on a cliff of 72 m above sea level and has a 220° ocean view stretching from southeast to north. Cheona station is located at 700 m above sea level and surrounded by vegetation, namely mixed deciduous trees and conifers. This station is very close to the downtown of the Jeju City so its atmosphere is likely influenced by the atmospheric properties of Jeju City.

Four monitoring stations are shown in Fig. 1 and the elevations and instrumental information on ozone measurement are given in Table 1. The measurements are recorded in hourly intervals which make them applicable for diurnal as well as seasonal studies.

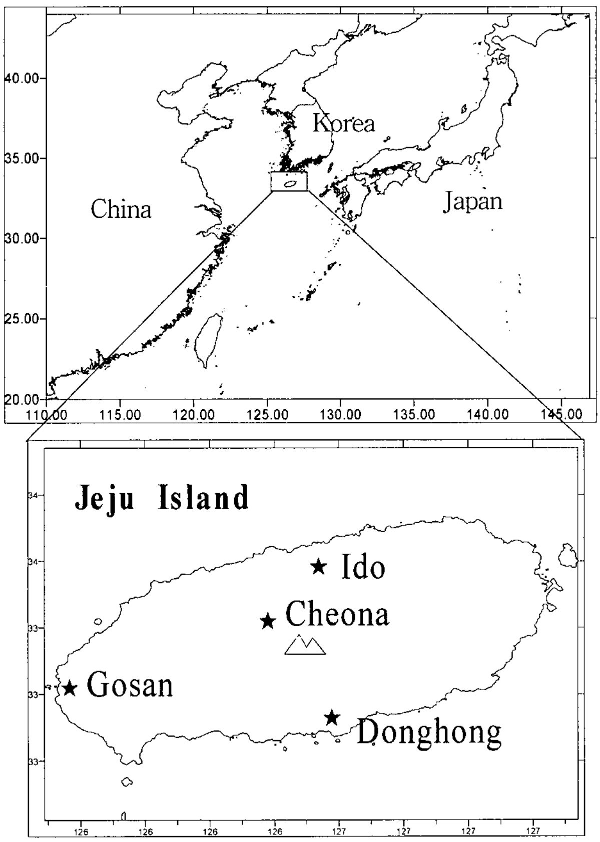

Fig. 1. Geographical location of observatory sites in this study.

Table 1. The location and instrumentation employed at the four ozone monitoring stations

Station	Lat. (°N)	Lon. (°E)	Elev. (m)*	Instrument used (O_3/NOx/CO)	Detection method
Ido	33.50	126.53	59	API/ 300A/200A/400A	
Donghong	33.26	126.58	49	Dongan/DA 4300/2300/3300	O_3 : UV absorption CO : IR absorption NO_x: Chemiluminescence
Gosan	33.28	126.17	76	TEI/ 49C/42C/48C	
Cheona	33.39	126.46	704		

* : the elevation of sampling inlet

Further details on the specifics of the measurements can found in the document[7].

The examined period was basically from 2000 to 2004. However, the measurements of Donghong and Cheona stations were started from 2002 and 2003, respectively. The ozone concentrations during the whole year of 2003 at Gosan station were unusually lower than those for years from 1995 to 2002 and 2004. We need to check closely. Therefore, the data set for 2003 at this station was excluded in this work.

2.2. Meteorological Information

The data for surface meteorological parameters for Ido, Donghong, and Gosan were provided by National Weather Station. At Cheona station, the meteorological parameters were directly measured in the field.

Backward trajectories used in the data analysis were calculated using the HYSPLIT4 model. The meteorological data set for this model was provided by the NOAA (National Oceanic and Atmospheric Administration) and the initial altitude was set to 1500 m.

3. Results and Discussion

3.1. General Characteristics

Statistical data of ozone concentration for the examined period are summarized by the site in Table 2. Attention should be paid to the differences among the measurement periods of the four stations. Table 2 shows that there are the significant differences of local mean ozone concentrations. This result suggests that the ozone production and destruction mechanisms differ between sites. That is, the complicated interplay of the many factors influenced the ozone formation and destruction. However, it can be found that maximum values were similar at the three sites. These similar values of maximum ozone concentrations are attributed to the effects of the external ozone supply.

Based on the data shown in Table 2, it seems that mean ozone levels on an annual basis over Jeju area are similar to the background ozone levels. Many studies revealed that background ozone concentrations in the Northern Hemisphere are in the range of 35~40 ppb[9,10].

