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Boundary Layer Ozone Transport from Eastern China to Southern Japan: Pollution Episodes Observed during Monsoon Onset in 2004

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ABSTRACT

The trajectory analysis of boundary layer ozone data at four regional sites in the East Asian outflow regions in Japan was carried out together with boundary layer ozone data observed at Mt. Tai and Mt. Huang in the source region of central eastern China during the monsoon onset in May-June 2003 and 2004. At all sites, the influences of anthropogenic emissions from East Asia have been found. During May and June 2004, the evidences of direct pollution transport from central eastern China to Hedo, an outflow site in Okinawa Island were observed. Ozone mixing ratios associated with air masses from central eastern China averaged 45 ppb while those associated with clean air masses from the Pacific were only 14 ppb, which resulted in averaged 31 ppb increase of ozone mixing ratios during the pollution episodes from central eastern China at Cape Hedo. Using transport time analysis and averaging all ozone episodes transported from central eastern China, the ozone dilution rate of 5.4 ppb per day was roughly estimated during air masses transported from source to outflow regions at Hedo. In the regions nearby Japanese mainland, however ozone increases by long-range transports were more related to both domestic and East Asian sources as a whole.

Key words: Anthropogenic emission, Central eastern China, Long-range transport, Ozone, Regional pollution

1. INTRODUCTION

We may criticize that human activities polluted the atmosphere leading us to both air pollution problems and the present situation of climate change and global warming. Air pollution and climate change are directly related. Both are resulted from emissions of the specific kinds of gases though with different time frame. The long lifetimes of greenhouse gases allow them to transport around the world and result in the global effect. Meanwhile, other air pollutions usually have shorter lifetimes and their impacts are often limited to local or regional scale. However, many studies showed that air pollution can affect a wide range of areas by chemical transformation and by long-range transport depending on climatology at that time. The distances that air pollution transports could be used to explain the air pollution scales. Local scale air pollutions are emitted and affect the local areas, normally 10-50 km distance. Regional scale air pollution could be transported in the ranges of hundreds to thousands kilometers. Hemispherical scale air pollution may be transported from continent to continent in the scale of several thousand kilometers. Transboundary air pollution transports which are the focus of this work, are mainly considered the regional scale and hemispherical scale air pollution.

East Asia consists of two regions, northeast and southeast. Northeast Asia includes China and Taiwan, Japan, North Korea, South Korea, Mongolia and Eastern Siberia. This region is sometimes simply referred to as East Asia. Apart from Japan and South Korea which has been highly industrialized, some of northeast Asia countries, especially China, are heading towards being an industrialized country at very fast pace. Many industrialized countries move their manufacture units and productions to Asia such as China, India and Southeast Asia. This leads to a change of pollution source regions from Europe and America to Asia. Air pollution problems arise as a result of rapid industrialization and urbanization. Most of cities with largest population are now in Asia. The study by Akimoto (2003) showed the increasing trend of NO_x in three largest sources of the world. It is noticed that NO_x in Asia is continuously increasing in contrast to North America and Europe which are more stabilized. This matches with Ohara and Sakata (2003) and Ohara *et al.* (2007) works which concluded that nowadays Asia turned to be the highest anthropogenic NO_x source of the planet.

The large amounts of air pollution emission can be further transported to another region. Significant factors which helped air pollution to be transboundary transported in and out of the region are climate and meteorology, in particular the monsoon which plays significant role in East Asia (Pochanart *et al.*, 1999). In mid-latitude normally between June and August, a summer monsoon brings air masses from the Pacific Ocean to the continent while in cold season of the year, a winter monsoon brings air masses from Siberia to the Pacific ocean and Southeast Asia (Pochanart *et al.*, 1999; Ahrens, 1994). Several studies, both model and observation have shown the transboundary transport evidences as a results of monsoon and climatology (Pochanart *et al.*, 2004, 2002, 1999; Yamaji *et al.*, 2008, 2006).

