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Quantitative verification of the turbulence barrier effect during heavy haze pollution events

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Abstract

Under calm and steady weather conditions with low wind speeds, turbulent intermittency frequently occurs in the atmospheric boundary layer (ABL), which can significantly weaken the turbulent diffusion of matter and energy between the surface and atmosphere. The turbulence barrier effect is defined as the phenomenon in which turbulence may disappear at certain heights, and during periods of heavy haze, creating what can seem like a barrier layer that hinders vertical transmissions. Although the turbulence barrier effect can explain the physical mechanisms behind the rapid accumulation of PM₂.₅ (fine particulate matter with diameters smaller than 2.5 μm) and the influence of turbulent diffusion conditions on the vertical distribution of PM₂.₅, more direct perspectives such as turbulent flux is still required for quantitative verification. Due of challenges in the acquisition of PM₂.₅ turbulent flux, carbon dioxide (CO₂), which has relatively mature flux acquisition technology, was used as a substitute means of verifying and quantifying this phenomenon. The turbulence data collected during heavy haze events, at from five levels of a 255 m meteorological tower located in Tianjin, were analyzed and used to quantitatively verify the influence of the turbulent barrier effect on PM₂.₅. The results also revealed that the vertical changes in the turbulent barrier effect were consistent with those of the concentrations and flux of CO₂. This means that this knowledge about the turbulent barrier effect can be extended to other mass-transfer processes. The analysis also found that the proportion of counter-gradient transport increases when the occurrences of the turbulent barrier effect are frequent. This work validates the presence of the turbulent barrier effect and is an important foundation for its future parameterization, which will help to accurately identify the matter transport processes in the stable boundary layer and under extreme weather conditions, such as intense pollution events.

1. Introduction

Haze pollution, with PM₂.₅ as the primary pollutant, has always attracted broad attention (Fan et al 2020, Chen et al 2021), even during the COVID-19 epidemic (Beeler and Chakrabarty 2021, Wu 2021). Meteorological conditions play an important role in the occurrence and development of haze pollution events (Wang et al 2021), especially in the ABL, where haze pollution occurs. Heavy haze pollution is often linked to the stable boundary layer (SBL) (Pierce et al 2019, Li et al 2020, Yuval et al 2020) under small and stagnant wind speeds (Jacobson and Kaufman 2006, Zhong et al 2018). There are interactions between aerosols and turbulence in ABL which can
enhance haze pollution (Ding et al. 2016, Zhao et al. 2021) and change the vertical distribution of aerosols (Ren et al. 2021a).

Turbulence is the main form of motion in the ABL, which plays an important role in the transportation and exchange of energy and matter between the surface and the atmosphere. Aerosols in the ABL are strongly affected by atmospheric turbulence. Conversely, aerosols can regulate turbulence structure through radiative effects (Bharali et al. 2019). Aerosols inhibit turbulence strength and increase atmospheric stratification by diminishing incoming solar radiation and surface heat flux (Barbaro et al. 2013, Wang et al. 2019, Su et al. 2020). The opposite effect is that absorbing aerosols concentrated near surface strengthen atmospheric instability by heating the atmosphere near the surface (Wang et al. 2018a, 2018b, Ma et al. 2020). However, Ding et al. (2016) proposed ‘dome effect’ which demonstrates that black carbon induces heating in the ABL, depresses the development of ABL and consequently enhances the occurrences of extreme haze pollution episodes. Zhang et al. (2022) found that cooling effect dominates warming effect induced by aerosols, generates negative net heating in low background aerosol extinction coefficient experiments, and eventually suppresses turbulence strength.

The characteristics of the SBL, such as turbulence intermittency, significantly influence haze pollution (Wei et al. 2018, Ren et al. 2021a). Turbulent intermittency has a significant effect on the exchanges between the surface to the atmosphere, and causes overestimations of earth-atmosphere exchanges in heavy pollution processes (Ren et al. 2019a, 2019b). This may be one of the possible reasons why simulated peak values of PM2.5 are always lower than their actual observed values in severe haze pollution processes (Wang et al. 2018a, 2018b). Furthermore, the influence on turbulent transport will cause issues related to the applicability of the Monin-Obukhov similarity theory which relates turbulent flux to the gradient profiles of meteorological factors (Mahrt 2010, Acevedo et al. 2014). This can significantly affect the accuracy of numerical simulations (Wilson 2008, Wood 2010). Intermittent turbulence events may cause the instantaneous values of meteorological elements (such as temperature or pollutant concentration) to be lower than their critical threshold values, resulting in frost disasters or harm to human health (Geiss and Mahrt 2015). Salmond (2005) proved that intermittent turbulence plays an important role in the ozone transport within SBLs at night. Further, Wei et al. (2018) revealed that the type of ‘upside-down’ turbulence structure caused by low-level jets intermittently induces increased turbulence, which aids the diffusion of PM2.5. Additionally, Ren et al. (2021a) found that during heavy haze pollution processes, weak turbulence, the quiescent and active period of turbulence intermittency, can affect changes in the rapid accumulation and vertical concentration of PM2.5, defined as the turbulence barrier effect.