It is interesting to note that, as shown in Table 3, the average concentration of 29.7 ppb at Ido station in Jeju City with the population of 300,000 is much lower than that of 34.0 ppb at Donghong station in Seogwipo City with the population of 83,000. We can easily consider that the emission rate of precursors differs between two cities.

The 5th, 50th (median), 95th and 99th percentiles of ozone hourly mean values at the four sites were also shown in Table 2. 95th percentile concentration of ozone, an indicator of high ozone concentrations[11], takes the value of 52, 61, 65, and 66 ppb at Ido, Donghong, Gosan and Cheona, respectively. This means that, for example, at Ido station the ozone concentration is not exceeded 52 ppb during 95% of time

Table 2. Statistical data for hourly ozone concentration in Jeju Island during 2000~2004

Station (target period)		Ido ('00~'04)	Donghong ('02~'04)	Gosan ('00~'02, '04)	Cheona ('03~'04)
Mean		30	34	41	37
Max. hourly		100	97	102	127
Percentile (annual)	99	63	76	75	82
	95	52	61	65	66
	50	30	33	42	37
	5	7	7	15	10

Table 3. The t-test result for equality of means of ozone concentrations between Ido and Donghong stations

Station	mean	SD	t-test for equality of means			
			variances	t-value	df	2-Tail Sig.
Ido	29.74	13.492	Equal	-29.843	43550	.000
Donghong	34.02	16.309	Unequal	-28.746	32447.871	.000

on five-year basis.

3.2. Seasonal variations in Ozone

To characterize monthly ozone variations in Jeju Island, 1-hour ozone concentrations of four monitoring stations were gathered, and the percentiles and the monthly mean concentrations were calculated and depicted in Fig. 2.

Figure 2 shows the seasonal variations of surface ozone as the multi-years monthly averaged concentrations at Ido, Donghong, Gosan and Cheona. All the sites in Jeju Island have very similar temporal patterns of ozone behaviors. In general, monthly mean ozone levels exhibit a noticeable drop in July and August, following a spring peak, and then escalate again in fall. The spring maximum is a common occurrence in remote background site around the world and also distinct springtime ozone maximum is a general feature in many mid-latitude regions of the Northern Hemisphere[8,12]. This can be attributed to stratospheric intrusion[13,14] and the longer lifetime of ozone during winter and the resultant accumulation of man-made ozone[15]. Similar seasonal behavior of ozone, showing a spring maximum and a summer minimum, was reported by several researchers[2,9,16].

It can be also confirmed a noticeable decrease of ozone level in July and August. This summer monsoon introduces unstable conditions, with cloud, boundary layer venting and heavy rainfall. These conditions are not conducive to photochemical production of ozone and accumulation of air pollutants[17]. The inflow of maritime air masses from the Pacific Ocean can also affect the low level of ozone concentration during summer over Jeju Island.

Figure 2 also shows that monthly patterns for the 5th percentile concentrations do not resemble those for the 95th percentile concentrations. The monthly patterns for the 5th percentile concentrations roughly resemble among stations and the 5th percentile concentrations still remain significantly high at all the sites. These patterns in Jeju Island are similar to those found in other coastal areas in Korea[16]. These results suggest the contribution of the external ozone supply can be a more important factor than the influence of local emissions of ozone precursors in Jeju area.

An re-elevation in ozone levels is also shown in fall. It is well known that the re-elevation of ozone in

