Distribution and Determinants of Correlation between PM2.5 and O3 in China Mainland: Dynamitic simil-Hu Lines

Chen Chenru, Xu Miaoqing, Liu Shuyi, Zhu Dehai, Yang Jianyu, Gao Bingbo*, Chen Ziyue*

Abstract

In recent years, China has made great efforts to control air pollution. During the governance process, it is found that fine particulate matter (PM2.5) and ozone (O3) change in the same trend among some areas and the opposite in others, which brings some difficulties to take measures in a planned way. Therefore, this study adopted multi-year and large-scale air quality data to explore the distribution of correlation between PM2.5 and O3, and proposed a concept called dynamic simil-Hu lines to replace the single fixed division in the previous research. Furthermore, this study discussed the causes of distribution patterns quantitatively with geographical detector and random forest. The causes included natural factors and anthropogenic factors. And these factors could be divided into three parts according to the characteristics of spatial distribution: broadly changing with longitude, changing with latitude, and having local characteristics. Overall, regions with relatively more densely population, higher GDP, lower altitude, higher humidity, higher atmospheric pressure, higher surface temperature, less sunshine hours and more accumulated precipitation often corresponds to positive correlation coefficient between PM2.5 and O3, no matter in which season. The parts with opposite conditions that mentioned above are essentially negative correlation coefficient. And what’s more, humidity, global surface temperature, air temperature and accumulated precipitation are four decisive factors to form the distribution of correlation between PM2.5 and O3. In general, collaborative governance of atmospheric pollutants should consider particular time and space background and also be based on the local actual socio-economic situations, geography and geomorphology, climate and meteorology and other comprehensive factors.

Keywords: PM2.5 and O3; correlation analysis; geographical detector; random forest; collaborative governance of atmospheric pollutants;

1. Introduction

Fine particulate matter (PM2.5) and ozone (O3) exist widely as atmospheric pollutants and appear simultaneously as governed objects. They are generated by secondary reaction processes and have common precursors, such as VOCs and NOx. These rigmaroles are influenced by several elements, including natural and anthropogenic aspects[1][2]. China has taken a series of air pollution control measures since 2013, especially for PM2.5. But at the
same time, O3 pollution has increased in some areas. Verifying the association between PM2.5 and O3 in a certain area and finding out the origin of this phenomenon are important premises to clarify the route of collaborative governance of them.

From the previous studies, it seems that the existing investigations on correlation of PM2.5 and O3 largely centered in trying to reveal the temporal and spatial features. Xie et al.[3] analyzed air quality data monthly that obtained from 286 monitoring sites during a year period, and considered that the relations between PM2.5 and O3 is either weak or uncorrelated in most of China. In contrast, Chen et al. [4] discovered that a positive correlation in summer and a negative correlation in winter, and also presented a significant spatial difference in north–south and coastland–inland China. Similarly, Jia et al. [5] revealed that inverse relations of PM2.5 and O3 between cold and hot seasons. Since then, numerous studies mentioned that the correlation of PM2.5 and O3 relevance to time and region exactly[6][7]. The current collaborative governance performed mainly focuses on human activities[8][9], and so do researches[10][11][12], they deliberately compared PM2.5 and O3 during COVID-19 period with the past. However, there is a lack of quantitative understanding of the impact of both natural factors and anthropogenic factors for the correlation between PM2.5 and O3, which can be used as a data basis of air quality control.

It should be noted that natural factors, especially geographical and meteorological factors, are inextricably linked. For example, due to the monsoon climate, those places with more total accumulative surface precipitation tend to have higher temperature. Thus, how to deal with multicollinearity of independent variables before traditional quantitative analysis (e.g. simple linear regression) has become a very complex problem[13]. Given the above, this study attempted to start from the basic theoretical principles of spatial analysis and solve problems from new ways. The second law of geography discussed by Goodchild et al., implies that geographic variables exhibit uncontrolled variance[14]. And spatial stratified heterogeneity proposed by Wang et al. referring to the within-strata variance less than the between strata-variance, implying potential distinct mechanisms by strata, suggesting possible determinants of the observed process[15]. The statistical method (q-value) based on the above principles can quantitatively analyze the force of influencing factors, which adopted by this research.

In summary, the objectives of this study are: (1) to verify the seasonal and spatial patterns of the correlation between PM2.5 and O3 through multi-year and large-scale air quality data; (2) to explore the determinants of the correlation distribution between PM2.5 and O3 then extract general character for air pollution control.

2. Materials and Method

2.1 Study area and data sources

The study area is Chinese Mainland. Most of it is in temperate zone, and a small part is in tropical zone, without frigid zone. Northern China has a temperate monsoon climate, with
four distinctive seasons. And subtropical monsoon climate dominates the South, which is characterized by hot rainy summer and mild humid winter. In terms of population and economy, Hu line which summarized by Hu, 1935[16] is recognized as an effective way to depict spatial heterogeneity. During 2015-2020, there are about 1500-1700 stations collecting air quality index (AQI) annually in China Mainland. However, in case of instruments maintenance, instruments failure, communication failure, power failure, etc., some stations were missing monitoring data. If there is a large amount of data incompleteness in a site, the experiments here may draw inaccurate or even completely opposite conclusions. Therefore, for threshold setting, only the sites with annual missing data less than 10% participate in spatial analysis. The details of data processing are shown in part of Table 4.

