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1. Calculation of PM$_{2.5}$ concentrations

1.1 Satellite-derived aerosol optical depth (AOD)

The assumption is that the surface of the study area is a Lambert surface and levels of gases in the atmosphere is uniform. The top of the atmospheric apparent reflectance $\rho_{\text{toa}}$ can be approximated by [1]:

$$\rho_{\text{toa}}(\theta_s, \theta_v, \phi) = \rho_0(\theta_s, \theta_v, \phi) + T(\theta_s) \cdot T(\theta_v) \cdot \frac{\rho_s(\theta_s, \theta_v, \phi)}{[1 - \rho_s(\theta_s, \theta_v, \phi) \cdot S]}$$  \hspace{1cm} (1)

where $\theta_s$, $\theta_v$, and $\phi$ are solar zenith angle, satellite zenith angle, and solar/satellite relative azimuth angles, respectively. $\rho_0$ is the atmospheric path reflectance; $\rho_s$ is the angular surface reflectance; $\rho_s$ is the function applied to $\theta_s$, $\theta_v$, and $\phi$; $r$ is the reflectance of the Lambert surface; $S$ is the atmospheric backscattering ratio; and $T$ is atmospheric transmission. As we can see from Equation (1), $\rho_0$, $T(\theta_s) \cdot T(\theta_v)$, $\rho_s$, and $S$ are unknown parameters. We have to quantify aerosols to resolve two key problems:

In the first step, we used 6S simulation of different optical thicknesses of the lookup table (LUT) to solve $\rho_0$, $T(\theta_s) \cdot T(\theta_v)$, $\rho_s$, and $S$. In the second step, we calculated surface reflectance. Kaufman et al. proposed the dark pixel method to eliminate the contribution from surface reflection noise. Kaufman et al. observed that, over vegetated surfaces, the surface reflectance in some visible (VIS) wavelengths correlated with the surface reflectance at 2.12 $\mu$m, and in fact, were nearly ratios in the 2.12 $\mu$m channel. Levy et al. discussed surface reflectance assumptions and introduced the V5.2 algorithm. This algorithm uses a new VIS/SWIR ratio to obtain the surface reflectance of visible wavelengths. The relationship between VIS/SWIR and $\rho_s^{2.2}$, $NDVI_{\text{swir}}$, and $\phi$ is defined as follows:

$$\rho_s^{\text{red}} = \rho_s^{2.2} \cdot \text{slope}_{\text{red/2.2}} + \text{yint}_{\text{red/2.2}}$$  \hspace{1cm} (2)

$$\rho_s^{\text{blue}} = \rho_s^{\text{red}} \cdot \text{slope}_{\text{blue/red}} + \text{yint}_{\text{blue/red}}$$  \hspace{1cm} (3)

where

$$\text{slope}_{\text{red/2.2}} = \text{slope}_{\text{x/2.2}} + 0.002 \phi - 0.27$$  \hspace{1cm} (4)

$$\text{yint}_{\text{red/2.2}} = 0.00025 \phi + 0.033$$  \hspace{1cm} (5)

$$\text{slope}_{\text{blue/red}} = 0.49$$  \hspace{1cm} (6)

$$\text{yint}_{\text{blue/red}} = 0.005$$  \hspace{1cm} (7)

where, in turn

$$\phi = \cos^{-1}(-\cos \theta_s \cos \theta_v + \sin \theta_s \sin \theta_v \cos \phi)$$  \hspace{1cm} (8)

$$\text{slope}_{\text{x/2.2}} = \begin{cases} 0.48 & \text{if } NDVI_{\text{SWIR}} < 0.25 \\ 0.58 & \text{if } NDVI_{\text{SWIR}} > 0.75 \\ 0.48 + 0.2 \cdot (NDVI_{\text{SWIR}} - 0.25) & \text{if } 0.25 < NDVI_{\text{SWIR}} < 0.75 \end{cases} \hspace{1cm} (9)$$

$$NDVI_{\text{swir}} = \frac{(\rho_{\text{toa}}^{1.64} - \rho_{\text{toa}}^{2.2})}{(\rho_{\text{toa}}^{1.64} + \rho_{\text{toa}}^{2.2})}$$  \hspace{1cm} (10)

where $\theta_s$, $\theta_v$, and $\phi$ are the solar zenith, the satellite zenith, and the relative azimuth angles (between the sun and satellite), respectively.

