Dear Dr Sally E. Pusede:

Thanks so much for giving us a chance to revise and submit our manuscript for the potential publication in ACP. According to the reviewers' comments, we managed to collect additional two years' PM_{2.5} and meteorological data and conduct a multiple year analysis using the CCM method. By comparing the one year's result in the previous manuscript with the multiple-year analysis result in the revised version, we found that there is no large differences at both regional and national scale, especially for the heavily polluted North China region. At the national scale, the dominant meteorological factors remain temperature, wind and humidity. The similar analysis results acquired using one year and multiple-year analysis indicated that meteorological influences on PM_{2.5} concentrations across China are generally stable at the inter-annual scale. In addition to the requirement for multiple-year analysis, we also added many details to the revised manuscript and the response file to explain the mechanisms of CCM. In this case, the advantage of CCM compared with the correlation analysis can be clearly demonstrated.

Reviewers suggested that the PM_{2.5}-meteorology relationship is well-known to the chemistry society and thus we deleted this part of discussion. Instead, according to their suggestions, we added more in-depth discussion concerning the potential reasons for the large variations of meteorological influences on PM_{2.5} concentrations across China and potential applications of employing the p value to better estimate and predict PM_{2.5} concentrations.

We also addressed all the cartography and technical issues raised by reviewers.

Thanks again for considering our manuscript. We are willing to conduct any further revisions according to other requirements from you and the reviewers.

The very best

Ziyue Chen

acp-2017-376-RC1 Anonymous Referee #3

R: Dear Referee, thanks so much for your comments, which helped us improve this manuscript a lot. We have fully revised this manuscript according to your general and detailed comments.

Yes, as you pointed out, the principle of CCM and its advantages compared with the correlation analysis are not well-known to all scholars and thus a better explanation of the CCM method would be needed. Due to limited space, we did not add all details to the modified manuscript and some responses and explanations are given here according to your specific comments. Meanwhile, some other issues were also addressed. As your suggested that multiple year data are required, we have extended the research period from one year to three years using latest published data. Other issues are responded as follows. Thanks again for all your valuable comments and we are willing to conduct further revisions if you have further requirements.

General Comments This is an interesting paper that applies an exciting and fairly new statistical method (convergent cross mapping) to quantify the relationship between local air quality and local meteorology (wind, temperature, precipitation, etc.). The authors argue that unlike a simple correlation analysis, this method is able to demonstrate causal relationships between variables. My understanding is that this method is quite new, and it is central to this study, so I think this paper would benefit from a clearer discussion of why it is better than correlation at determining causal relationships. Would the authors' findings have differed significantly if they used correlation instead of CCM?

R: The CCM method was proposed by Sugihara et al. (2012).

1 Sugihara, G., May, R., Ye, H., Hsieh, C., Deyle, E., Fogarty, M., Munch, S. 2012. Detecting Causality in Complex Ecosystems. Science, 338, 496-500. Sugibara et al. (2012) pointed out that correlation analysis could extract mirage correlations, especially in complicated ecosystems. For instance, two variables A and B that have no causality may demonstrate significant correlations due to the existence of an agent variable C, which interacts with both A and B. Through a series of experiments, Sugihara et al. (2012) proved that this type of mirage correlations could be detected using the CCM method by calculating a p value of 0. The CCM method not only performed better than the correlation analysis in causality analysis by excluding the influence of other variables, but also demonstrated the advantage of detecting weak causality compared with other causality analysis method (e.g. Granger Causality), which may fail to detect weak to moderate coupling between variables.

In our previous studies, we employed both the correlation and the CCM method to examine the influence of individual meteorological factors on PM_{2.5} concentrations in the Beijing-Tianjin-Hebei region and compared the performance of correlation and CCM method.

Chen, Z., Cai, J., Gao, B.B., Xu, B., Dai, S., He, B., Xie, X.M. Detecting the causality influence of individual meteorological factors on local PM2.5 concentration in the Jing-Jin-Ji region. Scientific Reports 2017. 7:407352

The comparison suggests that the causality influence of individual meteorological factors on PM2.5 concentration is better revealed using the CCM method than the correlation analysis. By comparing the correlation coefficient and p value in Table 2, one can see that some correlations between meteorological factors and PM2.5 concentration may result from mirage correlations (e.g. the correlation between meanRHU and PM2.5 concentration in Hengshui in summer). Secondly, CCM analysis reveals weak or moderate coupling (e.g. the interactions between SSD and PM2.5 concentration in Cangzhou in summer) whilst correlation analysis cannot. Additionally, due to interactions between different meteorological factors, the value of correlation coefficients cannot interpret the quantitative influence of individual meteorological factors on PM2.5 concentration. Instead, the ρ value from CCM method is designed to understand the coupling between two variables by excluding influences from other factors. Through comparison, the value of the correlation coefficient for some meteorological factors is notably different from the ρ value for these meteorological factors. A large correlation coefficient for one meteorological factor may correspond to a much smaller ρ value from the CCM analysis, indicating that the value of the correlation coefficient usually overestimates the influence of individual meteorological factors on PM_{2.5} concentration.

The previous research (Chen et al., 2017) proved that the CCM method outperform the correlation analysis in many aspects. And this research extended the study area from the Beijing-Tianjin-Hebei region to a national scale, so the CCM method, instead of the correlation analysis, remains the ideal tool for quantifying meteorological influences on PM_{2.5} concentration over China.

There are some aspects of CCM that were not clear to me. Does CCM account for relationships between meteorological factors? E.g. if wind is affecting both precipitation and PM, how is the affect of wind on precipion PM counted?

R: Yes, you made a very good point here. The exclusion of influences from other variables and solely focus on the interactions between two target variables are the most important advantages of the CCM. Sugihara et al. (2012) proposed the CCM method and examined its performance in removing the influence of agent variables through a diversity of experiments. The result proved that mirage correlations caused by the influence of other variables could be detected and removed by the CCM method. For instance, Sugihara (2012) suggested that the CCM method could quantify the bi-directional interactions between two individual variables without being affected other variables, which were also proved by a diversity of studies. So for your question, for each calculation, the CCM method simply examine the bi-directional coupling between two selected variables in complicated ecosystem whilst the influence of other factors was excluded. For your 4 instance, we could calculated the coupling between PM and wind speed, and the coupling between precipitation and wind speed using the CCM method respectively, and the results for both calculations would not influence each other.

Another question I have is what produces a large value. For example, are the values higher in winter simply because there is more PM available to be effected?

R: Generally, the CCM method simply calculates the quantitative influences of individual meteorological factors on PM concentrations, whilst the mechanisms were not revealed. The mechanisms how even one individual meteorological factor influences local PM concentrations can be highly complicated. The chemical compositions, size and mass concentrations of particulate matters actually vary significantly across locations and seasons. There is no comprehensive research on how one individual meteorological factor influence PM2.5 concentrations through different mechanisms. The large ρ value in winter may result from both the much higher PM2.5 concentrations, which may be easily influenced by meteorological factors, and unique meteorological conditions. For instance, PM_{2.5} induced haze weather occurred frequently in winter in the Beijing, indicating a much higher PM_{2.5} concentration. Meanwhile, some meteorological factors in Beijing in winter were quite different from those in other seasons. For instance, the northwest wind prevails in winter and it has become a common scene and a popular saying that "the best solution for haze in Beijing is to wait for the wind". Thus the p value for wind speed in many cities within the Beijing-Tianjin-Hebei region was much larger in winter than in other seasons for both PM2.5 concentrations due to both high PM concentrations and strong wind speeds. Another instance is that the chemical composition of PM, which is related to photochemical reaction and solar radiations, also varied significantly in different seasons. Hence, the reason for the variation of p value across seasons is highly complicated, and the high PM2.5 concentration is one of major reasons. Thanks so much for pointing this out. I believe systematic research on the influencing mechanism of individual meteorological factors should be examined in-depth by scholars from a diversity of backgrounds.

Or as another example, is precipitation more effective at removing PM along the coasts because it rains more? If there were a way to normalize by the amount of total rainfall, would precip still be more important along the coasts than in the drier interior?

R: Yes, it is highly possible that precipitation is more effective at removing PM along the coasts because it rains at a higher frequency and intensity. As we know that the PM_{2.5} concentrations drop significantly after a heavy rain whilst light rain may not reduce PM_{2.5} concentrations significantly. Meanwhile, PM_{2.5} concentrations may also affect the influence of precipitation. Light rains may have limited washing-off effects on high-concentration PM_{2.5} concentrations and may increase the relative humidity in the environment, which is favorable for the rising of PM_{2.5} concentrations. In the drier interior, the PM_{2.5} concentrations are usually much higher and the intensity and frequency of precipitation are much lower than those along coasts. These two factors may both be the reason that precipitation is more effective at removing PM along the coasts. Due to these influencing factors, precipitation may still be a less important meteorological driving force for PM_{2.5} concentrations in the drier interior, even if there were a way to normalize by the amount of total rainfall. In the revised manuscript, we added a new part to briefly introduce the underlying reasons for the variation of

My second major concern is that this study uses a single year of data to make general comments about PM-meteorology relationships. This gives us little sense of how much year-to-year variability may exist in these relationships and generally weakens the conclusions. If CCM is too computationally expensive to use on multiple years, perhaps a different method could be used to supplement it. R: This is a very good point. When we were preparing this manuscript, there were only one-year's data available. And now, we endeavored to acquire another two years' data for multiple year analysis. Although the CCM is computationally expensive, we still believe it is more persuasive to use multiple years' data for drawing conclusions. So thanks so much for this suggestions. We have re-calculated the CCM results using three years' data, and the updated results have been added to the revised manuscript. By comparing the mean annual p value for eight variables for 2014 with the mean annual p value for eight variables for 2014 with the mean annual p value for eight variables during 2014-2016, we can see that the meteorological influences on PM_{2.5} concentrations at the national scale did not change significantly in the past three years.

Finally, I would like to see a deeper discussion of the scientific significance of this work. As it is written currently, this paper is almost purely descriptive. The new method is interesting, but the paper could do a better job of articulating what we are learning from it. Perhaps some discussion about why different meteorological factors are more/less important in different regions/seasons, for example, would help give the paper more depth. I would also suggest spending more time discussing the implications for modeling, especially in the introduction and conclusions, as that was what I took away as the most important implication in this paper.

R: Thanks so much for this valuable suggestion. Yes, it is important to provide more indepth discussion for the acquired results based on the CCM. And as you suggested, some discussion, e.g. the mechanisms between meteorology factors and PM_{2.5} concentrations were well-known, and it is not necessary to introduce it in details. So in the revised manuscript, we have fully removed this part and left more space for the parts you suggested. We added some introduction on the potential applications the CCM method, instead of the correlation analysis, in research concerning meteorology-included PM_{2.5} concentration estimation prediction. Meanwhile, the potential reason for large variations of meteorological influences on PM_{2.5} concentrations across China has been added to the revised version. Thanks again for this valuable comment.

Specific Comments

pp 3, ln 93-95 - Can you elaborate further on how your previous study showed that CCM is better than correlation for the benefit of readers who have not read that paper. It is important for your results here to make as clear as possible why CCM is a better method/provides new insights.

R: This is a very good point. We have mentioned the advantages of the CCM method, compared with the correlation analysis in the method parts. However, as you pointed out, we should give more information on how the CCM method performed better than the correlation analysis. So we added some extra details concerning the CCM method in the method part (since Line 197).

As mentioned above, our previous study (Chen et al., 2017) employed the correlation analysis and the CCM method to examine the influence of individual meteorological factors on PM_{2.5} concentrations in the Beijing-Tianjin-Hebei region. In the paper, we demonstrated detailed results of both correlation and causality influence in two large tables. By comparing these results, we found that (1) some correlations between meteorological factors and PM2.5 concentration may result from mirage correlations. For instance, there was a correlation between meanRHU and PM2.5 concentration in Hengshui in summer whilst the causality influence of meanRHU on PM_{2.5} concentrations is 0, indicating a mirage correlation.

(2) CCM analysis reveals weak or moderate coupling whilst correlation analysis cannot. For instance, the correlation between SSD and PM2.5 concentration in Cangzhou in summer was not significant whilst there is a weak causality influence of SSD on PM_{2.5} concentrations detected. (3) Due to interactions between different meteorological factors, the value of correlation coefficients cannot interpret the quantitative influence of individual meteorological factors on PM2.5 concentration. Instead, the ρ value from CCM method is designed to understand the coupling between two variables by excluding influences from other factors. Through comparison, the value of the correlation coefficient for some meteorological factors is notably different from the ρ value for these meteorological factors. A large correlation coefficient for one meteorological factor may correspond to a much smaller ρ value from the CCM analysis. We found that the correlation coefficient between individual meteorological factors and PM2.5 concentrations was usually much larger than the value. This indicated that the causality of individual meteorological factors on PM2.5 concentrations was generally overestimated using the correlation analysis, due to the influences from other meteorological factors.

So the ρ value is a more reliable indicator for understanding and comparing quantitative influences of different individual meteorological factors than correlation coefficient.

Due to limited spaces, in the previous manuscript, we did not give all information on the comparison between correlation analysis and the CCM method. But according to your suggestion, more details have been added to the revised manuscript

pp 8 ln 221 - Are the results sensitive to the choice of parameters?

R: The CCM detects the bi-directional coupling between two variables highly automatically and only several parameters are required to run this model. However, the CCM result mainly depends on the time series data of the two variables and the several parameters mainly influence the presentation effects of the Convergent maps. We have tested different setting of these parameters, and the extracted ρ value was simply the same, only the presentation of convergent maps were more smooth with different settings. So the results were not sensitive to the choice of parameters, which is also the major advantage of the CCM method. Since CCM method is not sensitive to the choice of parameters and the reliability of the causality analysis result has been proved by hundreds of studies from different ecosystems, this method has been widely employed.

pp 9 ln 237 – how is the value of determined? Are you calculating the limit or taking the value at a specific time series length?

