Dear reviewer,

We all appreciate your hard work on this paper. These constructive opinions help to improve our work to a great extent. We did our best to respond to each comment and make this work well-organized. With the help of your detailed comments, some mistakes in the original manuscript were found and revised. Details are listed as follows:

General comments:

This manuscript analyzed the turbulence data observed from several severe haze pollution episodes in Beijing and its nearby suburbs by using the developed automated algorithm of identifying the spectral gap to separate pure turbulence and submesoscale motions from a 30-min signal based on the arbitrary-order Hilbert spectral method. Although I agree that the motivation of this study is good and its results are interesting, the presented study still needs some minor revision including the improvement in English before consideration for publication.

Specific comments:

- 1. In the abstract, "Urbanization seems to help reduce the consequences of pollution" may be somewhat misleading.
- **Response:** Thanks for your comments. It was really somewhat misleading. We changed the expression to "The turbulent effects caused by urbanization seems to help reduce the consequences of pollution within the same weather and pollution source condition, because the turbulence intermittency is weaker and the reduction of turbulence exchange is smaller over urban underlying surface." in the abstract. Next, I will explain in detail why we made such changes.

Suburban site is located at locally flat underlying surface, as shown in Fig.1, is 20 km away from the urban site. At this distance, we believe that the impact of the large-scale weather situation on both sites is consistent. Recent studies have shown that Beijing's winter pollution process can be divided into pollutant transport stage and accumulation stage (Zheng et al., 2015a; Liu et al., 2013; Guo et al., 2014; Zhong et al. 2017; Zhong et al., 2018). Some studies have shown that emissions from outside Beijing can contribute 28-70% of the ambient PM_{2.5} concentrations in Beijing (An et al., 2007; Streets et al., 2007; Wang et al., 2014; Chang et al. 2018). During the period we are concerned, as shown in Fig. 2, Zhong et al.'s research indicates that the pollution process is composed of the transportation of pollutants by southerly wind and accumulation of pollutants under the static wind (wind speed less than 2 m/s). Pollutant transport is dominated by large-scale weather processes, so the contribution to the source of $PM_{2.5}$ due to transport can be considered to be substantially consistent between the two sites separated by 20 km. In the accumulation stage of pollutants, it can be seen that the wind speed is less than 2 m/s in the layer below 1 km, and there is no longer large-scale transportation. At this time, the contribution of the local source may be highlighted. Due to the regulatory measures such as factory shutdowns implemented by the government in the fall of 2016 ("Bulletin on the State of China's Ecological Environment in 2016"), there were few major industrial sources in Beijing during our research phase, and

there are only a small number of residential sources. In fact, in the government regulation in 2017, the residential sources had also been strictly controlled. Although the suburban site is located in a relatively flat farmland, it is still in the vicinity of the Changping county. Therefore, the residential sources between the two sites may be different, but this difference is not significant compared to the large number of sources in the transportation process. In summary, we think that the sources of $PM_{2.5}$ of the two sites are generally consistent.

So, what we concluded in the manuscript is based on the background that the urban and suburban site are under the same weather and pollution source condition. That is why we changed the expression in that way.



Figure 1 Google earth map of the suburban site within a range of approximately 1 km.



Figure 2 Temporal variations in PM_{2.5}, PLAM, and vertical distributions of meteorological factors from 1 December 2016 to 10 January 2017 by Zhong et al. (2017), their Fig.1. (a) PM_{2.5} mass concentration, (b) wind vector and wind velocity (shading; m s⁻¹), (c) temperature (shading; °C), (d) RH (shading; %), and (e) PLAM. Red boxes correspond to original/transport explosive growth processes, while green boxes correspond to subsequent/cumulative explosive growth processes.

According to your comments, the following changes were made:

Changed the description in the abstract on the first page, line 25-28:

"The turbulent effects caused by urbanization seems to help reduce the consequences of pollution under the same weather and pollution source condition, because the turbulence intermittency is weaker and the reduction of turbulence exchange is smaller over urban underlying surface."