The Landsat8 OLI sensor provides global images of the visible wavelength band, near infrared band, and shortwave infrared band at 30 meters resolution, which have been used in urban environmental quality monitoring. In this paper, the short-wave infrared channel (2.10-2.30) was used to obtain the ground reflectivity of the red and blue bands to realize decoupled
earth–atmosphere and Landsat8 was used to quantify terrestrial aerosols using the dark target method.

(1) Determination of dark pixels and calculation of surface reflectance

SWIR (2.2 μm) can be used to identify dark pixels [2]. Since the relationship between reflectance in the visible and the 2.2μm channels is chaotic, reflectances in the 2.2μm band that are less than 0.15 are usually considered dark pixels, while reflectances in the 2.2μm band that are greater than 0.15 are not considered dark pixels [3].

Levy [13] showed that the reflectivity of dense vegetation at a wavelength of 2.1μm and the reflectance at a wavelength of 0.47μm and a wavelength of 0.66μm. This relationship is not only related to the scattering angle [4], but also to the canopy of vegetation. The scattering angle can be derived from Equation (3). The degree of vegetation is usually expressed as a normalized vegetation index, but the normalized vegetation index calculated using the red and near infrared bands is affected by aerosols. Considering that the shortwave infrared band is less affected by aerosols, we used shortwave infrared to calculate the normalized vegetation index NDVI_{swir}. The formula is shown in Equation (5). The surface reflectance of the red and blue bands can be obtained by calculating the surface reflectance in the 2.1μm band. The formula is shown in Equation (4).

(2) Construction of the lookup table (LUT)

The construction of the LUT is a key step in determining aerosol optical thickness. We used the second simulation of the satellite signal in the solar spectral radiative transfer code to construct the LUT [1].

As the Landsat8 satellite imaging range is small and under the observation limits of the satellite, its solar zenith angle and azimuth, satellite zenith angle, and azimuth angle can be considered constant. Where the solar zenith and azimuth can be obtained from the image header file, the satellite zenith angle and the azimuth angle can be regarded as a constant 0°. Therefore, this paper does not consider changes in the sun zenith angle and azimuth, satellite zenith angle, and azimuth.

The relevant parameters of the LUT are set to: aerosol type=custom, atmospheric mode=tropical atmosphere, altitude=0.03km, and time=imaging date. The spectral parameters are set to the blue and red bands of the Landsat8 OLI/. The aerosol optical thickness varies from 0.001 to 1.95, with an interval of 0.1.

In order to improve the accuracy of the LUT, we used the 6S model to define the spectral parameters of the sensor’s blue and red bands, because the 6S model does not have a standard definition of the OLI sensor’s spectral response function. The process involves two steps: first we entered the wavelength range of the blue and red bands, and then input the spectral response function of the blue and red bands with an interval of 0.0025μm.

(3) AOD calculation

First, we entered the parameters into the inversion program; the parameters include sun azimuth, observation zenith angle, observation azimuth, aerosol type, and observation date. Second, we generated a LUT of parameters, such as $\rho_0 \cdot (T(\theta_s) \cdot T(\theta_a))$, S, and other parameters containing different aerosol thicknesses in combination with the 6S radiation transmission
model. Third, we calculated $\rho_{\text{toa}}$ using the radiation calibration calculation. Fourth, we used the V5.2 algorithm to calculate the red and blue bands of surface reflectivity. Fifth, we substituted the equivalent reflectance $\rho_0$ of the red and blue bands, the hemispheric reflectance $S$ of the Lambertian surface reflectance $r$, and the lower hemisphere reflectivity $S$ of the atmosphere into formula (2) to solve the assumed apparent reflection rate. Finally, we compared the assumed apparent reflectance to the true apparent reflectance and used the LUT to find the apparent reflectance value with the smallest difference. The corresponding aerosol optical thickness in the same row is the value of the pixel.

(4) AOD spatial interpolation

Finally, spatial interpolation is required to produce the full AOD image. Ordinary kriging is a widely used geostatistical interpolation technique [5]. In this paper, AOD was interpolated using the ordinary kriging method in ArcGIS. See the supporting information for details.

