Dear Editors and Reviewers:

Thank you all for your comments concerning our manuscript entitled “Mixing layer transport flux of particulate matter in Beijing, China” (Manuscript ID: acp-2019-141). The comments were all valuable and very helpful for revising and improving the manuscript and provided important guiding significance for our research. We have studied the comments carefully and made corrections that we hope will be met with approval. The revised parts of the manuscript are shown using the “Track Changes” feature in Word. Below, we have provided the reviewers’ comments for ease of reading and have added our response after each comment.

List of Responses

Responses to the comments from Reviewer #1

We would like to thank you for your comments and helpful suggestions. We have revised the manuscript accordingly.

General Comments:

To quantifying the transport flux of atmospheric pollutants for understanding the causes of atmospheric pollution levels and development of decisions regarding the prevention and control of atmospheric pollution, the mixing layer height and wind profile inside the mixing layer were measured by ceilometer and doppler wind radar, respectively. The variation characteristics of atmospheric transport capacity (TC) were analyzed on this data base: TC is strongest in spring and weakest in autumn. The TC influence on the $PM_{2.5}$ concentration was determined and there shows a strong inverse correlation between the $PM_{2.5}$ and TC in spring, autumn and winter and a weak positive correlation in summer. The transport flux (TF) of fine particles in Beijing is highest in spring and lower in the other three seasons. The transport occurs mainly between 14:00 and 18:00 LT. The TF was large in the pollution transition period and decreased during heavy pollution periods.

Comment 1:

The application of TC, TF and VC should be explained in more detail: why these parameters are used and which advantages it provides in comparison to alternative parameters.

Response 1:

Thank you for your helpful suggestion. After careful consideration, we think that “atmospheric transport capacity” is prone to ambiguity, so we changed this term to “atmospheric dilution capability”. Atmospheric dilution is composed of vertical and horizontal dilutions, which can be characterized by the mixing layer height (MLH) and wind speed in the mixing layer (WS_{ML}), respectively. The ventilation coefficient (VC) is obtained by combining MLH and WS_{ML} and can be used for a comprehensive evaluation of the vertical and horizontal dilutions, where a higher VC indicates a stronger dilution capability. The TF represents the transport flux of $PM_{2.5}$, which can quantify the amount of pollutants passing through the area to assess the impact of regional transport. To avoid confusion, changes were made in the paper.

Comment 2:

It is concluded that the transportation influence in southern regions is of higher influence in the transition period of pollution, while local emissions are more important in the heavy pollution period. My main concern is why the whole discussion with TC, TF and VC up to section 3.2 is without wind direction. In section 3.3 it would be helpful to discuss MLH also.

Response 2:

Thank you for your helpful suggestion. After careful consideration, we have revised the structure of the paper according to your suggestion. Section 3.1 mainly discusses the seasonal and diurnal variations of the atmospheric dilution capability and $PM_{2.5}$ concentration; section 3.2 mainly discusses the evolution of the TF, both temporally and spatially; and section 3.3 analyzes the evolution of the TF under different pollution degrees in detail. The revised structure will make it easier for readers to understand. Thank you very much for your suggestions.

In addition, we have added the evolution of the MLH under different pollution degrees in section 3.3 as suggested. We found that the MLH decreases gradually with the worsening of the pollution (Fig. 1). This result also supports the conclusion that the transport is weak during heavy pollution.

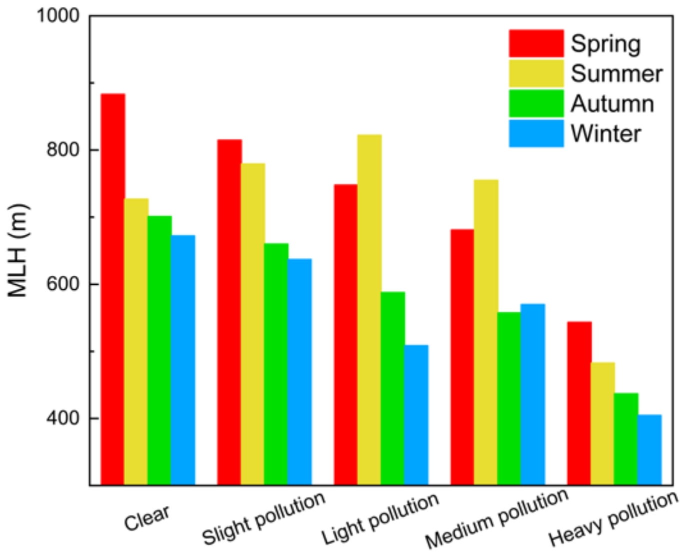

Fig. 1 Mixing layer height under different degrees of pollution in different seasons in Beijing.

Comment 3:

The conclusions are a summary and in this summary no relation to the existing knowledge / papers are given. What is new and what is supported by this study? The paper addresses relevant scientific tasks. The paper presents novel concepts, ideas and tools. The scientific methods and assumptions are valid and clearly outlined so that substantial conclusions are reached. The description of experiments and calculations allow their reproduction by fellow scientists.

Response 3:

Thank you for your helpful suggestion. Joint prevention and control have been recommended for a long time to solve the problem of heavy pollution in northern China. Even so, no concrete implementation plan has been established. To break through this embarrassing situation, this study quantifies the transport flux to explain the time period when the transport occurs, the main areas affected in Beijing and the height of transport. The important role of transport in the initial period of pollution is emphasized. The innovation of this study has been added to the conclusion.

Comment 4:

The quality of the figures is good. The figure captions should be improved so that these are understandable without the overall manuscript: terms must be explained, description of parameters.

Response 4:

Thank you for your helpful suggestion. According your suggestion, we added more detail to make the figures more readable, such as descriptions of the parameters and explanations of the abbreviations.

Specific Comments:

Comment 1:

Line 46: The values are valid for which time period?

Response 1:

Thank you for your helpful suggestion. The phrase has been revised to “the annual average fine particulate matter concentration”.

Comment 2:

Line 57: How TC is defined? Reference? Line 59: What about wind direction? Line 64: How VC is defined? Reference?

Response 2:

Thank you for your helpful suggestion. As mentioned in the response to comment 1 in the “General Comments”, we changed “TC” to “atmospheric dilution capability”. Definitions of the atmospheric dilution capability and VC have also been described in the beginning of section 2.4. The wind direction in this study refers to the average wind direction in the mixing layer. For ease of understanding, we modified the expression to “average wind direction in the mixing layer”.

Comment 3:

Line 81: When this happened?

Response 3:

Thank you for your helpful suggestion. This event happened in 2016, and this information has been added to the paper.

Comment 4:

Lines 110 – 113: This explanation is not correct. Explain clearly what do you mean.

Response 4:

Thank you for your helpful suggestion. This section was removed during the revision process.

Comment 5:

Line 116: What is $-(d\beta/dx)$?

Response 5:

Thank you for your helpful suggestion. β is the backscatter coefficient, and x is the distance between the lidar and scattering volume (Münkel et al. 2007). $-(d\beta/dx)$ represents the maximum negative gradient value in this paper. Considering that $-(d\beta/dx)$ has no practical meaning in the paper, it has been deleted.

Comment 6:

Line 128: time resolution not time accuracy

Response 6:

Thank you for your helpful suggestion. This section was corrected the revision process. The phrase "A time accuracy of 1 h" has been revised to "hourly".

Comment 7:

Lines 142 – 144: Why this is an explanation? Height profile instead of "by height"

Response 7:

Thank you for your helpful suggestion. Although previous studies have shown that the concentration of particulate matter in the mixing layer is basically uniform, there are still large differences in some time periods, especially in time periods with transport effects. Based on your suggestion and that of Reviewer 2, we find it inappropriate to so rashly use the near-surface $PM_{2.5}$ concentration as the concentration in the mixing layer. Because the ceilometer can measure the atmospheric backscattering coefficient, it is possible to obtain the vertical profile of the particles. Therefore, in the revised draft, we analyzed the relationship between the backscattering coefficient at 100 m measured by the ceilometer and the near-surface $PM_{2.5}$ concentration, discussed their correlations in different seasons, and obtained the fitting curves of different seasons. Using these four equations, we obtained the $PM_{2.5}$ concentration at different heights in different seasons. According to this result, we have recalculated the TF in the revised draft.

Comment 8:

Line 353: How $PM_{2.5}$ concentration is related to photochemical reactions?

Response 8:

Thank you for your helpful suggestion. Through subsequent analysis, we found that our previous inference was wrong. Considering that this part is not closely related to the topic, it has been deleted from the manuscript.

Comment 9:
Line 366: concentration column? What do you mean? Technical corrections Indicate if there are papers in Chinese.
Response 9:
Thank you for your helpful suggestion. We apologize for this mistake. We have revised "concentration column" to "column concentration".

Responses to the comments from Reviewer #2

We would like to thank you for your comments and helpful suggestions. We revised our manuscript accordingly.

General Comments:
The current study explores the seasonal source of $PM_{2.5}$ pollution in Beijing by quantifying the transport flux based on measurements of mixing layer height and wind profile. In particular, this study raises two questions that are rarely addressed in previous studies: (1) effects of ventilation coefficient on $PM_{2.5}$, and (2) observational quantification of transport fluxes. This topic is of broad interest to both the scientific community and policy-makers. The datasets analyzed in the study is valuable. However, the current analyses do not clearly address the questions raised in the beginning. In addition, the data and method section require some clarification. Therefore, I recommend major revision.

Specific Comments:
Comment 1:
I suggest changing the second question to emphasize its scientific merit. By quantifying transport fluxes from observation, what scientific question do you want to address?
Response 1:
Thank you for your helpful suggestion. We have emphasized the scientific merit of the second question and added it to the introduction, as follows:
Although the problem of heavy pollution in northern China has improved in recent years, regional pollution problems remain, especially in the Beijing-Tianjin-Hebei region (Shen et al. 2019). To solve the regional pollution problem, joint prevention and control have been recommended for a long time. Many studies on regional transport have been carried out, but most observational studies cannot easily quantify the transport flux due to the lack of particle and wind vertical profiles, and it is still unclear when we need to control the emission sources and in which areas. In this study, we used the backscattering coefficient measured by a ceilometer and wind profile to quantify the transport fluxes to solve the problems mentioned above.

Comment 2:

Section 2.2 describe the method to determine MLH. Although details are provided in earlier papers, necessary steps should be clearly mentioned in the current paper, e.g. line 113-115 averaging the profile over time? If so, over what time window, daily, hourly?

Response 2:

Thank you for your helpful suggestion. The text has been revised to “the MLH was calculated by the improved gradient method after smoothly averaging the profile data”. More details are as follows:

Because the lifetime of the particles can be several days or even weeks, the distribution of the particle concentration in the MLH is more uniform than that of the gaseous pollution. However, the particle concentration in the mixing layer and that in the free atmosphere are significantly different. In the attenuated backscatter coefficient profile, the position at which a sudden change occurs in the profile indicates the top of the atmospheric mixing layer. In this study, we used the Vaisala software product BL-VIEW to determine the MLH. The time averaging is dependent on the current signal noise. Height averaging intervals range from 80 m at ground level to 360 m at a 1600 m height and beyond. Additional features of this algorithm, which is used in the Vaisala software product BL-VIEW, include cloud and precipitation filtering and outlier removal. Because the aerosol concentrations are particularly low above the BLH and the BLH in the Beijing area is usually lower than 4 km, we halved the detection range to 7.7 km to reinforce the echo signals and reduce the detection noise.

Comment 3:

Section 2.4 cited a previous study to support the assumption that backscattering coefficient is relatively uniform in the mixing layer. I think your ceilometer observations include backscatter profile. Does your data quantitatively support this assumption?