In this study, PM2.5 and O3 24-hour smoothed data from 1 January 2015 to 31 December 2020 in the research area were picked up from the processed air quality index. Since the data smoothing process, such as moving average or using the maximum value continuously in per time period is a special low-pass filter[17]. It can reduce randomness in the sequence. For this study, specifically, the data released by the National Urban Air Quality Real-time Release Platform is updated once an hour. It can be seen that an obvious peak change in 24-hour O3 data. And the crest value often appears at midday because O3 concentrations is related to the intensity of sunlight radiation. On the contrary, PM2.5 has no obvious characteristics in a single day. While the relevance of PM2.5 and O3 are explored seasonally, the regular pattern difference in the time granularity based on days will interfere with the reflection of the overall law. Hence, daily smoothing of the original data is a necessary preparation, as shown in Figure 1.

![Figure 1. Raw data and 24-hour smoothed data of PM2.5 and O3](image)

The second part of the data employed in this study is influencing factors, including anthropogenic factors and natural factors, which are obtained from the WorldPop Country Datasets (https://www.worldpop.org/), National Bureau of Statistics (http://www.stats.gov.cn/) and National Meteorological Science Data Center (http://data.cma.cn/data/) in 2020. Anthropogenic factors include population density (PD) and production that may have a direct impact on the environment in the traditional view. Gross domestic product (GDP) is calculated by the production approach rather than the income or expenditure method in China, it is for this reason that the study adopted annual GDP to reflect productive activities. With regarding to natural factors, there are nearly 700 meteorological stations in China Mainland can be used this year. For each station, the study extracted a representative value (mean or total) of each meteorological factor quarterly, including the altitude (ALT), mean wind speeds (WIN), mean relative humidity (RHU), mean atmospheric pressure (PRS), mean global surface temperature (GST), mean air temperature (TEM), accumulated duration of sunshine (SSD) and accumulated
precipitation (PRE) at the location of stations, since the change of these factors highly coincides with the seasonal change. Usually, regarded March to May as spring, June to August as summer, September to November as autumn, and December, January and February for winter in the Northern Hemisphere. This study also follows the convention. Overview of geographical and meteorological factors as shown in Figure 2. The seasonal average of meteorological factors from stations were interpolated into 10 by 10 km square grids covering the study area through ordinary kriging method.

Figure 2. Overview of meteorological stations and natural factors

2.2 Method

2.2.1 Correlation Analysis Method

Pearson correlation coefficient is one of the most commonly used correlation indicators. It is a statistical concept used to measure the correlation degree of two variables. Usually, p-value is used for testing statistical hypothesis, and $p \leq 0.05$ indicates the correlation is significant. On the premise of meeting the significant test, the closer the coefficient is to $|1|$, the stronger the collinearity of the two variables, which means that there is a high positive or negative correlation between them. Instead, they are probably linearly independent of each other. However, linearly independent is not equal to no association between paired data, and the Spearman’s rank coefficient can be utilized to reveal whether there is a non-linear relationship. Based on mechanism of these two methods, Pearson’s approach works and then so does Spearman’s, it can be concluded that variables are linear dependent. Alternatively, Pearson correlation coefficient is close to 0 but Spearman’s method works, indicating that there is a monotone and non-linear relationship between variables. And when the data are roughly elliptically distributed, the Pearson and Spearman correlation give similar values that implied insignificant association between variables.

It is noteworthy that quarterly correlation coefficient should be calculated directly rather than
extract quarterly average through the daily correlation coefficient. This corresponds to an alternative explanation of Pearson’s, that is, the numerator measures the trend consistency between variables, and the denominator judge the dispersion degree of each variable. For instance, there are two pairs of arrays representing PM2.5 and O3 concentrations and relation for two days, respectively, as shown in Table 1. And it should be noted that the data here is not actual measurements, but abstracted and exaggerated for illustration.

| Table 1. Comparison of correlation coefficient calculation |
|----------------------------------------------------------|
|                | DAY_1                  | DAY_2                  | DAY_1 and DAY_2             |
| PM2.5          | 10, 11, 15, 13, 14     | 100, 110, 150, 130, 140| 10, 11, 15, 13, 14, 100, 110, 150, 130, 140 |
| O3             | 1, 2, 4, 3.5           | 1, 2, 4, 3.5           | 1.0, 2.0, 4.0, 3.0, 3.5, 1.0, 2.0, 4.0, 3.0, 3.5 |
| \( r_{xy} \)   | 0.9911901              | 0.9911901              | 0.1736953                    |
| Correlation    | Strong Positive        | Strong Positive        | Very Weak Positive           |

