Research Article

Analysis of Long-Range Transport of Carbon Dioxide and Its High Concentration Events over East Asian Region Using GOSAT Data and GEOS-Chem Modeling

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Received 30 January 2015; Revised 19 May 2015; Accepted 8 June 2015

Academic Editor: Ugo Cortesi

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This study aims to evaluate the long-range transport of CO₂ in East Asian region, using concentration data in a surface measurement site (Gosan Station), column averaged concentration data of satellite-borne instrument (GOSAT), and GEOS-Chem modeling results for the period of June 2009 to May 2011. We perform a validation of the data from GOSAT and GEOS-Chem with total column observations (TCCON). The analysis of the long-range transport and high concentration (HC) events using surface/satellite observations and modeling results is conducted. During the HC events, the concentrations in CO₂ and other air pollutants such as SO₂ and CO are higher than that of all episodes. It means that CO₂, known as a globally well-mixed gas, may also act as a fingerprint of human activity with unique regional characteristics like other air pollutants. This comprehensive analysis, in particular with GOSAT CO₂ observation data, shows that CO₂ plume with high concentration can be long-range transported with 1-2 days' duration with regional scale. We can find out with GEOS-Chem tagging simulation that more than 45% of the elevated CO₂ concentration over central/eastern China, Korea, and Japan on high concentration days can be explained by emission sources of East Asia mainland.

1. Introduction

Since the industrial revolution, the current level of CO₂ emitted into the atmosphere has increased by nearly 41% because of fossil fuel combustion. This contributes to a higher global warming potential (approximately 82%) as compared to other greenhouse gases, such as CH₄ and N₂O [1]. In general, the global CO₂ concentration is 2.0–2.5 ppmv higher in the northern hemisphere than in the southern hemisphere because of the greater dependence on fossil fuel combustion [2]. In particular, among the countries in the northern hemisphere, the ground-based CO₂ concentration in Asia for year 2007 is relatively high (387.6 ppmv), as compared to Europe (386.3 ppmv) and North America (385.1 ppmv) because of the rapid industrialization [3]. According to the International Energy Agency report [4], the 2007 CO₂ emissions by regions

in Asia, such as China, India, Korea, and Iran, have increased by over 100%, compared to the 1990 level.

The atmospheric CO₂ concentration is determined by the average concentration and its short-term variation mainly due to the long-range transport and meteorological conditions [5]. It is reported that an increase in CO₂ concentration in the Arctic region is affected by the impact of long-range transport of CO₂ emissions from the midlatitude northern hemisphere [6–12]. In case of Asian regions, some studies [3, 13, 14] showed that long-range transport of CO₂ from the Asian continent contributed to the variations of CO₂ concentration in Japan and Korea, using surface measurement data. In addition, using airborne and model data, Shirai et al. [15] mentioned that the CO₂ concentration in the free troposphere was significantly affected by the long-range transport of CO₂.

FIGURE 1: The location of Gosan site, Jeju Island in Korea.

In order to investigate the impact of global/regional carbon cycle on the regional change of CO_2 concentration and air quality [16], it is important to understand the comprehensive feature of long-range transport events utilizing the observation data on the surface level and vertically integrated column data and modeling results, simultaneously. However, the above-mentioned studies were carried out with temporally and spatially limited measurement data. For example, ground-based observations have much higher accuracy rather than the modeling approach and the satellite data, while their spatial resolution is very coarse. As for the modeling approach, there are no restrictions on spatial and temporal coverage, while it has lower confidence rather than observation data due to uncertainties in emission inventory, meteorological fields, and its initial and boundary conditions. On the other hand, the satellite data have less restriction on its spatial and temporal resolution rather than surface observations, and those are more reliable than modeling results; however, satellite data has still observational errors which are due to radiometric/geometric/atmospheric correction, surface reflection and cloud masking, and the smearing effect in high peak concentrations due to much less spatial resolution compared to that of modeling approach. Therefore, it is necessary to integrate the ground-based observations, the satellite data, and the modeling results in order to compensate possible drawbacks of the measurement and the modeling and utilize their advantages.