Response 3:

Thank you for your helpful suggestion. Although previous studies have shown that the concentration of particulate matter in the mixing layer is basically uniform, there are still large differences in some time periods, especially in the time periods with transport effects. Based on your suggestions and those of Reviewer 2, we find it inappropriate to so rashly use the near-surface $PM_{2.5}$ concentration as the concentration in the mixing layer. Because the ceilometer can measure the atmospheric backscattering coefficient, it is possible to obtain the vertical profile of the particles. Therefore, in the revised draft, we analyzed the relationship between the backscattering coefficient at 100 m measured by ceilometer and the near-surface $PM_{2.5}$ concentration, discussed their correlations in different seasons, and obtained the fitting curves of different seasons. Using these four equations, we obtained the $PM_{2.5}$ concentration at different heights in different seasons. According to this result, we have recalculated the TF in the revised draft.

Comment 4:

On line 156-158 and following statements, what is the number behind the ï´C´s sign?

Response 4:

Thank you for your helpful suggestion. I guess you mean “±”. The number after the “±” represents the standard deviation, a measure of the dispersion of the data. An explanation has been added where the notation first appeared.

Comment 5:

I suggest using the same color scheme for each season in Fig. 2 and Fig. 3.

Response 5:

Thank you for your helpful suggestion. The color scheme has been unified.

Comment 6:

Why didn’t you show diurnal variations and growth rates of $PM_{2.5}$ in Fig. 2? It seems directly relevant to the first scientific question.

Response 6:

Thank you for your helpful suggestion. The diurnal variations of the $PM_{2.5}$ and the corresponding analysis have been added. More details are as follows:

Notable differences are present when we compare the dilution-related parameters to $PM_{2.5}$. The daily maximum $PM_{2.5}$ concentrations in the spring, summer, autumn and winter were 73 μg m^{-3} (11:00 LT), 56 μg m^{-3} (09:00 LT), 78 μg m^{-3} (23:00 LT) and 101 μg m^{-3} (01:00 LT), respectively. The differences between the maximum and minimum were 14 μg m^{-3}, 10 μg m^{-3}, 20 μg m^{-3} and 38 μg m^{-3}, respectively. Thus, the diurnal variation of $PM_{2.5}$ can be divided into two categories: (1) the highest value occurs in the midday in the spring and summer and the overall change is small and (2) the highest value occurs during the night in the autumn and winter and differs greatly from the lowest value (Fig. 2). The main causes of air pollution are local emissions and regional transport. Thus, these results indicate that there is a greater local contribution in the autumn and winter and higher regional transport in the spring and summer.

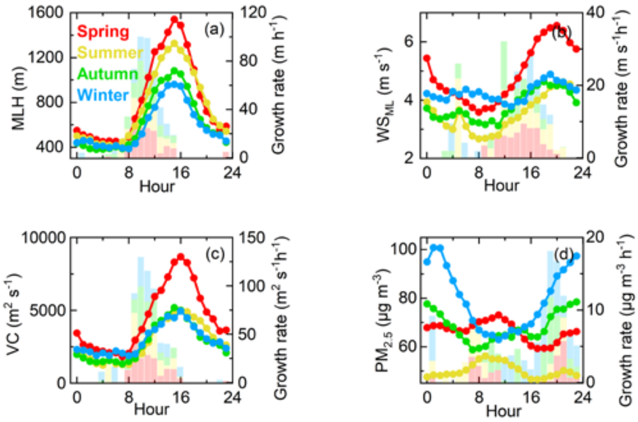

Fig. 2 Diurnal variations and growth rates of the MLH (a), WS_{ML} (b), VC (c) and $PM_{2.5}$ (d) in the spring, summer, autumn and winter in Beijing. Diurnal variations are represented by lines and scatters.

Growth rates are represented by columns, and only positive values are shown in the figure.

Comment 7:

In Fig.3, it is worth discussing higher frequency of high VC (> 10^3 m^2 s^{-1}) in winter, is it due to high wind speed associated with frontal passage?

Response 7:

Thank you for your helpful suggestion. We agree with you. In winter, when the Siberian High transits, strong northwest winds prevail in the Beijing area (Fig. 3), resulting the higher frequency of the VC in the range of 1000-2000 m^2 s^{-1}. We explained this point in section 3.1.1 of the revised draft.

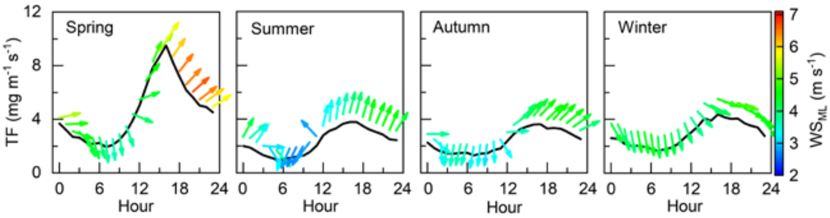

Fig. 3 Diurnal variations in the mixing layer transport flux of $PM_{2.5}$ and transport direction during different seasons in Beijing.

Comment 8:

In Fig.4, it seems to me that the dominant southerly wind partly explains the positive correlation between wind speed and $PM_{2.5}$ in summer.

Response 8:

Thank you for your helpful suggestion. The southern wind generally appeared at 12:00-2:00 LT, and the high $PM_{2.5}$ concentration generally appeared at 6:00-13:00 LT; therefore, there was no significant relationship between the two. In addition, due to the improper discussion of this section in the original text, we have deleted this section to avoid confusion.

Comment 9:

I don't think the conclusion on lines 289-294 that southerly wind is "dirtier" directly comes from Figure 5 and 6 Flux variation comes from $PM_{2.5}$ and wind speed, it could be that southerly wind are generally stronger. In order to demonstrate this point, it will help to add $PM_{2.5}$ fields in Figure 5 and Figure 6. Another way to demonstrate this conclusion is to show wind rose and flux rose, and $PM_{2.5}$ composite in different wind directions.

Response 9:

Thank you for your helpful suggestion. According your suggestion, the diurnal variation of the $PM_{2.5}$ concentration and the wind radar were added, and we found that the level of the TF is determined by two factors, the WS and $PM_{2.5}$ concentration. In the spring, summer and autumn, the strong south wind prevails in the afternoon. As the south wind is often accompanied by a high $PM_{2.5}$ concentration (Fig. 4), the TF is high. In the winter, the whole day is dominated by westerly and northerly winds. Although the northerly winds are strong, the TF is not high due to the low $PM_{2.5}$ concentration. Generally, a high WS means fast mixing, and the corresponding MLH is also high. At this time, the TF is mainly controlled by the WS. When the WS is low, the mixing speed is slow, and the MLH is low. At this time, the TF is mainly controlled by the $PM_{2.5}$ concentration. From the above analysis, it can be inferred that if the MLH and WS gradually decrease with the worsening of the pollution, the mixing layer TF is controlled by the WS first and then by the $PM_{2.5}$ concentration, and the maximum TF may occur at a critical moment. This moment is neither the moment of the maximum WS nor the moment of the maximum $PM_{2.5}$ concentration but should be somewhere in between.

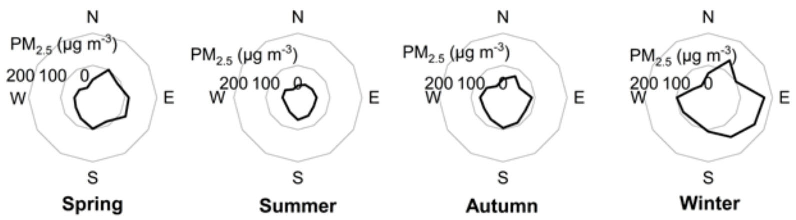

Fig. 4 The wind radar in different seasons in Beijing.

Responses to the comments from Reviewer #3

We would like to thank you for your comments and helpful suggestions. We revised our manuscript according to these comments and suggestions.

General Comments:

The manuscript presents a good investigation by studying the transport flux of particulate matter in the mixing layer over Beijing area, one of the heavily polluted places in the country. The study employs ceilometer, Doppler wind radar, and other meteorological measurement techniques to determine the transport flux in the region. Overall, the manuscript constitutes a good research article with clear conclusions, high quality figures, and great organization of the data. However, there seems to be a lot of room for English language improvement.

Specific Comments:

Comment 1:

Line 26, define “fine particle” for its first appearance, e.g., $PM_{2.5}$ or something else.

Response 1:

Thank you for your helpful suggestion. The definition of “fine particle” has been added to the paper.

Comment 2:

Line 31, recommend changing to “Transport mainly occurs between 14:00 and 18:00 LT”.

Response 2:

Thank you for your helpful suggestion. The text has been revised accordingly.

Comment 3:

Line 41, recommend changing “other provinces and cities” to “surrounding provinces and cities”

Response 3:

Thank you for your helpful suggestion. The text has been revised accordingly.

Comment 4:

Line 46, define fine particulate matter as $PM_{2.5}$ also if it is what the authors mean

Response 4:

Thank you for your helpful suggestion. The definition of “fine particle” has been added to the paper.

Comment 5:

Line 49, recommend changing “a steady decrease in poor air quality” to “steady improvement in air quality”

Response 5:

Thank you for your helpful suggestion. The text has been revised accordingly.

Comment 6:

Line 77, recommend changing “...1.2% yr-1...” to “1.2 percent per year”

Response 6:

Thank you for your helpful suggestion. The text has been revised accordingly.

Comment 7:

Line 86, recommend changing “...the reliability of the model will decrease” to “...the reliability of the model cannot be guaranteed”

Response 7:

Thank you for your helpful suggestion. The text has been revised accordingly.

Comment 8:

Line 91, recommend organizing it as “...transport flux (TF) in the mixing layer...”

Response 8:

Thank you for your helpful suggestion. The text has been revised accordingly.

Comment 9:

Line 156-158, the way this sentence and next one were constructed will really confuse the readers. “Seasonal variation” means and focuses on the variation, i.e, the standard deviation. I think the authors is trying to express something like this: “In terms of seasonal variation, the means of MLH for spring and summer are relatively higher than those of fall/autumn and winter. However, WS was quite different from MLH, ...". For Line 166-169, according adjustment is recommended for the discussion of $PM_{2.5}$ to avoid confusion.

Response 9:

Thank you for your helpful suggestion. We apologize for this mistake. Similar errors in the full text have been corrected accordingly.

Comment 10:

Line 163-164, recommend changing to "...The average TC for summer, winter, and autumn were quite similar, with the VC values...."

Response 10:

Thank you for your helpful suggestion. The text has been revised accordingly.

Comment 11:

Line 233-234, does the authors want to express this: "When MLH, WS_{ML} and VC were lower than 400 m, 2.5 m s^{-1} and 1500 m^2 s^{-1}, respectively, the $PM_{2.5}$ concentration decline sharply with these parameters increasing"? It is hard to imagine air pollution declines at these conditions not in favor of atmospheric dispersion.

Response 11:

This section has been deleted. Thank you for your helpful suggestion, and we apologize for this mistake.

Comment 12:

Line 261, I think May TF of 269 mg m^{-1} s^{-1} was 1.5 times higher than August TF of 106 mg m^{-1} s^{-1}. Alternatively, you can express it as "May TF was 2.5 times of August TF".

Response 12:

Thank you for your helpful suggestion. The text has been revised accordingly.

Comment 13:

A general comment: when using "transport" and "transportation", try to clarify it and avoid the ambiguity by meaning the transportation sector like vehicle emissions, since it is also great contributing factor for fine particle concentration.

Response 13:

Thank you for your helpful suggestion. Some ambiguity has been eliminated through the revision process, while the other instances can be understood by the context.

Comment 14:

Line 361-364, the expression in this segment could be revised to avoid negative image of the conclusion.

Response 14:

Thank you for your helpful suggestion. To avoid a negative image of the conclusion, this expression has been removed.

Technical corrections:

Comment 1:

Line 20, change “atmospheric pollution” to “air pollution”

Response 1:

Thank you for your helpful suggestion. The text has been revised accordingly.

Comment 2:

Line 24, change “weakens” to “weaker” or make alternative grammar corrections

Response 2:

Thank you for your helpful suggestion. The text has been revised accordingly.

