Exposure Risk of Global Surface O₃ During the Boreal Spring Season

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Abstract

Surface ozone (O₃) is an oxidizing gaseous pollutant; long-term exposure to high O₃ concentrations adversely affects human health. Based on daily surface O₃ concentration data, the spatiotemporal characteristics of O₃ concentration, exposure risks, and driving meteorological factors in 347 cities and 10 major countries (China, Japan, India, South Korea, the United States, Poland, Spain, Germany, France, and the United Kingdom) worldwide were analyzed using the MAKESSENS model, Moran's *I* analysis, and Generalized additive model (GAM). The results indicated that: in the boreal spring season from 2015 to 2020, the global O₃ concentration exhibited an increasing trend at a rate of 0.6 µg/m³/year because of the volatile organic compounds (VOCs) and NO_x changes caused by human activities. Due to the lockdown policies after the outbreak of COVID-19, the average O₃ concentration worldwide showed an inverted U-shaped growth during the study period, increasing from 21.9 µg/m³ in 2015 to 27.3 µg/m³ in 2019, and finally decreasing to 25.9 µg/m³ in 2020. According to exposure analytical methods, approximately 6.32% of the population (31.73 million people) in the major countries analyzed reside in rapidly increasing O₃ concentrations. 6.53% of the population (32.75 million people) in the major countries were exposed to a low O₃ concentration growth environment. Thus, the continuous increase of O₃ concentration worldwide is an important factor leading to increasing threats to human health. Further we found that mean wind speed, maximum temperature, and relative humidity are the main factors that determine the change of O₃ concentration. Our research results are of great significance to the continued implementation of strict air quality policies and prevention of population hazards. However, due to data limitations, this research can only provide general trends in O₃ and human health, and more detailed research will be carried out in the follow-up.

Keywords O₃ · Spatiotemporal variation · Exposure risk · Global · GAM

Introduction

O₃ is a primary pollutant produced by photochemical reactions, and its formation is driven by precursor emissions, chemical conversion, and weather conditions (Streets et al. 2007; Jerrett et al. 2009). Moreover, surface O₃ exposure poses a severe potential risk to human health (Yin et al. 2017; Huang et al. 2018; Lin et al. 2019), and numerous studies have demonstrated a significant correlation between spatiotemporal changes in O₃ and its impact on human health (Wang et al. 2021). Under the global warming and emission scenario RCP8.5, acute excess mortality associated with O₃ will increase in the future (Chen et al. 2018). Therefore, exploring the change regularities and exposure risk of global surface O₃ is of great significance for the implementation of strategies to reduce the negative impacts associated with O₃ exposure.

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In the recent years, numerous studies have investigated the changes in O_3 concentrations on a spatial and temporal scale. However, researchers are mostly concerned about the risk of human and vegetation exposure to surface O_3 on the city scale, as well as the driving factors of O_3 . For example, Seltzer et al. (2020) explored the spatiotemporal characteristics of surface O_3 concentrations in the United States and estimated the population exposure risk of surface O_3 during the summer months. Xue et al. (2020) used satellite remote-sensing data to investigate the spatiotemporal trends of O_3 exposure from 2013 to 2017 and concluded that O_3 is becoming a crucial player in the burden of disease caused by air pollutants in China. Feng et al. (2019) assessed the exposure of China's population and vegetation (crops and forests) to O_3 pollution in 2015, and proposed that decision-makers should develop strategies for the protection of human and vegetation health from O_3 . Wang et al. (2020a) revealed that aerosols are one of the primary driving factors increasing atmospheric O_3 concentrations, which requires the reduction of O_3 precursor emissions to reduce O_3 concentrations. Jeong et al. (2020) analyzed the impact of meteorological factors on the annual O_3 variability in South Korea and proposed that the O_3 -meteorology relationship showed spatiotemporal differences depending on the topographical and emission distribution characteristics of each area.

Although current research on the spatiotemporal distribution of global surface O_3 and risk exposure assessments has achieved fruitful results, there are still the following shortcomings: First, owing to the limitations of global ground data accuracy and workload, previous studies have predominately focused on city-scale research, and changes in the O_3 concentration on a global, continental, and national scale have rarely been investigated. However, O_3 pollution is fluid and transregional; thus, it is necessary to conduct in-depth analyses on changes in O_3 concentrations and their development trends from a global perspective. Second, owing to the lack of surface O_3 monitoring data, most of the existing literature has used the O_3 concentration dataset generated by remote sensing inversion and other methods to for their

analyses. This type of data is grid data with a large coverage area, and its timing changes are easily limited by accuracy. Third, previous studies have focused exclusively on the driving factors and sources of O_3 ; however, in the context of societal and economical development, it is crucial to analyze the effects of exposure to O_3 pollution.

In this study, we used daily O_3 concentrations in the boreal spring season from 2015 to 2020 and socioeconomic data worldwide from 2015 to 2020 to conduct the following analyses: First, using the MAKESENS model, Moran's I analysis, and Hotspot analysis, we quantitatively estimated global spatiotemporal patterns of O_3 concentration changes in the boreal spring season from 2015 to 2020; Second, population exposed to surface O_3 concentrations in key countries around the global were analyzed; Third, the meteorological driving factors of O_3 were discussed based on these datasets.

