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A case study of surface ozone source apportionment during a high concentration episode, under frequent shifting wind conditions over the Yangtze River Delta, China

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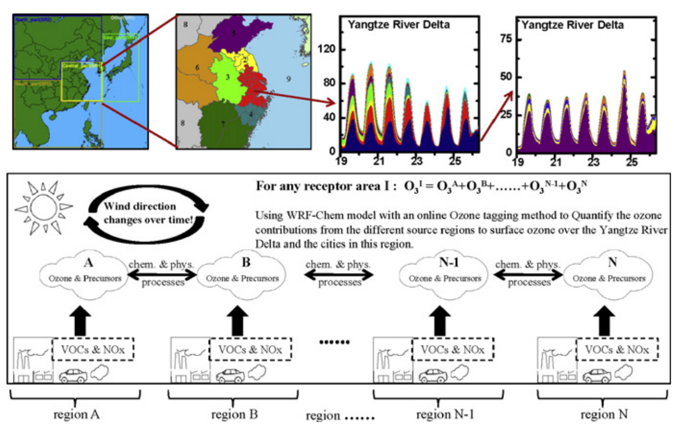
HIGHLIGHTS

- The online ozone tagging method was developed in the WRF-Chem model.
- Ozone from upwind-polluted areas is crucial to high ozone events over YRD.
- Diurnal super regional ozone contribution is significant and relatively stable.
- Vertical mixing plays an important positive role to surface ozone over YRD.

GRAPHICAL ABSTRACT



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ABSTRACT

Surface ozone is an environmental issue occurring at several scales, ranging from local to continental. One of the most developed regions in China, the Yangtze River Delta (YRD), experiences severe tropospheric ozone problem. Hence, quantifying the contributions from various geographical source regions is helpful for better understanding the regional ozone problem. Ozone source apportionment studies can provide relevant information for designing suitable air pollution protection strategies. In the present work, the WRF-Chem model coupled with an online ozone tagging method is applied to a case study, with the objective of exploring the ozone contributions to the surface ozone from different source regions over the YRD region, during a frequent wind-shifting period. Our results show that the YRD was highly affected by the upwind source regions bearing high values both ozone and its precursors. The contribution from the source region outside the main air pollution zones in the Central Eastern China (super regional contribution) was also important, accounting for more than 30 ppb of daytime maximum mean ozone concentrations. Ozone arising from increased local and regional emissions during high-concentration events was more significant than super regional contribution. It reveals that the ozone from Anhui region was transported through vertical mixing and horizontal advection to receptor areas in the YRD during the study time focus. Chemical process contributed significantly at ground and high altitude levels of 500 and

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1000 m. However, most of the ozone from the remote regions of Henan and Hubei provinces was transported to the receptor of Nanjing through physical processes. The vertical mixing process played a crucial positive role at super regional scales, with regard to the formation of surface ozone over the YRD region during the addressed time interval.

1. Introduction

Tropospheric ozone (O_3) is a major air pollutant formed in the troposphere through a series of physical processes and complex photochemical reactions (Crutzen, 1973). The formation mechanism is nonlinearly related to carbon monoxide (CO), nitrogen oxide ($NO_x = NO + NO_2$), and volatile organic compounds (VOCs) with the presence of solar radiation. It's well known that high concentrations of ozone near the surface can affect both vegetation growth and human health (NRC, 1991), being a source for public concern. The surface ozone problem is often governed by local and regional emission factors, such as O_3 lifetime and transport mechanism (Li et al., 2007; Wang et al., 2006; Geng et al., 2011), or even by intercontinental long-range transport (Li et al., 2002; Sudo and Akimoto, 2007; Pfister et al., 2013). Quantifying the ozone contribution from each source to a certain place or region can help investigating ozone source-receptor relationship. In this case, it's effective to divide the study area into several source sub-regions, within which ozone and its precursors are subsequently tracked, allowing the determination of each source contribution to ozone levels in a certain point or district.