Comment 3:

Line 35, change “transportation influence” to “influence/impact of (air pollutants) transport”, otherwise it seems to mean the influence of transportation section like vehicles

Response 3:

Thank you for your helpful suggestion. The text has been revised accordingly.

Comment 4:

Line 45, change “the Beijing’s air quality” to “Beijing’s air quality”

Response 4:

Thank you for your helpful suggestion. The text has been revised accordingly.

Comment 5:

Line 48, change “Although Beijing’s government has been dedicated...” to “Although Beijing government has dedicated...”

Response 5:

Thank you for your helpful suggestion. The text has been revised accordingly.

Comment 6:

Line 49-50, change “...ensure the continuous decline...” to “...ensure continuous decline...” or “...ensure the continued decline...”

Response 6:

Thank you for your helpful suggestion. The text has been revised accordingly.

Comment 7:

Line 109, change “...More detail descriptions...” to “More detailed descriptions...”

Response 7:

Thank you for your helpful suggestion. The text has been revised accordingly.

Comment 8:

Line 116, change “...remote sensor method...” to “remote sensing method...”

Response 8:

Thank you for your helpful suggestion. The text has been revised accordingly.

Comment 9:

Line 120, change the long dash to short dash or change it to “to”

Response 9:

Thank you for your helpful suggestion. The text has been revised accordingly.

Comment 10:

Line 150, change “...we carried out continuously measured...” to “...we continuously measured...” or “we carried out continuous measurement of...”

Response 10:

Thank you for your helpful suggestion. The text has been revised accordingly.

Comment 11:

Line 184, change “stable” to “relatively smaller”

Response 11:

Thank you for your helpful suggestion. The text has been revised accordingly.

Comment 12:

Line 185, recommend changing to “which are 4 h later than the peak and trough of MLH...”

Response 12:

Thank you for your helpful suggestion. The text has been revised accordingly.

Comment 13:

Line 193, change “at the latest” to “later than other seasons”. “At the latest” means something else like a deadline.

Response 13:

Thank you for your helpful suggestion. The text has been revised accordingly.

Comment 14:

Line 195, change “TC” to “VC” or change “VC” to “TC”, so that the same parameter is compared, even though we VC is used to express the magnitude of TC.

Response 14:

Thank you for your helpful suggestion. After careful consideration, we think that “atmospheric transport capacity” is prone to ambiguity, so we changed “atmospheric transport capacity (TC)” to “atmospheric dilution capability”.

Comment 15:

Line 236, change to “...than other seasons...”

Response 15:

Thank you for your helpful suggestion. The text has been revised accordingly.

Comment 16:

Line 243, change “indicator factors” indicators” or “indicating factors”

Response 16:

Thank you for your helpful suggestion. The text has been revised accordingly.

Comment 17:
Line 255-256, need improvement for this expression: “The northwesterly and westerly directions were the main transport sources of the cold period in Beijing.”
Response 17:
Thank you for your helpful suggestion. This phrase has been revised to “The transport sources of the cold period in Beijing were predominantly from the northwesterly and westerly directions.”

Comment 18:
Line 257, change “increased” to “changed”
Response 18:
Thank you for your helpful suggestion. The text has been revised accordingly.

Comment 19:
Line 286, change “rules” to “patterns”
Response 19:
Thank you for your helpful suggestion. The text has been revised accordingly.

Comment 20:
Line 297, change “4” to “four”, please refer to manuscript preparation guidance about numbers.
Response 20:
Thank you for your helpful suggestion. We apologize for our carelessness. The text has been revised accordingly.

Comment 21:
Line 299, recommend changing “and we must pay attention to local pollutant emission control” to “and local pollutant emission control is the most effective way of mitigating pollution levels”
Response 21:
Thank you for your helpful suggestion. The text has been revised accordingly.

Comment 22:
Line 346-347, change “the concentration of pollutants has a good relationship with VC” to “the concentration of pollutants is significantly correlated with VC”
Response 22:
This section has been removed. Thank you for your helpful suggestion.

Special thanks to you for your good comments.

We tried our best to improve the manuscript and made some changes accordingly. These changes do not influence the content or framework of the paper. We did not list all the changes here, but they are shown in red in the revised manuscript. Furthermore, to make the article more readable, we have had the manuscript professionally edited for language.

We earnestly appreciate the Editor's/Reviewers' earnest work and hope that the corrections will be met with approval.

Once again, thank you very much for your comments and suggestions.

Yours sincerely,

Dr. Tang

References:

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Shen, Y., L. Zhang, X. Fang, H. Ji, X. Li, and Z. Zhao: Spatiotemporal patterns of recent PM2.5 concentrations over typical urban agglomerations in China, Sci Total Environ, 655, 13-26, doi: 10.1016/j.scitotenv.2018.11.105, 2019.

Mixing layer transport flux of particulate matter in Beijing, China

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Abstract

Quantifying the transport flux (TF) of atmospheric pollutants plays an important role in understanding the causes of ~~atmospheric~~air pollution and in making decisions regarding the prevention and control of ~~atmospheric~~regional air pollution. In this study, the mixing layer height and wind profile ~~of the mixing layer~~ were measured by a ceilometer and Doppler wind radar, respectively, and the ~~variation~~ characteristics of the atmospheric dilution capability~~transport capacity (TC)~~ were analyzed using these two datasets. T~~he research showed that t~~he ventilation coefficient (V~~T~~C) appears to be highest~~strongest~~ in the spring (3940 ± 2110 m^2 s^{-1}) and ~~weake~~lower~~ns~~ in the summer (2953 ± 1322 m^2 s^{-1}), autumn (2580 ± 1601 m^2 s^{-1}) and winter (2913 ± 3323 m^2 s^{-1}). ~~Combined with the near-surface fine particle concentration data, the TC influence on the $PM_{2.5}$ concentration was studied, and there is a strong inverse correlation between the $PM_{2.5}$ and TC in spring, autumn and winter (R = -0.66, -0.65 and -0.80, respectively) and a weak positive correlation in summer (R = 0.33).~~ Combined with the backscatters measured by the ceilometer, vertical profiles of the $PM_{2.5}$ concentration were obtained, and~~By calculating~~ the $PM_{2.5}$ ~~transport flux~~ TF in the mixing layer was calculated~~of fine particles (TF),~~. ~~t~~The TF was the ~~in Beijing was found to be the~~highest in the spring, at 4.33 ± 0.69~~226 ± 294~~ mg $m^{-1}s^{-1}$,~~$m^{-1}s^{-1}$~~ and lower ~~in the other three seasons~~ in the summer, autumn and winter, when the TF values were 2.27 ± 0.42 mg $m^{-1}s^{-1}$, 2.39 ± 0.45 mg $m^{-1}s^{-1}$ and 2.89 ± 0.49 mg $m^{-1}s^{-1}$, respectively~~at approximately 140 mg $m^{-1}s^{-1}$~~. Air pollutant ~~Transport~~ ~~transport mainly~~ occurs between 14:00 and 18:00 LT. The ~~Except for during spring, the~~ TF was large in the pollution transition period (spring: 5.50 ± 4.83 mg $m^{-1}s^{-1}$, summer: 3.94 ± 2.36 mg $m^{-1}s^{-1}$ ~~328 ± 280 mg $m^{-1}s^{-1}$~~, autumn: 3.72 ± 2.86 mg $m^{-1}s^{-1}$ ~~280 ± 336 mg $m^{-1}s^{-1}$~~ and winter: 4.45 ± 4.40 mg $m^{-1}s^{-1}$~~240 ± 297 mg $m^{-1}s^{-1}$~~) and decreased during the heavy pollution period (spring: 4.69 ± 4.84 mg $m^{-1}s^{-1}$, summer: 3.39 ± 1.77 mg $m^{-1}s^{-1}$ ~~295 ± 215 mg $m^{-1}s^{-1}$~~, autumn: 3.01 ± 2.40 mg $m^{-1}s^{-1}$ ~~243 ± 238 mg $m^{-1}s^{-1}$~~ and winter: 3.25 ± 2.77 mg $m^{-1}s^{-1}$ ~~212 ± 209 mg $m^{-1}s^{-1}$~~). Our results indicate that the influence of the air pollutant transport~~ation influence~~ in the southern regions should receive more focus in the transition period of pollution, while local emissions should receive more focus in the heavy pollution period.

1. Introduction

With the rapid development of its economy and industry, as well as ~~the~~ its unique local topography, Beijing has become one of the cities in the world that is most seriously affected by air pollution. As early as before the 2008 Olympic Games, to fulfill the promise of a "Green Olympics", Beijing's industries were relocated to ~~other~~ surrounding provinces and cities. After the Olympic Games, with the promulgation of the "Action Plan for Prevention and Control of Air Pollution", Beijing implemented a series of measures to reduce pollutants, such as raising the emission standards of motor vehicles and fuel standards for vehicles, changing coal to natural gas, coal to electricity and so on. These measures have gradually improved ~~the~~ Beijing's air quality, with the annual average~~concentration of~~ fine particulate matter ($PM_{2.5}$) concentration decreasing from 90 μg m^{-3} in 2013 to 58 μg m^{-3} in 2017~~–~~ (Cheng et al. 2018a)~~(http://www.cnemc.cn/)~~.

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Although the Beijing~~'s~~ government has ~~been dedicated~~ been committed in recent years to taking measures that could ensure a steady improvement in the air quality~~a steady decrease in poor air quality~~, there is still great pressure to ~~ensure~~ achieve a ~~the~~ continuous decline in the particulate matter concentration. Beijing is in the north of the North China Plain, the south side and the west side are the Yanshan Mountains and the Taihang Mountains, respectively. Affected by the mountains to the northwest, there are more subsiding airflows, a lower mixing layer height and an extremely limited atmospheric ~~diffusion~~dilution ~~capacity~~capability. In addition, pollutants tend to accumulate in front of the mountains due to the influence of southerly winds and mountain obstruction. In central and northern China, the increase in $PM_{2.5}$ during winter is closely related to adverse atmospheric ~~transport~~ dilution conditions (Wang et al. 2016). Therefore, in addition to primary emissions and secondary formation, weak atmospheric dilution capability ~~transport capacity (TC)~~ is an important factor leading to the frequent occurrence of serious air pollution in Beijing.

In recent decades, mixing layer height (MLH) and wind speed (WS) are two major factors that lead to the annual increase in aerosol concentration and haze days during winter in China (Yang et al. 2016). Additionally, low MLH and low WS are important characteristics of weak atmospheric ~~TC~~dilution capability (Huang et al. 2018; Liu et al. 2018; Song et al. 2014; Tang et al. 2015). The change in MLH represents the vertical ~~TC~~dilution capability of pollutants, and the change in WS represents the horizontal ~~TC~~dilution capability of pollutants. To better characterize atmospheric ~~the TC~~dilution capability, the ventilation coefficient (VC) is usually used to evaluate the vertical and horizontal ~~transport~~ dilution capability~~capacity~~ of the atmosphere (Nair et al. 2007; Tang et al. 2015; Zhu et al. 2018). Thus, it is a good choice to use VC to evaluate the relationship between atmospheric ~~TC~~dilution capability and air pollution in Beijing. Although previous studies have analyzed the relationship between MLH and pollutants (Geiß et al. 2017; Miao and Liu 2019; Schäfer et al. 2006; Su et al. 2018), studies on the effects of VC on particle concentration are extremely rare.

