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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Intermittent turbulence contributes to vertical diffusion of $PM_{2.5}$ in the North China Plain" *by* Wei Wei et al.

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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:

Major comments: There are substantial differences between the CS and TS in TKE, u*, and even W and U (Fig. 2). One can use these quantities in the explanation of accumulation and diffusion of PM2.5. Why do we need an IF index? In other words, what is the advantage or superiority of using IF compared with other quantities? This

point should be discussed in the paper.

Response: Thank you for your comments. In previous works, some traditional variables (i.e. TKE, u*, and W/U) are commonly applied to indicate the behavior of turbulence. And the results in our work (Fig. 2) also show the relationship between those variables and the accumulation and diffusion of PM2.5 to some extent. Indeed, those quantities are useful for the description of turbulent characteristics including strength and variation, but fail to reveal the intermittency of turbulence. As mentioned in the introduction, the reason why we focus mainly on the influence of intermittent turbulence is that the intermittent turbulence accounts for a large amount of vertical fluxes in stable boundary layers but the discussion of the effects of intermittent turbulence on the transport of PM2.5 is still limited. At this point, we need an effective way to describe the characteristics of turbulent intermittency. The IF index was developed from the arbitrary-order Hilbert spectral analysis (arbitrary-order HSA, Huang et al., 2008). Compared with some classic methods (such as Fourier analysis and wavelet transform), the arbitrary-order HSA technique is intuitive, direct, and adaptive, with a posteriori-defined basis, from the decomposition method, based on and derived from the data, it is more appropriate for the analysis of nonlinear and non-stationary turbulence signals. Our some previous work (Wei et al. 2017) has addressed the intermittency of turbulence in the SBL using the arbitrary-order HSA technique. But we should admit that, as a newly-developed approach, arbitrary-order HSA still suffers some disadvantages. For example, the spline fitting and the end effects need more improvements. In the case of weak signals imbedded in stronger ones, differentiation should be applied if needed. In spite of these problems, HSA is still the best available nonlinear and non-stationary data analysis method so far. Based these considerations, we used the arbitrary-order HSA in this work to study the behavior of turbulent intermittency. The advantages of IF or arbitrary-order HSA are addressed in the revision according to your advice: "Based on the arbitrary-order HSA, we proposed an index, called intermittency factor (IF), to quantify the level of turbulent intermittency, which is assumingly more effective compare with some classic quantities." (page 5 lines 10-13) and "As one of the most important steps through this method, the empirical mode decomposition separates the original time series into different modes based on its own physical characteristics without any predetermined basis, implying an intuitive, direct, adaptive, and data-based nature" (page 6 lines 22-24). Some more detailed information is given in the supplement: "The reason why the arbitrary-order HSA is applied to this work is that this method is more suitable for the analyses of nonlinear and non-stationary turbulent signals, compared with traditional techniques, such as Fourier analysis and Wavelet transform. It is known that, the data, whether from physical measurements or numerical modeling, most likely will have some problems: (a) the total data span is too short; (b) the data are non-stationary; and (c) the data represent nonlinear processes. While the Fourier analysis has some crucial restrictions: the system should be linear; and the data should be strictly periodic or stationary; otherwise, the resulting spectrum will make little physical sense. However, for lack of alternatives, Fourier spectral analysis is still applied to many kinds of data which may result in misleading results. On the other hand, the wavelet approach is essentially an adjustable window Fourier spectral analysis, with the basic wavelet function that satisfies certain very general conditions. As the traditional technique for the analysis of intermittency, the structure function is essentially associated with the Fourier decomposition, which means that the scaling exponent function ζ(q) has some limitations in the application of nonlinear and non-stationary turbulence signals." (page 4 lines 9-21 in the supplement) "As discussed by Huang et al. (1998, 1999), the arbitrary-order HSA technique is intuitive, direct, and adaptive, with a posteriori-defined basis, from the decomposition method, based on and derived from the data, it is more appropriate for the analysis of nonlinear and non-stationary turbulence signals. Since its introduction, the HSA method has been successfully applied into different fields, including climatology (Molla et al. 2006; Hu et al. 2014), meteorology (Karipot et al. 2009; Vincent et al. 2011) and oceanography (Chen et al. 2014), to name just a few. One of the authors (Wei et al. 2017) used arbitrary-order HSA technique to separate fine-scale and large-scale motions in the stable boundary layer (SBL) and obtained a better approximation to the Monin-Obukhov similarity the-

ory than using bandpass filtering method. Bases on these considerations and previous work, we believe that the arbitrary-order HSA technique is a suitable method for the study of turbulence intermittency in the SBL." (page 4 line 21 and page 5 lines 1-9 in the supplement)