Although CO_2 is a globally well-mixed gas in a climatological sense, it has also the unique regional characteristics over the source and downwind area because of the same anthropogenic emission sources driven by human socioeconomic activity like other air pollutants (e.g., CO , SO_2 , and aerosols), and few studies for long-range transport of CO_2 and its high concentration events have been conducted at East Asia where it is one of the most hot spots of CO_2 emission in the world.

In this study, we investigate CO_2 long-range transport phenomena and high concentration events on the downwind regions of the Asian continent and provide observational evidences showing regionally the same behavior of CO_2 as

other air pollutants over East Asia, using all the available data, CO_2 and other air pollutants concentration data observed at surface monitoring sites, satellite data, and results of a chemical transport model.

2. Data and Methodology

This study focused on the CO_2 concentration data available for the period of June 2009 to May 2011, which was observed by the Greenhouse gases Observing SATellite (GOSAT) and surface monitoring stations. Surface observation data were collected from the Gosan site in Korea (<http://ds.data.jma.go.jp/gmd/wdcgg/cgi-bin/wdcgg/accessdata.cgi?index=GSN233N01-GERC>), which is located on the eastern side of the Asian continent. Satellite data, column-averaged CO_2 concentrations, were utilized along with GOSAT dataset L2 V02.xx (http://www.gosat.nies.go.jp/index_e.html). The Goddard Earth Observing System with Chemistry Model (GEOS-Chem, v9.1.2), a global chemical air quality transport model, was used for simulating high concentration events and their source/sink apportionment. In addition, we also evaluated satellite data and modeling results with CO_2 concentration data from the World Data Centre for Greenhouse Gases (WDCGG; <http://ds.data.jma.go.jp/gmd/wdcgg/>) and column-averaged concentration data from the Total Column Observing Network (TCCON; <http://www.tccon.caltech.edu/>). Also, the Hybrid Single Particle Lagrangian Integrated Trajectory Model (HYSPLIT) [17] was used for a trajectory analysis.

2.1. Ground-Based Observation Data. Hourly averaged CO_2 concentration and other air pollutants (SO_2 , NO_2 , CO , O_3 , PM_{10} , and $\text{PM}_{2.5}$) concentration data have been measured at Gosan Station in Korea. Gosan Station is located at the west end of Jeju Island, Korea ($33^\circ 17' \text{N}$, $126^\circ 10' \text{E}$) (Figure 1), in the middle among the Korean Peninsula, China, and Japan, so that it is well suited for the research on long-range transport of greenhouse gases and other air pollutants in East Asia. In this area, PEM-WEST A and B, ACE-Asia, and ABC's

International Joint Observational Campaign had been conducted [18–21].

The CO₂ sampling from a 10 m tower is automatically analyzed every 30 seconds, through a Nondispersive Infrared Analyzer (NDIR). All CO₂ data are reported using the WMO CO₂ mole fraction scale. We perform a statistical analysis after QA/QC (Quality Assurance/Quality Control) processes to determine whether it is a clean or a polluted atmosphere [22]. We followed Advanced Global Atmospheric Gases Experiment (AGAGE) statistical pollution identification procedure by O'Doherty et al. [22] to select clean atmosphere cases from Gosan observation data by removing polluted cases affected by local and regional sources. A polluted atmosphere is then divided as either a local atmosphere or a regional atmosphere by using the criterion according to wind direction and speed shown in Figure 1. For excluding data in a local atmosphere, the data in which wind direction is 22.5°–112.5° and the wind speed is less than 11 m/s were discarded [23]. The local atmosphere refers to areas affected by the urban scale emission sources, while the regional atmosphere reflects the effect of the East Asian regions, in which the effect of a local atmosphere could be minimized. In this study, CO₂ concentration data collected only in clean atmosphere and regional atmosphere were used to assess the effect of long-range transport.

Also, in order to make direct comparison of GOSAT data and GEOS-Chem model results with surface measurement data, we used TCCON's column-averaged concentration. TCCON is a network of ground-based Fourier transform spectrometers that measure column-averaged concentrations of CO₂, CO, CH₄, and H₂O in the atmosphere. For this study, the TCCON data were selected at each site, within about ±1 hour of the GOSAT overpass time. Also, the GOSAT and GEOS-Chem data were selected within about ±1 degree of the area centered at each TCCON site.

