To Dr Sally Pusede:

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R: Thanks so much for providing us some many valuable comments on our manuscript. Through two rounds of major revision, many new content has been added to the manuscript, and thus the focus of the manuscript has been changed significantly. Hence, the introduction and discussion part should be revised accordingly. As a result, your valuable comments on the structural revision on the introduction and discussion part are so important, and we do realize that your suggestion help to improve this manuscript significantly. In addition to these structural revision suggestions, we also fully revised this manuscript according to all your general and detailed comments, including figure revision, and details on the methodology and data explanation. Furthermore, more quantitative discussion has also been added to corresponding parts, according to your suggestions. Thanks again for processing and carefully reviewing this manuscript. We are more than willing to conduct additional revisions if you have further revision suggestions.

The manuscript is improved, but please address the concerns below prior to publication.

The Introduction does not properly frame the analysis. As I understand it, the manuscript describes an application of CCM to regional-scale relationships between PM2.5 and meteorological variables and discusses these results in the context of previous statistical approaches. There is no need for elementary detail on PM2.5 health effects or on trends in pollutants other than PM2.5 (e.g., ozone), especially outside of China. Focus the introductory content on material relevant to the manuscript, especially research on PM2.5-meteorology relationships in China. The CCM method should be introduced, since application of CCM is the heart of the work. The paragraph summarizing past work on PM2.5-meteorology correlations needs to be edited (begins line 104). The first two sentences are unnecessary, replace them with a sentence summarizing what has been observed so the reader is not presented with a listing of past results without context. I encourage you to add a concluding paragraph to the Introduction that describes the analysis that follows.

R: Thanks so much for your detailed suggestions on revising the introduction part. Yes, the introduction part should focus more on the previous studies concerning meteorology-$PM_{2.5}$ concentrations in China, and thus we have significantly reduced the introduction of $PM_{2.5}$ induced diseases and the meteorological influences on $PM_{2.5}$ concentrations in other countries. Furthermore, the interactions between other airborne pollutants (e.g. O_3 and PM_{10}) and meteorological factors have been completely deleted in the revised manuscript. Meanwhile, more relevant studies concerning meteorological influences on $PM_{2.5}$ concentrations in China have been added to the revised manuscript. These studies examined the correlation between $PM_{2.5}$ concentrations and different meteorological factors in specific cities. However, findings from these studies conducted at a local scale cannot reveal regional and national patterns of meteorological influences on PM2.5 concentrations in China. In addition, these studies mainly employed short-term observation data (e.g. one season or one year) and thus revealed characteristics of meteorological influences on $PM_{2.5}$ concentrations may be biased by inter-annual variations.

The correlation analysis employed in previous studies may lead to mirage correlations and can be biased significantly by influences from other variables. So it is necessary to introduce the CCM method briefly. A short explanation of CCM, especially its advantages compared with the correlation analysis, was added to the introduction. Finally, according to your comment, we added a conclusion part to the introduction part that describes the following analysis.

The added text in the revised manuscript is as follows:

Recent studies conducted in different countries proved that $PM_{2.5}$ were closely related to temperature (Pearce et al., 2011; Yadav et al., 2014; Grundstrom et al., 2015)，wind speed (Galindo et al., 2011; El-Metwally and Alfaro, 2013; Yadav et al., 2014) and precipitation (Yadav et al., 2014). Meanwhile, meteorological influences on $PM_{2.5}$ concentrations across China have also become a hot research topic. Yao (2017) revealed a generally negative correlation between evaporation and $PM_{2.5}$ concentrations in a series of cities within the North China plain. Huang et al. (2015) and Yin et al., (2016) found a negative influence of sunshine duration and a positive influence of relative humidity on $PM_{2.5}$ concentrations in Beijing. Li et al. (2015) suggested that air pressure and temperature was positively correlated with $PM_{2.5}$ concentrations in Chengdu. For Nanjing (Chen, T. et al., 2016) and Hong Kong (Fung et al., 2014), precipitation exerted a strong influence on $PM_{2.5}$ concentrations in winter, when the influence of wind speed on $PM_{2.5}$ concentrations was weak. Meanwhile, wind speed exerted a major influence on $PM_{2.5}$ concentrations in Beijing in winter. Through experiments, Guo (et al., 2016) found that the influence of precipitation on $PM_{2.5}$ concentrations in Xi'an was weaker than that in Guangzhou. Zhang et al. (2015b) quantified the correlations between meteorological factors and main airborne pollutants in three megacities, Beijing, Shanghai and Guangzhou, and pointed out that the influences of meteorological factors on the formation and concentrations of airborne pollutants varied significantly across seasons and geographical locations. Chen, Z. et al. (2017) quantified the meteorological influences on local $PM_{2.5}$ concentrations in the Beijing-Tianjin-Hebei region and revealed that wind, humidity and solar radiation were major meteorological factors that significantly influenced local $PM_{2.5}$ concentrations in winter. These studies revealed the correlations between $PM_{2.5}$ concentrations and a diversity of meteorological factors in some specific cities. However, findings from these studies conducted at a local scale cannot reveal regional and national patterns of meteorological influences on $PM_{2.5}$ concentrations in China. In addition, these studies mainly employed short-term observation data (e.g. one season or one year) and thus revealed characteristics of meteorological influences on $PM_{2.5}$ concentrations may be biased by inter-annual variations.

Due to complicated interactions between different factors, Sugihara et al. (2012) suggested that correlation analysis between two variables in a complicated ecosystem might lead to mirage correlations and the extracted correlation coefficient between two variables could be influenced significantly by other variables in the ecosystem. To better examine the coupling between two variables in a complicated system, Sugihara et al. (2012) proposed a CCM (Cross Convergent Mapping) method to qualify the bi-direction coupling between two variables without the influence from other variables. Therefore, the CCM method can effectively remove mirage correlations and extract reliable causality between two variables.

According to these challenges, this research aims to analyze and compare the influence of individual meteorological factors on $PM_{2.5}$ concentrations across China. Based on the CCM causality analysis, we quantified the influence of eight meteorological factors on $PM_{2.5}$ concentrations in 188 monitoring cities across China using the observation data from March, 2014 to February, 2017. To comprehensively understand the spatio-temporal patterns of meteorological influences on $PM_{2.5}$ concentrations across China, we a). investigated comprehensive meteorological influences on $PM_{2.5}$ concentrations for 37 regional representative cities, b) extracted the seasonal dominant meteorological factor for each monitoring city, and c) conducted a comparative statistics of the influence of different meteorological influences on $PM_{2.5}$ concentrations.

I am not familiar with meteorological measurement evaporation. Please clarify. The footnote is not helpful and is not encouraged in ACP.

R: Thanks so much for this comment. The evaporation measurement has been added to the revised manuscript and the use of footnote has been removed.

The added explanation of evaporation is as follows:

Evaporation indicates the amount of evaporation-induced water loss during a certain period and is usually calculated using the depth of evaporated water in a container. For this research, small (large) evaporation indicates the amount of evaporated water measured using a container with a diameter of 10cm (30cm) during 24 hours (unit: mm). Generally, the measured values using the two types of equipment are of slight differences.

The meteorological variables should not be italicized.

R: Corrected

Line 157: Write out the word “minimum.”

R: Corrected

Variable abbreviations like meanTEM and maxPRS reduce readability, rather than improve it. I recommend they are all removed.

R： Thanks so much for this comment. We have removed all these inappropriate abbreviations.

Page 6: Footnotes should be avoided. Place the information in the main text.

R: All these footnotes have been removed from the manuscript and placed in the main text.

Line 175: Remove etc., instead begin the list with "e.g."

R：All etc. in the manuscript have been replaced with "e.g.".

Line 234: Say where.

R: In the revised manuscript, we have specified these locations with seasonal variations of $PM_{2.5}$ concentrations.

Seasonal variations of $PM_{2.5}$ concentrations have been revealed in Beijing (Chen et al., 2015; Chen, Y. et al., 2016; Chen, Z. et al., 2016), Nanjing (Shen et al., 2014), Shandong Province (Yang and Christakos, 2015) and the Beijing-Tianjin-Hebei region (Wang et al. 2015; Chen, Z. et al., 2017). In addition to these local and regional studies, Cao et al. (2012) further compared seasonal variations of $PM_{2.5}$ concentrations in seven southern cities (Chongqing, Guangzhou, Hong Kong, Hangzhou, Shanghai, Wuhan, and Xiamen) and seven northern cities (Beijing, Changchun, Jinchang, Qingdao, Tianjin, Xi'an, and Yulin) across China.

Avoid use of the word "prove."

R: All use of the word "prove" has been removed.

Remove all uses of the word "haze." Be specific, if you mean PM2.5, say that.

R: All the use of "haze" has been removed in the revised manuscript.

The first sentence of many paragraphs in the paper is superfluous and should be deleted to improve readability.

R: Thanks again for this comment. We again reviewed this manuscript and deleted the first sentence of many paragraphs.

Fig. 1: Use full titles and full axis labels so the figure is more easily read.

R：This figure has been reproduced according to your comments.

Fig. 1: Explain why these four panels were selected.

R: We selected the winter 2014 in Beijing as an instance to demonstrate how the CCM figure explain the bi-directional coupling between meteorological factors and $PM_{2.5}$ concentrations. For winter, 2014, Beijing was one of heavily polluted cities with extremely high $PM_{2.5}$ concentrations and the calculated p value of meteorological factors on $PM_{2.5}$ concentrations was very large. So the coupling between $PM_{2.5}$ concentrations and meteorological factors in winter, 2014 is an ideal example to demonstrate how CCM works. To better present the effects of CCM method, we specifically selected four meteorological factors, which had the strongest influences on local $PM_{2.5}$ concentrations. Meanwhile, $PM_{2.5}$ concentrations also had notable feedback effects on these meteorological factors. By selecting these four major meteorological factors, the output CCM is more likely to provide readers a general comparison of the magnitude of simultaneous influences of meteorological factors on the local $PM_{2.5}$ concentration and its feedback effects. If other factors that exerted weak influences on $PM_{2.5}$ concentrations were selected, small p values would make the curves from exemplary CCMs difficult to understand and compare.

In the revised manuscript, we have added the following explanation:

As a heavily polluted city, we presented the interactions between $PM_{2.5}$ concentrations and meteorological factors in Beijing in winter, when the local $PM_{2.5}$ concentration was the highest, as an example. Four major meteorological factors, wind, humidity, radiation and temperature, which exerted much stronger influences on $PM_{2.5}$ concentrations than other factors, were employed. Due to the strong bidirectional coupling between $PM_{2.5}$ concentrations and these meteorological factors, Figure 1 not only demonstrates how CCM output could be interpreted, but also provides readers with a general comparison of the magnitude of simultaneous influences of different meteorological factors on the local $PM_{2.5}$ concentration and its feedback effects.

Line 300: This entire paragraph can be deleted.

R: We have deleted this paragraph.

Figs 2 will not reproduce well. It is difficult to distinguish the gray scale. The gray scale limits should be rounded to integers. I recommend the wind roses be made larger and the legends labeled with words rather than abbreviations.

R： Thanks so much for this comment. We have rounded the gray scale to integers and reproduced the legends with words as you suggested. We also tried to make these wind roses a bit larger, yet we cannot make these wind roses much larger. The reason is that we used a unified wind rose legend scale for all seasons to give readers a comparable presentation of how the magnitude of meteorological influences varied across different seasons and regions. Since the p value of different factors ranged from around 0.1 to 0.8 in different seasons, the size of rose pedals also varied significantly. I understand the size of wind roses in some regions in summer (or other seasons) is a bit small. However, if we further extend the legend scale of the wind roses, although those small wind roses can be presented better, there will be severe overlapping effects for those large wind roses for those representative cities in the North China plain in winter. Since a clear presentation of these large wind roses in heavily polluted cities is of great importance, some very small wind roses caused by extreme small p values cannot be further made bigger.

Figs 2 and 3: Remove the map inset and the yellow dashed boundary. These do not contribute the display of the data.

R: Thanks so much for this suggestion. We have removed the inset map and the yellow dashed boundary and this revision indeed improves the display of the data.

Line 323: This paragraph is too vague. You are reporting on your quantitative analysis here. This text is too general and could be known without your CCM results.

R: Thanks so much for this comment on this paragraph and other parts. We do realized that without quantitative p value presented here, the simple qualitative explanation is too vague. So in the revised manuscript, we included more details on the quantitative explanation of these patterns.

The following text has been added to the revised manuscript.