Although the problem of heavy pollution in northern China has improved in recent years, regional pollution problems remain, especially in the Beijing-Tianjin-Hebei region (Shen et al. 2019).~~In addition, with the reduction in local emission sources, the contribution of regional transport becomes particularly important.~~ There are three main transport routes affecting Beijing: the northwest path, the southwest path and the southeast path (Chang et al. 2018; Li et al. 2018; Zhang et al. 2018). The occurrence of heavy pollution in Beijing is closely related to the transportation of pollutants in southern regions, mainly in southern Hebei, northern Henan and western Shandong, while the high-speed northwest air mass is conducive to the removal of pollutants in Beijing (Li et al. 2018; Ouyang et al. 2019; Zhang et al. 2018; Zhang et al. 2017). In recent years, the contribution of regional transport to Beijing has been increasing annually, with a trend of 1.2 % per year~~% year^{-1}~~, which reached 31-73% in summer and 27-59% in winter (Chang et al. 2018; Cheng et al. 2018b; Wang et al. 2015). High $PM_{2.5}$ concentrations are usually accompanied by high transport flux (TF) within a day in Beijing~~,~~. As pollution worsens, the contribution of the surrounding areas to the $PM_{2.5}$ in Beijing has risen from 52% to 65% in a month on average in 2016 (Zhang et al. 2018). However, during heavy pollution, the ~~transport flux~~TF decreased in Beijing (Chang et al. 2018; Tang et al. 2015; Zhu et al. 2016).~~Although many studies on regional transport have been carried out, most observational studies cannot easily quantify transport flux due to the lack of wind profile data. Therefore, transport flux can only be obtained by models. When the model lacks verification data, the reliability of the model will decrease. Thus, it is imperative to quantify the transport flux through observations.~~

To solve the regional pollution problem, joint prevention and control have been put forward for a long time. Many studies on regional transport have been carried out, but most observational studies cannot easily quantify TF due to the lack of particle and wind vertical profiles, and it is still unclear when do we need to control the emission sources in which areas. To solve the above problems, we

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conducted 2 years of continuous observations on MLH and wind profiles in the Beijing mixing layer and analyzed the mixing layer dilution capability of atmosphere. Afterwards, using backscattering coefficient profile, we obtained the vertical $PM_{2.5}$ concentration profiles, and calculated TF profile and mixing layer TF. Finally, using the near-surface $PM_{2.5}$ concentration as an indicator to classify the air pollution degree, we analyzed the TF during the transitional and heavily polluted period in Beijing and illuminated the main controlling factors.~~To solve the above two problems, we conducted 2 years of continuous observations on MLH and wind profiles in the Beijing mixing layer and analyzed the mixing layer TC of pollutants and their relationship with particulate matter. Then, combined with the concentration of particulate matter, we analyzed fine particulate matter transport flux in the mixing layer (TF). Finally, using the $PM_{2.5}$ concentration as an indicator to classify the air pollution degree, we analyzed the TF in Beijing during the transitional and heavily polluted period and illuminated the main controlling factors.~~

2. Methods

2.1 Observational station

To understand the ~~TC~~dilution capability characteristics in Beijing, two years of observations were conducted in Beijing (2016.1.1-2017.12.31). The observational site (BJT) is in the Institute of Atmospheric Physics of the Chinese Academy of Sciences, located west of the Jiande Bridge in the Haidian District, Beijing (39.98° N, 116.38° W). The north and south sides of the station are the north third and north fourth ring roads respectively, and the eastern side is Beijing-Tibet expressway. The altitude (a.s.l.) is about 60 m. There is no obvious emission source around the observational site except motor vehicles~~the highway~~.

2.2 Observations of MLH and wind profiles

To analyze ~~TC~~dilution capability, MLH was observed by a single-lens ceilometer (CL51, Vaisala, Finland), and the wind profile ~~in the mixing layer~~ was simultaneously observed by doppler wind radar (Windcube 100s, Leosphere, France).

A single-lens ceilometer measures the attenuated backscatter coefficient profile of atmosphere~~ic aerosols~~ by pulsed diode laser lidar technology (910 nm waveband) within a 7.7 km range, and determine the MLH through the position of abrupt changes in the backscattering coefficient profile. In the actual measurement, the measurement interval was 16 s and the measurement resolution was 10 m. More detailed descriptions are presented in the published literature (Tang et al. 2016; Zhu et al. 2016). In this study, the gradient method (Steyn et al. 1999) is used to determine the MLH; that is, the top of the mixing layer was determined by the maximum negative gradient value~~ (-dβ/dx)~~ in the profile of the atmosphere backscattering coefficient. Moreover, to eliminate the interference of aerosol layer structure and the detection noise to data, the MLH was calculated by the improved gradient method after, smoothly ~~-~~averaging the profile data (Münkel et al. 2007; Tang et al. 2015). Doppler wind radar uses the remote sensing~~or~~ method of laser detection and ranging technology and measures the doppler frequency shift generated by the laser through the backscatter echo signal of particles in the air. Windcube 100s can provide 3D wind field data within a 3 km range from the system, including u, v and w vectors. In the actual measurement, starting from 100 m, the spatial resolution is 50 m, the WS accuracy is < 0.5 m s^{-1}, and the radial WS range is -30 m s^{-1} to ~~-~~30 m

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2.3 Other data

During the observations, the hourly $PM_{2.5}$ ~~and ozone surface~~ concentrations of the Beijing Olympic Sports Center (39.99° N, 116.40° W) were obtained from the Ministry of Environmental Protection of China (http://www.zhb.gov.cn/).

2.4 Analytical method

Atmospheric dilution is composed of vertical and horizontal dilution, which can be characterized by MLH and wind speed in the mixing layer (WS_{ML}), respectively. VC ($m^2\ s^{-1}$) was obtained by combining MLH (m) and ~~wind speed in the mixing layer (~~WS_{ML} ~~,~~ (m s^{-1}), which can be used ~~to~~ for ~~characterize~~comprehensive ~~–~~ evaluation of vertical and horizontal dilution~~TC~~. ~~A h~~Higher ~~VC~~ dilution-related parameters (MLH, WS_{ML} and VC) indicate~~s~~ a stronger ~~TC~~dilution capability, which is conducive to the transport and ~~diffusion~~dilution of heavy air pollution.

The VC calculation method is as follows:

$$VC = H_{ML} \sout{MLH} \times WS_{ML}, \quad (1)$$

$$WS_{ML} = \frac{1}{n}\sum_{i=1}^{n} WS_i, \quad (2)$$

$$\sout{WS} WS_i = \sqrt{\sout{u} u_i^2 + \sout{v} v_i^2}, \quad (3)$$

where WS_{ML} is the average WS within the mixing layer, calculated by Eq. (2); H_{ML} is the height of the mixing layer; WS_i is the WS observed at a certain height~~all heights~~, calculated by ~~the mean value of~~ u_i and v_i in the wind profile according to Eq. (3); and n is the number of measurement layers in the mixing layer (Nair et al. 2007).

TF (mg m^{-1}~~s~~^{2}s^{-1}) is determined by ~~TC~~ WS and the $PM_{2.5}$ concentration in the area under analysis. The calculation method for a certain height is shown in Eq. (4):

$$TF_{u_i} = u_i \times C_i \sout{C_{PM_{2.5}} \times MLH}, \quad (4)$$

where C_i is the concentration of $PM_{2.5}$ at a certain height. However, it is extremely difficult to observe the vertical $PM_{2.5}$ concentration in the mixing layer. To obtain the $PM_{2.5}$ concentration profile, we studied the backscattering coefficient measured by ceilometer, and found that the concentration of near-surface $PM_{2.5}$ is strongly correlated with the backscattering coefficient at 100 m (Fig. S1). Thus, based on the relationship between the two, backscattering coefficient profile can be used to inverse the vertical $PM_{2.5}$ concentration profile. ~~It is extremely difficult to observe the $PM_{2.5}$ concentration in the mixing layer by height, but previous observations have shown that the backscattering coefficient profile in the mixing layer is relatively uniform .~~ Then~~Assuming that the particle concentration in the mixing layer is uniform~~, the TFs in the mixing layer are calculated as follows:

$$TF_u = \int_{i=1}^{n} (u_i \sout{\frac{1}{n}\sum_{i=1}^{n} u_i} \times \sout{C_{PM_{2.5}}} C_i) \sout{\times MLH}$$

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$$TF_v = \int_{i=1}^{n} (v_i \times C_i) \quad \text{~~}TF_v = \frac{1}{n}\sum_{i=1}^{n} v_i \times C_{PM_{2.5}} \times MLH\text{~~} \tag{5}$$

Through the above method, radial and zonal ~~transport flux~~TFes can be obtained, and vector synthesis in two directions can be conducted to obtain the main transport direction to find the transport source area.

3. Results and discussions

3.1 Boundary layer meteorology~~Mixing layer transport capacity (TC)~~

3.1.1 Seasonal variation

To understand the variations of atmospheric ~~TC~~dilution capability, we carried out continuous~~ly~~ measurement of~~ed~~ MLH and wind profile~~s~~ within the mixing layer over a 2-year period (2016.1.1-2017.12.31). The availability was verified after MLH elimination by Tang et al. (Tang et al. 2016). After the exclusion of the data of MLH under rainy, sandstorm and windy conditions, data availability was 95% over the 2-year period, higher than that of previous studies (Mues et al. 2017; Tang et al. 2016). The availability was lowest in February at 86% and highest in July at 99%. In terms of ~~The~~ seasonal variation, the averages of ~~in~~ MLH for spring~~was higher in spring~~ (781 ± 229 m) (value ± standard deviation) and summer (767 ± 219 m) were higher than those of ~~and lower in~~ autumn (612 ± 166 m) and winter (584 ± 221 m) (Fig. 1). However, WS_{ML} was ~~quite~~ different from MLH in terms of seasonal variation, with the largest value at 4.6 ± 1.6 m s^{-1} in spring, followed by winter (4.1 ± 2.7 m s^{-1}) and autumn (3.7 ± 1.6 m s^{-1}), and the smallest value at 3.6 ± 1.1 m s^{-1} in summer. VC was calculated by the MLH and wind profile, and t~~he seasonal variation in TC over 2 years was analyzed (Fig. 1). T~~he results demonstrate that the ~~TC~~dilution capability was strongest in spring, as the VC reached as high as 3940 ± 2110 m^2 s^{-1}. Atmospheric~~The~~ ~~TC~~dilution capability ~~differences~~ ~~for~~among summer, winter and autumn were ~~small~~ similar, w~~ith~~hen the VC values were 2953 ± 1322 m^2 s^{-1}, 2913 ± 3323 m^2 s^{-1} and 2580 ± 1601 m^2 s^{-1}, respectively. A monthly analysis shows that atmospheric ~~the TC~~dilution capability was ~~the~~ strongest in May, the VC was as high as 5161 ± 2085 m^2 s^{-1}~~, ; the TC was the~~and worst in December, and the VC was only 1690 ± 1072 m^2 s^{-1}. The VC value in May was 3.1 times ~~higher than~~of that in December. To analyze the impacts of dilution capacity on $PM_{2.5}$, the seasonal variation of $PM_{2.5}$ were analyzed. The averages of $PM_{2.5}$ concentration for winter (80 ± 87 μg m^{-3}) was highest, followed by autumn (68 ± 54 μg m^{-3}) and spring (67 ± 60 μg m^{-3}), and summer (51 ± 29 μg m^{-3}) was lowest. The lowest monthly average $PM_{2.5}$ concentration was 42 ± 26 μg m^{-3} in August. The highest monthly average was in January at 94 ± 100 μg m^{-3}, 2.2 times of that in August (Fig. 1).~~The seasonal variation in the $PM_{2.5}$ concentration was the highest in winter (80 ± 87 μg m^{-3}), followed by autumn (68 ± 54 μg m^{-3}) and spring (67 ± 60 μg m^{-3}), and the seasonal variation was the lowest in summer (51 ± 29 μg m^{-3}). The lowest monthly average $PM_{2.5}$ concentration was 42 ± 26 μg m^{-3} in August. The highest monthly average was in January at 94 ± 100 μg m^{-3}, 2.2 times higher than that in August (Fig. 1). Thus, the vertical and horizontal diffusion capacities are strong in spring and weak in autumn and winter. In summer, the vertical diffusion capacity is strong, while the horizontal diffusion capacity~~

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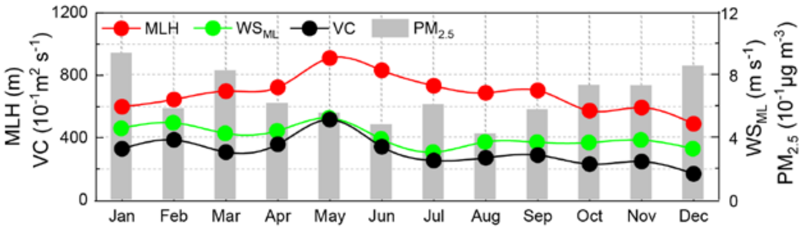
~~is weak. Overall, high $PM_{2.5}$ concentrations are associated with poor TC.~~



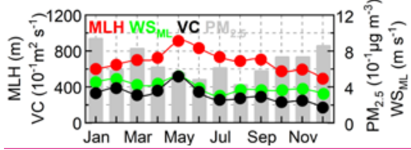


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Fig. 1~~.~~ Monthly variations in mixing layer height (MLH), wind speed in the mixing layer (WS_{ML}), the ventilation coefficient (VC) and $PM_{2.5}$ in Beijing.

