Environ Geochem Health
https://doi.org/10.1007/s10653-021-00931-0

ORIGINAL PAPER

Effects of air pollutant emission on the prevalence of respiratory and circulatory system diseases in Linyi, China

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Received: 15 July 2020 / Accepted: 3 April 2021
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Abstract As a typical industrial city, Linyi has suffered severe atmospheric pollution in recent years. Meanwhile, a high incidence of respiratory and circulatory diseases has been observed in Linyi. The relationship between air pollutants and the prevalence of respiratory and circulatory system diseases in Linyi is still unclear, and therefore, there is an urgent need to assess the human health risks associated with air pollutants. In this study, the number of outpatient visits and spatial distribution of respiratory and circulatory diseases were first investigated. To clarify the correlation between diseases and air pollutant emissions, the residential intake fraction (IF) of air pollutants was calculated. The results showed that circulatory and respiratory diseases accounted for 62.32% of the total causes of death in 2015. The incidence of respiratory diseases was high in the winter, and outpatient visits were observed for more males (60.9%) than females (39.1%). The spatial distribution suggested that outpatient visits for respiratory and circulatory diseases were concentrated in the main urban area of Linyi, including the Hedong District, Lanshan District, and Luozhuang District, and especially at the junction of these three areas. After calculating the IF combined with the characteristics of pollution sources, meteorological conditions, and population data, a high IF value was concentrated in urban and suburban areas, which was consistent with the high incidence of diseases. Moreover, high R values and a significant correlation ($R > 0.6$, $p < 0.05$) between outpatient visits and residential IF of air pollutants imply similar spatial distributions of outpatient visits and IF value of residents. The spatial similarity of air pollution and outpatient visits

Supplementary Information The online version contains supplementary material available at https://doi.org/10.1007/s10653-021-00931-0.

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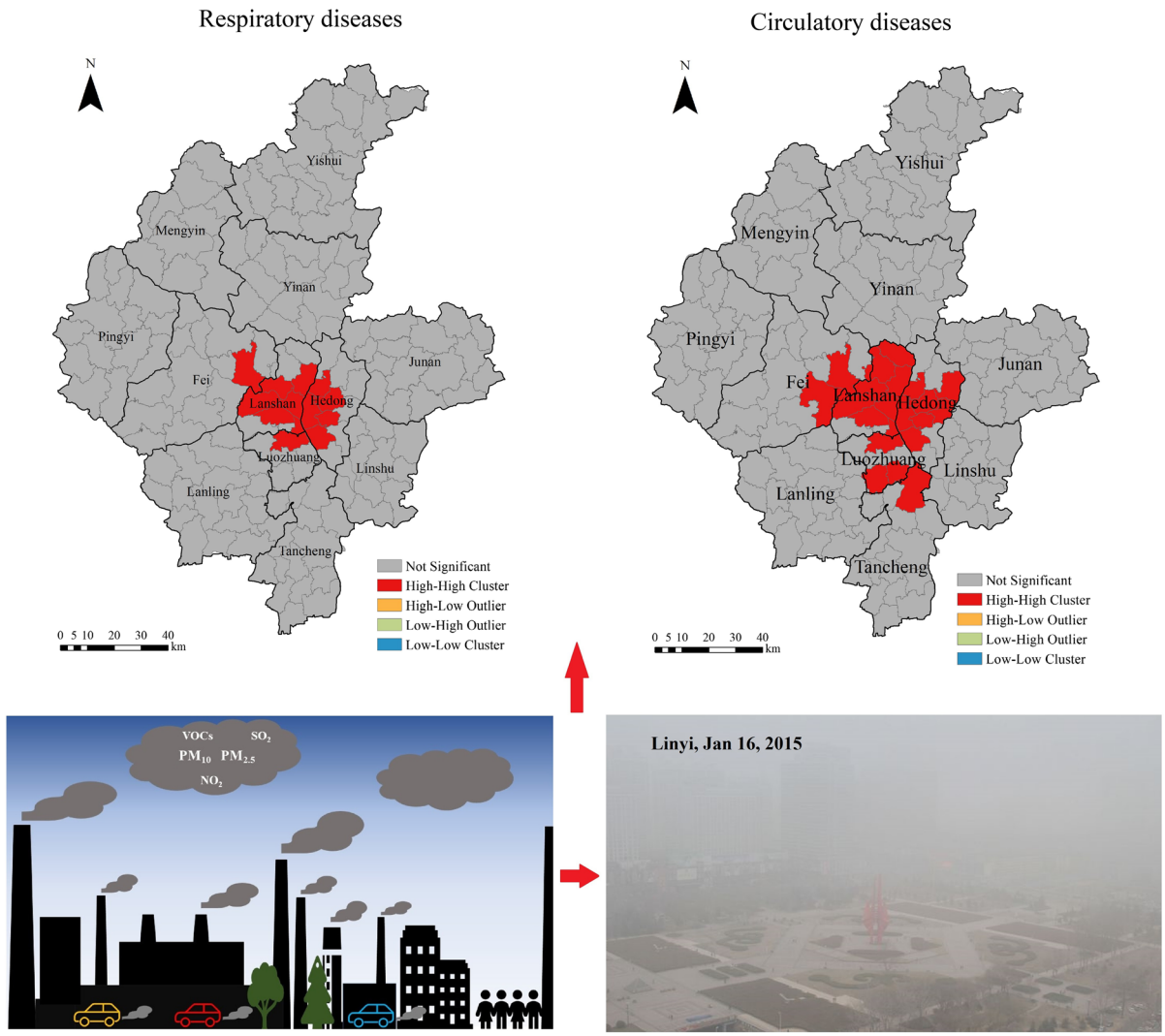
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Published online: 23 April 2021

suggested that future air pollution control policies should better reflect the health risks of spatial hotspots. This study can provide a potentially important reference for environmental management and air pollution-related health interventions.

Graphical abstract



Keywords Respiratory diseases · Circulatory system · Pollutants emission · Air quality · Intake fraction (IF)

Introduction

Rapid industrial development has led to frequent air pollution in China (Liu et al., 2017; Yao et al., 2019). A series of measures for the prevention and control of particulate pollution have achieved remarkable results

in recent years, and the concentrations of particulate matter in key regions of China, such as the Beijing-Tianjin-Hebei, the Yangtze River Delta, the Pearl River Delta have been significantly reduced year by year (Zhang et al., 2019; Tang et al., 2019; Zhai, 2019). However, heavy polluting processes still existed in this region and the number of premature deaths due to $PM_{2.5}$ pollution has still increased (Yue et al., 2020). Air pollutants can cause regional air pollution through diffusion and transmission (Ostro, 1994; Alberini et al., 1997; Steven, 2006; Coker & Kizito, 2018). Numerous epidemiological studies have shown that regional air pollution is a key factor affecting the morbidity and mortality of respiratory and cardiovascular diseases (Li et al., 2018; Liu et al., 2019; Sarizadeh et al., 2020; Yang et al., 2019). Exposure to air pollution can lead to adverse health effects, and air pollution has contributed to 1.6 million premature deaths every year in China (Maji et al., 2018; Rohde & Muller, 2015). Exposure to low levels of particulate matter also increases the risk of mortality (Yue et al., 2020). Therefore, the relationship between pollution and human disease needs to be studied continuously.