Figure 1 on page 6 in the revised manuscript has been modified to reflect the condition of the underlying surface around the two sites. The new Fig. 1 for the manuscript is shown as Fig. 3 here:



116.1 °E 116.2 °E 116.3 °E 116.4 °E 116.5 °E 116.6 °E

Figure 3 Figure 1 in the manuscript: Google Earth map of the observation sites in Beijing: (a) the observation site located in the urban underlying surface region (marked by the red pentagram) and the observation site located in the suburban underlying surface region with a flat terrain (marked by the red circle). The corresponding terrains (within a range of approximately 1 km) around the observation sites are shown in (b) and (c), respectively.

Details of the suburban site are added on page 3, lines 29-30:

"Data over a locally flat underlying surface were collected at a continuous measurement site (40.16° N, 116.28° E) in the Beijing suburb. The observational site was set up in the middle of a vast and horizontal farmland, near the Changping county."

We added a description of the location relationship between the two sites on page 4, lines 20-32:

"Suburban site is 20 km away from the urban site. At this distance, the large-scale weather background is consistent. As flat terrain of the suburban site, it was used as a reference. The sources of $PM_{2.5}$ of the two sites are generally consistent."

2. Line 16 in Page 3, "turbulence data observed from several severe haze pollution episodes", "from" should be changed to "in" or "during".

Response: Thanks for pointing out that. "from" has been corrected to "during".

- 3. The data source of $PM_{2.5}$ mass concentration, horizontal wind speed, virtual temperature and water vapor mixing ratio need be described. The details of all the data used in this study should be included in the Section.
- **Response:** Thanks for your suggestion. We have supplemented the details of the observation instruments and data of the two sites respectively. We added some descriptions of the suburban site at page 4, lines 2-4. Details are

given as follows:

"The turbulence data such as the horizontal wind vector, virtual temperature and water vapor content were collected using a data logger (CR3000, Campbell Scientific, Inc., USA) at a frequency of 10 Hz and were averaged over an interval of 30 min for the analysis of meteorological elements."

We added some descriptions of the urban site at page 4, lines 11-17. Details are given as follows:

"The concentrations of PM_{2.5} were collected using a Thermo-Fisher Sci. Co. instrument (series FH-62-C14), and 30-min averaging time series were performed to remove outliers. The system was equipped with an integrated CO₂/H₂O openpath gas analyzer (LI-7500, LI-COR Biosciences, Inc., USA) and threedimensional sonic anemometer-thermometer (IRGASON, Campbell Scientific, Inc., USA). The IRGASON was leveled and pointed north. The turbulence data such as the horizontal wind vector, virtual temperature and water vapor content were collected using a data logger (CR3000, Campbell Scientific, Inc., USA) at a frequency of 10 Hz and were averaged over an interval of 30 min for the analysis of meteorological elements."

We added a description of the location relationship between the two sites on page 4, lines 20-24:

"Suburban site is 20 km away from the urban site. At this distance, the large-scale weather background is consistent. As flat terrain of the suburban site, it was used as a reference. The sources of $PM_{2.5}$ of the two sites are generally consistent. The observations of $PM_{2.5}$ at urban sites are used to represent the evolution of the entire pollution process, as this study focuses on pollution processes rather than specific values. Since the observations at both sites are located in the surface layer, i.e. the constant flux layer, the values of turbulence flux are comparable."

- 4. It may be better to modify the title of Section 3 as "Methodology" or "Methodology of reconstructing signals". Section 2.2 may be merged into "Methodology". In a word, Section 2 and Section 3 should be rearranged.
- **Response:** Thanks so much for your constructive advice. We modified the title of Section 3 as "Methodology". You are right that the description of method of calculating the turbulent quantities in Section 2.2 should be merged into Section 3. So we retain the content of data processing in Section 2.2 and move the content of description of method to the Section 3 which was rearranged.