Take several mega cities in different regions for instance. During 2014-2016, the three major meteorological influencing factors for $PM_{2.5}$ concentrations in Beijing, a mega city in the North China plain, were as follows: humidity (0.48)，wind (0.37) and evaporation (0.31) for spring, humidity (0.39)，temperature (0.34) and SSD (0.25) for summer, humidity (0.56)，evaporation (0.51) and wind (0.41) for autumn, and humidity (0.76), wind (0.57) and evaporation (0.52) for winter. The three major meteorological influencing factors for $PM_{2.5}$ concentrations in Shanghai, a mega city in the Yangtze River Basin, were as follows: temperature (0.264), air pressure (0.260) and wind (0.25) for spring, temperature (0.40), wind (0.38) and humidity (0.27) for summer, temperature (0.39), wind (0.28) and humidity (0.17) for autumn, and precipitation (0.36), wind direction (0.25) and humidity (0.19) for winter. The three major meteorological influencing factors for $PM_{2.5}$ concentrations in Wuhan, a major city in Central China region, were as follows: precipitation (0.18), wind (0.16) and temperature (0.09) for spring, humidity (0.47), temperature (0.41) and wind (0.34) for summer, wind (0.44), precipitation (0.31) and temperature (0.26) for autumn, and precipitation (0.33), temperature (0.19) and wind (0.15) for winter. The three major meteorological influencing factors for $PM_{2.5}$ concentrations in Guangzhou, a major city in Southern China region, were as follows: wind (0.31), precipitation (0.24) and air pressure (0.23) for spring, air pressure (0.51), temperature (0.41) and wind (0.37) for summer, temperature (0.47), wind (0.36) and precipitation (0.29) for autumn, and temperature (0.52), wind (0.48) and air pressure (0.33). Notable seasonal variations of meteorological influences on $PM_{2.5}$ concentrations were found in these mega cities across China.

Line 331: Same comment. This paragraph is too vague. You are reporting on your quantitative analysis here. This text is too general and could be known without your CCM results.

R: Thanks so much for this comment. More detailed quantitative analysis result has been included in this part.

The following text has been added to the revised manuscript.

Take four major cities, Beijing, Tianjin, Taiyuan and Shijiangzhuang, in the North China Plain for example. For winter, SSD, evaporation, humidity and wind were the major meteorological factors for $PM_{2.5}$ concentrations in the four cities and the ρ value of these four factors was 0.50, 0.52, 0.76 and 0.57 for Beijing, 0.41, 0.44, 0.56

and 0.50 for Tianjin, 0.44, 0.36, 0.61 and 0.41 for Taiyuan, and 0.62, 0.58, 0.56 and 0.60 for Shijiazhuang respectively, presenting a similar regional pattern. Meanwhile, meteorological influences on $PM_{2.5}$ concentrations in cities within the Yangtze River Basin, especially the dominant factors, were also of some regional similarities. Take four major cities in the Yangtze River Basin, Shanghai, Nanjing, Hangzhou and Nanchang for example. For summer, precipitation, humidity, temperature and wind were the major meteorological factors for $PM_{2.5}$ concentrations in these four cities and the ρ value of these factors was 0.21, 0.27, 0.40 and 0.38 for Shanghai, 0.29, 0.41, 0.34 and 0.33 for Nanjing, 0.28, 0.27, 0.23 and 0.27 for Hangzhou, and 0.24, 0.33, 0.21 and 0.29 for Nanchang. Despite of some differences in the ρ values, similar dominant meteorological factors and the similar magnitude of meteorological influences demonstrated regional similarities of meteorological influences on $PM_{2.5}$ concentrations in the Yangtze River Basin.

Line 340: Same comment. This paragraph is too vague. You are reporting on your quantitative analysis here. This text is too general and could be known without your CCM results.

R: Thanks so much for this comment. More detailed quantitative analysis result has been included in this part.

The following text has been added to the revised manuscript.

Take four major cities in the North China region for instance. For Beijing, the major influencing meteorological factors in summer were temperature (0.34), humidity (0.39) and SSD (0.25) whilst the major influencing meteorological factors in winter were humidity (0.76), wind (0.57), evaporation (0.52) and SSD (0.5). For Tianjin, the major influencing meteorological factors in summer were precipitation (0.34), temperature (0.22) and air pressure (0.25) whilst the major influencing meteorological factors in winter were humidity (0.76), wind (0.57), evaporation (0.52) and SSD (0.50). For Shijiazhuang, the major influencing meteorological factors in summer were SSD (0.4), humidity (0.26) and evaporation (0.26) whilst the major influencing meteorological factors in winter were SSD (0.62), wind (0.60), evaporation (0.58) and humidity (0.56). For Taiyuan, the major influencing meteorological factors in summer were temperature (0.32), air pressure (0.23) and precipitation (0.20) whilst the major influencing meteorological factors in winter were humidity (0.61), SSD

(0.44) and wind (0.41).

Line 352: This entire paragraph can be deleted.

R：This paragraph has been fully removed in the revised manuscript.

Line 366: Likewise, points a–c are quite general. Can you talk about these results in specific quantitative terms?

R: Thanks so much for this comment. More detailed quantitative analysis result has been included in this part.

The following text has been added to the revised manuscript to a.

Here we analyzed the ρ value of precipitation in cities within the Yangtze River Basin and cities within the Beijing-Tianjin-Hebei region respectively. For winter, precipitation was the dominant factor for $PM_{2.5}$ concentrations in Shanghai, Hangzhou and Nanchang within the Yangtze River Basin and the ρ value of precipitation was 0.36, 0.29 and 0.31 respectively. Meanwhile, the ρ value of precipitation in Beijing, Tianjin and Shijiazhuang within the Beijing-Tianjin-Hebei region was 0.08, 0.01 and 0.06 respectively.

The following text has been added to the revised manuscript to b.

The prevalence of different meteorological factors across China can also be reflected according to the number of cities where this specific factor is the dominant factor for local $PM_{2.5}$ concentrations. For winter, the number of cities with temperature, wind or humidity as the dominant factor was 56，48 and 44 respectively. Meanwhile, the number of cities with SSD or wind direction as the dominant factor was 3 and 1 respectively.

The following text has been added to the revised manuscript to c.

Take some major cities in North China region for instance. For winter, the dominant meteorological factors for Beijing, Tianjin, Taiyuan, Zhangjiakou, Handan and Jining was humidity (0.76)，humidity (0.56), humidity (0.61), wind (0.62), humidity (0.43) and humidity (0.52) respectively. Meanwhile, for summer, the dominant meteorological factors for Beijing, Tianjin, Taiyuan, Zhangjiakou, Baoding, Handan and Jining was humidity (0.39), precipitation (0.28), temperature (0.23), temperature (0.47), air pressure (0.21) and SSD (0.18).

Fig. 3: Same comments as on Fig 2. Can you make these figures more readable? Is the size of the symbol important? They appear to vary.

R: Thanks so much for this comment. We have reproduced these symbols

and attempts to make different samples appear with a similar size.

Line 458: Do you mean “across different regions?”

R: Yes, and we have revised the use of “in” to “ across”.

Lines 486–488: What is the evidence for this?

R: Sorry that we did not make this clear in the previous manuscript. The main reason why local-scale studies cannot reveal regional patterns are as follows: a. Firstly, local-scale studies mainly focuses on specific cities and thus regional similarities of meteorological influences on PM$_{2.5}$ concentrations may not be revealed. E.g. a case study in Nanjing cannot reflect the spatio-temporal patterns in the Yangtze-River Basin. b. Previous local-scale studies were conducted at different time and thus findings from these studies could not be compared. c. Due to highly complicated interactions between meteorological factors in the atmospheric environment, the correlation coefficient between PM$_{2.5}$ concentrations and different meteorological factors was not a reliable indicator to compare across different regions. In this case, based on the CCM method, this research examined meteorological influences on PM$_{2.5}$ concentrations for 188 monitoring cities across China using a unified research period of three consecutive years and a unified set of meteorological factors and better understood the regional patterns of meteorological influences on PM$_{2.5}$ concentrations across China. The revised content was detailed explained in the following responses.

Line 532: Much of the content of this paragraph appears to be irrelevant to the manuscript.

R：This paragraph has been entirely deleted in the revised manuscript.

The Discussion should be refocused. The purpose of this analysis was to apply CCM to a wide region. The Discussion should then consider the difference between the author’s CCM results and past analyses on more local areas and to compare CCM to other statistical approaches. First, it is well known that PM2.5

correlates with meteorological variables. Second, ACP is not an appropriate journal to expound upon government policies and public response unrelated to analysis perform. These are not the discussion point, instead the authors must answer:

What new do we learn about PM2.5-meteological relationships by using CCM over a large spatial region?

R: Thanks so much for this comment. We have fully revised the discussion part. The discussion on the government policies and public responses unrelated to analysis perform have been massively reduced or removed. We have added some new content concerning the comparison between this large-scale research using the CCM method. and previous local-regional scale research using the correlation analysis. The added content concerning "What new do we learn about PM2.5-meteological relationships by using CCM over a large spatial region" is as follows:

Compared with studies at a local or regional scale, this research conducted at the national scale provided a better understanding of spatial and temporal patterns of meteorological influences on $PM_{2..5}$ concentrations across China, for the following reasons. a. A national perspective. Previous studies conducted at a local scale mainly focused on a specific city (e.g. Beijing), and can hardly reveal spatio-temporal patterns of meteorological influences on $PM_{2.5}$ concentrations at a large scale (e.g. the North China plain). This research, on the other hand, quantified the influence of meteorological factors on $PM_{2.5}$ concentrations for 188 cities across China, and thus revealed some regional patterns of meteorological influences on $PM_{2.5}$ concentrations in some typical regions (e.g. North China region or Yangtze River Basin). b. A unified research period and set of meteorological factors. Previous studies employed short-term observation data (e.g. one season or one year) to examine the meteorological influences on local $PM_{2.5}$ concentrations in specific cities. Due to the discrepancy in research periods and sets of meteorological factors, the findings from different local-scale studies cannot be compared and comprehensively understood. This research employed daily $PM_{2.5}$ and meteorological data of three consecutive years and a unified set of eight meteorological factors for all 188 monitoring cities and thus meteorological influences on $PM_{2.5}$ concentrations across China can be effectively compared without significant influences from inter-annual variations. c. A robust causality analysis method. Due to complicated interactions between different meteorological factors, correlations analysis, as introduced above, may lead to large bias in quantifying the meteorological influences on $PM_{2.5}$ concentrations. Similarly, the correlation coefficient between individual meteorological factors and $PM_{2.5}$ concentrations cannot be used as a reliable indicator to compare quantitative influences of individual meteorological factors on $PM_{2.5}$ across different cities. This research employed a robust CCM method, which removes the influence of other factors, and effectively quantified the coupling between $PM_{2.5}$ concentrations and a set of meteorological factors. The ρ value of each meteorological factor on $PM_{2.5}$ concentration can be compared between different cities. Based on national statistics across China, this research concluded that the influence of temperature, humidity and wind, especially temperature, on $PM_{2.5}$ concentrations was much larger than that of other meteorological factors, which could not be revealed by previous local and regional scale studies. _

Understanding meteorological influences on $PM_{2.5}$ concentrations across China: a temporal and spatial perspective

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Abstract

With frequent ~~haze~~ air pollution episodes~~vents~~ in China, growing research emphasis has been put on quantifying meteorological influences on $PM_{2.5}$ concentrations. However, these studies mainly focus on isolated cities whilst meteorological influences on $PM_{2.5}$ concentrations at the national scale have yet been examined comprehensively. This research employs the CCM (Cross Convergent Mapping) method to understand the influence of individual meteorological factors on local $PM_{2.5}$ concentrations in 188 monitoring cities across China. Results indicate that meteorological influences on $PM_{2.5}$ concentrations are of notable seasonal and regional variations. For the heavily polluted North China region, when $PM_{2.5}$ concentrations are high, meteorological influences on $PM_{2.5}$ concentrations are strong. The dominant meteorological influence for $PM_{2.5}$ concentrations varies across locations and demonstrates regional similarities. For the most polluted winter, the dominant meteorological driver for local $PM_{2.5}$ concentrations is mainly the wind within the North China region whilst precipitation is the dominant meteorological influence for most coastal regions. At the national scale, the influence of temperature, humidity and wind on $PM_{2.5}$ concentrations is much larger than that of other meteorological factors. Amongst eight factors, temperature exerts the strongest and most stable influence on national $PM_{2.5}$ concentrations in all seasons. Due to notable temporal and spatial differences in meteorological influences on local $PM_{2.5}$ concentrations, this research suggests pertinent environmental projects for air quality improvement should be designed accordingly for specific regions.

Keywords: $PM_{2.5}$; Meteorological factors; Causality analysis; CCM

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Introduction

With rapid social and economic growth in China, both the government and residents are placing more and more emphasis on the sustainability of the ambient environment, and air quality has become one of the most concerned social and ecological issues. Recently, the frequency of air pollution episodes with high $PM_{2.5}$ concentrations and the number of cities influenced by $PM_{2.5}$ pollution have increased notably in China since 2013. Statistical records from the national air quality publishing platform (http://113.108.142.147:20035/emcpublish/) revealed that $PM_{2.5}$ induced pollution events occurred in 25 provinces and more than 100 middle-large cities whilst there were on average 30 days with hazardous $PM_{2.5}$ concentrations for each monitoring city in 2014.

High $PM_{2.5}$ concentrations not only influence people's daily life (e.g. high $PM_{2.5}$ concentrations caused ~~the cause of~~ severe traffic jam ~~during haze epsiodes~~), but also severely threaten the health of residents that suffer from polluted air quality. Recent studies ~~(Garrett and Casimiro, 2011; Qiao et al., 2014; Pasca et al., 2014; Lanzinger et al., 2015; Li et al., 2015a; etc.)~~ have ~~proven~~ suggested that airborne pollutants, $PM_{2.5}$ in particular, are closely related to cardiovascular disease-related mortality (Garrett and Casimiro, 2011, Li et al., 2015a), emergency room visits (Qiao et al., 2014), all year non-accidental mortality (Pasca et al., 2014) and cardiovascular mortality (Lanzinger et al., 2015).