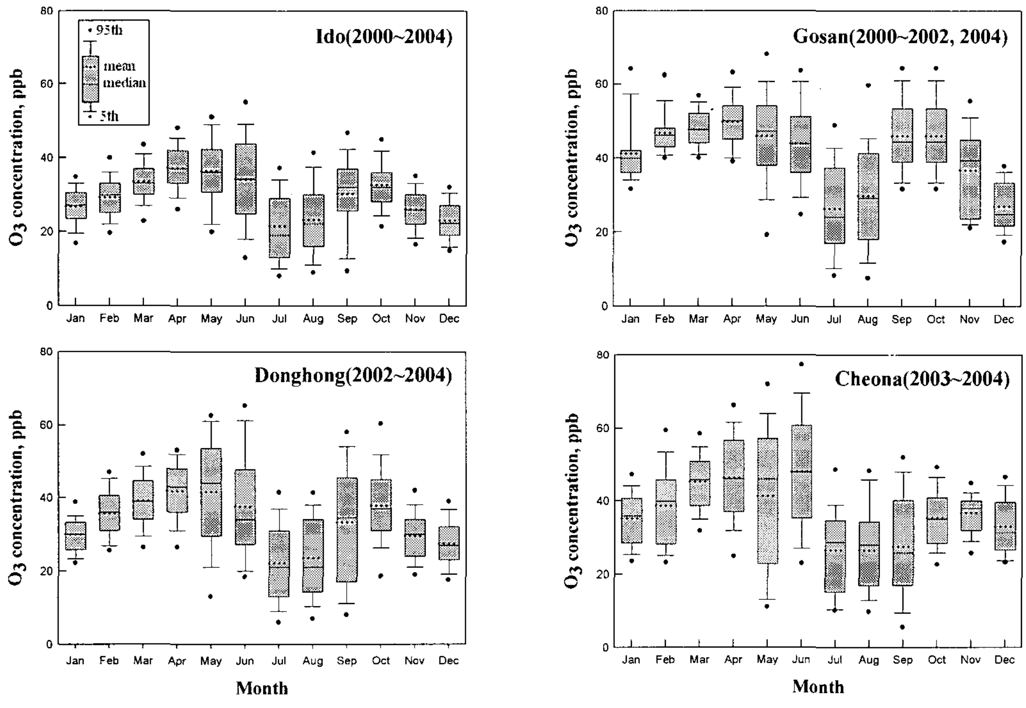

Fig. 2. Multi-year monthly averaged O_3 seasonal variations. The periods of O_3 monitoring varied among sites from 2000 to 2004.

fall is connected with the continental outflow from Eurasian continent. Sunwoo *et al.*[12] noted bimodal peaks in fall as well as in spring at southern island stations in Japan. This study also revealed that the cause of the fall peak is due to the inflow of ozone rich continental air mass in this season.

3.3. Diurnal variations in Ozone

Ozone variation over the diurnal scale can provide insight to the interplay of emissions, chemical and physical processes that operate on a diurnal cycle. Average diurnal variations of ozone for different months of the period 2000~2004 were shown in Fig. 3. It can be shown a clear diurnal pattern on each month at each station. During the day, ozone concentration starts increasing gradually after sunrise, attains maximum value during the afternoon and then decreases. All the sites closely follow this diurnal pattern during daytime. However, the nighttime ozone patterns become significantly different between urban and rural sites. The urban site obviously has a nighttime maximum concentration of ozone but there is no obvious maximum point (the flat curve in Fig. 3) in nighttime ozone concentration for all the rural sites.

In the diurnal cycle of ozone, the minimum ozone concentrations are observed during morning hours, that is, rush hours (07:00~09:00), as illustrated in Fig. 3. It is well known that indications of titration of ozone are obviously observed at urban sites. It can be also found that the appearance time of minimum ozone concentration by the month tends to follow the time of sunrise. Low levels of ozone during morning hours are due to the combined effects of chemical loss by NO and NO_2 species and suppressed boundary layer mixing processes[18]. During morning hours, lower boundary layer height reduces the mixing processes between the ozone poor surface layer and the ozone rich upper layer largely in comparison to the noontime and contribute to morning low O_3 levels.

The Ido and Donghong stations located at urban area have not only a nocturnal ozone peak but a broad daytime ozone peak. The ozone enhancement in the afternoon suggests an active in situ photochemical production and/or mixing of ozone-rich air mass from aloft following the breakup of the nocturnal boundary. At these sites, diurnal patterns in ozone, exhibiting daytime high concentrations with large fluctuation were observed throughout the year except summer months. As shown in Fig. 3, during monsoon months (July and August) the diurnal fluctuation of ozone is found to be small, of the order of 7~20 ppb amount-