The new Section 3.1 at page 8, lines 9-24 are given as follows:

"3.1 Turbulent kinetic energy and turbulent fluxes

The physical quantities used in this paper are turbulent kinetic energy (TKE),

several variances (σ_u , σ_v , σ_w , σ_θ and σ_q), friction speed (u_*), and fluxes ($-\overline{u'w'}$,

 $\overline{w'\theta'}$ and $\overline{w'q'}$). Among these, the TKE is calculated as:

$$e = \frac{1}{2} \left(\overline{u'^2} + \overline{v'^2} + \overline{w'^2} \right), \tag{1}$$

the variance is calculated as

$$\begin{aligned}
\sigma_{u} &= u'u', \\
\sigma_{v} &= \overline{v'v'}, \\
\sigma_{w} &= \overline{w'w'}, \\
\sigma_{\theta} &= \overline{\theta'\theta'}, \\
\sigma_{q} &= \overline{q'q'},
\end{aligned}$$
(2)

the turbulence flux is calculated as

$$\tau = \rho u_*^2 = \rho \overline{u'w'},$$

$$H = \rho C_p \overline{w'\theta'},$$
(3)

$$E = \rho \overline{w'q'}$$
,
and the friction speed is calculated as:

$$u_{*} = \left[\left(-\overline{u'w'} \right)^{2} + \left(-\overline{v'w'} \right)^{2} \right]^{\frac{1}{4}}.$$
(4)

The original Section 3.1 and Section 3.2 became the new Section 3.2 and Section 3.3

- 5. In Figure4, the comparison is made between the new half-hour results with those from the old results. Which is the reference? Is the overestimation of the variations in the variables calculated by the traditional EC method for 30 min referenced to the results using the new method? Then, what is the reference to assess the new method? Here, the description is confused.
- **Response:** Thanks for your suggestion. Yes, the overestimation of the variations in the variables calculated by the traditional EC method for 30 min is referenced to the results using the new method. In this work, we recognize that the new method can get the pure turbulence part and eliminate the effects caused by sub-mesoscale motion. Because we can get the conclusion by comparison of the spectra between the raw data and reconstructed data. For example, Fig. 3 in the manuscript shows the second-order Hilbert spectra from the newly reconstructed data and raw data. The raw data spectrums, which are shown by the black solid lines in Fig. 3 in the manuscript, are inconsistent with the structure of the turbulent energy spectrum in the classic theory on frequency bands with lower frequency, that is, smaller than the frequency indicated by the grey solid lines. The new spectrum, which is shown by the black dotted lines in Fig. 3 in the manuscript, is consistent with the structure of the turbulent energy spectrum in the classic theory. It is obvious that the reconstruction successfully eliminated the energy contained by large-scale motion while retaining turbulent energy. Under the situation that there are spectra gaps, the turbulence data we obtained during the heavy pollution process contains the sub-mesoscale motion signal. Turbulent flux calculated by the traditional timeaveraging method is contaminated by sub-mesoscale motions during the heavy pollution process. Similarly, this kind of contamination to turbulence flux caused by sub-mesoscale motion was also studied in some other works (Vickers and

Mahrt, 2006; Acevedo et al., 2006, 2007; Aubinet, 2008; Mahrt, 2010). All in all, we can find that the method we developed in this paper which is based on the Hilbert-Huang transform can get more realistic exchange between the surface and atmosphere during the heavy haze pollution.

However, you are right that the description here may easily cause confusion. We changed the expression of "old results" to "original results" in the manuscript which maybe can make it more clear. The figures involved the expression of "old" in the manuscript are Fig. 4 (on page 13) and Fig.5 (on page 15) which were also modified.



Figure 4 in the manuscript: Comparison of σ_u (a), σ_v (b), σ_w (c), σ_θ (d), σ_q (e) and TKE (f) from the new half-hour results with those from the original results from 16 December 2016 to 8 January 2017 at the suburban site. The black dotted line represents the 1:1 line in the figures. The black solid line represents the fitted results.