2.2. Satellite Data. GOSAT is a sun-synchronous satellite, which provides a global coverage in three days and crosses the equator at about 13:00 local time. Its swath width is 790 km and its spatial resolution at nadir is 10.5 km. GOSAT's wavelength ranges between 0.76 and 14.3 μm, consisting of two sensors—one is the Thermal and Near-Infrared Sensor for Carbon Observation-Fourier Transform Spectrometer (TANSO-FTS), which measures CO₂ and CH₄, and the second is the Thermal and Near-Infrared Sensor for Carbon Observation-Cloud and Aerosol Imager (TANSO-CAI), which measures clouds and aerosols. TANSO-FTS is composed of four bands. Bands 1 to 3 are Shortwave Infrared (SWIR) bands, which provide the total column amount of CO₂ and CH₄, and Band 4 is a Thermal Infrared (TIR) band, which produces a profile of CO₂ and CH₄ concentration at 1000, 700, 500, 300, 100, 50, and 10 hPa [24]. It is well known that the SWIR absorption bands near 1.6 μm and 2.0 μm provide better information on the near-surface concentrations. On the other hand, the TIR absorption band around 14 μm is used to obtain information on the profiles of CO₂ and CH₄, mainly above 2 km altitudes, so TIR data are sensitive to the middle and upper troposphere [25, 26]. With this reason, we use SWIR XCO₂ data, column-averaged concentrations,

in order to compare directly to the surface observation data of CO₂ in this study.

2.3. Modeling Data. We have used version 9-02-01 of GEOS-Chem, a global 3D atmospheric chemistry transport model developed by the National Aeronautics and Space Administration (NASA) and Harvard University. It is driven by assimilated meteorology from the NASA Global Modeling Assimilation Office (GMAO) and contains approximately 300 photochemical reaction mechanisms of the O₃-NO_x-hydrocarbon chemistry [27]. The original GEOS-Chem CO₂ simulation was developed by Suntharalingam et al. [28]. A current major update including the CO₂ source/sink inventory data has been carried out by Nassar et al. [29].

In this study, we used a globally uniform CO₂ field of 375 ppmv as an initial condition, which was measured on January 1, 2004. This simulation is performed for the target period of June 2009 to May 2011 using the GEOS-5 meteorological field with a horizontal resolution of 2° × 2.5° and 47 vertical levels that reach up to approximately 80 km above the surface of the Earth.

The CO₂ concentration (CO_{2*i*}) and air density (A_{*i*}) at each layer were used in the following equation [30] to calculate the column-averaged CO₂ concentrations (XCO₂) of GEOS-Chem [28] for comparison with CO₂ column-averaged mixing ratio of GOSAT defined as the number of gas molecules in a vertical unit column stretching from the ground surface to the top of the atmosphere [25]:

$$XCO_2 = \frac{\sum_{i=1}^{47} (A_i \times CO_{2i})}{\sum_{i=1}^{47} A_i}. \quad (1)$$

In addition, GEOS-Chem can carry out tagged simulations for CO, O_x, and CO₂ to better identify the contributions from various source regions. These contributions were calculated using tagged tracer for each geographical region and source [31, 32]. We estimated the contributions of each region and source/sink to atmospheric CO₂ concentration in East Asian regions by adding CO₂ tracer of different source produced in various geographical regions.

2.4. Selection of Long-Range Transport and High Concentration Events. It is difficult to show the long-range transport phenomena of CO₂ in the regional scale because its lifetime is very long enough to be well-mixed even globally. Moreover timely continuous CO₂ measurements with spatially wide coverage are very restricted so far. So, to distinguish each high concentration event from measurements of long-term period and to analyze the composite field of them are an easy way to identify the long-range transport phenomena.