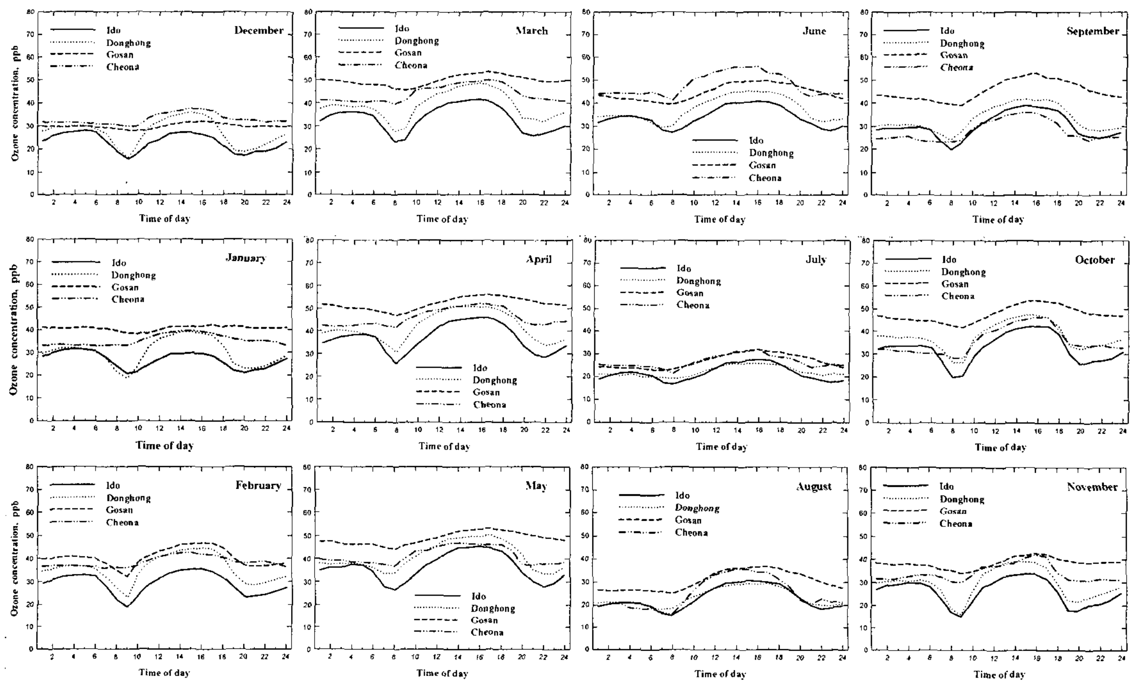

Fig. 3. Diurnal variations of average O_3 concentrations in different months for the 2000~2004 period observed in Jeju Island.

ing to only 33~75% of average ozone levels.

During the warm season, the rural sites, especially Cheona station, have a photochemistry induced diurnal cycles with an afternoon peak that is roughly 6~15 ppb higher than the nighttime concentrations. Figure 3 also shows the flat variability for rural site in the nighttime hours in contrast to the greater variability for urban site. Flat patterns of nighttime ozone concentrations in rural sites indicate that the ozone concentration is directly influenced by the external supply of ozone. Rodriguez and Guerra[19] also suggested that the ozone could reach the background concentrations in the oceanic boundary layer under nocturnal conditions. This means that the influences of local sources on the rural ozone concentration seem to be insignificant. In general, the diurnal fluctuation at Gosan station seems to be relatively small comparing to that at Cheona station. Because of absence of emission sources around Gosan station. it can be ignored the effects of local sources. These observations also reflect the differences, from site to site, in the competition between deposition and convective advection of ozone.

3.4. Diurnal variations in NOx and CO

Figures 4 and 5 show the average diurnal variations of NO_2 and CO during different months. In urban sites, both, NO_2 and CO show buildup during morning (07:00-09:00) and late evening hours (18:00-22:00) which is different from the variations in ozone. Higher levels of NOx and CO during morning and late evening hours were due to the combinations fo anthropogenic emissions, boundary layer processes, chemistry as well as local surface wind patterns. During these hours of maximum concentrations of NOx and CO, anthropogenic emissions are also prominent due to rush hours. It is important to note that the major anthropogenic source for CO and NOx are the fossil fuel burning, that is, the combustion in motor vehicles.

The lowest concentrations for both the species in urban sites are observed between 04:00 and 06:00 in all the months as shown in Fig. 4 and 5. The nighttime ozone peaks in urban sites also appear during this period, as illustrated in Fig. 3. In rural sites, no definite diurnal variations of both the species can be observed.

It was confirmed that in urban sites, an increase in the concentrations of NOx and CO (Fig. 4 and 5) results in a decrease of the ozone levels (Fig. 3). It can be seen that the daily patterns of ozone concentrations are opposite to the trend of daily NOx and CO cycles.