R: This is a good question. When we determine whether a curve is convergent or not, we set a Δ to represent the variation of ρ value along with increasing time series length. If the Δ was less than a given threshold (e.g. 0.01) for a consecutive several date until the end of the time series, we consider that the ρ was convergent to the p value of last date.

What about cases such as PM2.5 xmap minTEM, which (at least by eye) does not appear to be converging? Wouldn't that suggest that minTEM was not influencing PM2.5 at all?

R: The CCM method considers strict causality influence of one variable on the other one and if the curve is convergent to 0 or demonstrates no generally convergent trend, then no causality influence exists based on long-term time series analysis of two variables. And for your instance, PM2.5 xmap minTEM demonstrate a generally convergent trend to a value approximating to 0.2. You may argue that this curve is not strictly convergent. However, the CCM method does not actually conducts a limit calculation and the CCM curve simply demonstrates a general trend of convergence. According to a diversity of instances provided by Sugihara et al. (2012), the curve shape of PM2.5 xmap minTEM can be regarded as a convergent curve, indicating a detected causality. Those instances from Sugihara et al. (2012) that demonstrated a non-convergent trend are near-linear shape or totally irregular shapes, which are quite different from the PM2.5 xmap minTEM. In summary, according to the PM2.5 xmap minTEM, minTEM exerted a weak influences on PM_{2.5} concentrations.

pp 9 ln 244 - It's not clear here how PM is changing wind speed

R: High PM_{2.5} concentrations may lead to haze episodes, which usually result in generally stable atmospheric environment. In this case, the formation of winds, especially strong 10

winds within this atmospheric environment are influenced significantly. And this is the reason why there are few winds within the urban areas during severe haze episodes. Meanwhile, winds across regions with haze episodes are also influenced notably.

Yang et al. (2015) observed four haze episodes during Oct to Nov, 2014 and during these four haze episodes in the North China plain, the very high PM2.5 concentrations all led to stagnant condition and weak high-pressure systems, which further led to slowed wind speed and disturbed wind direction. This phenomenon was also observed by Liu et al. (2014) in haze episodes in Beijing in 2013. Very high PM2.5 concentrations induced haze episodes further led to stagnant and stable high-pressure systems, which made megacities served as obstacles to significantly slowed down the wind speed (Yang et al., 2015). Therefore, the effects of aerosols, especially high-concentration PM2.5 concentrations, prevented the wind occurrence mainly through indirect mechanisms.

Yang, Y. R., Liu, X. G., Qu, Y., An, J. L., Jiang, R., & Zhang, Y. H., et al. (2015). Characteristics and formation mechanism of continuous hazes in china: a case study during the autumn of 2014 in the north china plain. Atmospheric Chemistry & Physics, 15(14), 10987-11029.

Liu, X. G., Li, J., Qu, Y., Han, T., Hou, L., & Gu, J., et al. (2013). Formation and evolution mechanism of regional haze: a case study in the megacity beijing, china. Atmospheric Chemistry & Physics, 13(9), 4501-4514.

pp 10 ln 270 - I would guess that (out of the 189 cities) those clustered regionally would show similar maps. Is that true? I.e. are the 37 cities shown representative of the cities not shown?

R: The main reason we selected some representative cities, instead of all cities for presenting wind rose maps is that there is limited space in the map. As one can see, it is already very crowded to demonstrate 37 wind roses and we should pay special attention to the selection of representative cities. Since the provincial capital in each province is the most important city(and usually the largest city) within the province and thus the meteorological influences on $PM_{2.5}$ concentrations for the provincial capital city usually receive the most emphasis. In this case, we selected the provincial capital for 31 provinces to present meteorological influences on $PM_{2.5}$ concentration in each province, which considers both the spatial locations and the importance of the representative cities. For most provinces, especially provinces with low $PM_{2.5}$ concentrations, the number of monitoring cities and the variations of $PM_{2.5}$ concentrations are small. For regions with heavy air pollution (e,g, , the number of monitoring cities and the variations of $PM_{2.5}$ concentrations are small. In that case, we believe that the selective cities can be representative of no-shown cities within the same province.

pp 13 ln 292-296 - This only seems to be true in some seasons.

R: Thanks so much for this point. We also noticed that this part should be described more rigorously. So in the revised manuscript, we have fully revised this part.

pp 13 ln 299-300 - Can you quantify this more rigorously? By eye, it seems like there are enough outliers to call this in to question

R: Thanks for pointing this out. We realized that this conclusion may not apply to all regions in China. Instead, this pattern " the higher PM_{2.5} concentrations, the stronger meteorological influences on PM_{2.5} concentrations" was most obvious for the North China region, which is the most polluted region in China. In the revised manuscript, we have rephrased this statement to "For the North China region". Thanks again for this suggestions.

pp 14 ln 337-339 - This seems true for winter vs. summer, but what about spring vs autumn?

R: Again, thanks for pointing this out. Generally, $PM_{2.5}$ concentrations for one specific city are highest in winter and the lowest in summer. So in the paper, when we mention the season when $PM_{2.5}$ concentration is high (low), we mainly mean winter (summer). Meanwhile, the characteristics of $PM_{2.5}$ concentrations in spring and autumn are not obvious, so we mainly compare the characteristics of $PM_{2.5}$ concentrations in winter and summer, which provides the most important reference for the management of $PM_{2.5}$ induced haze episodes in winter. So thanks again for this comments and we realized that we should make the statement clearer to avoid some confusion. In the revised manuscript, this sentence has been rephrased.

pp 18 Fig 4 caption – Is there a particular argument for only including the dominant factor in each city?

R: Thanks for this comment. As explained in the text, there are more than 189 cities for this research and it requires some space in the map to place the wind rose map for each city without overlapping with each other. And as you can see, the wind rose map for 37 representative cities has already been filled with 37 wind roses with different sizes and it is impossible to present 189 wind roses on a single map without severe overlapping effects. So we employed an alternative approach to only present the dominant meteorological factor for all cities and an entire wind rose for 37 representative cities in separate maps.

Section 5.1 of the discussion feels out of place. This is more of a discussion of what we already know about aerosol-meteorology interactions than a discussion of the implications of the work done in this study. I would recommend either rewriting it so that it builds more on the results from this paper, or cut it and integrate the important information into earlier sections.

R: Thanks so much for your comments. As also suggested by another referee, this part has been well known to scholars with relevant background. So in the revised manuscript,

this part has been totally cut. Meanwhile, we have added a separate paragraph to explain the underlying reasons for large variations of meteorological influences

pp 24 ln 562-566 – as per my earlier comment, I am not sure that you have shown this. If Beijing were to receive the same amount of precipitation that a coastal city does, is it possible that precipitation would become more important in Beijing? Does looking at how a specific factor changed PM in a year tell us about how effective changes in that factor would be?

R: Thanks for pointing this out. I thought it is a bit difficult to test the hypothesis using the CCM method. This is because the precipitation amount for most days in Beijing is 0 and thus it is difficult to normalize the 0 value to a value similar to that in coastal cities. As an alternative solution, as responded above, we added another two years data to conduct a multiple-year analysis and checked whether the influence of precipitation on PM_{2.5} concentrations in Beijing vary with longer time series.

Technical Corrections pp 1, ln 17 (and later occurrences) - "causality influence" is redundant. "Influence" already implies causality.

R: All Corrected.

pp 2, ln 35-36 - "Amongst these environmental elements, . . . concerned social and ecological issues." The wording of this sentence is unclear.

R: This sentence has been rephrased.

pp 2, ln 42 - "Serious haze not only influences peoples daily life," this wording is vague. How does haze influence peoples daily lives?

R: Serious haze episodes, usually presented as very thick and heavy black fogs, cause serious negative influences on people's daily life, especially the traffic. During a severe 14

haze episode in Beijing in January, the extreme high fog episodes led to very low visibility and heavy traffic jam. This is one instance how haze influence people's daily life.

pp 2, ln 57 - "controversy" should be changed to "controversial"

R: Corrected.

pp 3, ln 68 - "... fractions of three different sizes. .." this is unclear. Authors should indicate that they are talking about aerosols, and specify the sizes.

R: Thanks for pointing this out. We have corrected this in the revised manuscript.

pp 3, ln 7 - what region is this study referring to?

R: The missing "research region" content has been added to the revised manuscript.

pp 3, ln 86 - word choice: I would suggest "well studied" instead of "massively studied"

R: Corrected.

pp 4, ln 119 - what does API stand for?

R: API (Application Programming Interface).

pp 5, ln 136-137 - I'm not clear on what small and large evaporation are.

R: Small evaporation indicates the evaporation amount calculated using a small-diameter measurement equipment and large evaporation indicates the evaporation amount calculated using a large-diameter measurement equipment. Generally, the amount calculated using the two types of equipment is usually the same, although slightly difference may exist between measured evaporation values. And the p value for the factor evaporation is decided by the larger value of the two indicators.

pp 5, ln 144 – sunshine duration for the day is a less widely used term and should be defined here

R: Thanks so much for pointing this out. Sunshine duration or sunshine hours is a climatological indicator, measuring duration of sunshine in given period (usually, a day or a year) for a given location on Earth. This definition has been added to the modified manuscript.

pp 5 ln 146 - what qualifies as extreme wind speed?

R: 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.

Thanks for pointing this out and the definition has been added to the modified manuscript.

pp 5 ln 146 - how is max wind direction defined?

R: The max wind direction indicates the dominant wind direction for the period with the max wind speed.

pp 6 ln 174 – "Two time series" is unnecessary. Suggest changing the sentence to " $\{X\}=[X(1), \ldots X(L)]$ and $\{Y\}=[Y(1), \ldots Y(L)]$ are defined as the temporal variations of variables X and Y."

R: Corrected according to your suggestions.

pp 6 ln 175 – It's unclear what r and S are.

R: Thanks for pointing this out.

r is the current position in the time series and the S presents the start position in the time series.

pp 8 ln 217 – why can E be 2 or 3?

R: This parameter decides at what dimension the CCM was calculated. When E equals 3, the calculation accuracy is higher. Through experiments, we found that the results were generally

the same using the value of 2 or 3. For this research, we set the value of E 3 for a theoretically optimal CCM result.

pp 14 ln 331-332 - the phrasing here is unclear.

R: This sentence has been rephrased.

pp 18 ln 374-376 - is there a way to test if these values are significant?

R: The CCM method is different from the correlation analysis and another classic causality analysis method , Granger causality, which provides readers with the significance for the coupling between two variables. The CCM did not give us a value to present the significance for the revealed causality. However, while the Granger causality mainly revealed the qualitative causality between two variables, the p value from the CCM method revealed the quantitative causality between them. And the CCM method suggests that if the significance between two variables was not significant, then the calculated p value would be 0. So the p value was a direct metric for the quantitative influence and an indirect metric for the significance.

pp 20 ln 419 - this wording is unclear

R: This part has been removed in the revised manuscript.

pp 20 ln 426 – Wikipedia is not an appropriate source. Better to cite a scientific paper that defines SSD.

R: In the revised manuscript, the PM_{2.5}-meteorology interaction part has been removed according to your and other referees' comments. However, here we would like specifically add the explanation here that SSD could reduce PM_{2.5} concentrations not only through

atmospheric photolysis, but also by enhancing surface temperature and promoting upward movement of aerosols.

pp 20 ln 440 – Temperature inversion is certainly important, but none of the metrics in this study measure it directly.

R: Thanks for pointing this out. Although this part has been removed in the revised manuscript, we would like to add some other mechanisms. Actually, in addition to temperature inversion, another important mechanism is that Temperature is closely related to pollutant concentrations by affecting atmospheric turbulence and chemical reactions. The temperature is positively correlated with pollutant concentrations in the majority of cities (He et al., 2017).

He, J., Gong, S., Ye, Y., Yu, L., Lin, W., Mao, H., et al. (2017). Air pollution characteristics and their relation to meteorological conditions during 2014–2015 in major chinese cities. Environmental Pollution, 223, 484-496.

pp 21 ln 447 - what about horizontal transport (advection)?

R: Yes, horizontal transport of airborne pollutants should be major reason for the variation of PM_{2.5} concentrations. i.e. Anticyclones (i.e., high pressure systems) induced low wind speed was not favorable for the dispersion of pollutants. On the other hand, low pressure systems may lead to large wind speeds, favorable for the dispersion of PM_{2.5}.

pp 21 ln 446 – change "social economic" to "socio-economic" pp 22 ln 492 – change "negative causality on" to "decreases" and "positive causality on" to "increases"

R: Corrected.

pp 23 ln 527-528 - do you have citations for this?

R: We have added more relevant references to the revised manuscript according to your

comments.

pp 24 ln 544 – can you give more details/citations about the controversy?

R: Some reports concerning different effects of this project has been added to the revised manuscript.

acp-2017-376-RC2 Anonymous Referee #1

Interactive comment on "Understanding meteorological influences on PM2.5 concentrations across China: a temporal and spatial perspective" by Ziyue Chen et al. Anonymous Referee #1 Received and published: 21 August 2017 Chen et al. present an interesting analysis of the spatial and temporal variation of the relationships of meteorology and PM2.5 over most of China. The cross convergent mapping analysis provides a unique method of understanding the causality of the relationships, which might otherwise be missed with typical correlation analysis. They highlight that the meteorological influence on PM2.5 varies widely by location and season, and that attempts to engineer favorable air quality meteorology should take these differences into account. The paper is well-written and relatively thorough, however it requires some additional explanations and detail. Thus I recommend publication following minor revision.