Linyi is located in the southeast of Shandong Province, China (Fig. 1). The administrative area includes nine counties and three districts, with a total population of 10,810,000 and a total area of 17,200 square kilometers. Both the population and area rank first in Shandong Province (Zhao et al., 2017). Recently, Linyi has witnessed rapid economic development, increasing energy consumption and severe air pollution (Fig. S1). In 2015, the Ministry of Environmental Protection of the People's Republic of China (MEP) supervised the air pollution in Linyi (MEP, 2015a, b, c). The increasingly prominent air pollution problem seriously affects the local residential's health and the city's sustainable development. Moreover, the incidences of respiratory and circulatory system diseases in Linyi have been increasing in recent years (Li et al., 2020). Numerous researchers have studied the emission inventory, air pollution and human health in the cities of Shandong Province, China (Gao et al., 2016; Xiong et al., 2016; Fan & Wang, 2016; Wang et al., 2005; Zhang et al., 2008; Maji et al., 2018). However, air pollution emissions and human health in Linyi have not been comprehensively investigated.

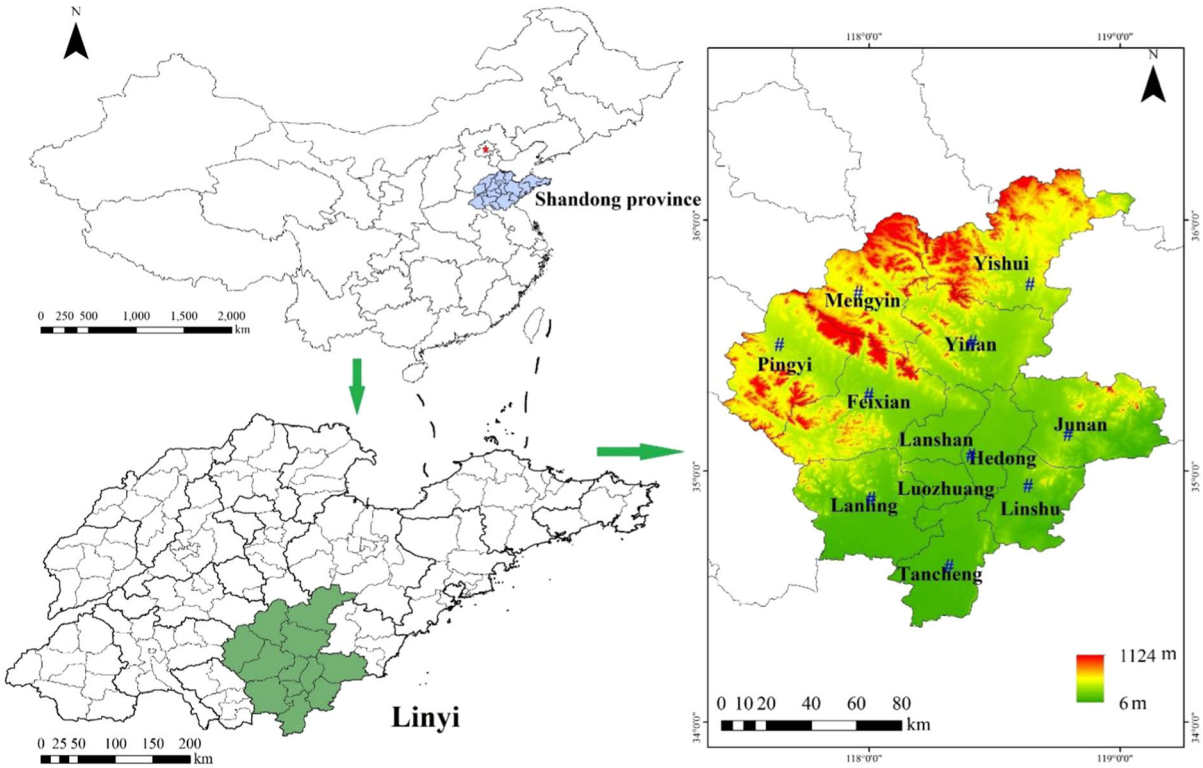


Fig. 1 Study area, showing the terrain elevation, surface meteorological monitoring stations, and main meteorological stations in Linyi

The detailed emission list of air pollution sources in Linyi City is scarce and cannot explain the characteristics of pollution discharge or the effect of air pollutants on human health. As Linyi is a typical industrialized city, the relationship between its air quality and high incidence of respiratory and circulatory systems diseases should be considered. The research results can provide a reference for the formulation of relevant urban air quality policies and disease interventions in Linyi that can also be implemented in other industrialized cities.

In this study, the time series and spatial distribution of respiratory and circulatory diseases in Linyi in 2015 were investigated. Then, an industrial air pollutant emission inventory was established based on the data of environmental statistics and the yearbook of Linyi. Combined with information from agricultural sources and the civilian and traffic source emissions of Multi-resolution Emission Inventory for China (MEIC), a comprehensive air pollutant emission inventory of Linyi City in 2015 was established. The CALPUFF model was used to simulate the contribution of air pollution emissions to the atmosphere and the residential intake fraction (IF) in Linyi. Finally, to clarify the correlation between diseases and air pollutant emissions, the Pearson correlation was calculated between residential IF and outpatient visits. This study succeeded in clarifying the relationship between regional emissions of air pollutants, population exposure and spatial clustering of diseases.

Methodology and key parameters

The data of human health

The outpatient visit data for respiratory and circulatory diseases were collected from the main hospital and clinic of Linyi. It contained the daily outpatient information of most patients from January 1, 2015, to December 31, 2015, regarding a total of 94,154 and 38,324 cases of respiratory diseases and circulatory diseases, respectively. The medical record information extracted from the outpatient data included the patient's gender, age, home address, disease diagnosis, department of treatment, date of treatment, and ID number. The data of non-residents in Linyi were excluded according to the patient's home address information to ensure that all the included data were collected from local residents. A data quality control protocol, involving missing data detection, error detection and possible error corrections (Zahumenský 2004), was conducted for the raw data, and the results support the applicability and usability of the data. According to the tenth edition of the *International Classification of Diseases* (ICD-10), respiratory diseases (ICD-10 code: J00-J99) and circulatory diseases (ICD-10 code: I00-I99) were selected as the end points of health effects.

The regional distribution of disease incidence (I) was calculated using the following formula:

$$I = \frac{\mathrm{POP}_{\mathrm{patients}}}{I_{\max} \cdot \mathrm{POP}_{\mathrm{total}}} \quad (1)$$

where $\mathrm{POP}_{\mathrm{patients}}$ is the population with disease in a certain area, $\mathrm{POP}_{\mathrm{total}}$ is the total population in the area, $I_{\max}$ is the maximum disease incidence, and the value was 1.

Spatial autocorrelation analysis was performed to understand the spatial dependence between the outpatient visit data. Spatial autocorrelation is the value of similar variables for regions with similar positions. If the high values clustered together with the other high values, and the low values clustered with the other low values, it was considered to present "positive spatial autocorrelation"; conversely, if the high values clustered together with the low values, it was considered "negative autocorrelation"; if the high and low values were completely randomly distributed, then there was no spatial autocorrelation. Moran's I index has been widely used in spatial autocorrelation analysis (Zou et al., 2014; Zhang et al., 2020). The formula is as follows:

$$\text{Moran's I} = \frac{\sum_{i=1}^{n}\sum_{j=1}^{n}\omega_{ij}(x_i-\overline{x})(x_j-\overline{x})}{S^2\sum_{j=1}^{n}(x_i-\overline{x})^2} \quad (2)$$

where $S^2 = \frac{\sum_{i=1}^{n}(\mathrm{xi}\ -\ \overline{x})^2}{n}$ is the variance of the samples, and ω_{ij} is the (i, j) element of the spatial weight matrix (measure of the distance between regions i and j). $\sum_{i=1}^{n}\sum_{j=1}^{n}\omega_{ij}$ is the sum of all weights.

