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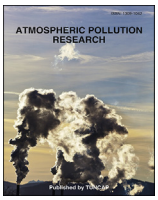

Identification of the potential regions contributing to ozone at a coastal site of eastern China with air mass typology

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ABSTRACT

The concentrations of O_3 and CO at a coastal site of eastern China were analyzed using hourly data from February 2015 to January 2016. Conditional probability function, bivariate correlation, trajectory clustering and potential source contribution function were applied to determine the potential regions contributing to surface O_3 at the study site. The O_3 and CO concentrations reached their maxima in autumn and winter, respectively. Relatively high exceeding rate (8.8%) over the national 8-h O_3 standard of China (75 ppb) was observed, which indicated significant health risks of O_3 exposure to people. The results of all the analyses suggested that the major potential regions contributing to O_3 were located in the coastal and marine areas of eastern China. The highest O_3 levels occurred with relatively high wind speeds (5 m/s), revealing significant contributions of external sources. Based on the cluster analyses of backward trajectories, the air masses associated with the coastal and marine clusters contributed to 32.2% and 37.4% of surface O_3 at the study site, respectively. The domain of major potential region varied with seasons, with larger areas appearing in summer and autumn, and smaller area in winter, which suggested more contributions to the O_3 pollution from distant transport during warm seasons. As to the influence mechanisms on surface O_3, the spatial position of the study site, mesoscale process and long-range transport of pollutants from northern China might play important roles. Besides, favorable climate conditions (e.g. high temperature) could also significantly contribute to the O_3 pollution during the study period.

1. Introduction

Surface ozone (O_3) is a typical secondary air pollutant mainly produced from photochemical mechanisms. The processes consist of oxidation reactions involving its precursors such as nitrogen oxides (NO_x), non-methane volatile organic compounds (NMVOCs) and carbon monoxide (CO) in the presence of solar radiation (Fishman and Crutzen, 1978; Sillman, 1999). A portion of O_3 could also be downward transported from stratosphere (Cuevas et al., 2013). Ozone depletion usually occurs by deposition and consumption in atmospheric chemistry. At a specific location, the O_3 budget is comprehensively affected by precursor concentrations, meteorological conditions and regional transport. The oxidative characteristics of O_3 have negative effects on human health (e.g. lung irritations) and plant growth (e.g. chlorosis), and large amounts of human deaths and crop yield reduction have been confirmed to be O_3-related (Wang and Mauzerall, 2004; Fann et al., 2012).

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With the increasing anthropogenic emissions of O_3 precursors, a steady rise of background O_3 level has been observed worldwide since 1950s (Royal Society, 2008). For the region of western North America, the increase rate was as high as 0.70 ppb yr^{-1} for springtime O_3 during 1984–2008 (Cooper et al., 2010). Benefiting from the active emission reduction of O_3 precursors, O_3 decreases have been observed in Europe and the eastern America since 2000 (Monks et al., 2015). However, in Asian countries like China and India, the O_3 pollution is still deteriorating due to the rapid industrialization and urbanization (Reddy et al., 2012; Tang et al., 2012). During warm seasons (e.g. late spring and summer) with strong solar radiation, O_3 has frequently become the top air pollutant replacing particulate matter (PM) in many areas subjected to haze problem (http://datacenter.mep.gov.cn/). With the decrease in PM level under the policy control on fine particle, O_3 pollution is predicted to be more prominent in Asian region (Liu et al., 2013).

As an oxidative gas species, O_3 has a lifetime from several hours to days in the boundary layer and a few weeks in the free troposphere (Jacob et al., 1996; Monks et al., 2015). Thus, it can be transported over long distance via large-scale atmospheric circulations (Cooper et al., 2010). Besides, O_3 precursors (e.g. CO and NO_x) with longer atmospheric lifetime in the free troposphere (days to months) could also be transported on large spatial scale, during which process more O_3 could be produced and accumulated (Jeon et al., 2014). Determining the spatial regions which could contribute to surface O_3 at a specific area is critical for further pollution control on both local and regional scales. It should be noted that, as the surface O_3 at the target area could be contributed not only from the direct O_3 transport but also from the transport of O_3 precursors, which could be converted into O_3 by photochemical reactions during the transport to the receptor site, the potential contributing regions to surface O_3 should involve both the areas directly exporting background O_3 and the areas exporting O_3 precursors. For the former areas, the surface O_3 is either coming from outside of its region by background transport or being formed locally by photochemical conversion. In the following discussion of this study, the 'potential contributing regions' was used to represent the comprehensive influences of both processes on surface O_3 at the study area.