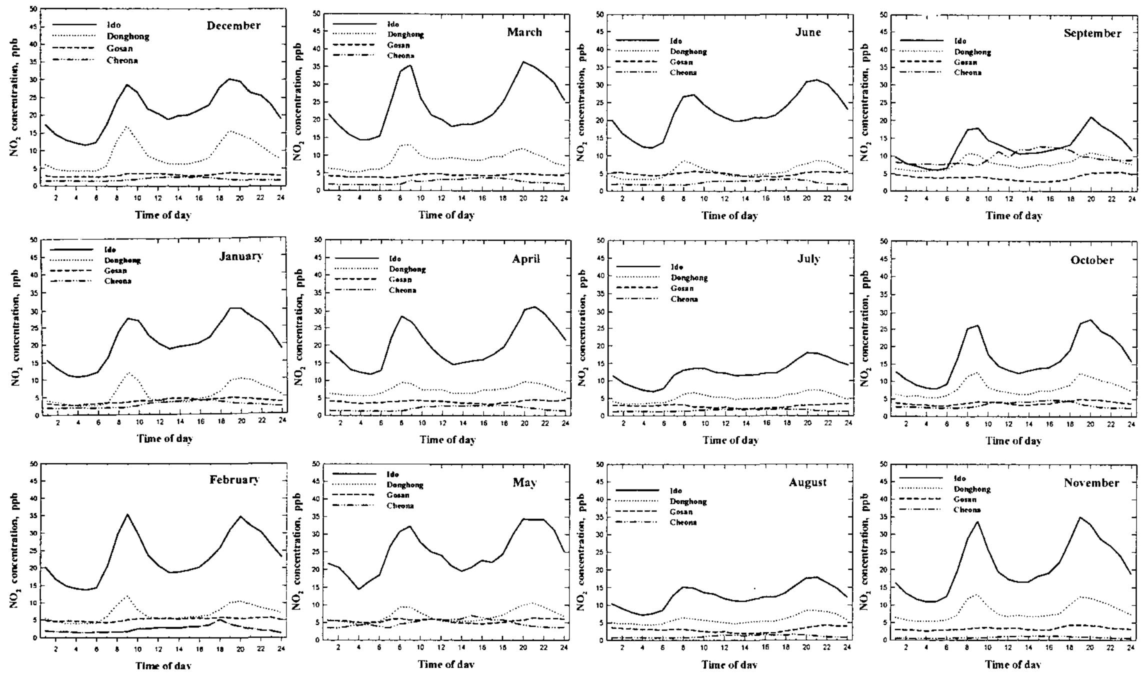

Fig. 4. Diurnal variations of average NO_2 concentrations in different months for the 2000~2004 period observed in Jeju Island.