As mentioned above, Gosan surface measurement site observed both CO₂ and other air pollutants simultaneously and its geographical location is almost a middle zone of Korean Peninsula, China, and Japan. So, this site is one of the best stations for analyzing the characteristics of a long-range transport that occurred in the East Asia regions. The observed CO₂, SO₂, and CO concentrations of Gosan site for the period of June 2009 to May 2011 were used. SO₂ and CO are well-known tracers of long-range transport, which are

FIGURE 2: Comparison of the column-averaged CO₂ concentration (a) between TCCON and GOSAT, (b) between TCCON and GEOS-Chem, and (c) between GOSAT and GEOS-Chem. N and R indicate number and correlation coefficient of matched data, respectively.

mostly emitted by anthropogenic activities and their lifetime (several weeks to months) is long enough to trace air pollutant plumes at the intercontinental scale [33–38].

In addition, high concentration (HC) events were defined as CO₂ concentration at Gosan Station was higher than the mean + 1σ (σ : standard deviation) and the SO₂ or CO concentration was also higher than the mean + 1σ for at least three consecutive hours. The day before HC and the day after HC will be defined as HC – 1 and HC + 1, respectively.

3. Results

3.1. Evaluation of GOSAT Data and Modeling Results. Compared to TCCON's column-averaged concentration, GOSAT concentrations underestimated nearly by 2.4 ± 5.3 ppmv ($0.6 \pm 1.4\%$), while GEOS-Chem overestimated nearly 0.8 ± 1.5 ppmv ($0.2 \pm 0.4\%$) (Figures 2(a) and 2(b)). Morino et al. [25] also reported that GOSAT V01.xx data underestimated approximately 8.57 ± 4.44 ppmv ($2.2 \pm 1.2\%$) as compared

TABLE 1: Daily averaged surface concentrations of CO₂ and air pollutants on HC, HC – 1, and HC + 1 case and their occurrences.

	CO ₂ (ppmv)	SO ₂ (ppbv)	CO (ppmv)	O ₃ (ppbv)	NO ₂ (ppbv)	PM ₁₀ ($\mu\text{g}/\text{m}^3$)	PM _{2.5} ($\mu\text{g}/\text{m}^3$)	Number of days
All	395.9	2.9	0.7	43.5	3.9	49.1	27.0	606
HC – 1	398.3	2.8	1.0	44.9	3.8	48.4	22.8	25
HC	403.1	5.0	1.2	47.7	5.5	68.1	40.3	67
HC + 1	398.0	2.8	1.0	46.9	3.9	55.2	25.5	25

to TCCON due to multiple errors (e.g., solar irradiance database, handling of aerosol scattering). An improved retrieval algorithm (V02.xx) shows much smaller bias and standard deviation (-1.48 ± 2.9 ppmv) than V01.xx [39]. Fraser et al. [40] indicated that the overestimation of observed concentration under GEOS-Chem was largely due to the high anthropogenic emissions. Figure 2(c) shows a comparison result of the GOSAT and the GEOS-Chem, highlighting that GEOS-Chem generally overestimated approximately 2.6 ± 2.5 ppmv ($0.7 \pm 0.7\%$) with the relatively small spatial/temporal variation, overestimation in low concentration, but underestimation in high concentration.

Figure 3 indicates the comparison between the GOSAT SWIR measurement data and GEOS-Chem modeling results for each season. The number of GOSAT data is not enough to make comparison with the modeling results at the northern hemisphere in DJF and North Pacific in all time, because of issues regarding the elevation angle of the Sun and the effect of cloud [41, 42].

Nevertheless, the GOSAT and GEOS-Chem data showed very similar patterns of the seasonal variation and spatial distributions. The seasonal variation for both data is obvious that CO₂ decreases in summer because of photosynthesis by vegetation while it increases in winter because of the increased usage of fossil fuel and decreased vegetation. The amplitude of the CO₂ seasonal cycle is higher in the northern hemisphere than in the southern hemisphere. Also, the spatial distributions represent that CO₂ concentration is higher in the northern hemisphere than in the southern hemisphere because of the higher population and greater number of industrial activities. In addition, the spatial distribution of CO₂ concentration showed the possibility of a long-range transport of CO₂ because relatively high concentration was detected over an ocean region far from the land source. For spring and winter period of northern hemisphere (MAM and DJF), high concentration plume of CO₂ emitted from the Asian continent was expanded down to North Pacific, and for spring period of southern hemisphere (SON), the CO₂ emitted from South America and Africa has been expanded to the Atlantic and the Indian Ocean.