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Although there is little difference in dilution capability between summer, autumn and winter, there is serious pollution in autumn and winter. To analyze this problem, the VC frequency distribution was studied. The results show that VC had a high frequency in the range of 1000-4000 m^2 s^{-1} from 2016 to 2017, but the frequency distribution was different in different seasons (Fig. 2). The VC showed a strong dilution capability in spring, mainly in the range of 2000-5000 m^2 s^{-1}, with the highest frequency (24%) in the range of 2000-3000 m^2 s^{-1}. In summer, the high frequency of VC occurred in the range of 1000-4000 m^2 s^{-1}, which was slightly lower than that in spring, and the highest frequency (27%) occurred in the range of 3000-4000 m^2 s^{-1}. Additionally, the VC high frequency appeared in a lower range in autumn and winter. The VC occurred at a high frequency of 1000-3000 m^2 s^{-1} in autumn, and the highest frequency occurred within the range of 2000-3000 m^2 s^{-1}, accounting for 33%. In winter, VC appeared more frequently in the range of 0-2000 m^2 s^{-1} and was the highest in the range of 1000-2000 m^2 s^{-1}, which was 28%. In winter, when the Siberian high transit, strong northwest winds prevail in the Beijing area (Fig. 5), which resulting the higher frequency of VC in the range of 1000-2000 m^2 s^{-1}. Particularly, the VC frequency of 0-1000 m^2 s^{-1} in winter was significantly higher than that of the other seasons, up to 22%, which was 7 times of that in spring, 5 times of that in summer and 2 times of that in autumn. According to the seasonal variation in $PM_{2.5}$ concentration, heavy pollution in autumn and winter is related to the high frequency of poor atmospheric dilution capability.

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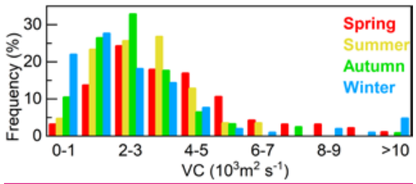

Fig. 2 Frequency distribution of the daily VC from January 2016 to December 2017 in Beijing.

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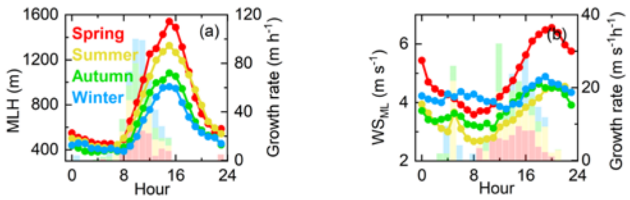
3.1.2 Diurnal variation

Moreover, the diurnal variations in dilution-related parameters~~meteorological factors~~ during different seasons were analyzed to reveal the diurnal evolution~~ characteristics~~ of atmospheric ~~TC~~dilution capability. The peak and trough values of MLH and VC appeared simultaneously at approximately 15:30 LT and 05:30 LT, respectively. Generally, the daily variation in MLH is characterized by a low value at night, which increases rapidly after sunrise and reaches the maximum value in the afternoon (Fig. ~~2a~~3a). The daily maximum value of MLH is seasonal, where it is higher in spring and summer and lower in autumn and winter. The daily minimum value of MLH generally occurs when the mixing layer is stable and is closely related to WS. The diurnal variation in WS_{ML} is smaller~~stable~~, with a peak at approximately 19:30 LT and a trough at approximately 10:00 LT, which ~~is~~ are ~4 h later than the peak and ~~valley~~trough of MLH (Fig. ~~2b~~3b). The diurnal variation in VC is similar to MLH, showing that the ~~TC~~dilution capability is strong before sunset, gradually weakens after sunset and remains stable at night. The ~~TC~~dilution capability in spring was significantly stronger than that during other seasons, and the maximum daily value reached 8678 $m^2 s^{-1}$ (Fig. ~~2c~~3c). In addition to spring, the daily maximum values of VC in summer, autumn and winter were close at approximately 5000 $m^2 s^{-1}$ (Fig. ~~2c~~3c). The ~~TC~~ VC growth rate in spring was significantly higher than that in other seasons, reaching a maximum at approximately 09:00 LT. ~~Late i~~In autumn, the ~~TC~~ VC growth rate peaked at approximately 10:00 LT. Summer and winter peaked at approximately 11:00 LT. Throughout the year, VC began to increase during winter ~~at the latest~~later than other seasons, at approximately 09:00 LT, indicating that the weaker ~~TC~~dilution capability remained for a longer period during winter. ~~TC~~ VC was weakened most rapidly in spring; however, ~~the TC~~it was still higher than that ~~he VC~~ of other seasons after declining. In addition to spring, the ~~TC~~ VC in autumn and winter weakened the most rapidly and the slowest in summer. In general, vertical and horizontal ~~diffusion~~dilution ~~is very~~are strong in the spring during both day and night. In winter, vertical ~~diffusion~~dilution is weak during the day, and horizontal ~~transportation~~ dilution during the night is the main~~ transportation~~. In summer, vertical ~~diffusion~~dilution during the day is dominant.

Notable differences are present when we compare dilution-related parameters to $PM_{2.5}$ concentration. The daily maximum of $PM_{2.5}$ concentration in spring, summer, autumn and winter were 73 μg m^{-3} (11:00 LT), 56 μg m^{-3} (09:00 LT), 78 μg m^{-3} (23:00 LT) and 101 μg m^{-3} (01:00 LT), respectively. The differences between the maximum and minimum were 14 μg m^{-3}, 10 μg m^{-3}, 20 μg m^{-3} and 38 μg m^{-3}, respectively. Thus, the diurnal variation of $PM_{2.5}$ can divided into two categories: (1) high value occurs in the midday in spring and summer and the overall change is small, (2) high value occurs during the night in autumn and winter and differ greatly from low value (Fig. 3d). The main causes of air pollution are local emissions and regional transportation. Thus, these results indicating that more local contribution in autumn and winter, and higher regional transport in spring and summer.



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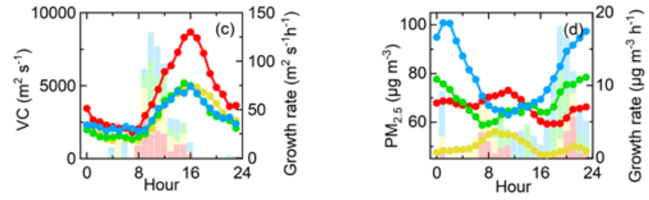

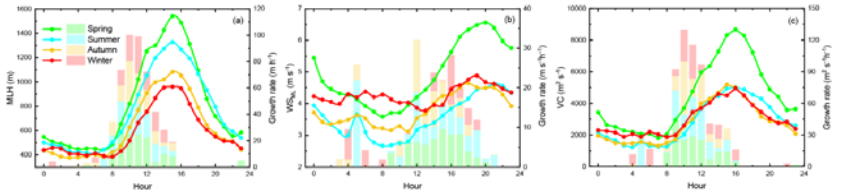

Fig. 23. Diurnal variations and growth rates of MLH (a), WS_{ML} (b) and , VC (c) and $PM_{2.5}$ (d) in spring, summer, autumn and winter in Beijing. Diurnal variations are represented by lines and scatters. Growth rates are represented by columns, and only positive values are shown in the figure.

3.1.3 Frequency distribution

Although there is little difference in TC between summer, autumn and winter, there is serious pollution in autumn and winter. To analyze this problem, the VC frequency distribution was studied. The results show that VC had a high frequency in the range of 1000-4000 $m^2 s^{-1}$ from 2016 to 2017, but the frequency distribution was different in different seasons (Fig. 3). The VC showed a strong TC in spring, mainly in the range of 2000-5000 $m^2 s^{-1}$, with the highest frequency (24%) in the range of 2000-3000 $m^2 s^{-1}$. In summer, the high frequency of VC occurred in the range of 1000-4000 $m^2 s^{-1}$, which was slightly lower than that in spring, and the highest frequency (27%) occurred in the range of 3000-4000 $m^2 s^{-1}$. Additionally, the VC high frequency appeared in a lower range in autumn and winter. The VC occurred at a high frequency of 1000-3000 $m^2 s^{-1}$ in autumn, and the highest frequency occurred within the range of 2000-3000 $m^2 s^{-1}$, accounting for 33%. In winter, VC appeared more frequently in the range of 0-2000 $m^2 s^{-1}$ and was the highest in the range of 1000-2000 $m^2 s^{-1}$, which was 28%. However, the VC frequency of 0-1000 $m^2 s^{-1}$ in winter was significantly higher than that of the other seasons, up to 22%, which was 7 times higher than that of spring, 5 times higher than that of summer and 2 times higher than that of autumn. According to the seasonal variation in $PM_{2.5}$ concentration, heavy pollution in autumn and winter is related to the high frequency of poor TC.

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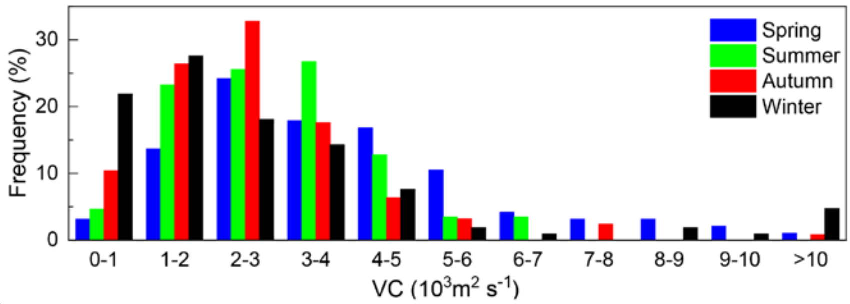

~~Fig. 3. Frequency distribution of the daily VC from January 2016 to December 2017 in Beijing.~~

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3.2 Mixing layer TF of $PM_{2.5}$mixing layermixing layer

$_{2.5}$Response of particulate matter to TC

Studies have found that air pollution worsens when TC weakens . To further understand the response of fine particles to TC in different seasons, the correlations between meteorological factors and $PM_{2.5}$ concentration were analyzed (Fig. 4). From 2016 to 2017, the annual average $PM_{2.5}$ concentration was 66 ± 62 μg m^{-3}, the maximum concentration was 898 μg m^{-3}, and the minimum concentration was only 1 μg m^{-3}, which showed high concentrations in autumn and winter. As shown in Fig. 4, $PM_{2.5}$ concentrations increased exponentially with decreases in MLH, WS_{ML} and VC, indicating that the concentration of fine particles was highly sensitive to these meteorological factors. When MLH, WS_{ML} and VC were lower than 400 m, 2.5 m s^{-1} and 1500 m^2 s^{-1}, respectively, the air pollution declines sharply. VC had a better correlation with the $PM_{2.5}$ concentration than MLH and WS_{ML}, indicating that VC can better characterize pollution dissipation. The $PM_{2.5}$ concentration in winter had a better response to TC than the other seasons, with the correlation coefficient with VC reaching -0.80, followed by spring and autumn, with correlation coefficients of -0.66 and -0.65, respectively (Fig. 4). The correlation in spring and autumn may decrease due to dust. In summer, $PM_{2.5}$ had a poor relationship with WS_{ML} and even had weak positive correlations with MLH ($R = 0.42$) and VC ($R = 0.33$). A high ozone concentration existed in the high MLH (Fig. 4), which will promote the transformation of gaseous precursors to secondary particles. Therefore, the weak positive correlation in summer was related to a strong photochemical reaction.

