2014年冬季珠三角区域典型城市PM$_{2.5}$污染时空关联特征

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摘要：为探讨冬季珠江三角洲（简称珠三角）区域污染物的空间传输延迟性及其与气象、地理的关联性，利用2014年12月1日—2015年1月9日天气图、珠三角区域4个典型城市——韶关、广州、深圳、香港的地面气象数据及$\rho$(PM$_{2.5}$)小时均值、日均值的时间序列相关性分析等方法，分析了2014年冬季各城市大气$\rho$(PM$_{2.5}$)变化相关特征以及受天气过程的影响。结果表明：在研究时段内，受11次冷空气南下和3次西南暖湿气流控制的典型天气过程影响，4个典型城市$\rho$(PM$_{2.5}$)小时均值、日均值的变化趋势具有一致性，并且4个城市间的$\rho$(PM$_{2.5}$)相关性均呈现广州>深圳>香港的规律。在冷空气南下的典型天气过程中，4个城市$\rho$(PM$_{2.5}$)小时均值在显著相关。其中，韶关与广州的相关系数为0.84，广州与深圳的相关系数为0.80，深圳与香港的相关系数为0.92；4个城市间的$\rho$(PM$_{2.5}$)变化存在一定的滞后现象，其中，广州较韶关延迟1～4 h，深圳较广州延迟3 h，香港较深圳延迟1 h；而在西南暖湿气流控制的典型天气过程中，4个城市间的$\rho$(PM$_{2.5}$)变化的关联特征不明显。研究显示，冬季珠三角区域污染物在典型冷空气南下过程中存在较明显的空间传输延迟特征，并且各典型城市间浓度变化相关性较显著。

关键词：珠三角；冬季；PM$_{2.5}$；时空关联

Investigation of Spatial and Temporal Association of PM$_{2.5}$ Pollution during Winter 2014 in Typical Cities of Pearl River Delta

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Abstract: This study explored the time-lag in regional transport of pollutants over the Pearl River Delta (PRD), and the relationship between the time lags and the surface synoptic processes as well as geographic positions, in order to provide reference for the regional warming forecast and pollution prevention over PRD. Based on weather charts, surface synoptic data and PM$_{2.5}$ concentration data from four cities (Shaoguan, Guangzhou, Shenzhen and Hong Kong) from 1st December 2014 to 9th January 2015, the trend of PM$_{2.5}$ concentration over the aforementioned four cities was analyzed, and the relationship between the surface synoptic processes and PM$_{2.5}$ concentration levels were identified by time series and correlation analysis. The results showed that, due to the influence of synoptic process, including cold airflow moving southward (11 episodes) and warm-wet southwest airflow control (3 episodes), the overall trend of PM$_{2.5}$ concentrations for the four cities was consistent in both hourly and daily concentrations.
Moreover, the correlation coefficients decreased in the following order: Shenzhen & Hong Kong > Guangzhou & Shenzhen > Shaoguan & Guangzhou. During the process of cold airflow moving southward, there was noticeable correlation between PM$_{2.5}$ concentrations of the four cities, and the correlation coefficients of the hourly correlation were: 0.84 between Shaoguan and Guangzhou, 0.80 between Guangzhou and Shenzhen, and 0.92 between Shenzhen and Hong Kong. There were time lags for the concentrations of those cities: Guangzhou four hours later than Shaoguan, Shenzhen three hours later than Guangzhou, and Hong Kong one hour later than Shenzhen. However, the correlation between the four cities was not significant during the process of warm-wet southwest airflow control process. Overall, the process of cold airflow moving southward considerably affected the relevance of PM$_{2.5}$ pollution during winter in the Pearl River Delta. Therefore, there was an obvious time-lag character of the regional transport of pollutants over the PRD during typical process of cold airflow moving southward, and the relationship between the air pollutant concentration trends of the typical PRD cities was remarkable.