Dear referee:

Thanks so much for your encouragement and valuable comments. We have fully revised this manuscript according to your general and detailed comments, as well as comments from other reviewers. We would like to make further revision in due stages if you have further requirements.

Page 2, lines 56-57: "Although quantitative contributions of different sources (e.g. coal burning and automobile exhaust) to airborne pollutants remain controversy" – It's not clear what you mean here with the "controversial" – politically or scientifically? If scientifically – the direct emissions and/or subsequent chemistry?

R: Yes, the controversy mainly comes from the mixed understanding of relative contribution. For instance, some scholars claimed that automobile exhaust took up only 4% of relative contributions to PM_{2.5} concentrations. However, many following papers

argued that the actual contribution from automobile exhaust took up more than 20%. The difference was that the former mainly considered the direct emissions whilst the latter ones comprehensively considered the direct emission and following secondary pollutants. So yes, your point was exactly the situation.

Page 4, section 2.1.1: How do you quality control the data and/or deal with missing data?

R: For this research, all released data were previously maintained by specific institutions and there are several stations for each city to report hourly PM_{2.5} concentrations conditions. For some stations, missing data lead to 0 value. If there are stations with Non-0 value, then the mean PM_{2.5} concentration for a specific city was calculated using these stations. So for most days in each city, a valid mean PM_{2.5} concentration value could be calculated. For days when the measured PM_{2.5} concentration from all stations was 0, then mean PM_{2.5} concentration was 0. The record for this day was deleted. Only a very small proportion of cities experienced days with no daily average data. And since few missing records would not influence the order of time series of PM_{2.5} and meteorological data, the CCM result would not be influenced by the missing data. Meanwhile, for cities (e.g. Liaocheng and Zhuji) with a large amount of missing meteorological data, we deleted this city for this research.

CCM method: How does the time lag parameter affect the results? The resolution of the map is mentioned but how does it affect the physical interpretation of the results? – Especially for those variables that may act on a shorter time scale.

R: We compared the CCM analysis result calculated using different parameters: 2,5, 10, 20 and the result was generally the same. Just the resolution of the map was higher with a small time lag. And you made a good point here that the physical interpretation of the results may lead to biased p value. Actually, the presented CCM map was simply for a 21

basic demonstration about how CCM works. For exact p value, the provided CCM algorithm actually calculated an accurate p value with the increase of time series, and the CCM map was produced based on a series of accurate p values. So for this research, it is not feasible and reliable to physically interpret the p value for 190*18*4 CCM maps and the p value used for producing the wind-rose and other maps were extracted directly from the program.

Page 13, lines 295-296: This causality seems to be backwards: i.e., why would differences in PM levels cause differences in meteorological influences? What mechanism would cause this?

R: This is a very good question. Actually, this phenomenon was revealed and proposed in Chen et al. (2017). Chen et al. (2017) found that in the Beijing-Tianjin-Hebei region, the causality influence of individual meteorological factors on PM_{2.5} concentrations was the strongest in winter, when the PM_{2.5} concentrations were the highest, for all cities, Meanwhile, the causality influence of individual meteorological factors on PM_{2.5} concentrations was the weakest in winter, when the PM_{2.5} concentrations were the lowest. The potential mechanism could be that similar meteorological conditions may lead to large variations of PM_{2.5} concentrations were high and may lead to haze episodes, a strong northwester wind may immediately reduce the PM_{2.5} concentrations to a very low level. Meanwhile, high wind-speed in summer may lead to small variations of PM_{2.5} concentrations, as the original PM_{2.5} concentrations are low. Similarly, other meteorological factors are more likely to change PM_{2.5} concentrations significantly when the PM_{2.5} concentrations are high. In the revised manuscript, we have rephrased this part to avoid unnecessary confusions.

Page 14, lines 330-333: This sentence is very vague - can you be more specific?

R: Thanks so much for pointing this out. Yes, this part should be explained with more details. Actually, what we mean here is that some meteorological factors can be dominant factors across China. For instance, according to Fig 3, you can see such factors as temperature and wind were dominant meteorological factors in many regions, including Northeast, Northwest, coastal areas and inland areas; Meanwhile, some meteorological factors for limited regions (Mainly middle inland cities). This part has been added to the revised manuscript.

Figure 2 and 3: I would suggest the background of concentrations be in a gray scale so the colored icons/wind roses stand out more. Also, how different would the maps be if the correlation coefficient were used instead? A statement or two would reinforce the argument for the use of CCM rather than correlation coefficient.

R: Thanks so much for the cartography suggestions. We have updated the Fig 2 and 3 according to your suggestions and made some further revisions to improve the quality of maps.

Another referee also mentioned that the difference between the p value and the correlation coefficient. And we are sorry that we did not make this clear in previous manuscript. Here we simply explained the advantages of CCM method and some findings concerning the comparison between correlation analysis and the CCM method from our previous studies.

The CCM method was proposed by Sugihara et al. (2012).

1 Sugihara, G., May, R., Ye, H., Hsieh, C., Deyle, E., Fogarty, M., Munch, S. 2012. Detecting Causality in Complex Ecosystems. Science, 338, 496-500.

Sugibara et al. (2012) pointed out that correlation analysis could extract mirage correlations, especially in complicated ecosystems. For instance, two variables A and B that have no causality may demonstrate significant correlations due to the existence of an agent variable C, which interacts with both A and B. Through a series of experiments, Sugihara et al. (2012) proved that this type of mirage correlations could be detected using the CCM method by calculating a p value of 0. The CCM method not only performed better than the correlation analysis in causality analysis by excluding the influence of other variables, but also demonstrated the advantage of detecting weak causality compared with other causality analysis method (e.g. Granger Causality), which may fail to detect weak to moderate coupling between variables.

In our previous studies, we employed both the correlation and the CCM method to examine the influence of individual meteorological factors on PM_{2.5} concentrations in the Beijing-Tianjin-Hebei region and compared the performance of correlation and CCM method.

Chen, Z., Cai, J., Gao, B.B., Xu, B., Dai, S., He, B., Xie, X.M. Detecting the causality influence of individual meteorological factors on local PM2.5 concentration in the Jing-Jin-Ji region. Scientific Reports 2017. 7:407352

The comparison suggests that the causality influence of individual meteorological factors on PM2.5 concentration is better revealed using the CCM method than the correlation analysis. By comparing the correlation coefficient and ρ value in Table 2, one can see that some correlations between meteorological factors and PM2.5 concentration may result from mirage correlations (e.g. the correlation between meanRHU and PM2.5 concentration in Hengshui in summer). Secondly, CCM analysis reveals weak or moderate coupling (e.g. the interactions between SSD and PM2.5 concentration in Cangzhou in summer) whilst correlation analysis cannot. Additionally, due to interactions 24 between different meteorological factors, the value of correlation coefficients cannot interpret the quantitative influence of individual meteorological factors on PM2.5 concentration. Instead, the ρ value from CCM method is designed to understand the coupling between two variables by excluding influences from other factors. Through comparison, the value of the correlation coefficient for some meteorological factors is notably different from the ρ value for these meteorological factors. A large correlation coefficient for one meteorological factor may correspond to a much smaller ρ value from the CCM analysis.

The previous research (Chen et al., 2017) proved that the CCM method outperform the correlation analysis in many aspects.

Page 20, lines 414-420: While higher relative humidity does lead to hygroscopic growth of aerosols, this is probably not evident in the observed concentrations since most measurements are taken at a constant relative humidity (e.g, 35% in US and Europe). Measurements in China may not do this, and if so, should be explicitly stated since this can have a major effect to aerosol mass depending on the composition of the aerosol.

R: Thanks so much for this explanation. This information is very useful for future comparison of meteorological influences, especially the humidity factor, on PM_{2.5} concentrations in China and other regions. The reason we added the general introduction of mechanisms how meteorological factors may interact with PM_{2.5} concentration is that one referee during the first stage of ACPD review process suggested we do so. However, during this round of ACPD review, you and other referee all suggested that the part of introduction is well known to scholars with meteorological background and we have deleted this part in the revised manuscript.

Page 23, lines 515-535: This paragraph seems out of place with the rest of the section. Page 23, line 525: I am not able to read Cheng et al. (2015), but I'm wondering what the model is using for predictors? If they are "static" models, isn't that just the mean state? I'm having a hard time understanding. If the argument is to use CCM instead of correlations, an example (see above) would help to reinforce this.

R: Static statistical models did not consider the influence of meteorology on PM_{2.5} concentrations whilst dynamic models select some reliable and key meteorological influencing factors for better predicting PM_{2.5} concentrations. The advantage of p value compared with the correlation analysis has been explained above and added to the revised manuscript. As suggested by another referee, the improvement of models based on the CCM method could be important practical applications of the meteorological influences on PM_{2.5}. So more in-depth discussion concerning this part has been added to the revised manuscript.

Page 24, lines 562-566: How does the frequency of precipitation affect this statement? For example, if precipitation is rare in Beijing during winter, especially compared to the Yangtze River Basin.

R: This is a very good point and has also been pointed out by another referee. Yes, it is highly possible that precipitation is more effective at removing PM along the coasts because it rains at a higher frequency and intensity. As we know that the $PM_{2.5}$ concentrations drop significantly after a heavy rain whilst light rain may not reduce $PM_{2.5}$ concentrations significantly. Meanwhile, $PM_{2.5}$ concentrations may also affect the influence of precipitation. Light rains may have limited washing-off effects on highconcentration $PM_{2.5}$ concentrations and may increase the relative humidity in the 26 environment, which is favorable for the rising of $PM_{2.5}$ concentrations. In the drier interior, the $PM_{2.5}$ concentrations are usually much higher and the intensity and frequency of precipitation are much lower than those along coasts. These two factors may both be the reason that precipitation is more effective at removing PM along the coasts. Due to these influencing factors, precipitation may still be a less important meteorological driving force for $PM_{2.5}$ concentrations in the drier interior, even if there were a way to normalize by the amount of total rainfall. We have added more discuss on this in the revised manuscript.

Results/Discussion: Much of this review of meteorology-PM2.5 relationships in the discussion would probably be better suited in the introduction and in the results as it pertains to different locations within China. Many of the statements in the results are rather vague (e.g., page 14, line 330-333) and could be elaborated to include specific meteorological factors and specific locations.

R: Thanks so much for your comments. As explained above, the review of meteorology-PM2.5 relationships has been removed in the revised manuscript. And some vague statements in the previous manuscript have been re-phrased with more details.

Minor comments

Page 2, line 61: were correlated

R: Corrected

Page 3, line 68: "fractions of three different sizes" of particulate matter

R: Corrected

Page 4, lines 119-120: What does this sentence mean?

R: Sorry that we did not make this clear. The API (Application Programming Interface) tool we programmed can automatically downloaded hourly air pollution data since the execution of this tool.

Page 12, line 288: Awkward wording

R: Rephrased

Page 20, line 426: Wikipedia is not an appropriate citation.

R: Other definition has been added in the revised manuscript in other parts.

Page 20, line 427 and elsewhere: Check your usage of "by analogy" – you may be looking for a different phrase.

R: Thanks so much for this suggestion. We have changed this to "similarly".

Page 20, line 433 and elsewhere: Check subject verb agreement, specifically for "PM2.5" and "concentration(s)"

R: Thanks so much for this. We have corrected all these incorrectly used format.

acp-2017-376-RC3 Anonymous Referee #2

Dear Referee:

Thanks so much for your valuable suggestions. We have fully revised the manuscript according to your suggestions in the revised manuscript. And we are willing to conduct further revision if you have additional requests.

This paper attempts to investigate the meteorological influence on PM2.5 concentra- tions in China at the national scale using a convergent cross-mapping (CCM) method. This method is somewhat new to the atmospheric chemistry community, but the physi- cal mechanism as discussed in this paper is very descriptive and already well-known. Overall I don't feel these results are significant enough to warrant publication in ACP. Here are my major concerns.

R: Thanks so much for pointing this out. Actually, the major aim of this research is to quantify the causality influence of individual meteorological factors on $PM_{2.5}$ concentrations in 190 monitoring cities across China. The spatial and seasonal variations of meteorological influences on $PM_{2.5}$ concentrations at a national scale have rarely been examined before. Meanwhile, previous studies for meteorological influences on $PM_{2.5}$ concentrations at local and regional scale mainly employed the Correlation analysis, which can lead to mirage correlations and unreliable correlation coefficient, due to complicated interactions between different meteorological factors. Thus the use of CCM method has the advantage to remove potential influences from other variables when analyzing the bi-directional coupling between two variables. The comparison and patterns of calculated p value (quantitative causality influence) of individual meteorological factors on $PM_{2.5}$ concentrations across China is the key findings for this research.

Yes, as you pointed out, the e physical mechanism We did not add the physical mechanism of $PM_{2.5}$ -meteorology relationship in the first version of manuscript. One reviewer in the 29

first stage of ACPD review suggested this, and thus we added a brief discussion. However, in this round review, you and other referees all pointed out that this part was off the structure and was already well known to scholars with relevant background. So in the revised manuscript, according to your suggestions, we have deleted this part. And for other major issues you pointed out, e.g. the lack of multiple year analysis ,we have fully revised this manuscript accordingly and explained as follows. Thanks again for your valuable comments and we would like to make further revision in due stages if you have further requirements.

First, the authors just use the PM2.5 observations in one year, from Mar 2014 to Feb 2015, which is far from sufficient to draw any convincing conclusions. In Figure 2, they evaluate the influence of 8 different variables on PM2.5 in each season. This means they make these conclusions using only 90 data values, which is far from enough. When the authors prepare this manuscript, observations in 2015 and 2016 should already be available. Why not include a longer time series of observations into this study?