~~all-cause and cause-specific mortality. Garrett and Casimiro, (2011) revealed that the relative risk for cardiovascular disease-related mortality for alder groups (>65 years) was 2.39% (95%C.I. 1.29%, 3.50%) for each 10 μ g/m^3 $PM_{2.5}$ increase. Guaita et al. (2011) Qiao et al. (2014) found an interquartile range increment in $PM_{2.5}$ concentration (36.47 μg/m^3) led to a 0.57% [95% confidence interval (CI): 0.13%, 1.01%] increase in emergency room visits. Through experiments in nine French cities, Pasca et al. (2014) observed a notable effect of $PM_{2.5}$ (+0.7%, [−0.1; 1.6]) on all year non-accidental mortality for all age groups. In five European cities, estimation results suggested that a 12.4 μg/m^3 increase in the $PM_{2.5}$ concentration can lead to 3.0% [− 2.7%; 9.1%] increase in cardiovascular mortality (Lanzinger et al., 2015). Li et al. (2015a) found that temperature played an important role in $PM_{2.5}$ induced mortality in Beijing. Under the condition of the lowest temperature range (-9.7~2.6 °C), a 10 μg/m^3 increase in PM2.5~~

~~concentration led to an increase of 1.27 % (95 % CI 0.38~2.17 %) in the relative risk (RR) of cardiovascular mortality, which was the highest for all temperature ranges.~~ Due to its strong negative influences on public health~~,~~ , scholars have been working towards a better understanding of sources (Guo et al., 2012; Zhang et al., 2013；Gu et al., 2014; Liu et al., 2014; Cao et al., 2014), characteristics (Wei et al., 2012; Zhang et al., 2013; Hu et al., 2015; Zhang, F. et al., 2015; Zhen et al., 2016; Zhang et al., 2016) and seasonal variations (Cao et al., 2012; Shen et al., 2014; Yang and Christakos, 2015; Wang et al., 2015; Chen et al., 2015; Chen, Y. et al. 2016; Chen, Z. et al., 2016) of $PM_{2.5}$ and other airborne pollutants. Meanwhile, large-scale research on the variation and distribution of $PM_{2.5}$ has been conducted using a variety of remote sensing sources and spatial data analysis methods (Ma et al., 2014; Kong et al., 2016~~.~~).

One key issue for air quality research is to find the source and influencing factors for airborne pollutants. Although quantitative contributions of different sources (e.g. coal burning and automobile exhaust) to airborne pollutants remain controversial, meteorological influences on airborne pollutants have been examined in depth by more and more scholars. Recent ~~Recently, massive studies have been conducted to extract quantitative correlations between meteorological factors and air pollutants.~~ studies conducted in different countries indicated that $PM_{2.5}$ were closely related to temperature (Pearce et al., 2011; Yadav et al., 2014; Grundstrom et al., 2015)，wind speed (Galindo et al., 2011; El-Metwally and Alfaro, 2013; Yadav et al., 2014) and precipitation (Yadav et al., 2014). Meanwhile, meteorological influences on $PM_{2.5}$ concentrations across China have also become a hot research topic. Yao (2017) revealed a generally negative correlation between evaporation and $PM_{2.5}$ concentrations in a series of cities within the North China plain. Huang et al. (2015) and Yin et al., (2016) found a negative influence of sunshine duration and a positive influence of relative humidity on $PM_{2.5}$ concentrations in Beijing. Li et al. (2015) suggested that air pressure and temperature was positively correlated with $PM_{2.5}$ concentrations in Chengdu. For Nanjing (Chen, T. et al., 2016) and Hong Kong (Fung et al., 2014), precipitation exerted a strong influence on $PM_{2.5}$ concentrations in winter, when the influence of wind speed on $PM_{2.5}$ concentrations was weak. Meanwhile, wind speed exerted a major influence on $PM_{2.5}$ concentrations in Beijing in winter. Through experiments, Guo (et al., 2016) found that the influence of precipitation on $PM_{2.5}$ concentrations in Xi'an was weaker than that in Guangzhou. ~~Blanchard et al. (2010) indicated that ozone concentrations were linearly correlated with temperature and humidity, and non-linearly correlated with other meteorological factors. Juneng et al. (2011) suggested that such meteorological factors as temperature, humidity and wind speed, dominated the fluctuation of PM_{10} over the Klang Valley during the summer monsoon. In Melbourne, Pearce et al. (2011) found that local temperature led to strongest responses of different pollutants (PM, ozone and NO_2), whilst other meteorological factors (e.g. winds, water vapor pressure, radiation, precipitation) affected one or more specific pollutants. In the city of Elche, Spain, Galindo et al. (2011) revealed that fractions of three different PM sizes (PM_1, $PM_{2.5}$ and PM_{10}) were negatively correlated with wind speed in winter, whilst coarse fractions were strongly correlated with temperature and solar radiation. At a site of the Egyptian Mediterranean coast, El Metwally and Alfaro (2013) found that the wind speed not only influenced the dilution of airborne pollutants, but also affected the composition of airborne pollutants. For a Western Indian location, Udaipur, Yadav et al. (2014) proved that precipitation exerted a stronger influence on PM_{10} than on $PM_{2.5}$. High temperature diluted the emission of surface pollutants whilst strong winds diminished the trend of air pollution in May. Grundstrom et al. (2015) suggested that low wind speeds and positive vertical temperature gradients were favorable meteorological conditions for elevated NOx and particle number concentrations (PNC).~~ Zhang et al. (2015b) quantified the correlations between meteorological factors and main airborne pollutants in three megacities, Beijing, Shanghai and Guangzhou, and pointed out that the influences of meteorological factors on the formation and concentrations of airborne pollutants varied significantly across seasons and geographical locations. Chen, Z. et al. (2017) quantified the meteorological influences on local $PM_{2.5}$ concentrations in the Beijing-Tianjin-Hebei region and revealed that wind, humidity and solar radiation were major meteorological factors that significantly influenced local $PM_{2.5}$ concentrations in winter. These studies revealed the correlations between $PM_{2.5}$ concentrations and a diversity of meteorological factors in some specific cities. However, findings from these studies conducted at a local scale cannot reveal regional and national patterns of meteorological influences on $PM_{2.5}$ concentrations in China. In addition, these studies mainly employed short-term observation data (e.g. one season or one year) and thus revealed characteristics of meteorological influences on $PM_{2.5}$ concentrations may be biased by inter-annual variations. ~~.~~

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~~Although correlations between airborne pollutants and meteorological factors have been well studied, analyzing the sensitivity of airborne pollutants to individual meteorological parameters remains challenging (Pearce et al., 2011). This is because different meteorological factors are inherently interacting and can thus influence airborne pollutants through direct and indirect mechanisms.~~ Due to the diversity of meteorological factors and complicated interactions between them, Pearce et al (2011) suggested that multiple models and methods should be comprehensively employed to quantify the influence of meteorological factors on local airborne pollutants. Due to complicated interactions between different factors, Sugihara et al. (2012) suggested that correlation analysis between two variables in a complicated ecosystem might lead to mirage correlations and the extracted correlation coefficient between two variables could be influenced significantly by other variables in the ecosystem. To better examine the coupling between two variables in a complicated system, Sugihara et al. (2012) proposed a CCM (Cross Convergent Mapping) method to qualify the bi-direction coupling between two variables without the influence from other variables. Therefore, the CCM method can effectively remove mirage correlations and extract reliable causality between two variables. Our previous research (Chen, Z., 2017) ~~proved~~ found that the CCM (Cross Convergent Mapping) method performed better in quantifying the influence of individual meteorological factors on $PM_{2.5}$ concentrations than traditional correlation analysis through comprehensive comparison. However, this study mainly focused on the meteorological influences on $PM_{2.5}$ concentrations in a specific region. As pointed out by some scholars, interactions between meteorological factors and airborne pollutants are of great variations for different regions, yet most relevant studies have been conducted at the local or regional scale. China is a large country, including many regions with completely different air pollution levels, geographical conditions and meteorological types. To better understand the variations of meteorological influences on $PM_{2.5}$ concentrations, a comparative study at the national scale is required.

~~In accordance with~~According to these challenges, this research aims to~~–~~ analyze and compare~~quantify and compare~~ the influence~~s~~ of individual meteorological factors on $PM_{2.5}$ concentrations ~~in different cities~~ across China. Based on the CCM causality analysis, we quantified the influence of eight meteorological factors on $PM_{2.5}$ concentrations in 188 monitoring cities across China using the observation data from

March, 2014 to February, 2017. To comprehensively understand the spatio-temporal patterns of meteorological influences on $PM_{2.5}$ concentrations across China, we a). investigated comprehensive meteorological influences on $PM_{2.5}$ concentrations for 37 regional representative cities, b) extracted the seasonal dominant meteorological factor for each monitoring city, and c) conducted a comparative statistics of the influence of different meteorological factors on $PM_{2.5}$ concentrations at the national scale. ~~Based on the causality analysis, dominant meteorological factors for $PM_{2.5}$ concentrations can be extracted for each city and spatio-temporal patterns of meteorological influences on $PM_{2.5}$ concentrations across China can be revealed. In addition to its theoretical significance, this research may provide useful reference for evaluating pertinent environmental projects and enhancing air quality through meteorological measures.~~

2 Materials

2.1 Data sources

2.1.1 $PM_{2.5}$ data

$PM_{2.5}$ data are acquired from the website PM25.in. This website collects official data of $PM_{2.5}$ concentrations provided by China National Environmental Monitoring Center (CNEMC) and publishes hourly air quality information for all monitoring cities. Before Jan 1st, 2015, PM25.in publishes data of 190 monitoring cities. Since Jan 1st, 2015, the number of monitoring cities has increased to 367. By calling specific API (Application Programming Interface) provided by PM25.in, we collect hourly $PM_{2.5}$ data for target cities. The daily $PM_{2.5}$ concentrations for each city is calculated using the averaged value of hourly $PM_{2.5}$ concentrations measured at all available local observation stations. For a consecutive division of different seasons and multiple-year analysis, We collected $PM_{2.5}$ data from March 1st, 2014 to February 28th, 2017 for the following analysis.

2.1.2 Meteorological data

The meteorological data for these monitoring cities are obtained from the “China Meteorological Data Sharing Service System”, part of National Science and Technology Infrastructure. The meteorological data are collected through thousands of observation stations across China. Previous studies (Zhang et al., 2015b; Pearce et al., 2011; Yadav et al., 2014) ~~proved~~ revealed that such meteorological factors as relative humidity, temperature, wind speed, wind direction, solar radiation, evaporation, precipitation, and air pressure may be related to $PM_{2.5}$ concentrations. Therefore, to comprehensively understand meteorological driving forces for $PM_{2.5}$ concentrations in China, all these potential meteorological factors were selected as candidate factors. To better quantify the role of these meteorological factors in affecting local $PM_{2.5}$ concentrations, these factors are further categorized into some sub-factors: evaporation (small evaporation and large evaporation~~, short for smallEVP and largeEVP[2]~~), temperature (daily max temperature, mean temperature, minimum temperature, and largest temperature difference for the day~~, short for maxTEM, meanTEM, minTEM and difTEM~~), precipitation (total precipitation from 8am-8pm, total precipitation from 8pm-8am and total precipitation for the day~~, short for PRE8-20, PRE20-8 and totalPRE~~), air pressure (daily max pressure, mean pressure and minimum pressure~~, short for maxPRS, meanPRS and minPRS~~), humidity (daily mean and minimum relative humidity~~, short for meanRHU and minRHU~~), radiation (sunshine duration~~[3]~~ for the day, short for SSD), wind speed (mean wind speed, max wind speed and~~,~~ extreme wind speed~~[4], short for meanWIN, maxWIN and extWIN~~), wind direction (max wind direction~~[5]~~ for the day~~, short for dir_maxWin~~). Some meteorological factors are briefly explained here. Evaporation indicates the amount of evaporation-induced water loss during a certain period and is usually calculated using the depth of evaporated water in a container. For this research, small (large) evaporation indicates the amount of evaporated water measured using a container with a diameter of 10cm (30cm) during 24 hours (unit: mm). Generally, the measured values using the two types of equipment are of slight differences. SSD represents the hours of sunshine measured during a day for a specific location on earth. The max wind speed indicates the max mean wind speed during any 10 minutes within a day's time. The extreme wind speed indicates the max instant (for 1s) wind speed within a day's time. The max wind direction indicates the dominant wind direction for the period with the max wind speed. As there are one or more observation stations for each city, the daily value for each meteorological factor for each city was calculated using the mean value of all available observation stations within the target city. To conduct time series

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~~[2] SmallEVP and LargeEVP indicate the evaporation amount measured using small-diameter and large-diameter equipments respectively. Generally, the measured values using the two types of equipment are of slight differences.~~

~~[3] Sunshine duration represents the hours of sunshine measured during a day for a specific location on earth.~~

~~[4] The max wind speed indicates the max mean wind speed during any 10 minutes within a day's time. The extreme wind speed indicates the max instant (for 1s) wind speed within a day's time.~~

~~[5] The max wind direction indicates the dominant wind direction for the period with the max wind speed.~~

comparison, we also collected meteorological data from March 1st, 2014 to February 28th, 2017.

