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Original article

Seasonal relationship between meteorological conditions and surface ozone in Korea based on an offline chemistry–climate model

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A B S T R A C T

Tropospheric ozone plays an important role in climate variation and air quality, and it has seen a dramatic rise in East Asia due to the region's rapid economic growth. The relationship between meteorological conditions and surface ozone in Korea varies seasonally, and to obtain a better understanding of this process, this study performed offline simulations using a climate–chemistry model. The model represented the observed annual cycle of surface ozone over East Asia well, including the spring/autumn peaks and summer troughs. Increases in ozone were associated primarily with the westerly wind anomaly during spring and with surface warming during the autumn and summer. Moreover, a decrease in ozone during the summer likely resulted from the transportation of ozone-depleted air masses by anomalous southeasterly winds. Reduced cloud cover increased ozone levels significantly during all seasons except winter. The relationship between the El Niño and Southern Oscillation and ozone concentrations in Korea was also examined. Spring ozone levels tended to be elevated following mature-phase El Niño winters, whereas elevated levels during summer and autumn followed La Niña winters.

1. Introduction

Tropospheric ozone is considered the third most important greenhouse gas after CO_2 and CH_4 (Forster et al., 2007) and it plays a crucial role in climatic variability and air quality (Allen et al., 2012; U.S. EPA, 2015). Ozone concentration has increased, mainly due to emissions from ozone-forming precursors (e.g., NO_x, non-methane volatile organic compounds, CH_4, and CO), which has led to a global mean radiative forcing of $+0.35$ Wm^{-2} ($+0.25$ to $+0.65$ Wm^{-2}) (Crutzen, 1974; Derwent et al., 1996; Ziemke et al., 2005). This elevated concentration of tropospheric ozone has detrimental effects on human health and crop production (Wang and Mauzerall, 2004; Chang et al., 2005).

Recently, East Asia in general and China in particular, have seen dramatic rises in tropospheric ozone in relation to their rapid economic growth (Shindell et al., 2006; Tanimoto, 2009; Wang et al., 2009; Jeong and Park, 2013). Some studies have suggested East Asia to be the region with the highest rate of premature mortality caused by ozone exposure, followed by India (Nawahda et al., 2012; Silva et al., 2013). Furthermore, modeling studies have predicted that surface ozone concentrations in East Asia will continue to increase in the near future (Dentener et al., 2005). Consequently, changes in tropospheric ozone in Korea are now of great concern (Ghim and Chang, 2000; Kim et al., 2002a, 2013; Yoo et al., 2005), leading to a call for the prevention of air pollution and the mitigation of climate change.

Significant attention has been paid to the analysis of meteorological effects on observed surface ozone in major Korean cities. For example, Kim et al. (2002b) showed that high concentrations of surface ozone in Seoul were associated with both high temperatures and weak winds; conditions favorable for in situ production and accumulation of ozone. Furthermore, high concentrations of ozone in China can have considerable effect on surrounding areas and even on the rest of the Northern Hemisphere via long-range transport (Akimoto, 2003; Liu et al., 2003; Cooper et al., 2010). In some Korean cities, the relationship between meteorological conditions and surface ozone has been found to vary seasonally (Kim et al., 1997; Kim and Park, 1998); however, the manifestation of this relationship over the entire Korean Peninsula remains poorly understood because of limited distribution of surface ozone observations. The objective of this study was to improve the

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understanding of meteorological effects on surface ozone in all seasons in Korea by performing simulations of tropospheric ozone.

2. Model and simulation

To simulate tropospheric ozone concentration and its changes associated with meteorological conditions, a global 3-D chemical transport model (GEOS-Chem v. 8-01-03) was used, which included full coupling of the tropospheric ozone–NO_x–VOC gas-phase chemical mechanism (Park et al., 2003, 2004). Originally, this model was developed to be forced by meteorological field data from the Goddard Earth Observing System (GEOS) of the National Aeronautics and Space Administration Global Modelling and Assimilation Office (Bey et al., 2001). The GEOS-Chem model has $2^\circ \times 2.5^\circ$ (latitude × longitude) horizontal resolution and 26 vertical layers from the surface to 3.5 hPa. Instead of using GEOS meteorological input, the GEOS-Chem code was modified slightly to use meteorological data from the third version of the National Center for Atmospheric Research Community Atmosphere Model (CAM3) (Collins et al., 2006). CAM3 is configured with a finite volume dynamic core with $2^\circ \times 2.5^\circ$ (latitude × longitude) horizontal resolution and 26 vertical levels.

The simulation was performed in two steps: initially, CAM3 was run from 1970 to 2011 with forcing from the Hadley Centre sea surface temperature, producing meteorological data; then, GEOS-Chem was run using the CAM3 output. To isolate the role of meteorology on tropospheric ozone, GEOS-Chem was driven using annually constant values for biomass and anthropogenic emissions. Therefore, the meteorological fields strongly determined the simulated variations in the concentration of chemicals through transport, washout, and chemical reaction rates, independently of either natural or anthropogenic emission-induced variability.