The value of Moran's I is generally between -1 and 1, and a value greater than 0 indicates that there is a positive autocorrelation. The high value is adjacent to the high value, and the low value is adjacent to the low value. A value less than 0 indicates a negative autocorrelation, and a high value is adjacent to the low value. In general, positive autocorrelation is more

common than negative autocorrelation. If the Moran index I is close to 0, it indicates that the spatial distribution is random and there is no spatial autocorrelation. The Moran's I can be regarded as the correlation coefficient between the observed value and its spatial lag.

When there is no spatial autocorrelation in the total index, spatial autocorrelation may still exist in the local region. The local Moran's I (MI) of the LISA proposed by Anselin was used to test the spatial aggregation of the region (Anselin & Florax, 1995; Sang, 2001). MI measures the degree of correlation between region i and region j. The specific calculation formula is as follows:

$$\mathrm{MI} = \frac{x_i - \overline{x}}{S_y^2} \sum_{j=1}^{n} \left[\omega_{\mathrm{ij}} \left(x_j - \overline{x}\right)\right] \tag{3}$$

The spatial correlation and clustering of the number of outpatients in Linyi was drawn using ArcGIS software. The local correlation of outpatient data in Linyi shows the existence of H–H, L–L, L–H, and H–L. H–H denotes "high-high", meaning an area with a high number of outpatient cases was surrounded by a region with a high number of outpatient cases. L–L denotes "low-low", meaning an area with a low number of outpatient cases was surrounded by a region with low number of outpatient cases. L–H denotes "low–high", meaning an area with low number of outpatient cases surrounded by a region with a high number of outpatient cases. H–L denotes "high-low", meaning an area with a high number of outpatient cases surrounded by a region with a low number of outpatient cases.

Emission inventory method

Emission inventory is an important basic parameter data of environmental air quality management. It is not only the starting point of scientifically describing the impact of air pollution, but also the final destination for implementing pollution control measures (Streets et al., 2000; Liu et al., 2015; Shahbazi et al., 2016). Based on the technical guidelines on the preparation of the inventory published by MEP, we categorized the emission sources into industrial sources, agricultural sources, civilian sources, and traffic sources, focusing on seven kinds of air pollutants: $PM_{2.5}$, PM_{10}, SO_2, NOx, CO, VOCs, and NH_3. Industrial source emissions included stationary combustion sources and industrial process sources. The stationary combustion sources referred to the combustion equipment that used the heat generated from fuel combustion to provide heat and power for electricity generation, industrial production and life. Industrial processes referred to as industrial activities with the purpose of realizing physical and chemical transformations of industrial materials during industrial production and processing. Basic data on industrial resources mainly came from the environmental statistics of Linyi in 2015, which included the name, address, longitude and latitude, production of main products, industry sector, annual energy consumption, and other information about a total of 835 industrial enterprises (including 15 power plants) in the urban area of Linyi. Based on the detailed information of the enterprises' names and addresses, remote sensing images were used to check and correct the longitudes and latitudes of each enterprise, which provided accurate data support for the analysis of the temporal and spatial distribution characteristics of the point-source air pollutants.

In the present study, the enterprises involving industrial process sources were divided into 21 subdivisions as the key research objects. In accordance with the corresponding production volume and technology relating to the emission sources, the author adopted the emission factor method to obtain the estimated emission of pollutants. The formula is as follows:

$$E_i = \sum_{j,m} A_{i,j,m} \times EF_{i,j,m} \times \left(1 - \eta_{i,j,m}\right) \tag{4}$$

where E_i is the pollution emissions; A is the production volume of the industrial product under the third-level emission sources and the technology under the fourth-level emission source; i, j, and m are the pollutant, enterprise, and production volume/technology type, respectively; EF is the coefficient of the production of primary pollutants; and η is the terminal control technology's pollutant-removing efficiency.

The data on power plants came from a high-resolution power emission inventory for China based on CEMS in 2015 (HPEC) (Cui et al., 2018). Meanwhile, considering the completeness of the emission inventory, the MEIC was used for the agricultural sources, civilian sources, and traffic source emissions. However, it is noteworthy that there were certain uncertainties in the emission inventory.

First, the uncertainty of activity level: the activity level data of industrial pollution sources was from data on coal burning and other raw fuel consumption of environmental statistics, and its uncertainty was lower. Environmental statistics generally only created statistics for enterprises above a designated size and lacked information on the activity level data of small enterprises (small scattered pollution). Second, the uncertainty of emission factors, some industries use the emission factors of the *Technical Manual* in the estimation of inventory, which led to the uncertainty of the emission inventory.

Model parameters

CALPUFF is an integrated Lagrangian puff modeling system that has been widely used for the simulation of atmospheric pollution dispersion (Elbir, 2004; Levy et al., 2002; Ozkurt et al., 2013; Sagan et al., 2018). The modeling system is recommended by the US Environmental Protection Agency (EPA) (Ranzato et al., 2012), and it is also one of the regulatory dispersion models recommended by the Ministry of Ecology and Environmental of the People's Republic of China (Wang et al., 2005; Cai et al., 2015).

The modeling system uses terrain data of 90 m from the United States Geological Exploration Agency (USGS) and land data with an accuracy of 30 m (GeoPortal C, 2018). Surface meteorological monitoring data (Surf.dat) and 3-mesoscale prediction data (3d.dat) are input files for CALMET, where Surf.dat uses meteorological data from nine monitoring stations in Shandong province (except Weinan Station). The meteorological data of Yinan station were used to verify the results of the CALMET meteorological simulation. The Universal Transverse Mercator (UTM) was used to simulate the year 2015. The grid resolution was 3000 m, and there were 66×83 grid points in both the east–west and north–south directions. Theten vertical layers were set at heights of 20, 40, 80, 160, 300, 600, 1000, 1500, 2200, and 3000 m. The MESOPUFF II chemical mechanism was chosen and hourly ozone monitoring data were inputted in the model to estimate the daytime oxidation of SO_2, NOx, PM_{10} and $PM_{2.5}$. The dry and wet deposition of each pollutant was considered during the simulation, and a one-hour simulation time step was used.

Relative studies about the evaluation of air pollutants using CALPUFF have suggested that the overall uncertainty of CALPUFF model is roughly − 32.88 to 32.67% (Cai et al., 2015), which was similar with the uncertainty in our study (− 63.25 to 36.47%). Zhou et al. (2003) and Hu (2007) estimated the population exposure to power plant emissions using CALPUFF and indicated that the CALPUFF model was suitable for assessing the public health risk posed by power plants. Levy et al. (2002) also suggested that the inherent limitations of CALPUFF may cause the additional uncertainties. However, the degree of uncertainty in atmospheric modeling such as CALPUFF will not dominate the total uncertainty associated with health impact estimation.

Intake fraction

Particulate matter (PM), SO_2, and NOx can enter the human body mainly through the respiratory exposure pathway (Davidson et al., 2005; Johns & Linn, 2011; Park & Kwan, 2017). The IF is defined as the fraction of pollutants inhaled from the total emission of a source (Bennett et al., 2002). It is a relatively simplified linkage of population exposure with pollutant emissions. In this study, the characteristics of pollution sources (chimney height, diameter, emission temperature, and velocity), meteorological characteristics (wind speed, wind direction, and atmospheric stability), pollutant characteristics (vapor–liquid–solid phase, reactivity, and solubility), and receptor characteristics (location and population density) were integrated into the model analysis.

