# Response to reviewers

**Dear Professor Xavier Querol:**

**Thanks so much for providing us a chance to revise and resubmit our manuscript. In the past weeks, we have fully revised the manuscript according to all the general and detailed comments raised by two reviewers. The English has been fully polished. Most figures have been reproduced. And the structure and some other issues were also carefully addressed. And for some limitations raised by the reviewers, we have provided detailed explanation in the responses letter and the revised manuscript.**

**Please feel free to contact us if additional revisions are requested and we are more than willing to conduct further revisions.**

**The very best**

**Ziyue**

**To Reviewer 1:**

**Thanks so much for providing us some many general and detailed comments, which helped us so much improve this manuscript. We have fully advised this manuscript according to your constructive comments. We are more than willing to conduct further revisions if you have additional comments.**

**Thanks again for your time and help.**

The article shows an innovative methodology to evaluate the effect of meteorological temporal scales on the pollutant-meteorology association. The collection of a large amount of data on measured pollutant concentrations and meteorological variables makes its results relevant and interesting to address the problem of air pollution in China, more specifically where most of its population is concentrated. The methodology is also interesting for the rest of the scientific community as it is a methodology applicable to other regions.

Although the innovative part of the methodology is clear, it is not clear what is the innovative value of the results obtained. Some of the results seem somewhat obvious or certainly expected, for example, the relationship of temperature with the pollutants analyzed, or a greater association of ozone to the temporal scale of 3h, given its dependence on temperature and solar radiation (in this article not studied as already mentioned by the authors), or the high relationship in winter with precipitation (due to higher frequency of bad weather?). However, the results are no less important for being obvious, but they are shown as if it were a mere descriptive report and there is a lack of greater depth in the analysis of the results and their possible applicability in future strategies for air quality control.

**R: Thanks so much for pointing this out. Yes, we mentioned that temperature was the dominant factor for PM and ozone generally at the national scale, which was clear in our previous studies. However, this was not the major conclusion and findings of this research. As stressed in the manuscript, the major innovation and findings of this research was the temporal scale played an important role in the extracted pollutant-meteorology association. Specifically, despite a generally strong influence of temperature, we found that the number of cities with temperature as the dominant meteorological and the mean ρ value of temperature presented notable differences at the 3h and 24h scale. The difference of pollutant-meteorology association at different temporal scales was not investigated quantitatively before and was the major innovation and contribution of this research.**

**Thanks so much for your comment. In the revised manuscript, we further pointed out that the dominant temperature was clear in previous studies and the temporal effects on revealed difference of pollutant-meteorology association was the new knowledge to the field. In this case, the major innovation of this research was clearly highlighted.**

1. In order to improve this analysis, I propose a series of improvements for the authors to consider:

The reference year studied should appear in the abstract: 2020.

**R: Thanks so much for this comment. We have corrected it accordingly in the revised manuscript.**

2. The structure of the article could be clearer if section 2 were titled Methodology, and 2.2. Advanced Causation Model.

Following the latter, it would be clearer to join section 3 and section 4, since the way the article is presented, it is difficult to follow the relationship between the results and the authors' discussion of them. It would be advisable to make the discussion at the same time as the results are shown.

**R: Thanks so much for this comment. It's a very good suggestion and we have corrected it accordingly in the revised manuscript.**

3. In the introduction, the sentence starting on line 46 "Zhai et al. (2019) estimated... for PM2.5 variations" is not understood.

**R: Thanks so much for this. Here we meant that Zhai et al. (2019) estimated the correlation between $PM_{2.5}$ concentration and meteorological factors at the 10-day scale, found that the variation trend of $PM_{2.5}$ and $SO_2$, $NO_2$ and CO was consistent, also $SO_2$ emission-control was the main driving factor for $PM_{2.5}$ variations. This result was inconsistent with findings from other studies. In the revised manuscript, we have corrected this to make this part clearer.**

4. The introduction should briefly mention why the 3h time scale was chosen and not another. This question is also not addressed in the methodological part of the article. It would also be advisable

to add in the introduction what the purpose of this study is, beyond evaluating the methodology and its results, what future applicability it has, or why this study's results are interesting or innovative.