Keywords: Pearl River Delta; winter; PM$_{2.5}$; spatial and temporal association

The Pearl River Delta (also known as Pearl River Delta) city groups are the most prosperous and congested city groups in China, which contribute a lot to the economic development of China. Guangzhou, the capital of Guangdong Province, is an important industrial city and has a large population. The city is surrounded by the Pearl River Delta, which is a major economic region in China. The city is located at the mouth of the Pearl River, which flows into the South China Sea. Guangzhou has a humid subtropical climate, with hot and humid summers and mild winters. The city is known for its delicious Cantonese cuisine, which is famous throughout China.

The city is also a major center for business and trade, with numerous international companies operating in the region. The city is home to several universities and research institutes, which contribute to its status as a major intellectual center. Guangzhou is also an important cultural and artistic center, with a rich history and a vibrant arts scene.

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2 结果与讨论

为了探究冬季珠三角区域 PM$_{2.5}$污染时空关联特征，该研究首先对研究时段内韶关、广州、深圳、香港4个典型城市的PM$_{2.5}$污染时空关联特征进行总体识别，再进一步对研究时间段内的PM$_{2.5}$典型污染过程与天气过程的关联特征进行分析。

2.1 PM$_{2.5}$污染时空关联特征

2.1.1 $\rho$(PM$_{2.5}$)小时及日际变化特征

图2、3分别为研究时段内韶关、广州、深圳、香港的$\rho$(PM$_{2.5}$)小时均值、日均值变化。由图2可见，2014年12月1-4日，上述4个城市的$\rho$(PM$_{2.5}$)呈先升后降的变化过程；12月5-10日4个城市$\rho$(PM$_{2.5}$)均持续上升，12月11日韶关出现下降，而其余3个城市的$\rho$(PM$_{2.5}$)则达到峰值；12月12日韶关$\rho$(PM$_{2.5}$)再次上升，直至15日再次达到峰值。而其余城市的$\rho$(PM$_{2.5}$)则在12日降到谷值后，在13-15日上升并达到峰值，其中，香港15日$\rho$(PM$_{2.5}$)略有降低；12月16-17日，除了香港$\rho$(PM$_{2.5}$)略有下降，其余城市均有下降；12月18-23日，4个城市的$\rho$(PM$_{2.5}$)一致先升后降再升；12月24-28日，广州$\rho$(PM$_{2.5}$)为先上升后下降，而其余城市则一致有不同程度的下降；2014年12月29日-2015年1月3日，韶关的$\rho$(PM$_{2.5}$)处于较低水平，略微上升，香港与深圳的$\rho$(PM$_{2.5}$)在波动中微升后下降，广州则波动较大，呈先升后降再升的过程；1月4-5日韶关的$\rho$(PM$_{2.5}$)大幅上升至观察时段内的最高值，随后至7日持续下降，而广州则先下降再上升再下降，香港与深圳为略微下降后略微上升；8日4个城市的$\rho$(PM$_{2.5}$)一致。上述4个城市的$\rho$(PM$_{2.5}$)在日均值序列上变化也保有一定的一致性。
从图3可见，上述4个城市的\( \rho(\text{PM}_{2.5}) \)小时均值亦呈现一致性，尤以2014年12月1日-4日的变化最为明显，这说明研究区域受到了较大范围的天气过程的影响。此外，注意到2015年1月4-7日期间，上述4个城市的\( \rho(\text{PM}_{2.5}) \)变化存在较为明显的差异，并且韶关的\( \rho(\text{PM}_{2.5}) \)小时均值长时间（近48 h）保持在75 mg/m\(^3\)以上，在研究时段（约1个月）属较高水平。基于天气图及气象参数信息（包括风速、风向、温度、相对湿度、大气压等资料），识别出研究时间段内有14次天气过程（见图3中英文序号“1"1", 2", ..., 14"8"），可大致分为冷空气南下、西南暖湿气流控制两大典型类。其中，冷空气南下出现11次（天气过程编号为1-5"8", 7"4", 9"4", 12"8", 14"8"），西南暖湿气流控制过程共3次（天气过程编号为6"8", 8"4", 13"8"）。而2014年12月1-4日、2015年1月4-7日两个过程恰分属上述两种类别。

\[ \text{小时均值为标准值。其他3个城市为目标城市的相关系数图，时间差} < 0 \text{ h 表示目标城市的} \rho(\text{PM}_{2.5}) \text{变化先于基准城市，时间差} > 0 \text{ h 则表示目标城市的} \rho(\text{PM}_{2.5}) \text{变化迟于基准城市。由图5可见，城市间} \rho(\text{PM}_{2.5}) \text{变化存在时间差，其中，广州较韶关延迟} 2 \text{ h，深圳较广州延迟} 2 \text{ h，香港较深圳延迟} 1 \text{ h，表明} \rho(\text{PM}_{2.5}) \text{变化相关性与地理位置相关，而地理位置对相关性的影响主要由气象因素的影响所表征。} ]^{[18]} \]