The spatial distribution data of Linyi's population use the LandScan population dataset released by the American Oak Ridge National Laboratory with a spatial resolution of 1 km (Dobson et al., 2000; Liu et al., 2018). The respiratory rate of Shandong residents in the *Exposure Factors Handbook of the Chinese Population* was selected as a relatively localized respiratory rate among Linyi residents. IF was calculated using the following equation (Hao et al., 2007):

$$\mathrm{IF}_{xy} = \frac{\sum_i^N \mathrm{POP}_i C_{xi} \mathrm{BR}}{Q_{xy}} \quad (5)$$

where IF_{xy} is the IF of the pollutant x emitted by the pollution sources y, N is the total number of grids in the study region, i is the grid number, POP_i is the total population of the ith grid, C_{xi} is the concentration of the ith grid contaminant x calculated in μg/m^3, BR is the respiratory rate in m^3/d per person, Q_{xy} is the emission of pollutant x from pollution sources y in μg/d, and x includes $PM_{2.5}$, PM_{10}, SO_2, and NOx.

Results and discussion

Respiratory and circulatory system diseases in Linyi

In 2015, the top five most prevalent health disease with high mortality in Linyi were cardiovascular diseases, cerebrovascular diseases, malignant tumors, respiratory diseases (chronic bronchitis, pneumonia, chronic obstructive pulmonary emphysema), and injuries. Among these, cardiovascular, cerebrovascular, and respiratory diseases account for 62.32% of the total causes of death, and the mortality rate of cardiovascular diseases is still on the rise (Linyi Center for Disease Control, 2016). Air pollution has a more severe effect on cardiovascular and respiratory diseases have also been reported in relevant studies (Lelieveld et al., 2020; Li et al., 2018; Liu et al., 2019; Yang et al., 2019). Therefore, to reveal air pollution-related health problems in Linyi, the relationship between air pollution and respiratory system and circulatory system diseases (hypertension, coronary heart disease, acute cerebrovascular disease, heart failure, arrhythmia, and rheumatic heart disease) in Linyi was analyzed in this study. In 2015, the total number of outpatient visits in Linyi was approximately 2.06 million, of which 94,154 visits were for respiratory diseases, accounting for 4.57% of the total outpatient visits. For circulatory diseases, the total number of outpatient visits was approximately 38,324, accounting for 1.86% of the total outpatient visits. The average number of daily outpatient visits for respiratory diseases was 310.8 in Linyi (Table S1). In winter, the number of outpatient visits for respiratory diseases was significantly higher than that in other seasons, especially in spring, with the lowest outpatient visits (Fig. 2). The highest number of daily outpatient visits in winter was approximately 3.6 times higher than the lowest daily outpatient visits in spring. For respiratory system diseases, high PM concentrations in winter showed strong synchronization with the high outpatient visits (Fig. S2). Significant correlation was observed between PM concentration and outpatient visits of respiratory diseases ($p < 0.05$). The number of male patients with respiratory diseases was significantly higher than that of female patients. Among them, there were 189.3 males (60.9%) and 121.5 females (39.1%). Epidemiological studies have indicated that air pollution has significantly different effects on males and females, which may be related to sex-related physiological or lifestyle differences, such as females being more sensitive to air pollution; however, the observed high morbidity for males may be related to smoking or more outdoor activities (Clougherty, 2009; Miraglia et al., 2013; Zhang et al., 2011). The number of children under 6 years of age with respiratory diseases was much higher than that in other age groups, accounting for almost 50% of the annual outpatient visits for respiratory system diseases, followed by juvenile patients aged 7–17 years. Relevant studies have suggested that age modifies outdoor air pollution-related mortality, and air pollution-induced mortality increases with age (Kan et al., 2008; David et al., 2002). However, the morbidity was high for children under 6 years in the present study, implying that the influence of air pollution on children may be more complicated due to physiological characteristics (Clougherty, 2009). The number of outpatient visits for circulatory diseases was lower, and there were no significant seasonal trends or gender differences. The number of outpatient visits for circulatory diseases in middle-aged people aged 41–65 is much higher than that in other age groups, accounting for 52.2% of the total annual number of outpatient visits of circulatory system diseases, followed by elderly patients over 66 years old. The sum of the number of outpatient visits in these two age groups accounts for 85.46% of the total annual number of outpatient visits of circulatory system diseases. Since environmental exposures are complex, a comprehensive and in-depth investigation across multiple life stages from infancy, childhood, puberty, adulthood, and the elder is suggested for future research.

From the perspective of spatial distribution, the distribution of outpatient visits for respiratory and circulatory diseases indicated that high number of outpatient visits were concentrated in the main urban