Nowadays, Asia is considered to be main region of environmental problems as a result of a rapid economic transformation. Asia is becoming an air pollution exporter, to both countries within and the other continents. This study will focus on how to identify the evidence of long-range transport of ozone from one country to another country using a real simultaneously measurement data from both source and receptor regions which up to now has been confirmed mainly bases on modeling studies. In this work ozone data in Japan and central eastern China were compared and studied. The former is located in the outflow path of the latter which is considered largest source region in East Asia. Ozone data at several sites in Japan were screened and analyzed together with ozone data from observations at mountain sites in central eastern China during the late spring and early summer seasons of 2003 and 2004 looking for evidences of direct transport from central eastern China to Japan. The boundary layer ozone transport characteristics and the relationship of ozone according to its transport between outflow and source regions using air mass trajectories analysis were explained, then the episodic ozone pollution export from China to Japan were identified and quantified.

2. OBSERVATION AND METHODOLOGY

Ozone data in Japan were obtained from four monitoring sites (Happo, Oki, Tsushima, and Hedo; see Fig. 1) representing the regional data in the outflow region of East Asia. These data were obtained from the Acid Deposition Monitoring Network in East Asia (EANET). However, the main focus is mostly at data from Cape Hedo as the station is most relevant to the ideal outflow condition due to the minimized effects of emissions from Japan mainland and Korea. Previous studies at most of these sites confirmed the regional representative of the sites and more details have been previously explained and could be found elsewhere (Pochanart *et al.*, 2004, 2003, 2002, 1999). In Fig. 1, Mondy observatory in Siberia, though not used in this analysis, is also shown as a representative of Eurasian background or clean air masses in the inflow region of East Asia before the perturbation by source region in China (Pochanart *et al.*, 2003).

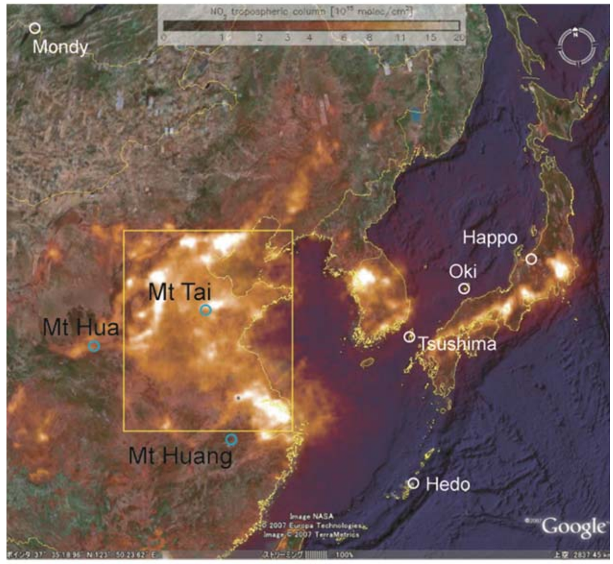

Fig. 1. Location of the observation sites and the OMI NO_2 data in June 2004. Map is from Google Earth. OMI NO_2 data from Tropospheric Emission Monitoring Internet Service (www.temis.nl).

Ozone data in China were obtained from the observatory sites that were established in 2003. The ozone and carbon monoxide monitoring at three sites in China have been carried out, and later intensive monitoring campaign at some of these sites were set up (Kanaya *et al.*, 2013). Mt. Tai, Mt. Huang, and Mt. Hua are located in central eastern China (CEC). These mountain sites are located higher on the top of the boundary layer and are less susceptible to the local emissions than surface sites. Each site is separated by the distance of about 1000 km and is considered the representative for regional scale study. These three sites are located on the mountain summits and are accessible only by cable car and foot walk. The measurements were made on a temporally basis in 2003 and followed by a full year continuous measurement in 2004. These data are among the first available ozone data set monitored at the regional mountain sites above boundary layer in China. Prior to 2004 ozone studies in China were often from lower elevation sites (with the exception of Mt. Waliguan in western China) and details could be referred elsewhere (for e.g. Xu *et al.*, 2008; Wang *et al.*, 2006, 2001; Zhu *et al.*, 2004; Chueng and Wang, 2001; Aunan *et al.*, 2000; Luo *et al.*, 2000; Chameides *et al.*, 1999).