Turbulent intermittency is an important feature of turbulence in the SBL (Mahrt 1999). It can be described as the short-term turbulent motion that occurs during periods of relatively weak pulsation or even laminar flow (Van de Wiel et al. 2002), consisted by two elements: long-term quiescent periods and occasionally turbulent motion that interrupts the quiescent periods, called active ‘bursts’ in some papers (Mahlr 1999, Coulter and Doran 2002, Ohya et al. 2008). There are many studies devoted to identify and quantitatively describe turbulent intermittency. For example, some studies look for periods where turbulence occurs intermittently within a time series of wind speed fluctuations or temperature fluctuations (Sun et al. 2004, Ohya et al. 2008) by subjective judgment. Turbulence statistics (Viana et al. 2008, Vindel et al. 2008, Mahrt 2011) or threshold related to turbulent flux (Nakamura and Mahrt 2005, Acevedo et al. 2006, Steeneveld et al. 2006, Drüe and Heinemann 2007, Mahrt 2010) are also applied to identify occurrences of turbulence intermittency. In addition, some other studies have used mathematical methods such as Fourier transform, wavelet transform (Vickers and Mahrt 2003, Muschinski et al. 2004) and the Hilbert-Huang transform technique (Wei et al. 2016, 2017) for analyses. Unfortunately, there are still few widely recognized and unified standards for quantifying turbulent intermittency because of the highly complicated mechanism by which turbulent intermittency is generated (Linden et al. 2020). Researchers currently generally believe that the occurrence of turbulent intermittency can be driven by a variety of different non-stationary motions, and different sub-mesoscale motions may overlap and influence each other (Mahrt 2010, Mortarini et al. 2019).

The turbulent barrier effect link turbulent intermittency to the vertical distribution of pollutants firstly (Ren et al. 2018, 2019c, 2021a). Turbulent intermittency is a natural feature of SBL (Costa et al. 2011), can affect any matter transportation process. Therefore, the turbulent barrier effect should be extended to the turbulent transport of other substances, but the extension was not studied yet. Although the turbulent barrier effect performs well in the pollution process, it is only quantitatively analyzed from the perspective of turbulent motion. The turbulent barrier effect needs to be verified from a more direct perspective, such as turbulent transport, but the verification was not studied yet. The extension and quantitative verification of turbulence barrier effect are two concerns this manuscript going to answer. One of the reasons why the quantitative verification of the turbulent barrier effect is not addressed is that the mass flux of PM2.5 is difficult to obtain (Ren et al. 2020, 2021b). There are some past studies which used the eddy covariance technique to determine the number concentration flux of fine particles in Stockholm (Vogt et al. 2011), London (Harrison et al. 2012), Helsinki (Ripamonti et al. 2013) and so on. However, obtaining the fine particles mass flux is more important for
many applications (Yuan et al. 2016). The number concentration flux of fine particles is always dominated by high concentrations of smaller particles, which cannot represent the mass flux directly. There are less significant studies about high frequency measurements of the mass concentration of fine particles (Yuan et al. 2019) which causes ambiguity about the turbulent characteristics of fine particle concentration and uncertainty in numerical model simulations (Jia et al. 2021).

In this study, CO₂, which has a relatively mature flux acquisition technology, was used as a substitute to extend and verify the turbulent barrier effect. The turbulent barrier effect is crucial for the accurate identification of material transport processes in SBLs. Studying the two concerns this manuscript focuses on, the extension and quantitative verification of turbulence barrier effect, is an important step in the parameterization of the turbulent barrier effect in the future.