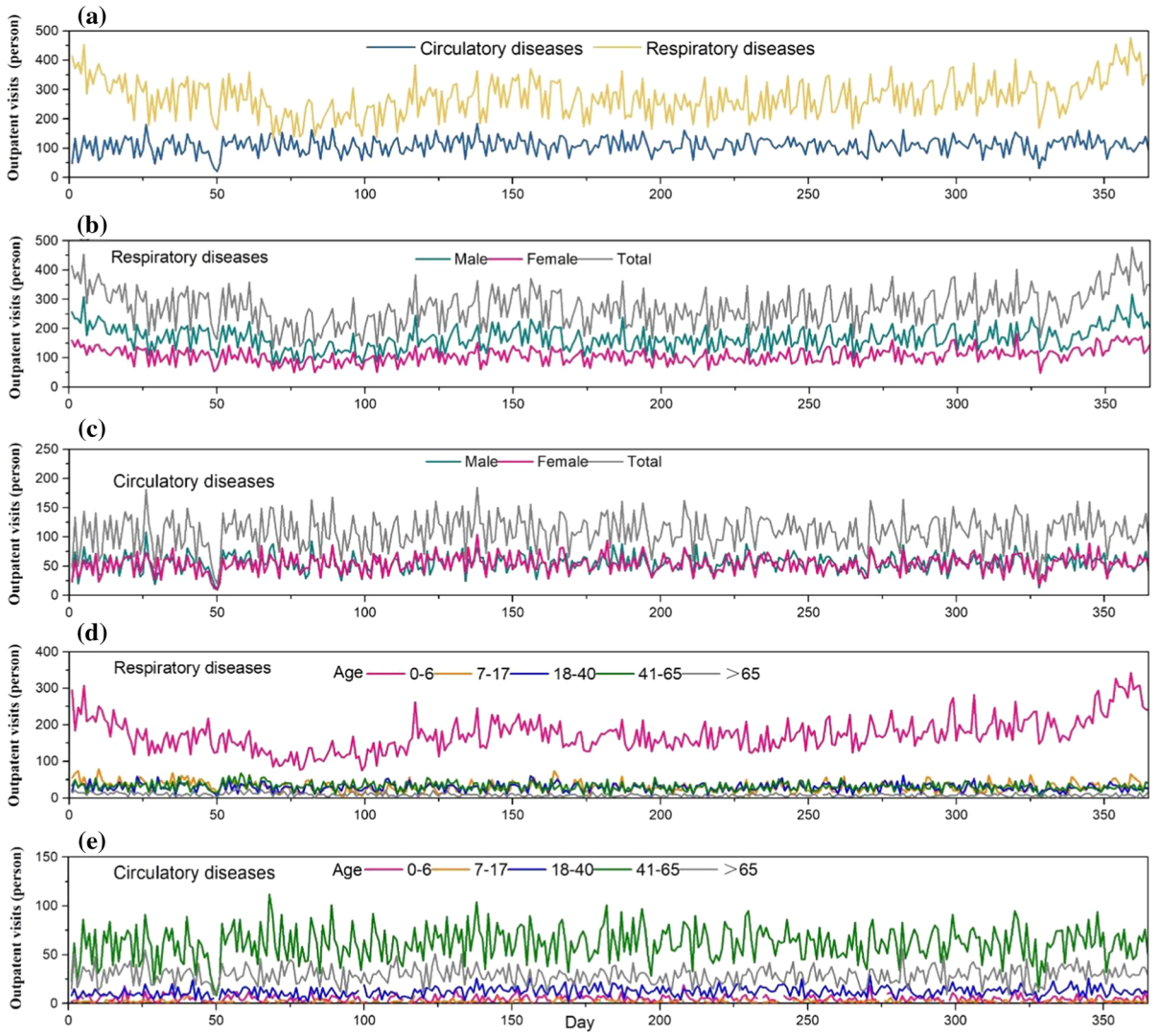


Fig. 2 Time series of outpatient visits for respiratory and circulatory diseases (**a**) with different genders (**b**, **c**) and ages (**d**, **e**) reported by Linyi in 2015

area of Linyi (Hedong District, Lanshan District, and Luozhuang District), especially at the junction of these three areas (Fig. 3). Most of the above-mentioned areas located in urban and suburban areas. On the one hand, the above areas have a large population and large traffic exhaust emissions. On the other hand, the large amount of pollutants from industrial activities in the suburbs can be easily transported, leading to high pollutant concentrations. Therefore, the spatial distribution of air pollutant emissions in Linyi will be discussed to determine whether there is a correlation between the incidence of diseases and air pollution in Linyi.

Air pollution concentration of Linyi in 2015

The total emissions of $PM_{2.5}$, PM_{10}, SO_2, NOx, CO, VOCs and NH_3 in Linyi in 2015 were 1.09×10^5, 1.43×10^5, 8.80×10^4, 1.5×10^5, 1.49×10^6, 4.93×10^5, and 1.24×10^5t, respectively (Table S2). According to the emission sources, civilian sources and industrial process sources were major contributors. For $PM_{2.5}$, civilian sources and process sources accounted for 52.6% and 36.2%, respectively. For industrial process sources, $PM_{2.5}$ mainly comes from the nonferrous metal metallurgy and calendaring industry, foundry industry, and

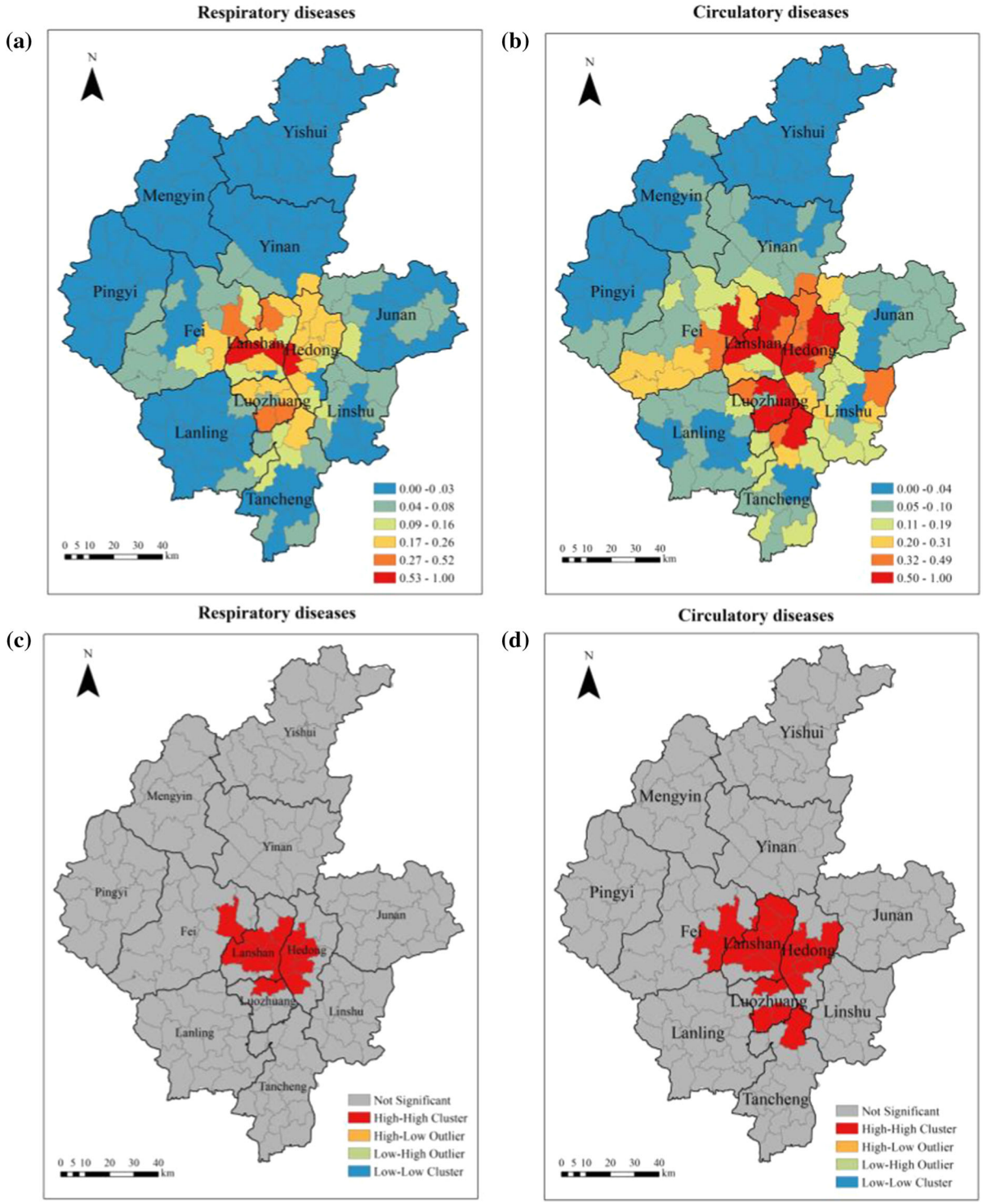


Fig. 3 Distribution of outpatient visits for respiratory and circulatory diseases in counties of Linyi in 2015. **a**, **b** Show the standardized distribution of these diseases, **c**, **d** indicate the regional clustering of the diseases

nonmetallic mineral products industry, accounting for 54.7%, 16.9%, and 12.2% of the total $PM_{2.5}$ emissions of the process sources, respectively. For PM_{10}, civilian source and process source contribution rates of civilian and technological processes were 44.2% and 46.3%, respectively. For the process source, PM_{10} mainly came from nonferrous metal smelting and calendar processing, nonmetallic mineral products, and foundry industries, accounting for 39.2%, 33.1% and 14.1% of the total PM_{10} emissions from the process sources, respectively. Civilian sources, stationary combustion sources, and industrial process sources of fossil fuel were major contributors to SO_2 emissions, accounting for 47.4%, 31.9%, and 16.9%, respectively. Among them, the SO_2 emissions from industrial boilers accounted for 79.6% of the total emissions from stationary combustion sources. For the industrial process source, the discharge of nonferrous metal smelting and calendar processing industries and nonmetallic mineral products industry accounted for 39.2% and 33.1% of the total SO_2 emissions from the process source, respectively. NO*x* was mainly from traffic sources and stationary combustion sources, accounting for 65.5% and 16.1%, respectively. Civilian sources, process sources and traffic sources were the main sources of CO, accounting for 65.5%, 16.6% and 15.7%, respectively. VOCs mainly came from process sources and a civilian source, which accounted for 76.7% and 18.1%, respectively. Among these, the chemical industry was the main emission industry of atmospheric VOCs in Linyi, accounting for 58.5% of total VOCs in the process sources. About 90.1% of NH_3 emissions were from agricultural sources, and the industrial process sources contributed 5.82% of the chemical raw materials involved in the production of urea.