**R: Thanks so much for pointing this out. This is a good point. Currently, most research concerning pollutant-meteorology association was conducted based on a daily basis (24 hour). This is because meteorology data with a higher temporal resolution is not available. Due to the lack of data sources, there is no research conducted on the temporal effects on the pollutant-meteorology association.**

**We recently obtained the 3h meteorological data sources from China Meteorological Administration, which was so far the meteorological data set with the highest temporal resolution. When hourly data set is not being produced now, we can consider the 3h meteorological data source was the existing data set with the highest temporal scale. Although 3h meteorological source has yet been publically available now, it will become available in the future. In that case, the resolution of 3h and daily (24h) will become the most frequently data set for most researchers. Meanwhile, the comparison between 3h and 24h data set presented a strong difference, indicating the temporal effects on pollutant-meteorology association was strong. With the growing availability of long-term meteorological and pollutant data, multi-scale research is recommended to comprehensively understand the short- and long-term meteorological influences on different airborne pollutants.**

**What you mentioned here was very important. In the revised manuscript, we added relevant explanation.**

5. Regarding the first paragraph of section 2.1. Data sources: Why was the year 2020 selected, is it a question of data availability? Since an annual seasonal study is conducted, how did the emissions change over the year due to the lockdown? This should be stated clearly. Additionally, when analyzing or grouping stations, have any criteria been used to discard any stations?, for example, some with high industrial character or downwind of an industry, or less exposure to the population, or have all of them been taken into account?

**R: Thanks so much for this. As we acknowledged, the 3h meteorological data set was a very rare data set and unfortunately, we did not have a long-term data series with such high temporal-resolution and only got the data set for 2020. This was admitted in the discussion part and we suggested that we would like to explore the multi-year variations of pollutant-meteorology associations. For now, no other studies, to our best knowledge, have conducted analysis using data sets with temporal resolution better than daily. However, the one year data set for this research, including four seasons (four complete time series with more than 90 records (24h) and 720 records (3h)), was sufficient for conducting this comparison analysis. CCM is sensitive to weak to moderate coupling in ecological time series and can effectively extract the association between two variables based on more than 30 records. Therefore, CCM was an ideal tool for conducting this comparison research. Secondly, yes, you are right, the COVID-19 in 2020 may cause may cause a difference in the extracted pollutant-meteorology association. However, by comparing the output of this research with**

**our previous studies, the extracted p value was slightly smaller in 2020, yet the general trend was highly consistent with previous studies. Since the major aim of this research was to examine whether temporal scales can cause a difference on extracted pollutant-meteorology association (and it is effectively proved by this research), the specialty of 2020 caused limited influence on the outputs.**

**Thanks again for your comment more information on the availability, advantages, and limitations of this data set has been added to the revised manuscript, which helped us improve this manuscript.**

6. How are the location of the 101 cities relevant? Are they the ones shown on the maps in the figures? Do they have any geographic biases that might be relevant to the study, such as different climate zones influencing the results?

**R: Thanks so much for this. The locations of 101 cities are not correlated, they are the ones shown on the maps in the figures. Here we considered all the available cities in this data set, which can demonstrate the overall influence of temporal effects on the extraction of pollutant-meteorology association. Amongst the 101 cities, half of them demonstrated notable difference of dominant meteorological factors between the 3h and 24h scale. So the inclusion of as many as cities across China can effectively remove the potential bias caused by specific regions and highlight the strong temporal effects on pollutant-meteorology association.**

7. It is understood that data used is based on hourly measurements, but they are not mentioned until line 78, is that so?

**R: Thanks so much for this. For this research, the available meteorological data of 3h and 24h were collected respectively. Meanwhile, since the publicly released pollution data was hourly based, to match the temporal scale of meteorological data, the per-3h and per-24h pollutant data were produced by conducting average operation on hourly concentration data.**

8. In section 3.1., moving on to the results, the tables are referenced all at once, they must be referenced one by one as the specific results of each of them are discussed. How relevant are the results shown in this section? The influence in winter is discussed, is winter one of the seasons of concern in terms of pollution problems? And if so, what do these results imply?

**R: Thanks so much for this. This is a very good point. We have corrected it accordingly in the revised manuscript. For all three airborne pollutants, the dominant meteorological factor at the 3h and 24h scale was the same in only a third of cities, indicating the temporal scale played a large role in the analysis of pollutant-meteorology association. From the seasonal scale, the consistence between dominant meteorological factors extracted at 3h and 24h in autumn and winter was higher than that in spring and summer. This phenomenon indirectly suggested that meteorological influences on airborne pollutants were stronger in autumn and winter, and thus the role of dominant meteorological factor was highlighted and notably larger than other factors.**

**Thanks again for this comment. According to this, this part has been largely improved.**

9. In section 3.2. it is mentioned that the relationship between PM and $O_3$ is different, and likewise for the meteorological variables, have the authors identified any dominating meteorological driver that might not be expected or so obvious?