2. Data and methods

2.1. Field campaign
Tianjin (39.00° N, 117.21° E, altitude is 3.4 m) is a large city with a population more than 13 million and covering area about 11 300 km² in the North China Plain. Tianjin neighbors Bohai Sea to the east and neighbors Beijing to the northwest. The Tianjin 255 m meteorological observation tower is located in the southern part of Tianjin, situated in the Tianjin Municipal Meteorological Bureau, where surrounded by residential areas. Three types of observational data are used in this manuscript. The first type is turbulence data. Five levels of the tower are equipped with sonic anemometers (CSAT3, Campbell Scientific Co., USA) and infrared gas analyzers (LI-7500, Li-COR Bioscience, USA). These levels are at the heights of 40, 80, 120, 160, and 200 m. Probes of sonic anemometers are all facing north. Turbulence data such as the fluctuation of wind speed, temperature, H₂O and CO₂ were collected at these five levels at high resolution (10 Hz) using a data logger (CR3000, Campbell Scientific, Inc., USA). The second type is meteorological element data collected at 15 levels of the tower. Fifteen humidity and temperature observation sensors (HMP45C, Campbell Scientific Co., USA) were installed on the tower at the heights of 5, 10, 20, 30, 40, 60, 80, 100, 120, 140, 160, 180, 200, 220, and 250 m. Humidity and temperature data were collected every minute. The third type of observational data is pollution monitoring. The PM₂.₅ mass concentration was observed using a 1405-DFTOM (taper element oscillating microbalance) system at two levels, 120 m and 3 m above the ground, and 30-min averaging were performed to remove outliers from the time series.

2.2. Preprocessing of turbulence data
Eddy Pro software was used (Advanced 4.2.1, Li-COR Biosciences, Inc., USA) to process the turbulence data before the intermittent turbulence analysis. At first, the turbulent datasets were pre-processed over an averaging period of 30 min, including error flags, despiking (following Vickers and Mahrt 1997), double rotation (following Wilczak et al. 2001), and detrend (operated using the block averaging method). Additionally, to erase the inaccuracies caused by field observation, datasets meeting any of the following conditions were eliminated: (1) the angle between direction of the sonic anemometer sensing probe and the wind direction is greater than ±120°, (2) the angle between the horizontal anemometer plane and the wind direction is >3°, (3) data groups that contained obvious errors were also removed. In this manuscript, the dataset covering December 15 to 23, 2016 was analyzed in depth.

2.3. Intensity of turbulent intermittency
As introduced in section 1, turbulent intermittency is driven by sub-mesoscale motions. Salmond (2005) indicated that intermittency can be defined by the turbulent and nonturbulent portions of a signal once the criteria for identifying the boundary between them have been established. Ren et al. (2019a) developed an algorithm that uses spectral gaps to distinguish turbulence and sub-mesoscale motion, and then reconstructs the original data through the Hilbert-Huang transform technique to obtain the intensity of turbulent motion and sub-mesoscale motion. Based on this, Ren et al. (2019c) proposed two indicators, local intermittency strength of turbulence (LIST) and intermittency strength (IS), to express the intensity of turbulent intermittency; and some studies (Ren et al. 2019b, Wei et al. 2021, Ren et al. 2021a) have validated the effectiveness of these indicators. However, they are both relative values, and changes in IS can represent changes in the strength of intermittent turbulence during a pollution event, during the transition from a clean state to a polluted state; as described by \( \Delta IS = IS - IS_{clear} \), where \( IS_{clear} \) is the mean value of IS in the 24 h prior to the beginning of the pollution event. \( IS_{clear} \) characterizes the mean non-stationary state of the atmosphere during the clean period and can be regarded as a reference value. \( \Delta IS = 0.1 \) can be a threshold value that delineates the atmospheric states of light pollution and heavy pollution processes (Ren et al. 2019c). However, in any case, the processes of light pollution and heavy
pollution are just a representation, and the threshold essentially reflects the state of intermittent turbulence. In this study, $\Delta IS$ is used for discussion and analysis.

$IS$ directly describes the strength of intermittent turbulence and is defined as (Ren et al 2019c):

$$IS = \frac{V_{\text{smeso}}}{V_{\text{turb}}}$$  \hspace{1cm} (1)

where $V_{\text{turb}} = \sqrt{u_{\text{turb}}^2 + v_{\text{turb}}^2 + w_{\text{turb}}^2}$ is the turbulent velocity scale, $V_{\text{smeso}} = \sqrt{u_{\text{smeso}}^2 + v_{\text{smeso}}^2 + w_{\text{smeso}}^2}$ is the velocity scale of sub-mesoscale motion. $u'_{\text{turb}}(u'_{\text{smeso}})$, $v'_{\text{turb}}(v'_{\text{smeso}})$, and $w'_{\text{turb}}(w'_{\text{smeso}})$ represent the deviations in the turbulent (sub-mesoscale) motion component within each 30 min interval. The algorithm developed by Ren et al (2019a) is used to get $u'_{\text{turb}}(u'_{\text{smeso}})$, $v'_{\text{turb}}(v'_{\text{smeso}})$, and $w'_{\text{turb}}(w'_{\text{smeso}})$ which consists of the following three main steps. First, the empirical mode decomposition method is used to decompose the preprocessed 10 Hz three-dimensional wind speed data into several intrinsic mode functions (IMFs) with different frequencies. Then, the second-order Hilbert spectrum of the wind speed is calculated in every half hour, the position of the spectral gap is found on the spectrum, and the frequency of the spectral gap position is obtained. Finally, the corresponding IMF whose frequency is close to the frequency of the spectral gap is found. All IMFs whose frequency smaller than the frequency of spectral gap are reconstructed to obtain $u'_{\text{turb}}$, $v'_{\text{turb}}$, and $w'_{\text{turb}}$, and the remaining IMFs whose frequency greater than the spectral gap are reconstructed to obtain $u'_{\text{smeso}}$, $v'_{\text{smeso}}$, and $w'_{\text{smeso}}$.

