

## Authors' Response to Comments of Reviewer #1

*Referee comments are in black. Author responses are in blue and revised manuscript in blue marked by underline.*

Authors present research on an interesting topic: the influence of meteorological parameters and aerosol concentrations measured at ground level on radiative forcing. They use up-to-date methods and data sources; however, in my opinion, the manuscript in its current form is not suitable for publication, and some points should be explained, changed, or even require additional data analysis.

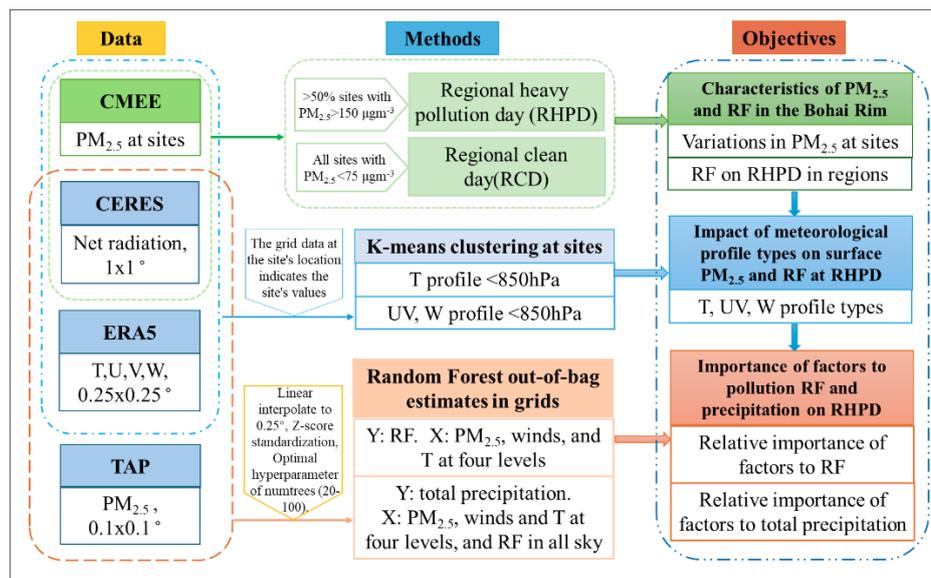
**Response:** We are grateful to you for your time, insightful comments, and constructive suggestions, which have been invaluable in helping us to significantly improve the quality and clarity of our work. We have carefully considered each point raised and have made revisions to the manuscript accordingly.

1. Moreover, the way the authors present their findings is chaotic and sometimes hard to follow. The figures are sometimes unreadable, captions are too small, etc. There is probably too much data presented in the manuscript; maybe moving some data to an appendix would help and allow readers to focus on the main scope of the manuscript.

**Response:** ①To present our work more clearly, we added a Methodology section in Section 2 as “2.4 Methodology”, incorporating “2.4.1 k-means clustering method” and “2.4.2 Importance estimation based on Random Forest algorithms” in this section. And a new subsection entitled “2.4.3 The data usage workflow” has also been added after that. This section specifically addresses ambiguities and unclear points regarding methods in the original text, and includes an additional figure: Figure 1. The data usage workflow and framework in this work. The figure clearly illustrates the data, methods, and objectives employed in this work, along with the data and methods used for each specific objective. Besides, we added the introduction of the data usage workflow and framework as “Figure 2 exhibited the data usage workflow and framework in this work. The CMEE PM<sub>2.5</sub> data at sites and CERES net radiation flux data were used to show the characteristics of PM<sub>2.5</sub> and RF on regional heavy pollution days (RHPD) in the Bohai Rim.....”