Through correlation analysis and comparison of spatial distribution among the comprehensive dataset, TCCON, GOSAT, and GEOS-Chem, we find that, at least in East Asian region, GOSAT and GEOS-Chem data are comparable to each other in temporal/spatial distribution with well-known differences with TCCON data, so that it might be reasonable to use those data for this study.

3.2. Analysis of the Long-Range Transport and High Concentration Event of CO₂ Using Surface Measurement. Annual averaged CO₂ concentration observed at Gosan Station in 2010 was 398.3 ppmv, which is 13.1 ppmv higher than the global average CO₂ concentration of 389.0 ppmv [43]. For the past nine years (2002–2010), its growth rate was approximately 2.2 ppmv per year, which is faster than the global average growth rate (1.98 ppmv per year).

Table 1 shows the concentrations of CO₂ and air pollutants for the HC, HC – 1, and HC + 1 days defined in Chapter 2.4. During all of the target period (June 2009 to May 2011; 606 days) of this research, the averaged CO₂ concentration observed at Gosan Station is 395.9 ppmv. The ratio of HC days to all periods is 11.1% (67 days) with the mean CO₂ concentration of 403.1 ppmv, which was 7.2 ppmv higher than that of all periods. Also, the concentration of other air pollutants was higher than that of all periods. In particular, the concentrations of SO₂ (5.0 ppbv) and CO (1.2 ppmv) were more than 1.7 times higher than those for all periods. HC was almost 5 ppmv higher than HC – 1 (398.3 ppmv) and HC + 1 (398.0 ppmv). It is obvious that Gosan Station is more directly affected by a polluted plume transported from China on HC days rather than for a day before/after HC days (HC – 1/HC + 1).

Backward trajectory and forward trajectory analyses (Figure 4) were conducted for ± 3 days on HC case, at a height of 1 km above the ground using the HYSPLIT model which was developed by NOAA to compute air trajectories and dispersion of atmospheric tracers [17]. It clearly demonstrated that the air originating from/out of Asian continent traveled through the polluted areas in China and flowed into the Korean Peninsula. Also, the analysis of the forward trajectory clearly showed that the air that once flowed into the Korean Peninsula moved away to the northwestern Pacific through Japan.

Figure 5 shows the monthly occurrence of HC case at Gosan Station. HC occurred for 48 days in winter (20 days in December, 10 days in January, and 18 days in February), for 11 days in spring (4 days in March, 5 days in April, and 2 days in May), for 7 days in autumn (5 days in October and 2 days in November), and for 1 day in summer (1 day in August). It means that long-range transport occurred mostly in winter because the CO₂ gases emitted from fuel combustion in China were affected by the continental high pressure, thus moving into the downwind regions. In summer, because the high pressure in the North Pacific expanded to the Korean Peninsula, they are mainly affected by the oceanic weather

FIGURE 3: Seasonal variation and global distribution of the column-averaged CO₂ concentration (a) measured by GOSAT SWIR and (b) simulated by GEOS-Chem. MAM denotes the period from March to May, JJA from June to August, SON from September to November, and DJF from December to February.

FIGURE 4: (a) Backward trajectories and (b) forward trajectories at 1 km altitude for the cases of high concentration (HC) case.

FIGURE 5: Monthly distribution in number of HC days classified from CO₂ concentration data observed at Gosan Station.

pattern, thus reducing the impact caused by the Asian continent.

3.3. Analysis of the Long-Range Transport and High Concentration Event of CO₂ Using GOSAT Data and GEOS-Chem Results. In this section, further analysis on the long-range transport of high concentration CO₂ was conducted using the GOSAT data and the GEOS-Chem modeling results, in order to expand its spatial coverage.