(2) Are there any significant correlations between IF and TKE as well as other parameters? If there are, they should be presented and discussed.

Response: Thank you for your suggestion. We examined the relationship between IF and other quantities, including u* as the dynamic parameter and z/L as the thermodynamic variable, and the results are shown as follows:

Figure 1 Scatter plot comparing IF and other variables (u_* and nighttime z/L) for two cases at 40 m. The dashed lines are the fittings from least-squares regression and the triangle marks the cross point.

Generally, the values of IF decrease with increasingly stronger turbulence, which meets our expectation. Under extremely strong stable conditions, the turbulence in the ABL is suppressed, accompanied by very small dynamic quantities (such as u* in this case). At this point, the values of IF are nearly zero, representing the extremely weak turbulent fluctuation. With the increase of turbulence strength, the abstract value of IF rises, indicating that the relatively stronger turbulence in the ABL is intermittent but not continuous or fully-developed. In order to confirm these conclusions, the distribution of IF with stability function z/L during the nighttime is given in Figure 1 as well. Under strong stable conditions (i.e. z/L » 1), turbulence is weak and IF is nearly zero. While the weak stable cases (i.e. z/L≈0.1) are accompanied by active turbulence but larger negative IF. This part is added to the revision as in: "Fig. 7 further confirms the relationship between IF and u_* or z/L, in which dots of strong turbulence (u_*) and weak stable stratification (z/L≈0.1) mainly come from the TS. Larger deviation of IF occurs accompanied by increasing turbulent strength when stability in the ABL becomes weaker. That is, intermittent turbulence (marked by large negative values of IF) leads to strong fluxes during the TS." (page 13 lines 7-10) and Fig.7 (page 14).

(3) Your measurements are from Tianjin, which is just west of the Bohai Bay. The emission and formation of PM2.5 over the land areas are much stronger than over the sea. Therefore, I guess air from the Bohai Bay was much cleaner. During each TS the prevailing wind was either southeasterly or northeasterly, different from that during the CSs. Did the change in horizontal air flow contribute also to the decrease in the PM2.5 concentration? And how significant?

Response: We really appreciate your constructive questions. Firstly, we must apologize that there is something wrong with the wind vector in Fig. 2. We have carefully checked through the raw data and corrected the drawing program. The right wind vector at three levels (40, 120, and 200 m) for two cases is shown in the following Figure 2 and Figure 3 is the corresponding rose diagram of wind direction. From the results of Figure 2, the CS is characterized by south-easterly wind. When it comes to the TS, the wind predominately originates from west or north-west. Although the wind vector of Case-2 in Figure 2 is relatively disordered, the rose diagram in Figure 3 reveals that the most common wind direction for the CS ranges from east to south-east and the flow during the TS is mainly from the west, which is consist with the results of Case-1. Previous works have given plenty of solid evidence on the impacts of local and synoptic circulation on the accumulation and transport of air pollutants in the North China Plain. Considering that the main purpose of this work focuses on the vertical transport by turbulent mixing, we just cited some recently published paper (including Zhang et al., 2017; Miao et al., 2017; Ye et al., 2016; Zhang et al., 2012; Zheng et al., 2015a; Jiang et al., 2015) in the introduction (see page 2 line 4). Here we try to address it in detail. Tianjin is surrounded by Hebei province and also located to the north of western Shandong and northern Henan province, which are the most densely pullulated regions with the fastest growing economy in Northern China recently (Wang et al., 2014). In this case, the contribution to the PM concentration by cross-city transport from neighboring province cannot be neglected. Using modeling study, Jiang et al. (2015) revealed