### 2.2.2 Geographical Detector Model

Geographical detector is a new statistical model to detect spatial heterogeneity and reveal the driving factors behind it. This method has clear physical meaning and no linear hypothesis. It based on the assumption that if an independent variable has an important impact on the dependent variable, the spatial distribution of them should be similar, which can be described by \( q \)-statistic, as shown in Equation 1:

\[
q = 1 - \frac{\sum_{h=1}^{L} N_h \sigma^2_h}{\sigma^2} = 1 - \frac{SSW}{SST} \tag{1}
\]

\[
SSW = \sum_{h=1}^{L} N_h \sigma^2_h \quad \text{or} \quad \sum_{h=1}^{L} \sum_{i=1}^{N_h} (Y_{hi} - \bar{Y}_h)^2 \tag{2}
\]

\[
SST = \sum_{i=1}^{N} (Y_i - \bar{Y})^2 \tag{3}
\]

where \( h = 1, 2, 3, \ldots, L \) refers the strata or classes of spatial phenomenon, \( N_h \) is the number of parcels in \( h \)th class, \( \sigma^2_h \) is the variance within \( h \)th class, \( N \) is the total number of parcels, \( \sigma^2 \) is the total variance, \( SST \) and \( SSW \) are statistical terms representing total variation and within group variation, respectively.

The physical meaning of \( q \)-value depends on the division approach of strata (or class) applying to \( SSW \) part. There are two perspectives to explain it. First, depending on \( Y \) itself completely, and \( q \)-value is exactly the goodness of variance fit (GVF), which normally used to evaluate classification methods, through the smaller the intra-strata difference and the larger the inter-strata difference. The second explanation is the most common uses of geographical detector method (GDM) that to explore the similarity of spatial distribution between explanatory variable and target variable, as shown in Equation 2 and 3.

In addition, this study considers that the \( SSW \) part and \( SST \) part of the equation are not required to use the value of dependent variable \( Y \) rigidly. It means that Equations 1, 2, 3 can be calculated with the independent variable \( X \). Especially, the packet mood of \( X \) is determent by \( Y \), which ensures that it can be used to verify the spatial distribution similarity of \( X \) and \( Y \).
As for how to confirm those variables are the influencing factors and the others are determined, it can be judged by common sense in many circumstances. This formula also contains an implicit limit thought in calculus that $q$-value probably increases with L when L is large enough and they both in the condition of optimum stratification, ultimately L approaches N meanwhile $q$-value approaches 1. Such a view required that the choice of L should be considerate, because different results may be obtained with the same data, which also affect the further analysis according to the $q$-value in the study.

The geographical detector is designed for estimating power of determinants regarding the theory of spatial stratified heterogeneity. On this basis, the model is further developed to be used for revealing interactive influence (Interaction Detector), calculating the average values in each stratum of determined variable (Risk Detector), and testing significant differences between two influencing factors (Ecological Detector). All of these can be implemented from R package[18]. Significantly, there are some factors with small $q$-values originally and their weights of effect are amplified after interactive influence detector. Two kinds of enhancement effects are appeared in the results, named *enhance, bi-* and *enhance, nonlinear-* by Wang et al. [19] respectively. The former term means that the interactive influence of two factors is greater than the maximum of them. The latter means that the interactive influence of two factors is greater than the sum of both, which is the strongest case in the detector. This research utilized geographical detector model to determine the influence of those factors, including natural and anthropogenic, on atmospheric phenomenon separately and jointly.

### 2.2.3 Multiscale Discretization Method

The principle of geographical detector model involves stratification, which requires to discrete the input continuous variables. There are various classical discretization ways, such as k-means method, Jenks natural breaks method (one and the same as k-means when dealing with one-dimensional data), equal interval method, standard deviation method (suitable for data with normal distribution), geometrical interval method (developed by ESRI co.) etc. Beyond these approaches, multiscale discretization (MSD) method proposed by Meng et al., considering both independent variable X and dependent variable Y to except the optimal discretization results when using geographical detector model[20].

Starting from the Equation 1, the essence of MSD method is to employ exhaustive search method to minimize SSW part for achieving the highest $q$-value which means stratified variables optimally. More specifically, assuming that N different values are classified into L strata, thus $p = L - 1$ cut points exist. Searching for all possibilities, there are $C_{N-1}^p$ kinds of ways to select cut points and therefore obtain $C_{N-1}^p$ $q$-values. After comparison, the best data discretization algorithm with the largest $q$-value is acquired. It is absolutely accurate. There is still one small problem that the research cases usually contain a large amount of data, reducing the algorithm complexity from binomial coefficient to an acceptable range and balancing the accuracy of the results are urgently necessary. Accordingly, researchers Meng et al. continue to push forward on this basis, established an alternative route named Upscaling
and Downscaling, as shown in Table 2.

### Table 2. Procedure of MSD method

| Sequences | Max Strata | Best Cut Points in This Step | Algorithm Complexity |
|-----------|------------|------------------------------|---------------------|
| Original Data | X₁, X₂, X₃, X₄, ..., Xₙ | N \ (hard to calculate) | CₚN⁻¹ |
| Upscaling Results | ⌊X₁/k⌋, ⌊X₂/k⌋, ⌊X₃/k⌋, ..., ⌊Xₙ/k⌋ | N/k | CₚN/k⁻¹ |
| Downscaling Results | X₁, X₂, X₃, X₄, ..., Xₙ | p*k | Cₚk¹ + Cₚk² + ... + Cₚkᵢ |

In principle, multiscale discretization method is used to stratified independent variable X for minimizing the intra-strata difference and maximizing the inter-strata difference of the corresponding dependent variable Y. This is a solid foundation to use geographical detector model. Besides that, MSD procedures excelled at discretizing integers. As this study did for part of variable X, including WIN, PRS, GST, TEM, SSD and PRE, adjust the units to enlarge them before using this method.