2.2 Study sites

For a comprehensive understanding of meteorological influences on local $PM_{2.5}$ concentrations across China，all monitoring cities (except for Liaocheng and Zhuji, where continuous valid meteorological data were not available) during the study period were selected for this research. The 188 cities included most major cities (Beijing, Shanghai, Guangzhou, etc.) in China. For regions (e.g. Beijing-Tianjin-Hebei region) with heavy air pollution, the density of monitored cities was much higher than that in regions with good air quality.

3 Methods

Due to complicated interactions in the atmospheric environment, it is highly difficult to quantify the causality of individual meteorological factors on $PM_{2.5}$ concentrations through correlation analysis. Instead, a robust causality analysis method is required.

To extract the coupling between individual variables in complex systems, Sugihara et al. (2012) proposed a convergent cross mapping (CCM) method. Different from Granger causality (GC) analysis (Granger, 1980), the CCM method is sensitive to weak to moderate coupling in ecological time series. By analyzing the temporal variations of two time-series variables, their bidirectional coupling can be featured with a convergent map. If the influence of one variable on the other variable is presented as a convergent curve with increasing time series length, then the causality is detected; If the curve demonstrates no convergent trend, then no causality exists. The predictive skill (defined as ρ value), which ranges from 0 to 1, suggests the quantitative causality of one variable on the other.

The principle of CCM algorithms is briefly explained as follows (Luo et al. 2014). Two time series $\{X\}= [X(1), \ldots, X(L)]$ and $\{Y\} = [Y(1), \ldots, Y(L)]$ are defined as the temporal variations of two variables X and Y. For r = S to L (S < L), two partial time series $[X(1), \ldots, X(L_P)]$ and $[Y(1), \ldots, Y(L_P)]$ are extracted from the original time series (r is the current position whilst S is the start position in the time series). Following this, the shadow manifold M_X is generated from $\{X\}$, which is a set of lagged-coordinate vectors $x(t) = <X(t), X(t-\tau), ..., X(t-(E-1)\tau)>$ for $t = 1+(E-1)\tau$ to $t = r$. To generate a cross-mapped estimate of Y(t) ($\hat{Y}$ (t)|M_X), the contemporaneous lagged-coordinate vector on M_X, x(t) is located, and then its E+1 nearest neighbors are extracted, where E+1 is the minimum number of points required for a bounding simplex in an E-dimensional space (Sugihara and May, 1990). Next, the time index of the E+1 nearest neighbors of x(t) is denoted as t_1, ..., t_{E+1}. These time index are used to identify neighbor points in Y and then estimate Y(t) according to a locally weighted mean of E+1 $Y(t_i)$ values (Equation 1).

$$\hat{Y}(t)|M_X = \sum_{i=1}^{E+1} w_i Y(t_i) \qquad \text{（E1）}$$

Where w_i is a weight calculated according to the distance between X(t) and its ith nearest neighbor on M_X. $Y(t_i)$ are contemporaneous values of Y. The weight w_i is determined according to Equation 2.

$$w_i = u_i \Big/ \sum_{j=1}^{E+1} u_j \qquad \text{（E2）}$$

Where $u_i = e^{-d[\underline{x}(t),\underline{x}(t_i)]/d[\underline{x}(t),\underline{x}(t_1)]}$ whilst $d[x(t), x(t_i)]$ represents the Euclidean distance between two vectors.

In our previous research, interactions between the air quality in neighboring cities (Chen, Z. et al., 2016), and bidirectional coupling between individual meteorological factors and $PM_{2.5}$ concentrations (Chen, Z. et al., 2017) were quantified effectively using the CCM method. By comparing the performance of correlation analysis and CCM method, Chen, Z. et al. (2017) suggested that correlation analysis may lead to a diversity of biases due to complicated interactions between individual meteorological factors. Firstly, some mirage correlations (two variables with a moderate correlation coefficient) extracted using the correlation analysis were revealed effectively using the CCM method (the ρ value between two variables was 0). Secondly, some weak coupling, which was hardly detected using the correlation analysis (the correlation between the two variables were not significant), was extracted using the CCM method (a small ρ value). Meanwhile, as Sugihara et al. (2012) suggested, the correlation between two variables could be influenced significantly by other agent variables and thus the value of correlation coefficient between two variables could not reflect the actual causality between them. Chen et al. (2017) further revealed that the correlation coefficient between individual meteorological factors and $PM_{2.5}$ concentrations was usually much larger than the ρ value. This indicated that the causality of individual meteorological factors on $PM_{2.5}$

concentrations was generally overestimated using the correlation analysis, due to the influences from other meteorological factors. In this case, the CCM method is an appropriate tool for quantifying bidirectional interactions between $PM_{2.5}$ concentrations and individual meteorological factors in complicated atmospheric environment.

4 Results

Seasonal variations of $PM_{2.5}$ concentrations have been ~~proved by a large body of studies~~revealed in Beijing (Chen et al., 2015; Chen, Y. et al., 2016; Chen, Z. et al., 2016), Nanjing (Shen et al., 2014), Shandong Province (Yang and Christakos, 2015) and the Beijing-Tianjin-Hebei region (Wang et al. 2015; Chen, Z. et al., 2017). In addition to these local and regional studies, Cao et al. (2012) further compared seasonal variations of $PM_{2.5}$ concentrations in seven southern cities (Chongqing, Guangzhou, Hong Kong, Hangzhou, Shanghai, Wuhan, and Xiamen) and seven northern cities (Beijing, Changchun, Jinchang, Qingdao, Tianjin, Xi’an, and Yulin) across China. ~~(Cao et al., 2012; Shen et al., 2014; Yang and Christakos, 2015; Wang et al., 2015; Chen et al., 2015; Chen, Y. et al. 2016; Chen, Z. et al., 2016).~~ Hence, the research period was divided into four seasons. According to traditional season division for China, spring was set as the period between March 1st, 2014 and May 31st, 2014; summer was set as the period between June 1st, 2014 and August 31st, 2014; autumn was set as the period between September 1st, 2014 and November 30th, 2014; and winter was set as the period between December 1st, 2014 and February 28th, 2015. For each city, the bidirectional coupling between individual meteorological factors and $PM_{2.5}$ concentrations in different seasons was analyzed respectively using the CCM method. The CCM method is highly automatic and only few parameters need to be set for running this algorithm: E (number of dimensions for the attractor reconstruction), τ (time lag) and b (number of nearest neighbors to use for prediction). The value of E can be 2 or 3. A larger value of E produces more accurate convergent maps. The variable b is decided by E (b = E + 1). A small value of τ leads to a fine-resolution convergent map, yet requires much more processing time. Through experiments, we found that the final results were not sensitive to the selection of parameters and different parameters mainly exerted influences on the presentation effects of CCM. In this research, to acquire optimal interpretation effects of convergent cross maps, the value of τ was set as 2 days and the value of E was set 3. For each meteorological factor, its causality coupling with $PM_{2.5}$ concentrations can be

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represented using a convergent map. Since it is not feasible to present all these convergent maps here, we simply display some exemplary maps to demonstrate how CCM works (Fig 1). As a heavily polluted city, we presented the interactions between $PM_{2.5}$ concentrations and meteorological factors in Beijing in winter, when the local $PM_{2.5}$ concentration was the highest, as an example. Four major meteorological factors, wind, humidity, radiation and temperature, which exerted much stronger influences on $PM_{2.5}$ concentrations than other factors, were employed. Due to the strong bidirectional coupling between $PM_{2.5}$ concentrations and these meteorological factors, Figure 1 not only demonstrates how CCM output could be interpreted, but also provides readers with a general comparison of the magnitude of simultaneous influences of different meteorological factors on the local $PM_{2.5}$ concentration and its feedback effects.

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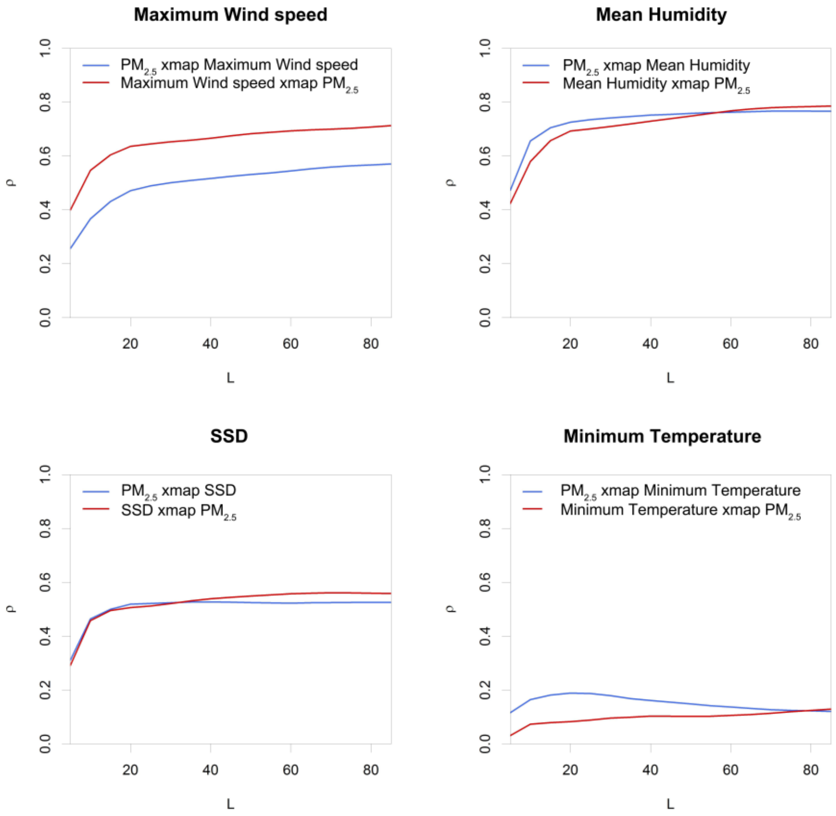

Fig 1. Illustrative CCM results to demonstrate the bidirectional coupling between

meteorological factors and $PM_{2.5}$ concentrations in Beijing (2014, winter)

ρ: predictive skills. L: the length of time series. A xmap B stands for convergent cross mapping B from A, in other words, the causality of variable B on A. For instance, $PM_{2.5}$ xmap meanRHU stands for the causality of meanRHU on $PM_{2.5}$ concentrations. meanRHU xmap $PM_{2.5}$ stands for the feedback effect of $PM_{2.5}$ on meanRHU concentrations. ρ indicates the predictive skills of using meanRHU to retrieve $PM_{2.5}$ concentrations.

According to Fig 1, one can see that the quantitative influence of individual meteorological factors on $PM_{2.5}$ was well extracted using the CCM method whilst the feedback effect of $PM_{2.5}$ on specific meteorological factors was revealed as well. For Beijing, meanRHU and maxWIN exerted a strong influence on local $PM_{2.5}$ concentrations in Winter (ρ > 0.4) whilst SSD and minTEM also had a weaker influence on local $PM_{2.5}$ concentrations. (ρ close to 0.2). On the other hand, ~~serious haze weather (~~high $PM_{2.5}$ concentrations~~)~~ had an even stronger feedback influence on meanRHU, maxWIN and SSD (ρ close to 0.6) whilst $PM_{2.5}$ had little influence on minTEM (ρ close to 0). The bidirectional coupling between $PM_{2.5}$ concentrations and individual meteorological factors provides useful reference for a better understanding of the form and development of ~~serious haze events~~$PM_{2.5}$-induced air pollution episodes. For Beijing, low wind speed (high humidity and low SSD) in winter results in high $PM_{2.5}$ concentrations, which in turn causes lower wind speed (higher humidity and lower SSD). In consequence, $PM_{2.5}$ concentrations are increased further by the changing wind (humidity and SSD) situation. This mechanism causes a quickly rising $PM_{2.5}$ concentrations, which brings the atmospheric environment to a comparatively stable status. In this case, ~~the haze is unlikely to disperse and~~ persistent ~~haze weather~~high-concentration $PM_{2.5}$ is unlikely to disperse and –usually lasts~~s~~ for a long period in this region. Similarly, bidirectional interactions between $PM_{2.5}$ concentrations and other meteorological factors can as well be quantified using the CCM method. Since the main aim of this research is to understand the influence of individual meteorological factors on $PM_{2.5}$ concentrations across China, the feedback effect of $PM_{2.5}$ concentrations on specific meteorological factors is not explained in details herein.