Materials and Methods

Study Area

This research was conducted on a global scale, mainly focusing on five typical continents: Asia, Europe, North America, South America, and Oceania. Africa was not included in our research because previous studies have demonstrated less significant O_3 concentration changes for this continent compared to other global regions (Zhang et al. 2020; Klimont et al. 2017). According to the continent grouping provided by Natural Earth (<https://www.naturalearthdata.com>), our study area was divided into 11 areas (Fig. 1): Eastern Asia (EA), Southern Europe (SE), Northern North America (NN), Central South America (CS), Northern Europe (NE), Western Europe (WE), Western Asia (WA), Eastern Europe (EE), South-Central Asia (SA), Oceania (AO), and Southern South America (SS). In order to avoid too many groupings, we have carried out grouping induction according to the data attributes provided by Natural Earth. We grouped

Fig. 1 Spatial distribution map of 347 key cities and 11 main research areas explored in this paper

small categories into large categories. For example, New Caledonia and French Polynesia are grouped in Melanesia and Polynesia based on the data from Natural Earth respectively, but these two countries belong to Oceania, so we divide them into Oceania (AO). In addition, we selected ten major countries based on factors such as GDP, population, proportion of secondary industry, and degree of development, namely China, Japan, India, South Korea, the United States, Poland, Spain, Germany, France, and the United Kingdom, for our analyses. The selected Asian countries include China, Japan, South Korea, and India, all of which are among the most polluted counties in the world. The main cause of pollution in these counties is overpopulation. However, due to the air quality control policies of various countries, the impact of human activities on O₃ pollution has been reduced. Few counties like France, Germany, and Spain are the center of attracting tourists throughout the year. The main cause of air pollution in these counties is road traffic. The United States and the United Kingdom, as established countries with rapid economic development, are also indispensable for their exploration. As a highly industrialized country, Poland has heavy industry, manufacturing and high-polluting enterprises. The main source of air pollution is industrial activities, including production, import and export transportation, etc. In addition, there are certain differences in climatic conditions in these countries. For example, China is mostly located in the monsoon region, India has a tropical monsoon climate, the UK has a temperate maritime climate, and so on. More detailed information about these 10 countries can be found in Supplementary Table S1.

Data Sources

Daily O₃ concentration and meteorological data (dew point, relative humidity, precipitation, pressure, temperature, wind speed) from March 1 to May 31 of each year from 2015 to 2020 were obtained from the World Air Pollution: Real-time air quality index (WAQI project, <https://aqicn.org/>). These data were compiled from 12,000 ground-based monitoring stations spatially distributed over 1000 major cities in more than 100 countries worldwide (Supplementary Fig. S1). We preprocessed the collected stations data and mainly followed the following principles: First, we excluded from the raw data daily values ≤ 0 and missing values. Second, to calculate mean values, if monitoring data for a given month covered fewer than 27 days due to missing data, data for that station were excluded. Third, we removed abnormal values (> 1000 daily; Guo et al. 2017). Finally, we take the average of all the station data owned by each city, which is the daily O₃ concentration of that city. Generally, each city has 1–6 stations. Ultimately, we obtained O₃ concentration data for 347 cities (Fig. 1 and Supplementary Table S2), and carried out follow-up research on this basis. The population

distribution data for 2020 were obtained from the World Bank (<https://data.worldbank.org/indicator/>), to explore the effects of O₃ exposure among global populations.

Methods

Research Framework

This paper used the MAKESENS model, the spatial autocorrelation model, and the GAM method to analyze the O₃ concentration data in the boreal spring season from 2015 to 2020. First, we used the MAKESENS model to calculate the change trend of O₃ concentration; Second, we chose the spatial autocorrelation model to explore the spatiotemporal changes on the trend of O₃ concentration, including cold/hot spot analysis and cluster analysis; Third, we explored the populations of 10 major countries under different levels of O₃ concentration; Finally, we used the GAM model to discuss the impact of meteorological factors on the changes in O₃ concentration. Figure 2 shows the research framework of this paper.

Fig. 2 Research framework of this paper

MAKESENS model

The MAKESENS model is often used to detect and estimate change trends in interannual atmosphere and precipitation (Sarkar and Ali 2009; Ali et al. 2012). This model is based on the nonparametric Mann–Kendall test for the trend and the nonparametric Sen's method for the magnitude of the trend. The MAKESENS model does not assume the distribution of data; thus, outliers and missing values do not severely affect the model results (Partial and Kahya 2006). Therefore, we use the MAKESENS model to calculate the change trend of O₃ concentration during the study period.

For time series with less than 10 data points, the MAKESENS model uses the *S*-test, and for time series with 10 or more data points, the *Z*-test is used. Since the research period of this paper is in the boreal spring season from 2015 to 2020, this time series contain 6 data points. Thus, *S*-test statistics based on the Mann–Kendall test were calculated as follows:

$$S = \sum_{k=1}^{n-1} \sum_{j=k+1}^n \text{sgn}(x_j - x_k) \quad (1)$$

where x_j and x_k are the O₃ concentrations at years j and k respectively, $j > k$, and,

$$\text{sgn}(x_j - x_k) = \begin{cases} 1, & \text{if } x_j - x_k > 0 \\ 0, & \text{if } x_j - x_k = 0 \\ -1, & \text{if } x_j - x_k < 0 \end{cases} \quad (2)$$

The value of the *S*-statistics indicates the direction of the trend. A positive value of *S*-statistics indicates an increasing trend, whereas a negative value indicates a decreasing trend.