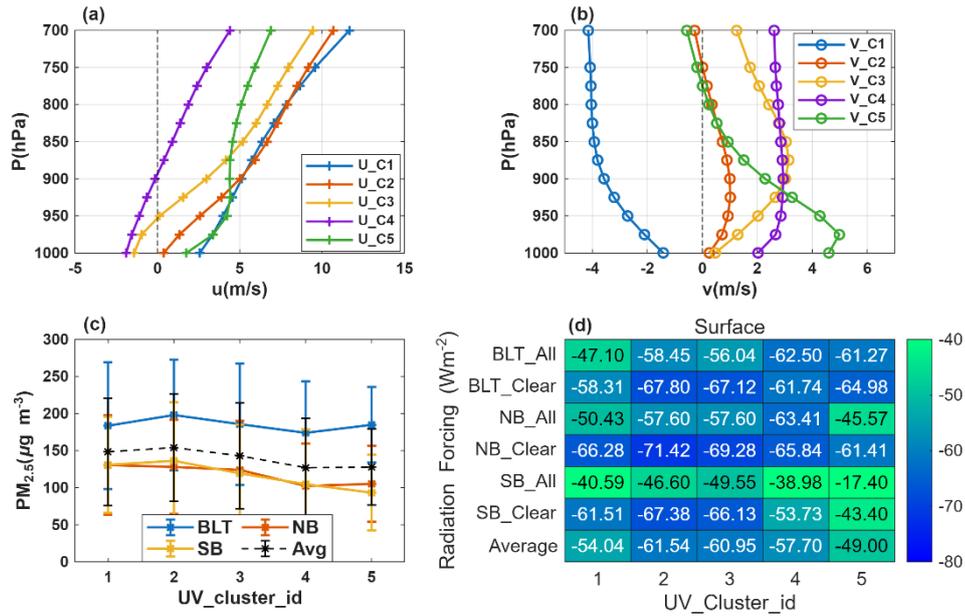
Subsequently, CMEE, CERES, and ERA5 data were combined to analyze the impact of meteorological profile types (temperature and winds) on surface  $PM_{2.5}$  and RF on regional heavy pollution days by using the k-means clustering method at the stations.....

To explore the relative importance of factors to pollution RF and precipitation on regional heavy pollution days in the Bohai regions, the gridded data of CERES, ERA-5, and TAP were jointly applied to the random forest algorithm....”

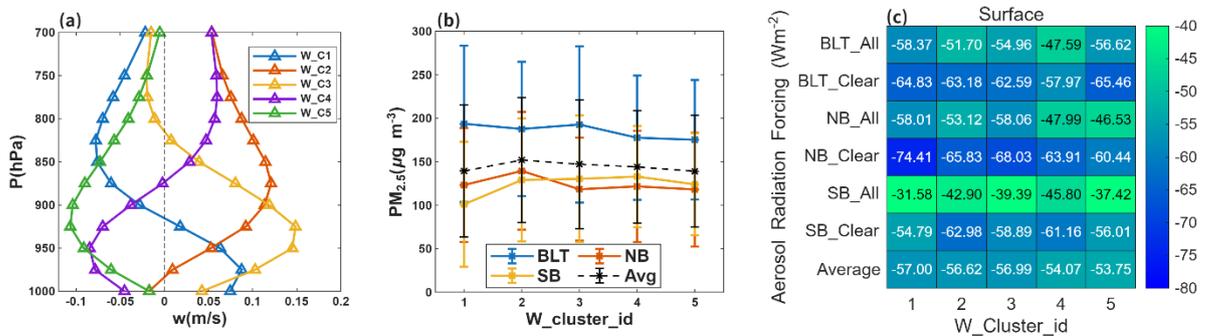


**Figure 1.** The data usage workflow and framework in this work. (Figure 2 in the revised manuscript).

②Regarding your concerns about image clarity and excessive data presentation, we have revised Figures 5 and 6 in the original manuscript. We have enlarged the text within the figures and condensed the original Figures 5 and 6 to include only wind classification,  $PM_{2.5}$ , and surface RF subplots (as shown below: Figures 2 and 3). The remaining sections of the original text (percentage statistics at stations, and radiative forcing (RF) at the top and in the atmosphere) have been relocated to supplementary file (Figure S6 and Figure S7). This approach enables readers to focus on the core content and objectives of the manuscript.



**Figure 2.** The five horizontal wind profiles under 850 hPa by k-means clustering on the regional heavy pollution days: (a) u wind, (b) v wind, (c) the PM<sub>2.5</sub> concentrations, and (d) the radiative forcing in the three regions in clear- and all-sky at the surface at the five clusters. (the Figure 5 in origin version and Figure 6 in revised version).



**Figure 3.** The five vertical wind profiles under 850 hPa by k-means clustering on the regional heavy pollution days: (a) the w wind, (b) the PM<sub>2.5</sub> concentrations, and (c) the radiative forcing in the three regions in clear- and all-sky at the surface. (the Figure 6 in origin version and Figure 7 in revised version).

2. The main issue of the manuscript, in my opinion, is how the authors “compare” in-situ data with satellite measurements. They use multisource data, which is fine; however, the relationship between them and the transition from in-situ measurement data to

gridded data is unclear.

