Response to Referee #2

We would like to thank the referee for reviewing the manuscript and providing the valuable comments. We will update our manuscript following the suggestions. Below we answer the specific comments point by point. For readability the comments are shown in bold and italics.

Review comments:

The study made the first attempt to apply the new observation strategy "target observation" to improve the air quality forecasts. A new approach of conditional nonlinear optimal perturbation (CNOP) was applied to find the sensitive area for targeting observations associated with the $PM_{2.5}$ ***forecast of a heavy haze event that occurred in the Beijing-Tianjin-Hebei region. Then several OSSEs, with different lead times and observation distances, were designed to illustrate the sensitivity of the target observations. They also evaluate this new observation strategy through the comparison with other observation strategies revealed by other studies. In addition, they provided the physical reasons why the target observation strategy can greatly improve the PM2.5 forecasts.***

The paper is well written, clearly structured. The study provides a new perspective on understanding the sensitivity of air quality forecasts to the meteorological initial field and can serve as a theoretical guidance on practical observation tasks for $PM_{2.5}$ ***forecast. In the summary part, the authors also present a few sound recommendations for future work, which I think are worthy in-depth study and discussed. Overall this study will make a valuable contribution to the air quality studies. I recommend acceptance after addressing the issues as listed below.***

Response: We appreciate your encouraging comments.

Major comments:

1. ***The authors adopted different observing distances but the same observation number to examine the role of observing distances in the sensitive areas in improving*** $PM_{2.5}$ ***forecast. It was suggested that the observation arrays of large observing distances generally play important role in improving the forecast skill of*** $PM_{2.5}$***. Actually, it is not surprised because the observing array with larger observation distance covers larger area and more meteorological information are captured, which are then much favorable for improving PM 2.5 forecast skill. So I suggest the authors to conduct the following experiments and further examine the validity of the sensitive areas. For a given size of sensitive area, the observing arrays of different observation distances are assimilated to evaluate the role of observing distance. If the large observing distance is still much important for improving PM25 forecast (in this situation, the number of observations is much small), the original result would be assured.***

Response: We thank the referee's comments. In fact, in the 4 forecasts concerned in the manuscript, not all of the observation deployments with the large distance (150km) are the optimal deployments (Table 2 and 3 in the manuscript). For the forecasts at the AT with a 24 hour lead time and the forecast at the DT with a 12 hour lead time, the observation array with the distance of 90km shows higher $PM_{2.5}$ forecast skills than those of 150km. So the observing array with a larger observation distance will not necessarily lead to higher forecast skills.

Moreover, we also conducted the following experiment as the referee suggested. Specifically, we first select a number of 120 most sensitive grids, according to the VI value in the four forecasts. For the given size of the sensitive area, the observations with the distance of 30, 60, 90, 120 and 150km are assimilated. In this situation, the number of the observations differs at different observing distances. The AE_V/AE_M of the forecast at the AT and DT with lead times of 24 and 12 hours are shown in Table R1.

For the forecasts at the AT with lead times of 24 and 12 hours, the observations with a distance of 30km shows the largest improvement in both AE_V/AE_M. It implies that in the given size of sensitive area, denser observation sites can better resolve the synoptic initial conditions within the sensitive area, which in turn enhance the forecasting skills more effectively. In detail, the improvements of AE_V/AE_M become slightly as the observation number increases from around 15 (90km) to 120 (30km) in the two forecasts, indicating that adding more observation sites only results in a small additional benefit.

For the forecast at the DT, the observations with the distance of 30km also shows the largest improvement with the lead time of 24 hours. However, when the lead time is reduced to 12 hours, the observations with the position distance of 90km show the largest improvement. It implies that in this forecast, an appropriate observation distance is much important for improving the $PM_{2.5}$ forecasts.

Compared the results with those in the manuscript, we found that deploying more meteorological observations on the given size of sensitive area is more effective on improving the $PM_{2.5}$ forecast skills. Thus, it is suggested that, if we have a fixed number of observation equipment, an appropriate observing distance is essential to obtain the largest improvement of forecast skills. The observations with the large distance, which cover large areas, will not necessarily lead to higher forecast skills. If we have adequate observation equipment and the observations should be deployed in a given size of area, deploying more observations will be beneficial to enhance the $PM_{2.5}$ forecasting skills. In fact, deploying more observations in the certain range of area with high sensitivity is more realistic, and it could further emphasize the effect of targeted observation.

Table R1 The AE_V/AE_M of the forecasts at the AT and DT with lead times of 24 and 12 hours, when the additional observations in the sensitive region (CNOP) are assimilated. The respective optimal observation array is marked in bold.