$IS$ refers to the ratio of sub-mesoscale motion to the turbulent components. $\Delta IS$ is the change value in $IS$ during a pollution event. We explain these indicators in the following two situations. When the intensity of sub-mesoscale motion characterized by $\Delta IS$ is relatively strong and the turbulent component is relatively weak, sub-mesoscale motion has an effect on the overall signal. There are two possibilities. One possibility is that turbulent motion is suppressed by sub-mesoscale motion and very weak, which characterizes the quiescent period of turbulent intermittency. At this time, the barrier effect is formed between layers of different heights, which hinders the substance exchange among them. Another possibility is that a turbulent burst is formed in the mesoscale motion characterized by $\Delta IS$ is weak and the turbulence part is strong. The collected signal is almost completely turbulence and the turbulent energy is strong; thus, the turbulence barrier effect is weak.

In this study, to more accurately capture small-scale intermittent changes in turbulence, we combined the data for the preceding and post 10 min before and after each 10 min interval, to form a data set with 30 min resolution, for data analysis using the algorithms described above. Thus, we obtained 1152 data points for each variable, for the period between December 15 to December 23, 2016. Another point that needs to be emphasized is that all the turbulence variables (e.g., TKE, standard deviation, turbulent flux) used in this study exclude consideration of sub-mesoscale motion. Because the goal of this work was to focus on vertical intermittent turbulence, only the $w$ component of these indicators was used in subsequent analysis.

### 2.4. Quadrant analysis

Quadrant analysis is a widely-used method for investigating the characteristic variables of turbulent transport (Dupont and Patton 2012, Wallace 2016, Dupont et al 2019). Quadrant analysis decomposes fluxes into four quadrants (i.e., Q1, Q2, Q3, and Q4) based on the sign of the turbulent deviations of quantities. The definition of each quadrant in our analysis was the same as previous studies. The plotted diagram for the four quadrant definitions for CO$_2$ flux is shown in figure 4.

To obtain the contributions of CO$_2$ fluxes in each quadrant, the time averaged fluxes in each quadrant are calculated using:

$$\overline{w'c_{o2}}_{k=1:4} = \frac{1}{T_N} \int_0^{T_N} \overline{w'c_{o2}I_{k=1:4}} dt$$ \hspace{1cm} (2)

where $k$ indicates the quadrant ($k = 1, 2, 3, 4$), and $T_N$ is the averaged period, which is 30 min here; where:

$$I_{k=1:4} = \begin{cases} 1, & \text{if } \overline{w'c_{o2}} \text{ is in quadrant } k \\ 0, & \text{otherwise} \end{cases}$$ \hspace{1cm} (3)
Then, the contributions of the CO$_2$ fluxes within quadrant $k$ are defined by:

$$E_{k|Q}^{w'co_2} = \frac{|w'co_2|_{k|Q}}{\sum_{Q=1}^{4} |w'co_2|_{Q}}$$  

(4)

The sum of the total contributions of the four quadrants is 1.

3. Results

3.1. Trends of CO$_2$ concentrations and turbulent variables during the six study stages

As described above, two concerns of this study are extension and verification of turbulent barrier effect. CO$_2$, which has a relatively mature flux acquisition technology, was used as a substitute to extend and verify the turbulent barrier effect. The entire haze pollution is divided into six stages (S1, S2, S3, S4, S5, and S6, as shown in figure 1). The characteristics of the meteorological elements of each stage are shown in figure 1. In this section, the trends of CO$_2$ concentrations at five heights and the average states of turbulence variables in each stage are carefully introduced.

