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Nationwide ground-level ozone measurements in China suggest serious risks to forests☆

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ABSTRACT

We processed hourly ozone (O_3) concentrations collected in 2015 and in 2016 by a network of 1497 stations across China, with the main aim of assessing the risk that present ambient O_3 exposure is posing to Chinese forests. Our results indicate that the values of the metrics AOT40 (the accumulated hourly O_3 concentrations above 40 ppb during daylight hours) recommended as European Union standard, and W126 (the sum of weighted hourly concentrations from 8:00 to 20:00) recommended as USA standard for forest protection, exceeded the critical levels (5 ppm h across 6 months for AOT40 and 7–21 ppm h over 3 months for W126) on average by 5.1 and 1.2 times, respectively. N100 showed on average 65 annual exceedances of 100 ppb as hourly value. The 12-h and 24-h averages showed a small difference, suggesting high concentrations also at night. Risk was higher for the northern temperate climate than for the southern tropical and sub-tropical climates, and overall for the northern regions than for the southern regions. Higher risk occurred in the non-urban areas than in the urban areas in northern, south-west and north-west China, whereas risk was higher at urban areas in eastern and southern China. The overall results of this first nationwide assessment suggest a significant risk for forests over the entire China and warrant for urgent measures for controlling O_3 precursor emissions and establishing standards of protection.

1. Introduction

China is the most populous country in the world and has experienced explosive economic growth and profound social transformation since the 1970s; in the last decades, China became the larger emitter of air pollutants worldwide (Lefohn et al., 2017; Liu and Diamond, 2005; Liu et al., 2014; Quéré et al., 2015; Richter et al., 2005; Wang et al., 2017). Tropospheric ozone (O_3) is a strong oxidant gas pollutant, which is formed by reactions among primary pollutants, namely nitrogen oxides, volatile organic compounds, carbon monoxide and methane. Satellite measurements of tropospheric O_3 showed that concentrations over China increased by about 7% between 2005 and 2010, mostly due to a 21% rise in Chinese emission of precursors (Verstraeten et al., 2015). Ozone concentrations exceeding the ambient air quality standard of 100 ppb as hourly value (The Ministry of Environmental Protection of China, 2012) have been observed in China's major metropolitan agglomerations such as Jing-Jin-Ji, the Yangtze River delta, and the Pearl River delta (Wang et al., 2017). Such high O_3 concentrations are expected to cause harmful effects on agricultural crops, forest productivity and human health (Feng et al., 2014, 2015; Li et al., 2017; Paoletti et al., 2007a; Sicard et al., 2016a; Wang et al., 2017; Wittig et al., 2009).

Knowledge on forest responses to O_3 pollution in China is limited to the results of experimental studies (e.g. Li et al., 2016; Yuan et al., 2016; Zhang et al., 2012). To date, O_3 risk assessment

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to vegetation in China at nationwide level has been carried out by modelling crop responses (e.g. rice, Tang et al., 2013, 2014; wheat), as large-scale data from ground-level monitoring of O_3 and its precursors were not available in earlier years. In 2012, China adopted the new Ambient Air Quality Standard (The Ministry of Environmental Protection of China, 2012) and started regularly measuring hourly O_3 values at a large number of air quality stations distributed at urban and non-urban sites across the country.

Many O_3-exposure metrics are widely used in the European Union and United States to summarize risk to vegetation (Paoletti et al., 2007b; Lefohn et al., 2017). Such assessments in China from nationwide air monitoring networks are lacking although some pieces of information on AOT40 at some sites are now available (Tian et al., 2016; Lefohn et al., 2017). Here, we aggregated *in-situ* hourly O_3 concentrations from a nationwide network, with the main aim of assessing the risk that present ambient O_3 exposure is posing to Chinese forests. We selected the most widely used O_3 metrics to protect forests in North America and Europe (Paoletti et al., 2007b), i.e., the growing season average of hourly values (M24), the growing season average of daily 12 h (M12), the USA secondary standard for the protection of ecosystems (W126) and the EU standard for the protection of vegetation (AOT40, as adapted to forests by CLRTAP, 2015), as well as N100 (the number of 1-h concentrations exceeding 100 ppb over the growing season). Stomatal O_3 flux was not used because of missing stomatal conductance parameterization for local species and regional meteorological data like soil moisture, although it has been developed for O_3 risk assessments in Europe (LRTAP, 2015). We postulated that potential O_3 impacts varied depending on the site location because O_3 formation is seriously affected by local environmental conditions; in particular, it is stimulated by high air temperature, solar radiation, air layer mixing and air stagnation (Butkovic et al., 1990). At a regional scale, the surface O_3 concentration in China strongly differs by latitude and longitude (large range) and bio-geographical zone, e.g. the peak month of surface O_3 changed from October in the Pearl River Delta (Wang et al., 2009), to May in the Yangtze River Delta (Wang et al., 2001a), and to June in the North China Plain (Ding et al., 2008; Feng et al., 2015; Wang et al., 2011).