**R: Thanks so much for this. This is a very interesting point. Actually, yes, there seemed some unexpected outputs here. We originally thought that the extracted Pollutant-Meteorology association was always stronger at 3h scale. Interestingly, we found that for some meteorological factors, this was not the same as we expected. For instance, we thought precipitation could exert a stronger influence on PM at 3h scale. However, actually, we found the influence of precipitation on PM was stronger at 24h scale. We assume this was attributed to the characteristics of recording the amount of precipitation at different temporal scales. At the 3h scale, the amount of precipitation at some time slots may be too limited to record, which influenced the calculation outputs.**

10. In section 3.3. the spatial distribution of the dominant meteorological variables is discussed, as in section 3.1., it is necessary to reference each figure and to highlight the relevant results of each one of them.

**R: Thanks so much for pointing this out. We have corrected it accordingly in the revised manuscript.**

11. In line 189 of section 3.3, "heavily polluted summer" is mentioned, does this happen for the whole territory? and is it supposed to be in the areas where a higher concentration of ozone has been plotted? Line 194 talks about "not severe and homogeneous", where readers can observe this?

**R: Thanks so much for this. In summer, ozone pollution is worse than in other seasons. This is because high-temperature and low-humidity are the major driver for ozone pollution, which is often observed in summer. And this scenario is applied to the entire China, as revealed by many previous studies.**

**For PM pollution across China, we generally considered that it was most severe in the three major city-clusters, Beijing-Tianjin-Hebei region, Yangtze-River Delta and Pearl-River Delta. For other regions, especially those inland regions, PM pollution was relatively low and presented less heterogeneous distribution.**

**Thanks so much for your comment. We have revised it accordingly.**

12. Section 3.3. should be accompanied by an indication on the map of "Yangtze River Delt" and "Shandong Peninsula" locations.

**R: Thanks very much for this comment. These figures have been reproduced accordingly.**

13. Regarding section 3.3., the figures (maps) associated with this section are not in the same color scale (for concentrations), nor do they indicate what these concentrations are, the hourly average for each season?

**R: Thanks very much for this comment. Actually, the color bar was set for each season respectively. So the concentration map is only drawn for each season, without overall unification. And these concentrations are the hourly average for each season.**

14. In the first paragraph of Section 4, the limitations of the method are mentioned, it would be more advisable to mention it in the methodology. Lines 211 to 213 reinforce the potential of the study that should have been mentioned previously.

**R: Thanks so much for this. We have corrected it accordingly in the revised manuscript.**

15. The sentence starting on line 225 is not understandable.

**R: Thanks so much for this. Here it means that, Both from a qualitative perspective (the identification of dominant meteorological factors for pollutants) and quantitative perspective (the details of p value of individual meteorological factors on pollutants), the extracted pollutant-meteorology association at 3h and 24h presented large differences, indicating strong temporal effects on pollutant-meteorology associations.**

**Thanks again for your comment. We have revised this part accordingly.**

16. Line 235 talks about "heavily polluted and less polluted seasons", but at no point does it mention which is which.

**R: Thanks so much for this comment. Heavily polluted and less polluted seasons are indicated in the revised version.**

17. The sentence starting on line 237 and ending in 240 should be included into results.

**R: Thanks so much for this. Actually, we have already listed some similar outputs in the result parts "For $PM_{2.5}$ and $PM_{10}$, the calculated influence of temperature at 24h scale was consistently larger than that at the 3h scale. For the relatively cold season winter and spring, when $O_3$ concentrations were relatively low, the influence of temperature at 24h scale was larger than that at the 3h scale. For summer, when $O_3$ concentrations were the highest, the influence of temperature at 3h scale was much larger than that at the 24h scale".**

18. The paragraph starting on line 248 could go to the introduction or part to the method, it does not add value to the discussion of the results.

**R: Thanks so much for this. According to your comment, we have moved this part to the introduction section.**

19. The conclusions in Section 5 are succinct and the results here exposed are clear, although there is no mention of the spatial distribution of the results. This section may be a reference for the authors to improve sections 3 and 4 and to highlight or develop these relevant aspects that are mentioned here.

**R: Thanks so much for this. We have corrected it accordingly in the revised manuscript.**

20. In line 289 "the secondary reaction of which was relatively slow" is not understood.

**R: Thanks so much for this. This means "the secondary reaction between precursors was relatively slow".**

21. Some comments on the wording:

-In "extracted pollutant-meteorolgy", what is meant by "extracted"?

**R: Thanks so much for this. It can also be interpreted as extracted pollutant-meteorology (using CCM).**

The h in 24 is missing: 24h

**R: Corrected. Thanks so much for pointing this out.**

What does the "composite" of "composite airborne pollution" mean?