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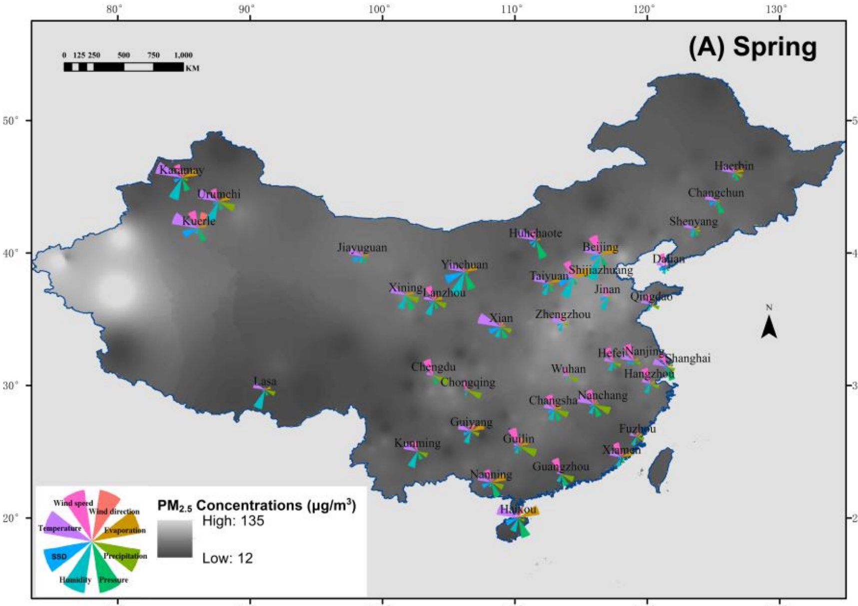
The ρ value is a direct indicator of quantitative causality. For this research, the maximum ρ value of all sub-factors in the same category was used as the causality of this specific meteorological factor on $PM_{2.5}$ concentrations. E.g. for a specific city, the maximum ρ value of max temperature, mean temperature, minimum temperature, and largest temperature difference for the day ~~maxTEM, meanTEM, minTEM and difTEM~~ is used as the influence of temperature on local $PM_{2.5}$ concentrations. For this research, we collected meteorological and $PM_{2.5}$ data for three consecutive years. To avoid the analysis of inconsecutive time series, which may influence the CCM result, we did not calculate the general influence of individual meteorological factors on $PM_{2.5}$ concentrations during 2014-2016 by analyzing three isolated periods (e.g. April- June, 2014, April-June, 2015, and April- June, 2016) as a complete data set. Instead, for each city, we quantified the influence of individual meteorological factors on $PM_{2.5}$ concentrations for each season in 2014, 2015 and 2016 respectively and calculated the mean ρ value during 2014-2016 for each city.

~~Generally, it is difficult to properly demonstrate the influence of eight meteorological factors on $PM_{2.5}$ concentrations for all 188 cities on a comprehensive map. Therefore, two cartography strategies were employed to explain the meteorological influences on $PM_{2.5}$ concentrations across China.~~

4.1 Comprehensive meteorological influences on $PM_{2.5}$ concentrations in some regional representative cities

When the ρ value for each meteorological factor was calculated, a wind rose, which presents the quantitative influences of all individual meteorological factors on $PM_{2.5}$ concentrations, can be produced for each city. It is not feasible to present all 188 wind roses simultaneously, due to severe overlapping effects. Thus, considering the social-economic factors, 37 regional representative cities (including all 31 provincial capital cities in mainland China), which are the largest and most important cities for specific regions, were selected to produce a wind rose map of meteorological influences on $PM_{2.5}$ concentrations across China (Fig 2).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Beijing
Haerbin
Changchun
Shenyang
Dalian
Taiyuan
Shijiazhuang
Jinan
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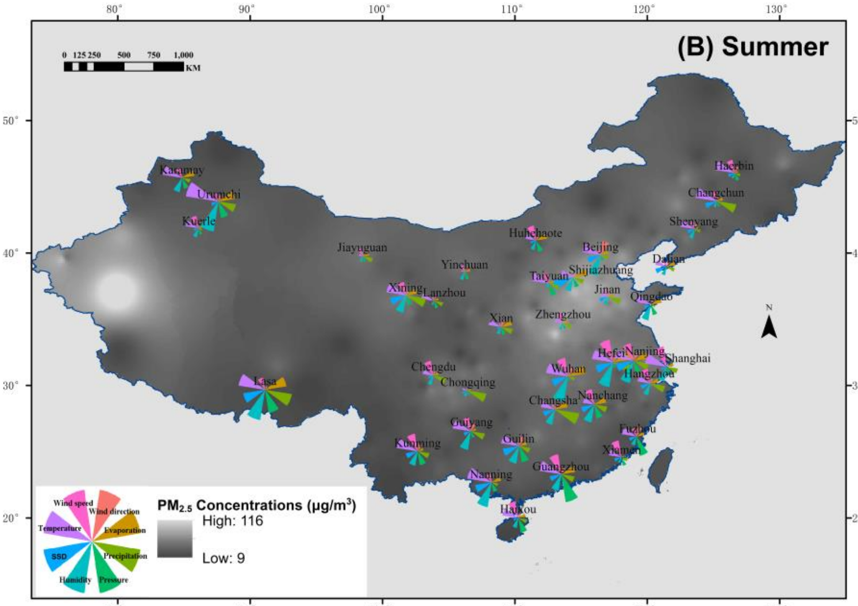
Chengdu
Chongqing
Changsha
Nanchang
Guiyang
Kunming
Guilin
Fuzhou
Xiamen
Guangzhou
Nanning
Haikou
Wind speed
Wind direction
Temperature
Evaporation
SSD
Precipitation
Humidity
Pressure
PM$_{2.5}$ Concentrations (μg/m^3)
High: 135
Low: 12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Yinchuan
Huhehaote
Beijing
Haerbin
Changchun
Shenyang
Dalian
Taiyuan
Shijiazhuang
Jinan
Qingdao
Xining
Lanzhou
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Nanjing
Shanghai
Hangzhou
Wuhan
Lasa
Chengdu
Chongqing
Changsha
Nanchang
Guiyang
Kunming
Guilin
Fuzhou
Xiamen
Guangzhou
Nanning
Haikou
Wind speed
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Temperature
Evaporation
SSD
Precipitation
Humidity
Pressure
PM$_{2.5}$ Concentrations (μg/m^3)
High: 116
Low: 9

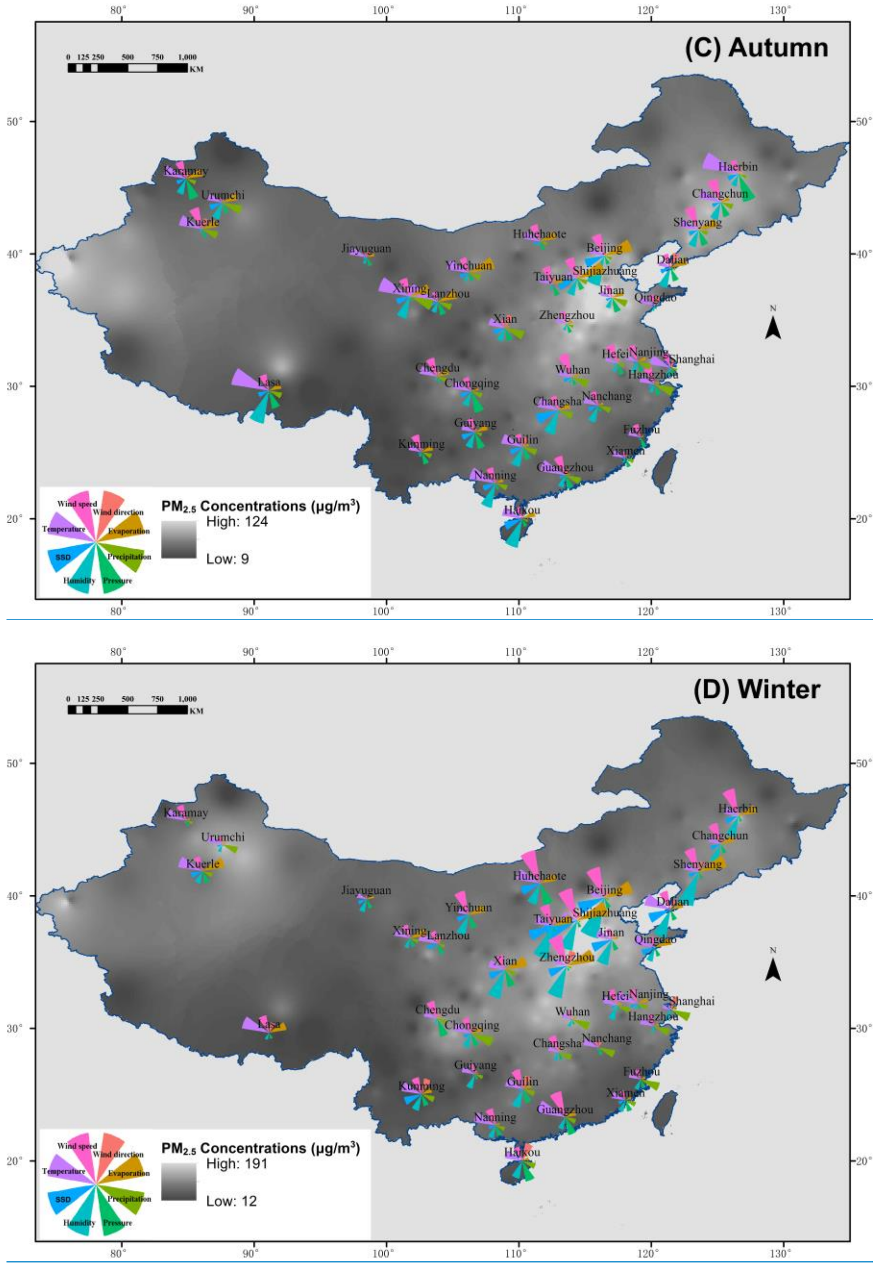

Fig 2. Wind rose map of influences of eight individual meteorological factors on $PM_{2.5}$ concentrations across China (37 representative cities) during 2014-2016

According to Fig 2, some spatial and temporal patterns of meteorological influences on $PM_{2.5}$ concentrations at the national scale can be found as follows:

a. Like seasonal variations of $PM_{2.5}$ concentrations, the influences of individual meteorological factors on local $PM_{2.5}$ concentrations vary significantly. For a specific city, the dominant meteorological driver for $PM_{2.5}$ concentrations in one season may become insignificant in another season. E.g. in winter, one major meteorological influencing factor for Beijing is wind (The mean ρ value during 2014-2016 was 0.57), which exerts little influence on $PM_{2.5}$ concentrations in summer (The mean ρ value during 2014-2016 was 0.10). Furthermore, it is noted that seasonal variations of meteorological influences on $PM_{2.5}$ concentrations apply to all these representative cities, as the shape and size of wind rose for each city change significantly across different seasons. Take several mega cities in different regions for instance. During 2014-2016, the three major meteorological influencing factors for $PM_{2.5}$ concentrations in Beijing, a mega city in the North China plain, were as follows: humidity (0.48)， wind (0.37) and evaporation (0.31) for spring, humidity (0.39)， temperature (0.34) and SSD (0.25) for summer, humidity (0.56)， evaporation (0.51) and wind (0.41) for autumn, and humidity (0.76), wind (0.57) and evaporation (0.52) for winter. The three major meteorological influencing factors for $PM_{2.5}$ concentrations in Shanghai, a mega city in the Yangtze River Basin, were as follows: temperature (0.264), air pressure (0.260) and wind (0.25) for spring, temperature (0.40), wind (0.38) and humidity (0.27) for summer, temperature (0.39), wind (0.28) and humidity (0.17) for autumn, and precipitation (0.36), wind direction (0.25) and humidity (0.19) for winter. The three major meteorological influencing factors for $PM_{2.5}$ concentrations in Wuhan, a major city in Central China region, were as follows: precipitation (0.18), wind (0.16) and temperature (0.09) for spring, humidity (0.47), temperature (0.41) and wind (0.34) for summer, wind (0.44), precipitation (0.31) and temperature (0.26) for autumn, and precipitation (0.33), temperature (0.19) and wind (0.15) for winter. The three major meteorological influencing factors for $PM_{2.5}$ concentrations in Guangzhou, a major city in Southern China region, were as follows: wind (0.31), precipitation (0.24) and air pressure (0.23) for spring, air pressure (0.51), temperature (0.41) and wind (0.37) for summer, temperature (0.47), wind (0.36) and precipitation (0.29) for autumn, and temperature (0.52), wind (0.48) and air pressure (0.33). Notable seasonal variations of meteorological influences on $PM_{2.5}$ concentrations

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were found in these mega cities across China.

b. In spite of notable differences in the shape and size of wind roses, meteorological influences on $PM_{2.5}$ concentrations cities are of some regional patterns. ~~For instance,~~ $PM_{2.5}$ concentrations in cities within the North China region are influenced by similar dominant meteorological factors, especially in winter, when $PM_{2.5}$ concentrations in these cities was high. Take four major cities, Beijing, Tianjin, Taiyuan and Shijiangzhuang, in the North China Plain for example. For winter, SSD, evaporation, humidity and wind were the major meteorological factors for $PM_{2.5}$ concentrations in the four cities and the ρ value of these four factors was 0.50, 0.52, 0.76 and 0.57 for Beijing, 0.41, 0.44, 0.56 and 0.50 for Tianjin, 0.44, 0.36, 0.61 and 0.41 for Taiyuan, and 0.62, 0.58, 0.56 and 0.60 for Shijiazhuang respectively, presenting a similar regional pattern. Meanwhile, meteorological influences on $PM_{2.5}$ concentrations in cities within the Yangtze River Basin, especially the dominant factors, were also of some regional similarities. Take four major cities in the Yangtze River Basin, Shanghai, Nanjing, Hangzhou and Nanchang for example. For summer, precipitation, humidity, temperature and wind were the major meteorological factors for $PM_{2.5}$ concentrations in these four cities and the ρ value of these factors was 0.21, 0.27, 0.40 and 0.38 for Shanghai, 0.29, 0.41, 0.34 and 0.33 for Nanjing, 0.28, 0.27, 0.23 and 0.27 for Hangzhou, and 0.24, 0.33, 0.21 and 0.29 for Nanchang. Despite of some differences in the ρ values, similar dominant meteorological factors and the similar magnitude of meteorological influences demonstrated regional similarities of meteorological influences on $PM_{2.5}$ concentrations in the Yangtze River Basin.