In addition, forest types differ in their responses to O_3 with evergreen broadleaf and conifer species usually considered more O_3 tolerant than deciduous broadleaf species (Anav et al., 2011; Calatayud et al., 2010; Li et al., 2016, 2017; Wittig et al., 2007, 2009). China's forests span over tropical, subtropical and temperate climate zones from south to north (Fang et al., 2001). Such broad climatic gradient supports diverse forest types ranging from evergreen broadleaf species in the southern tropical forests to deciduous broadleaf species in the central temperate forests to coniferous species in the northern boreal forests (Fig. 1). It is therefore of interest to assess O_3 risk in climatically-different zones of China. It is also well known that urban sites experience lower O_3 levels than nearby rural sites (Paoletti et al., 2014), in particular due to fast O_3 titration by nitrogen monoxide (Madronich, 2014; Querol et al., 2016; Sicard et al., 2016b). Similar patterns between urban and rural areas were also found in several Chinese cities (Tong et al., 2017; Xu et al., 2011), but a comprehensive assessment of the magnitude of this response across China and an assessment of the risk to forests are missing.

China has implemented several large-scale afforestation and forest conservation programs and has undergone rapid forest expansion since 1990s (Zhang et al., 2017), resulting in the largest annual net increment of forest area (FAO, 2010). In particular, China's growing economy associated with increasing demand for forest products has brought huge pressure on forests (Liu and Diamond, 2005; Xu and White, 2004) with impacts on the global economy (Hu et al., 2015b) and air quality (Verstraeten et al., 2015). The roles that forests play in climate change mitigation, biodiversity conservation, air and water quality provision and forest product production are increasingly awarded, and are attracting the global interest in O_3 impacts (Proietti et al., 2016; Sitch et al., 2007). Quantifying the potential impacts of O_3 on Chinese forests is thus of interest to policy makers and forest ecology worldwide.

Our specific aims were: (1) to assess the O_3 risk to: (i) urban and non-urban forests; (ii) temperate, sub-tropical and tropical forests; and (iii) different geographic regions of China; and (2) to discuss which metrics are the most suitable for the protection of forests in China.

2. Materials and methods

2.1. Database collection and categories

From 2013, the Ministry of Environmental Protection of China has adopted National Ambient Air Quality Standard released in 2012 (NAAQS-2012) and publicized hourly air quality monitoring data of six major pollutants ($PM_{2.5}$, PM_{10}, O_3, SO_2, NO_2 and CO) at each monitoring station in a national web platform (http://113.108.142.147:20035/emcpublish/). There are 1497 stations along with 190 cities since 2014. Due to download restrictions on the official Chinese urban air quality web platform, *PM25.in* as the third-party source was used following to Rohde and Muller (2015). *PM25.in* is a direct mirror of data from the 1497 stations in China's national network. In this paper, we collected hourly air monitoring data from January 2015 to December 2016 of all air quality monitoring stations across China (Table 1). All instruments were run, corrected and maintained by the third-party to assure the monitored data quality. Consistency, quality control, and validation checks were applied to the raw data prior to further analysis in order to reduce the impact of outliers, badly calibrated instruments, and other problems. Following a data quality control with selection of data below a cut-off threshold of 300 ppb and above 0 ppb, stations with a minimum data capture of 85% over the index time-window, as recommended by Querol et al. (2016), were processed (Table 1). Station distribution is shown in Fig. 1.

We divided the stations into three types of category: (i) urban and non-urban stations; (ii) temperate, subtropical and tropical climates; (iii) geographic zones, i.e., northern China (NC), northeast China (NE), eastern China (EC), southern China (SC), southwest China (SW) and north-west China (NW) (Fig. 1). To assign the stations to the urban or non-urban category, the boundaries of Chinese cities were defined according to China's official definition of their administrative areas (Chan, 2007). In this study, urban land was defined as areas dominated by human-made surfaces (e.g., roads and buildings), including residential, commercial, industrial, and transportation space within the administrative boundary, following the definitions and practices of previous studies (Zhao et al., 2016). Urban land can therefore be considered interchangeable with impervious surface or the built environment. Landsat Thematic Mapper remote sensing images covering China at 1:100,000-scale from the national land-use database for 2010 (Liu et al., 2014) were used to obtain the information on urban and non-urban land. The database was provided by Data Center for Resources and Environmental Sciences, Chinese Academy of Sciences (RESDC) (http://www.resdc.cn). It is worth noting that the non-urban category includes both suburban and rural areas.

To assign the stations to one of the three climatic zones of China, we calculated the Kira's warmth index (WI) (Fang and Li, 2002) as follows: WI (^{o}C month) = $\sum$ (t−5), Where t is > 5 °C average monthly temperature, obtained from World Climate database (www.worldclim.org) with a spatial resolution of 30 s (*c.*