R: This is a very good point. Long-term observation data are more likely to present reliable causality influence of 8 different variables on PM_{2.5} in each season, as one-year data may be influenced by abnormal meteorological conditions. So according to your suggestions, we managed to collect the PM_{2.5} and meteorological data from Mar 2014 to Feb 2017. In the revised manuscript, we have added additional two years' data for multiple-year analysis and thus a comprehensive CCM analysis based on three year's analysis has been conducted. Thanks again for pointing this out, as the inclusion of multiple-year analysis made the results more robust.

Second, the discussion of the scientific significance of this work looks very superficial and unprofessional. Throughout Section 5.1, the authors made a lot of descriptive statements with

little reference. For example in Line 410-413, the authors claim that rising PM2.5 concentrations prevents the occurrence of winds. Is this true? Can the authors list some references? In my understanding, the effect of aerosols on wind occurrence is much smaller than that from synoptic circulation patterns.

R: Thanks so much for this valuable suggestions. As explained above, we also know that $PM_{2.5}$ —meteorology interactions, as you and another two referees pointed out, the mechanisms were well-known and may be off the focus of this manuscript. For this reason, we did not add this introduction of this part to the original manuscript. During the first stage of ACPD discussion, a referee kindly suggested that a brief introduction of PM2.5-meteorology relationship can be added, and thus we provided a general introduction of mechanisms in the previous manuscript. In the revised manuscript, according to the suggestions of you and other referees, we have deleted this part to make the aim and key findings highlighted. In addition, according to the comments of you and other referees, we have added some more in-depth discussion, concerning the potential applications of this research and underlying reasons for the large variations of meteorological influences on $PM_{2.5}$ concentrations across China, has been added to the revised manuscript.

Although the PM_{2.5}—meteorology interaction part has been removed, we would like to give some explanations on the example you suggested. Yes, we understood that synoptic circulation patterns were the major causes for wind occurrence and we are not claiming that the effects of aerosols were large enough compared with the synoptic circulations. We just pointed out that the potential mechanisms of the negative feedbacks of high PM_{2.5} concentrations

Yang et al. (2015) observed four haze episodes during Oct to Nov, 2014 and during these four haze episodes in the North China plain, the very high PM_{2.5} concentrations all led to

stagnant condition and weak high-pressure systems, which further led to slowed wind speed and disturbed wind direction. This phenomenon was also observed by Liu et al. (2014) in haze episodes in Beijing in 2013. Very high PM_{2.5} concentrations induced haze episodes further led to stagnant and stable high-pressure systems, which made megacities serve as obstacles to significantly slow down the wind speed (Yang et al., 2015). Therefore, the effects of aerosols, especially high-concentration PM_{2.5} concentrations, prevented the wind occurrence mainly through indirect mechanisms.

Yang, Y. R., Liu, X. G., Qu, Y., An, J. L., Jiang, R., & Zhang, Y. H., et al. (2015). Characteristics and formation mechanism of continuous hazes in china: a case study during the autumn of 2014 in the north china plain. Atmospheric Chemistry & Physics, 15(14), 10987-11029.

Liu, X. G., Li, J., Qu, Y., Han, T., Hou, L., & Gu, J., et al. (2013). Formation and evolution mechanism of regional haze: a case study in the megacity beijing, china. Atmospheric Chemistry & Physics, 13(9), 4501-45

Understanding meteorological influences on PM_{2.5} concentrations across China:

`a temporal and spatial perspective

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Abstract

With frequent haze events in China, growing research emphasis has been put on quantifying meteorological influences on PM2.5 concentrations. However, these studies mainly focus on isolated cities whilst meteorological influences on PM2.5 concentrations at the national scale have yet been examined comprehensively. This research employs the CCM (Cross Convergent Mapping) method to understand the causality influence of individual meteorological factors on local PM2.5 concentrations in 189188 monitoring cities across China. Results indicate that meteorological influences on PM2.5 concentrations are of notable seasonal and regional variations. GenerallyFor the heavily polluted North China region, the higher PM25 concentrations, the larger influences meteorological factors exert on PM2.5 concentrations. 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 PM2.5 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 and air pressure exert stronger influences on PM2.5 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 PM2.5 concentrations, this research suggests pertinent environmental projects for air quality improvement should be designed accordingly for specific regions.

Keywords: PM2.5; Meteorological factors; Causality analysis; CCM

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1 Introduction

2 With rapid social and economic growth in China, both the government and residents are 3 placing more and more emphasis on the sustainability of the ambient environment-4 Amongst these environmental elements, ambient, and air quality has become one of the 5 most concerned social and ecological issues. Recently, the frequency of haze events and 6 the number of cities influenced by haze have increased notably in China since 2013. 7 Statistical records from the national air quality publishing platform 8 (http://113.108.142.147:20035/emcpublish/) revealed that haze events occurred in 25 9 provinces and more than 100 middle-large cities whilst there were on average 30 days with

10 haze for each monitoring city in 2014.

11 Serious haze not only influences people's daily life; (e.g. the cause of severe traffic jam 12 during haze epsiodes), but also severely threatens the health of residents that suffer from 13 polluted air quality. Recent studies (Garrett and Casimiro, 2011; Guaita et al., 2011; Qiao 14 et al., 2014; Pasca et al., 2014; Lanzinger et al., 2015; Li et al., 2015) have proven that 15 airborne pollutants, PM2.5 in particular, are closely related to all-cause and cause-specific-16 cause mortality. In consequence, scholars have been working towards a better 17 understanding of sources (Guo et al., 2012; Zhang et al., 2013; Gu et al., 2014; Liu et al., 18 2014; Cao et al., 2014), characteristics (Wei et al., 2012; Zhang et al., 2013; Hu et al., 2015; 19 Zhang, F. et al., 2015; Zhen et al., 2016; Zhang et al., 2016) and seasonal variations (Cao 20 et al., 2012; Shen et al., 2014; Yang and Christakos, 2015; Wang et al., 2015; Chen et al., 21 2015; Chen, Y. et al. 2016; Chen, Z. et al., 2016) of PM_{2.5} and other airborne pollutants. 22 Meanwhile, large-scale research on the variation and distribution of $PM_{2.5}$ has been 23 conducted using a variety of remote sensing sources and spatial data analysis methods (Ma 24 et al., 2014; Kong et al., 2016.)

25 One key issue for air quality research is to find the source and influencing factors for 26 airborne pollutants. Although quantitative contributions of different sources (e.g. coal 27 burning and automobile exhaust) to airborne pollutants remain controversycontroversial, 28 meteorological influences on airborne pollutants have been examined in depth by more 29 and more scholars. Recently, massive studies have been conducted to extract quantitative 30 correlations between meteorological factors and air pollutants. Blanchard et al. (2010) 31 indicated that ozone concentrations was linearly correlated with temperature and humidity, 32 and non-linearly correlated with other meteorological factors. Juneng et al. (2011) 带格式的: 字体: Cambria

33 suggested that such meteorological factors as temperature, humidity and wind speed, 34 dominated the fluctuation of PM10 over the Klang Valley during summer monsoon. In 35 Melbourne, Pearce et al. (2011) found that local temperature led to strongest responses of 36 different pollutants, whilst other meteorological factors (e.g. winds, water vapor pressure, 37 radiation, precipitation) affected one or more specific pollutants. In the city of Elche, Spain, 38 Galindo et al. (2011) revealed that fractions of three different PM sizes (PM1, PM2.5 and 39 PM_{10}) were negatively correlated with wind speed in winter, whilst coarse fractions were 40 strongly correlated with temperature and solar radiation. At a site of the Egyptian 41 Mediterranean coast, El-Metwally and Alfaro (2013) suggested found that the wind speed 42 not only influenced the dilution of airborne pollutants, but also affected the composition 43 of airborne pollutants. For a Western Indian location, Udaipur, Yadav et al. (2014) proved 44 that precipitation exerted a stronger influence on PM₁₀ than on PM_{2.5}. High temperature 45 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 46 47 vertical temperature gradients were favorable meteorological conditions for elevated NOx 48 and particle number concentrations (PNC). Zhang et al. (2015b) quantified the correlations 49 between meteorological factors and main airborne pollutants in three megacities, Beijing, 50 Shanghai and Guangzhou, --and pointed out that the influences of meteorological factors 51 on the formation and concentrations of airborne pollutantpollutants varied significantly 52 across seasons and geographical locations. Chen, Z. et al. (2017) quantified the 53 meteorological influences on local PM2.5 concentrations in the Beijing-Tianjin-Hebei 54 region and revealed that wind, humidity and radiation were major meteorological factors 55 that significantly influenced local PM2.5 concentrations in winter.

56 Although correlations between airborne pollutants and meteorological factors have been 57 massivelywell studied, analyzing the sensitivity of airborne pollutants to individual 58 meteorological parameters remains challenging (Pearce et al., 2011). This is because 59 different meteorological factors are inherently interacting and can thus influence airborne 60 pollutants through direct and indirect mechanisms. Due to the diversity of meteorological 61 factors and complicated interactions between them, Pearce et al (2011) suggested that 62 multiple models and methods should be comprehensively employed to quantify the 63 influence of meteorological factors on local airborne pollutants. Our previous research 64 (Chen, Z., 2017) proved that the CCM (Cross Convergent Mapping) method performed

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65 better in quantifying the causality influence of individual meteorological factors on PM2.5 66 concentrations than traditional correlation analysis through comprehensive comparison. However, this study mainly focused on the meteorological influences on PM2.5 67 68 concentrations in a specific region. As pointed out by some scholars, interactions between 69 meteorological factors and airborne pollutants are of great variations for different regions, 70 yet most relevant studies have been conducted at the local or regional scale. China is a 71 large country, including many regions with completely different air pollution levels, 72 geographical conditions and meteorological types. To better understand the variations of 73 meteorological influences on PM2.5 concentrations, a comparative study at the national 74 scale is required.

75 In accordance with these challenges, this research aims to quantify and compare influences 76 of individual meteorological factors on PM2.5 concentrations in different cities across 77 China. Based on the causality analysis, dominant meteorological factors for PM_{2.5} 78 concentrations can be extracted for each city and spatio-temporal patterns of 79 meteorological influences on PM2.5 concentrations across China can be revealed. In 80 addition to its theoretical significance, this research may provide useful reference for 81 evaluating pertinent environmental projects and enhancing air quality through 82 meteorological measures.

83 2 Materials

84 2.1 Data sources

85 2.1.1 PM_{2.5} data

86 $PM_{2.5}$ data are acquired from the website PM25.in. This website collects official data of 87 PM_{2.5} concentrations provided by China National Environmental Monitoring Center 88 (CNEMC) and publishes hourly air quality information for all monitoring cities. Before 89 Jan 1st, 2015, PM25.in publishes data of 190 monitoring cities. Since Jan 1st, 2015, the 90 number of monitoring cities has increased to 367. By calling specific API (Application 91 Programming Interface) provided by PM25.in, we collect hourly PM_{2.5} data for target cities. 92 The daily PM_{2.5} concentrations for each city is calculated using the averaged value of 93 hourly PM2.5 concentrations measured at all available local observation stations. For a 94 consecutive division of different seasons, and multiple-year analysis, We collected PM2.5 95 data from March 1st, 2014 to February 28th, 2015 were employed 2017 for the following

96 analysis.

97 2.1.2 Meteorological data

98 The meteorological data for these monitoring cities are obtained from the "China Meteorological Data Sharing Service System", part of National Science and Technology 99 100 Infrastructure. The meteorological data are collected through thousands of observation 101 stations across China. Previous studies (Zhang et al., 2015b; Pearce et al., 2011; Yadav et 102 al., 2014) proved that such meteorological factors as relative humidity, temperature, wind 103 speed, wind direction, solar radiation, evaporation, precipitation, and air pressure may be 104 related to PM2.5 concentrations. Therefore, to comprehensively understand meteorological 105 driving forces for PM2.5 concentrations in China, all these potential meteorological factors 106 were selected as candidate factors. To better quantify the role of these meteorological 107 factors in affecting local PM2.5 concentrations, these factors are further categorized into 108 some sub-factors: evaporation (small evaporation and large evaporation, short for 109 smallEVP and largeEVP²), temperature (daily max temperature for the day, mean 110 temperature for the day, min temperature for the day, and largest temperature difference 111 for the day, short for maxTEM, meanTEM, minTEM and difTEM), precipitation (total 112 precipitation from 8am-20pm, total precipitation from 20pm-8am and total precipitation 113 for the day, short for PRE8-20, PRE20-8 and totalPRE), air pressure (daily max pressure, mean pressure and min pressure, short for maxPRS, meanPRS and minPRS), humidity 114 115 (daily mean and min relative humidity, short for meanRHU and minRHU), radiation 116 (sunshine duration^{$\frac{3}{2}$} for the day, short for SSD), wind speed (mean wind speed, max wind 117 speed, extreme wind speed⁴, short for meanWIN, maxWIN and extWIN), wind direction 118 (max wind direction⁵ for the day, short for dir_maxWin). As there are one or more 119 observation stations for each city, the daily value for each meteorological factor for each 120 city was calculated using the mean value of all available observation stations within the 121 target city. To conduct time series comparison, we also collected meteorological data from 122 March 1st, 2014 to February 28th, 2017.

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.

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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.

⁵ The max wind direction indicates the dominant wind direction for the period with the max wind speed. 37

123 2.2 Study sites

For a comprehensive understanding of meteorological influences on local PM_{2.5} concentrations across China, all monitoring cities (except for Liaochen<u>g and Zhuji</u>, where continuous valid meteorological data were not available) during the study period were selected for this research. The <u>189188</u> 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 <u>the densitythat</u> in regions with good air quality.