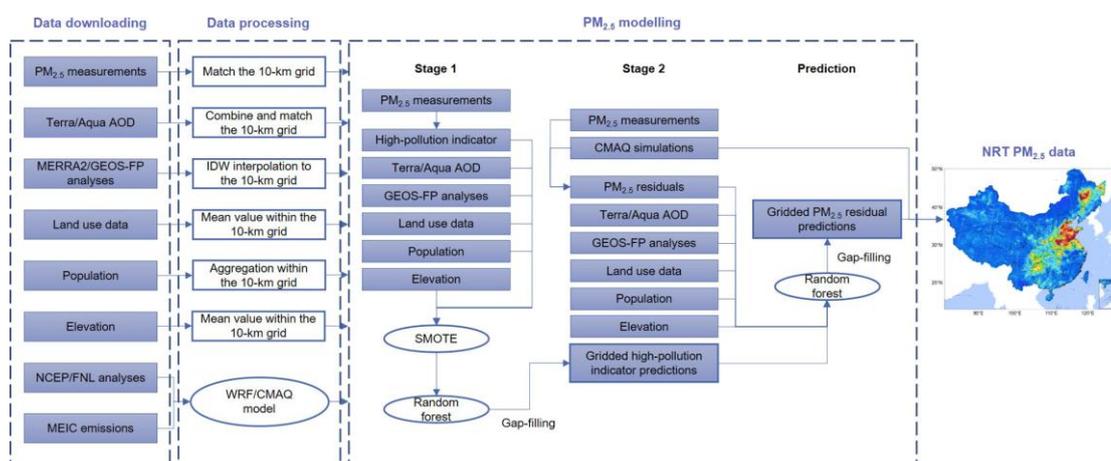
**Response:** This comment helped us recognize that the original manuscript lacked sufficient explanation regarding the comparison between ground-based observational data and satellite measurement data. To address this, we have expanded [“2.4.3 the data usage workflow”](#) section within Section 2, and explicitly describe the comparison between in-situ and grid data as [“...CMEE, CERES, and ERA5 data were combined to analyze the impact of meteorological profile types \(temperature and winds\) on surface PM<sub>2.5</sub> and RF on regional heavy pollution days by using the k-means clustering method at the stations. The RF and meteorological parameters at stations were extracted from grid data of CERES and ERA5, respectively. The extracting method is that the grid data at the site's location indicates the site's values...”](#). For Section 3.2, site-based PM<sub>2.5</sub> data (derived from in situ observations) were employed. Radiative forcing data were calculated using satellite CERES data from the grid cell containing the site. Vertical meteorological profiles were sourced from ERA5 data corresponding to the grid cell at the site location. For Section 3.3, all data employed are grid-based. A random forest importance analysis was conducted using grid-based TAP, ERA5, and CERES data; this section did not utilize site-based observational data.

3. For instance, they use a monitoring network to identify high-concentration episodes, which is acceptable. Then, some gridded data are used in statistical analysis. What happens between in-situ and gridded data? How does TAP work? A brief description is needed in the manuscript. By the way, why do the authors use ERA-5 data while the TAP website claims that meteorological data are combined with aerosol data? If ERA-5 data are better, then what is the quality of TAP aerosol data? The authors use PM<sub>2.5</sub> data as a predictor in the Random Forest model. They claim it is a proxy for anthropogenic sources; however, elsewhere they discuss the influence of transport on local concentrations, and in another place, they state that columnar optical properties determine radiative forcing. So, is local PM<sub>2.5</sub> a predictor of radiative forcing or not? Maybe it would be better to use emission inventories—TAP claims that one is incorporated—to estimate anthropogenic sources instead of PM<sub>2.5</sub> concentrations? Moreover, in section 3.3, the authors again claim that PM<sub>2.5</sub> is related to emissions while meteorological profiles reflect diffusion. I cannot agree: emissions together with diffusion factors influence PM<sub>2.5</sub> concentrations. So, PM<sub>2.5</sub> is not an independent

variable, contrary to what is stated in the conclusion.

Response: ① Regarding the insufficient description of data usage and the handling of in-situ and grid data, we have already outlined our revisions in the first two responses and will not elaborate further here.

② For the TAP data, Figure 4 shows the operational process of the near real-time PM<sub>2.5</sub> data generated from TAP (Geng et al., 2021). In the revised version, additional descriptions have been included to demonstrate the quality of the data, as follows: “The TAP PM<sub>2.5</sub> is estimated based on a two-stage machine learning model coupled with the synthetic minority oversampling technique and a tree-based gap-filling method, which improves the PM<sub>2.5</sub> estimations on highly polluted days. The TAP PM<sub>2.5</sub> showed a higher regression slope (0.97) when evaluated against ground measurements (Geng et al., 2021).”.