1.2 Calculation of PM$_{2.5}$ from AOD data

To determine ground-level PM$_{2.5}$ concentrations from AOD, vertical and humidity corrections are required. The concept of scale height is incorporated into the vertical correction, while the effects of hygroscopic growth, mass extinction efficiency (MEE), and finemode fraction (FMF) are incorporated in the humidity correction. The calculation consists of three parts:

(1) Vertical correction for AOD

AOD is a columnar measurement of aerosol light extinction. We assumed a plane parallel atmosphere. To connect AOD with ground-level PM$_{2.5}$ concentrations, the relationship between AOD and the surface aerosol extinction coefficient needs to be elucidated [6]. By assuming a negatively exponential form for the vertical distribution of the aerosol extinction coefficient, AOD can be derived using the following equation [7]:

$$ AOD = \int_0^{\infty} k_{\text{ex}} dz = k_{\text{ex},0} \cdot H $$

where $k_{\text{ex},0}$ is the surface aerosol extinction coefficient at 0.55 μm and $H$ represents the aerosol scale height. $H$ can be estimated by empirical relationship [8]. The equation is expressed as:

$$ k_{\text{ex},0} = \frac{3.91}{\text{VIS}} $$

where $k_{\text{ex},0}$ is the surface aerosol extinction coefficient at 0.55 μm and VIS is visibility.

We used AOD and visibility data to calculate $k_{\text{ex},0}$, where the visibility data comes from the Jinjiang Meteorological Bureau.

(2) RH correction of the aerosol extinction coefficient ($k_{\text{ex},0}$)

We can define a hygroscopic growing factor, $f$(RH), as the ratio of “wet” $k_{\text{ex},0}$, obtained at the ambient RH, to “dry” $k_{\text{ex},0}$, obtained under relatively dry conditions (e.g. at an RH below 40%). And based on previous studies [9], the $f$(RH) used in this paper can be represented as:
\[ f(RH) = (1 - \frac{RH}{100})^{-g} \]  
where \( g \) is an empirical fit coefficient. Hence the “dry” \( k_{ex,0} \) is obtained through the RH correction:

\[ k_{AOD,\text{dry}} = k_{ex,0} \cdot \frac{1-RH}{1-40\%} \]  

where \( k_{AOD,\text{dry}} \) represents the aerosol extinction coefficient in dry conditions and \( RH \) is set at 40\%, the reference \( RH \) value in dry conditions.

### (3) Correlation between the corrected \( k_{AOD,\text{dry}} \) and PM\(_{2.5}\) concentrations

Wang [29] found that \( k_{AOD,\text{dry}} \) and PM\(_{2.5}\) concentrations show a linear correlation. We drew a scatter plot which confirms this linear correlation between \( k_{AOD,\text{dry}} \) and PM\(_{2.5}\) concentrations. Based on the above analysis, we established linear correlative models between \( k_{AOD,\text{dry}} \) and PM\(_{2.5}\) concentrations. The equation is:

\[ PM_{2.5} = a_1 k_{AOD,\text{dry}} + b_1 \]  

where \( a_1 \) and \( b_1 \) are fit coefficients. These coefficients were obtained through the linear regression based on the vertical-RH-corrected AOD and the corresponding PM\(_{2.5}\) concentrations.

Due to a low number of environmental monitoring sites, we used leave-one-out cross validation to validate the Landsat-estimation of PM\(_{2.5}\). Our results indicated that the Landsat-estimation of PM\(_{2.5}\) correlates with ground-based measurements on 13 December 2014, 29 December 2014, and 14 January 2015, with \( R^2 = 0.68, 0.66, \) and 0.72; MAE=4.31, 3.08, and 8.39; and RMSE=5.38, 3.83, and 10.465, respectively.

### 2. Geographical detector model

The geographical detector model detects various factors that influence urban PM\(_{2.5}\) concentrations, the degree of influence of each factor, and the interactions between factors based on spatial analysis of variance [10-11]. The model integrates the power of the determinant indicator, logical inference, and existing geostatistical techniques [12] and has been used to comprehensively analyze the processes that influence ecological forecasting. The model has obvious benefits over traditional models that only rely on specific types of variables, thereby limiting data input. In contrast, the geographical detector model processes both categorical and numerical variables, in addition to providing quantitative calculations. Furthermore, the model quantitatively expresses various types of interactions and directions among different variables. It is a spatial heterogeneity hypothesis-based approach suitable for studying spatially heterogeneous ecological regions. Thus, the model may be applied to study the effects of multiple ecological factors and analyze differences in spatial heterogeneity. Influential ecological factors can be identified and the interactions among them can be quantified.