During S1, the horizontal wind speed was relatively high, and the wind direction was mostly southwesterly (as shown in figures 1(c) and (d)). PM$_{2.5}$ at 120 m and on the surface increase simultaneously. The CO$_2$ concentrations at the five heights varied slightly and synchronously, and had a relatively small value of standard deviation (as shown in figure 3(b)). The temperature profile indicated that there was no inversion, the turbulence developed well at noon. Correspondingly, the $\Delta S_{\omega}$ profile during S1 shows that the intensity of turbulent intermittency at different heights was low, with a value of less than 0.1. TKE decreased with height. There was no turbulent barrier effect, and the observed values of the CO$_2$ flux at different heights of the tower represent the normal state. As shown in figure 3(c), the values of CO$_2$ flux at the heights of 40 m (0.18 mg m$^{-2}$ s$^{-1}$) and 80 m (0.17 mg m$^{-2}$ s$^{-1}$) during S1 were close, but less than the values at 120 m (0.29 mg m$^{-2}$ s$^{-1}$) and 160 m (0.32 mg m$^{-2}$ s$^{-1}$). It is speculated that the layer below 80 m belongs to the roughness layer of the urban ground surface (Sha et al 2021), while the layer above 80 m belongs to the constant flux layer, in the traditional sense.
During S2, the wind speed decreased (less than 2 m s\(^{-1}\)), the humidity increased, and the concentration of PM\(_{2.5}\) at the surface and 120 m height, increased significantly. The CO\(_2\) concentrations at different heights were also increased, compared to S1, with corresponding increases in their standard deviations. However, the magnitudes of the increases varied across heights. The changes in the trends of the CO\(_2\) concentrations at heights of 160 m and 200 m were more consistent, which was different with that at the other three heights. Weak winds across the layer mean less turbulence due to wind shear. $\Delta IS_w$ increased significantly during S2, and the development of the turbulent barrier effect was enhanced, turbulent flux and TKE close to zero.

During S3, the wind speed was less than 5 m s\(^{-1}\), and temperature inversion occurred. There was a rapid decrease and increase in PM\(_{2.5}\) concentrations on the surface and at 120 m. The CO\(_2\) concentrations at heights of 40 m, 80 m, 120 m, and 160 m underwent similar declines and rapid increases. However, the variations in CO\(_2\) concentrations at 200 m were relatively small and stable. The turbulence was weak with low wind and temperature inversion. There was a maximum value of $\Delta IS_w$ at a height of 120 m, indicating the presence of a stronger turbulent barrier. Ren et al. (2021a) believed that the temporary disappearance of the turbulent barrier effect at heights of 40 m and 120 m helped the diffusion of PM\(_{2.5}\). However, the turbulent barrier effect was once again strengthened, which prevented pollutants from continuing to dissipate and the transition to a clear state.

During S4, the wind speed did not change significantly, and the humidity decreased slightly. The PM\(_{2.5}\) concentrations at 120 m were reduced to a clean state, but the PM\(_{2.5}\) concentrations on the surface were reduced to 280 $\mu g$ m\(^{-3}\) in a short time and subsequently maintained. The CO\(_2\) concentrations at the heights of 40 m, 80 m, and 120 m also exhibited a similar rapid declining pattern (the maximum decrease was up to 0.5 g m\(^{-3}\)). However, unlike the PM\(_{2.5}\) concentration at the 120 m height, they directly transitioned to clean states. Interestingly, the CO\(_2\) concentrations at 160 m and 200 m also experienced a significant drop, but later. The CO\(_2\) concentrations at all five heights increased rapidly at 04:00 on December 20, as demarcated by the blue dotted line in figure 2. Ren et al. (2021a) speculated that the disappearance of the turbulent barrier at 80 m, 120 m, 160 m, and 200 m or higher caused a sharp decrease in PM\(_{2.5}\) at 120 m. However, the presence of a turbulent barrier at heights of 40 m or lower hinders the diffusion of surface contaminants. Moreover, it is also speculated that at the moment demarcated by the blue dotted line, the reopening of the turbulent barrier at various heights.
leads to the downward transport of higher-level pollutants, which leads to an increase in PM$_{2.5}$ at 120 m at the beginning of S5.

The rapid change in CO$_2$ concentrations at different heights during S3 and S4 also increased the standard deviations. The larger downward flux values during S3 and S4 were linked to the penetration of the turbulent barrier effect caused by the intermittent bursts of turbulence, the analysis of which is described in detail below.

During S5, the PM$_{2.5}$ concentrations at 120 m increased, but the PM$_{2.5}$ concentrations on the ground changed slightly. The CO$_2$ concentrations at the five heights also increased after a period of fluctuation, and were similar to the PM$_{2.5}$ concentrations at 120 m. During S6, wind speed increased (greater than 5 m s$^{-1}$). The PM$_{2.5}$ concentrations at 120 m and on the ground transitioned to a clean state. The CO$_2$ concentrations at different heights were significantly reduced. The large value of TKE during S6 was corresponded to the large value of wind speeds (see figure 1). During the entire pollution process, the CO$_2$ concentration at a height of 80 m was higher than that at 40 m, which may be due to the local CO$_2$ emission background value of the city site.