As observed from satellite sensors such as SCIAMACHY, GOME and OMI (also shown in Fig. 1 for June 2004 OMI data) (Buchwitz *et al.*, 2007; Uno *et al.*, 2007), these sites are located in the high emission areas of China. The sites also represent the interface between urban and rural regions in China. For example, Shandong province (where Mt. Tai is located) together with its surrounding provinces have more than 400 million population. In this work, despite data available from three sites, only data from Mt. Tai and Mt. Huang were found to have direct correlation to the ozone data in Japan. Data from Mt. Hua did not show any direct correlation with ozone data in Japan. Apart from results in this paper, data from these sites, either campaign base or seasonal/annual base have also been reported (Kanaya *et al.*, 2013).

The instrumental systems and monitoring criteria for ozone in this study are similar to those of our previous studies and details regarding ozone measurement, calibration, regular maintenance and data handling have been reported elsewhere (Tanimoto *et al.*, 2007; Pochanart *et al.*, 2003, 1999). The isentropic backward trajectories were applied in order to look for the evidence of direct episodic transboundary ozone transport events from a region nearby Chinese sites in central eastern China to Japanese site. Details on trajectory have been reported previously (Pochanart *et al.*, 2003, 1999).

3. RESULTS AND DISCUSSION

3.1 Export of Ozone from Source Regions to the Outflow Regions

The export events of ozone pollution from the source regions in central eastern China to the outflow region in Japan have been investigated using ozone database obtained in both regions. The focus is on ozone data and air masses transport during the ozone peaks period in May-June which is also the same period of summer monsoon onset. The clear evidences of ozone outflow were likely observed during this period because there were continental and oceanic air masses exchanges. In winter and early-spring when the influences of Siberian outflow are strongest, because of the less photochemical activities in the air masses the differentiation of "source regions" outflow events and "non-source regions" outflow events in the boundary layer could not give a very clear picture of ozone pollution episodes. The monthly averages ozone between the two transport events could be different (Pochanart *et al.*, 2004, 1999) but the pollution episodes would not be clearly identified. In contrast, the summer months are also not favorable conditions as the main inflows are summer monsoon from the Pacific Ocean. Without strong perturbation by continental air, the observed low ozone associated with clean marine air masses during summer would make it difficult to identify the pollution episodes.

Based on the available data in 2003 and 2004 backwards trajectories and ozone data during May-June were then investigated at Happo (2003, 2004), Oki Islands (2004), Tsushima Island (2003) and Cape Hedo, Okinawa Island (2003, 2004) (see Fig. 1). The air masse transports at these sites were checked for the outflow events directly from the high emission regions in Central eastern China (the polluted domain in Fig. 1), Japan, and East Asia as a whole. As mentioned above, while this period is the time when ozone mixing ratios in the source regions are highest, it is also the same period when the summer monsoon starts to influences the East Asia. During the summer monsoon onset, transport patterns of air masses are more complicate. Air masses switching between different origins, mainly continental and oceanic, are often observed, and provide more opportunity to identify ozone in different air mass categories in details.

At all observatories, the ozone pollution transport episodes have been observed. Most of the episodes are associated with transport from East Asia, including China, Korea, and Japan. The results in Table 1 show the source regions that influence the Japanese sites.

Table 1. Evidences of ozone pollution export to Japan from East Asia/central eastern China based on trajectory analysis.

Year	Monitoring site	Significant sources of ozone pollution		
		Whole East Asia	Japan	Central Eastern China
2003	Happo	Yes	Yes	No
	Tsushima	Yes	Yes	No
	Hedo	Yes	Yes	No
2004	Happo	Yes	Yes	No
	Oki	Yes	Yes	No
	Hedo	Yes	Yes	Yes

The main influences that affect all the sites are from Japan itself and East Asia as a whole. However, the clear evidence of ozone transport from central eastern China to the outflow region in Japan was found at Cape Hedo, Okinawa Island in southern Japan in 2004.