由于香港与深圳同处海边，距离较较近，受海陆风影响的规律较一致，因此，两地\( \rho(\text{PM}_{2.5}) \)小时均值的相关性最高。

2.1.2 各城市污染的相互关联特征

对4个城市间的\( \rho(\text{PM}_{2.5}) \)时间序列相关性进行分析，结果如图4所示。4个城市的\( \rho(\text{PM}_{2.5}) \)相关性中，韶关与广州、广州与深圳、深圳与香港的较大，并且呈现出从区与香港 > 广州与深圳 > 韶关与广州的规律，说明韶关、广州、深圳、香港的\( \rho(\text{PM}_{2.5}) \)变化相关性与地形分布有关，距离较近的城市相关性较大。由表1可见，上述4个城市间\( \rho(\text{PM}_{2.5}) \)小时均值的相关系数显著高于市均值，因此利用小时均值序列进行时间延迟相关性分析结果如图5所示，图5(a) - (d)分别为以韶关、广州、深圳、香港\( \rho(\text{PM}_{2.5}) \)小时均值为基准，其他3个城市为目标城市的相关系数图，时间差 < 0 h 表示基准城市的\( \rho(\text{PM}_{2.5}) \)变化先于目标城市，时间差 > 0 h 则表示目标城市的\( \rho(\text{PM}_{2.5}) \)变化迟到基准城市。由图5可见，城市间\( \rho(\text{PM}_{2.5}) \)变化存在时间差，其中，广州较韶关延迟2 h，深圳较广州延迟2 h，香港较深圳延迟1 h，表明\( \rho(\text{PM}_{2.5}) \)变化相关性与地理位置相关，而地理位置对相关性的影响主要由气象因素的影响所表征。
表1 研究区域各城市$\rho(\text{PM}_{2.5})$日均值/小时均值显著水平
Table 1 The significance level of $\text{PM}_{2.5}$ daily/hourly concentration among the cities within the research area

<table>
<thead>
<tr>
<th>城市组</th>
<th>$\rho(\text{PM}_{2.5})$日均值</th>
<th>$\rho(\text{PM}_{2.5})$小时均值</th>
<th>$P$值</th>
<th>显著水平</th>
</tr>
</thead>
<tbody>
<tr>
<td>韶关与广州</td>
<td>$1.15 \times 10^{-3}$</td>
<td>不显著</td>
<td>6.86 $\times 10^{-18}$</td>
<td>显著</td>
</tr>
<tr>
<td>韶关与深圳</td>
<td>$3.37 \times 10^{-1}$</td>
<td>不显著</td>
<td>2.41 $\times 10^{-7}$</td>
<td>显著</td>
</tr>
<tr>
<td>韶关与香港</td>
<td>$3.37 \times 10^{-1}$</td>
<td>不显著</td>
<td>7.35 $\times 10^{-9}$</td>
<td>显著</td>
</tr>
<tr>
<td>广州与深圳</td>
<td>$2.56 \times 10^{-4}$</td>
<td>显著</td>
<td>2.98 $\times 10^{-56}$</td>
<td>显著</td>
</tr>
<tr>
<td>广州与香港</td>
<td>$7.74 \times 10^{-2}$</td>
<td>显著</td>
<td>7.35 $\times 10^{-9}$</td>
<td>显著</td>
</tr>
<tr>
<td>深圳与香港</td>
<td>$1.32 \times 10^{-12}$</td>
<td>显著</td>
<td>1.88 $\times 10^{-17}$</td>
<td>显著</td>
</tr>
</tbody>
</table>