Using an ensemble of techniques including aircraft, satellite, and surface observations, Zhang et al. (2008) found that Asian pollution had enhanced surface O_3 concentrations by 5–7 ppb over western North America in spring 2006. Choi et al. (2014) has also found that the NO_x and VOC emissions from China could significantly contribute to the O_3 level of South Korea. Besides being an active source region for exporting O_3 and its precursors, Asia has also been affected by pollutants outflow from other continents. Based on the global chemical transport model (MOZART-4), Li et al. (2014) found that transport of O_3 originating from anthropogenic emissions from Europe could also strongly influence O_3 in western China. Given the relatively long lifetime (ca. two months) of CO, which is a good tracer of anthropogenic pollution, the long-range transport of air pollutants has also been widely confirmed by the positive correlation between O_3 and CO levels (Cristofanelli et al., 2013; Cuevas et al., 2013). Eurasian plumes with correlated CO and O_3 are often observed in the free troposphere over North America (Price et al., 2004). Recently, trajectory analysis based on Hybrid Single-Particle Lagrangian Integrated Trajectory model (HYSPLIT) has also been applied to determine the potential source of O_3 (e.g. Ding et al., 2013; Vellingiri et al., 2016), with considerable amount of surface O_3 being accounted for by long-range transport.

China is now experiencing unprecedent air pollution due to large consumption of fossil fuels (Gao et al., 2017). O_3 pollution with high level above the national air quality standard has frequently been observed, especially at developed eastern and southern areas (Shan et al., 2008; Liu et al., 2013). So far, numerous studies have analyzed the pollution characteristics of surface O_3 in China (e.g. Cheung and Wang, 2001; Tu et al., 2007; Wang et al., 2015b). Using the modeling techniques, Ding et al. (2013) found strong influence of regional transport of precursors from eastern China on O_3 level in Hongkong. Li et al. (2016) found that O_3 produced in the Pacific Ocean could significantly contribute to concentrations in different regions of China via long-range transport. Ningbo (28°51′–30°33′ N, 120°55′–122°16′E) is one of the economic and traffic centers of Zhejiang Province, China and the world's fourth largest container port city. There are more than 60 ten-thousand-ton level berths distributed here, which have greatly favored the development of local industry. Considerable amounts of O_3 precursors were released into the atmosphere annually in Ningbo (Zheng et al., 2014), which might significantly aggravate local O_3 pollution. However, very limited studies have analyzed the variations of surface ozone in this city (Jiang et al., 2016), while no published articles have reported the possible sources of O_3 here.

In this study, the O_3 pollution at a coastal site of Ningbo over the period of 2015–2016 was analyzed based on backward trajectory analyses, conditional probability function (CPF), bivariate correlation analysis and potential source contribution function (PSCF). The major objectives are to (1) identify the typology of air masses; (2) determine the potential regions contributing to surface O_3 in the study area; and (3) reveal the possible causes for the distribution characteristics of these potential regions.

2. Materials and methods

2.1. Study area

The study site is located at Ningbo Urban Environment Observation and Research Station (NUEORS) (29°45′ N, 121°54′ E), Ningbo city, Zhejiang province. NUEORS is built at a region under rapid development of southeastern Ningbo, which is to the west of East China Sea (Fig. 1). A large area of land in this region has been created by tidal flat landfill, and many projects on school and housing construction are under way here. NUEORS is 37 km away from the city center with much lower traffic density nearby. It is close to an artificial lake and undeveloped land at distances <0.2 km. A mountain range (southwest to northeast trend) is 2.5 km away from the west of NUEORS while the sea is 0.6 km away from the east of this site. Some mechanical fitting factories are located at the foot of the mountain.

As to the climate conditions, the study area belongs to the humid subtropical climate (i.e. climate group of Cfa in the Köppen climate classification) with humid summers and mild to cool winters (Peel et al., 2007). The annual mean air temperature and precipitation are 16.4 °C and 1480 mm, respectively. Monthly mean air temperature reaches its maximum (28.0 °C) and minimum (4.7 °C) in July and January, respectively. About 60% of the annual precipitation occurs from May to September. Annual wind speed is over 2 m/s with northwest and southeast winds being dominant in winter and summer, respectively.

2.2. Data collection

Commercial trace gas instruments were utilized for air pollutant measurements at NUEORS. The O_3 analyzer (Model 49i, Thermo Fisher Scientific Inc., USA) operates based on the principle that O_3 molecules absorb ultraviolet (UV) light at the wavelength of 254 nm. The O_3 concentration is directly related to the absorbance of UV light. The lower detectable limit and precision of the instrument are 1 ppb. The CO analyzer (Model 48i, Thermo Fisher

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