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As we can see, meteorological influences on $PM_{2.5}$ concentrations in China are mainly controlled by geographical conditions (e.g. terrain and landscape patterns).

c. For the heavily polluted North China region, the higher the local $PM_{2.5}$ concentrations, the larger influence meteorological factors exerts on $PM_{2.5}$ concentrations. $PM_{2.5}$ concentrations are usually the highest in winter, causing serious ~~haze~~ air pollution episodes across China, the North China region in particular. Meanwhile, $PM_{2.5}$ concentrations in spring and summer are comparatively low. Accordingly, there are more influencing meteorological factors on $PM_{2.5}$ concentrations for cities within this region and the ρ value of these meteorological factors is notably larger in winter. Take four major cities in the North China region for instance. For Beijing, the major influencing meteorological factors in summer were temperature (0.34), humidity (0.39) and SSD (0.25) whilst the major influencing meteorological factors in winter were humidity (0.76), wind (0.57), evaporation (0.52) and SSD (0.5). For Tianjin, the major influencing meteorological factors in summer were precipitation (0.34), temperature (0.22) and air pressure (0.25) whilst the major influencing meteorological factors in winter were humidity (0.76), wind (0.57), evaporation (0.52) and SSD (0.50). For Shijiazhuang, the major influencing meteorological factors in summer were SSD (0.4), humidity (0.26) and evaporation (0.26) whilst the major influencing meteorological factors in winter were SSD (0.62), wind (0.60), evaporation (0.58) and humidity (0.56). For Taiyuan, the major influencing meteorological factors in summer were temperature (0.32), air pressure (0.23) and precipitation (0.20) whilst the major influencing meteorological factors in winter were humidity (0.61), SSD (0.44) and wind (0.41). As explained, bidirectional interactions between meteorological factors and $PM_{2.5}$ concentrations may lead to complicated mechanisms that further enhance local $PM_{2.5}$ concentrations significantly. Therefore, strong meteorological influences on $PM_{2.5}$ concentrations in winter are a major cause for the form and persistence of ~~haze events~~high $PM_{2.5}$ concentrations within the North China region.

~~Although some general patterns of meteorological influences on $PM_{2.5}$ concentrations across China may be concluded according to Fig 2, spatial and temporal variations of meteorological influences on $PM_{2.5}$ concentrations should be further examined in depth based on the statistics of all 188 monitoring cities. Hence, we employed another cartography strategy to demonstrate spatial and temporal variations of meteorological influences on local $PM_{2.5}$ concentrations across China.~~

4.2 Spatial and temporal variations of the dominant meteorological influence on local $PM_{2.5}$ concentrations across China

Through statistical analysis, we selected the factor with the largest ρ value as the dominant meteorological factor for local $PM_{2.5}$ concentrations. The spatial and temporal variations of the dominant meteorological influence on local $PM_{2.5}$ concentrations across China are demonstrated as Fig 3. According to Fig 3, some spatio-temporal characteristics of meteorological influences on $PM_{2.5}$ concentrations can be further concluded:

a. The dominant meteorological factor for $PM_{2.5}$ concentrations is closely related to geographical conditions. For instance, the factor of precipitation may exert a key influence on local $PM_{2.5}$ concentrations in some coastal cities and cities within the Yangtze River ~~basin~~ Basin whilst this meteorological factor exerts limited influence on $PM_{2.5}$ concentrations within some inland regions ~~(e.g. the Beijing-Tianjin-Hebei region)~~. Here we analyzed the ρ value of precipitation in cities within the Yangtze River Basin and cities within the Beijing-Tianjin-Hebei region respectively. For winter, precipitation was the dominant factor for $PM_{2.5}$ concentrations in Shanghai, Hangzhou and Nanchang within the Yangtze River Basin and the ρ value of precipitation was 0.36, 0.29 and 0.31 respectively. Meanwhile, the ρ value of precipitation in Beijing, Tianjin and Shijiazhuang within the Beijing-Tianjin-Hebei region was 0.08, 0.01 and 0.06 respectively.

b. Some meteorological factors can be the dominant factor for cities within different regions but some (e.g. evaporation and SSD) are mainly the dominant meteorological factor for $PM_{2.5}$ concentrations in cities within some specific regions. In other words, some factors can be regarded as regional and national meteorological factors for $PM_{2.5}$ concentrations, yet some meteorological factors are context-related influencing factors for local $PM_{2.5}$ concentrations. ~~For instance~~Specifically, such factors as temperature, wind and humidity serve as the dominant meteorological factors in many regions, including Northeast, Northwest, coastal areas and inland areas; Meanwhile, such factors as SSD and w~~W~~ind direction serve as the dominant meteorological factors mainly in some inland regions. The prevalence of different meteorological factors across China can also be reflected according to the number of cities where this specific factor is the dominant factor for local $PM_{2.5}$ concentrations. For winter, the number of cities with temperature, wind or humidity as the dominant factor was 56，48 and 44 respectively. Meanwhile, the number of cities with SSD or wind direction as the dominant factor was 3 and 1

respectively.

c. Similar to patterns revealed in Fig 2, the ρ value for the dominant meteorological factors is much larger in winter than that in summer. Furthermore, it is noted that the dominant meteorological factors demonstrate more regional similarity in winter. ~~For instance~~Specially, the dominant meteorological factors for $PM_{2.5}$ concentrations in the

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heavily polluted North China region are more concentrated and homogeneously distributed in winter (mainly the wind and humidity factor) whilst a diversity of dominant meteorological factors (includes ~~wind~~humidity, temperature, SSD and air pressure) for $PM_{2.5}$ concentrations is irregularly distributed within this region in summer. Take some major cities in North China region for instance. For winter, the dominant meteorological factors for Beijing, Tianjin, Taiyuan, Zhangjiakou, Handan and Jining was humidity (0.76)，humidity (0.56), humidity (0.61), wind (0.62), humidity (0.43) and humidity (0.52) respectively. Meanwhile, for summer, the dominant meteorological factors for Beijing, Tianjin, Taiyuan, Zhangjiakou, Baoding, Handan and Jining was humidity (0.39), precipitation (0.28), temperature (0.23), temperature (0.47), air pressure (0.21) and SSD (0.18). According to this pattern, when a regional ~~haze~~ $PM_{2.5}$-induced air pollution episode occurs in winter, the regional air quality is more likely to be simultaneously improved by the same meteorological factor. This is consistent with the common scene in winter that regional ~~haze events~~air pollution episodes in the Beijing-Tianjin-Hebei region can be considerably mitigated by strong northwesterly synoptic winds， which are produced by presence of high air pressure in northwest Beijing (NW-High) （Tie et al., 2015; Miao et al., 2015）. On the other hand, regional air pollution in summer can hardly be solved simultaneously through one specific meteorological factor.

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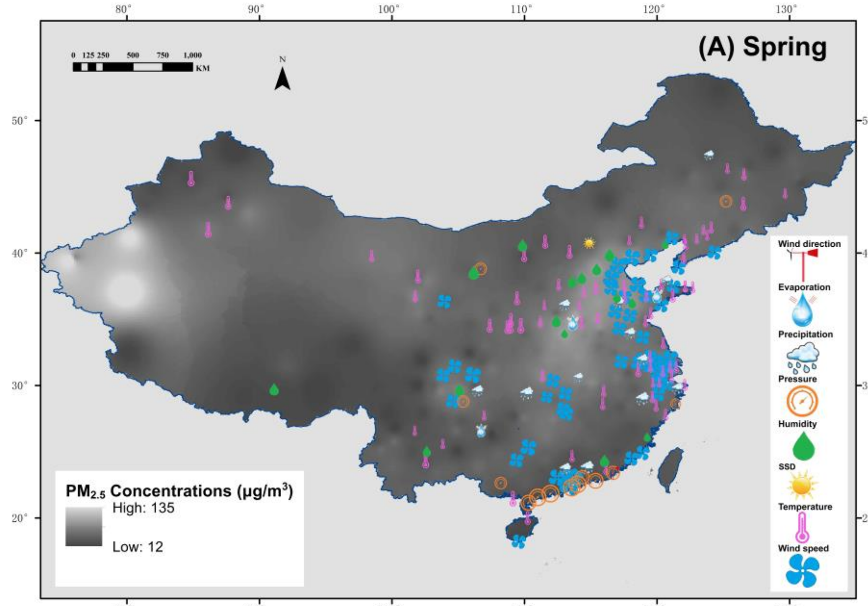
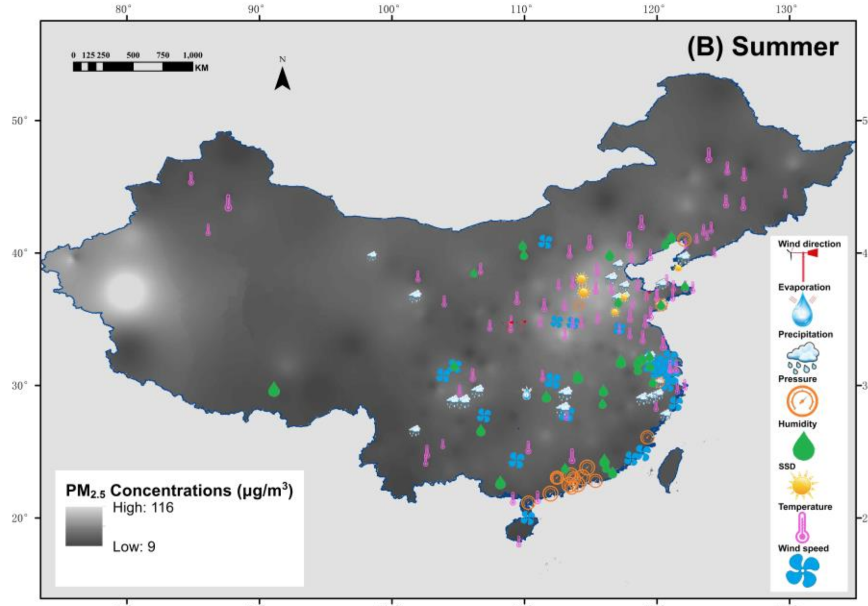

(A) Spring
0 125 250 500 750 1,000
KM
N
80° 90° 100° 110° 120° 130°
50° 40° 30° 20°
PM$_{2.5}$ Concentrations (μg/m^3)
High: 135
Low: 12
Wind direction
Evaporation
Precipitation
Pressure
Humidity
SSD
Temperature
Wind speed


(B) Summer
0 125 250 500 750 1,000
KM
N
80° 90° 100° 110° 120° 130°
50° 40° 30° 20°
PM$_{2.5}$ Concentrations (μg/m^3)
High: 116
Low: 9
Wind direction
Evaporation
Precipitation
Pressure
Humidity
SSD
Temperature
Wind speed

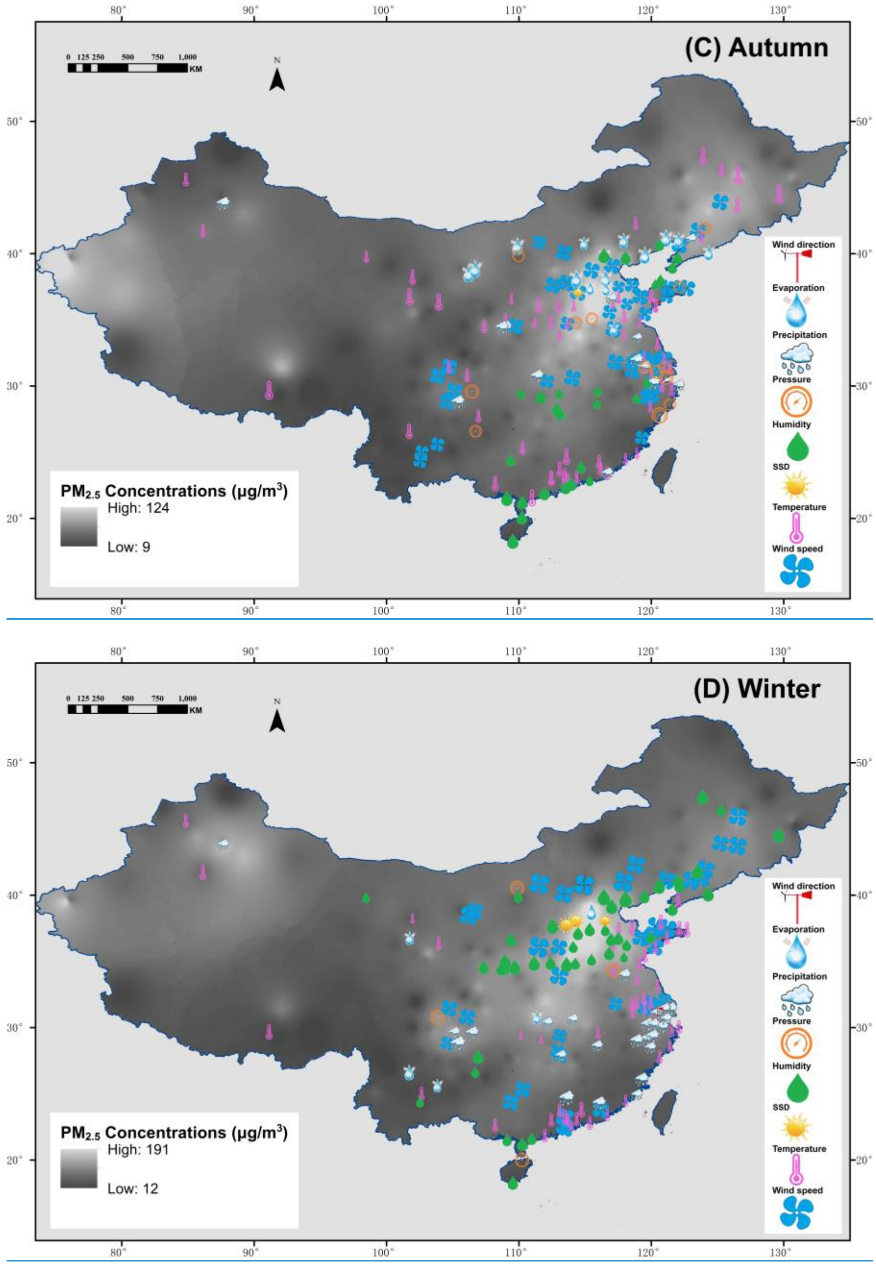


Fig 3. The dominant meteorological factor for local $PM_{2.5}$ concentrations in 188 monitoring cities across China

The size of symbols indicates the ρ value of the meteorological factor on local $PM_{2.5}$ concentrations.