Regarding the spatial distribution of the industrial process source for particles ($PM_{2.5}$ and PM_{10}), Luozhuang District had the largest number of enterprises; the highest emission amounts were from Junan County, Linshu County, and Hedong District, and some enterprises had relatively large amounts of emissions (Table S3), which was related to major factors such as economic development, industrial structure, enterprise management, and technical level. Moreover, the Luozhuang District has the largest emission amount of SO_2, NO*x*, and CO. High emissions of SO_2 in Yishui County, Fei County, and Tancheng County were related to industrial combustion. The GDP of Lanshan, Luozhuang, Yishui and Feixian County accounted for 50.5% of the total industrial GDP in Linyi. The industrial enterprises above a designated size accounted for only 36.7% of total enterprises. $PM_{2.5}$, PM_{10}, SO_2, NO*x*, CO, VOCs and NH_3 emissions from industrial enterprises above designated size accounted for 24.2%, 27.2%, 56.5%, 67.4%, 42%, 39.5% and 14.3%, respectively. Particularly for Lanshan District, which has the highest GDP and a relatively low pollutant emission (Fig. S3). However, according to the spatial distribution of respiratory and circulatory system diseases, high numbers of outpatient visits were from Lanshan, Luozhuang, and Hedong District. Therefore, pollutant emissions might be significantly different from atmospheric concentrations, and airborne concentrations are closely related to the chemical characteristics of pollutants, meteorological conditions, and regional transportation (Li et al., 2019; Zhang et al., 2019).

The simulation results from CALPUFF indicated that the pollutants concentration were high in the central areas of Linyi, mainly concentrated in Lanshan, luozhuang, Feixian, Lanling and Hedong (Fig. 4). The different spatial distribution of pollutant emissions and atmospheric concentration revealed that such distribution might be associated with the transport of pollutants, topography, and other meteorological factors (Li et al., 2019; Zhang et al., 2019). For instance, regional transport significantly influences air quality in Beijing during severe haze events (Zheng et al., 2015). In 2015, the prevailing north wind at the northern areas, and the prevailing south wind in southern area of Linyi, resulted in the high centration of air pollutants in the central area of Linyi, including the Lanshan, Hedong and Luozhuang District (Fig S4. A). Meanwhile, effected by topography, high altitude in the northwest of Linyi was unfavorable for the diffusion of air pollutants in the central area, which further exacerbated air pollution in the central areas. Moreover, influencing by the prevailing north wind in winter, the regional transmission of pollutants in the Beijing-Tianjin-Hebei region of northern China is also an important reason for the high concentration of pollutants in Linyi (Fig. S4 B). High terrain in the north of Linyi and the prevailing northerly winds in winter were favorable for pollutant diffusion in the northern part of Linyi to the central and southern area of Linyi. For the central and southern areas, the lower terrain, more industrial emission sources, and a dense

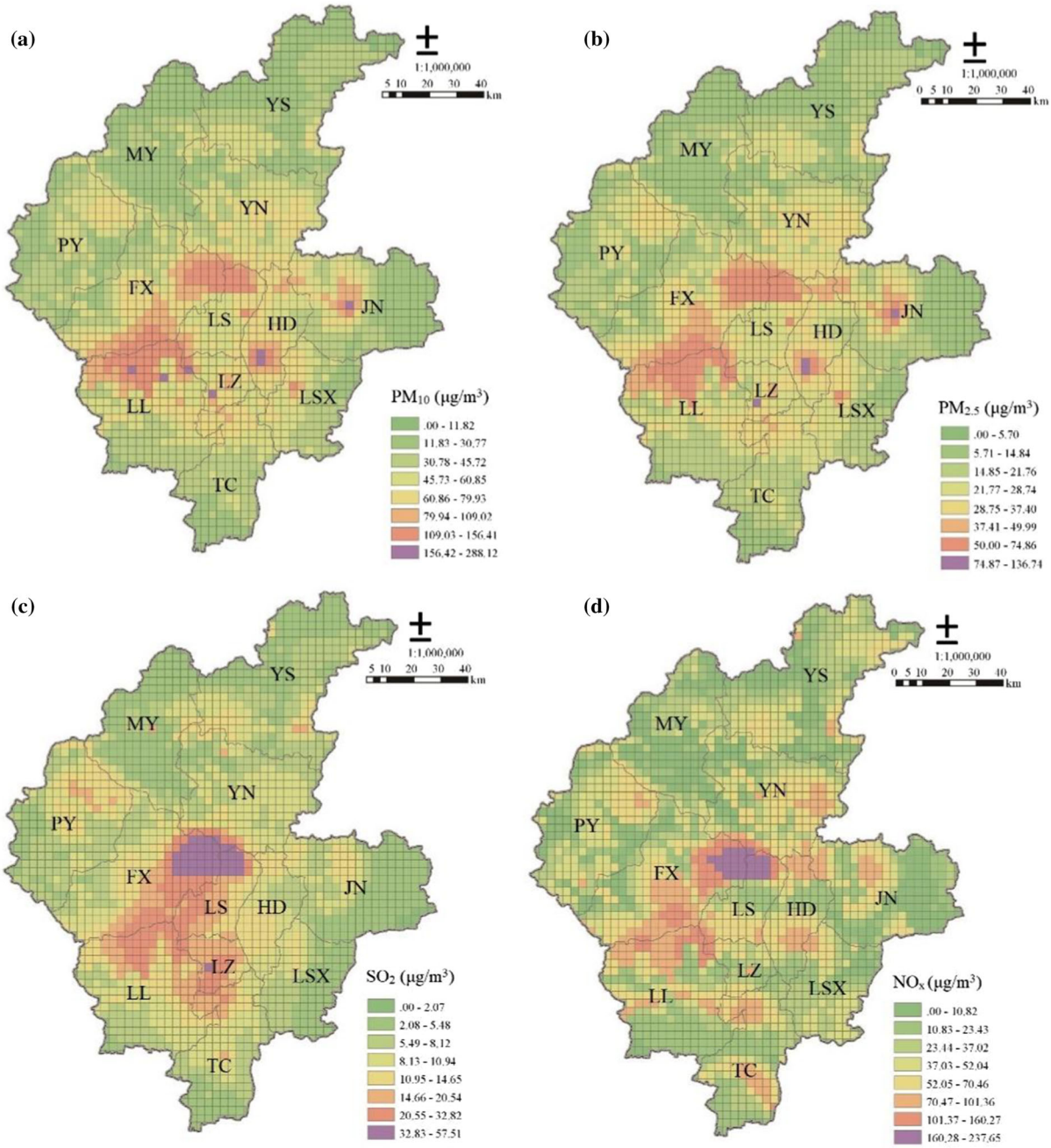


Fig. 4 Simulation of the average annual concentrations of four pollutants ($PM_{2.5}$, PM_{10}, SO_2, and NO*x*)

population were unfavorable for the diffusion of pollutants. Therefore, the spatial distribution of pollutant concentration does not necessarily agree with the distribution of emissions; it might be closely related to the location of emission sources, the height of emission, the topography and meteorological conditions.