Sen's method is typically used to estimate the true slope of an existing trend. It is usually assumed that the trend is linear. To get the slope Q , we first calculate the slopes of all data value pairs.

$$Q_i = \frac{x_j - x_k}{j - k} \quad (3)$$

where $j > k$. x_j and x_k are the O₃ concentration at times j and k respectively. i represented each data value pair, $i = 1, 2, 3, \dots$

If there are n values x_j in the time series we get as many as $N = n(n-1)/2$ slope estimates Q_i . The Sen's estimator of slope is the median of these N values of Q_i . The N values of Q_i are ranked from the smallest to the largest and the Sen's estimator is.

$$Q = \frac{1}{2}(Q_{[N/2]} + Q_{[(N+2)/2]}), \text{ if } N \text{ is even} \quad (4)$$

$$Q = Q_{[(N+1)/2]}, \text{ if } N \text{ is odd} \quad (5)$$

More details about MAKESENS model can be found in Salmi et al. (2002).

Spatial Autocorrelation Analysis

Spatial autocorrelation refers to the potential interdependence of variables in the same distribution area (Shan et al. 2020). Global spatial autocorrelation reflects the general trend of the spatial autocorrelation of raw data in the entire study area. Generally, Global Moran's I is used as the measure index. The value range of Global Moran's I is $[-1, 1]$. At a certain significance level, there is a positive correlation if the Global Moran's I value is > 0 , which denotes a high-high clustering or low-low clustering, and a negative correlation if the Global Moran's I value is < 0 , which presents a spatial dispersion pattern. There is no spatial autocorrelation if the Global Moran's I is 0. A global spatial autocorrelation analysis is used to judge the aggregation trend of data, but the instability of the local space is not reflected (Song et al. 2020). To analyze spatial autocorrelation more accurately, we measured the local spatial autocorrelation to explore spatial heterogeneity. The Anseline Local Moran's I can distinguish spatial clustering with statistical significance, such as high-value clustering (hot spots) and low-value clustering (cold spots).

In this study, the Global Moran's I was used to characterize the spatial autocorrelation of the change trend of O₃ concentration based on 347 cities. This study used the Anseline Local Moran's I to explore the local spatial autocorrelation of the change trend of O₃ concentration. Then, we used a Hotspot analysis (Getis-Ord G_i^*) for the high-low clustering test. For the i spatial unit, the Global Moran's I and Local Moran's I were calculated as follows:

$$I_G = \frac{\sum_{i=1}^n \sum_{j=1}^n W_{ij}(x_i - \bar{x})(x_j - \bar{x})}{S^2 \sum_{i=1}^n \sum_{j=1}^n W_{ij}} \quad (6)$$

$$I_L = \frac{\sum_{i=1}^n \sum_{j=1}^n W_{ij}(x_i - \bar{x})(x_j - \bar{x})}{S^2} \quad (7)$$

$$S = \frac{1}{n} \sum_{i=1}^n (x_i - \bar{x})^2 \quad (8)$$

$$Z(I) = \frac{[I - E(I)]}{\sqrt{V[I]}} \quad (9)$$

$$E[I] = -1/(n-1) \quad (10)$$

$$V[I] = E[I^2] - E[I]^2 \quad (11)$$

where n is the number of spatial units (347 cities in this study), x_i and x_j are the change trend of O₃ concentrations of spatial units i and j , respectively, and $\bar{x}$ is the average change trend of O₃ concentration of all the units. W_{ij} is the spatial weight matrix of the units i and j . W_{ij} value of 1 indicates a common edge between spatial units i and j , otherwise $W_{ij}=0$. For Moran's I , the standardized statistic $Z(I)$ can be used to test whether there is a spatial autocorrelation relationship. The value of Moran's I ranges between $[-1, 1]$ and the value of $Z(I)$ is between $[-1.96, 1.96]$. At a significance level of 0.05, four different spatial autocorrelation clustering relationship types between Moran's I value and $Z(I)$ can be obtained (Zhou et al. 2019; Zhao et al. 2020): If Moran's $I > 0$ and $Z(I) > 1.96$, the correlation is an HH type, which means that the increasing trend of O₃ concentration of this unit and neighboring units are higher than the average; thus, these areas are "hot spot" areas. If Moran's $I > 0$ and $Z(I) < -1.96$, the correlation is an LL type, which indicates that the increasing trend of O₃ concentration of this unit and neighboring units are lower than average, and these areas are "cold spot" areas. If Moran's $I < 0$ and $Z(I) > 1.96$, the correlation is an HL type, suggesting that the cell with a high increasing trend of O₃ concentration is surrounded by a cell with low increasing trend of O₃ concentration. If Moran's $I < 0$ and $Z(I) < -1.96$, the relationship is an LH type, meaning that the cell with a low increasing trend of O₃ concentration is surrounded by a cell with a high increasing trend of O₃ concentration.