Process	Lead Times	30 km	60 km	90 km	120 km	150 km
Accu	24 hour	**22.98/33.94**	20.85/29.95	19.95/26.59	14.31/26.00	11.87/23.28

Diss	12 hour	**46.50/57.62**	43.09/54.12	42.98/51.88	40.87/49.24	40.72/48.00
	24 hour	**58.95/49.81**	55.18/47.41	51.34/44.37	47.28/41.66	42.26/41.07
	12 hour	29.58/39.60	27.57/37.27	**31.48/40.01**	23.22/32.35	19.52/26.36

2. ***Section 5, Line 593-597, the interpretations for the improvements during the accumulation process is a bit weak. Actually, there are two areas identified as sensitive areas for the forecasts at the AT. One lies in the south of BTH, the other is located at central Inner Mongolia. What role did each area play on improving the*** $PM_{2.5}$ ***of BTH? Are there any relation between the meteorological field on these two areas? Such details are needed to be addressed and will help understand the meaning of sensitive areas.***

Response: We thank your comments. To detect each role of the meteorological initial conditions of the two sensitive areas on the $PM_{2.5}$ forecasts, we assimilated the same number of meteorological observations with the same observing distance in the two areas separately. When we only assimilated the observations in the sensitive area near the Dezhou city, which lie to the southeast of Hebei province, the forecast error of $PM_{2.5}$ decreased by 5.49% measured by AE_V and 16.02% measured by AE_M. The assimilation run increases the southerly wind component by 0.05m/s and increases the temperature by 0.1°C at the AT over the BTH region. When we assimilated the observations in the sensitive area near the central Inner Mongolia, the values of AE_V and AE_M are 14.00% and 22.08%, respectively. The assimilation run increases the southerly wind component by 0.16m/s and increases the temperature by 0.21°C at the AT. So assimilating the observations in each of the two areas will result in an increase of $PM_{2.5}$ forecast skills. The sensitive area near the Inner Mongolia plays a more dominant role on the $PM_{2.5}$ forecast of BTH region, by inducing a larger southerly wind component.

We think it is hard to quantify the relations between the meteorological fields over these two regions. When we assimilated the observations on each of the two regions, only the local meteorological condition is improved. Two areas are defined as the sensitive areas because there are two sources of initial errors contributing to the forecast errors of BTH. The role of the north sensitive area is to weaken the northerly wind and the role of the south sensitive area is to strengthen the southerly wind. They both increase the southerly wind component of BTH region, which is helpful for transporting southern pollution to the BTH region in the control run.

Minor comments:

1. ***The "PM 2.5 concentration" in the whole paper means "***$PM_{2.5}$ ***surface air concentrations" (PM 2.5 can be aloft). Please define "PM 2.5 concentration" as "surface air concentrations of PM 2.5" when it is first appeared.***

   Response: We will define the "PM 2.5 concentration" as "surface air concentrations of PM 2.5" when it is first appeared in the revised manuscript.

2. ***Line 40, “relative moisture” is few used. Modify it to “relative humidity”.***
   Response: We will modify the “relative moisture” to “relative humidity” in the revised manuscript.

3. ***Figure 2-5, the color bars of T and QVAPOR are too small. Please modify.***
   Response: We will modify the size of color bars in the revised manuscript.

4. ***Line 75, “assimilating more observations may not lead to higher forecast benefits”. References are needed.***
   Response: We will add the references (Janjic et al., 2017; Zhang et al., 2019) in the revised manuscript.

5. ***Line 339-343, this is not clear to me. Please rephrase it.***
   Response: We will rephrase it in the revised manuscript. “In this situation, the $PM_{2.5}$ forecast could be very sensitive to the combined effect of initial errors of the meteorological fields in the area with larger VI, and preferentially reducing the meteorological initial errors in these sensitive areas will lead much larger improvements of meteorological forecasts over the BTH region, then significantly improve the $PM_{2.5}$ forecasts.”

6. ***Line 585. Clarify which observation array in CNOP-EXP is used when comparing the forecast differences between the CNOP-EXP and control run.***
   Response: We will clarify the observation array in the revised manuscript.

References:

Janjić, T, Bormann, N, Bocquet, M, Carton, JA, Cohn, SE, Dance, SL, Losa, SN, Nichols, NK, Potthast, R, Waller, JA, Weston, P. On the representation error in data assimilation, Q J R Meteorol Soc. 144: 1257– 1278, 2018.

Zhang, K. , Mu, M. , Wang, Q. , Yin, B. , Liu, S. . CNOP-based adaptive observation network designed for improving upstream kuroshio transport prediction. *Journal of Geophysical Research: Oceans, 124*, 4350-4364, 2019.