4.3 Comparative statistics of the influence of individual meteorological factors on local $PM_{2.5}$ concentrations across China

In addition to meteorological influences on $PM_{2.5}$ concentrations for individual cities, we examined and compared the comprehensive influence of individual meteorological factors on $PM_{2.5}$ concentrations at a national scale. The results are presented as Table 1 and Fig 4.

Table 1. The comparison of the influence of individual meteorological factors on $PM_{2.5}$ concentrations in 188 cities across China (2014-2016)

Season	Factor	TEM	SSD	PRE	EVP	PRS	RHU	WIN	Dir_WIN
Spring	**No. of cities**[1]	76	1	13	3	13	17	64	1
	Mean ρ value	0.254	0.102	0.143	0.108	0.177	0.161	0.222	0.094
	SD of ρ value	0.106	0.071	0.088	0.081	0.123	0.105	0.102	0.077
	Max ρ value	0.572	0.366	0.385	0.397	0.653	0.475	0.595	0.429
Summer	**No. of cities**	78	5	22	1	20	32	27	3
	Mean ρ value	0.272	0.136	0.183	0.137	0.163	0.219	0.191	0.087
	SD of ρ value	0.098	0.086	0.099	0.088	0.109	0.118	0.095	0.062
	Max ρ value	0.604	0.433	0.536	0.399	0.518	0.562	0.453	0.311
Autumn	**No. of cities**	70	1	13	15	13	27	48	1
	Mean ρ value	0.316	0.164	0.191	0.181	0.199	0.247	0.265	0.104
	SD of ρ value	0.109	0.098	0.093	0.117	0.091	0.125	0.089	0.074
	Max ρ value	0.702	0.479	0.430	0.514	0.524	0.662	0.488	0.331
Winter	**No. of cities**	56	3	27	5	4	48	44	1
	Mean ρ value	0.306	0.183	0.166	0.190	0.180	0.304	0.299	0.119
	SD of ρ value	0.094	0.129	0.115	0.130	0.086	0.161	0.136	0.092
	Max ρ value	0.527	0.615	0.473	0.595	0.427	0.755	0.623	0.560

[1] No. of cities: the number of cities with this factor as the dominant meteorological factor (its ρ value is the largest amongst eight factors) on local $PM_{2.5}$ concentrations.

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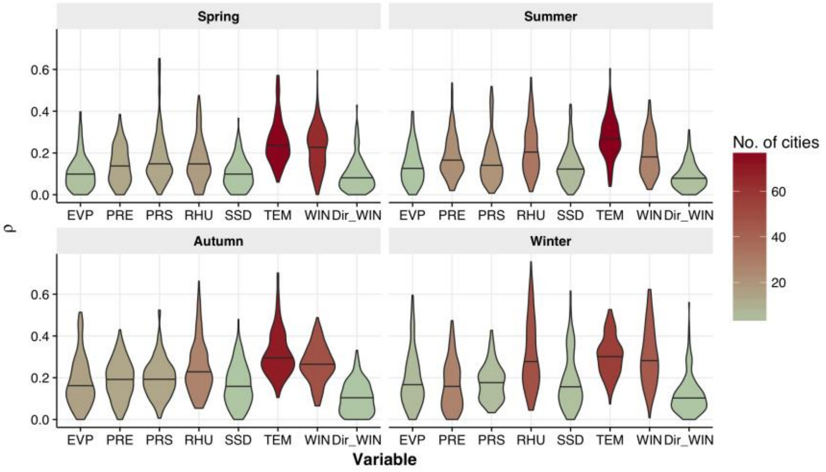

Fig 4. Violin plots of the influence of eight different meteorological factors on local $PM_{2.5}$ concentrations in 188 cities across China

No. of cities: the number of cities with this factor as the dominant meteorological factor (its ρ value is the largest amongst eight factors) on local $PM_{2.5}$ concentrations. The shape of the violin bars indicated the frequency distribution of ρ value for 188 cities.

We compared the influence of individual meteorological factors on $PM_{2.5}$ concentrations from different perspectives.

a. From a national perspective, temperature, humidity, and wind exert stronger influences on local $PM_{2.5}$ concentrations than other factors. The annual mean ρ value for temperature, wind and humidity was 0.287, 0.244 and 0.233, compared with wind direction (0.101), SSD (0.146), evaporation (0.155), precipitation (0.171) and air pressure (0.180). Amongst the eight factors, temperature was ~~proved~~ found to be the most influential meteorological factor for general $PM_{2.5}$ concentrations in China. In addition to the largest mean ρ value, temperature was the dominant meteorological factors for most cities in all seasons. Furthermore, the Coefficient of Variation (SD /mean×100%) for temperature was much smaller than other factors, indicating the consistent influence of temperature on local $PM_{2.5}$ concentrations across China.

b. Although some meteorological factors exert a limited influence on $PM_{2.5}$ concentrations at a national scale, these factors may be a key meteorological factor for local $PM_{2.5}$ concentrations. As shown in Table 1, the max ρ value for each

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meteorological factor was large than 0.35 for all seasons (except for the wind direction factor in summer and autumn), indicating a very strong influence on local $PM_{2.5}$ concentrations in some specific regions. As a result, when analyzing meteorological influences on local $PM_{2.5}$ concentrations for a specific city, meteorological factors that have little influence on $PM_{2.5}$ concentrations at a large scale should also be comprehensively considered.

c. Some factors (e.g. precipitation in summer and winter) may be the dominant meteorological factors for a large number of cities, though the mean ρ value remained small. This may be attributed to the fact that these meteorological factors mainly exert influence on local $PM_{2.5}$ concentrations in those cities (seasons) where (when) the general $PM_{2.5}$ concentrations is not high. Taking the precipitation as an example. Luo et al. (2017). pointed out that there may be thresholds for the negative influences of precipitations on PM2.5 concentrations and Guo et al. (2016) found that the same amount of precipitation led to a weaker washing-off effect in areas with higher $PM_{2.5}$ concentrations. Hence, precipitation mainly exerts a dominant influence on local $PM_{2.5}$ concentrations in winter for Yangtze River Basin or coastal cities, where the amount of precipitation is large and the $PM_{2.5}$ concentration is low, whilst precipitation exerts a limited role in northern China, where the amount of precipitation is small and the $PM_{2.5}$ concentration is high. Therefore, as explained above, comprehensive meteorological influences on $PM_{2.5}$ concentrations are limited considerably.

5 Discussion

Despite the lack of a comprehensive comparison of meteorological influences on $PM_{2.5}$ concentrations ~~in~~ across different regions, ~~some studies concerning meteorology-$PM_{2.5}$ relationship in specific areas have been conducted and~~ correlations between individual meteorological factors and $PM_{2.5}$ concentrations have been analyzed in such mega cities as Nanjing (Chen, T. et al., 2016; Shen and Li., 2016;), Beijing (Huang et al., 2015; Yin et al., 2016), Wuhan (Zhang et al., 2017), Hangzhou (Jian et al., 2012), Chengdu (Zeng and Zhang, et al. 2017) and Hong Kong (Fung et al., 2014). These studies mainly employed correlation analysis to quantify the influence of several meteorological factors on $PM_{2.5}$ concentrations and suggested that meteorological influences on $PM_{2.5}$

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concentrations varied significantly across regions. The dominant meteorological factors for P$_{2.5}$ concentrations (presented as the largest correlation coefficients in previous studies and the largest ρ value in this research) demonstrated notable regional differences. For Nanjing (Chen, T. et al., 2016), a mega city in the Yangtze River, and Hong Kong (Fung et al., 2014), a mega coastal city, precipitation exerted the strongest influence whilst wind speed exerted a weak influence on PM$_{2.5}$ concentrations in winter. On the other hand, for winter, wind speed was the dominant meteorological factor for PM$_{2.5}$ concentrations in Beijing (Huang et al., 2015.) , a mega city in North China, and precipitation played a weak role in affecting local PM$_{2.5}$ concentrations . These studies generally analyzed and compared the influences of different meteorological factors on PM$_{2.5}$ concentrations and extracted the dominant meteorological influencing factors for specific areas. ~~However, most studies were conducted at the local scale and few studies have focused on the comparison and statistics of meteorological influences on PM$_{2.5}$ concentrations in different areas. Meanwhile, although the correlation coefficient can be used to understand and compare the general magnitude of the influence of individual meteorological factors, the correlation analysis, as explained above, may lead to large bias in quantifying the meteorological influences on PM$_{2.5}$ concentrations.~~

~~Different from previous studies conducted at the local scale, this research conducted at the national scale better understood spatial and temporal patterns of meteorological influences on PM$_{2.5}$ concentrations that will not be revealed in small-scale studies. The finding from this research was consistent with and a major extension of that from previous studies by quantifying the influence of individual meteorological factors in a large number of cities across China, instead of several scattered cities, using a more robust causality analysis method, other than the correlation analysis.~~ Compared with studies at a local or regional scale, this research conducted at the national scale provided a better understanding of spatial and temporal patterns of meteorological influences on PM$_{2.5}$ concentrations across China, for the following reasons. a. A national perspective. Previous studies conducted at a local scale mainly focused on a specific city (e.g. Beijing), and can hardly reveal spatio-temporal patterns of meteorological influences on PM$_{2.5}$ concentrations at a large scale (e.g. the North China plain). This research, on the other hand, quantified the influence of meteorological factors on $PM_{2.5}$ concentrations for 188 cities across China, and thus revealed some regional patterns of meteorological influences on $PM_{2.5}$ concentrations in some typical regions (e.g. North China region or Yangtze River Basin). b. A unified research period and set of meteorological factors. Previous studies employed short-term observation data (e.g. one season or one year) to examine the meteorological influences on local $PM_{2.5}$ concentrations in specific cities. Due to the discrepancy in research periods and sets of meteorological factors, the findings from different local-scale studies cannot be compared and comprehensively understood. This research employed daily $PM_{2.5}$ and meteorological data of three consecutive years and a unified set of eight meteorological factors for all 188 monitoring cities and thus meteorological influences on $PM_{2.5}$ concentrations across China can be effectively compared without significant influences from inter-annual variations. c. A robust causality analysis method. Due to complicated interactions between different meteorological factors, correlations analysis, as introduced above, may lead to large bias in quantifying the meteorological influences on $PM_{2.5}$ concentrations. Similarly, the correlation coefficient between individual meteorological factors and $PM_{2.5}$ concentrations cannot be used as a reliable indicator to compare quantitative influences of individual meteorological factors on $PM_{2.5}$ across different cities. This research employed a robust CCM method, which removes the influence of other factors, and effectively quantified the coupling between $PM_{2.5}$ concentrations and a set of meteorological factors. The ρ value of each meteorological factor on $PM_{2.5}$ concentration can be compared between different cities. Based on national statistics across China, this research concluded that the influence of temperature, humidity and wind, especially temperature, on $PM_{2.5}$ concentrations was much larger than that of other meteorological factors, which could not be revealed by previous local and regional scale studies.