Exposure Analytical Methods

The average O₃ concentration in each city is calculated by averaging the concentration of all air quality monitoring stations in a city. Based on the average O₃ concentration of 347 cities in the boreal spring season from 2015 to 2020, the change trend of O₃ concentration of each city is calculated. The natural breakpoint method (Yao et al. 2020) is used to divide all the change trend of O₃ concentration into four levels: extreme, strong, moderate, weak. According to the change trend of O₃ concentration, each city can be classified into these four levels. The population of cities at different levels will be classified as the exposed population of that level. The cities are aggregated to determine the number of exposed populations of different levels in each country (Guo et al. 2017).

GAM method

Meteorological factors are important driving factors affecting O₃ concentration and its change trend (Hu et al. 2021). Therefore, we believe that it is necessary to explore the

driving meteorological factors of changes in O₃ concentration. Generalized additive model (GAM) is a flexible free regression model. By controlling the influence of confounding factors on the research object, we analyzed the complex nonlinear relationship between the response variables and other explanatory variables (Wood and Augustin 2002). It is more flexible in exploring the relationship between response variables and explanatory variables, and its results have a higher reliability (Gong et al. 2018). The general form of the GAM model is as follows:

$$Y = \beta_0 + \varphi_1(x_1) + \varphi_2(x_2) + \varphi_3(x_3) + \dots + \varphi_n(x_n) + \varepsilon \quad (12)$$

where Y is the dependent variable; $x_1, x_2, \dots, x_n$ are the explanatory variables of Y ; β_0 is the intercept; $\varphi_1, \varphi_2, \dots, \varphi_n$ are the smoothing functions of the explanatory variable; ε is the residual.

Six meteorological factors were selected as explanatory variables: mean dew point (DEWP, °F), relative humidity (Rel.hum, %), precipitation (PRCP, inches), mean pressure (P , hPa), maximum temperature ($T_{\max}$, °F), and mean wind speed (WDSP, knots), and the O₃ concentration was selected as the response variable to construct a basic model. Moreover, F -statistics calculated by the GAM model were used to rank the importance of each meteorological factor. F -statistics comprehensively reflect the degree of freedom (e.d.f.) and the p value of each variable. In general, the larger the F -statistic, the more important the meteorological factor (Yang et al. 2019).

Results and Discussion

Spatiotemporal Patterns and Variations in O₃

To reduce the cross-regional linkage pollution of cities worldwide, this study explored the temporal and spatial variation characteristics of the O₃ distribution by analyzing the dynamic evolution process of O₃ in the boreal spring season from 2015 to 2020 (Figs. 3 and 4). In general, the annual O₃ concentrations showed distinct spatial patterns and strong variations across the whole study area. At the regional scale, areas with high O₃ concentrations (average concentration $> 20 \mu\text{g}/\text{m}^3$) are mainly located in EA, entire Europe, and NN, which are characterized by intensive human activities and well-developed economies. Conversely, low O₃ concentrations (average concentration $< 10 \mu\text{g}/\text{m}^3$) were predominately distributed in the AO and SS regions (Fig. 4).

On a temporal scale, the global average O₃ concentration in the boreal spring season was $21.9 \mu\text{g}/\text{m}^3$ in 2015 and $25.9 \mu\text{g}/\text{m}^3$ in 2020. That is, the annual average concentration

Fig. 3 Spatiotemporal patterns of global annual O_3 concentrations in the boreal spring season from 2015 to 2020

Fig. 4 Interannual variations in annual averaged O_3 in each region worldwide. Where GL is represented the globe average data

of O_3 worldwide showed a slight increase rate of $0.6 \mu\text{g}/\text{m}^3/\text{year}$. The gradual increase in O_3 concentration around the globe was also confirmed in other studies (Finch and Palmer 2020; Lu et al. 2020). Previous studies have shown that O_3 is a secondary pollutant that depends on its precursor emissions (He et al. 2021). Therefore, we hold that the increase in O_3 concentration is inseparable from the change in the content of its precursors. On the urban scale, the formation of O_3 was limited by the VOCs– NO_x ratio (Sillman and He 2002). There are characteristics of high O_3 with low NO_x concentrations in urban areas, especially in cities where the

economy is developed and people live in abundance. For example, Li et al. (2013) found that when the O_3 formation regime was limited to volatile organic compounds (VOCs), NO_x reduction increased the mean O_3 concentration. That is, the reduction of VOCs emissions hindered the formation of O_3 , whereas the reduction of NO_x emissions promoted the formation of O_3 . Over the past years, the reduction of O_3 precursor emissions has been insufficient to shift from VOCs-limited to NO_x -limited conditions, leading to an increase in O_3 concentrations in cities because of the emission of vehicles (Sicard, 2021). Therefore, an effective strategy to reduce the O_3 concentration is to reduce VOCs emissions and control NO_x emissions (Oak et al. 2019).