China has been experiencing a period of rapid economic expansion and industrial development, with the correspondent increases in surface ozone concentration, arising from enhanced fossil fuel burning emissions. Surface ozone varies both seasonally and spatially over China (Wang et al., 2011). Li et al. (2008) studied the source-receptor relationship of near-ground ozone in Central Eastern China (CEC), demonstrating that nearly half of the near-ground mean O_3 at mountain Tai relates to regional-scale transported contributions of chemically produced ozone in other areas in CEC during June. Wang et al. (2009a) reported that the ozone pollution in Beijing urban areas is affected by emissions both local and surrounding areas. Furthermore, their study also revealed the importance of transport effect, as a major factor governing ozone pollution in Beijing urban areas. Similar studies were also conducted in the Pearl River Delta (PRD) region, located in south China, where ozone problem exhibits both regional and super regional (outside the PRD region) properties (Wang et al., 2009b; Zheng et al., 2010). Li et al. (2012, 2013) studied the ozone source contributions in the PRD region, showing that the super regional contributions are dominant under mean ozone conditions (ozone concentration are in a normal levels), while local and regional sources are very important in generating high ozone episodes. The relevance of local and regional contributions suggests that regional joint control of ozone precursors at a regional scale is decisive for achieving lower tropospheric ozone concentrations in the receptor zones, particularly concerning high ozone events.

The Yangtze River Delta (YRD), being one of the most developed areas in China, has become an economic center in the recent decades. However, the fast economic development also resulted in significantly increased air pollutant emissions (Wang et al., 2007), leading to a number of severe photochemical contamination problems. Ozone pollution in the YRD has been previously approached (Wang et al., 2006; Geng et al., 2008; Ran et al., 2009; Shan et al., 2010; Jiang et al., 2012; Tie et al., 2013), but some properties controlling O3 behavior in this area remain uncertain. A better understanding of O3 source-receptor relationship in the YRD can be accomplished through the identification of the different sources involved, especially during shifting wind conditions. Furthermore, such information is vital for designing adequate control strategies regarding tropospheric ozone concentration.

In this work, we firstly developed the tagging method in the WRF-Chem model, subsequently applying the model to a case study concerning the identification of contributions from different geographical source regions to the surface ozone in the YRD region when the receptor region, under frequent shifting wind conditions at the receptors. The rest of this work is organized as follows. Section 2 introduces the model configurations, the relevant data, and model evaluations. Section 3 presents the ozone contributions from source regions in the YRD region and the mechanism of ozone formation from source regions to receptors. Finally, we close by summarizing our findings.

2. Model simulations and evaluation

2.1. Model description and used data

The version 3.4 of WRF-Chem model (Weather Research and Forecasting with Chemistry) was used in the study and a detailed description of this approach can be consulted in Skamarock et al. (2008). The chemistry component, fully coupled online with the WRF model, is also defined in Grell et al. (2005). The model was set up considering two domains (Fig. 1) with horizontal resolutions of 36 and 12 km, and 140 × 140 and 126 × 126 grids, for domains d01 and d02, respectively. Domain d01 covered most of East Asia and the corresponding simulation provided meteorological and chemical boundary conditions to the inner domain. The second domain covered middle and east China, where the high concentration of ozone is mainly addressed during the simulation period, being suitable for modeling chemical reactions and transport processes of air pollutants relevant for the assessment of local and regional contributions to the surface ozone over the YRD area. The vertical structure of the atmosphere was represented with 38 sigma levels between surface and 50 hPa, including 12 levels within the lowest 2 km. Simulation time spanned from 1 to 27 May 2013, with

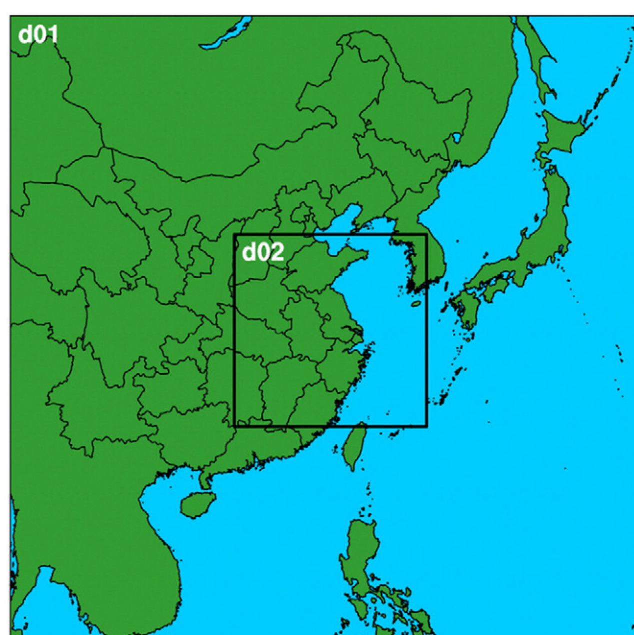

Fig. 1. Map of the two model domains.