Both the simulated meteorological fields and the surface ozone concentrations were monthly mean data based on the same horizontal grid ($2^\circ \times 2.5^\circ$; latitude × longitude) for the period 1970–2011 and they were used to calculate the seasonal means. To identify statistical relationships between the meteorological fields and surface ozone concentrations, linear regression and Pearson correlation analyses were employed. These methods determine whether one variable is associated with another variable (Wilks, 2011). The statistical significance of the regression and correlation was computed using the Student t-test with a null hypothesis that $R^2 = 0$ (R is the correlation coefficient), i.e., no linear relationships between the variables. Furthermore, some individual correlation coefficients were computed together with the so-called p-values (p), which indicated the statistical significance of a linear relationship. If the value of probability p is small (e.g., $p < 0.05$), the null hypothesis may be rejected with a >95% confidence level. Qualitatively, the regression and correlation analyses provided similar results and therefore, the present study focused mainly on the correlation analysis, except for the relationship between wind and surface ozone concentration. Since regression analysis involves the variance of predictors and predictants, regression coefficients between wind and ozone have a specific dimension (e.g., m/s/ppbv) which represents the velocity changes given a unit departure in ozone concentration over Korea.

3. Results and discussion

3.1. Simulated climatology and surface ozone

Initially, the performance of CAM3 in simulating surface wind and precipitation over East Asia was evaluated. The seasonal mean distributions of precipitation and 850 hPa winds from both CAM3 and observations for the period 1970–2011 are presented in Fig. 1. The results clearly show important aspects of the summer/winter East Asian monsoon system. Seasonal reversals in low-level winds and associated precipitation arise from the differential heating of land and ocean (Webster and Fasullo, 2002). Although CAM3 is reasonably good at simulating these dramatic reversals, some large-scale biases exist, including reduced levels of precipitation over Korea and excessive easterly oceanic winds during the summer. Similar errors can also be seen during the other seasons, including small precipitation anomalies and a change in the sign of the anomaly in spring. Despite these biases, which are known as common biases in the field of climate modeling (ul Islam et al., 2013; Song and Zhou, 2014), this modeling system has been used successfully in previous research to study climate–chemistry interactions in East Asia (e.g., Moon et al., 2011; Youn et al., 2011) and the tropical Pacific (Moon et al., 2013).

The horizontal distributions of simulated surface ozone concentration, wind fields, and sea level pressure in East Asia were averaged over all months and for each season from 1970 to 2011 (Fig. 2a and b). Most of the continent, including the Korean Peninsula, and the adjacent Yellow and East/Japan seas are shown to be within a zone of high total mean ozone concentration (Fig. 2a). Both winds and surface ozone concentration are characterized by dramatic seasonal variability (Fig. 2b). In spring, Korea is influenced mainly by westerly and southwesterly winds with associated high concentrations of ozone transported from the continent. The wind becomes more southerly (30°N–40°N) during summer, because of the subtropical high, which transports cleaner air from the Pacific Ocean across Korea, southern China, and Japan. During autumn, the concentration of surface ozone becomes higher over the Yellow Sea, East/Japan Sea, and southern China in association with the prevalence of northerly winds. In winter, Korea is affected mostly by clean air and northwesterly winds driven by the Siberian High system. In contrast to the other seasons, a zone of high ozone concentration is located over the western North Pacific. Other modeling studies have reported similar seasonal changes in surface ozone concentrations (e.g., Chatani and Sudo, 2011).

These results clearly show that Korea experiences a spring/autumn maximum and summer/winter minimum in surface ozone concentration, which could be attributed to continental–oceanic air mass exchange resulting from the East Asian monsoon (Pochanart et al., 2002; He et al., 2008). The simulations show that ozone concentrations over Korea decrease sharply from spring to summer, coincident with the transition from easterly to southerly winds at approximately 30°N (Fig. 2b). Ozone levels are then shown to recover in autumn following the reemergence of westerly winds. To confirm the robustness of the simulation of ozone over Korea, the simulated monthly mean ozone concentrations, obtained by averaging 15 model grid points (red dots in Fig. 2c) were compared with the surface ozone concentrations from 13 ground-based stations (blue dots in Fig. 2c). Note that these model grid points are surrounded by the rectangle shown in Fig. 2a, and that the observed data of surface ozone concentrations for 2006–2014 are available on the Korea Environment Corporation website (KECO, 2015). Fig. 2d shows bimodal seasonality of the surface ozone concentrations in both the simulation and the observational data, which is consistent with previous ground-based measurements (Kim and Park, 1998; Ghim and Chang, 2000). However, some biases are evident in the model simulation, including relatively high levels of ozone and early peaks (by one month). Such discrepancies are probably due to the inability of the model to capture the meteorological conditions over East Asia accurately (as shown in Fig. 1), which could be considered a limitation of this study.