In the entire scenario, there were both similarities and differences between the dynamic trends in CO$_2$ and PM$_{2.5}$ concentrations at different heights. Ren et al. (2021a) explained the turbulent barrier effect and proposed a mechanism for intermittent explosive turbulent transport at each stage. In the following analysis, we use CO$_2$ flux observations at five heights to verify if the interpretation and conjecture of the turbulent barrier effect is reasonable.

3.2. Quantitative verification of the turbulent barrier effect

During S1, the $\Delta IS_w$ values at each height were all less than 0.1, and only exceeded 0.1 at a few moments that were not sustained. A turbulent barrier effect did not exist. During S2, the value of $\Delta IS_w$ gradually increased at different heights and exceeded the threshold value of 0.1. At 08:00 on December 17, the intensity of turbulence intermittency at the heights of 40 m, 80 m, and 120 m began to develop gradually. At approximately 10:00 on December 17, the turbulence intermittency at 160 m and 200 m also began to develop, and their intensities were stronger (the $\Delta IS_w$ value was greater) and lasted longer, indicating that the turbulent barrier effect was also stronger.

From 12:00 on December 17 to 07:00 on December 18 (the period demarcated by Square 1 in figure 4), the turbulent barrier effect at the heights of 160 m and 200 m was strong and was sustained, impeding the transport of matter below 160 m. The value of $\overline{w^2CO_2}$ was significantly lower than that of S1. The average values of $\overline{w^2CO_2}$ at 200 m and 160 m were $-0.02$ mg m$^{-2}$ s$^{-1}$ and $0.012$ mg m$^{-2}$ s$^{-1}$, respectively; and the summed values of $\overline{w^2CO_2}$ at 200 m and 160 m were $-2.5$ mg m$^{-2}$ s$^{-1}$ and $1.51$ mg m$^{-2}$ s$^{-1}$, respectively. The turbulent transfer was close to zero, and the CO$_2$ concentrations at these two heights remained stable. From a vertical comparison perspective, the turbulent barrier effects at 40 m, 80 m, and 120 m were weaker than those at 160 m and 200 m, and CO$_2$ can still be transported within the region of air between 40 m and 120 m. However, the upward transfer of CO$_2$ cannot continue after encountering the turbulent barriers at 160 m and 200 m heights, resulting in an increase in the concentration of CO$_2$ at the three heights of 40 m, 80 m, and 120 m. Moreover, there were differences in the time at which the concentrations of CO$_2$ increased at the three heights: 40 m first, 80 m followed, and 120 m lagged slightly behind the first two. This time lag was reasonable.

From 08:00 to 12:00 on December 18 (the period demarcated by square 2 in figure 3), the intensity of the turbulent barrier effects at the height of 160 m weakened. During this period, the intensity of turbulent intermittency at the heights of 40 m, 80 m, 120 m, and 200 m were maintained at low levels. The synchronous turbulent barrier at these five heights was broken and penetrated, and there was a brief upward transport of CO$_2$, which caused its concentrations at all five heights to rise.

At approximately 12:00–24:00 on December 18 (the period demarcated by Square 3 in figure 3), the turbulent barrier effect at heights 80 m and above were enhanced, and the vertical transportation of CO$_2$ was weakened. The $\Delta IS_w$ at the 40 m height also increased, but the turbulent barrier effect was relatively weak. The transportation of CO$_2$ was reduced under the obstruction of the turbulent barrier, and the concentration of CO$_2$ at 40 m increased. However, the concentrations of CO$_2$ at heights above 80 m did not change significantly. The turbulent transport of CO$_2$ during S2 thoroughly confirmed the conclusion of Ren et al. (2021a).

At the beginning of S3, at around 02:00 on December 19, the $\Delta IS_w$ at the heights of 40 m and 80 m decreased, the strength of the turbulent barrier weakened, the downward turbulent transport of CO$_2$ appeared, and the concentration of CO$_2$ began to decrease. During this process, the total fluxes at 40 m and 80 m were $-6.70$ mg m$^{-2}$ s$^{-1}$ and $-4.79$ mg m$^{-2}$ s$^{-1}$, respectively; and the mean fluxes at 40 m and 80 m were $-0.32$ mg m$^{-2}$ s$^{-1}$ and $-0.23$ mg m$^{-2}$ s$^{-1}$, respectively. The net flux at the two heights was still downward, indicating that CO$_2$ was transported to the lower atmosphere. The descent time of $\Delta IS_w$ at the height of 120 m was later than that for 40 m and 80 m, and there was also downward CO$_2$ turbulent transport, which corresponded to the decrease in CO$_2$ concentrations at a later time. From 02:00 to 07:00 on December 19, the turbulent barrier effects at the heights of 160 m and 200 m were strong, and the concentrations of CO$_2$ did not change significantly.
From 07:00 to 12:00 on December 19 (the period demarcated by Square 4 in figure 4), the intensity of turbulence intermittency at the heights of 40–160 m decreased simultaneously for a short period of time, and the turbulent barrier dissipated. It can be seen that there were strong intermittent turbulent bursts at the height of 160 m, which transported turbulent CO₂ downward. The net downward CO₂ flux caused the CO₂ concentrations at 40 m, 80 m, and 120 m heights to increase instead. The analyses for S3 confirmed the speculations regarding the changes in the intensity of the turbulent barrier effects and the intermittent turbulent transport when PM₂.₅ concentrations change.