Specifically, the annual average O_3 concentration in the study area showed an inverted U-shaped growth during this period. The lowest O_3 concentrations were observed in the boreal spring season of 2015, with an average of $21.9 \mu\text{g}/\text{m}^3$, and a peak O_3 concentration of $27.3 \mu\text{g}/\text{m}^3$ was recorded in 2019, resulting in an increasing number of people being exposed to O_3 pollution. Afterward, the O_3 concentration decreased slightly, with an average concentration of $25.9 \mu\text{g}/\text{m}^3$ recorded in 2020. The decrease in the O_3 concentration from 2019 to 2020 can be explained by the COVID-19 outbreak at the end of 2019. It can be seen that numerous countries worldwide experienced lockdown during this period, and the restriction on human activities reduced man-made

Fig. 5 Interannual trends in annual averaged O₃ in each region worldwide. Where, GL is represented the globe average data

emissions. These phenomena indicate that the strict lockdown measures reduced NO_x and VOCs emissions in the studied cities, contributing to low VOCs–NO_x ratios, and therefore lower O₃ concentrations. Therefore, global O₃ pollution in 2020 improved to a certain extent (He et al. 2021; Zhang et al. 2020).

Spatial Difference of O₃ Change Trend

Spatial distributions of the interannual trends global O₃ concentrations in the boreal spring season from 2015 to 2020 are shown in Figs. 5 and 6. In this study, the MAKESENS model was used to calculate changes in the O₃ concentration in the boreal spring season from 2015 to 2020. The O₃ concentration for the most of the regions shows significant increasing trends, particularly EA, WE, and NN (Figs. 5 and 6). According to the analysis of O₃ concentration pattern before, these regions not only have high O₃ concentration, but also have a fast growth rate. The significant upward O₃ trends can be attributed to a lower titration of O₃ by NO due to the reduction in NO_x emissions from road transport following the implementation of stringent vehicle emission standards in these regions (Wang et al. 2020b; Seo et al. 2018). Only a few areas showed a downward trend or no obvious changes.

We calculated the proportion of cities with increasing O₃ concentration trends in the 347 cities, which is approximately 83.86%. That is, among the 347 cities studied, 291 cities showed an increase in the O₃ concentration during the study period. Among them, a total of 157 cities had an average O₃ concentration growth rate of 0–1 µg/m³/year. A total of 111 cities had an O₃ concentration growth rate of 1–2 µg/m³/year, and 22 cities had an O₃ concentration growth rate of 2–4 µg/m³/year. Only one city (Harbin, China) had an O₃ concentration growth rate of > 4 µg/m³/year. This can be attributed to the high heterogeneity of land use types in Harbin. The higher rate of increase in O₃ concentration may be due to higher vegetation coverage, which emission release

Fig. 6 **a** the spatiotemporal distribution of annual changes in the global O₃ concentration in the boreal spring season from 2015 to 2020, **b** the significance level of these changes

higher concentrations of VOCs which were precursor pollutants of O₃ (Li et al. 2020).

From a regional perspective, the growth trend varied between the regions, with significant spatial differences (Figs. 5 and 6). The CS region showed the most significant O₃ concentration growth trend, with an average annual growth of more than 2.0 µg/m³/year. Moreover, a significantly negative trend was observed for SA (− 1.2 µg/m³/year). Other regions that recorded significant increases in the O₃ concentrations during the study period included NE (1.3 µg/m³/year), WE (1.0 µg/m³/year), EA (0.9 µg/m³/year), SE (0.8 µg/m³/year), EE (0.7 µg/m³/year), SS (0.4 µg/m³/year), NN (0.4 µg/m³/year), WA (0.3 µg/m³/year), and AO (0.0 µg/m³/year). We further analyzed why the O₃ concentration change trend only showing a downward trend in SA. SA mainly contains three countries: India, Thailand, and Singapore. These three countries include ten cities: Delhi, Lampang, Bangkok, Seltzer nai, Chiang Mai, Rayong, Chon

Buri, Hyderabad, Lucknow, and Singapore. With the exception of Delhi, O_3 concentrations have shown a downward trend in other cities. The climate of these cities is affected by the northeast monsoon in winter, and the northeast wind will bring dryness, breeze, and poor air quality to these cities in winter. In spring, the monsoon begins to turn, and the southwest monsoon from the Indian Ocean and South China Sea will bring warm, humid, and unstable air masses. Compared with winter, these cities will usher in more precipitation and air quality will also improve (Janjai et al. 2016). Moreover, different countries have formulated a series of prevention and control policies for air pollution. For example, the Central Pollution Control Board (CPCB) of India has established a national environmental air quality standard to minimize the health-related pollution risks of the population (CPCB 2010). However, due to the rapid economic growth in Delhi, India, the number of vehicles per thousand in the population increased considerably from 317 in 2006 to 598 in 2018 (Tiwari et al. 2015). Thus, the surface O_3 concentration in Delhi often exceeds the standard. This also shows that the

influence of human factors on changes in O_3 concentration is far greater than natural factors.