Figure 6 indicates the spatial distribution of the deviation of the column-averaged CO₂ concentrations for HC to those for all the periods observed by GOSAT within the area of Eastern Asia (10–70°N, 80–170°E). The mean column-averaged concentration (389.4 ppmv) for HC days was 3.6 ppmv higher than the one for all of the periods, showing a similar pattern to the result from the surface observation. As compared to the reported retrieval error (2 ppmv) of GOSAT SWIR measurement [25, 41], the increased concentration is 1.8 times higher than the measurement error. It is interesting that high concentrations are also dominating in central and eastern China and Japan when a surface site, the Gosan Station in Korea, experiences high concentration events. This means that CO₂ plume with high concentration is prevailing over those areas through long-range transport

and mixing processes like other air pollutants. It implies that the regional/national action to reduce CO₂ emission and its adverse impact might be taken with the same way/time as that of air pollutant reduction for the sake of socioeconomic cost, that is, cobenefit strategy.

We focused on 3 subregions (Region I, Region II, and Region III) for providing some observational evidence of long-range transport of high concentration CO₂ over downwind area from Asian continent; Region I (30–60°N, 110–120°E) covers the eastern region of China, and Region II (30–60°N, 120–130°E) covers the Yellow Sea and the Korean Peninsula, while Region III (30–60°N, 130–140°E) represents Japan.

Figure 7(a) shows that the concentrations on HC – 1, HC, and HC + 1 were 389.7 ppmv, 390.4 ppmv, and 390.7 ppmv in Region I, respectively. Their amplitude of variation is less than 1 ppmv, thus maintaining a certain level of concentration because Region I is one of the world most populated areas and major emission sources. However, in Region II, concentrations on HC – 1, HC, and HC + 1 were 389.3 ppmv, 391.5 ppmv, and 388.7 ppmv, respectively (Figure 7(b)). In this case, the concentration on HC is 2.2 ppmv higher than that of HC – 1 and then it dramatically decreases on HC + 1. Unlike Regions I and II, the concentration (392.2 ppmv) on HC + 1 in

FIGURE 6: Difference in CO_2 concentration between HC and all periods of the East Asian region using GOSAT SWIR. Region I ($30\text{--}60^\circ\text{N}$, $110\text{--}120^\circ\text{E}$) covers the eastern region of China, and Region II ($30\text{--}60^\circ\text{N}$, $120\text{--}130^\circ\text{E}$) covers the Yellow Sea and the Korean Peninsula, while Region III ($30\text{--}60^\circ\text{N}$, $130\text{--}140^\circ\text{E}$) represents Japan.

FIGURE 7: Regional mean of the column-averaged CO_2 concentration and its standard deviation on HC - 1, HC, and HC + 1 and for all periods (All) using GOSAT SWIR. (a) Region I ($30\text{--}60^\circ\text{N}$, $110\text{--}120^\circ\text{E}$), (b) Region II ($30\text{--}60^\circ\text{N}$, $120\text{--}130^\circ\text{E}$), and (c) Region III ($30\text{--}60^\circ\text{N}$, $130\text{--}140^\circ\text{E}$).

FIGURE 8: Distribution of the column-averaged CO₂ concentrations for all periods (All) and high concentration cases (HC – 1, HC, and HC + 1) simulated by GEOS-Chem.

Region III was 1.3–1.4 ppmv higher than the ones (390.8 ppmv and 390.9 ppmv) on HC – 1 and HC (Figure 7(c)). In this analysis, we can clearly find out with satellite observations that CO₂ plume with high concentration could be long-range transported with 1–2 days' duration.

Figure 8 shows the simulation results of GEOS-Chem for the same domain and period of GOSAT. Given the distribution of CO₂ concentration in all periods, most inland regions where anthropogenic emission sources are mainly concentrated, including China, Korea, and central Tokyo in Japan, reached more than 392 ppmv of CO₂ concentration. It is also shown that high concentration (almost up to 400 ppmv) plume of CO₂ emitted from China on HC – 1 days moved into the Korean Peninsula passing through the Yellow Sea on HC days and then ran away to the Pacific through the eastern coast of Korea and Japan, a day after the high concentration days (HC + 1). In particular, on HC days, high CO₂ concentrations are shown over marine area near the Korean Peninsula.

In order to clarify the source apportionment on high CO₂ concentration days, we performed the so-called tagging simulation [31, 32] using GEOS-Chem modeling.