that the southerly wind at lower layer contributes to transport PM from the southern neighboring cities with serious pollution. Furthermore, air masses from the south are warmer and wetter than the northern air masses, thus possessing a higher specific humidity, which facilitates the secondary formation by heterogeneous reactions (Zheng et al., 2015a). In terms of transport stage, Zheng et al., (2015a) found that weather pattern for the clean hours are normally characterized by strong high-pressure centers northwest of the polluted region in winter (i.e., the Siberian anticyclone), resulting in strong north-westerly wind. We are also thankful for your revealing comment on the sea breeze. It is reasonable to expect that the flows from the Bohai sea would be helpful to improve the air quality in Tianjin, but the work by Miao et al. (2015) provides an opposite conclusion. Their modeling results show that the southerly ambient wind brings lots of aerosols emitted from southern region to the Bohai sea and then sea breeze transports marine air together with the aerosols to the land. All of these works above present the importance of horizontal circulation in the transport of PM2.5. However, some works (Zheng et al., 2015a; Chan and Yao, 2008) mentioned that in the case of city clusters, air pollution may not be eliminated solely by advection. This is because the pollution is formed in the city cluster, there is no clean air from upwind, resulting in more persistent pollution events. There is no doubt that horizontal transport is crucial to the decrease in the PM2.5 concentration, but the mechanisms of polluted events are complicated and we try to explore the effect of intermittent turbulence from a new angle. We are so sorry for the errors in the wind vector in Fig. 2 and they have been corrected in the revision. Besides, in order to enrich this work, some previous results on the local circulation are introduced in page 7 lines 14-22: "For Case-1, wind at lower levels mainly comes from the south-east during the CS, while the dominant wind direction turns into west when it comes to the TS. Although the wind direction for Case-2 is seemingly unsteady in Fig. 2, the statistical the rose diagrams (see Figure S8) confirm a similar result, with south-easterly flows dominating the CS and westers for the TS. This wind-direction pattern is in agreement with previous works (Zhang et al., 2017; Miao et al., 2017; Zheng et al., 2015a; Jiang et al., 2015). They found that south-easterly wind can bring the aerosols emitted by the surrounding cities to this region while the clean hours is normally characterized by strong high-pressure centers northwest of the polluted region in winter. However, in the region with densely distributed mega-cities (as in the case of Tianjin), because the upwind flows is polluted, mere advection may not be enough to disperse pollutants, thus resulting in persistent air pollution events (Zheng et al., 2015a; Chan and Yao, 2008)."

Figure 2 Wind vector at three levels. The left panel is for Case-1 and the right panel is for Case-2.

Figure 3 Comparison of rose diagram between the CS and the TS for two cases at three levels.

(4) You show in Fig. 3 vertical profiles of changes in potential temperature during the CSs. I think similar results for the TSs should be presented and discussed as well. You may show how rapidly the stable condition formed during the CSs was broken by intermittent turbulence. In addition, some discussions about the evolution of the PBL height may be also good for a more complete picture.

Response: Thank you so much for your constructive advice. The change in potential temperature during the CS and the TS is both presented in Fig.3 (a–f) in the reversion. Compared with the clear warming at high levels during CSs, the high-level cooling during TSs is significant (see Fig. 3 d–f), implying the collapse of inversion layer when it comes to the TS. Meanwhile, Fig. 3g gives the daily mean potential temperature profiles from 23 to 28 November to illustrate the evolution of inversion layer. We can see that the inversion layer gradually developed from 23 to 27 November but rapidly collapsed on 28 November. Since there is no radio soundings available near Tianjin site, we simulated the Planetary Boundary Layer Height using WRF model. The model configuration can refer to the work by Zheng et al. (2015c) which focused on a haze event in 2013 of this region. Details are given as follows: "Fig. 3 depicts the distribution of Planetary Boundary Layer Height (PBLH) and the daily mean potential temperature