Over the remaining duration of S3, the turbulent barrier effect between 40 m and 160 m was further strengthened, especially at the 120 m height where a stronger turbulent barrier was sustained over a long period of time, and where material transfer was hindered. The CO₂ concentrations between 40 m and 120 m did not change significantly. Over the course of S3, the turbulent barrier at the 200 m height was strong in the early stages, and the turbulent barrier effects at the heights of 160 m and 120 m developed later in the stage. This caused the CO₂ concentrations at 200 m not to change much, regardless of the changes in the CO₂ concentrations at other heights.

At the beginning of S4, the turbulent barrier between 120 m and 160 m was briefly penetrated, and the strong turbulent transport of CO₂ from 40 m to 160 m was observed, accompanied by corresponding reductions in the CO₂ concentrations. The CO₂ concentrations at the 120 m height exhibited the largest decreases. However, with the redevelopment of the 40 m, 80 m, 120 m turbulent barrier, the net downward CO₂ transportation flux from the 160 m height led to increases in the CO₂ concentrations between 40 m and 120 m, while the CO₂ concentration at 160 m decreases. During this process, that is, from 16:00 to 22:00 on December 19 (the period demarcated by Square 5 in figure 4), the turbulent barrier at the 200 m height was always present, and the CO₂ concentrations did not change significantly. The downward turbulent transport of CO₂ during this period also confirmed the speculation of Ren et al. (2021a) that there is indeed a downward transportation flux caused by the weakening of the turbulent barrier effect, and which corresponds to decreases in the concentrations of substances at various heights. However, there was also a slight difference. While the PM₂.₅ concentrations at 120 m fell to clean levels during this process, the CO₂ concentration at 120 m did not drop completely to 0. This may be related to the differences in the source distributions of PM₂.₅ and CO₂.

At 4:00–10:00 on December 20 (the period demarcated by the square 6 in figure 4), the turbulent barrier between 40 m and 160 m disappeared for a short time. Although the CO₂ concentration at the 160 m height was absent, the downward CO₂ flux from 40 m to 120 m proved that there was a downward transport in substances, causing the CO₂ concentrations at these three heights to increase again. This is in full agreement with the speculation of Ren et al. (2021a): that the turbulent downward transport of PM₂.₅, from a higher level causes an increase in the PM₂.₅ concentrations at 120 m to increase. During this process, the turbulent barrier at 200 m also weakened, corresponding to the upward transportation of CO₂ and the reductions in the CO₂ concentrations, which may be associated with the changes in the barrier effects above 200 m, but may be irrelevant to the changes below 200 m.
During S5, the strength of the turbulent barrier between 40 m to 120 m underwent continuous development and changes, and the CO\(_2\) concentrations at these three heights increased slowly. During S6, the intensity of turbulent intermittency at all five heights was weakened to less than the threshold, and there was high upward transportation of CO\(_2\) and decreases in the CO\(_2\) concentrations. The relationship between turbulent transportation and the barrier effect during S5 and S6 also confirmed the results of Ren et al. (2021a).

**Figure 4.** The time series of $\Delta IS_w$ (red solid line), $\bar{w} CO_2$ (light blue bar) and CO\(_2\) (green solid line) at (a) 200 m, (b) 160 m, (c) 120 m, (d) 80 m, and (e) 40 m, from December 15 to December 22, 2016. The blue solid lines represent the beginnings and ends of the six stages.
3.3. The characteristics of turbulent transfer under the influence of the turbulent barrier effect

The above analysis shows that the enhancement and sudden disrupting process of the turbulent barrier effect will significantly affect the turbulent transport of materials. The turbulent transport during this period was different from that for normal conditions. In order to discuss this issue, we chose the turbulent transport of CO\textsubscript{2} during S\textsubscript{1} as the normal state. The time periods demarcated by the six squares in figure 4 represent the cases where the turbulent barriers were either strengthened or broken. Figure 5 illustrates the differences between the two.