Figure 7 indicates trends in the global O_3 concentration in ten countries in the boreal spring season from 2015 to 2020. The average O_3 concentration of each country in the boreal spring season from 2015 to 2020 was: Japan ($32.4 \mu\text{g}/\text{m}^3$), Korea ($29.3 \mu\text{g}/\text{m}^3$), China ($29.3 \mu\text{g}/\text{m}^3$), France ($26.1 \mu\text{g}/\text{m}^3$), Spain ($26.0 \mu\text{g}/\text{m}^3$), US ($24.8 \mu\text{g}/\text{m}^3$), Germany ($24.4 \mu\text{g}/\text{m}^3$), UK ($23.8 \mu\text{g}/\text{m}^3$), Poland ($23.6 \mu\text{g}/\text{m}^3$), and India ($14.6 \mu\text{g}/\text{m}^3$). All the countries except for India showed an increasing O_3 concentration trend, whereas India recorded a decrease of $1.1 \mu\text{g}/\text{m}^3/\text{year}$ for the study period. Moreover, the UK recorded the highest growth rate of O_3 , reaching $1.4 \mu\text{g}/\text{m}^3/\text{year}$, followed by Korea and France, with a growth rate $> 1.0 \mu\text{g}/\text{m}^3/\text{year}$. In fact, various countries have formulated certain policies for air pollution. In the past period, the emission control strategies made by government have focused more on NO_x rather than VOCs. And these policies were insufficient to shift the chemical regimes from VOCs-limited to

Fig. 7 O_3 concentration and its trend in the 10 major countries analyzed in the boreal spring season from 2015 to 2020

NO_x-limited conditions, leading to O₃ formation in countries due to reduced O₃ titration by NO (Sicard 2021). The increase in O₃ concentration observed in South Korea is mainly due to the increase in VOCs and NO_x emissions since 2010 (Seo et al. 2014). As a result, continuing NO_x controls can reduce O₃ levels.

Spatial Clustering Pattern of the O₃ Change Trend

Figure 8a shows the distribution of the global autocorrelation significance test results. By calculating the Global Moran's *I* index, it was determined that the Global Moran's *I* index was positive during the study period and passed the significance test of 0.01 ($p=0.00$). This indicates that the variation in the O₃ concentration in the study area had an

enhanced positive spatial autocorrelation and showed an obvious aggregation state.

Figure 8b and c show the results of the Hotspot analysis and local spatial autocorrelation analysis. The results revealed two significant hot spots and two significant cold spots based on global O₃ concentration changes. Hot spots (i.e., high-value concentration areas (HH) of O₃ concentration growth rate) were mainly distributed in SE, EA, and the coastal areas of NN, which are characterized by the most severe O₃ pollution. Cold spots (i.e., low-value concentration areas (LL) of O₃ concentration growth rate) were predominately distributed in SS and SA, where O₃ pollution is relatively low. This is similar to the spatial agglomeration pattern of PM_{2.5} concentration around the globe (Yang et al. 2021). Li et al. (2019) believed that an important factor for the aggravation of O₃ pollution is the decrease of

Fig. 8 Spatial clustering characteristics of O₃ concentration changes in each city-level unit in the boreal spring season from 2015 to 2020. **a** Result of spatial autocorrelation test (Moran's *I* Index), **b** spatial

distribution of cold and hot spots of O₃ concentration changes, and **c** spatial distribution of spatial clusters of the changes in O₃ concentration

PM_{2.5} concentration, because a sharp reduction of PM_{2.5} will cause the heterogeneous absorption of O₃ precursors, which will further aggravate the photochemical reaction of O₃. In addition, the sub-figures of Fig. 8c shows that sporadically appear as high-low (HL) clusters were observed in hot spots areas, suggesting that cities with high O₃ concentration growth rates are surrounded by cities with low O₃ concentration growth rates, such as Chongqing, Kunming, Harbin, and Beijing in China. In other words, although the overall O₃ concentration growth rate in hotspots is relatively high, there are still some cities that have higher O₃ concentration growth rates than the surrounding cities, thus forming a pattern of high-low (HL) clusters. We found that most of these cities are provincial capitals with rapid economic development and high population density in China. We believe that these provincial capitals need more winter coal and biomass combustion than surrounding areas (Wang et al. 2014), which has caused a rapid increase in O₃ concentration. However, the current changes in O₃ concentration are caused by a variety of factors, and the analysis of meteorological elements is also indispensable.

The Human Health Risk Exposure to O₃

The sustainable development goals (SDGs) propose to comprehensively control air pollution and strive to reduce the impact of environmental pollution on health (United Nations General Assembly, 2015). The third goal of the SDGs is good health and well-being, ensured by regular testing of the environmental quality. Long-term exposure to O₃ pollution has become a major global public health problem affecting human health (Zhang et al. 2019). Exposure to O₃ for

18–20 h can change the permeability of lung epithelial cells, and the mucociliary tissue of the lung is also stimulated by O₃. These effects increase the susceptibility to respiratory bacterial infections (Karthik et al. 2017). Therefore, there is an urgent need to explore the degree of population exposure to O₃ pollution.

The exposed population is calculated based on the population and O₃ concentration in each year. Thus, we explored the relationship between O₃ and the population in the boreal spring season from 2015 to 2020 (Fig. 9a). The outer circle is the horizontal axis, and the radius is the vertical axis, which are shown sequentially from the outside to the inside: First, the distribution relationship between population (y-value) and O₃ (x-value) in each year; Second, an O₃ data histogram distribution; Third, we selected O₃ data as the x-value and population data as the y-value to rank O₃ in descending order; Fourth, the center line segment from thin to thick indicates that the average O₃ concentration increases, and vice versa. Analyzing from the outer circle to the inner circle, it can be found that: (1) the O₃ concentration in each year ranged from 10–40 µg/m³, and there is no absolute linear relationship between the O₃ concentration and the population size; (2) most of the O₃ data used in this article are normally distributed; (3) when the O₃ concentration was between 30 and 40 µg/m³, the population is the largest; (4) The annual average O₃ concentration only declined from 2019 (27.34 µg/m³) to 2020 (25.86 µg/m³). In the remaining years, the average O₃ concentration was on the rise. In the boreal spring season of 2015, the average O₃ concentration was 21.88 µg/m³, which increased by 3.98 µg/m³ in 2020.