131 3 Methods

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

136 To extract the coupling between individual variables in complex systems, Sugihara et al. (2012) proposed a convergent cross mapping (CCM) method. Different from Granger 137 138 causality (GC) analysis (Granger, 1980), the CCM method is sensitive to weak to moderate 139 coupling in ecological time series. By analyzing the temporal variations of two time-series 140 variables, their bi-directional bidirectional coupling can be featured with a convergent map. 141 If the causality influence of one variable on the other variable is presented as a convergent 142 curve with increasing time series length, then the causality is detected; If the curve 143 demonstrates no convergent trend, then no causality influence exists. The predictive skill 144 (defined as $\rho \rho$ value), which ranges from 0 to 1, suggests the quantitative causality 145 influence of one variable on the other.

147 Two time series $\{X\} = [X(1), \dots, X(L)]$ and $\{Y\} = [Y(1), \dots, Y(L)]$ are defined as the 148 temporal variations of two time series variables X and Y. For r = S to L(S < L), two partial

- time series $[X(1), ..., X(L_P)]$ and $[Y(1), ..., Y(L_P)]$ 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
- 152 vectors $\mathbf{x}(t) = \langle X(t), X(t-\tau), \dots, \tau \rangle$, $X(t-(E-1)\tau \tau) >$ for $t = 1+(E-1)\tau \tau$ to t = r. To generate
- a cross-mapped estimate of $Y(t) \leftarrow \hat{Y}(\hat{Y}(t)|M_X)$, the contemporaneous lagged-coordinate

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154	vector on M_x , x(t) is located, and then its E+1 nearest neighbors are extracted, where E+1
155	is the minimum number of points required for a bounding simplex in an E-dimensional
156	space (Sugihara and May, 1990). Next, the time index of the $E+1$ nearest neighbors of $x(t)$
157	is denoted as t_1 ,, t_{E+1} . These time index are used to identify neighbor points in \underline{Y} and
158	then estimate $Y(t)$ according to a locally weighted mean of E+1 $Y(t_i)$ values (Equation 1).

159

$$\hat{Y}(t)|M_x = \sum_{i=1}^{k+1} w_i Y(t_i)$$
(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

162

2.

163	$w_i = u_i \Big/ \sum_{j=1}^{E+1} u_j$	(E2)

164 165 Where $u_i = e^{-d[\underline{x}(t),\underline{x}(t_i)]/d[\underline{x}(t),\underline{x}(t_i)]}$ whilst $d[\underline{x}(t), \underline{x}(t_i)]$ represents the Euclidean distance between two vectors.

166 In our previous research, interactions between the air quality in neighboring cities (Chen, 167 Z. et al., 2016), and bidirectional coupling between individual meteorological factors and 168 PM_{2.5} concentrations (Chen, Z. et al., 2017) were quantified effectively using the CCM 169 method. By comparing the performance of correlation analysis and CCM method, Chen, 170 Z-et al. (2017) proved that the CCM method not only detected mirage correlations, but 171 also extracted weak coupling, which may not be detected by correlation analysis. Additionally, Chen, Z et al. (2017) indicated that the -P-value was a more reliable 172 173 indicator of quantitative meteorological influences on PM2.5 concentrations than the 174 correlation coefficient. et al. (2017) suggested that correlation analysis may lead to a 175 diversity of biases due to complicated interactions between individual meteorological 176 factors. Firstly, some mirage correlations (two variables with a moderate correlation 177 coefficient) extracted using the correlation analysis were revealed effectively using the 178 <u>CCM method (the ρ value between two variables was 0). Secondly, some weak coupling</u>, 179 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). 180 181 Meanwhile, as Sugihara et al. (2012) suggested, the correlation between two variables 182 could be influenced significantly by other agent variables and thus the value of correlation 183 coefficient between two variables could not reflect the actual causality between them. 39

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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.

191 4 Results

192 Seasonal variations of PM2.5 concentrations have been proved by a large body of studies 193 (Cao et al., 2012; Shen et al., 2014; Yang and Christakos, 2015; Wang et al., 2015; Chen 194 et al., 2015; Chen, Y. et al. 2016; Chen, Z. et al., 2016). Hence, the research period was 195 divided into four seasons. According to traditional season division for China, spring was 196 set as the period between March 1st, 2014 and May 31st, 2014; summer was set as the 197 period between June 1st, 2014 and August 31st, 2014; autumn was set as the period between 198 September 1st, 2014 and November 30th, 2014; and winter was set as the period between 199 December 1st, 2014 and February 28th, 2015. For each city, the bidirectional coupling 200 between individual meteorological factors and PM2.5 concentrations in different seasons 201 was analyzed respectively using the CCM method. The CCM method is highly automatic 202 and only few parameters need to be set for running this algorithm: E (number of 203 dimensions for the attractor reconstruction), τ (time lag) and b (number of nearest 204 neighbors to use for prediction). The value of E can be 2 or 3. A larger value of E produces 205 more accurate convergent maps. The variable b is decided by E(b = E + 1). A small value 206 of τ leads to a fine-resolution convergent map, yet requires much more processing 207 time. Through experiments, we found that the final results were not sensitive to the 208 selection of parameters and different parameters mainly exerted influences on the 209 presentation effects of CCM. In this research, to acquire optimal presentationinterpretation 210 effects of convergent cross maps, the value of $\tau \tau$ was set as 2 days and the value of E 211 was set 3. For each meteorological factor, its causality coupling with PM_{2.5} concentrations 212 can be represented using a convergent map. Since it is not feasible to present all these 213 convergent maps here, we simply display some exemplary maps to demonstrate how CCM 214 works (Fig 1).





















217 $\rho \div \rho$: predictive skills. $t \div L$: the length of time series. A xmap B stands for convergent cross

mapping B from A, in other words, the causality influence of variable B on A. For instance, PM_{2.5}

xmap meanRHU stands for the causality influence of meanRHU on PM_{2.5} concentrations. meanRHU

220 xmap PM_{2.5} stands for the feedback effect of PM_{2.5} on meanRHU concentrations- ρ -indicates the

221 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 $(\rho_{\perp} \rho_{\perp} > 0.4)$ whilst SSD and minTEM also had a weaker influence on local PM_{2.5} concentrations. $(\rho_{\perp} \rho_{\perp} > 0.4)$

 $(\rho \text{ close to } 0.2)$. On the other hand, serious haze weather (high PM_{2.5} concentrations) had

228 an even stronger feedback influence on meanRHU, maxWIN and SSD (ρ (ρ close to 0.6)

whilst PM_{2.5} had little influence on minTEM $(\rho (\rho \text{ close to } 0))$. The bidirectional coupling

230 between PM_{2.5} concentrations and individual meteorological factors provides useful

231 reference for a better understanding of the form and development of serious haze events.

For Beijing, low wind speed (high humidity and <u>low</u>SSD⁶) in winter results in high PM2.5

concentrations, which in turn causes lower wind speed (higher humidity and lower SSD).

In consequence, PM_{2.5} concentrations isare increased further by the changing wind

⁶ The interaction between some individual meteorological factors (e.g. SSD) and PM_{2.5}-concentrations may be difficult to understand, and a brief explanation is given in the discussion part.-

In this case, the haze is unlikely to disperse and persistent haze weather usually lasts for a
long period in this region. By analogySimilarly, 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.

243 The $\rho \rho$ value is a direct indicator of quantitative causality influences. For this research, 244 the maximum ρ value of all sub-factors in the same category was used as the causality influence of this specific meteorological factor on $PM_{2.5}$ concentrations. E.g. for a specific 245 246 city, the maximum $\rho \rho$ value of maxTEM, meanTEM, minTEM and difTEM is used as 247 the influence of temperature on local PM2.5 concentrations. For this research, we collected 248 meteorological and PM2.5 data for three consecutive years. To avoid the analysis of 249 inconsecutive time series, which may influence the CCM result, we did not calculate the 250 general influence of individual meteorological factors on PM2.5 concentrations during 251 2014-2016 by analyzing three isolated periods (e.g. April- June, 2014, April-June, 2015, 252 and April- June, 2016) as a complete data set. Instead, for each city, we quantified the 253 influence of individual meteorological factors on PM2.5 concentrations for each season in 254 2014, 2015 and 2016 respectively and calculated the mean ρ value during 2014-2016

255 <u>for each city.</u>

Generally, it is difficult to properly demonstrate the influence of eight meteorological factors on PM_{2.5} concentrations for all <u>189188</u> cities on a comprehensive map. Therefore, two cartography strategies were employed to explain the meteorological influences on PM_{2.5} concentrations across China.

260 4.1 Comprehensive meteorological influences on PM2.5 concentrations in some

261 regional representative cities

When the $\not - \rho$ value for each meteorological factor was calculated, a wind rose, which presents the quantitative influenceinfluences of all individual meteorological factors on PM_{2.5} concentrations, can be produced for each city. It is not feasible to present all 189188 wind roses simultaneously, due to severe overlapping effects. Thus, considering the socialecologicaleconomic 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













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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:
- 279 a. Like seasonal variations of PM2.5 concentrations, the influences of individual 280 meteorological factors on local PM2.5 concentrations vary significantly. For a specific city, 281 the dominant meteorological driver for PM2.5 concentrations in one season may become 282 insignificant in another season. E.g. in winter, one major meteorological influencing factor 283 for Beijing is wind, which exerts little influence on PM2.5 concentrations in summer. 284 Furthermore, it is noted that seasonal variations of meteorological influences on PM2.5 285 concentrations apply to all these representative cities, as the shape and size of wind rose 286 for each city change significantly across different seasons.
- 287 b. In spite of notable differences in the shape and size of wind roses, meteorological 288 influences on PM_{2.5} concentrations cities are of some regional patterns, subject to local 289 PM2.5-concentrations. For instance, PM2.5 concentrations in cities within the Beijing-290 Tianjin HebeiNorth China region (or Norththe Northeast China region) is influenced by 291 similar dominant meteorological factors, especially in winter, when PM_{2.5} concentrations 292 in these cities was high. By analogyMeanwhile, meteorological influences on PM2.5 293 concentrations in the Kuerle and Karamay (cities within Xinjiang province) are 294 generally the Yangtze River basin were also highly similar, especially in winter. However, 295 meteorological influences on PM2.5-concentrations in their neighboring city, Urumchi, are 296 quite different. This may attribute to the fact that PM2.5 concentrations in Urumchi is much higher than that in Kuerle and Karamayall seasons. As we can see, meteorological 297 298 influences on PM_{2.5} concentrations in China are mainly controlled by both-geographical 299 conditions (e.g. terrain and landscape patterns) and local PM2.5 concentrations per se.).

300 c. Except for some specific cities (e.g. Lasa), c. For the heavily polluted North China region, 301 the higher local PM_{2.5} concentrations, the larger influence meteorological factors exerts on 302 $PM_{2.5}$ concentrations. $PM_{2.5}$ concentrations is are usually the highest in winter, causing 303 serious smog eventshaze episodes across China, the North China region in particular, 304 whilst. Meanwhile, PM_{2.5} concentrations in spring and summer is comparatively low. 305 Accordingly, there are more influencing meteorological factors on PM2.5 concentrations for most cities within this region and the $\frac{\rho}{\rho}$ value of these meteorological factors is 306 307 notably larger in winter. As explained above, bidirectional interactions between 308 meteorological factors and PM2.5 concentrations may lead to complicated mechanisms that

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- 309 further enhance local PM_{2.5} concentrations significantly. Therefore, strong meteorological
- 310 influences on PM_{2.5} concentrations in winter are a major cause for the form and persistence
- 311 of haze events within the North China region, which experiences the most frequent and
- 312 severe air pollution in China..
- 313 Although some general patterns of meteorological influences on PM2.5 concentrations
- 314 across China may be concluded according to Fig 2, spatial and temporal variations of
- 315 meteorological influences on PM2.5 concentrations should be further examined in depth
- 316 based on the statistics of all 189188 monitoring cities. Hence, we employed another
- 317 cartography strategy to demonstrate spatial and temporal variations of meteorological
- 318 influences on local PM2.5 concentrations across China.

319 4.2 Spatial and temporal variations of the dominant meteorological influence on local

320 PM_{2.5} concentrations across China

- 321 Through statistical analysis, we selected the factor with the largest \mathcal{P}^{ρ} value as the 322 dominant meteorological factor for local PM2.5 concentrations. The spatial and temporal 323 variations of the dominant meteorological influence on local PM2.5 concentrations across 324 China are demonstrated as Fig 3. According to Fig 3, some spatio-temporal characteristics
- 325 of meteorological influences on PM2.5 concentrations can be further concluded:
- 326 a. The dominant meteorological factor for PM2.5 concentrations is closely related to
- 327 geographical conditions. For instance, the factor of *precipitation* may exert a key influence
- 328 on local PM2.5 concentrations in some coastal cities and cities within the Yangtze River
- 329 basin whilst this meteorological factor exerts limited influence on PM2.5 concentrations 330
- within some inland regions (e.g. the Beijing-Tianjin-Hebei region).
- 331 b. Some meteorological factors (e.g. temperature, wind and humidity) can be the dominant 332 factor for cities within different regions whilstbut some (e.g. evaporation and SSD) are 333 mainly the dominant meteorological factor for PM2.5 concentrations in cities within some 334 specific regions. In other words, some factors can be regarded as regional and national 335 meteorological factors for PM_{2.5} concentrations, yet some meteorological factors are 336 context-related influencing factors for local PM2.5 concentrations. For instance, such 337 factors as temperature, wind and humidity serve as the dominant meteorological factors in
- 338 many regions, including Northeast, Northwest, coastal areas and inland areas; Meanwhile,
- 339 such factors as SSD and Wind direction serve as the dominant meteorological factors

340 mainly in some inland regions.