S\textsubscript{1} belongs to the normal state, and the proportions of Q\textsubscript{1} and Q\textsubscript{3} were significantly high; that is, the transportation caused by the gradient motion contributed most to the total flux. During the period demarcated by Square 1, the turbulent barrier effect gradually developed and strengthened, especially at the 200 m height. It can be seen that the significant advantage of the gradient motion is lost at the height of 200 m, and the proportion of the gradient motion at other heights also decreased. During the time demarcated by Square 2, the turbulent barrier was briefly penetrated, and there was no significant advantage in the gradient motion compared with the counter-gradient motion. During the period demarcated by Square 3, the turbulent barrier developed again. Similar to the case for Square 2, there was no significant advantage in the gradient motion compared with the counter-gradient motion, even though the counter-gradient motion contributed slightly more at 200 m and 120 m. The situation changed during the periods demarcated by Squares 4, 5, and 6. During this time, the turbulent barrier was briefly penetrated at heights of 40 m, 80 m, 120 m, and 160 m, and even at 200 m during the period denoted by Square 6. However, unlike the results obtained from the periods denoted by Squares 1, 2, and 3, the gradient motions still dominate during the periods demarcated by Squares 4, 5, and 6. This may be related to the strong turbulent exchange caused by the intermittently increasing turbulence in these three cases. In the case of strong turbulent transport capacity, the gradient motion still dominated. The turbulent barrier at the 200 m height was not broken during the period demarcated by Squares 4 and 5, and the counter-gradient motions and the gradient motions had equal proportions (as shown in figures 5(e)–(f)), which illustrates these observations from the side profile.

4. Discussion and conclusion

Under calm and steady weather conditions with low wind speeds, turbulent intermittency occurs frequently in the atmospheric boundary layer, and can significantly weaken the turbulent diffusion of matter and energy between the surface and atmosphere. Ren et al (2021a) proposed the physical concept of the turbulence barrier effect, to define the phenomenon in which turbulence may dissipate at certain heights, forming a laminar flow.
that acts like a barrier layer that hinders vertical transmission during periods of heavy haze. Although the effectiveness and important influence of the turbulent barrier effect have been revealed through the use of turbulence data and the PM$_{2.5}$ from five elevations, observations at the ground and 120 m in Ren et al (2021a), quantitative verification and expansion in the application of the turbulent barrier effect are required. This was the main question discussed in this manuscript. Because of the lack of PM$_{2.5}$ flux data, a relatively easy-to-obtain substitute, carbon dioxide (CO$_2$) was used as a 'tracer' to verify and quantify the targeted patterns using data from five levels of the Tianjin 255 m meteorological tower. The results revealed that the vertical dynamics in the turbulent barrier effect were consistent with that of the concentration and flux of CO$_2$. This means that the application of the turbulent barrier effect can be extended to other mass-transfer processes. The analysis also found that the proportion of counter-gradient transport increased during the frequent occurrence of the turbulent barrier effect. Locally, as far as Tianjin is concerned, the turbulent barrier effect at heights of 200 m and above are often stronger under static and stable weather.

In order to summarize the findings of this study, figure 6 depicts the schematic diagram of the relationships between the intensity of the turbulent barrier, the turbulent transport of material, and the trends in the concentrations of the simple case where the material source is from ground level. As figure 6(a) shows, in the normal turbulent state, there is no turbulent barrier, substance concentrations decrease with height, and the turbulent flux remains unchanged in the near-surface layer. Under calm and steady weather conditions with low wind speeds, such as those associated with heavy haze pollution, the intensity of the turbulent barrier effect varies across different heights. The enhancements in the turbulent barrier effect at specific heights will result in the obstruction of turbulent vertical transportation at that height. As shown in figure 6(b), the turbulent barrier effect increases with height, leading to a larger increases in the concentrations of matter in the lower layer, smaller increases in the concentrations of matter at higher layers; turbulent transport at the lower layer is stronger than that at higher layers. If the intensity of the turbulent barrier effect at different heights is simultaneously weakened, it will cause the material to be transported in a certain region of air. As shown in figure 6(c), the intensity of the turbulent barrier at h2 and h3 are simultaneously weakened, which will lead to the mixing of substances in this space, uniform concentration, and local flux. In addition, as shown in figure 6(d), the weakening of the turbulent barrier at h2 will result in an increase in turbulent transport and a decrease in the concentrations of matter at this height.

This work validates the turbulent barrier effect and is an important foundation of its parameterization in the future, which will aid the accurate identification of the transport process for matter in the SBL or under extreme weather conditions such as heavy pollution events.

**Data availability statement**

The data generated and/or analysed during the current study are not publicly available for legal/ethical reasons but are available from the corresponding author on reasonable request.
Competing interests

The authors declare that they have no conflict of interest.

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