profiles at 15 different heights, including change of θ over the CS (Fig. 3a–c) / TS (Fig. 3d–f) and the development during the whole polluted event (Fig. 3g). The $\Delta\theta$ at given height of CSs was calculated by subtracting the value of θ on the last day from that on the first day. And so it does for TSs. For Case-1, $\Delta\theta$ during the CS at the lowest level (5 m) is only 5.2 K. But for the top level at 250 m, $\Delta\theta$ is relatively larger with a value of 6.8 K. This result confirms that the warming of upper layers is stronger than that of lower layers, implying an increasingly stably stratified boundary layer during polluted days. Figs. 3b and 3c for Case-2 verify this conclusion as well. On the contrary, $\Delta\theta$ during TSs (Fig. 3d–f) presents a significant cooling at higher levels, denoting the collapse of inversion layer at the end of the polluted event. Taking Case-1 as an example, Fig. 3g depicts the evolution of inversion layer. It can be seen that the inversion layer was gradually enhanced from 23 to 27 November but quickly depressed on 28 November, which verifies the results of Fig. 3a–f. Fig. 3h illustrates the distribution of PBLH, which is simulated with the Weather Research & Forecasting (WRF) Model (Zheng et al., 2015c). In Fig. 3h, the PBLH for Case-1 gradually decreased and reached its minimum on the night of 26–27 November. Then the PBLH redeveloped to higher than 1,300 m during the daytime of 28 November." (page 7 lines 29-33 and page 8 lines 1-9)

Figure 4 Vertical distribution of daily mean potential temperature. The change of daily mean potential temperature of CSs is showed in (a)–(c) and (d)–(f) are for TSs. (g) illustrates the evolution of inversion layer of Case-1. (h) is the PBLH simulated with WRF Model.

(5) Your results and conclusions are based on cases study. I think it is better to add "cases from Tianjin" or similar subtitle. And "vertical diffusion" in the title can be questionable if you cannot prove that the decrease in PM2.5 was solely due to the vertical diffusion.

Response: Thank you for your suggestion. We specified "cases from Tianjin" in the revision. But considering that there have been a lot of work aiming on the horizontal transport of PM2.5, we mainly focus on the effect of vertical mixing of intermittent turbulence. Indeed, the reasons for the transport of particles are complicated, including climate change, synoptic circulation, and boundary layer structures and we cannot address them all. So far, there is limited works on the intermittent turbulence under strongly stable conditions. Therefore, we keep “vertical” in the new title to emphasize the effects of vertical turbulent mixing and we hope you will approve of our modification. The new title is “Intermittent turbulence contributes to vertical dispersion of PM2.5 in the North China Plain: cases from Tianjin”.

Minor points: Page 1 line 21: What do you mean by “wind filed”? Wind profile?

Response: Yes, it should be “wind profile” and has been rewritten.

Page 2 line 21: Define “FI”.

Response: “FI” is defined as flux intermittency by Mahrt (1998). FI = σF/abs[F], in which σF is the standard deviation of the 5-minute averaged flux and abs[F] is the absolute value of the one-hour average of the flux. The manuscript has been corrected as well. “FI index (Flux Intermittency, Eq. (9) in Mahrt, 1998)” (page 2, lines 21-22)

Page 2 line 32: Delete “respectively”.

Response: Yes.

Page 3 line 4: Change “east” to “southeast”.

Response: Yes.

Page 3 line 11: I think “HMP45C” is the type name of the probe and should be put in the brackets.

Response: This has been corrected.

Page 3 line 16: Was the TEOM system installed near the tower or the WPR? Please make it clear.

Response: Thank you for your question. The TEOM system used in this study is

mounted near the 255-m tower. The distance between the 255-m tower and the TEOM system is around 2.3 km. The location of the TEOM system is specified as follows: “The 1405-DF TEOM system is located nearly 2.3 km away from the 255-m tower to the east and installed at a height of 3 m to monitor the surface PM2.5.” (page 3 lines 15-17)

Table 1: “c: 300-366 m s-1”. Is this the range of wind speed that the sonic anemometer can measure? 300 is a very strange number here.

Response: Here c represents the speed of sound which is used to calculate the sonic virtual temperature. According to the instruction manual of CSAT3 Three Dimensional Sonic Anemometer, the range of speed of sound is from 300 to 366 m s–1 (–50 to +60 $^{\circ}$C). The definition of c is added to Table 1.