Figure 5 in the manuscript: A comparison of the vertical heat flux $(\overline{w'\theta'})$ (a), vertical water-vapor flux $(\overline{w'q'})$ (b) and momentum flux $(-\overline{u'w'})$ (c) from the new half-hour results with those from the original results from 6 December 2016 to 8 January 2017 at the suburban site. The black dotted line represents the 1:1 line in the figure. The black solid line represents the fitted results.

6. What's the difference of PM_{2.5} in Fig.6-8 and Fig.2? How about the difference of

data source? More details need be described.

Response: Thanks for your suggestion. The data source of PM_{2.5} is the same in Fig.6-8 and Fig.2. We are sorry that we have not described clearly. In order to facilitate comparative analysis and display intuitively, the time series of PM_{2.5} was added in Fig.6-8. The observations of PM_{2.5} at urban sites are used to represent the evolution of the entire pollution process, as this manuscript focuses on pollution processes rather than specific values. As mentioned in the answer of the first question, the sources of PM_{2.5} of the two sites are not much different. In fact, the trends in concentration of PM_{2.5} across all environmental monitoring sites throughout the Beijing area are consistent, although there are some numerical differences. We choose three environmental monitoring stations, Changping (116.23°N, 40.22°E), Haidian (116.29°N, 39.99°E) and Daxing (116.40°N, 39.72°E), to prove that. The time series of mass concentration of PM_{2.5} at the three environmental monitoring stations are shown in Fig. 6, their locations are shown in Fig. 7.

The data of $PM_{2.5}$ used in this manuscript is mainly to show the corresponding relationship between the trend of pollution development and intermittent turbulence, as shown in Fig.7 and Fig.8 in the manuscript. For the purposes of this work, the difference in the magnitude of the $PM_{2.5}$ values between the two sites does not affect the results. And because the pollution data from environmental monitoring sites have a large number of missing measurements during the study period, so we still use the observations from the urban site to represent the evolution of the entire pollution process.

An explanation of the data problem of $PM_{2.5}$ is added to Section 2, page 4, lines 21-22:

"The observations of PM_{2.5} at urban sites are used to represent the evolution of the entire pollution process, as this study focuses on pollution processes rather than specific values."



Figure 6 The time series of mass concentration of $PM_{2.5}$ at the three environmental monitoring stations in Beijing.



Figure 7 Google Earth map of the observation sites in Beijing. The locations of the three environmental monitoring stations are marked by the red pentagram. The urban and suburban sites in manuscript are marked by red circle.

7. In Fig. 9-11, the description of lines is wrong.

Response: Thanks for your suggestion. We are sorry for these faults. We corrected the descriptions of lines in Fig. 9-11 as follows:

"Figure 9: Normalized standard deviations in wind speed in the horizontal and vertical directions (σ_u/u_* , σ_v/u_* , and σ_w/u_*) as functions of the stability parameter ζ . The red (blue) solid line in the figure represents the results under polluted (clear) weather conditions. Observations marked with * (°) were made under polluted (clear) weather conditions." (page 18).

"Figure 10: Normalized standard deviations in the potential temperature $(\sigma_{\theta}/|\theta_*|)$ and moisture content $(\sigma_q/|q_*|)$ as functions of the stability parameter ζ . The red (blue) solid line in the figure represents the results under polluted (clear) weather conditions. Observations marked with * (°) were made under polluted (clear) weather conditions." (page 19, lines 2-4).

"Figure 11: Diurnal variations in the mean vertical heat flux $(\overline{w'\theta'})$ (a), vertical water-vapor flux $(\overline{w'q'})$ (b), momentum flux $(-\overline{u'w'})$ (c) and TKE (d) under polluted weather (red solid line) and clear weather (blue solid line) conditions over the urban site. Diurnal variations in these variables over the suburban site are shown in (e), (f), (g), and (h), respectively."

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