341 c. Similar to patterns revealed in Fig 2, the $\frac{\rho}{\rho}$ value for the dominant meteorological factors is the largestmuch larger in winter than that in summer. Furthermore, it is noted 342 343 that the dominant meteorological factors demonstratesdemonstrate more regional 344 similarity when PM2.5 concentrations is highin winter. For instance, the dominant 345 meteorological factors for PM_{2.5} concentrations in the heavily polluted North China region 346 are more concentrated and homogeneously distributed in winter (mainly the wind and 347 humidity factor) whilst a diversity of dominant meteorological factors (includes wind, 348 temperature, wind direction and air pressure) for PM2.5 concentrations is irregularly 349 distributed within this region in summer. Based on According to this pattern, when a 350 regional haze eventepisode occurs in winter, the regional air quality is more likely to be 351 simultaneously improved by the same meteorological factor. This is consistent with the 352 common scene in winter that regional haze events in the Beijing-Tianjin-Hebei region can 353 be considerably mitigated by strong winds. On the other hand, regional air pollution in 354 summer can hardly be solved simultaneously through one specific meteorological factor.-











362 **4.3** Comparative statistics of the influence of individual meteorological factors on

363 local PM_{2.5} concentrations across China

 $364 \qquad \text{In addition to meteorological influences on $PM_{2.5}$ concentrations for individual cities,}$

365 we examined and compared the comprehensive influence of individual meteorological

 $366 \qquad factors \ on \ PM_{2.5} \ concentrations \ at \ a \ national \ scale. \ The \ results \ are \ presented \ as \ Table \ 1$

367 and Fig 4.

B68 Table 1. The comparison of the influence of individual meteorological factors on

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Season	FactorsFa	TEM	SSD	PRE	EVP	PRS	RHU	WIN	Dir_W
	<u>ctor</u>								IN
	No. of	<u>4576</u>	<u>81</u>	22<u>13</u>	<u>83</u>	31<u>13</u>	34<u>17</u>	35<u>64</u>	6 1
	cities ¹								
	Mean $-\!$	0. 281 2	0. 138<u>1</u>	0. <u>1521</u>	0. 131<u>1</u>	0. 209<u>1</u>	0. 204<u>1</u>	0. 215 2	0. 104<u>0</u>
Sprin	$\frac{\rho}{value}$	<u>54</u>	<u>02</u>	<u>43</u>	<u>08</u>	<u>77</u>	<u>61</u>	<u>22</u>	<u>94</u>
g	SD of -P-	0. 024<u>1</u>	0. 019 0	0. 024<u>0</u>	0. 021<u>0</u>	0. 028<u>1</u>	0. 028<u>1</u>	0. 019<u>1</u>	0. 015 0
	$\frac{\rho}{value}$	<u>06</u>	<u>71</u>	<u>88</u>	<u>81</u>	<u>23</u>	<u>05</u>	<u>02</u>	<u>77</u>
	Max $\not\!$	0. 747<u>5</u>	0. 617<u>3</u>	0. 723<u>3</u>	0. 610<u>3</u>	0. 714<u>6</u>	0. 796<u>4</u>	0. 555<u>5</u>	0. 502<u>4</u>
	$\frac{\rho}{value}$	<u>72</u>	<u>66</u>	<u>85</u>	<u>97</u>	<u>53</u>	<u>75</u>	<u>95</u>	<u>29</u>
	No. of	38<u>78</u>	4 <u>5</u>	37<u>22</u>	7 <u>1</u>	35<u>20</u>	41 <u>32</u>	23<u>27</u>	7 <u>3</u>
	cities								
	Mean 🗜	0. 244<u>2</u>	0. 107 1	0. 179 1	0. 119<u>1</u>	0. 175<u>1</u>	0. 221 2	0. 168 1	0. 067<u>0</u>
Summ	<u>ρ</u> value	<u>72</u>	<u>36</u>	<u>83</u>	<u>37</u>	<u>63</u>	<u>19</u>	<u>91</u>	<u>87</u>
er	SD of 🗜	0. 019 0	0. 014<u>0</u>	0. 023 0	0. 014<u>0</u>	0. 021<u>1</u>	0. 024<u>1</u>	0. 015 0	0. 007<u>0</u>
	<u>ρ</u> value	<u>98</u>	<u>86</u>	<u>99</u>	<u>88</u>	<u>09</u>	<u>18</u>	<u>95</u>	<u>62</u>
	Max $-\!$	0. <u>6116</u>	0. <u>5074</u>	0. 716<u>5</u>	0. 625 3	0. 676<u>5</u>	0. 69 4 <u>5</u>	0. 536<u>4</u>	0. 364<u>3</u>
	$\frac{\rho}{value}$	<u>04</u>	<u>33</u>	<u>36</u>	<u>99</u>	<u>18</u>	<u>62</u>	<u>53</u>	<u>11</u>
	No. of	58 70	<u>31</u>	18<u>13</u>	21<u>15</u>	4 <u>313</u>	20 27	<u>2348</u>	3 1
A 4	cities								
Autu	Mean 🗜	0. 330<u>3</u>	0. <u>1321</u>	0. 159<u>1</u>	0. 176<u>1</u>	0. 271<u>1</u>	0. 225 2	0. 230 2	0. 082 1
11111	ρ_{value}	<u>16</u>	<u>64</u>	<u>91</u>	<u>81</u>	<u>99</u>	<u>47</u>	<u>65</u>	<u>04</u>
	SD of P	0. 020<u>1</u>	0. 014<u>0</u>	0. 025 0	0. 027<u>1</u>	0. 029<u>0</u>	0. 028<u>1</u>	0. 018<u>0</u>	0. 009<u>0</u>

PM2.5 concentrations in 189188 cities across China (2014-2016)

	<u>value</u>	<u>09</u>	<u>98</u>	<u>93</u>	<u>17</u>	<u>91</u>	<u>25</u>	<u>89</u>	<u>74</u>
	Max 🗜	0. 641<u>7</u>	0.4 <u>724</u>	0. 714<u>4</u>	0. 637<u>5</u>	0. 697<u>5</u>	0. 773<u>6</u>	0. 556<u>4</u>	0.4 <u>523</u>
	<u>value</u>	<u>02</u>	<u>79</u>	<u>30</u>	<u>14</u>	<u>24</u>	<u>62</u>	<u>88</u>	<u>31</u>
	No. of	4 <u>356</u>	<u>83</u>	4 <u>027</u>	<u>85</u>	<u>444</u>	<u>3448</u>	4 <u>044</u>	<u>21</u>
	cities								
	Mean \mathcal{P}	0. 310<u>3</u>	0. 172<u>1</u>	0. 200<u>1</u>	0. 185<u>1</u>	0. 198<u>1</u>	0. <u>3003</u>	0. 255<u>2</u>	0. 115<u>1</u>
Winte	<u>value</u>	<u>06</u>	<u>83</u>	<u>66</u>	<u>90</u>	<u>80</u>	<u>04</u>	<u>99</u>	<u>19</u>
r	SD of $\stackrel{-\rho}{\to}$	0. 017<u>0</u>	0. 019<u>1</u>	0. 045<u>1</u>	0. 025<u>1</u>	0. 019<u>0</u>	0. 028<u>1</u>	0. 033<u>1</u>	0. 015 0
	<u>value</u>	<u>94</u>	<u>29</u>	<u>15</u>	<u>30</u>	<u>86</u>	<u>61</u>	<u>36</u>	<u>92</u>
	Max $-\!$	0. 626<u>5</u>	0. 611<u>6</u>	0. 770<u>4</u>	0. 591<u>5</u>	0. 634<u>4</u>	0. 721<u>7</u>	0. 746<u>6</u>	0. 525 5
	<u></u> value	<u>27</u>	<u>15</u>	<u>73</u>	<u>95</u>	<u>27</u>	<u>55</u>	<u>23</u>	<u>60</u>

¹No. of cities: The the number of cities with this factor as the dominant meteorological factor (its $-\rho \rho$

 $371 \qquad \text{value is the largest amongst eight factors) on local PM_{2.5} concentrations.}$





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Fig 4. The comparison<u>Violin plots</u> of the influence of eight different meteorological factors on local PM_{2.5} concentrations in <u>189188</u> cities across China (violin plot) No. of cities: <u>Thethe</u> number of cities with this factor as the dominant meteorological factor (its $\frac{\rho}{2}$ value is the largest amongst eight factors) on local PM_{2.5} concentrations. The shape of the violin

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380 We compared the influence of individual meteorological factors on $PM_{2.5}$ 381 concentrations from different perspectives.

bars indicated the <u>frequency</u> distribution frequency of $\frac{\rho}{\rho}$ value for 189188 cities.

382 a. From a national perspective, temperature, humidity, wind and air pressure wind exert 383 stronger influences on local PM2.5 concentrations than other factors. The annual mean 384 $-\frac{\rho}{\rho}$ value for temperature, humidity, wind and air pressure humidity was 0.291287, 0.238, 0.217244 and 0.213233, compared with wind direction (0.092101), SSD 385 386 (0.137146), evaporation (0.153) and 155), precipitation (0.173171) and air pressure 387 (0.180). Amongst the eight factors, temperature was proved to be the most influential 388 meteorological factor for general PM2.5 concentrations in China. In addition to the 389 largest mean $\frac{\rho}{\rho}$ value, *temperature* was the dominant meteorological factors for 390 most cities in all seasons. Furthermore, the Coefficient of Variation (SD/mean $\times 100\%$) 391 for temperature was much smaller than other factors, indicating the consistent influence 392 of temperature on local PM2.5 concentrations across China.

393 b. Although some meteorological factors exert a limited influence on $PM_{2.5}$

394 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 $\frac{-\rho}{P}$ value for the eighteach meteorological factors in each seasonfactor was large than 0.535 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, the influence of some-meteorological factors, which that have little influence on PM_{2.5} concentrations at a large scale, should be carefully examined at the local scale.

402 c. Some factors (e.g. *precipitation* in summer and winter) may be the dominant 403 meteorological factors for a large number of cities, though the mean $\frac{\rho}{P} \frac{\rho}{P}$ value 404 remained small. This may be attributed to the fact that these meteorological factors 405 mainly exert influence on local PM_{2.5} concentrations in those cities (seasons);) where 406 (when) the general PM_{2.5} concentrations is not high. In this case, as explained above, 407 comprehensive meteorological influences on PM_{2.5} concentrations are limited 408 considerably.

409 5 Discussion

10 5.1 Underlying mechanisms for bidirectional coupling between PM2.5

11 concentration and individual meteorological factors

412 Although the CCM method quantified the causality between PM2.5 concentration and 413 individual meteorological factors, it did not explain how these variables were interacted. 414 To better understand meteorological influences on PM2.5 concentration and its feedback 415 effects, we attempt to give some brief explanation on the mechanisms of some typical 416 bidirectional coupling. As we know, that one meteorological factor may influence PM2.5 417 concentrations through different mechanisms and here we only explain some 418 fundamental interactions between PM2.5 concentrations and individual meteorological 419 factors.

- Interactions between wind and PM_{2.5}: On one hand, winds, especially strong winds
 blow airborne pollutants away and reduce PM_{2.5} concentration effectively. On the other
 hand, high PM_{2.5} concentration, especially a quickly rising PM_{2.5}-concentration brings
- 123 the atmospheric environment to a comparatively stable status, which prevents the form
- 424 of winds and reduces the wind speed in smog covered areas.

Interactions between humidity and PM_{2.5}. Higher humidity causes more vapor attached to the Particulate Matter and significantly increases the size and mass concentration of PM, namely the hygroscopic increase and accumulation of PM_{2.5} (Fu et al., 2016). On the other hand, the larger mass and higher concentration makes it difficult for PM_{2.5} to disperse and leads to a stable polluted atmospheric environment, which is not favorable for the vapor evaporation and further increase the environmental humidity.

432 Interactions between SSD and PM2.5: Previous studies (Guo et al., 2012; Zhang et al., 433 2013; Cao et al., 2014; etc) have proved that organic carbon (OC) is an important 434 component for PM2.5, and atmospheric photolysis could occur on OC to reduce PM2.5 435 concentration. Therefore, longer SSD has a negative influence on PM2.5 concentration. 436 On the other hand, SSD is a general indicator of cloudiness 437 -(https://en.wikipedia.org/wiki/Sunshine_duration-) -. The more cloud, the less SSD 438 received on the ground observation station. By analogy, serious smog (thick black fog) 439 caused by high PM2.5 concentration notably blocked radiation emitted to the ground and 440 thus the PM_{2.5} concentration has a negative feedback effect on the SSD.

Interactions between Precipitation and PM2.5. On one hand, previous studies (Tai et 441 442 al., 2010) show that an increase in precipitation causes a decrease in all PM25 443 components through scavenging. On the other hand, the influence of PM2.5 on 444 precipitation are more complex: PM25 can serve as cloud nuclei influencing 445 precipitation (suppressing the light rain and strengthen the heavy rain) by acting on the 446 size and number of cloud droplets (Rosenfeld et al., 2014). Meanwhile, PM2.5 can also 447 modulate precipitation by changing the atmospheric vertical static stability via the 448 aerosol radiative effect (Jacobson, 2001).