At Happo (central western Japan), Oki Islands (western Japan), and Tsushima Island (between Japan and Korea) the transport events from the source regions of central eastern China to the sites were observed. However, most of them are also associated with the highly anthropogenic emissions from Korea and Japan and should be regarded as the East Asian pollution as a whole. At Happo in May-June 2003 and 2004, trajectories that passed the sources regions in East Asia during the last five days before arrival at site were about 19% and 25%, respectively. Only less than 3% of air masses are transported directly from central eastern China with the least influences from Korea and Japan. Similar to Happo, while the air masses that passed polluted regions of East Asia were 17% in May-June 2004 at Oki, only about 4% of the total numbers of air masses show the direct transport from Central eastern China. At Tsushima which is an island located between Kyushuu Island of Japan and Korea in May-June 2003, 11% of transports from East Asia have been observed. No evidences of transports that were solely from central eastern China were found. Indeed, most of the polluted air mass transport patterns at Tsushima were either or both influenced by Korea and/or Japan rather than China. Ozone mixing ratios associated with the "regionally polluted" air masses categories are generally higher than the "non-polluted" categories. This is, however, nothing new and has been proved previously (Tsusumi *et al.*, 2006; Pochanart *et al.*, 2004, 2001, 1999). As mentioned, the increased ozone would be considered the results from the East Asian emissions. The distinct episodes of ozone pollution from China do not stand out from these data, at least from this study. To identify the influence from central eastern China on the ozone variations in Japan mainland based only on the observation data would be extremely difficult task or almost improbable, although recently the newest studies pointed out the importance of air pollution studies in China and reveal a lot of significant finding (for e.g. Kanaya *et al.*, 2013, 2009, 2008; Pan *et al.*, 2013, 2011; Li *et al.*, 2011; Suthawaree *et al.*, 2010; Yamaji *et al.*, 2010).

3.2 Ozone Export from Central Eastern China to Cape Hedo, Okinawa Island during Ozone Peak Period in 2004

In comparison with other observatories, Hedo is more favorable because it is located at the lower latitude, more isolated from sources in Korea and Japan mainland, and in the proper location for the northwesterly outflows from central eastern China. During May-June in 2003 and 2004, multiple ozone pollution episodes have been observed at Hedo at both years. Ozone episode here is defined by the significant increase of ozone where the peak mixing ratios exceeded 50 ppb and the event lasts longer than one day.

Even with more favorable location, in May-June 2003, most of the ozone episodes, five out of six, observed at Hedo were resulted from the pollution transported from Korea and Japan rather than China. About 32% of the air masses during this period were directly transported over Korea and Japan without passing any sources regions in central eastern China. Only 7% were found directly transported from central eastern China.

In contrast, the ozone climatology during May-June 2004 is quite unique compared to the others. About 21% of the transports were directly from central eastern China, apparently without the direct influences from Korea and Japan. Only 7% were transported from Korea and Japan without passing emission region in central eastern China. As shown in Fig. 2, seven ozone pollution episodes have been observed. Four of these episodes are the results directly from central eastern China transports. One is mainly from Korea and Japan. One is considered local ozone pollution and one is unidentified.

Although all data and analysis in this work concerns only observation aspect, it is noteworthy here that the ozone mixing ratios observed in our measurement in East Asia agree very well with the results from model studies, in particular regional chemical transport models. The recent publication of these model studies, i.e. CMAQ or NAQPMS which refers to our observation data show good reproducibility of the observation results. More details could be found in Yamaji *et al.* (2008, 2006), Li *et al.* (2007) and Wang *et al.* (2006) . With the observation data, CMAQ model simulation of ozone at Hedo is also depicted for the purpose of comparison. Most of them show good agreements with the observation except the episode 4 (local) and 5 (unidentified) when no significant changes are found in the model results.