Population exposure risk can be measured by the number of people exposed to various types of O₃ concentrations.

Fig. 9 **a** Relationship between population and O₃ in 2015–2020 in the boreal spring season. **b** Schematic diagram showing the increase trend of the four types of exposure (extreme, strong, moderate, and

weak) of O₃ concentration and the corresponding population. The numbers in brackets indicate the proportion of population at each level

Even if the exposure time is the same for different groups, there are certain risk differences for various O₃ concentrations. Using the natural breakpoint method to classify the growth trend of the O₃ concentration to explore the proportion of the population at potential exposure risk, we divided the exposure severity into four categories: extreme (1.88–5.31 $\mu\text{g}/\text{m}^3/\text{year}$), strong (0.69–1.88 $\mu\text{g}/\text{m}^3/\text{year}$), moderate (−0.92–0.69 $\mu\text{g}/\text{m}^3/\text{year}$), and weak (−4.17–0.92 $\mu\text{g}/\text{m}^3/\text{year}$) (Fig. 8b). Overall, in the boreal spring season from 2015 to 2020, the O₃ concentration in the environment where 31.73 million people live in key cities in the world has increased at the extreme level, accounting for approximately 6.32% population. 87.15% of the population (437.26 million people) live in an environment where the O₃ concentration increases between −0.92 and 1.88 $\mu\text{g}/\text{m}^3/\text{year}$. 6.53% of the population (32.75 million people) were exposed to a low O₃ concentration growth environment. However, as the O₃ concentration increases year by year, the number of people exposed to high concentrations of O₃ will continue to increase. We need to establish more stringent air pollution standards to prevent O₃ pollution from causing more and more damage to human health.

Combined with the spatial distribution of population density in the study area (Fig. 10a), it can be seen that the population density in EA, SA, and WE is higher. In general, the greater the population density, the higher the population exposed to O₃ pollution. It is therefore necessary to classify populations with different concentrations of O₃ pollution. Figure 10b shows the population at different levels of O₃ change trends in each country in the boreal spring season from 2015 to 2020. China has the largest population exposed to O₃ at the extreme level, up to 22.89 million. Moreover, Germany and the United Kingdom have > 1 million people exposed to O₃ at the extreme level, followed by Poland (0.87 million), South Korea (0.46 million), Spain (0.34 million), Japan (0.31 million), and France (0.11 million). India and the United States did not have any people exposed at the extreme level. The top three countries exposed at the strong level are China, South Korea, and Japan. China (13.55 million) and India (6.80 million) had more people exposed to O₃ at a weak level, far exceeding those of other countries. Among them, Japan, the United States, Poland, Spain, Germany, France, and the United Kingdom had zero exposed people at weak level. In short, the exposed population at each level is the largest in China.

Fig. 10 Distribution of the global population and number of exposed populations at different levels in 10 key countries. **a** World population distribution map; and **b** histograms of population numbers at different levels (weak, moderate, strong, and extreme) in the 10 key countries

As the country with the highest global population, China, with its rapid economic development, ranks first in terms of the number of people exposed to all levels of O₃. Moreover, researchers have reported that the number of people determines the O₃ pollution and its changing trend to a large extent (Li et al. 2020; Wang et al. 2020a). However, it is interesting that India, also with an exceptionally high population, recorded a significantly lower population exposure to O₃ at the extreme level compared to Germany and the United Kingdom. This observation indicates that the population is not the only factor that influences the O₃ concentration (Escudero et al. 2014; Kim et al. 2018). Meteorological factors will affect the formation of O₃ precursors, which in turn affect the changes in O₃ concentration. Compared with SA and NA, countries in EA and Europe face higher levels of exposure risk. The extreme and strong risk levels were observed in areas with relatively high population numbers and O₃ concentration growth rates. Moreover, the results exhibit strong spatial aggregation, which is consistent with previous analyses of spatiotemporal O₃ patterns.

Driving Meteorological Factors and the Spatial Effects on O₃

To determine the main meteorological factors of O₃ concentration change in 347 cities in the world, we statistically analyzed the *F*-statistics and significance of various meteorological factors in the GAM model (Fig. 11). Throughout the study period, 347 cities around the world have significant correlations with meteorological conditions. Many studies have proposed that climate conditions are major impact force for day-to-day variations of O₃ (Zhao et al. 2016; Wang et al. 2017). From a regional perspective, mean wind speed was the most important meteorological condition in WE and SE, with *F*-statistics of 32.65 and 30.20, respectively. These two values far exceeded the other meteorological factors affecting these regions. Relative humidity (31.59) and mean pressure (22.61) were the main factors affecting the change in O₃ in NE. Mean dew point and mean wind speed were the two main factors affecting the changes in O₃ concentrations in EA and AO. SA was mainly affected by two meteorological factors: mean wind speed (9.61) and precipitation (7.35). During the entire study period, mean wind speed (30.20) and maximum temperature (11.14) were the main meteorological factors affecting the change in O₃ concentration in SE. From a national perspective, China, Germany, and France were mainly affected by maximum temperature and mean wind speed. Mean wind speed and relative humidity were the two major factors affecting the United States, Poland, and Spain (Supplementary Table S3).