449 Interactions between Temperature and PM2.5: Temperature is one important 450 meteorological factors affecting the transformation of pollutants and the temperature 451 inversion is one major cause for haze episodes in winter. The temperature inversion 452 leads to an unfavorable condition for the dispersion of PM2.5 and an increase of PM2.5 453 concentrations. On the other hand, high PM2.5 concentrations may lead to a stable 454 atmospheric environment, and further improve the temperature inversion phenomenon. 455 Interactions between Air pressure and PM2.5. When the atmospheric environment is 456 controlled by low air pressure, it demonstrates an unstable status and the near-ground 59

457 air is pushed upward, which is favorable for the transportation of airborne pollutants 458 and the reduction of PM2.5 concentrations. On the other hand, high PM2.5 concentrations 459 may lead to the temperature inversion phenomenon, usually accompanied with a stable 460 atmospheric controlled by high air pressure. 461 Interactions between Evaporation and PM2.5: Liu et al (2015) suggested that the loss 462 of PM_{2.5}-concentrations increased with an increase of evaporation. Meanwhile, high PM2.5 concentrations lead to a stable atmospheric environment, in which the 463 464 evaporation rate is low. 465 Interactions between Wind direction and PM2.5: The influence of wind direction on 466 PM2.5 concentrations and its feedback effects is majorly dependent on the geographical 467 conditions and local landscape patterns. For instance, due to the specific geographical 468 conditions surrounded by hills on three sides, northwest wind in Beijing leads to an 469 improvement of air quality whilst southeast wind leads to the accumulation of airborne 470 pollutants. However, the influence of wind direction on PM2.5 concentrations varies 471 significantly in other cities. So the interactions between wind direction and PM25 is 472 context-related. 473 5.2 Understanding the formation mechanisms of haze episodes and improving air 474 quality from a meteorological perspectiveDue to different meteorological conditions 475 and complicated mechanisms of PM2.5-meteorology interactions, the influence of 476 individual meteorological factors on PM2.5 concentrations varied significantly at the 477 national scale. Firstly, notable differences existed in meteorological conditions across 478 China. For instance, in winter, the frequency and intensity of precipitation are much 479 higher and stronger in coastal areas than those in the North China region, where the 480 frequency of strong winds is high in winter. Therefore, precipitation exerts a large 481 influence on PM2.5 concentrations in coastal regions whilst wind is the key influencing 482 factor for PM2.5 concentrations in the North China region in winter. Secondly, the 483 interaction mechanisms between individual meteorological factors and PM2.5 484 concentrations may be influenced significantly by the magnitude of local 485 meteorological factors and PM2.5 concentrations. For instance, heavy precipitation can 486 have a strong washing-off effects on PM2.5 concentrations and notably reduce PM2.5 487 concentrations. Meanwhile, slight precipitation may not effectively remove the high-488 concentration PM_{2.5}. Instead, the slight precipitation may induce enhanced relative

489	humidity,	which leads to	the hygroscopi	c increase and	1 accumulation	of PM _{2.5}	(Fu et al.,

490 <u>2016</u>). In addition to precipitation, He et al. (2017) suggested that such meteorological

factors as temperature and humidity were either positively or negatively correlated with

PM_{2.5} concentrations in different regions of China. Given the complexity of interactions

between meteorological factors and PM_{2.5}, characteristics and variations of influences

494 of individual meteorological factors on PM_{2.5} concentrations should be further

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

- 496 observation data.
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498 With rapidly growing haze events, meteorological influences on PM_{2.5} concentrations

499 have become a hot social-economic topic not only studied by scholars, but also

500 considered by government officials and decision makers. On December 1st, 2016,

501 Beijing published the latest regulations for the prevention and control of

502 meteorological hazards

503 (<u>http://www.bjrd.gov.cn/zt/cwhzt1431/hywj/201612/t20161201_168233.html</u>) and

504 included haze events as one type of meteorological hazards, sparking widespread 505 controversy. Although the meteorological influences on PM2.5 concentrations are well 506 acknowledged, quantifying meteorological contribution, compared with exhaust 507 emission, to airborne pollution remains challenging. Hence, criticisms have been raised 508 that since traffic and industry induced exhaust emission is the main cause for airborne 509 pollution, the emphasis on the meteorological causes for haze hazards is to avoid 510 governmental responsibilities. Some of ourOur previous research may provide 511 reference for a better understanding of this issue from different perspectives. Chen, Z₂ 512 et al. (2016) pointed out that more than 180 days in Beijing experienced notable and 513 sudden air quality change (the Air quality Index, AQI, difference between one day and 514 its previous day is larger than 50) in 2014. Considering that the industrial, automobile 515 and household exhaust emission, which are main sources for PM2.5 and other airborne 516 pollutants, is unlikely to change dramatically in one day, meteorological factors seem 517 to exert an important influence on local PM2.5 concentrations. Chen, Z₂ et al. (2017) 518 proved that such meteorological factors as SSD, wind and humidity exerted strong 519 influences on winter PM_{2.5} concentrations in the Beijing-Tianjin-Hebei Region. 520 Furthermore, Chen, Z. et al. (2017) quantified the interactions between different 521 meteorological factors and suggested that one meteorological factor may influence

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522 PM_{2.5} concentrations through both direct and indirect means. Take winter PM_{2.5} 523 concentrations in Beijing for instance. The wind factor has a strong negative causality 524 influence on PM2.5 concentrations. In addition, the wind factor has a negative causality 525 ondecreases humidity, as well as positive causality onincreases SSD and evaporation. 526 Since the factor humidity (SSD and evaporation) has a strong positive (negative) 527 influence⁷ on local PM_{2.5} concentrations, increasing wind speeds can reduce PM_{2.5} 528 concentrations indirectly through reduced (increased) humidity (SSD and evaporation). 529 In this research, we further revealed that meteorological influences on PM2.5 530 concentrations varied significantly across China. In the most polluted winter, the 531 dominant meteorological factors for PM2.5 concentrations in the North China region are 532 mainly the wind and humidity factor whilst the dominant meteorological factor on PM2.5 533 concentrations in coastal cities are mainly precipitation and temperature. Furthermore, 534 this research proved that the meteorological influences on PM2.5 concentrations were 535 the strongest in winter, when the PM_{2.5} concentrations was the highest. With strong 536 bidirectional coupling between individual meteorological factors and PM2.5 537 concentrations in winter, PM2.5 concentrations can be further enhanced through 538 complicated atmospheric mechanisms, leading to more haze events. Based on these 539 studies, we are not attempting to challenge the fundamental contribution of human-540 induced exhaust emission to PM2.5 concentrations. Instead, our research suggested that 541 with a stable amount of exhaust emission, meteorology was a key factor for the 542 persistence and deterioration of haze events, especially in winter. On one hand, the 543 pollutant emission should be strictly restricted, as human-induced emission is the major 544 cause of haze pollution. Meanwhile, since meteorological factors play an important role 545 in the accumulation and dispersion of PM2.5, meteorological influences should be 546 comprehensively considered for a better understanding and management of haze 547 episodes. 548 In spite of a diversity of prediction models, air quality forecast, especially PM2.5

549 forecasting in China, remains challenging. Commonly used air quality forecast models

50 include CAMx (ENVIRON Company, US), CMAQ (Environmental Protection Agency,

551 US), WRFCHEM (National Center for Atmospheric Research, US) and NAQ PMS

552 (Institute of Atmospheric Physics, Chinese Academy of Sciences, China). Due to

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⁷ Although the CCM method did not give a positive(-negative) direction betweenof interactions between two variables, the direction of interactions can be easily understood according to the correlation coefficient. 62

553 highly complicated atmospheric environment and the difficulty in acquiring true data 554 of exhaust emission, these commonly used models (e.g. CAMx, CMAQ and 555 WRFCHEM) may lead to large biases and uncertainty when applied to China. On the 556 other hand, without prioriprior knowledge of mechanisms of haze formation and 557 information of exhaust emission, statistical models can achieve satisfactory forecasting 558 results based on massive historical data (Cheng et al., 2015). However, Cheng et al. 559 (2015) pointed out that most Compared with the static models, dynamic statistical 560 models were static model and did not comprehensively consider the meteorological 561 influences on airborne pollutants. Even if some models consider PM2.5 concentrations 562 and some meteorological factors that are of stable, representative and strong 563 correlations with PM_{2.5} are selected for forecasting PM_{2.5} concentrations. Meanwhile, 564 many recent studies (Cheng et al., 2017; Guo et al., 2017; Lu et al., 2017; Ni et al. 2017; 565 etc) have recognized the meteorological influences on PM2.5 concentrations, they only 566 employthe evolution of PM2.5 concentrations and included some key meteorological 567 factors in their models for PM2.5 estimation. However, most PM2.5 estimation and 568 forecasting models mainly employed correlation analysis, which has been proved to 569 problematic in reveal the influence of individual meteorological factors on PM2.5 570 concentrations. Due to complicated interactions in atmospheric environment-, the 571 correlation coefficient between meteorological factors and PM2.5 concentrations is usually much larger than the ρ value and overestimates the influence of individual 572 573 meteorological factors on PM2.5 concentrations. In this case, this research provides 574 useful reference for improving existing statistical models. The P-value is a better 575 indicator than the correlation coefficient to demonstrate the quantitative influence of 576 individual meteorological factors on local PM2.5 concentrations. By incorporating the 577 $-\frac{\rho}{\rho}$ value, instead of the correlation coefficient, of different factors into corresponding 578 GAM (Generalized Additive Models) and adjusting parameters accordingly, we 579 canmay significantly improve the reliability of future estimation and forecasting of 580 PM_{2.5} concentrations.

581 With the understanding of strong meteorological influences on PM_{2.5} concentrations 582 across China, especially in some heavily polluted regions, decision makers are placing 583 special emphasis on improving local and regional air quality through meteorological 584 means. Targeting this, quantified causality<u>influence</u> of individual meteorological 585 factors on PM_{2.5} concentrations provides useful decision support for evaluating relevant

586 environmental projects. Specifically, a forthcoming Beijing wind-corridor project

587 (http://www.bj.xinhuanet.com/bjyw/yqphb/2016-05/16/c_1118870801.htm)

588become a hot social and scientific issue, yet its potential effects arouse wide589controversies.Somescholars

590 (http://china.cnr.cn/yxw/201411/t20141123_516839830.shtml

591 http://health.people.com.cn/n1/2016/0413/c398004-28271979.html) pointed out that 592 the wind-corridor project could only exerted limited influence on the reduction of PM2.5 593 concentrations and major efforts should be made on emission-reduction. Herein, our 594 research suggests that wind is a dominant meteorological factor for winter PM2.5 595 concentrations in Beijing and can significantly influence PM_{2.5} concentrations through 596 direct and indirect mechanisms. In consequence, the wind-corridor project may directly 597 allow in more strong wind, which thus leads to a larger value of SSD and EVP and a 598 smaller value of RHU. The change of SSD, RHU and EVP values can further induce the 599 reduction of PM2.5 concentrations. From this perspective, the Beijing wind-corridor 600 project has good potential to improve local and regional air quality. In addition to the 601 wind-corridor project, some scholars and decision makers have proposed other 602 meteorological means for reducing PM_{2.5} concentrations. For instance, Yu (2014) 603 suggested that water spraying from high buildings and water towers in urban areas was 604 an efficient way to reduce PM2.5 concentrations rapidly by simulating the process of 605 precipitation. However, some limitations, such as the humidity control and potential 606 icing risk, remained. In the near future, with growing attention on the improvement of 607 air quality, more environmental projects should be properly designed and implemented. 608 According to this research, meteorological influences on PM_{2.5} concentrations vary 609 notably across China. Considering the diversity of dominant meteorological factors on 610 local PM2.5 concentrations in different regions and seasons, it is more efficient to design 611 meteorological means accordingly. For the heavily polluted North China region in 612 winter, meteorological means for encouraging strong winds are more likely to reduce 613 PM_{2.5} concentrations considerably whilst meteorological means for inducing precipitation are more likely to improve air quality in coastal cities and cities within the 614 615 Yangtze River basin.

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616 6 Conclusions

617 Based on the CCM method, we quantified the causality influence of eight 618 meteorological factors on local PM2.5 concentrations for 189188 monitoring cities 619 across China. The results suggest that meteorological influences on PM2.5 are of notable 620 seasonal and spatial variations. For most citiesFor the heavily polluted North China 621 region, the higher PM_{2.5} concentrations, the stronger influence meteorological factors 622 exert on local PM2.5 concentrations. The dominant meteorological factor for PM2.5 623 concentrations is closely related to geographical conditions. For heavily polluted winter, 624 precipitation exerts a key influence on local PM2.5 concentrations in most coastal areas 625 and the Yangtze River basin, whilst the dominant meteorological driver for PM2.5 626 concentrations is wind in the North China regions. At the national scale, the influence 627 of temperature, humidity, and wind and air pressure exert stronger influences on on 628 local PM2.5 concentrations is much larger than that of other factors-, and temperature 629 exerts the strongest and most stable influences on national PM2.5 concentrations in all 630 seasons. The-causality influence of individual meteorological factors on PM2.5 631 concentrations extracted in this research provides more reliable reference for better 632 modelling and forecasting local and regional PM2.5 concentrations. Given the 633 significant variations of meteorological influences on PM2.5 concentrations across 634 China, environmental projects aiming for improving local air quality should be 635 designed and implemented accordingly. 636 Acknowledgement This research is supported by National Natural Science Foundation of China (Grant 637 638 Nos. 210100066), the National Key Research and Development Program of China

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- 641 Project for excellent scholars (2015000020124G059).

References

 Blanchard, C., Hidy, G., Tanenbaum, S., 2010. NMOC, ozone, and organic aerosol in the southeastern United States, 1999-2007: 2. Ozone trends and sensitivity to NMOC emissions in Atlanta, Georgia. Atmospheric Environment. 44 (38), 4840e4849.

