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Atmospheric $CO₂$ at Waliguan station in China: Transport climatology, temporal patterns and source-sink region representativeness

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HIGHLIGHTS highlights are the control of

The transport climatology at Waliguan station in China was obtained by total monthly footprint.

CO2 background values were extracted from simulations, and temporal patterns thereof were compared with observations.

• A method is proposed to explore where the sources and sinks have the greatest impact on $CO₂$ background concentrations.

• The sources and sinks were analyzed in the representative source-sink regions of the monthly CO₂ background concentrations.

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In order to explore where the source and sink have the greatest impact on CO₂ background concentration at Waliguan (WLG) station, a statistical method is here proposed to calculate the representative sourcesink region. The key to this method is to find the best footprint threshold, and the study is carried out in four parts. Firstly, transport climatology, expressed by total monthly footprint, was simulated by FLEX-PART on a 7-day time scale. Surface CO₂ emissions in Eurasia frequently transported to WLG station. WLG station was mainly influenced by the westerlies in winter and partly controlled by the Southeast Asian monsoon in summer. Secondly, $CO₂$ concentrations, simulated by CT2015, were processed and analyzed through data quality control, screening, fitting and comparing. $CO₂$ concentrations displayed obvious seasonal variation, with the maximum and minimum concentration appearing in April and August, respectively. The correlation of CO₂ fitting background concentrations was $R^2 = 0.91$ between simulation and observation. The temporal patterns were mainly correlated with $CO₂$ exchange of biosphereatmosphere, human activities and air transport. Thirdly, for the monthly $CO₂$ fitting background concentrations from CT2015, a best footprint threshold was found based on correlation analysis and numerical iteration using the data of footprints and emissions. The grid cells where monthly footprints were greater than the best footprint threshold were the best threshold area corresponding to representative source-sink region. The representative source-sink region of maximum $CO₂$ concentration in April was primarily located in Qinghai province, but the minimum $CO₂$ concentration in August was mainly influenced by emissions in a wider region. Finally, we briefly presented the $CO₂$ source-sink characteristics in the best threshold area. Generally, the best threshold area was a carbon sink. The major source and sink were relatively weak owing to less human activities and vegetation types in this high altitude area. $CO₂$ concentrations were more influenced by human activities when air mass passed through many urban areas in summer. Therefore, the combination of footprints and emissions is an effective approach for assessing the source-sink region representativeness of $CO₂$ background concentration.

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1. Introduction

Carbon dioxide $(CO₂)$ is one of the most important greenhouse gases in atmosphere ([Xia et al., 2015](#page--1-0)). Biogeochemical cycling and

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atmospheric budget of $CO₂$ play the key role in global climate change ([Liu et al., 2014](#page--1-0)). The longest continuous record, starting in 1958, of $CO₂$ in the atmosphere has been called "Keeling Curve" at Mauna Loa [\(Keeling, 2008](#page--1-0)). The globally averaged mole fraction (concentration) of CO₂ in 2014 was 397.7 \pm 0.1 ppm, reaching new highs [\(WMO, 2015\)](#page--1-0). The anthropogenic $CO₂$ emissions were 10.0 ± 0.9 Pg C in 2010, estimated from fossil-fuel combustion, cement production, and land-use change [\(Friedlingstein et al.,](#page--1-0) [2010; Peters et al., 2011\)](#page--1-0). The major sinks of atmospheric $CO₂$ were ocean and terrestrial ecosystems, absorbing approximately 55% of the fossil fuel and cement emissions during the past 50 years ([Ballantyne et al., 2012](#page--1-0)).

Temporal patterns of atmospheric $CO₂$ are always concerned by scientists ([Steffen et al., 2015\)](#page--1-0). But temporal and spatial distributions of atmospheric $CO₂$ are inhomogeneous [\(WDCGG, 2013\)](#page--1-0). Therefore, various observations have been performed, such as optical remote sensing of ground, air-borne and space-borne ([Frankenberg et al., 2015; Qu et al., 2013; Wunch et al., 2011\)](#page--1-0). As early as 1985, the World Meteorological Organization (WMO) began to coordinate what is now called Global Atmosphere Watch (GAW), observing and analyzing atmospheric greenhouse gases and other trace species [\(WMO, 2015](#page--1-0)). At present, there are three regional atmospheric background stations with one global site ("Waliguan") in China under the framework of WMO/GAW [\(Liu](#page--1-0) [et al., 2009\)](#page--1-0). In order to get traceable, comparable and long-term CO2 dataset, previous studies were focused on the quality control, screening, interpolation and fitting methods of observed $CO₂$ concentrations [\(Zhou et al., 2005; Fang et al., 2014](#page--1-0)). Combining with δ^{13} C isotopic tracing and air-mass backward trajectories, the CO₂ dataset was used to analyze temporal patterns of atmospheric $CO₂$ ([Xia et al., 2015; Zhang and Zhou, 2013\)](#page--1-0). Also, atmospheric $CO₂$ concentrations measured at WMO/GAW stations were applied to infer through the use of inverse models global or regional $CO₂$ sources and sinks, which were influenced by limited data and uneven distribution of sites ([Deng et al., 2007; Maksyutov et al., 2013\)](#page--1-0). Then the simulated $CO₂$ concentrations could be obtained with the top-down approach [\(Nassar et al., 2011](#page--1-0)). The reliability of the simulation results was verified by the observational $CO₂$ data ([Cheng et al., 2013](#page--1-0)). But there has been little research on how to process the simulated $CO₂$ concentrations. The post-processing method of simulated CO₂ concentrations was so important that we could extract specific time series (such as $CO₂$ background concentrations) from them. Here, we have tried to screen the reliable $CO₂$ concentrations simulated by Carbon Tracker (2015) (CT2015) model into background and non-background. After background fitting, the seasonal variations of monthly $CO₂$ concentrations were analyzed by the comparison of simulation with observation.