#### 2.2.4 Random Forests

Random forest is an algorithm in prediction that established by Breiman, 2001[21]. Generally speaking, the essence of random forest is the integration of multiple decision trees. The data structure of each tree is determined by randomly selecting samples, and the prediction result of each tree is obtained by randomly selecting the characteristics of m-dimension. Then the results of multiple decision trees are voted or averaged to acquire the final outcome. The above two randomness ensure the robustness of the algorithm to the influence of noise.

More specifically, the first randomness of the algorithm avoid overfitting in prediction, which is based on the Law of Large Numbers. Since bootstrap sampling is employed in this process, when the sample size and sampling frequency are large enough, the following equation could be established:

$$\lim_{n \to \infty} \left(1 - \frac{1}{n}\right)^n = \frac{1}{e}$$

(4)

where \(n\) is the sample size and sampling frequency (they could use same representation when they are tend to infinite). And the whole formula means about one-third of the case are still left out after \(n\) rounds of sampling, which are called out-of-bag (OOB) samples. The OOB samples could be used for cross validation and fulfilling prediction. Back to the argument, it is precisely because of the existence of OOB samples are equivalent to the incomplete use of the training sets, which guarantee random forest to shield from most of the overfitting.

The second randomness of this algorithm involves the selection of the number of sample
features. Then a specific number of feature subsets (m-dimension) are randomly selected from the sample. In this process, the correlation within forest increases since the overlapping parts of trees become larger if m-dimension is too big. On the contrary, the prediction ability of a single decision tree is weak due to insufficient information on features. Neither of these situations are beneficial to obtain the optimal prediction results. Therefore, it is desirable to use the exhaustive search method to make the optimal selection of m-dimension under appropriate circumstances.

All machine learning prediction methods involve the selection on features of input variable. This study considers that there are two possibilities lead to the final prediction results are similar by using intact variable and its feature subset separately. The first is the principle of some methods, including random forest, which is destined to be insensitive to feature loss and missing values. The other is that the feature subset is more dominant for the generation of phenomena than other features of the input variable. Hence, the exact reason needs to be confirmed with other methods.

This study considered all determinants, including ALT, WIN, RHU, PRS, GST, TEM, SSD, PRE, PD and GDP, as input variables of random forest to predict correlation between PM2.5 and O3 firstly. And then utilizing subset of these above, which were detected by geographical detector model as the main influencing factors, to make prediction in the same route.

3. Result

3.1 The Seasonal and Spatial Patterns of the Correlation Between PM2.5 and O3

According to Pearson and Spearman correlation method, this study analyzed the 24-hour smoothing values of PM2.5 and O3 in China mainland AQI stations that meet the threshold setting (missing data less than 10%) from 2015 to 2020. However, in the calculation process of Pearson and Spearman, there are some sites that fail to significance test (p≤0.5) need to be excluded from the results, as listed in Table 4.

The Pearson’s and Spearman’s correlation value both range from -1 to 1, and the presence of a relationship between variables is primarily determined by this value. This study divided this value into three main intervals, in which [-1, -0.2] refers to the negative linear correlation, (0.2, 1] refers to the positive linear correlation, and (-0.2, 0.2] refers to almost uncorrelation. Accordingly, these three intervals are minced to seven sub-categories, as shown in Figure 3 and Figure 4, which described the strength of the correlation using the following guideline for the value of \( r_{xy} \) and \( \rho \)[22].
Table 3. Guideline for interpreting correlation coefficient

| Range of correlation coefficient | Interpretation            |
|----------------------------------|---------------------------|
| -1.00 to -0.60                   | strong negative           |
| -0.59 to -0.40                   | moderate negative         |
| -0.39 to -0.20                   | weak negative             |
| -0.20 to 0.20                    | very weak, almost uncorrelated |
| 0.21 to 0.40                     | weak positive             |
| 0.41 to 0.60                     | moderate positive         |
| 0.61 to 1.00                     | strong positive           |

The above results basically verified the previous research conclusions by processing multi-year and large-scale air quality data, and obtained further discoveries. Pearson’s and Spearman’s results are relative similar, which is not surprising in areas with strong correlation, but it can be asserted that there are very weak associations (whether linear or non-linear) between PM2.5 and O3 in those areas with coefficient approximate to 0. Hence, this study uses Pearson’s results directly in the follow-up exploration process. Furthermore, there are some new findings have emerged. The approximate dividing lines between positive correlation and negative correlation are similar to Hu line. But unlike Hu line, these dividing lines both move with seasonal variations from 2015 to 2020, which are named dynamic Simul-Hu lines in this study. Here’s another way to look at this phenomenon. From southeast to northwest of China, the correlation between PM2.5 and O3 actually changes from positive to weak correlation and then to negative, while the positions (or lines) of this transition are diverse in different seasons. From 2015 to 2020, the number of AQI sites is increasing but the regular pattern of each year is basically the same. It should be noted that AQI stations are mostly located in regions with high degree of urbanization depend on their own purpose and sparsely distributed in Northwest China. Therefore, although the division line in summer cannot be directly sought as other seasons in Figure 3 and Figure 4, the correlation coefficient is basically decreasing in numerical order from southeast to northwest.