Thus, MLH, WS_{ML} and VC can be used as indicator factors for the formation of air pollution, but the particle concentration responds best to VC. Additionally, the response of particle concentration to VC showed obvious seasonal differences, with the best in winter, followed by autumn and spring, and a weak positive correlation in summer.

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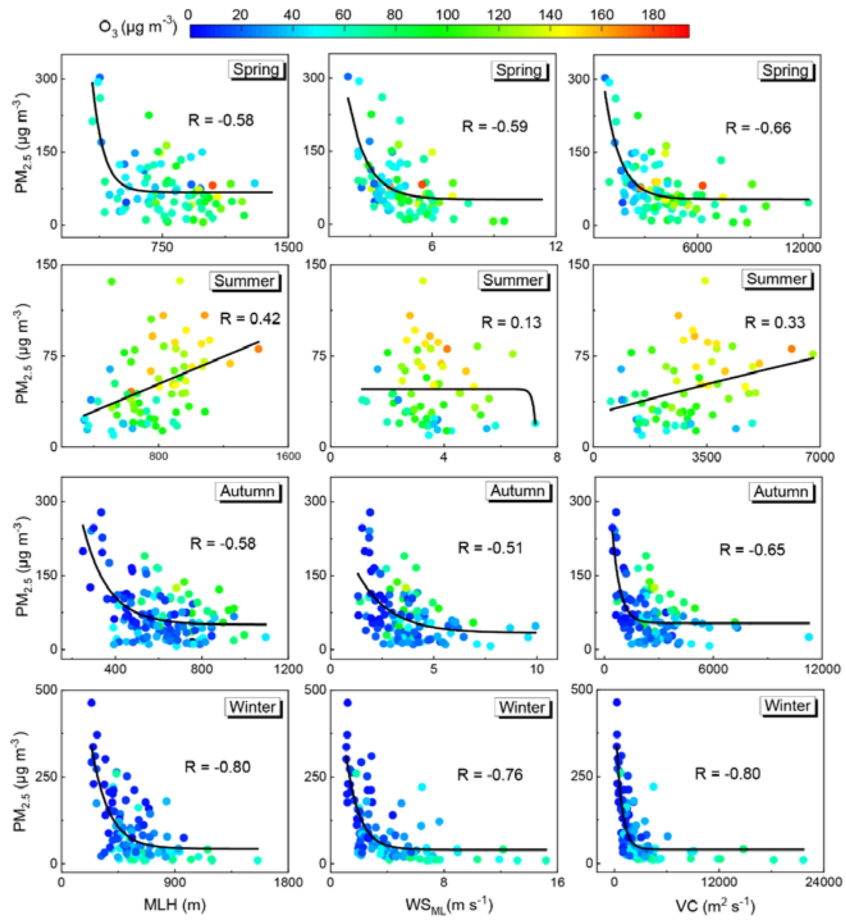

Fig. 4. Correlations among MLH, WS_{ABL} and VC and $PM_{2.5}$ under different ozone levels in Beijing.

3.3 Mixing layer transport flux of particulate matter

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To quantify the transport of pollutants in Beijing, the Beijing TF was analyzed, and the transport direction of fine particles was characterized by the wind direction in the mixing layer. As shown in Fig. 5, the TF in spring was the largest, reaching 226 ± 294 mg $m^{-1}s^{-1}$, and there was no significant difference in summer, autumn or winter, when the TF values were 147 ± 182 mg $m^{-1}s^{-1}$, 143 ± 194 mg $m^{-1}s^{-1}$ and 134 ± 179 mg $m^{-1}s^{-1}$, respectively. The northwesterly and westerly directions were the main transport sources of the cold period in Beijing. With temperature warming, the transport direction gradually increased from west to south, mainly as a southwesterly in spring and southerly in summer. The monthly average maximum value of TF occurred in May, as high as 269 ± 328 mg $m^{-1}s^{-1}$ and mainly originated from the southwest direction, which was accompanied by a strong wind. The minimum value appeared in August, as low as 106 ± 145 mg $m^{-1}s^{-1}$, which was mainly transported from western regions, with low WS values. The TF in May was 2.5 times higher than that in August (Fig. 5). Therefore, the change in transport direction leads to an obvious seasonal variation in TF. Overall, the regional transport contributes the most to the particulate matter concentration in spring, which is mainly related to increased dust activities; regional transport has the least contribution in winter, which indicates that more focus should be given to local emission source control; in summer and autumn, the southwest airflow transportation influence on Beijing should receive more focus.

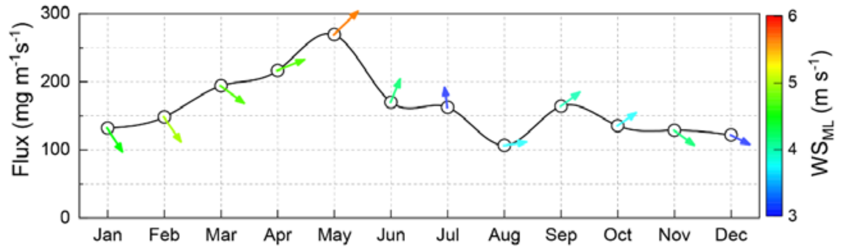
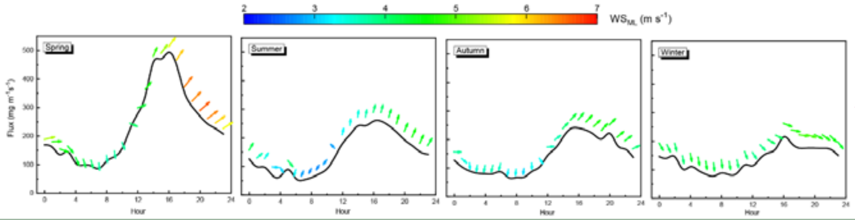


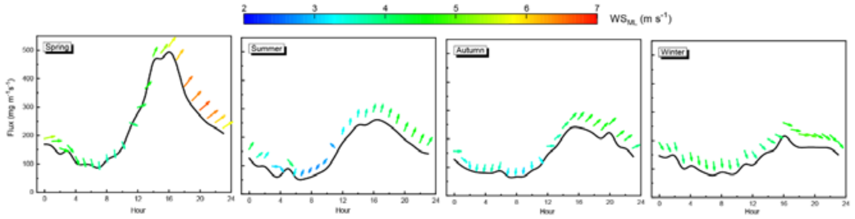
Fig. 5. Seasonal variations in the mixing layer transport flux of PM and transportation directions.

To understand the regional transport influence on the Beijing area, the diurnal variation characteristics of TF were analyzed during different seasons in Beijing. The daily minimum value of TF appeared at approximately 07:00 LT and was accompanied by a northerly wind. As the wind direction gradually turned south, the daily minimum value of TF continued to rise until the daily maximum value appeared at approximately 16:00 LT (Fig. 6). Transportation mainly occurred between 14:00 and 18:00 LT, which was consistent with the results of a previous study (Ge et al. 2018). In spring, the WS was higher, so the peak TF duration was shorter, at approximately 2 h. The maximum daily value was 494 mg $m^{-1}s^{-1}$, and the minimum was 87 mg $m^{-1}s^{-1}$ in spring. Therefore, the diurnal variation in TF during spring showed the characteristics of a rapid rise and rapid decline. The peak duration was approximately 4 h for a long time in summer and autumn, where the daily maximum values were 259 mg $m^{-1}s^{-1}$ and 240 mg $m^{-1}s^{-1}$, and the minimum values were 53 mg $m^{-1}s^{-1}$ and 66 mg $m^{-1}s^{-1}$, respectively. The diurnal variation in TF during summer and autumn showed the characteristics of a slow rise and slow decline. Specifically, the daily variation had a strong fluctuation in winter, which peaked at only 16:00 LT (215 mg $m^{-1}s^{-1}$), then dropped sharply to 193 mg $m^{-1}s^{-1}$, plateaued from 17:00 to 22:00 LT for approximately 5 h, maintained at approximately 176 mg $m^{-1}s^{-1}$, and then quickly dropped to 78 mg $m^{-1}s^{-1}$



~~Fig. . Diurnal variations in the mixing layer transport flux of PM and transportation directions during different seasons in Beijing.~~

~~The TF variation rules can be summarized as a high TF corresponds to a southerly wind and a low TF corresponds to a northerly wind. When the wind direction in the mixing layer changed from north to south, the wind gradually increased from the daily minimum to the daily maximum. The TF increased by 6 times in spring, 5 times in summer, 4 times in autumn and 3 times in winter. The current pattern is because areas south of Beijing are heavily polluted and southerly winds help transport pollutants into the city, leading to high transport flux in spring, summer and autumn afternoons (Fig. 6). The results further confirm the conclusion that the northwest wind in Beijing is a clean wind (Wang et al. 2015; Zhang et al. 2018). Thus, the north wind is conducive to the outward transport of pollutants from Beijing, which helps to alleviate pollution. As a result, there was no high TF in winter when the westerly wind and northerly wind prevailed. This finding also proves the important influence of local emissions on heavy pollution occurrence during winter in Beijing. In summary, there are 4 main transport routes that affect Beijing, including the northwest path, southwest path, west path and south path. The TF in winter is low, local emissions play an important role, and we must pay attention to local pollutant emission control.~~



~~Fig. 6. Diurnal variations in the mixing layer transport flux of PM and transportation directions during different seasons in Beijing.~~

3.2.1 Temporal evolution of TF

To quantify the transport of $PM_{2.5}$ in Beijing, the transport direction of $PM_{2.5}$ was characterized by the average wind direction in the mixing layer. As shown in Fig. 4, mixing layer TF in spring was largest, reaching 4.33 ± 0.69 mg m^{-1}s^{-1}, and there was no significant difference in summer, autumn

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or winter, when the TF values were 2.27 ± 0.42 mg $m^{-1}s^{-1}$, 2.39 ± 0.45 mg $m^{-1}s^{-1}$ and 2.89 ± 0.49 mg $m^{-1}s^{-1}$, respectively. The transport sources of the cold period in Beijing were predominantly from the northwesterly and westerly directions. With temperature warming, the transport direction gradually changed from west to south, mainly as a southwesterly in spring and southerly in summer. The monthly average maximum value of TF occurred in May, as high as 5.00 ± 5.21 mg $m^{-1}s^{-1}$ and mainly originated from the southwest direction, which was accompanied by a strong wind. The minimum value appeared in August, as low as 1.70 ± 1.73 mg $m^{-1}s^{-1}$, which was mainly transported from western regions, with small WS. The TF in May was 3 times of that in August (Fig. 4). Therefore, the change in transport direction leads to an obvious seasonal variation in TF. Overall, the regional transport contributes the most to the $PM_{2.5}$ concentration in spring, which is mainly related to increased dust activities; regional transport has smaller contribution in winter, but high near-surface $PM_{2.5}$ concentration, which indicates that more focus should be given to local emission source control; in summer and autumn, the southwest airflow transportation influence on Beijing should receive more focus.

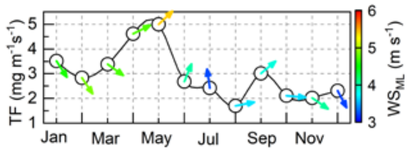

Fig. 4 Seasonal variations in the mixing layer TF of $PM_{2.5}$ and transportation directions.

