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The impacts of summer monsoons on the ozone budget of the atmospheric boundary layer of the Asia-Pacific region

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HIGHLIGHTS

- The Asia-Pacific monsoon greatly affects O_3 seasonal and inter-annual variations.
- The differences of emissions and zonal winds lead to pollutants transition zone.
- Advection plays a key role in the monsoon impact on O_3 inter-annual variation.

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ABSTRACT

The seasonal and inter-annual variations of ozone (O_3) in the atmospheric boundary layer of the Asia-Pacific Ocean were investigated using model simulations (2001–2007) from the Model of Ozone and Related chemical Tracers, version 4 (MOZART-4). The simulated O_3 and diagnostic precipitation are in good agreement with the observations. Model results suggest that the Asia-Pacific monsoon significantly influences the seasonal and inter-annual variations of ozone. The differences of anthropogenic emissions and zonal winds in meridional directions cause a pollutants' transition zone at approximately 20°–30°N. The onset of summer monsoons with a northward migration of the rain belt leads the transition zone to drift north, eventually causing a summer minimum of ozone to the north of 30°N. In years with an early onset of summer monsoons, strong inflows of clean oceanic air lead to low ozone at polluted oceanic sites near the continent, while strong outflows from the continent exist, resulting in high levels of O_3 over remote portions of the Asia-Pacific Ocean. The reverse is true in years when the summer monsoon onset is late.

1. Introduction

Tropospheric ozone is an important greenhouse gas, pollutant, and source of OH radicals. Its contribution to global warming from the preindustrial era to the present is regarded as the third most important, following those of carbon dioxide and methane (Solomon et al., 2007). The level of tropospheric ozone also affects human health and natural ecosystems. Elucidation of the processes determining spatial and temporal variations in tropospheric ozone is important for evaluating the effect of ozone on regional air quality and climate change.

Monsoons are a seasonal variation of wind, particular of wind directions, which result from the variations of meridional differences in solar radiation and the thermal difference between the land and sea. It is an important element of the global climate system. Asian monsoons are composed of three sub-systems, which are the tropical Indian monsoon, the tropical Western North Pacific (WNP) monsoon, and the subtropical East Asian (EA) monsoon (Zhu et al., 1986; Wang and Lin, 2002). Over the Asia-Pacific region, low-level winds seasonally reverse from winter easterlies to summer westerlies for the WNP monsoon and from winter northerlies to southerlies for the EA monsoon. The EA and WNP monsoons are considered together as the Asia-Pacific monsoon hereafter in this study. Whether considering the EA monsoon or the WNP monsoon, the transition of the winter monsoon to the summer monsoon results in a series of changes in weather, such as wind shifts, convection, precipitation, and air temperature. Together these changes significantly affect the transport paths and photochemical production of pollutants. The observed summer minimum of surface O_3 over the Asia-Pacific region was attributed to the incursion of the monsoon, which transports oceanic air with less background O_3 to the region, causing lower O_3 concentrations (Chan et al., 1998; Pochanart et al., 2002; Wang et al., 2006; Yamaji et al., 2006; Zbinden et al., 2006; He et al., 2008). Tanimoto et al. (2005) and He et al. (2008) identified a relationship between monsoons and the O_3 spring maximum at the surface. He et al. (2008) found that

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the basic common features of O_3 seasonal behaviors over the monsoon region are the pre- and post-monsoon peaks with a summer trough; these bimodal seasonal patterns become weaker or even disappear outside of the monsoon region. Kurokawa et al. (2009) investigated the influence of meteorological variability on the interannual variation of the springtime boundary layer ozone over Japan, and found some correlation between spring ozone over Japan and the El Niño-Southern Oscillation. Zhu (2012) determined that ozone seasonal cycle features were mainly controlled by the seasonal transitions of Asia-Pacific monsoon circulations by using monthly surface ozone data and related wind field and precipitation data.

This paper addresses the influence of the Asia-Pacific monsoon on seasonal and inter-annual variations of boundary layer ozone over the Asia-Pacific region, based on an analysis of the ozone budget using the chemical transport model (CTM), Model of Ozone and Related chemical Tracers, version 4 (MOZART-4). A fixed emission experiment was performed to identify the influence of early/late summer monsoon onset on the O_3 budget. For the Asia-Pacific monsoon region, various indices based on considerations of thermodynamics and dynamics from different aspects have already been defined. Each monsoon index pays attention to some specific physical processes and represents its own meanings, independent of others. In this study, meridional wind (Lau and Li, 1984; Chen et al., 1991) was used to reveal the seasonal march of the summer monsoon, and a dynamical normalized seasonality monsoon index developed by Li and Zeng (2003) was used to distinguish the early and late transition periods of the Asia-Pacific summer monsoon onset. The importance of the impact on the ozone levels by advection, convection, diffusion, and photochemistry over the Asia-Pacific monsoon region was evaluated.