Table 4. Number and distribution type of AQI stations

| Year | Season | Number of AQI stations | missing data less than 10% | significant correlation of Pearson | significant correlation of Spearman |
|------|--------|------------------------|----------------------------|-----------------------------------|-------------------------------------|
| 2015 | Spring | 1500                   | 1434                       | 867                               | 864                                 |
|      | Summer | 1500                   | 1409                       | 1392                              | 1392                                |
|      | Autumn | 1500                   | 1411                       | 840                               | 807                                 |
|      | Winter | 1500                   | 1398                       | 832                               | 827                                 |
| 2016 | Spring | 1500                   | 1413                       | 1362                              | 1370                                |
|      | Summer | 1500                   | 1380                       | 1027                              | 1026                                |
|      | Autumn | 1500                   | 1380                       | 1321                              | 1336                                |
|      | Winter | 1500                   | 1381                       | 1339                              | 1343                                |
| 2017 | Spring | 1579                   | 1455                       | 1384                              | 1391                                |
| Year | Season     | 2018       | 2019       | 2020       |
|------|------------|------------|------------|------------|
| 2018 | Summer     | 1583       | 1608       | 1677       |
|      | Autumn     | 1596       | 1608       | 1673       |
|      | Winter     | 1600       | 1609       | 1706       |
| 2019 | Spring     | 1608       | 1608       | 1677       |
|      | Summer     | 1608       | 1644       | 1673       |
|      | Autumn     | 1608       | 1644       | 1673       |
|      | Winter     | 1609       | 1644       | 1706       |
| 2020 | Spring     | 1608       | 1644       | 1677       |
|      | Summer     | 1608       | 1644       | 1673       |
|      | Autumn     | 1609       | 1644       | 1673       |
|      | Winter     | 1609       | 1644       | 1706       |
Figure 3. Correlation between PM2.5 and O3 calculated by Pearson’s method
Figure 4. Correlation between PM2.5 and O3 calculated by Spearman's method
3.2 The Determinants of the Correlation Distribution Between PM2.5 and O3.

3.2.1 Discretization of the Influencing Factors

As anthropogenic factors, population density and annual GDP were discretized according to the spatial granularity of prefecture-level city that kept the consistency with reality. Meanwhile, urban development was divided according to the GDP, and differentiated urban scale from population, as shown in Figure 5. These two factors can be regarded as stable elements in the year because their seasonal variations are negligible compared with some natural factors.

As mentioned above, this study picked up 8 natural influencing factors that obtained from meteorological stations, including ALT, WIN, RHU, PRS, GST, TEM, SSD and PRE in 2020. But it is hard to directly match the concentration of atmospheric pollutants and meteorological data for further analysis, since the stations measuring these two types of data are often not in the same location. According to the distribution type calculation[23], the meteorological stations are almost evenly allocated in China Mainland. Therefore, this study adopted Thiessen polygons of meteorological sites as the units to associate AQI data.

With regarding to discretizing the relatively continuous natural factors in space, this study adopts the MSD method referred before. For research purposes, the independent variable X of MSD method is designated as natural factors, and the dependent variable Y is the Pearson correlation coefficient between PM2.5 and O3. Since the correlation coefficient is divided into three intervals on the basis of its statistical meaning, even if its continuous numerical results are used in the exploration process, the number of the intervals is still a useful guideline in independent variables discretization. It should be noted that if stratified conventions and criteria has already existed, such as Three Gradient Terrain of China based on altitude and wind speeds measured by Beaufort scale, the breaks are identical all-round year. Especially, for those factors without generic standards the stratifications possibly change with season as shown in Figure 5 because the breaks generated by MSD method are used for optimizing discretization and lead to accurate GDM results, instead of creating a unified standard for each factor to demonstrate the temporal and spatial features.

3.2.2 Estimating Power of Determinants

By mapping the influencing factors to Pearson correlation coefficient that extracted from official statistics and calculated by AQI stations’ data, and stratifying these factors to use geographical detector model, there are two results obtained quarterly as follows: (1) $q$-values measured the determinant power and identified dominant factors by their sorted order, as detailed in Table 5. More specifically, there is no obvious dominant factor and the influences of RHU, GST, TEM, SSD and PRE are relatively balanced in spring; RHU and PRE are the deciding factors in summer, and other natural elements except WIN maintain about 10% - 20% explanatory power for the spatial distribution of correlation between PM2.5 and O3;
GST, TEM and PRE are the crucial factors in autumn, and RHU also contributes considerably; GST and TEM play a prominent role in winter, the similarity of these two factors and correlation on spatial distribution is about 40% respectively. It also can be seen intuitively from Figure 3, Figure 4 and Figure 5 that the part with positive correlation coefficient between PM2.5 and O3 often corresponds to relatively more densely population, higher GDP, lower altitude, higher humidity, higher atmospheric pressure, higher surface temperature, less sunshine hours and more accumulated precipitation, no matter in which season. The part with negative correlation coefficient is opposite to the above. From the perspective of data results, the magnitude of q-values reflected the degree of corresponding between influencing factors and the distribution of correlation coefficient. The larger the q-values, the better the correspondence, which further indicated that the influencing factors have stronger explanatory power for the phenomenon.