To understand the regional transport influence on the Beijing area, the diurnal variations of mixing layer TF were analyzed during different seasons in Beijing. The daily minimum value of TF appeared at approximately 07:00 LT and was accompanied by a northerly wind. As the average wind direction in the mixing layer gradually turned south, the daily minimum value of TF continued to rise until the daily maximum value appeared at approximately 16:00 LT (Fig. 5). Transportation mainly occurred between 14:00 and 18:00 LT, which was consistent with the results of a previous study (Ge et al. 2018). In spring, the WS was highest, so the peak TF duration was shortest, peaked at only 16:00 LT (9.50 mg $m^{-1}s^{-1}$), and then dropped sharply to 1.94 mg $m^{-1}s^{-1}$. Therefore, the diurnal variation in TF during spring showed the characteristics of a rapid rise and rapid decline. The peak duration was approximately 3 h for a long time in summer and autumn, where the daily maximum values were 3.79 mg $m^{-1}s^{-1}$ and 3.63 mg $m^{-1}s^{-1}$, and the minimum values were 1.00 mg $m^{-1}s^{-1}$ and 1.30 mg $m^{-1}s^{-1}$, respectively. The diurnal variation in TF during summer and autumn showed the characteristics of a slow rise and slow decline. Specifically, the daily variation had a strong fluctuation in winter, peaked three times at 14:00 LT (4.06 mg $m^{-1}s^{-1}$), 16:00 LT (4.38 mg $m^{-1}s^{-1}$) and 19:00 LT (4.07 mg $m^{-1}s^{-1}$), then dropped slowly to 1.66 mg $m^{-1}s^{-1}$. Another special point is that in spring, summer and autumn, high TF corresponds to southerly wind, while in winter, southerly wind does not appear in the whole transport process, instead, there is westerly wind, which influenced by the Siberian high.

Even so, the TF variation patterns can be summarized as a high TF corresponds to a southerly wind and a low TF corresponds to a northerly wind (Fig. 5). When the average wind direction in the mixing layer changed from north to south, the TF gradually increased from the daily minimum to the daily maximum. The TF increased by 5 times in spring, 4 times in summer, 3 times in autumn and winter. The current pattern is because areas located in south of Beijing are heavily polluted and

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southerly winds help transport pollutants into the city, leading to high TF in the afternoons (Fig. 5). However, due to the high mixing layer in spring, the concentration of near-surface $PM_{2.5}$ did not increase. The results further confirm the conclusion that the northwest wind in Beijing is a clean wind (Wang et al. 2015; Zhang et al. 2018). Thus, the northwest wind is conducive to the outward transport of pollutants from Beijing, which helps to alleviate pollution. As a result, there was no high TF in winter when the northwest wind prevailed. On the other hand, southerly winds are stronger than northerly winds (Fig. 5), which can also result high TF occurred. Therefore, the level of TF is determined by two factors, that of WS and $PM_{2.5}$ concentration. In spring, summer and autumn, strong south wind prevails in the afternoon. As the south wind is often accompanied by high $PM_{2.5}$ concentration (Fig. S2), TF is high. In winter, the whole day is dominated by westerly and northerly winds. Although the northerly winds are strong, TF is not high due to the low $PM_{2.5}$ concentration. Generally, high WS means fast mixing, and the corresponding MLH is also high. At this time, TF is mainly controlled by WS. While WS is low, the mixing speed is slow and MLH is low. At this time, TF is mainly controlled by $PM_{2.5}$ concentration. From the above analysis, it can be inferred that if MLH and WS gradually decrease with the worsen of pollution, the mixing layer TF is controlled by WS first and then by $PM_{2.5}$ concentration, which may appear a maximum TF at a critical moment. This moment is neither the moment of maximum WS nor the moment of maximum $PM_{2.5}$ concentration. It should be somewhere in between. This will be discussed in more detail in section 3.3.

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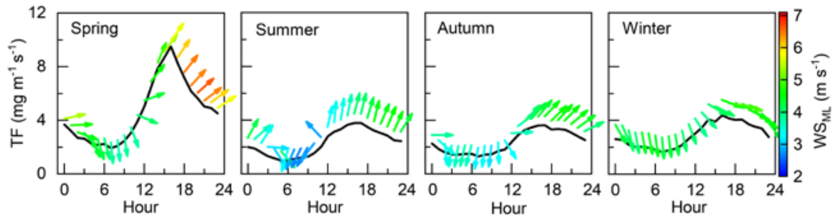

Fig. 5 Diurnal variations in the mixing layer TF of $PM_{2.5}$ and transportation directions during different seasons in Beijing.

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3.2.2 Vertical evolution of TF

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After the aforementioned analyses, the transportation period is known. To further explore the height of transport, we studied the seasonal variation of TF profile in combination with the vertical wind and $PM_{2.5}$ profile. With the increase of altitude, the WS first increases sharply at approximately 200 m, and then slowly increases, and the differences between different seasons gradually become significant. WS is always smallest in summer, and strongest in winter. It is same in spring and autumn at 1200 m. Above 1200 m, WS in autumn exceeds those in spring. The $PM_{2.5}$ concentration at 100 m obtained by inversion is highest in winter (93.7 mg $m^{-2}s^{-1}$), and similar in spring and autumn (80.3 mg $m^{-2}s^{-1}$ and 75.8 mg $m^{-2}s^{-1}$, respectively), and lowest in summer (53.5 mg $m^{-2}s^{-1}$). This is consistent with near-surface results. Below 200 m, $PM_{2.5}$ concentration is relatively uniform. As the height increased, the $PM_{2.5}$ concentration decreased gradually. Between 200-600 m, $PM_{2.5}$ concentration begins to decrease rapidly, but the rate of decline was obviously different in different seasons. In autumn and winter, the reduce rate of $PM_{2.5}$ concentration was significantly higher than that in spring and summer. As a result, the spring $PM_{2.5}$ concentration at 400 m began to be greater than that in winter; the summer $PM_{2.5}$ concentration at 650 m began to be greater than that in autumn, and was at the same level as that in winter. Over 600 m, there is no significant difference in $PM_{2.5}$ concentration between different seasons, while WS varies greatly. Therefore, TF is greatly affected by WS at high altitude, while it is greatly influenced by $PM_{2.5}$ concentration on the near ground. Besides, the TF in the mixing layer is also affected by MLH.

The vertical evolution of the TF is different from both WS and $PM_{2.5}$ concentration, and the seasonal variation remains consistent from near-surface to the upper air, which shows that the TF for spring was the highest, followed by winter and autumn, and summer was the lowest. The vertical variation of TF increases first~~ly~~ and then decreases, and a peak appears around 300 m, at 0.38 mg $m^{-2}s^{-1}$ in spring, at 0.19 mg $m^{-2}s^{-1}$ in summer, at 0.24 mg $m^{-2}s^{-1}$ in autumn, and at 0.31 mg $m^{-2}s^{-1}$ in winter. In the process of TF lowering, it has different performances in different seasons. In spring, the decline slowed down at about 1500 m. The change in summer and autumn is similar. After the peak, TF drops rapidly in summer and autumn. And the decrease rate above 500 m becomes slow, and then slows down again after 1500 m, finally, the TF profiles tend to be vertical. In winter, TF declines rapidly, followed by fluctuations around 1000 m. The above results can preliminarily indicate that the transportation mainly occurs within 200-1500 m, which will be dissect in Sec. 3.3. To sum up, in autumn and winter, the high concentration of $PM_{2.5}$ is concentrated in near the ground, while the TF is not large, again indicating that local emission is the main source of $PM_{2.5}$ in autumn and winter; in spring, affected by high-altitude transportation, $PM_{2.5}$ concentration is high; in summer, both TF and $PM_{2.5}$ concentration are at the lowest level, indicating that regional transport may play an important role in $PM_{2.5}$ concentration in summer.

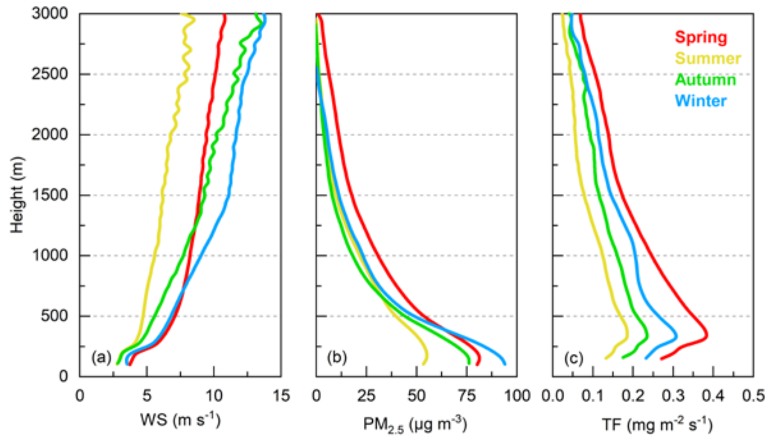

Fig. 6 Vertical profiles of WS (a), $PM_{2.5}$ (b) and TF of $PM_{2.5}$ (c) in different seasons in Beijing.

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3.~~4~~3 TF under different degrees of air pollution

Previous studies have demonstrated that transportation occurs only at the transition period of pollution, and transportation is weak at the peak of pollution (Tang et al. 2015; Zhu et al. 2016). To quantify the transport impact of different pollution levels, the $PM_{2.5}$ concentration was divided into five levels according to the “Technical Regulation on Ambient Air Quality Index (on trial)”

(HJ 633-2012): $PM_{2.5} \leq 35$ μg m^{-3} (clear days), $35 < PM_{2.5} \leq 75$ μg m^{-3} ((slight pollution), $75 < PM_{2.5} \leq 115$ μg m^{-3} (light pollution), $115 < PM_{2.5} \leq 150$ μg m^{-3} (medium pollution) and $PM_{2.5} > 150$ μg m^{-3} (heavy pollution). An interesting phenomenon is that with the increase in altitude, the heavier the pollution near the ground is, the greater the reduction rate of the $PM_{2.5}$ concentration is (Fig. 7). As a result, there is a reversal at 1000-1500 m. In other words, the more severe the near-surface pollution, the lower the high-altitude $PM_{2.5}$ concentration. This is particularly outstanding in the spring: from a clear to a heavy polluted day, the TF at 100 m was, in turn, 0.15 mg $m^{-2}s^{-1}$, 0.26 mg $m^{-2}s^{-1}$, 0.32 mg $m^{-2}s^{-1}$, 0.39 mg $m^{-2}s^{-1}$, 0.66 mg $m^{-2}s^{-1}$, and at 2600 m, the values dropped to 0.15 mg $m^{-2}s^{-1}$, 0.17 mg $m^{-2}s^{-1}$, 0.13 mg $m^{-2}s^{-1}$, 0.10 mg $m^{-2}s^{-1}$ and 0.07 mg $m^{-2}s^{-1}$, respectively. That is, the lower the pollution degree, the more vertical the TF tends to be. This is related to the MLH, because a high MLH is conducive to the diffusion of pollutants in the vertical direction. With the worsening of pollution, the MLH shows a downward trend (Fig. S2).slight haze), $75 < PM_{2.5} \leq 115$ μg m^{-3} (light haze), $115 < PM_{2.5} \leq 150$ μg m^{-3} (medium haze) and $PM_{2.5} > 150$ μg m^{-3} (heavy haze).