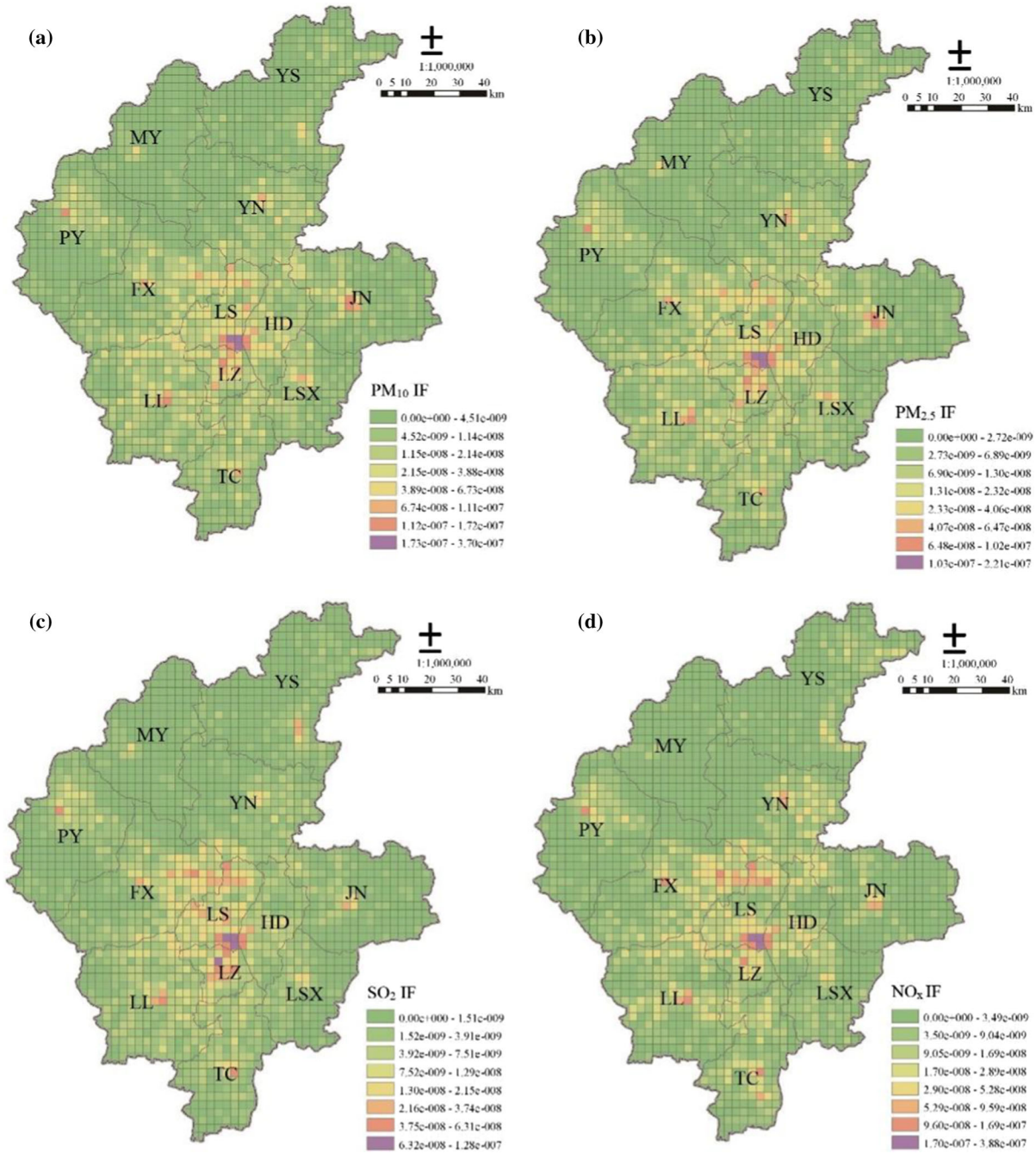


Fig. 5 Grid distribution of residential IF of four pollutants ($PM_{2.5}$, PM_{10}, SO_2 and NO*x*) in Linyi

Evaluation of residents IF of Linyi in 2015

The total IF of the Linyi residents to $PM_{2.5}$, PM_{10}, SO_2 and NO*x* was 1.2×10^{-5}, 2×10^{-5}, 0.7×10^{-5}, and 1.6×10^{-5}, respectively (Fig. 5). The high IF regions were mainly concentrated at the intersection of Lanshan, Luozhuang, and Hedong districts, which have large populations. From the IF of a single grid, the maximum IF of NO*x* in a grid in Linyi was 3.88×10^{-7}, followed by PM_{10}, $PM_{2.5}$ and SO_2, and the maximum IF was 3.70×10^{-7}, 2.21×10^{-7}, and 1.28×10^{-7}, respectively, which were closely related

to factors such as population density, pollutant discharge, etc. The IF of residents was closely related to population density. For PM_{10}, $PM_{2.5}$ SO_2 and NOx, areas with density populations showed higher IF for residents in Linyi.

From the aspect of the process source, the IF of $PM_{2.5}$ from different industries to residents in Linyi varied significantly (Fig.S5). The fiberglass yarn industry had little influence on the main urban area of Linyi City (Hedong District, Lanshan District, Luozhuang District), mainly concentrated in the southern part of Yishui County, the $PM_{2.5}$ emission of this industry was the smallest in the process sources. However, the IF was not the minimum, which may be related to the accumulation of pollutants in Yishui County and then influence $PM_{2.5}$ concentration in other areas through long-distance migration and diffusion. The maximum IF of ferrous metal smelting and calendaring industry, chemical raw materials, petroleum coking and nuclear fuel processing industries, nonferrous metal metallurgy and calendaring industry, nonmetallic mineral products industry, foundry industry were 3.36×10^{-6}, 1.44×10^{-7}, 2.05×10^{-7}, 4.19×10^{-7}, 6.06×10^{-7}, 6.31×10^{-7}, respectively. $PM_{2.5}$ emission from ferrous metal metallurgy and calendaring industry was the largest in Linyi, but its maximum IF was not the largest. The nonmetallic mineral products industry had a closer relationship with the population IF and population density, which may be related to the higher emission of the industry and the smaller contribution of the unit emission to the surrounding area. Due to the impact of the regional transmission of pollutants, locally generated pollutants may occur at high concentrations in the surrounding area and downwind areas (Li et al., 2019).