(2) interaction detector indexes revealed interactive influence, which discriminated the pair-wise factors whether weaken or strengthen determinant power through comparing the q-values of individuals and combination, as illustrated by Figure 7. Size of the bubble represents interactive influence index, while the dark color represents enhance, nonlinear- and the light color represents enhance, bi- . Generally speaking, the amplification effect is obvious when natural factors interact with anthropogenic factors. In addition, wind speeds also contribute to the enhancement of the influence of all other factors.

3.3 Spatial prediction Results by Random Forest

Through the results of geographical detector model, it can be seen that the listed intact features of independent variable, including 8 natural factors and 2 anthropogenic factors, contributed to the dynamic Simil-Hu lines. And according to Table 5 and Figure 7, RHU, GST, TEM, PRE as the feature subset, are dominant factors of the phenomenon from beginning to end of the year. Therefore, the intact features and feature subset are utilized separately with the corresponding existed Pearson correlation coefficients as training sets of random forest. Then the correlation between PM2.5 and O3 in the whole Chinese mainland are predicted.

Ultimately, the results include two parts, as shown in Figure 8. Firstly, through the prediction by intact features, it can be seen that dynamic Simil-Hu lines move with changing seasons: the advance and retreat from southeast to northwest. Secondly, feature subset was exploited and the predicted distributions of correlation between PM2.5 and O3 in the study area are basically similar to the results in the first part. Therefore, mutual corroboration between random forest and geographical detector model indicating that RHU, GST, TEM, PRE are the main meteorological factors contributed to dynamic Simil-Hu lines.
Figure 5. Anthropogenic and natural factors stratified by generic standards and MSD method.
Table 5. The q-values of the factor detector

| Influencing Factors | SPRING          | SUMMER          | AUTUMN          | WINTER          |
|---------------------|-----------------|-----------------|-----------------|-----------------|
|                     | q-value | p-value | q-value | p-value | q-value | p-value | q-value | p-value |
| Anthropogenic Factors | PD     | 5.35%    | 5.48E-10 | 12.75% | 7.40E-11 | 7.58%    | 2.54E-10 | 5.88%    | 1.98E-10 |
| GDP                 | 2.54%    | 1.31E-05 | 2.19%    | 1.43E-03 | 0.93%    | 1.45E-02 | 1.01%    | 6.57E-03 |
| ALT                 | 2.38%    | 1.79E-07 | 15.38%   | 6.35E-11 | 5.52%    | 8.44E-11 | 5.91%    | 2.27E-10 |
| WIN                 | 1.34%    | 6.75E-03 | 0.72%    | 8.19E-01 | 1.88%    | 7.14E-05 | 1.08%    | 5.14E-03 |
| RHU                 | 12.28%   | 8.53E-10 | 38.10%   | 9.09E-10 | 28.18%   | 3.67E-11 | 18.03%   | 9.41E-10 |
| Natural Factors     | PRS     | 4.44%    | 4.91E-11 | 19.69% | 6.16E-10 | 12.65%   | 1.39E-10 | 13.64%   | 5.23E-10 |
| GST                 | 14.76%   | 7.54E-10 | 12.14%   | 5.18E-10 | 41.97%   | 1.57E-10 | 39.58%   | 5.29E-10 |
| TEM                 | 13.43%   | 6.69E-11 | 14.80%   | 1.66E-10 | 40.55%   | 1.37E-10 | 38.71%   | 3.28E-10 |
| SSD                 | 14.24%   | 3.13E-11 | 13.68%   | 3.18E-10 | 14.68%   | 7.62E-10 | 6.76%    | 6.63E-11 |
| PRE                 | 15.86%   | 1.02E-11 | 31.26%   | 5.05E-11 | 39.12%   | 1.67E-10 | 18.91%   | 7.03E-10 |

Figure 6. Main factors annually affecting the correlation between PM2.5 and O3

Figure 7. Results of interaction detector
Figure 8. Using intact features and feature subset to predict correlation between PM2.5 and O3
- by intact features
- by feature subset
4. Discussion

In initial studies, especially those with small temporal and spatial ranges, considered that there is a weak or even irrelevant correlation between PM2.5 and O3, which is often covered by the fluctuation caused by random factors. In the later researches, there are plenty attempts have been made to explore the correlation between PM2.5 and O3 and elucidate their temporal and spatial distribution characteristics. Based on multi-year and large-scale air quality data, this study validated the correlation by using Pearson’s method. Owing to PM2.5 and O3 have a certain relationship with human life style and production mood, the differences in their concentration between densely and sparsely populated areas are inevitably existed. According to Figure 3 and Figure 4, however, wide seasonal variations over many consecutive years are almost impossible originated by anthropogenic factors alone, there necessarily emerge the influence of natural factors that change with the seasons. There is, in addition, one further point to emphasize. An artificial specific separation and opposition in time (winter and summer) and space (South and North) summarized by those previous studies, but the mechanical division is difficult to explain the parts with very weak correlation. This study considered from the perspective of dynamic Simil-Hu lines, the temporal and spatial continuity of correlation between PM2.5 and O3 have both been preserved, which are solid foundations for subsequent analysis.