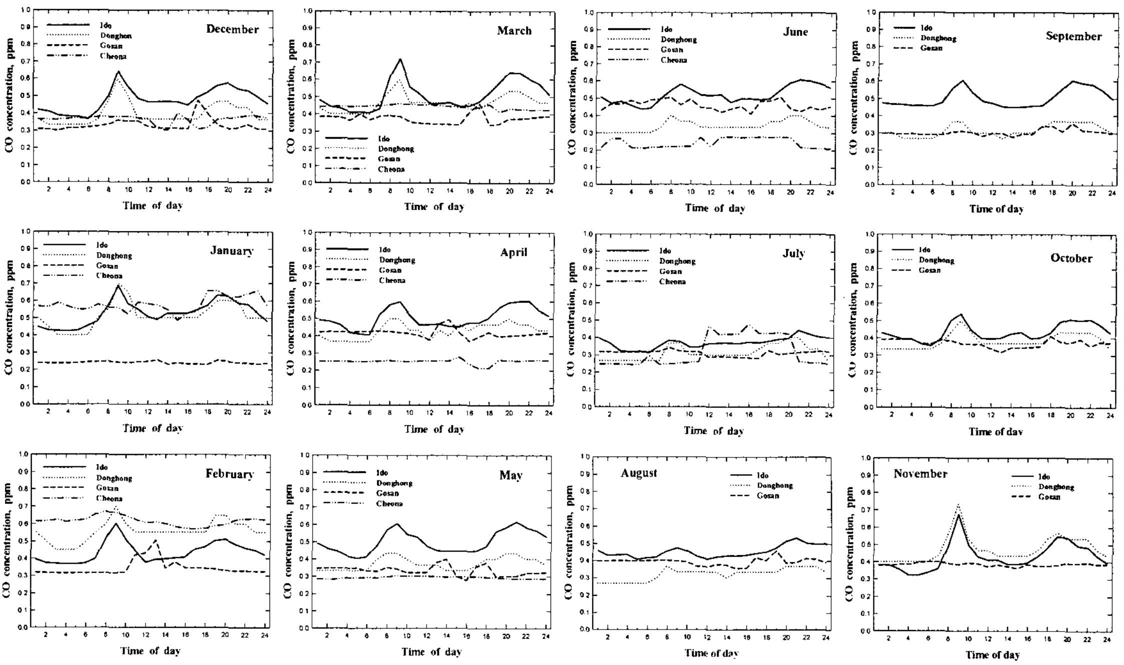

Fig. 5. Diurnal variations of average CO concentrations in different months for the 2000~2004 period observed in Jeju Island.

The average ozone concentration was lower and the average NO_2 concentration was higher at Ido station (Jeju City) than at Donghong station (Seogwipo City). NO_2 predominates on the NOx concentrations in typical air quality monitoring stations, which indicates that the sources of NOx are not very close to the monitoring site. However, a large number of motor vehicles always move around Ido station. It is noteworthy that the reaction of NO with O_3 is about two orders faster than any other chemical loss reactions. Therefore, the lower level of ozone concentration at Ido station is attributable to these reasons, a large number of motor vehicles and the O_3-NOx behaviors.

3.5. Trajectory analysis

To gain insight into the relationship between ozone episode and the meteorological conditions, surface meteorological data were analyzed. Table 4 compares the averaged weather parameters for the high ozone event days (with the ozone levels exceeding 8-hour averaged ozone concentration of 60 ppb during at least 3 consecutive days). It can be seen that for the high ozone event days, both hours of sunshine and the difference between maximum and minimum temperatures increased considerably in comparison with the average conditions. In addition the amount of rainfall is much less on high ozone event days. Intense solar radiation, high temperatures and light winds have found favorable for the formation of high levels of ozone. For the present study sites, significant difference in wind speed was found between high ozone event days and typical days.

To identify the sources of the high and low ozone concentrations recorded in Jeju Island the backward trajectory analysis was used. The examples of trajectories arriving at Jeju Island were shown in Fig. 6 for the high ozone events although more detailed studies will be needed for obtaining the objectivity for these results. The trajectories can be classified into three major routes (Route 1, 2, and Route 3) which are thought to represent airflow patterns arriving at the station. Route 1 (a, b in Fig. 6) and 2 (c in Fig. 6) typically originates from China continent and Mongolia, respectively and pass through the industrialized area of China and Yellow Sea directly reaching Jeju Island. Route 3 originates from the north of East Sea, moves toward the southeast across the Honshu of Japanese Islands, and then bends westward clockwise to reach Jeju Island via southern part of Japanese Islands (d in Fig. 6). These routes allow polluted air to mix and also have enough time for photochemical buildup of ozone.

4. Conclusion

From an analysis of the surface ozone data at the four sites in Jeju Island during 2000~2004 the following conclusions were found.

Surface ozone observed at urban and rural sites in Jeju Island showed the seasonal variation with spring and autumn peaks and summer minimum.

From our findings of moderate ozone levels at night at urban sites, relatively high levels of the 5th percentile concentrations, and no obvious diurnal variation of ozone concentration at Gosan station without the distinct anthropogenic emission sources of pollutants, it should be clear that though the surface ozone level in Jeju Island is, to some extent, directly influenced by the local anthropogenic emissions when atmospheric conditions are favorable, it would be very sensitive to the external ozone supply, originated from the regional anthropogenic emissions of NOx and other pollutants in the region of Northeast Asia. These facts are demonstrated from the results of trajectory analysis.