During all events, ozone mixing ratios clearly increased. As the results shown in Table 2, the ozone mixing ratios average of seven episodes is 43.2 ± 10.0 ppb while the non-episodes ozone mixing ratios average is 14.2 ± 7.9 ppb. The four episodes from central eastern China show the highest ozone mixing ratios, averaged 45.0 ± 9.8 ppb. About 31 ppb of ozone increase has been observed as a result of ozone transport from central eastern China. This evidence illustrates the significant increase of ozone mixing ratios at Japan-

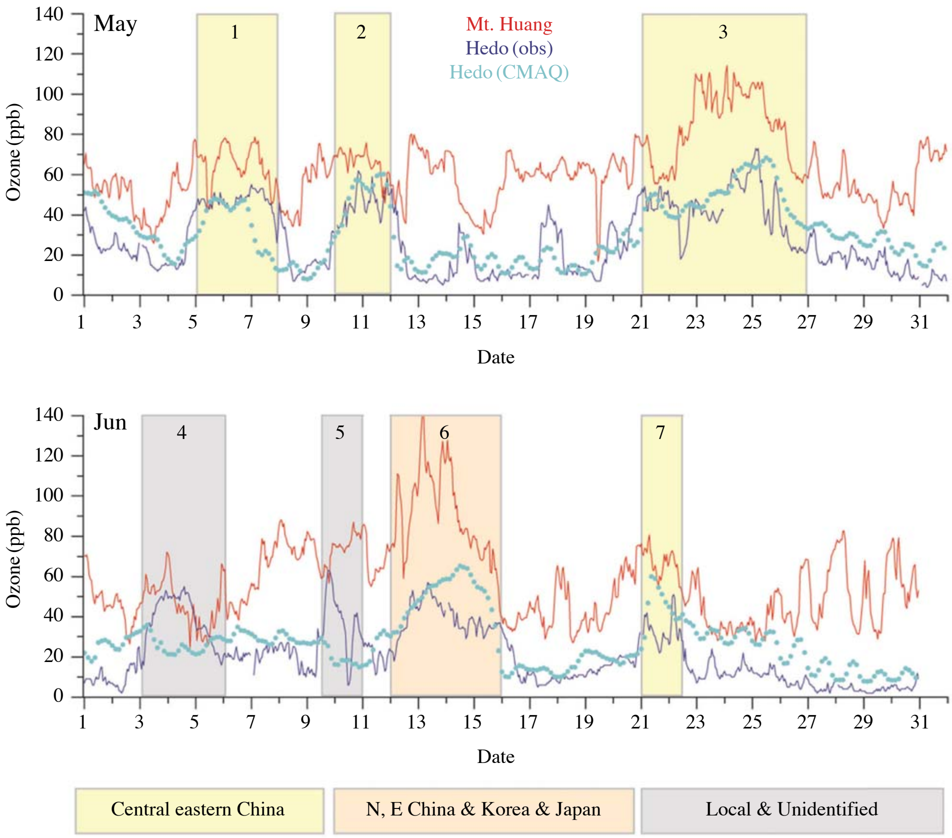

Fig. 2. Ozone pollution episodes observed at Hedo, Okinawa Island during May-June 2004. Hedo ozone data are shown in blue color. For comparison, Mt. Huang ozone data are shown in red color. The yellow, orange, and grey bars indicated the ozone episodes from central eastern China, from Korea and Japan, and from local and unidentified sources. CMAQ results courtesy of I. Uno (Kyushu U.) and K. Yamaji (FRCGC/JAMSTEC).

Table 2. Ozone Mixing Ratios During the Pollution Transport Episodes at Cape Hedo in May-June 2004.