2. Data and methods

2.1. Observed data

Observations of daily O_3 at seven regional stations were taken from the Acid Deposition Monitoring Network in East Asia (EANET): Rishiri, Tappi, Sado-Seki, Oki, Yusuhara, Hedo, and Ogasawara. Descriptions of these sites can be found at http://www.eanet.asia/site/index.html. Observations from two additional sites, Yonagunijima and Minamitorishima, were obtained from the WMO-World Data Centre for Greenhouse Gases (WDCGG) (http://ds.data.jma.go.jp/gmd/wdcgg/). The site location information including latitude, longitude and elevation height above sea level for all sites is provided in Fig. 1. Daily precipitation data collected from 2001 to 2007 are taken from the Global Precipitation Climatology Project (GPCP) with a resolution of $1^\circ \times 1^\circ$ (http://precip.gsfc.nasa.gov/). The GPCP dataset reflects the spatial and temporal distributions of precipitation very well, as a combination of GPCC (Global Precipitation Climatology Center) ground observations from precipitation gauges and a precipitation inversion from satellite remote sensing observations.

2.2. Model setup

A detailed description and evaluation of the standard version of MOZART-4, and the upgrade over MOZART-2 (Horowitz et al., 2003), is given by Emmons et al. (2010). MOZART-4 includes an updated chemical scheme of hydrocarbon and bulk aerosols (Tie et al., 2001, 2005), and improved emissions compared to MOZART-2. It does not include explicit stratospheric chemistry, but constrains the climatological mixing ratios of ozone and other species in the stratosphere. The Synoz (synthetic ozone) scheme (McLinden et al., 2000) is used as a flux upper boundary condition for ozone in the stratosphere and yields a cross-tropopause ozone flux of 500 Tg/yr. More details about the physical and chemical mechanisms were discussed in Emmons et al. (2010). In this study, MOZART-4 is run with the standard chemical mechanism [see Emmons et al., 2010 for details], with online calculation of dry deposition. It is driven by meteorological parameters from the NCAR reanalysis of the National Centers for Environmental Prediction (NCEP) forecasts. The output from the model run is available at a temporal resolution of 6 h, a horizontal spatial resolution of approximately $2.8^\circ \times 2.8^\circ$, and 28 hybrid levels in the vertical. The top model level is located at approximately 2 hPa. The initial condition and emissions are based on the NCAR Community Data Portal (http://cdp.ucar.edu/). The model was run in time steps of 20 min from June 2000 to December 2007, with the first seven months used to spin up. The experiments used fixed emissions from 2001 and the meteorological parameters were varied for each year over the simulations, with the first seven months as spin-up time. Note that the modeled results are based on the mean values in the atmospheric boundary layer (the six lowermost layers in the model, surface to ~2 km) except for the model validations in Section 3.1 and the special definitions in Section 3.2.

2.3. Monsoon index and ozone budget

Previous studies showed that the East Asian monsoon is characterized as a seasonal reversal of the lower-troposphere meridional wind direction (Lau and Li, 1984; Chen et al., 1991), and the 850 hPa wind is used to reflect the low-level atmospheric circulation (Lu and Chan, 1999). Therefore, the daily meridional wind at 850 hPa is used to show the seasonal march of the Asia-Pacific monsoon in this study. A pre-defined monsoon index (MI) can provide a useful insight into quantitatively examining the strength and variation of monsoon circulation in monsoon regions. For the Asia-Pacific monsoon region, many studies have already defined various indices based on considerations of dynamics and thermodynamics from different aspects, such as wind field, precipitation, difference of ocean–land temperature, and so on. Each MI is designed to capture specific physical processes. In this study, we used a dynamically normalized seasonality MI developed by Li and Zeng (2003) to examine the wind field to investigate the influence of the Asia-Pacific monsoon on the inter-annual variability of ozone.

The factors controlling ozone levels in the atmospheric boundary layer (transport, net photochemical production, and deposition) are discussed in this study. Each term affecting the ozone budget is evaluated quantitatively at 850 hPa in Section 3.2 and in a column from the surface to approximately 2 km (the atmospheric boundary layer) in Section 3.3. The rate of change of ozone can be expressed as:

$$\frac{dO_3}{dt} = \mathrm{Chem} + Adv + \mathrm{Con} + Dif - Dep$$

where *Chem* represents the net chemical production; *Adv*, *Con*, and *Dif* are the transport fluxes associated with advection, convection and diffusion, respectively; *Dep* is the dry deposition rate. Fig. 4 shows each of these components of the ozone budget for the study region: net chemistry, net transport flux (advection, convection, and diffusion), and O_3S. O_3S is an O_3 tracer that tags O_3 transported from the stratosphere (Sudo and Akimoto, 2007).

3. Results and discussion

3.1. Validation of simulated O_3 and precipitation

The modeling system described in Section 2.2 has previously been used for analyzing tropospheric O_3 over the northern hemisphere (Horowitz et al., 2003; Liu et al., 2005; Pfister et al., 2008a,b; Emmons et al., 2010), and, in these studies, the simulated results showed good agreement with observations. Stevenson et al. (2006) suggested that the tropospheric O_3 budget in MOZART-4 is in good agreement with the mean of 26 models, whereas the stratospheric input value has been determined to be realistic by Wild (2007). In this section, we further evaluate the general performance of our modeling system for O_3 over the Asia-Pacific region.