A total of two anthropogenic factors and eight natural factors were selected as determinants in this study for investigating possible causes on temporal and spatial distribution of the correlation between PM2.5 and O3. Especially, these factors could be roughly divided into three parts in line with their features. The first part is related to geography and geomorphology, including altitude (ALT) and mean atmospheric pressure (PRS), which basically presented the analogous distribution with the Three Gradient Terrain of China and almost changing with the span of longitude. The second part is related to climate and meteorology, including mean relative humidity (RHU), mean global surface temperature (GST), mean air temperature (TEM), accumulated duration of sunshine (SSD) and accumulated precipitation (PRE), which vary with the span of latitude and seasonal changes violently. In the third part, there are certain corresponding relationship between influencing factors and determined variables in some areas while undefined in totality, including mean wind speeds (WIN) and two anthropogenic factors (PD and GDP) that do have an impact on PM2.5 and O3. In general, the seasonal dynamic Simil-Hu lines of division into the correlation between PM2.5 and O3 are formed by the geography and geomorphology with latitudinal differences, climate and meteorology with longitudinal differences and wind speeds with local characteristics seasonally. These three parts’ regular patterns also can be realized from the Figure 5 intuitively. As for the effect of these factors, it should be further analyzed quantitatively.

Obtained data stratification through generic standards and multiscale discretization method, then utilized geographical detector model to quantitate power of those natural determinants. In Table 5, it can be seen that the certain factors are more at \( q \)-values than others, and these factors are more like determinants on temporal and spatial features of the correlation between PM2.5 and O3. And these four factors, including RHU, GST, TEM, PRE, are mainly contribution to form the patterns that verified once again by using random forest. Meanwhile,
compared with Figure 3 and Figure 4, the dynamic Simil-Hu lines are shown explicitly in Figure 8. It should be noted that the combination of other factors besides these four majors can also explain and predict phenomenon to a certain extent, but they worked in some seasons and not in others. This is because they maintained powerful influences only throughout the year, just as shown in Table 5. According to Figure 7 and referring to three parts mentioned above, if factors varied greatly in distribution from one to another, the interaction effect between them tended to be larger than two similar elements. And it should be noted additionally that the impact of population density and GDP quantified by $q$-values are much smaller than expected. In the overall feeling and common sense, the population and GDP are subjected to the form of Hu line and they should be regarded as the stability factors and main causes for the distribution of correlation between PM2.5 and O3. However, although the human activities are largely distributed in the southeast of Hu line, as shown in Figure 5, they are not continuous and often concentrated in larger developed cities then radiate to drive the surrounding areas. In a single administrative unit, it depends more on human initiative factors even on the basis of the same natural conditions. For instance, the population and GDP of two adjacent administrative units with similar area and same terrain (i.e. controlling natural variables) are also different. Moreover, the types of human production activities (measured only by the GDP in this study) are diverse and different industrial types have different impact on the environment. Therefore, this study considered that the anthropological factors have a certain effect on concentration of PM2.5 and O3, but the relationship (or correlation) between these two air pollutants mainly determined by atmospheric conditions.

Back to the purpose of this study, quantified the impact of natural and anthropogenic factors on correlation between PM2.5 and O3, and to govern these two pollutants collaboratively. As mentioned before, natural phenomena have certain temporal and spatial continuity, and PRE, RHU, GST and TEM are key factors annually as shown in Figure 6. A good case in point is if there are natural factors that make PM2.5 and O3 increase or decrease simultaneously in a place for a certain period of time, it should be treated according to the positive correlation, rather than depending on whether it belongs to the South or the North and in which season at present. Because the granularity of time and space was set artificially in the research, but they are continuous and dynamic indeed. It does not exist that the correlation between PM2.5 and O3 changes from positive to negative suddenly on a certain day of the handover between spring and summer. If there is a local area with higher humidity and temperature than in previous years due to the abnormal climate, then a positive correlation between PM2.5 and O3 is likely to appear and corresponding measures should be taken flexibly. As mentioned above, in terms of whether PM2.5 and O3 have the same or opposite trend, the changing meteorological factors would amplify the results caused by human society. In summary, taking the quarter as the time granularity during analysis but do not have to stop there, air quality control measures should be taken in the light of circumstances.

5. Conclusion

This study verified the spatial differential characteristics of the correlation between PM2.5 and O3 in China mainland from 2015 to 2020, and named the dynamic Simil-Hu lines as the regular pattern of seasonal variation. Then geographical detector and random forest adopted to quantitatively explore the influencing factors of above phenomena. The results show that the influence of natural factors on the distribution of correlation between PM2.5 and O3 is greater
than that of population-economy factors, especially in terms of temperature and humidity. For those stakeholders of air pollution control, the influencing factors may be partially uncontrollable, but strengthen understanding and attention would make the collaborative governance process more scientific and effective: considering the temporal and spatial differences of collaborative control of PM2.5 and O3 pollution, formulate collaborative emission reduction schemes under different natural and anthropogenic conditions.