According to the previous analysis, two peaks may appear in the TF profile, indicating that the transport occurs at two different heights, approximately 200 m (low-altitude transport) and 1000 m (high-altitude transport), respectively. Due to the sudden increase in the WS at approximately 200 m, the low-altitude transport at 200 m is the basic transport height, regardless of the season and the degree of pollution. In contrast, the high-altitude transport is quite special and mainly occurs in the winter when there is significant pollution. A small peak of in the TF can also be found on heavy polluted days in the summer. Although the change in the TF profile of medium pollution in the autumn is not as obvious as that in the summer and winter, a small increase can still be seen (Fig. 7). In the case of heavy pollution, the MLH is usually less than 1000 m, while in the case of clear and slight pollution, the MLH is close to the height of high-altitude transport (Fig. S2). Therefore, it can be inferred that the pollutants transported at a high altitude during heavy pollution are stored in the residual layer, and when the mixing layer becomes higher, the pollutants stored in the residual layer diffuse into the mixing layer, affecting the pollution level within the mixing layer. This may be a key contributor to the slight pollution in the summer, autumn and winter, but further research is needed. With pollution aggravation, the TF in Beijing increased by varying degrees during different seasons, and the transportation direction gradually shifted from northwest to south (except during winter) (Fig. 7). In particular, the TF continued to increase only in spring, from 93 ± 124 mg $m^{-1}s^{-1}$ on clear days to 382 ± 438 mg $m^{-1}s^{-1}$ on heavily polluted days, which may be caused by more dust during spring. With the except of during spring, with pollution deterioration, the TF showed an increasing trend at the initial stage of pollution and decreasing trend during the heavy pollution period. From medium haze to heavy haze, the TF decreased from 328 ± 280 mg $m^{-1}s^{-1}$ to 295 ± 215 mg $m^{-1}s^{-1}$in summer, from 280 ± 336 mg $m^{-1}s^{-1}$ to 243 ± 238 mg $m^{-1}s^{-1}$ in autumn, and from 240 ± 297 mg $m^{-1}s^{-1}$ to 212 ± 209 mg $m^{-1}s^{-1}$ in winter. These results indicate that although the region south of Beijing is the main transport source during summer and autumn in Beijing, this contribution is significantly reduced during the severe pollution period. In winter, with pollution aggravation, the transportation direction changed from northwest to southwest and finally to the north. In contrast to other seasons, the north wind with a low WS was the main wind during heavy pollution in winter, indicating that regional transport contributed less to heavy pollution during winter in Beijing. In general, the transport of pollutants from the southwest is the main controlling factor for pollution occurrence during spring in Beijing. In other seasons, regional transport plays an important role in the initial period of pollution, while local emissions during the period of heavy pollution are the

~~main controlling factor.~~

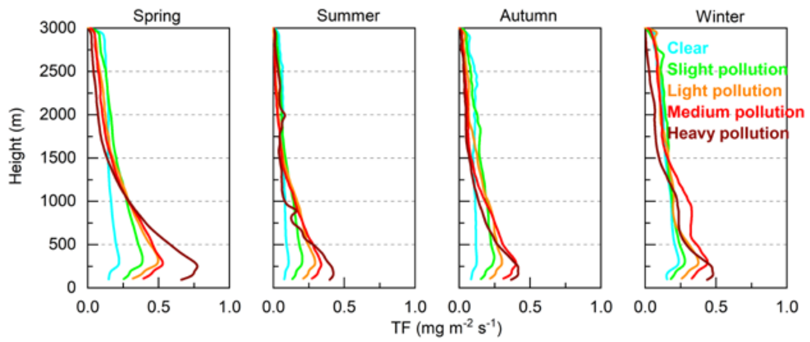
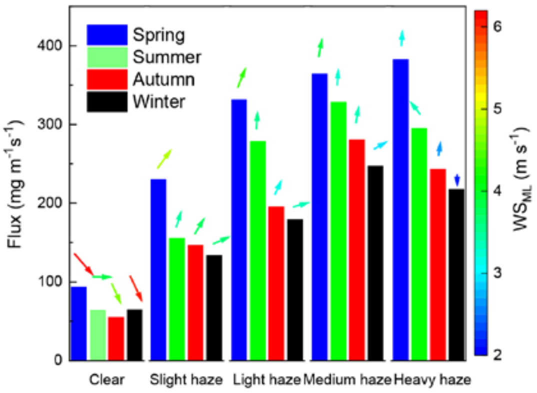


Fig. 7 Vertical profiles of TF of $PM_{2.5}$ under different degrees of pollution in different seasons in Beijing. Clear days: $PM_{2.5} \leq 35$ µg m^{-3}, slight pollution: $35 < PM_{2.5} \leq 75$ µg m^{-3}, light pollution: $75 < PM_{2.5} \leq 115$ µg m^{-3}, medium pollution: $115 < PM_{2.5} \leq 150$ µg m^{-3} and heavy pollution: $PM_{2.5} >$ 150 µg m^{-3}



According to the same division method, we further explored the seasonal variation of the TF and transport source in the mixing layer under different pollution degrees. With pollution aggravation, the mixing layer TF in Beijing increased by varying degrees during different seasons, and the transport direction gradually shifted from northwest to south (except during the winter) (Fig. 8). In particular, the mixing layer TF in the spring is significantly higher than that in the other seasons at all pollution degrees, which is 1.1-2.0 times that in the other seasons. This may be caused by the greater amount of dust during the spring. With the pollution deterioration, the TF showed an increasing trend in the initial stage of pollution and a decreasing trend during the heavy pollution period. From medium pollution to heavy pollution, the TF decreased from 5.50 ± 4.83 mg m^{-1}s^{-1} to 4.69 ± 4.84 mg m^{-1}s^{-1}in the spring, from 3.94 ± 2.36 mg m^{-1}s^{-1} to 3.39 ± 1.77 mg m^{-1}s^{-1} in the summer, from 3.72 ± 2.86 mg m^{-1}s^{-1} to 3.01 ± 2.40 mg m^{-1}s^{-1} in the autumn, and from 4.45 ± 4.40 mg m^{-1}s^{-1} to 3.25 ± 2.77 mg m^{-1}s^{-1} in the winter. Among them, the largest drop was found in the winter. In the winter, with the pollution aggravation, the transport direction changed from northwest to southwest and finally to the north. In contrast to in the other seasons, the weak north wind was the main wind during heavy pollution in the winter, indicating that regional transport contributed less to the heavy

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pollution during the winter in Beijing. In the initial stage of pollution, the TF continued to increase, but the rate of increase gradually slowed in the spring and summer. From light haze to medium haze, the TF decreased by 0.1mg $m^{-1}s^{-1}$ in the spring and increased by only 0.07 mg $m^{-1}s^{-1}$ in the summer. It is also not difficult to find from the changes in the TF profile (Fig. 7) that the regional transport has little impact on the medium pollution in the spring and summer. These results indicate that although the region south of Beijing is the main transport source in Beijing, its contribution is significantly reduced during the severe pollution period. In general, regional transport plays an important role in the initial period of pollution, while local emissions are the main controlling factor during the period of heavy pollution. The parabolic pattern of the TF is the result of a combination of the WS and $PM_{2.5}$ concentration. The TF reaches a threshold during medium pollution, which is the critical moment mentioned above.

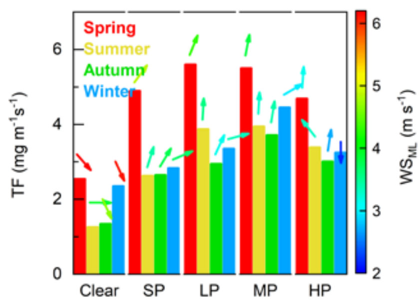

Fig. 2. The mixing layer ~~transport flux~~TF of $PM_{2.5}$ levels ~~of PM~~ and transportation directions under different degrees of pollution in different seasons in Beijing. (SP denotes slight pollution, LP denotes light pollution, MP denotes medium pollution and HP denotes heavy pollution.)

4. Conclusions

To understand the characteristics of $PM_{2.5}$ ~~fine particulate matter~~ transport flux in Beijing, the height of the atmospheric mixing layer and wind profile within the mixing layer in Beijing were observed for a 2-year period. The main conclusions are as follows:

(1) By analyzing the variations of VC, atmospheric dilution capability in Beijing is strongest in spring and weaker in summer, autumn and winter. In spring, vertical and horizontal dilution capacities are strong; in autumn and winter, vertical and horizontal dilution capacities are weak; in summer, vertical dilution capability is strong and horizontal dilution capability is weak. The diurnal variation in VC is consistent with MLH, which shows that the dilution capability is strongest before sunset, gradually weakens after sunset and remains stable at night. In spring, vertical and horizontal dilutions are strong during both day and night. In winter, vertical dilution is weak during the day, and horizontal dilution during the night is the main. In summer, vertical dilution during the day is dominant. Although there is little difference in diffusivity between summer, autumn and winter, poor dilution capability occurs more frequently in autumn and winter~~By analyzing the variation characteristics of VC, the TC in Beijing is strongest in spring and weaker in summer, autumn and winter. In spring, vertical and horizontal diffusion capacities are strong; in autumn and winter, vertical and horizontal diffusion capacities are weak; in summer, vertical diffusion capacity is strong~~

~~and horizontal diffusion capacity is weak. The diurnal variation in VC is consistent with MLH, which shows that the TC is strongest before sunset, gradually weakens after sunset and remains stable at night. In spring, vertical and horizontal diffusion are very strong during both day and night. In winter, vertical diffusion is weak during the day, and horizontal transportation during the night is the main means of transportation. In summer, vertical diffusion during the day is dominant. Although there is little difference in diffusivity between summer, autumn and winter, poor TC occurs more frequently in autumn and winter~~.

~~(2) $PM_{2.5}$ concentrations during different seasons have different responses to MLH, WS_{ML} and VC. During the three dry seasons of winter, spring and autumn, the concentration of pollutants has a good relationship with VC, indicating that the main dissipation method of pollutants is diffusion. In summer, there is a weak positive correlation between pollutant concentration and VC, which is related to strong photochemical reactions.~~

(~~3~~2) TF is largest in spring and smaller in summer, autumn and winter in Beijing. The high TF mainly comes from southward transport, while the low TF is accompanied by northwest transport. The transport mainly occurred between 14:00 and 18:00 LT. And the height of transport is around 200 m and 1000 m. Using the $PM_{2.5}$ concentration as a classified index of air pollution, the results show that the regional transport from the southern area plays an important role in the initial period of pollution, and local emissions are the main controlling factors in the heavy pollution period, especially in winter. ~~TF is largest in spring and smaller in summer, autumn and winter in Beijing. The high TF mainly comes from southward transport, while the low TF is accompanied by northwest transport. Using the $PM_{2.5}$ concentration as a classified index of atmospheric pollution, the results show that the regional transport of pollutants from the southwest is the main controlling factor of pollution during spring in Beijing, while during the other seasons, the regional transport from the southern area plays an important role in the initial period of pollution, and local emissions are the main controlling factors in the heavy pollution period, especially in winter.~~

To solve the problem of heavy pollution in north China, joint prevention and control has been put forward for a long time. Even so, there is still no concrete implementation plan. To break through this embarrassing situation, this study quantifies TF to explain the time period when the transport occurs and the main areas affected on Beijing. In this study~~, the response of particulate matter to meteorological conditions in the mixing layer was studied~~, ~~and the difference in the seasonal response was found. T~~atmospheric~~he transport~~ dilution ~~capacity~~capability during different seasons and the ~~transport flux~~TF during different pollution periods were also discussed. The important role of transport in the initial period of pollution is emphasized. And local pollutant emission control is the most effective way of mitigating pollution levels. The research results are of great significance to the early warning, prevention and control of atmospheric particulate pollution. ~~However, due to the limitation of observational data, the near-surface particle concentration was used to replace the concentration column for discussion purposes, resulting in uncertainty in the result. In the future, this issue will be further discussed in combination with ground-based telemetry lidar.~~

Data availability

The data in this study are available from the corresponding author upon request (tgq@dq.cern.ac.cn).

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Author contribution

GT and YW designed the research, LZ, BH, BL and YunL conducted the measurements. YusL and GT wrote the paper. SL reviewed and commented on the paper.

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Competing interests

The authors declare that they have no conflict of interest.

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