Page 4 line 16: Change “poor data” to “poor quality of data”.

Response: Yes, thank you.

Page 4 lines 19-21: What are the criteria for data that are suitable for this study?

Response: All of the data used in this study were checked strictly. The quality control for turbulence observations includes error flag, spike detection, cross wind correction, spectral loss correction, sonic virtual temperature correction, density fluctuation correction, and coordinate rotation. “If more than 20% points within a given 30-min time series were detected as outliers, then this 30-min observation was discarded.” (page 4 lines 10-11) The wind profiles were checked time by time. “First, data below 200 m were removed due to the interference of surrounding environment, including trees and buildings. Then each vertical profile was checked through and points with larger than 2.5 standard deviations were regarded as outliers and discarded. (page 4 lines 17-19)” And according to previous study in this region (Wei et al., 2014), a profile was discarded if more than 40% of the data points were outliers or missing.

Page 5 line 2: “local standard time” or “Beijing Time”?

Response: Yes, it should be "Beijing Time".

Page 5 line 9: "On this basis"? It is not clear what is denoted.

Response: We mean that based on the arbitrary-order HSA, we developed IF. This has been corrected.

Page 5 lines 11-12: Delete "(CSAT3, CAMPBELL Inc., USA)" because the same information is given on page 3.

Response: It has been deleted.

Page 5 line 19: Do you mean the local maxima that are found within every 30-min periods?

Response: Yes, here the local maxima are from the 30-min time series X(t). This has been corrected as: "The first step is to form the upper envelope e_max (t) based on the local maxima of 30-min X(t)" (page 5 lines 21).

Page 5 line 9, page 6 lines 12-13, and page 11 lines 18-19: You are proposing or defining IF at these three places. This is redundant. I think you should define the IF index at a suitable place and use it elsewhere.

Response: Thank you for pointing out that. The IF index is defined when it is first mentioned (page 5 line 11) and other definitions have been deleted.

Page 6 line 31 and page 7 line 1: ". . .increased to a maximum of 412 ug m-3 for PM2.5 and then dropped to a low level within a few hours no matter for Case-1 and Case-2". Please check you expression. I do believe the maximum values in Fig. 2a and Fig. 2b are all 412.

Response: Thank you for your question. The maximum for Case-1 is 263 μg m-3 and 412 μg m-3 for Case-2. This part has been rewritten in the revision. "it can be seen that the concentration of PM2.5 gradually increased to maxima (263μg m-3 for Case-1 and 412 μg m-3 for Case-2) and then dropped to a low level within a few hours." (page

7 lines 5-6)

Page 9 Fig. 2: Please add some ticks on the Y-axes of Figs. 2a and 2b.

Response: The Fig. 2a and 2b have been replotted with ticks.

Page 11 line 2: CSs or TSs?

Response: We are sorry for the slip of the pen. It should be "TSs".

Page 11 Fig. 5: Does each concave curve represent a 30-min result? Please make it clear.

Response: Yes, each curve in Fig. 5 is from a 30-min vertical wind speed signal, which has been clarified in the caption of Fig. 5. "Hilbert-based scaling exponent function at 40 m during different stages for (a) – (b) Case-1 and (c) – (d) Case-2, where each dashed curve represents the result of 30-min vertical wind speed signal and the black solid line denotes the K41 result q/3." (page 12 and lines 15-16)

Page 11 line 24: Delete "site".

Response: Yes.

Page 13 line 2: "under stable conditions"? Are you not talking about the TS?

Response: Thank you for your question. We have replaced "under stable conditions" with "in the ABL".

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Please also note the supplement to this comment:
https://www.atmos-chem-phys-discuss.net/acp-2018-121/acp-2018-121-AC1-supplement.pdf

Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-121, 2018.