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The findings from this research were consistent with and a major extension of those from previous studies by quantifying the influence of individual meteorological factors in a large number of cities across China using a more robust causality analysis method. Similar to previous studies, this study also revealed notable differences in meteorological influences on $PM_{2.5}$ concentrations at the national scale, the major reason for which was different meteorological conditions and complicated mechanisms of $PM_{2.5}$-meteorology interactions. Firstly, notable differences existed in meteorological conditions across China. For instance, in winter, the frequency and intensity of precipitation are much higher and stronger in coastal areas than those in the North China region, where the frequency of strong winds is high in winter. Therefore, precipitation exerts a large influence on $PM_{2.5}$ concentrations in coastal regions whilst wind is the key influencing factor for $PM_{2.5}$ concentrations in the North China region in winter. Secondly, in addition to the large variations in the values of correlation coefficients, the interaction mechanisms between individual meteorological factors and $PM_{2.5}$ concentrations may also vary significantly across regions. For such meteorological influences as wind speed, its negative effect on $PM_{2.5}$ concentrations was consistent in China (He e al., 2017). On the other hand, He et al. (2017) suggested that temperature and humidity were either positively or negatively correlated with $PM_{2.5}$ concentrations in different regions of China. In terms of humidity, when the humidity is low, $PM_{2.5}$ concentration increases with the increase of humidity due to hygroscopic increase and accumulation of $PM_{2.5}$ (Fu et al., 2016). When the humidity continues to grow, the particles grow too heavy to stay in the air, leading to dry (particles drop to the ground) (Wang, J., & Ogawa, S. (2015)) and wet deposition (precipitation) (Li et al., 2015b), and the reduction of $PM_{2.5}$ concentrations. Similarly, there may be thresholds for the negative influences of precipitations on $PM_{2.5}$ concentrations (Luo et al., 2017). Heavy precipitation can have a strong washing-off effects on $PM_{2.5}$ concentrations and notably reduce PM2.5 concentrations. Meanwhile, slight precipitation may not effectively remove the high-concentration $PM_{2.5}$. Instead, the slight precipitation may induce enhanced relative humidity and thus lead to the increase of $PM_{2.5}$ concentrations. Meanwhile, the washing-off effect from the same amount of precipitation on $PM_{2.5}$ concentrations in Xi'an, a city with higher $PM_{2.5}$ concentrations, was lower than that in Guangzhou (Guo et al., 2016), indicating local $PM_{2.5}$ concentrations also exerted a key role in the negative effects of precipitation. Meanwhile，temperature can either be negatively correlated with $PM_{2.5}$ concentrations by accelerating the flow circulation and promoting the dispersion of $PM_{2.5}$ (Li et al., 2015b), or positively correlated with $PM_{2.5}$ concentrations through inversion events (Jian et al., 2012). Given the complexity of interactions between meteorological factors and $PM_{2.5}$, characteristics and variations of influences of individual meteorological factors on $PM_{2.5}$ concentrations should be further

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investigated for specific regions across China respectively based on long-term observation data.

~~With rapidly growing haze events, meteorological influences on PM$_{2.5}$ concentrations have become a hot social-economic topic not only studied by scholars, but also considered by government officials and decision makers. On December 1st, 2016, Beijing published the latest regulations for the prevention and control of meteorological hazards (http://www.bjrd.gov.cn/zt/cwhzt1431/hywj/201612/t20161201_168233.html) and included haze events as one type of meteorological hazards, sparking widespread controversy. Although the meteorological influences on PM$_{2.5}$ concentrations are well acknowledged, quantifying meteorological contribution, compared with exhaust emission, to airborne pollution remains challenging. Hence, criticisms have been raised that since traffic and industry induced exhaust emission is the main cause for airborne pollution, the emphasis on the meteorological causes for haze hazards is to avoid governmental responsibilities. Our previous research may provide reference for a better understanding of this issue from different perspectives. Chen, Z. et al. (2016) pointed out that more than 180 days in Beijing experienced notable and sudden air quality change (the Air quality Index, AQI, difference between one day and its previous day is larger than 50) in 2014. Considering that the industrial, automobile and household exhaust emission, which are main sources for PM$_{2.5}$ and other airborne pollutants, is unlikely to change dramatically in one day, meteorological factors seem to exert an important influence on local PM$_{2.5}$ concentrations. Chen, Z. et al. (2017) proved that such meteorological factors as *SSD*, *wind* and *humidity* exerted strong influences on winter PM$_{2.5}$ concentrations in the Beijing-Tianjin-Hebei Region. Furthermore, Chen, Z. et al. (2017) quantified the interactions between different meteorological factors and suggested that one meteorological factor may influence PM$_{2.5}$ concentrations through both direct and indirect means. Take winter PM$_{2.5}$ concentrations in Beijing for instance. The *wind* factor has a strong negative influence on PM$_{2.5}$ concentrations. In addition, the *wind* factor decreases *humidity*, as well as increases *SSD* and *evaporation*. Since the factor *humidity* (*SSD* and *evaporation*) has~~

~~a strong positive (negative) influence[6] on local PM$_{2.5}$ concentrations, increasing *wind* speeds can reduce PM$_{2.5}$ concentrations indirectly through reduced (increased) *humidity* (*SSD* and *evaporation*). In this research, we further revealed that meteorological influences on PM$_{2.5}$ concentrations varied significantly across China. In the most polluted winter, the dominant meteorological factors for PM$_{2.5}$ concentrations in the North China region are mainly the *wind* and *humidity* factor whilst the dominant meteorological factor on PM$_{2.5}$ concentrations in coastal cities are mainly *precipitation* and *temperature*. Furthermore, this research proved that the meteorological influences on PM$_{2.5}$ concentrations were the strongest in winter, when the PM$_{2.5}$ concentrations was the highest. With strong bidirectional coupling between individual meteorological factors and PM$_{2.5}$ concentrations in winter, PM$_{2.5}$ concentrations can be further enhanced through complicated atmospheric mechanisms, leading to more haze events. Based on these studies, we are not attempting to challenge the fundamental contribution of human-induced exhaust emission to PM$_{2.5}$ concentrations. Instead, our research suggested that with a stable amount of exhaust emission, meteorology was a key factor for the persistence and deterioration of haze events, especially in winter. On one hand, the pollutant emission should be strictly restricted, as human-induced emission is the major cause of haze pollution. Meanwhile, since meteorological factors play an important role in the accumulation and dispersion of PM$_{2.5}$, meteorological influences should be comprehensively considered for a better understanding and management of haze episodes.~~

~~In spite of a diversity of prediction models, air quality forecast, especially PM$_{2.5}$ forecasting in China, remains challenging.~~ Due to highly complicated atmospheric environment and the difficulty in acquiring true data of exhaust emission, commonly used models for air quality prediction(e.g. CAMx, CMAQ and WRFCHEM) may lead to large biases and uncertainty when applied to China. On the other hand, without prior knowledge of mechanisms of high PM$_{2.5}$ concentrations ~~haze formation~~ and information of exhaust emission, statistical models can achieve satisfactory forecasting results based on massive historical data (Cheng et al., 2015). Compared with the static models, dynamic statistical models additionally consider the

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[6] ~~Although the CCM method did not give a positive(negative) direction of interactions between two variables, the direction of interactions can be easily understood according to the correlation coefficient (Chen et al., 2017)~~

meteorological influences on $PM_{2.5}$ concentrations and some meteorological factors that are of stable, representative and strong correlations with $PM_{2.5}$ are selected for forecasting $PM_{2.5}$ concentrations. Meanwhile, many recent studies (Cheng et al., 2017; Guo et al., 2017; Lu et al., 2017; Ni et al. 2017; etc) have recognized the meteorological influences on the evolution of $PM_{2.5}$ concentrations and included some key meteorological factors in their models for $PM_{2.5}$ estimation. However, most $PM_{2.5}$ estimation and forecasting models mainly employed correlation analysis to reveal the influence of individual meteorological factors on $PM_{2.5}$ concentrations. Due to complicated interactions in atmospheric environment, the correlation coefficient between meteorological factors and $PM_{2.5}$ concentrations is usually much larger than the ρ value and overestimates the influence of individual meteorological factors on $PM_{2.5}$ concentrations. In this case, this research provides useful reference for improving existing statistical models. By incorporating the ρ value, instead of the correlation coefficient, of different factors into corresponding GAM (Generalized Additive Models) and adjusting parameters accordingly, we may significantly improve the reliability of future estimation and forecasting of $PM_{2.5}$ concentrations.

~~With the understanding of strong meteorological influences on $PM_{2.5}$ concentrations across China, especially in some heavily polluted regions, decision makers are placing special emphasis on improving local and regional air quality through meteorological means. Targeting this, q~~Quantified causality of individual meteorological factors on $PM_{2.5}$ concentrations provides useful decision support for evaluating relevant environmental projects, which aim to improve local and regional air quality through meteorological means~~.~~ Specifically, a forthcoming Beijing wind-corridor project (http://www.bj.xinhuanet.com/bjyw/yqphb/2016-05/16/c_1118870801.htm) has become a hot social and scientific issue~~, yet its potential effects arouse wide controversies~~. ~~Some scholars (http://china.cnr.cn/yxw/201411/t20141123_516839830.shtml http://health.people.com.cn/n1/2016/0413/c398004-28271979.html) pointed out that the wind corridor project could only exerted limited influence on the reduction of $PM_{2.5}$ concentrations and major efforts should be made on emission reduction.~~ Herein, our research suggests that wind is a dominant meteorological factor for winter $PM_{2.5}$ concentrations in Beijing and can significantly influence $PM_{2.5}$ concentrations

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through direct and indirect mechanisms(Chen,Z. et al., 2017). In consequence, the wind-corridor project may directly allow in more strong wind, which thus leads to a larger value of SSD and EVP and a smaller value of RHU. The change of SSD, RHU and EVP values can further induce the reduction of $PM_{2.5}$ concentrations. From this perspective, the Beijing wind-corridor project has good potential to improve local and regional air quality. In addition to the wind-corridor project, some scholars and decision makers have proposed other meteorological means for reducing $PM_{2.5}$ concentrations. For instance, Yu (2014) suggested that water spraying from high buildings and water towers in urban areas was an efficient way to reduce $PM_{2.5}$ concentrations rapidly by simulating the process of precipitation. However, some limitations, such as the humidity control and potential icing risk, remained. In the near future, with growing attention on the improvement of air quality, more environmental projects should be properly designed and implemented. According to this research, meteorological influences on $PM_{2.5}$ concentrations vary notably across China. Given the diversity of dominant meteorological factors on local $PM_{2.5}$ concentrations in different regions and seasons~~, which has been proved by previous studies and this research~~, it is more efficient to design meteorological means accordingly. For the heavily polluted North China region, especially the Beijing-Tianjin-Hebei region, the northwesterly synoptic wind（Tie et al., 2015; Miao et al., 2015）is much stronger in winter than winds in summer and exerts a dominant influence on $PM_{2.5}$ concentrations (Chen et al., 2017). Furthermore, in North China, the $PM_{2.5}$ concentration is much more sensitive to the change of wind speed than that of other meteorological factors (Gao et al., 2016). Meanwhile, wind-speed induced climate change led to the change of $PM_{2.5}$ concentrations by as much as 12.0 μgm^{-3}, compared with the change of $PM_{2.5}$ concentrations by up to 4.0 μgm^{-3} in south-eastern, northwestern and south-western China (Tai et al., 2010). Considering the strong winds in winter, the dominant influence of wind speed on $PM_{2.5}$ concentrations and the sensitivity of $PM_{2.5}$ feedbacks to the change of wind speed, meteorological means for encouraging strong winds are more likely to reduce $PM_{2.5}$ concentrations considerably in North China. Similarly, Luo et al. (2017) suggested that only precipitation with a certain magnitude can lead to the washing-off effect of $PM_{2.5}$ concentrations whilst Guo et al. (2016) revealed that the variation of $PM_{2.5}$ concentrations was more sensitive to the same amount of precipitation in areas with lower $PM_{2.5}$ concentrations. Therefore,

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meteorological means for inducing precipitation are more likely to improve air quality in coastal cities and cities within the Yangtze River basin, where there is a large amount of precipitation and relatively low $PM_{2.5}$ concentrations.

6 Conclusions

Previous studies examined the correlation between individual meteorological influences and $PM_{2.5}$ concentrations in some specific cities and the comparison between these studies indicated that meteorological influences on $PM_{2.5}$ concentrations varied significantly across cities and seasons. However, these scattered studies conducted at the local scale cannot reveal regional patterns of meteorological influences on $PM_{2.5}$ concentrations. Furthermore, previous studies generally selected different research periods and meteorological factors, making the comparison of findings from different studies less robust. Thirdly, these studies employed the correlation analysis, which may be biased significantly due to the complicated interactions between individual meteorological factors. This research is a major extension of previous studies. Based on a robust causality analysis method CCM, we quantified and compared the influence of eight meteorological factors on local $PM_{2.5}$ concentrations for 188 monitoring cities across China using $PM_{2.5}$ and meteorological observation data from 2014.3 to 2017.2. Similar to previous studies conducted at the local scale, this research further ~~proved~~ indicated that meteorological influences on $PM_{2.5}$ concentrations were of notable seasonal and spatial variations at the national scale. Furthermore, this research revealed some regional patterns and comprehensive statistics of the influence of individual meteorological factors on $PM_{2.5}$ concentrations, which cannot be understood through small-scale case studies. For the heavily polluted North China region, the higher $PM_{2.5}$ concentrations, the stronger influence meteorological factors exert on local $PM_{2.5}$ concentrations. The dominant meteorological factor for $PM_{2.5}$ concentrations is closely related to geographical conditions. For heavily polluted winter, precipitation exerts a key influence on local $PM_{2.5}$ concentrations in most coastal areas and the Yangtze River basin, whilst the dominant meteorological driver for $PM_{2.5}$ concentrations is wind in the North China regions. At the national scale, the influence of temperature, humidity and wind on local $PM_{2.5}$ concentrations is much larger than that of other factors, and *temperature* exerts the strongest and most stable influences on national $PM_{2.5}$

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concentrations in all seasons. The influence of individual meteorological factors on $PM_{2.5}$ concentrations extracted in this research provides more reliable reference for better modelling and forecasting local and regional $PM_{2.5}$ concentrations. Given the significant variations of meteorological influences on $PM_{2.5}$ concentrations across China, environmental projects aiming for improving local air quality should be designed and implemented accordingly.

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