Episode	Date	Origin	Ozone mixing ratios (ppb)	Estimation of ozone dilution during transport			
				Averaged transport time (hr)	Ozone in CEC before transport (ppb)	Difference in both regions (ppb)	Dilution rate (ppb/day)
1	May 5-7	CEC (Huang)	47.5±3.6	57	46.3±12.0	−1.2	−0.5
2	May 10-11	CEC (Huang	47.6±6.7	39	65.8±5.6	18.2	11.2
3a	May 21-26 (1st half)	CEC (Huang)	42.2±8.2	114	61.7±3.0	19.5	4.1
3b	May 21-26 (2nd half)	CEC (Tai)	51.4±12.6	92	62.2±9.6	10.8	2.8
4	June 3-5	Local	42.9±8.4				
5	June 9-10	Unidentified	38.2±15.0				
6	June 12-15	Korea/Japan	40.0±7.6				
7	June 21-22	CEC (Both/M)	32.9±7.3	44	62.2±8.7	29.3	16.0
1-3 and 7		CEC	45.0±9.8	74	61.6±14.2	16.6	5.4
All		All	43.2±10.0				
Non-episode		Marine	14.2±7.9				

CEC refers to central eastern China. Episode 7 is transported from region around both Mt. Huang and Mt. Tai in central eastern China but was considered marine origin. Mt. Tai data was used as origin for this episode. If Mt. Huang was used, it would result in 59.4±12.8 ppb of ozone at the source regions.

ese outflow region by direct transport from central eastern China using surface observation. At Yonaguni-jima, an outflow site at lower latitude and much closer to Taiwan and Chinese mainland, about 15-20 ppb

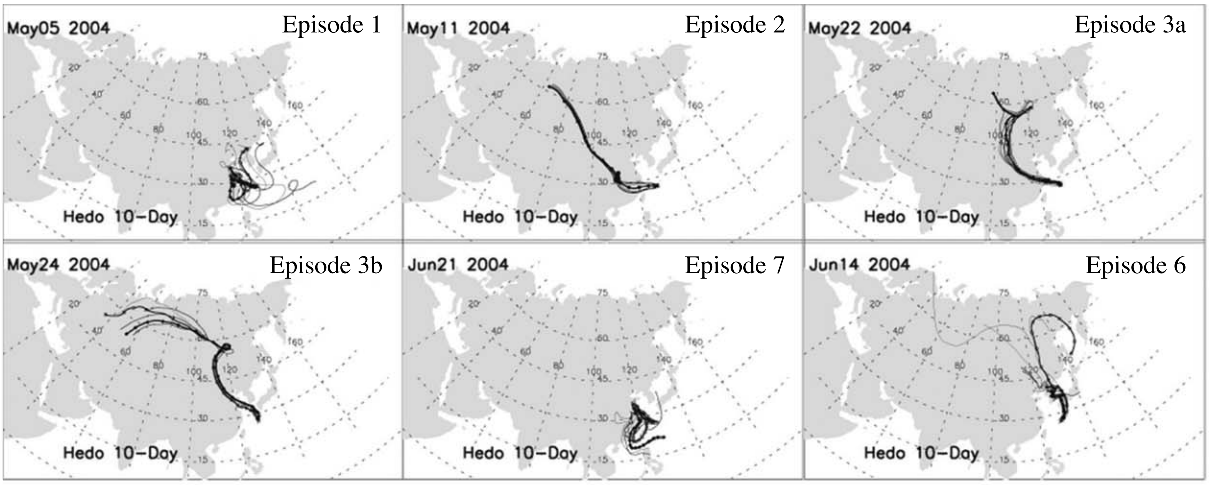

Fig. 3. Evidences of direct pollution transport from source regions in central eastern China to outflow regions at Cape Hedo, Okinawa Island from backward trajectories. Episodes 1, 2, 3a, 3b, 7 are directly from central eastern China. Episode 6, also shown for comparison, is from Korea and Japan.

difference of ozone between "regional events" and "background events" have regularly been observed in summer (Tsutsumi *et al.*, 2006).

It is also noticed that the four ozone episodes from central eastern China were also transported from the regions around Mt. Huang (episode 1, 2, first half of episode 3, and 6) and Mt. Tai (Second half of episode 3). The examples of the trajectories transport pattern are depicted in Fig. 3. Thus, this opportunity could be used to further investigate and compare the relationship between ozone in the source region and the outflow regions using observation data. The following test must be considered as only case study and could not be applied in other conditions. The four central eastern China ozone episodes consists about 20% of the total air masses arriving at site in May-June, in which 4% were of marine origin. From trajectories, the transport times from the source region in central eastern China (using 120E as the boundary) to Hedo were identified for each episode.