**R: Thanks so much for this. Composite airborne pollution is a commonly used term, which means the airborne pollution was caused by a variety of atmospheric pollutants.**

Line 87, the authors mean "complex ecosystems"? Instead of "complicated ecosystems"

**R: Corrected. Thanks so much for pointing this out.**

Line 87, what does casual influence mean?, do the authors mean causal influence?

**R: Corrected. Thanks so much for pointing this out.**

Line 92, what does "mirage" correlation mean?

**R: Thanks so much for this. Mirage correlation means the correlation between two variables was not because there are actual causation between them. Instead, there are no causation, and the calculated correlation between them was caused by an agent variable, which was correlated with both of them. Since CCM was the optimal model to remove the influence of other meteorological factors, the mirage correlation calculated using correlation analysis would be identified by CCM.**

Line 103, there is a typo.

**R: Corrected. Thanks so much for pointing this out.**

The figures are in low resolution, and some symbols are less visible than others, for example the one for temperature.

**R: Thanks so much for pointing this out. The low resolution was caused by the image compression during the uploading process. We have solved it by inserting images with higher resolution.**

**To Reviewer 2:**

Assessing the association between the concentrations of multiple airborne pollutants and driving factors is important for identifying the underlying mechanisms for explaining air pollutants' variations. Xu et al. investigate the effects of temporal scales on the identification of dominant meteorological factors for PM and ozone levels across China in 2020. The results showed that temperature is the most critical meteorological factor at both 3h and 24h scales and pollutant-meteorology associations are in a higher degree of agreement in highly polluted regions.

This work is a good contribution that is meaningful and fills some knowledge gaps to understand the influence of temporal scales in the attribution of airborne pollution in China. I would be glad to see its publication, yet there are still some questions, as elaborated in the article. Please correct and clarify them, which will make this manuscript more reasonable and better present the effects.

**R: Thanks so much for all your constructive remarks and useful suggestions, which has significantly raised the quality of the manuscript. We have addressed the issues you raised in the response letter and the revised manuscript. By clarifying the issues you suggested, the manuscript has been largely improved. Thanks again for all your encouragement and valuable comments.**

**Please feel free to contact us if additional revisions are required and we are more than willing to conduct further revisions according to your comments.**

**Specific comments:**

1. How to extract the reliable association between airborne pollutants and meteorological factors is the key to revealing the temporal efforts on pollutant-meteorology causation, and the selection of robust methods is crucial. Therefore, why CCM is suitable for this research and other models not suitable for such analysis should be clearly explained. In the current form, authors have briefly introduced CCM, yet its principle and advantage remained unclear to me. Please elaborate on the advantages and limitations of CMM model and the advantages and rationality of the CMM model compared with other mainstream models.

**R: Thanks so much for this comment. According to our recent model-comparison paper (Chen et al. 2022), CCM may be the most suitable model for causal inference of atmospheric environment. Theoretically, firstly, CCM is specifically designed to deal with the nonlinear relationship between two variables and is fully suitable for the nonlinear relationship between atmospheric factors. Secondly, CCM automatically considers all possible interaction forms and lag effects between the time series of two variables, which effectively reduces the influence of interference and avoids the influence of other factors. Third, CCM requires less parameter setting and prior knowledge, eliminating the uncertainty caused by improper parameter setting. Therefore, CCM model was an ideal tool for this research.**

**The relevant references and explanation has been added to the revised manuscript. Thanks so much for this comment, which improved the rationality of this research significantly.**

*Chen, Z., Xu, M., Gao, B., et al. Causation inference in complicated atmospheric environment. Environmental Pollution, 2022, 303, 119057.*

2. Some further details in the Discussion section can help explain the motivation and main findings of this study. In particular, some discussions on the related works revealed different dominant meteorological factors when the temporal scale is different. This can re-stress the necessity of considering temporal scales.

**R: As stressed in the manuscript, the major innovation and findings of this research was the temporal scale played an important role in the extracted pollutant-meteorology association. Specifically, despite a generally strong influence of temperature, we found that the number of cities with temperature as the dominant meteorological and the mean ρ value of temperature presented notable differences at the 3h and 24h scale. The difference of pollutant-meteorology association at different temporal scales was not investigated quantitatively before and was the major innovation and contribution of this research.**

**Thanks so much for your comment. In the revised manuscript, we further pointed out that the major meteorological elements of air pollutants was clear in previous studies and the temporal effects on revealed difference of pollutant-meteorology association was the new knowledge to the field. In this case, the major innovation of this research was clearly highlighted.**

**Technical comments:**

1. The time of the data (last access date) should be included according to the requirement of ACP.

**R: Corrected. Thanks so much for pointing this out.**

2. The English is understandable, yet with some typos. I suggest the authors carefully read through the manuscript and correct them. Some examples are listed below:

Line 13: Should be "24h".