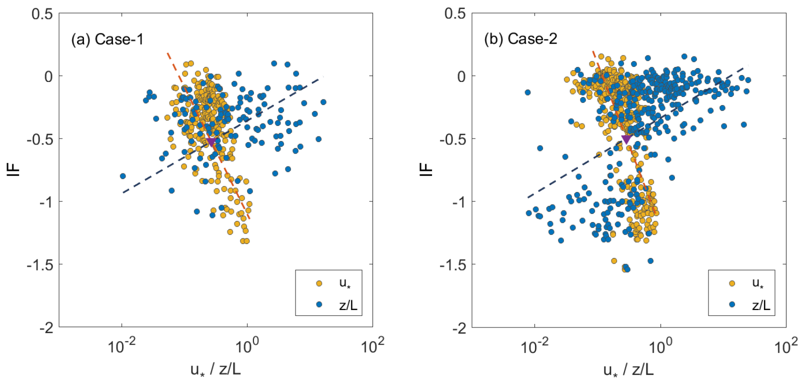

Fig. 1. Scatter plot comparing IF and other variables (u_* and nighttime z/L) for two cases at 40 m. The dashed lines are the fittings from least-squares regression and the triangle marks the cross point.

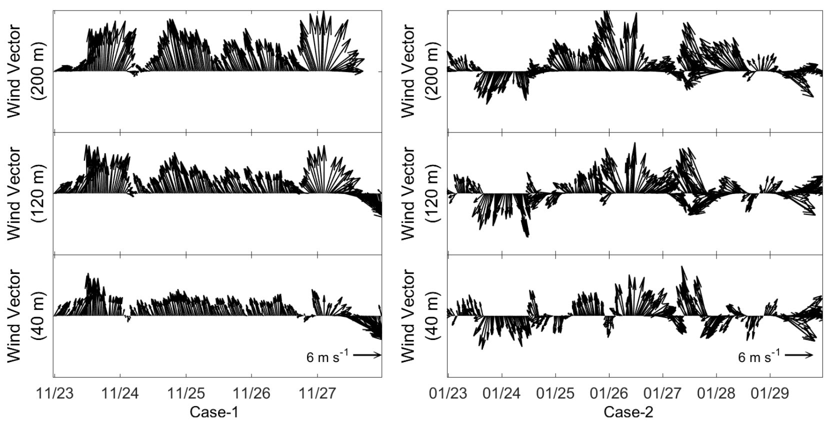

Fig. 2. Wind vector at three levels. The left panel is for Case-1 and the right panel is for Case-2.

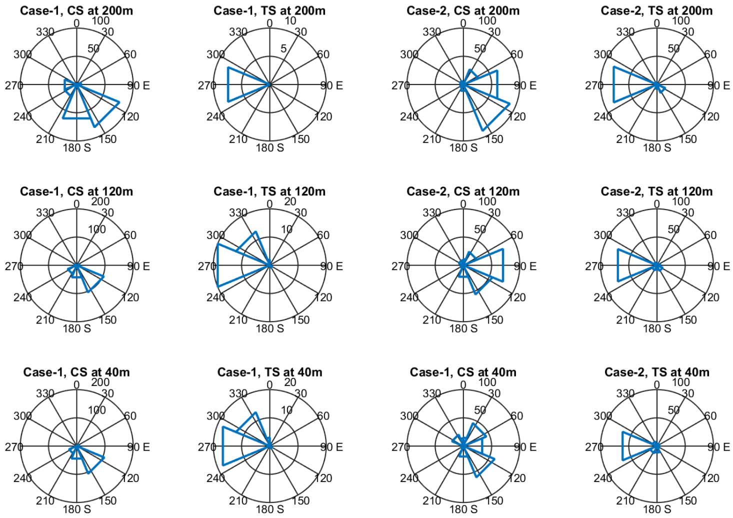

Fig. 3. Comparison of rose diagram between the CS and the TS for two cases at three levels.