Figures 9(a), 9(b), and 9(c) show the difference of the column-averaged CO₂ concentration between HC days and all of target periods from all sources and sinks, East Asia mainland, and South and Central Europe, respectively. Figure 10 indicates the contribution to East Asian regions as being caused by sources and sinks of 40 different regions around the world.

It is found that there are two major contributions to East Asia region for high concentration events from East Asia mainland and South/Central Europe. Over central and eastern China, Korea, and Japan, almost 45% of the contribution to China, Korea, and Japan inland area can be explained by emission sources of East Asia mainland (i.e., central/eastern China) and almost 20% to northern China by European emission sources out of the analysis domain (Figures 9(b), 9(c), and 10). In particular, it is reported from previous meteorological analysis [36] that the dynamic and meteorological mechanism for long-range transport phenomenon of high concentration CO₂ plume supposes that CO₂ emitted from China is lifted to the upper troposphere or tropopause by convection, midlatitude cyclones, and orographic lifting and is thus transported across the Pacific along with the westerlies in the upper atmosphere.

FIGURE 9: (a) Differences of the column-averaged CO₂ concentrations between high concentration cases (HC - 1, HC, and HC + 1) and all periods (All) simulated by GEOS-Chem. (b) and (c) represent differences of column-averaged CO₂ concentration contributed by emission from East Asia mainland and South and Central Europe by tagged simulation, respectively.

4. Conclusion

In this study, the ground-based data, the satellite-borne instrument (GOSAT), and the chemical transport model (GEOS-Chem) are used to investigate the long-range transport phenomena for high CO₂ concentration plume in Asian continent and the downwind regions for the period of June 2009 to May 2011. We also evaluate GOSAT data and GEOS-Chem results with TCCON data to show the reliability of the derived data. Those have very similar patterns of the seasonal variation and spatial distribution and have well-known differences with TCCON data, so that it could be reasonable to use GOSAT and GEOS-Chem data for this study.

The analysis of the long-range transport and high concentration event using observation data in a surface site, the Gosan Station in Korea, shows that the ratio of high concentration days to all periods is 11.1% with CO₂ concentration of

403.1 ppmv and 7.2 ppmv increases compared to the average CO₂ concentration (395.9 ppmv) in all periods. We can find out from backward/forward trajectory analysis that the air originating from/out of Asian continent may be transported through the polluted area in China and flowed into Korea and Japan.

The column-averaged concentration data by GOSAT and GEOS-Chem were analyzed to expand the spatial distribution of the long-range transport of high concentration CO₂ plume. With GOSAT data, concentration (389.4 ppmv) on HC days is 3.6 ppmv higher than that of all periods. It is obvious that high concentrations are also dominating in central and eastern China and Japan as a surface site, the Gosan Station in Korea, experiences high concentration events and CO₂ plume with high concentration is prevailing over those areas through long-range transport and mixing processes like other air pollutants. Also, with subregional analysis, it is found that CO₂

FIGURE 10: The contribution of sources and sinks to atmospheric CO₂ concentration of the East Asian regions simulated by GEOS-Chem tagged simulation.

plume with high concentration can be long-range transported with 1-2 days' duration with regional scale.

With tagging simulation using GEOS-Chem modeling, we can explain that more than 45% of the elevated CO₂ concentration over central and eastern China, Korea, and Japan on HC days may be attributed to emission sources of East Asia mainland, while almost 20% over northern China may be attributed to European emission sources.

In this study, we can understand that CO₂ which is known as a globally well-mixed gas may also act as a fingerprint of human activity with regional characteristics like other air pollutants. This comprehensive analysis with all of available data, although observational evidences are still quite few, would help better understand the long-range transport of CO₂ and its impact on climate change and the carbon cycle in the Asian regions.

Conflict of Interests

The authors declare that there is no conflict of interests regarding the publication of this paper.

Acknowledgments

The authors thank JAXA, NIES, and MOE for the GOSAT data and their continuous support as part of the Joint Research Agreement. TCCON data was obtained from the TCCON Data Archive, operated by the California Institute of Technology from the Website at <http://tccon.ipac.caltech.edu/>. They also thank WDCGG data contributors.

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