注：$\rho(\text{PM}_{2.5})$日均值样本人数为38个；$\rho(\text{PM}_{2.5})$小时均值样本人数为912个；$P<0.01$ 代表在0.01 水平上显著相关。

2.2 PM$_{2.5}$典型污染过程与天气过程的关联特征

由于天气过程从发展到结束过程变化较快，为反映$\rho(\text{PM}_{2.5})$变化与天气过程的关系，主要采用小时序列进行分析。

2.2.1 典型过程—2014年12月1—4日（冷空气南下过程）

由天气图及地面气象资料分析得知，2014年12月1—4日属于冷空气南下过程。4个城市$\rho(\text{PM}_{2.5})$小时均值变化见图6，4个城市地面气象参数变化见图7。

12月1日08:00左右，风速均呈现不同程度的减弱，风向以北风、偏北风为主，气压在波动中略

微上升，相对湿度在波动中略微下降，说明有冷空气影响但强度较弱，此时4个城市$\rho(\text{PM}_{2.5})$开始上升，到2日00:00左右，韶关迅速达到峰值（67 $\mu g/m^3$，增长近5倍），4 h 后是广州（69 $\mu g/m^3$，增长近9倍），而深圳及香港的$\rho(\text{PM}_{2.5})$则持续缓慢上升，直到2日21:00，22:00 两地才分别达到峰值（69、60 $\mu g/m^3$，增长近 3 倍）。

在12月2－3日白天，4个城市风速均较小（低于2 m/s），风向上除了韶关转为偏南风之外，其余城市以北风、东北风为主。在波动中下降，相对湿度在波动中上升并维持较长时间，说明该时段内冷空气影响减弱至结束，此时$\rho(\text{PM}_{2.5})$保持在较高范围内。

12月3日12:00左右，韶关风速开始大幅增加，风向转为偏南风，气温上升，相对湿度大幅下降，温度亦有所下降，$\rho(\text{PM}_{2.5})$大幅下降，随后广州、深圳、香港的气压参数相继出现相似的变化，说明新一轮冷空气南下影响研究区域。与此同时，韶关市$\rho(\text{PM}_{2.5})$首先快速下降，直至4日03:00 达到最低值（9 $\mu g/m^3$），而广州则从12月3日19:00 起迅速下降，直到4日05:00 达到最低值（1 $\mu g/m^3$），随后深
深圳、香港也分别于3日22:00, 4日00:00起开始大幅下降，直到4日06:00左右两地才分别达到最低值（10.2 μg/m³）。12月4日4个城市为风速均有不同程度的减弱，风向仍以北风为主，气压及相对湿度均较稳定，温度较前3d有所降低，4个城市ρ（PM₁₅）处在较低范围内。

12月1—2日ρ（PM₂.₅）上升期，4个城市ρ（PM₂.₅）小时均值每小时平均上升1～4 μg/m³，该时段内升幅分别为306%～886%。在12月3—4日ρ（PM₂.₅）下降期，4个城市ρ（PM₂.₅）小时均值每小时平均下降4～7 μg/m³，该时段内降幅为86%～99%。4个城市ρ（PM₂.₅）小时均值出现较明显的变相对湿度特征，各城市的ρ（PM₂.₅）小时均值相关系数均较大，其中韶关与广州为0.84，广州与深圳为0.80，深圳与香港为0.92。此外，由图8可见，各城市间ρ（PM₂.₅）存在一定的变化滞后现象。其中，广州较韶关延迟4 h，深圳较广州延迟3 h，香港较深圳延迟1 h。

综上，2014年12月1—3日是一次冷空气南下对珠三角区域造成影响的过程，而12月3—4日是另一次冷空气南下的过程。12月1日ρ（PM₂.₅）在较高水平，是因为该股冷空气较弱（4个城市24 h变压平均值为0.96 hPa），平流清除能力有限，在其减弱后，ρ（PM₂.₅）迅速反弹。该研究结果与文献[21]的研究结论相一致，而12月3—4日ρ（PM₂.₅）大幅下降，是因为此次的冷空气较强（4个城市24 h变压平均值为3.77 hPa），平流清除能力较强，4个城市ρ（PM₂.₅）的变化次序是由北到南逐一发生的，这与冷空气南下的影响路径较一致，此外，还发现当相对湿度较高、气压较低、风速较小时，ρ（PM₂.₅）较高；当相对湿度较低、气压较高、风速较大时，ρ（PM₂.₅）较低；风向有较大改变时会对ρ（PM₂.₅）有影响。在河北省，长三角等区域相关研究的结论[27—28]相似。