Line 95: There's a space missing between "(Chen et al., 2022)" and "proved".

Line 103: There's a missing word.

Line 196: Should be "was".

Line 267: Should be "NO2".

The first letter in " (e.g., ) " is not uppercase or lowercase.

**R: Thanks so much for this comment. We have corrected all these typos in the revised manuscript. Meanwhile, we have re-checked the manuscript carefully and polished the English. Thanks again for your comment.**

**List of all relevant changes made in the manuscript:**

Line 14: '$PM_{2.5}$, $PM_{10}$ and $O_3$ concentrations at the 3h and 24 scale' →'$PM_{2.5}$, $PM_{10}$ and $O_3$ concentrations in 2020 at the 3h and 24h scale respectively'.

Line 45: 'between PM2.5 concentration at the 10-day scale and various meteorological factors' → 'between PM2.5 concentration and meteorological factors at the 10-day scale'.

Line 53: 'In recent years, the research on pollutant-meteorology has been massively conducted since 2013 (Chen et al., 2020), yet some gaps remained. Due to the lack of high temporal-resolution data, previous studies were mainly conducted at the daily scale and many scholars may believe that the application of high-temporal-resolution data leads to a better extraction of pollutant-meteorology association.' has been added to the revised manuscript.

Line 62: 'Materials' →'Methodology'.

Line 81: 'Methods' →'Advanced Causation Model'.

Line 89: 'casual' →'causal'.

Line 98: 'CCM is specifically designed to deal with the nonlinear relationship between two variables and is fully suitable for the nonlinear relationship between atmospheric factors.' has been added to the revised manuscript.

Line 104: 'CCM automatically considers all possible interaction forms and lag effects between the time series of two variables, which effectively reduces the influence of interference and avoids the influence of other factors.' has been added to the revised manuscript.

Line 110: 'Based on the rarely employed 3h meteorological data sources, we compared the effects of temporal scales on the extracted pollutant-meteorology causation. Acknowledged, due to the data limitation at the 3h scale, which did not include humidity and sunshine duration, we could simply consider a limited number of meteorological factors (Temperature, Precipitation, Wind Speed, Wind Direction and Atmospheric Pressure), which was less than our previous studies based on meteorological data at the 24h scale, while some meteorological factors (e.g., humility and sunshine duration) were missed in this research. However, since we compared the same set of these major meteorological factors at both 3h and 24h scale, the calculated consistence and difference could effectively reveal the potential effects of different temporal scales on the quantitative (the detailed $\rho$ value) and qualitative (the dominant meteorological factor) findings of pollutant-meteorology association. Despite the limitation of number of meteorological factors, it caused limited influence on the temporal effects on pollutant-meteorology association. This is because CCM simply considers the causality between the target variable and one influencing variable, and removes the influence from other variables (Sugihara et al., 2012; Chen et al., 2020). Another limitation of this data was that this data set simply included one year's data and thus the inter-annual variation of temporal effects on pollutant-meteorology association could not be revealed. For this research, we majorly revealed the existence of strong temporal effects on pollutant-meteorology association, which can be fully supported by the one-year data with four seasons (four complete time series with more than 90 records (24h scale) and 720 records (3h scale)Meanwhile,

the temporal variation of temporal effects on pollutant-meteorology association and its influencing factors should be further investigated in future studies, when the long time series data sets of 3h meteorological data become available.' has been added to the revised manuscript.

Line 132: ' (As shown in Table 1, Table 2 and Table 3) ' →'$O_3$ (Table 1), $PM_{2.5}$ (Table 2) and $PM_{10}$ (Table 3)'.

Line 150: 'which was consistent with previous studies (Wang et al., 2018; Yang et al., 2021)' has been added to the revised manuscript.

Line 211: '(Figure 3)', '(Figure 4)' has been added to the revised manuscript.

Line 215: '(Figure 5)' has been added to the revised manuscript.

Line 221: 'are' →'was'.

Line 232: 'Based on the …' has been removed.

Line 261: '(e.g. The heavily polluted season for $O_3$ and PM was winter and summer respectively).' has been added to the revised manuscript.

Line 274: 'In recent years …' has been removed.

Line 290: '(e.g., humidity, boundary layer height)' →'(e.g., Humidity, Boundary layer height)'.

Line 231: 'the influence of meteorological factors on airborne pollutants has obvious seasonal variations and presented some regional similarity,' has been added to the revised manuscript.