Although Jeju City is the largest city in Jeju

Table 4. Comparison of monthly mean local meteorological data with those averaged for high ozone event days (8h $\geq$ 60 ppbv)at Gosan and Donghong stations[1)]

Station	Month	Temp. (°C)	Max. temp. (°C)	ΔT (°C)	Wind speed (ms^{-1})	Hour of sunshine (h)	Rainfall (mm)	Pressure (hPa)
Gosan	Jan-02	7.6 (12.0)	10.4 (14.8)	5.7 (7.0)	13.7 (7.4)	3.3 (6.5)	29 (0.3)	1021 (1018)
	Feb-02	7.2 (8.0)	10.3 (11.8)	6.2 (6.8)	9.9 (7.5)	6.7 (9.2)	49 (0.0)	1024 (1024)
	Mar-02	10.7 (11.9)	14.2 (15.7)	6.7 (7.8)	8.8 (6.7)	5.4 (6.5)	61 (3.5)	1018 (1022)
	Apr-02	13.7 (12.6)	17.1 (16.2)	6.2 (6.8)	8.1 (6.2)	4.4 (5.8)	71 (0.0)	1016 (1017)
	May-02	16.5 (16.9)	19.9 (20.7)	5.7 (6.3)	4.8 (4.5)	4.0 (7.5)	96 (1.7)	1012 (1012)
	Jun-02	20.9 (20.8)	24.4 (24.2)	6.3 (6.4)	4.4 (4.3)	6.7 (6.6)	52 (22.0)	1008 (1007)
	Jul-02	23.4 (22.7)	26.6 (27.0)	5.3 (6.2)	5.9 (4.9)	3.9 (4.3)	274 (1.9)	1005 (1001)
	Aug-02	24.9 (24.2)	27.7 (27.0)	5.2 (6.2)	6.1 (5.1)	5.4(10.2)	309 (0.0)	1008 (1011)
	Sep-02	22.4 (22.3)	26.0 (25.6)	6.2 (6.1)	5.2 (5.3)	7.7 (8.9)	110 (0.0)	1014 (1014)
	Oct-02	17.1 (18.8)	20.4 (22.5)	6.1 (7.3)	7.7 (5.7)	5.6 (8.0)	77 (0.0)	1017 (1018)
	Nov-02	10.8 (NA)	13.8 (NA)	6.1 (NA)	9.7 (NA)	4.5 (NA)	30 (NA)	1022 (NA)
	Dec-02	8.4 (NA)	10.9 (NA)	4.7 (NA)	9.7 (NA)	2.6 (NA)	67 (NA)	1024 (NA)
Dong-hong	Jan-02	9.1 (NA)	12.7 (NA)	6.7 (NA)	3.7 (NA)	5.7 (NA)	38 (NA)	1021 (NA)
	Feb-02	9.7 (NA)	13.8 (NA)	7.5 (NA)	3.0 (NA)	7.1 (NA)	52 (NA)	1023 (NA)
	Mar-02	12.9 (NA)	16.6 (NA)	7.2 (NA)	3.1 (NA)	5.7 (NA)	101 (NA)	1018 (NA)
	Apr-02	15.7 (NA)	19.2 (NA)	6.6 (NA)	3.4 (NA)	4.5 (NA)	133 (NA)	1016 (NA)
	May-02	19.2 (22.7)	22.7 (26.7)	6.4 (8.3)	2.9 (2.7)	4.7 (11.5)	195 (0.0)	1012 (1009)
	Jun-02	22.4 (23.0)	26.0 (27.4)	6.7 (8.2)	2.9 (2.6)	6.6 (8.1)	96 (0.0)	1008 (1008)
	Jul-02	25.4 (NA)	28.5 (NA)	5.3 (NA)	3.2 (NA)	3.7 (NA)	369 (NA)	1005 (NA)
	Aug-02	26.7 (26.1)	29.5 (29.9)	5.0 (7.7)	3.3 (2.4)	5.0 (10.7)	421 (0.0)	1008 (1011)
	Sep-02	24.0 (23.7)	27.6 (27.6)	6.6 (7.7)	3.3 (2.9)	6.5 (8.2)	133 (24.5)	1014 (1015)
	Oct-02	19.2 (21.6)	23.0 (25.6)	7.2 (7.7)	3.3 (3.1)	6.7 (7.4)	183 (117)	1017 (1011)
	Nov-02	12.3 (16.3)	16.3 (21.0)	7.5 (8.5)	3.2 (2.2)	5.9 (9.4)	28 (0.0)	1022 (1027)
	Dec-02	10.3 (NA)	13.9 (NA)	6.5 (NA)	3.3 (NA)	4.6 (NA)	98 (NA)	1024 (NA)

[1)] : Values in parenthesis are the high ozone event day averages.
NA: no high ozone event was observed in that month.