Assuming that human activities and certain ecological factors jointly influence PM\(_{2.5}\) concentrations, the temporal and spatial distribution patterns of PM\(_{2.5}\) concentrations can be
determined because they are similar to those of ecological factors. The geographical detector model, which is based on the consistency of variable spatial patterns, is freely available at http://www.sssampling.org/Excel-geodetector/. Geographical detector models have been successfully used to identify determinants and their interactions involved in neural tube defects, mortality in children under 5 years old, and fluoroquinolone residues in soil. These models typically include a factor detector, an ecological detector, and an interaction detector. The factor detector is used to explore the impact of different factors on the research target, the ecological detector is used to explore the impacts of different levels of significance on those factors, and the interaction detector is used to explore the impacts of combinations of different impact factors on the research target.

(1) **Factor detector**

This detector quantifies the impact of a factor $D$ on the observed spatial pattern of PM$_{2.5}$ concentrations residues, using $PD$, the dispersion variance ($\sigma^2_{D,p}$) of pooled data of the entire region, and the summation of stratified dispersion variance ($\sigma^2_{D,x}$). Therefore, the $PD$ value of factor $D$ can be expressed using Equations (16–18):

$$PD = 1 - \frac{\sigma^2_{D,p}}{\sigma^2_{D,x}}$$  \hspace{1cm} (16)

$$\sigma^2_{D,p} = \frac{1}{n_{D,p}} \sum_{p=1}^{n_{D,p}} (y_{D,p} - \bar{y}_D)^2$$  \hspace{1cm} (17)

$$\sigma^2_{D,x} = \sum_{z=1}^{L} \frac{R_z}{n_{x,p}} \frac{1}{n_{x,p}} \sum_{p=1}^{n_{x,p}} (y_{x,p} - \bar{y}_x)^2$$  \hspace{1cm} (18)

where $\bar{y}_D$ and $\bar{y}_x$ refer to the average FQs residues within the entire region with area $\hat{A}$ and a specific subregion with area $\hat{A}_x$ stratified by $D$, respectively.

(2) **Interaction detector**

This detector quantifies the combined effect of two risk factors $C$ and $D$ (e.g., by overlaying geographical layers $C$ and $D$ in GIS to form a new layer $E$). The attribute of layer $E$ is defined as the combination of the attributes of layers $C$ and $D$. From the $PD$ values for the layers $C$, $D$, and $E$, the interaction detector can determine whether two factors acting together have a stronger or weaker effect on PM$_{2.5}$ concentrations residues than when acting separately.

(3) **Ecological detector**

This detector uses the $F$-value test to compare $C$ and $D$ and explore whether $C$ is more significant than $D$ in controlling the spatial pattern of the PM$_{2.5}$ concentrations residues by comparing the $PD$ values of $C$ and $D$. If $C$ is more likely than $D$ to cause PM$_{2.5}$ concentrations residues over space, one would expect the dispersion variance of $C$ ($\sigma^2_{C,x}$) to be larger than that of $D$ ($\sigma^2_{D,x}$). The relevant test is calculated using Equation (19):

$$F = \frac{n_{c,p}(n_{c,p}-1)\sigma^2_{C,x}}{n_{D,p}(n_{D,p}-1)\sigma^2_{D,x}}$$  \hspace{1cm} (19)

where $n_{c,p}$ and $n_{D,p}$ denote the number of sample units, $p$ within the coverage of layers $C$ and $D$, respectively. This statistic is asymptotically distributed as $F(n_{c,p} - 1, n_{D,p} - 1)$, with $df = (n_{c,p}, n_{D,p})$. The null hypothesis is $H_0: \sigma^2_{C,x} = \sigma^2_{D,x}$. If $H_0$ is rejected conditionally at a significance
level of $\alpha$ (usually 5%), the effect of risk factor $C$ on PM$_{2.5}$ concentrations residues is significantly different from that of $D$.

References