Correlation analysis between IF and health

The spatial distribution of outpatient visits from respiratory and circulatory diseases was concentrated in the main urban area of Linyi City (Hedong District, Lanshan District, Luozhuang District) (Fig. 3). This was supported by the regions with high IF of PM_{10}, $PM_{2.5}$, SO_2, NOx, and main industrial sources (Fig. 5 and Fig. S5). The above areas have a large population, with relatively high GDP and more industrial emission sources. As shown in Fig. S3, for $PM_{2.5}$ and PM_{10}, the Luozhuang and Hedong district have fewer enterprises but with large emissions. Moreover, the Luozhuang and Lanshan district have relatively high emissions of SO_2 and NOx. The Pearson correlation coefficient (R) was conducted to reveal the relationship between the outpatient visits and IF of residents to NOx, SO_2, $PM_{2.5}$, and PM_{10} (Table 1). The high R values ($R > 0.6$, $p < 0.05$) imply similar spatial distribution of outpatient visits and IF of residents, and p value statistically supports this relationship at a confidence level of 95%.

The Pearson's correlation suggested that the high incidence of respiratory and circulatory diseases was significantly associated with exposure to air pollutants in Linyi; residents' exposure to NOx and SO_2 had the highest correlation with respiratory and circulatory diseases. Moreover, a high correlation coefficient between PM_{10} and $PM_{2.5}$ also suggested the high risk of respiratory and circulatory diseases induced by PM. Relevant studies have suggested that air pollution is one of the most dangerous health risks in the world (Kelly & Fussell, 2015; Ali et al., 2019). Air pollution causes damage to blood vessels by increasing oxidative stress, leading to increased blood pressure, diabetes, stroke, increased blood pressure, and heart failure, and can also lead to cardiovascular diseases and premature death (Lelieveld et al., 2020). Compared with adults, the elderly and children are more susceptible to the effects of air pollution (Maji et al., 2018). Liu et al. (2019) investigated ambient PM_{10}, $PM_{2.5}$ and daily mortality in 652 cities worldwide and found close link between mortality and PM concentration, especially mortality due to respiratory and circulatory diseases.

As mentioned in "Air pollution concentration of Linyi in 2015" section, the emission inventory suggested that civilian sources and process sources were major contributors to PM_{10} and $PM_{2.5}$ emissions in Linyi in 2015. Civilian sources, stationary combustion sources and industrial process sources were major contributors to SO_2 emissions. For NOx, the traffic sources and stationary combustion sources were the main emission sources. Among these, industrial and vehicle emissions were the main sources of atmospheric pollutants. Moreover, although pollutant emissions in urban areas were not the highest, urban areas with densely populated regions have a high correlation with disease prevalence. In recent years, the implementation of aggressive emissions control measures has significantly improved the air quality in

Table 1 Results of statistical analysis on the relationship of outpatient visits and IF of residents of four pollutants (NO*x*, SO_2, $PM_{2.5}$ and PM_{10})

	Total			Respiratory diseases			Circulatory diseases		
	N	*R*	*p* value	*N*	*R*	*p* value	*N*	*R*	*p* value
IF to NO*x*	12	0.626**	0.029	12	0.621**	0.031	12	0.642**	0.025
IF to SO_2	12	0.661**	0.019	12	0.657**	0.020	12	0.672**	0.017
IF to $PM_{2.5}$	12	0.600**	0.039	12	0.591**	0.043	12	0.628**	0.029
IF to PM_{10}	12	0.613**	0.034	12	0.603**	0.038	12	0.643**	0.024

**Significance at 5%

China. Strict clean air and emission reduction policies have reduced the population's prevalence rate and mortality risk from 2013 to 2017. The rapid decrease in $PM_{2.5}$ caused a 14% reduction in premature deaths due to long-term exposure, and number of deaths associated with acute exposure decreased by 61% during heavily polluted days (Xue et al., 2019). However, due to the complexity of exposure and lag between air pollution and disease morbidity, the incidence of respiratory and circulatory system diseases in Linyi has still been on the rise in recent years. Although the concentration of airborne pollutants is significantly reduced, persistent health risks still exist, and the health effects of short-term and long-term exposure still need further attention. Gu et al. (2019) and Yue et al. (2020) suggested that stronger emission reduction policies are still required to substantially reduce air pollution-associated morbidity and mortality. The spatial distribution of pollutants and the exposure risk in densely populated areas still require particular attention.

Aside from chemical compounds (such as PM, VOCs, SO_2, and NO*x*), there is also a large number of biological compounds in the air (e.g., bacteria, viruses, pollen spores, and other bioaerosols) (Humbal et al., 2018), which will influence the exposure–response relationship of air pollutants. Moreover, the multi-pollutant mixtures, multiple exposures (in both occupational and non-occupational environments), other environmental conditions such as geographic environment (including urban internal structures), extreme weather (extreme high or low temperature), and socioeconomic conditions increased the complexity and uncertainty of the simulation and prediction of the pollution-disease model (Bobb et al., 2018). Therefore, the occurrence of disease is related to individual genetic and living conditions (e.g., diet and economic conditions), and environmental factors (e.g., air, water and soil pollution; and local economic level and industrial institutions). Future research on pollution-exposure models will move toward multi-dimensional, nonlinear, and multiple statistical analyses (Stafoggia et al., 2017). The etiological associations between multiple environmental risk factors and human health will be targets of scientific inquiry and interest in the fields of environmental health, which may combine epidemiology, clinical trials, and large-scale data analysis.

Conclusions

In this study, air pollutant emissions and the exposure–response relationship between air pollutants and respiratory and circulatory diseases in Linyi were investigated to reveal the characteristics of air pollutants and potential health effects. In Linyi, circulatory and respiratory diseases accounted for 62.32% of total deaths in 2015. High air pollutant emissions in Linyi might be responsible for the incidence of regional respiratory and circulatory diseases, especially in males and young people. Further analysis of the impact of air pollutant emissions on diseases suggested that the simulated ambient annual average concentrations of four pollutants ($PM_{2.5}$, PM_{10}, SO_2 and NO*x*) were quite different from those of pollutant emissions. Regional transport, topography, and meteorological factors may be responsible for the disparity. However, significant spatial correlations between the residential IF of ambient air pollutants and outpatient visits of respiratory and circulatory diseases were observed. The high residential IF values of ambient air

pollutants and outpatient visits were concentrated in urban and suburban areas, with high population density and large traffic exhaust emissions. Therefore, to reduce ambient air pollutant concentrations in Linyi, persistent protective measures and efforts should be strengthened in the future, especially in densely populated areas.

Acknowledgements This study was supported by Special Fund of State Key Joint Laboratory of Environment Simulation and Pollution Control (16K01ESPCT), the National Research Program for Key Issues in Air Pollution Control (No. DQGG0209-07 and DQGG0304-07), and the National Natural Science Foundation of China (42075183).

Author contributions XW, DL and JY involved in outpatient visits data collection and manuscript writing; MF and HL involved in data analysis, illustration preparation and manuscript revision; Technical support was done by HL; Air pollutants emission inventory establishment was done by PW; CALPUFF model application: was done by XL; MW and XB involved in funding acquisition and conceived and designed the experiments.

Funding Air pollutant emission inventory and meteorological data collection, and CALPUFF model analysis, were funded by the Special Fund of State Key Joint Laboratory of Environment Simulation and Pollution Control (16K01ESPCT), the National Research Program for Key Issues in Air Pollution Control (Nos. DQGG0209-07 and DQGG0304-07), outpatient data collection and analysis were supported by the National Natural Science Foundation of China (42075183).

Data availability Data available on request from the authors. The data that support the findings of this study are available from the corresponding author, minwei@sdnu.edu.cn and boxin@acee.org.cn, upon reasonable request.

Declarations

Conflict of interest The authors declare no conflict of interest.

Animal research This study contains no animal research.

Consent to participate Not applicable.

Consent to publish Not applicable.

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