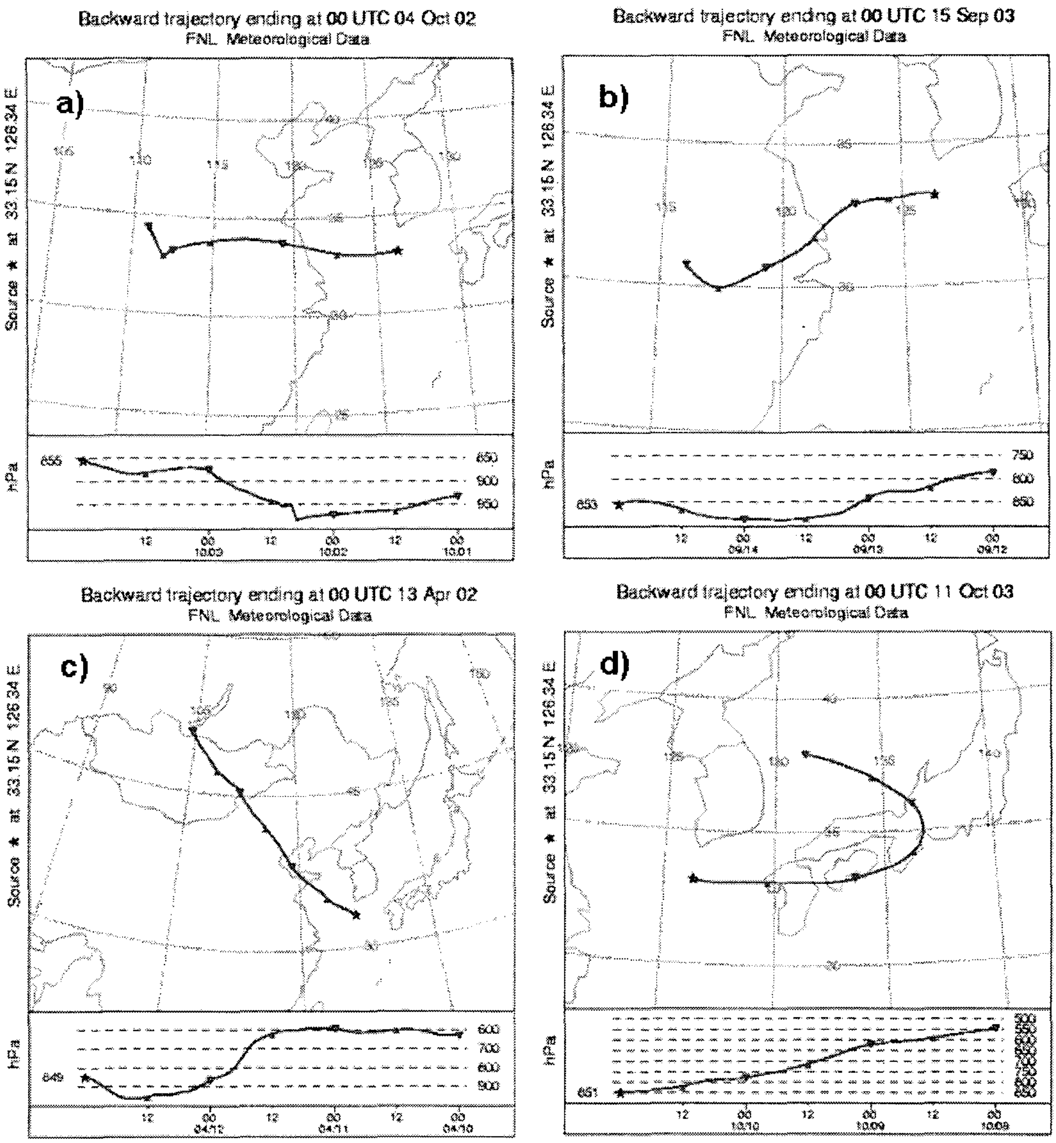

Fig. 6. Patterns of backward trajectories arriving at Jeju Island. The dates and arrival times are given in the upper right corners. Symbols(▾) are shown at 12h intervals.

Island, the average ozone level appears to be lower in Jeju City than in Seogwipo City. It can be also shown that the diurnal variation of ozone concentration is the reverse to the daily NOx and CO cycles and concentration of NO_2 is higher at Ido station than at other three stations.

Additional studies will be needed for the better understanding of the sources of emissions and also the sub-synoptic meterological transport processes in the study region.

Acknowledgements

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