由图6可见，两次冷空气南下均造成韶关、广州、深圳、香港存在一致的ρ（PM₂.₅）小时均值先升后降的变化趋势。究其原因：①可能为受风场（4个城市小时风速最大值均超过4 m/s）造成的外源污染输送的影响，冷空气将上风向的污染物携带至下风向，造成下风向的污染物浓度升高[21]，珠三角区域城市PM₂.₅，外源输送的贡献在35%～70%内（见表2）；②可能受气团压缩造成的污染的累积，一方面因为冷空气南下至韶关需翻越南岭山脉，存在一定的下沉气流[21—22]，另一方面因为冷气团与该区域原有气团存在挤压作用[26—30]。

### 图6 研究区域各城市2014年12月1—4日ρ（PM₂.₅）小时均值变化

![Fig. 6 The trend of PM₂.₅, hourly concentration of the cities within research area from 1st to 4th December 2014](image)

2.2.2 典型过程二：2015年1月4—7日（西南暖湿气流控制）

由天气图及地面气象资料分析可知，2015年1月4—7日主要为西南暖湿气流控制过程，4个城市ρ（PM₂.₅）小时均值变化见图9，4个城市地面气象参数变化见图10。

1月4日12:00左右开始，韶关以偏南风为主，风速有所增大，相对湿度在波动中升高，气压有所降低，韶关ρ（PM₂.₅）小时均值快速上升，20:00左右达到峰值（141 μg/m³），9 h增长了96 μg/m³（约213%），此后至1月6日12:00左右，一直基本维持在100 μg/m³以上。而广州、深圳、香港的风速较小，以东风向为主，并且相对湿度、气压均在较稳定的范围内波动，广州ρ（PM₂.₅）小时均值在50～80 μg/m³范围内波动，深圳、香港的ρ（PM₂.₅）小时均值变化较为一致，维持在20～40 μg/m³范围内。通过天气型分
Fig. 7 Variation of PM$_{2.5}$ and meteorological parameters from 1$^{st}$ to 4$^{th}$ December 2014

Fig. 8 The correlation coefficients of PM$_{2.5}$ hourly concentration among the cities within the research area from 1$^{st}$ to 4$^{th}$ December 2014
表2 外地源对不同地区PM$_{2.5}$贡献率对比
Table2 Contributions of external transmission to different areas

<table>
<thead>
<tr>
<th>模拟时间</th>
<th>模型</th>
<th>地区</th>
<th>贡献率%</th>
<th>数据来源</th>
</tr>
</thead>
<tbody>
<tr>
<td>2012年12月</td>
<td>CAMx</td>
<td>广州</td>
<td>69</td>
<td>文献[31]</td>
</tr>
<tr>
<td>2012年12月—2013年3月</td>
<td>CMAQ结合HYSPLIT</td>
<td>广州番禺</td>
<td>69</td>
<td>文献[32]</td>
</tr>
<tr>
<td>2013年1月21—24日</td>
<td>NAQPS</td>
<td>珠三角区域各城市</td>
<td>35.1~70</td>
<td>文献[33]</td>
</tr>
<tr>
<td>2006年10月27日</td>
<td>CMAQ结合HYSPLIT</td>
<td>广州万顷沙</td>
<td>37.6</td>
<td>文献[34]</td>
</tr>
</tbody>
</table>