References

[1] Miao Y, Che H, Zhang X, et al. Relationship between summertime concurring PM 2.5 and O3 pollution and boundary layer height differs between Beijing and Shanghai, China[J]. Environmental Pollution, 2020.

[2] Wu J, Wang Y, Liang J, et al. Exploring common factors influencing PM2.5 and O3 concentrations in the Pearl River Delta: Tradeoffs and synergies[J]. Environmental Pollution, 2021, 285(1):117138.

[3] Xie Y, Zhao B, Lin Z, et al. Spatiotemporal variations of PM2.5 and PM10 concentrations between 31 Chinese cities and their relationships with SO2, NO2, CO and O3[J]. 中国颗粒学报: 英文版, 2015, 020(003):141-149.

[4] Chen J, Shen H, Li T, et al. Temporal and Spatial Features of the Correlation between PM2.5 and O3 Concentrations in China[J]. International Journal of Environmental Research and Public Health, 2019, 16(23).

[5] Jia M, Zhao T, Cheng X, et al. Inverse Relations of PM2.5 and O3 in Air Compound Pollution between Cold and Hot Seasons over an Urban Area of East China[J]. Atmosphere, 2017, 8(12):59.

[6] Nguyen G, Shimadera H, Uranishi K, et al. Numerical assessment of PM2.5 and O3 air quality in continental Southeast Asia: Baseline simulation and aerosol direct effects investigation[J]. Atmospheric Environment, 2019, 219:117054.

[7] Zhao S, Yin D, Yu Y, et al. PM2.5 and O3 pollution during 2015–2019 over 367 Chinese cities: Spatiotemporal variations, meteorological and topographical impacts[J]. Environmental Pollution, 2020, 264:114694.

[8] Liu Z, Qi Z, Ni X, et al. How to apply O3 and PM2.5 collaborative control to practical management in China: A study based on meta-analysis and machine learning[J]. Science of The Total Environment, 2021, 772(10):145392.

[9] Huang L, Sun J, Jin L, et al. Strategies to reduce PM2.5 and O3 together during late summer and early fall in San Joaquin Valley, California[J]. Atmospheric Research, 2021, 258(32):105633.

[10] Marinello S, Butturi M A, Gamberini R. How changes in human activities during the lockdown impacted air quality parameters: A review[J]. Environmental Progress & Sustainable Energy, 2021.

[11] DEVENDRAASIINGH, Baudddh K, Shah M A, et al. Temporary reduction in air pollution due to anthropogenic activity switchoff during COVID19 lockdown in northern parts of India[J]. Environment Development and Sustainability, 2020(11).

[12] Li H, Ma Y, Duan F, et al. Stronger secondary pollution processes despite decrease in gaseous precursors: A comparative analysis of summer 2020 and 2019 in Beijing[J]. Environmental Pollution, 2021, 279(2):116923.

[13] Duan W, Wang X, Cheng S, et al. Influencing factors of PM2.5 and O3 from 2016 to
2020 based on DLNM and WRF-CMAQ[J]. Environmental Pollution, 2021, 285(1):117512.

[14] Goodchild M F . The Validity and Usefulness of Laws in Geographic Information Science and Geography[J]. Annals of the Association of American Geographers, 2004, 94(2):300-303.

[15] Wang J F , Zhang T L , Fu B J . A measure of spatial stratified heterogeneity[J]. Ecological Indicators, 2016, 67:250–256.

[16] Hu H Y. Population geography of China, Volume I [M]. East China Normal University Press, 1984.

[17] Zhao J , Han Y B , Estimation of correlation significance levels after moving average[J]. Journal of Beijing Normal University(Natural Science) , 2005, 41(2).

[18] Song, Y., Wang, J., Ge, Y. & Xu, C. (2020) “An optimal parameters-based geographical detector model enhances geographic characteristics of explanatory variables for spatial heterogeneity analysis: Cases with different types of spatial data”, GIScience & Remote Sensing. 57(5), 593-610. doi: 10.1080/15481603.2020.1760434.

[19] Wang JF, Li XH, Christakos G, Liao YL, Zhang T, Gu X & Zheng XY. 2010. Geographical detectors-based health risk assessment and its application in the neural tube defects study of the Heshun region, China. International Journal of Geographical Information Science 24(1): 107-127.

[20] Xiaoyu Meng, Xin Gao, Jiaqiang Lei & Shengyu Li (2021) Development of a multiscale discretization method for the geographical detector model, International Journal of Geographical Information Science, 35:8, 1650–1675, DOI: 10.1080/13658816.2021.1884686

[21] Breiman L . Random forest[J]. Machine Learning, 2001, 45:5-32.

[22] Yu H, Gao M, Zhang H, Chen Y, Dynamic modeling for SO2-NOx emission concentration of circulating fluidized bed units based on quantum genetic algorithm - Extreme learning machine, Journal of Cleaner Production (2021), doi: https://doi.org/10.1016/j.jclepro.2021.129170.

[23] Zhu G R , Zhang G S , Map Analysis[M]. Surveying and Mapping Publishing House, 1994, 99-101.