However, the frequent mention of "source-sink region representativeness" indicates where the sources and sinks have the greatest impact on specific time series of atmospheric $CO₂$ concentration. Studies of representativeness have been reported before. For example, spatial representativeness of flux tower eddycovariance measurements was characterized using remotely sensed vegetation indices and footprint analysis of land cover ([Chen et al., 2012](#page--1-0)). The representativeness of land cover and leaf area index (LAI) sampled by a global network of sites was investigated to be used for the evaluation of LAI, derived from satellites ([Baret et al., 2006](#page--1-0)). Gu-Ting (an urban air monitoring station) was well representative of outdoor concentrations of air pollutants for a period of three weeks within a 700 m radius around this station ([Chan and Hwang, 1996\)](#page--1-0). Spatial characteristics of PM2.5 were used to identify potential monitoring sites, which were most representative of the overall ambient exposures to $PM_{2.5}$ among susceptible populations in Seattle, Washington [\(Goswami et al., 2002\)](#page--1-0).

Parameters reflecting air quality monitoring site representativeness were evaluated in Europe by taking emissions, deposition, transport, and land use characteristics into account ([Henne et al., 2010;](#page--1-0) [Janssen et al., 2012](#page--1-0)). These studies were mainly focused on representativeness of fluxes, radiation, temperature, and air pollutants at meteorological observation stations, national reference climato-logical stations and urban environmental monitoring stations ([Xu](#page--1-0) [et al., 2015\)](#page--1-0). Different from them, we analyze the source-sink region representativeness of simulated $CO₂$ background concentrations at the WMO/GAW global background station of Waliguan, which is significant for the evaluation of site layout and $CO₂$ data application [\(Janssen et al., 2012](#page--1-0)).

In this study, the overall aim is to determine the source-sink region representativeness for the simulated $CO₂$ fitting background concentrations on monthly scale. Therefore, we will introduce $CO₂$ observation, transport and dispersion model, $CO₂$ inversion model, and the statistical analysis methods for representative source-sink regions in section 2. The results and discussion consist of four steps in section [3:](#page--1-0) (1) Transport climatology is analyzed by total monthly footprint distributions; (2) The simulated $CO₂$ concentrations are presented and compared with observation results through a series of post-processing; (3) Sourcesink region representativeness is discussed, i.e. representative source-sink regions are determined for the simulated $CO₂$ fitting background concentrations from March 2009 to December 2010 at Waliguan station; and (4) CO₂ source-sink characteristics in representative source-sink regions are briefly presented. Finally, conclusions are presented in Section [4](#page--1-0).

2. Methods

2.1. $CO₂$ observation

Flask sampling observation of $CO₂$ has been operational since 1991 at the Mt. Waliguan (WLG) station (36 $^{\circ}$ 17'N, 100 $^{\circ}$ 54'E, and 3810 m above sea level) [\(Liu et al., 2014](#page--1-0)). As the only global background station of WMO/GAW in the Central Eurasia Continent, WLG station is a multi-ethnic-populated area with the typical high plateau continental climate ([Zhang et al., 2013](#page--1-0)). The intensities of CO2 source and sink are weak within 100 km radius around WLG because of sparse vegetation, arid and semi-arid prairie, and little human activity [\(Zhou et al., 2003; Fang et al., 2014](#page--1-0)). WLG is relatively isolated from industrial and populated centers. Geographical location and surrounding cities of this station have been shown in [Fig. 1.](#page--1-0)

The discrete air sampling, measurement and analysis of $CO₂$ at WLG have been carried out using methods recommended by WMO/ GAW [\(Fang et al., 2015\)](#page--1-0). Before the use of flasks for sampling, there were a series of operations in order, such as gas leakage test and filling with the dry balance gas. Air samples were collected in series into a pair of flasks when atmospheric conditions were suitable for sampling [\(Tans et al., 1989](#page--1-0)). Generally, flask pairs at WLG were collected weekly in the morning local time to capture the downdraft and avoid interference of man-made activity [\(Li et al., 2014;](#page--1-0) [Xia et al., 2015](#page--1-0)). The $CO₂$ concentrations in air samples were analyzed in greenhouse gases research laboratory of China Meteorological Administration (CMA) in Beijing and National Oceanic and Atmospheric Administration (NOAA) in Boulder, Colorado. The processing of original $CO₂$ concentrations would be performed, which consisted of quality control, screening, and curve-fitting. In addition, both working and calibration standard gases are traceable to WMO X2007 standard scale. More details have been described by [Thoning et al. \(1989\), Masarie et al. \(2001\), Zhou et al. \(2005\), Liu](#page--1-0) [et al. \(2014\)](#page--1-0) and [Fang et al. \(2014\)](#page--1-0).

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