Many studies have proposed that meteorological conditions are the main factors affecting the change in O₃ concentration (Cao and Yin 2020; Adhikari and Yin 2020).

The results of the GAM model in the 10 countries selected for analysis indicated that maximum temperature, mean wind speed, and relative humidity play an important role in increasing the O₃ concentration. In addition, the O₃ concentrations in all 10 countries displayed significant correlations with meteorological conditions. For example, as the temperature continues to rise, the O₃ concentrations is rising in Europe (Hůnová et al. 2019) and China (Sun et al. 2019). That is, O₃ concentration increases with temperature. Moreover, significantly negative correlations between relative humidity and the O₃ concentration were observed in the countries during the study period. We found non-linear declines in the O₃ concentrations with increasing relative humidity in China, and this finding was consistent with those of studies conducted in Europe (Mertens et al. 2019; Ordóñez et al. 2020). Specifically, as other variables were controlled, high relative humidity results in a reduction in O₃ and its precursors. As discussed above, O₃, as a secondary pollutant, cannot be formed without VOCs and NO_x. According to previous studies, VOCs and NO_x monitoring also exist in surface water and groundwater (Li et al. 2014, 2018). Evaporation is an important process that affects groundwater chemistry. Therefore, whether organic substances in water can volatilize into the air and affect the formation and change of O₃ needs to be further considered (Li et al. 2016, 2017; Mthembu et al. 2020).

Our research results have revealed the impact of O₃ pollution on human health to a certain extent, and attention should be paid to the issue of O₃ pollution in the future. However, this study also has certain limitations and deficiencies: First, due to the large scope of the research and the difficulty of data collection, one of the main limiting factors of this study is that the amount of data used to explore global changes in O₃ concentration may not be sufficient. Therefore, the calculation of changes in O₃ concentration in this paper can only reflect certain trends. Second, the selection of the population exposure model is still to be considered. We only used a simple analysis model, and follow-up studies can use multiple models for comprehensive comparison. Last, we have considered meteorological factors such as precipitation, maximum temperature, and mean dew point, but there are other meteorological factors that affect the changes in O₃ concentration, such as visibility. Subsequent studies should make more in-depth and detailed discussions. Meanwhile, we hope that the authorities can implement more stringent air quality policies and emission control strategies to improve environmental conditions and human health.

Conclusions

In this study, we analyzed the spatiotemporal variation of O₃ concentration and its change trend in 347 cities and 10 key countries around the globe in the boreal spring season from

Fig. 11 F -statistics of dominant meteorological factors and their p value affecting O₃ concentration in each city during the study period

2015 to 2020. We estimated the number of people exposed to O₃ at different levels, and discussed the effects of meteorological factors on the O₃ concentration in these regions. The preliminary conclusions are as follows:

First, in the boreal spring season from 2015 to 2020, the average concentration of O₃ worldwide showed a slight increase rate of 0.6 $\mu\text{g}/\text{m}^3/\text{year}$. The world average O₃ concentration increased from 21.9 $\mu\text{g}/\text{m}^3$ in the boreal

spring season of 2015 to $27.3 \mu\text{g}/\text{m}^3$ in 2019, and finally decreased to $25.9 \mu\text{g}/\text{m}^3$ in 2020, showing an inverted U-shaped growth. The O_3 concentration in most parts of the world showed a significant upward trend, particularly in EA, WE, and NN. The results show that the O_3 concentration around the globe is rising year by year, and most regions are threatened by O_3 pollution.

Second, in the boreal spring season from 2015 to 2020, most of the population in major global cities were exposed to O_3 concentrations of $30\text{--}40 \mu\text{g}/\text{m}^3$. Among them, 31.73 million people (approximately 6.32% of the total population) were exposed to O_3 concentrations at the extreme level, and 32.75 million people (6.53%) were exposed to O_3 concentrations at the weak level. It is not that the larger the total population of each country, the greater the number of exposed populations at all levels. It shows that the O_3 concentration is not directly proportional to the number of total population. The concentration of O_3 is mainly affected by its precursors, and meteorological factors can often affect the changes of the precursors.

Third, according to the *F*-statistics results, maximum temperature, mean wind speed, and relative humidity are the most important meteorological factors affecting the O_3 concentration. As the temperature rises, the O_3 concentration gradually rises. However, relative humidity is inversely proportional to O_3 concentration.

To sum up, with the gradual increase in O_3 concentration, the population in all regions of the world is suffering from O_3 pollution. The responsible government departments need to improve and strengthen the monitoring of O_3 pollution to ensure that the emissions of O_3 precursors (VOCs, NO_x) comply with government regulations. In addition, various countries should conduct in-depth research on the meteorological factors that affect the changes in O_3 concentration, and explore the sustainable development path to reduce O_3 concentration.

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Data Availability The data used to support the results of this research are shown in the manuscript and available from the corresponding author upon request.

Declarations

Conflict of interest All authors have no conflicts of interests to declare.

Ethical Approval All authors agree to participate.

Consent to Participate All authors agree to participate.

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