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- Cao, C., Jiang, W., Wang, B., Fang, J., Lang, J., Tian, G., Jiang, J., Zhu, T. 2014.Inhalable Microorganisms in Beijing's PM2.5 and PM10 Pollutants during a Severe Smog Event. Environmental Science and Technology. 48, 1499–1507.
- Cao, J., Shen, Z., Chow, J., Watson, J. G., Leed, S., Tie, X., Ho, K., Wang, G., Han, Y., 2012, Winter and Summer PM_{2.5} Chemical Compositions in Fourteen Chinese Cities. Journal of the Air & Waste Management Association. 62(10), 1214-1226.
- Chen, W., Zhang, H.T., Zhao, H. M. 2015. Diurnal, weekly and monthly spatial variations of air pollutants and air quality of Beijing. Atmospheric Environment. 119. 21-34.
- Chen, Y., Schleicher, N., Fricker, M., Cen, K., Liu, X.L., Kaminski, U., Yu, Y., Wu, X.F., Norra, S. 2016. Long-term variation of black carbon and PM_{2.5} in Beijing, China with respect to meteorological conditions and governmental measures. Environmental Pollution. 212, 269-278.
- 6.<u>1.-Chen, Z.Chen, Z.Y., Xu, B., Cai, J., Gao, B.B. 2016. Understanding temporal</u> patterns and characteristics of air quality in Beijing: A local and regional perspective. Atmospheric Environment. 127, 303-315.
- 7.6. Chen, Z.-Y., Cai, J., Gao, B.B., Xu, B., Dai, S., He, B., Xie, X.M., 2017. Detecting the causality influence of individual meteorological factors on local PM2.5 concentrations in the Jing-Jin-Ji region. Scientific Reports, 7.
- 7. Chen, Z.Y., Xu, B., Cai, J., Gao, B.B. 2016. Understanding temporal patterns and characteristics of air quality in Beijing: A local and regional perspective. Atmospheric Environment. 127, 303-315.
- Cheng, N.L., Li, J.J., Li, Y.T., Sun, F. 2015 Development of PM2.5 dynamic partitioning statistical prediction model based on Matlab in Beijing (in Chinese). Chinese Journal of Environmental Engineering. 9(10), 4965-4970.
- 9. Cheng, Z., Li, L., & Liu, J. (2017). Identifying the spatial effects and driving factors of urban pm 2.5, pollution in china. Ecological Indicators, 82, 61-75.
- 9.10. El-Metwally, M., Alfaro, S.C. 2013. Correlation between meteorological conditions and aerosol characteristics at an East-Mediterranean coastal site. Atmospheric Research. 132–133, 76–90.
- 11. Fu, X., Wang, X., Hu, Q., Li, G., Xiang, D., Zhang, Y., et al. (2016). Changes in visibility with pm2.5 composition and relative humidity at a background site in the pearl river delta region. Journal of Environmental Sciences, 40(2), 10-19.

- 10.12. Galindo, N., Varea, M., Moltó, J.G., Yubero, E. Nicolás, J. 2011. The Influence of Meteorology on Particulate Matter Concentrations at an Urban Mediterranean Location. Water Air Soil Pollution.215, 365–372.
- 13. Garrett, P., Casimiro, E., 2011. Short-term effect of fine particulate matter (PM2.5) and ozone on daily mortality in Lisbon, Portugal. Environmental Science and Pollution Research. 18(9), 1585-1592.
- H-14. Granger, C. W. J. 1980. Testing for causality: A personal viewpoint. Journal of Economic Dynamics and Control. 2, 329-352.
- 12.15. Grundstrom, M., Hak, C., Chen, D., Hallquist, M., Pleije, H. 2015. Variation and co-variation of PM₁₀, particle number concentrations, NOx and NO₂ in the urban air- Relationships with wind speed, vertical temperature gradient and weather type. Atmospheric Environment. 120, 317-327.
- 13.1. Garrett, P., Casimiro, E., 2011. Short term effect of fine particulate matter (PM2.5) and ozone on daily mortality in Lisbon, Portugal. Environmental Science and Pollution Research. 18(9), 1585–1592.
- 14.16. Gu, J., Du, S., Han, D., Hou, L., Yi, J., Xu, J., Liu, G., Han, B., Yang, G., Bai, Z., 2014. Major chemical compositions, possible sources, and mass closure analysis of PM_{2.5} in Jinan, China. Air Quality, Atmosphere & Health. 7(3), 251-262.
- 15.17. Guaita, R., Pichiule, M., Maté, T., Linares, C., Día, J., 2011. Short-term impact of particulate matter (PM_{2.5}) on respiratory mortality in Madrid. International Journal of Environmental Health Research. 21(4), 260-274.
- 18. Guo, Y., Tang, Q., Gong, D. Y., & Zhang, Z. (2017). Estimating ground-level pm 2.5, concentrations in beijing using a satellite-based geographically and temporally weighted regression model. Remote Sensing of Environment, 198, 140-149.
- 16.19. Guo, S., Hu, M., Guo, Q., Zhang, X., Zheng, M., Zheng, J., Chang, C., Schauer, J.J., Zhang, R. Y. 2012. Primary Sources and Secondary Formation of Organic Aerosols in Beijing, China. Environmental Sciences & Technology, 46, 9846–9853.
- 20. He, J., Gong, S., Ye, Y., Yu, L., Lin, W., Mao, H., et al. (2017). Air pollution characteristics and their relation to meteorological conditions during 2014 - 2015 in major chinese cities. Environmental Pollution, 223, 484-496.
- Hu, J. Qi, Y., Wang, Y., Zhang, H. 2015. Characterizing multi-pollutant air pollution in China: Comparison of three air quality indices. Environment 67

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International, 2015, 84:17-25.

- 18.22. Jacobson, M. Z. 2001. Global direct radiative forcing due to multicomponent anthropogenic and natural aerosols. Journal of Geophysical Research Atmospheres, 106(D2), 1551-1568.
- 19.23. Juneng, L., Latif, M.T., Tangang, F. 2011. Factors influencing the variations of PM₁₀ aerosol dust in Klang Valley, Malaysia during the summer. Atmospheric Environment. 45, 4370-4378.
- 20.24. Kong, L.B., Xin, J.Y., Zhang, W.Y., Wang, Y.S. 2016. The empirical correlations between PM_{2.5}, PM₁₀ and AOD in the Beijing metropolitan region and the PM_{2.5}, PM₁₀ distributions retrieved by MODIS. Environmental Pollution. 216, 350-360.
- 21.25. Lanzinger, S., Schneider, A., Breitner, S., Stafoggia, M., Erzen, I., Dostal, M. et al. 2015. Associations between ultrafine and fine particles and mortality in five central European cities Results from the UFIREG study. Environment International. 88(2): 44-52.
- 22.26. Li, Y., Ma, Z., Zheng, C., Shang, Y., 2015. Ambient temperature enhanced acute cardiovascular-respiratory mortality effects of PM2.5 in Beijing, China International Journal of Biometeorology. 10.1007/s00484-015-0984-z
- 23.27. Liu, Q.Y., Baumgartner, J., Zhang, Y., Liu, Y., Sun, Y., Zhang, M. 2014. Oxidative Potential and Inflammatory Impacts of Source Apportioned Ambient Air Pollution in Beijing. Environmental Sciences & Technology. 48, 12920–12929.
- 24.28. Liu, C. N., Lin, S. F., Tsai, C.Lu, D., Xu, J., Wu, Y. C., Chen, C. F. 2015. Theoretical model for the evaporation lossYang, D., & Zhao, J. (2017). Spatiotemporal variation and influence factors of pm 2.5, during filter sampling. concentrations in china from 1998 to 2014. Atmospheric Environment, 109, 79-86Pollution Research.
- 25.29. Luo, C., Zheng, X., Zeng, D. 2014. Causal Inference in Social Media Using Convergent Cross Mapping. IEEE. Intelligence and Security Informatics Conference. 260-263.
- 26.30. Ma, Z., Hu, X., Huang, L., Bi, J., Liu, Y., 2014. Estimating Ground-Level PM_{2.5} in China Using Satellite Remote Sensing. Environmental Science & Technology. 48 (13), 7436–7444.
- 31. Ni, X. Y., Huang, H., & Du, W. P. (2017). Relevance analysis and short-term

prediction of pm2.5 concentrations in beijing based on multi-source data. Atmospheric Environment, 150, 146-161.

- 27.32. Pasca, M., Falq, G., Wagner, V., Chatignoux, E., Corso, M., Blanchard, M., Host, S., Pascala, L., Larrieua,S., 2014. Short-term impacts of particulate matter (PM₁₀, PM_{10-2.5}, PM_{2.5}) on mortality in nine French cities. Atmospheric Environment. 95, 175–184.
- 28.33. Pearce, J.L., Beringer, J., Nicholls, N., Hyndman, R.J., Tapper, N.J., 2011. Quantifying the influence of local meteorology on air quality using generalized additive models. Atmospheric Environment. 45, 1328-1336.
- 29.34. Qiao, L.P., Cai, J., Wang, H.L., Wang, W.L., Zhou, M., Lou, S.R., Chen, R.J., Dai, H.X., Chen, C.H., Kan, H.D. 2014. PM_{2.5} Constituents and Hospital Emergency-Room Visits in Shanghai, China. Environmental Science and Technology. 48 (17), 10406–10414.
- Rosenfeld, D., Andreae, M. O., Asmi, A., Chin, M., Leeuw, G., & Donovan, D. P., et al. 2014. Global observations of aerosol - cloud - precipitation - climate interactions. Reviews of Geophysics...
- 31.35. Shen, G., Yuan, S., Xie, Y., Xia, S., Li, L., Yao, Y., Qiao, Y., Zhang, J., Zhao, Q., Ding, A., Li,B., Wu, H. 2014. Ambient levels and temporal variations of PM_{2.5} and PM₁₀ at a residential site in the mega-city, Nanjing, in the western Yangtze River Delta, China. Journal of Environmental Science and Health, Part A: Toxic/Hazardous Substances and Environmental Engineering. 49(2), 171-178.
- 32.-36. Sugihara, G., May, R. 1990. Nonlinear forecasting as a way of disting uishing chaos from measurement error in time series. Nature, 344(6268), 7 34–741.
- 33.37. Sugihara, G., May, R., Ye, H., Hsieh, C., Deyle, E., Fogarty, M., Munch, S. 2012. Detecting Causality in Complex Ecosystems. Science, 338, 496-500.
- 34. Tai, A. P. K., Mickley, L. J., Jacob, D. J. 2010. Correlations between fine particulate matter (pm2.5) and meteorological variables in the united states: implications for the sensitivity of pm_{2.5} to climate change. Atmospheric Environment, 44(32), 3976–3984.
- 35.38. Wang, G., Cheng, S., Li, J., Lang, J., Wen, W., Yang, X., Tian, L. 2015. Source apportionment and seasonal variation of PM_{2.5} carbonaceous aerosol in the Beijing-Tianjin-Hebei Region of China. Environmental Monitoring and Assessment.

10.1007/s10661-015-4288-x.

- 36.39. Wang, G., Cheng, S., Li, J., Lang, J., Wen, W., Yang, X., Tian, L. 2015. Source apportionment and seasonal variation of PM_{2.5} carbonaceous aerosol in the Beijing-Tianjin-Hebei Region of China. Environmental Monitoring and Assessment. 10.1007/s10661-015-4288-x.
- 37.40. Wei, S., Huang, B., Liu, M., Bi, X., , Ren, Z.F., Sheng, G., Fu, J. 2012. Characterization of PM_{2.5}-bound nitrated and oxygenated PAHs in two industrial sites of South China. Atmospheric Research. 109-110, 76-83.
- 38.41. Yadav, R., Beig, G, Jaaffrey, S.N.A. 2014. The linkages of anthropogenic emissions and meteorology in the rapid increase of particulate matter at a foothill city in the Arawali range of India. Atmospheric Environment. 85, 147-151.
- 39.42. Yang,Y., Christakos, G. 2015. Spatiotemporal Characterization of Ambient PM2.5 Concentrations in Shandong Province (China). Environmental Sciences & Technology. 49 (22), 13431–13438.
- 40.43. Yu, S.C. 2014. Water spray geoengineering to clean air pollution for mitigating haze in China's cities. Environmental Chemistry Letters. 12(1), 109–116.
- 41.44. Zhang, F., Wang, Z., Cheng, H., Lv, X., Gong, W., Wang, X., Zhang, G., 2015, Seasonal variations and chemical characteristics of PM_{2.5} in Wuhan, central China. Science of The Total Environment. 518, 97–105.
- <u>45.</u> Zhang, H. F., Wang, Z. H., Zhang, W. Z. 2016. Exploring spatiotemporal patterns of PM_{2.5} in China based on ground-level observations for 190 cities. Environmental Pollution. 89-90, 212-221.
- 42.46. Zhang, H., Wang, Y., Hu, J., Ying, Q., Hu, X. 2015b. Relationships between meteorological parameters and criteria air pollutants in three megacities in China. Environmental Research, 140, 242-254.
- 43.47. Zhang, R., Jing, J., Tao, J., Hsu, S., C., Wang, G., Cao, J., et al., 2013. Chemical characterization and source apportionment of PM2.5 in Beijing: seasonal perspective. Atmospheric Chemistry and Physics, 13, 7053-7074.
- 44.<u>1.</u>Zhang, H. F., Wang, Z. H., Zhang, W. Z. 2016. Exploring spatiotemporal patterns of PM_{2.5} in China based on ground level observations for 190 eities. Environmental Pollution. 89-90, 212-221.
- <u>45.48.</u> Zhen, C, Luo L, Wang S, Wang, Y., Sharma, S., Shimadera, H., Wang, X., et al. 2016. Status and characteristics of ambient PM_{2.5} pollution in global megacities.
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Environment International. 89-90, 212-221.