Then the ozone changes during transport between the two regions were estimated using the ozone mixing ratios at Hedo, transport time from central eastern China to Hedo, and ozone mixing ratios at Mt. Huang/Mt. Tai (depending on transport routes) at the times air masses originally leaved central eastern China for each episode. The results are also shown in Table 2. It is found that transport times from central eastern China to Hedo varied among episodes, 39-114 hours, and averaged 74 hours or about 3 days. Meanwhile, the averaged ozone mixing ratios in central eastern China before outflow air masses were transported to Hedo are 61.6 ± 14.2 ppb, about 17 ppb higher than the average of 45.0 ± 9.8 ppb when the ozone pollution episodes reached Hedo. Except the first ozone episode in May 5-7 which shows no significant change in ozone at the source and the outflow regions, the others show significant decrease of ozone during transport from sources to the outflow regions. Episode 2 and 3 show the ozone decrease of 10.8-18.2 ppb during transport. The maximum decrease was observed in episode 7 when nearly 30 ppb of ozone decreases during transport. This episode is somewhat different from episode 1-3 because the air masses were of marine origin, then transported through regions nearby both Mt. Huang and Mt. Tai before circulated back to Hedo. The strong monsoon influence in late-June would also result in lower ozone at Hedo. Averaging all ozone episodes transported from central eastern China, the ozone dilution rate of 5.4 ppb per day was roughly estimated during air masses transport from source to outflow regions at Hedo. The uncertainties in this study are large mainly due to the assumption that data from Mt. Huang and Mt. Tai could be used as proxy for ozone in central eastern China. One must understand that pinpointing the exact numerical values using only observation data and this type of analysis is improbable, which is not the purpose of this study. Nonetheless, the evidences of direct ozone pollution transport from China to Okinawa Island were confirmed and during such transport ozone mixing ratios significantly decrease with the rate of about 5 ppb per day have been approximated.

The above case study pointed out that while recently it becomes serious concern whether the Chinese air pollution would affect air quality in Japan, the potential influence would occur mainly as the increases of background ozone mixing ratios in Japan as previously reported (Pochanart *et al*., 2004, 1999). In most circumstances, the sources of such increases must include Korea and domestic pollution in Japan as well. Only during the favorable and specific meteorological condition that the ozone episodes as a result of direct transport from China would be observed in Japan mainland. In the climatology aspect and from previous works, the northwesterly flows from Siberia dominate Japan from fall to spring and maritime air masses from the Pacific and partly from the Indian Ocean in summer. Thus, statistically Japanese mainland tends to get more air pollution from Korea and from the Japanese mainland itself rather than from the source regions in China.

4. CONCLUSIONS

This work presents the ozone data analysis from simultaneous observation in eastern China and southern Japan and identifies the evidences of direct transboundary air pollution transport from China to Japan based on data in late spring and early summer of 2003 and 2004. At that time, the issue was rather sensitive. A decade passed by, China is now more acknowledged its own environmental issues and the Chinese authorities are trying hard to solve the environmental problems. New data from many studies have proved the potential air pollution export from East Asia including China to other regions. This finding would reinforce the importance and urgency to reduce the East Asian air pollution emissions.

Using data both in the source regions and outflow regions, the evidences of direct ozone transport from central eastern China to Cape Hedo, Okinawa Island of Japan were found. In May-June of 2004, four ozone episodes from central eastern China resulted in an increase of about 31 ppb of ozone at Okinawa. It was estimated that ozone on averaged decreased with a rate of 5 ppb/day during transports from central eastern China to Okinawa. Meanwhile, based on the observation results at other sites, background ozone variations in Japan mainland are more likely influenced by the ozone from domestic production, Korea, or East Asia as a whole.

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