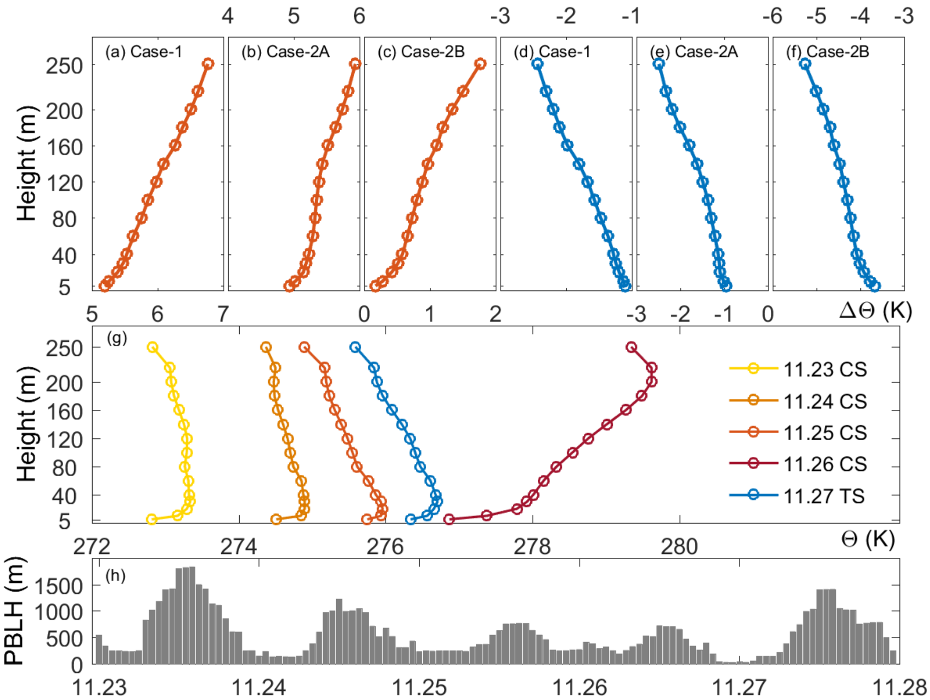

Fig. 4. Vertical distribution of daily mean potential temperature. The change of daily mean potential temperature of CSs is showed in (a)–(c) and (d)–(f) are for TSs. (g) illustrates the evolution of inversio

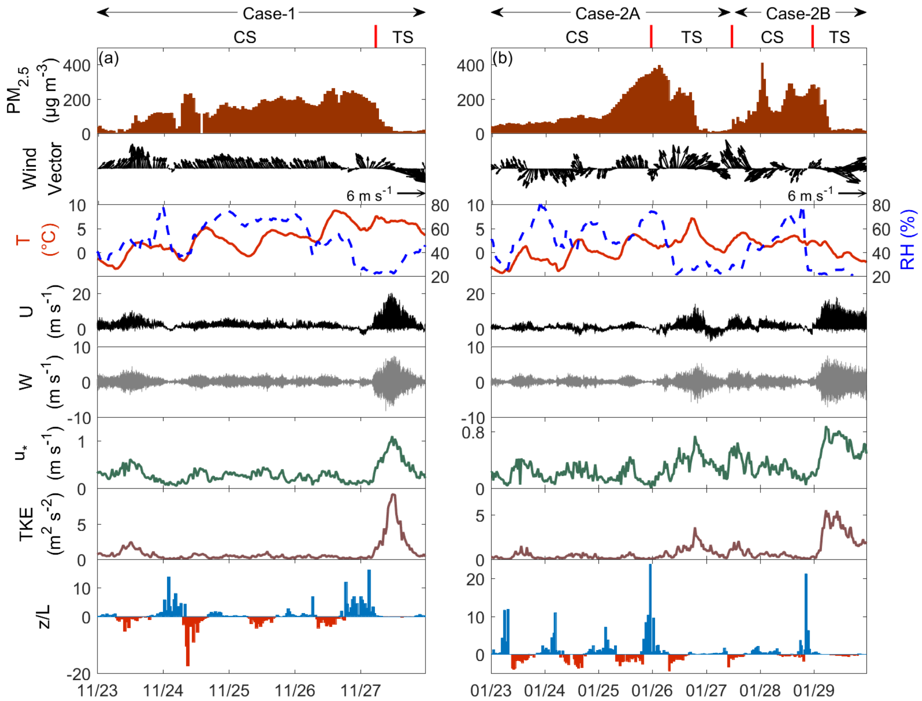

Fig. 5. Figure 2 in the manuscript. Time series of surface PM2.5, wind vector, temperature (T), relative humidity (RH), horizontal wind speed (U), vertical wind speed (W), friction velocity (u_*), turbulent