注：CAMx为区域空气质量模式；CMAQ为综合空气质量模式；HYSPLIT为气流后向轨迹模式；NAQPS为质谱网格空气质量模式系统。

析可知，2015年1月4—6日为西南暖湿气流控制过程，低压槽从广西方向影响珠三角北部区域（包括韶关），暖湿气流带来的高湿度（相对湿度接近100%）引起大气污染物的物理化学反应利于PM$_{2.5}$生成（9），此外受南岭山脉的阻挡作用，易形成污染的累积，对韶关的影响较为明显，因此韶关ρ（PM$_{2.5}$）较高；而广州、深圳、香港位于珠三角的南部，属于此西南暖湿气流控制区域的边缘，相对湿度较韶关低，大气污染物的液相物理-化学反应较韶关弱，且南岭山脉较远，因此上述3个城市的ρ（PM$_{2.5}$）较韶关低。此外，上述3个城市的风速均较小（小于2 m/s），污染受风场的输送作用不明显，主要受局地污染的影响，因此ρ（PM$_{2.5}$）在一定范围内波动。

1月6日12:00—7日10:00左右，韶关、广州、深圳、香港4个城市的ρ（PM$_{2.5}$）依次有所下降。由天气型分析可知，该时段主要为冷空气南下及冷空气持续补充过程对珠三角区域的逐步影响。

通过对典型过程一和典型过程二的分析可知，在冷空气南下过程中，受风场输送影响，污染区域影响较突出，韶关、广州、深圳、香港4个城市的ρ（PM$_{2.5}$）时间序列变化趋势一致性特征，小时均值的相关系数达到0.8以上，并且各城市间ρ（PM$_{2.5}$）变化存在一定的时间滞后性；而在暖湿气流控制过程中，上述关联特征不明显，以污染的局地影响为主。

由全研究时段，典型过程一、典型过程二的分析可知，在ρ（PM$_{2.5}$）变化一致性及ρ（PM$_{2.5}$）变化滞后性方面，虽然典型过程二上述特征并不明显，但是全研究时段与典型过程一均呈现出较为一致的上述特征。可能是因为研究时段内与典型过程一相似的天气
Fig. 10 Variation of PM$_{2.5}$ levels and meteorological parameters from 4$^{th}$ to 7$^{th}$ January 2015

Fig. 11 The correlation coefficients of PM$_{2.5}$ hourly concentration among the cities within the research area from 4$^{th}$ to 7$^{th}$ January 2015
过程较多(11次冷空气南下过程),对研究时间段整体特征有一定程度的影响。韶关、广州、深圳和香港等珠三角城市的\(\rho(\text{PM}_{2.5})\)在冬季大部分情况下所表现出的浓度变化一致性及滞后性特征,有利于珠三角区域冬季条件下区域联防联控措施的制订,更有效地预防和控制区域\(\text{PM}_{2.5}\)污染。

3 结论

a) 研究时段内,经历的14次天气过程可大致分为冷空气南下(11次)、西南暖湿气流控制(3次)两大典型类型。总体而言,韶关、广州、深圳、香港的\(\rho(\text{PM}_{2.5})\)小时均值、日均值的时间序列存在变化趋势一致性特征。4个城市间\(\rho(\text{PM}_{2.5})\)相关性呈现深圳与香港 > 广州与深圳 > 韶关与广州的规律,这表明\(\rho(\text{PM}_{2.5})\)小相关性与地理位置有关。

b) 在冷空气南下的典型过程中,4个城市\(\rho(\text{PM}_{2.5})\)小时均值变化的关联特征较明显,其中韶关与广州的相关系数为0.84,广州与深圳的相关系数为0.80,深圳与香港的相关系数为0.92;并且各城市间存在一定的\(\rho(\text{PM}_{2.5})\)变化滞后现象,其中,广州较韶关延迟4h,深圳较广州延迟3h,香港较深圳延迟1h。这与冷空气南下的影响路径(由北到南)相关。

c) 对于西南暖湿气流控制的典型过程,4个城市\(\rho(\text{PM}_{2.5})\)变化的关联特征不明显,相关系数最高仅为0.52。

d) 总体上,整个研究时段表现出的\(\rho(\text{PM}_{2.5})\)小时变化一致性特征与冷空气南下典型过程所展现的特征相似,可能是因整个研究时段内冷空气南下过程发生次数较多所致,也在一定程度上也说明了冷空气南下过程对冬季珠三角区域\(\text{PM}_{2.5}\)污染关联特征的影响较大。

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