**Figure 4.** Operational process of the near real-time PM<sub>2.5</sub> data generated from TAP (Geng et al., 2021).

③ Regarding the use of TAP PM<sub>2.5</sub> as a predictive variable, we are most grateful for your identification of this critical issue. We recognize that the original manuscript contained inconsistencies and imprecision in its discussion of PM<sub>2.5</sub>. Our original intent was to examine the contribution of surface PM<sub>2.5</sub> to RF by the Random Forest model, rather than treating it as an independent emission source. Consequently, characterizing it as an approximate emission source in the original text was indeed imprecise. In the revised manuscript, we have modified this section's wording, removed the statement that it represents anthropogenic emission sources and replaced it with “surface aerosol concentrations” (our work focuses on regional heavy pollution days with PM<sub>2.5</sub>>150

$\mu\text{gm}^{-3}$ ). The reason we did not employ emission inventories is that current inventories typically lack sufficient spatial-temporal resolution. In contrast, TAP PM<sub>2.5</sub> offers high spatiotemporal resolution (near real-time daily full-coverage PM<sub>2.5</sub> data at a spatial resolution of 10 km). Besides, it performs well under conditions of high pollution (a higher regression slope of 0.97 when evaluated against ground measurements in Beijing-Tianjin-Hebei (Geng et al., 2021)), and thus better reflects the actual amount of radiation-related surface aerosols formed. Furthermore, we explicitly recognize the limitations of TAP PM<sub>2.5</sub> being influenced by atmospheric diffusion transport in the discussion section of the paper as “TAP PM<sub>2.5</sub> data contains the meteorological information, and some other factors have not been taken into account in the machine learning such as terrain elevation.”

Reference: Geng, Guannan, Qingyang Xiao, Shigan Liu, et al. 2021. ‘Tracking Air Pollution in China: Near Real-Time PM<sub>2.5</sub> Retrievals from Multisource Data Fusion’. *Environmental Science & Technology* 55 (17): 12106–15. <https://doi.org/10.1021/acs.est.1c01863>.

4. Another example concerns the “mean profiles” of meteorological parameters. It is written that the authors used profiles at over 11 stations. How are they representative for the  $0.25 \times 0.25$  grid used in the Random Forest analysis? Is there any local orography favoring aerosol transport or accumulation in valleys? What about sea-land differences? I can understand that clustering is performed over land (land stations). It seems that clustering and Random Forest are independent. So, it should be explained somewhere why the authors perform such investigations.

**Response:** This opinion has provided us with inspiration and deeper insight.

①As previously addressed in our earlier response, clustering and random forests are distinct and independent methodologies. Cluster analysis focuses on station-based investigations, primarily examining how variations in temperature and wind profiles within the boundary layer (below 850 hPa) influence PM<sub>2.5</sub> concentrations and RF, all conducted at land-based observation stations. Random forests, however, represent a regional analysis, aiming to investigate the relative importance of PM<sub>2.5</sub> and temperature and wind at various altitudes (1000, 850, 700, and 500 hPa) for RF. To address this, we have incorporated relevant clarifications in the revised manuscript, such as the data utilization workflow in the Methods section in the above response, and

added the rationale for this analysis in the first paragraph of Section 3.3 as “.... Additionally, the above impact was based on stational statistics. Therefore, our analysis in this section was conducted at all the grids within the Bohai regions....”.

② Regarding the influence of topography and land-ocean differences, it is certain that these factors affect both  $PM_{2.5}$  and radiative forcing. Regarding topography, Figure 1 in the manuscript illustrates the terrain characteristics within the study area. The northwestern in study area is mountainous, where southerly or easterly winds favors pollution accumulation in land, while westerly winds facilitate pollutant transport from land to the ocean. In the regional variation analysis, we added the discussion of spatial heterogeneity related to driving factors such as aerosol concentrations, RF values, local topography and geographical location in section 3.2.1 as “The importance of influencing factors to pollution RF showed heterogeneity across different regions. This may be related to variations in aerosol concentrations and pollution RF. In clear sky, the BLT region featured high  $PM_{2.5}$  concentrations (Figure 3) coupled with high RF values (Figure 4a), whereas the SB region exhibited low  $PM_{2.5}$  concentrations alongside low RF, illustrating the higher importance of  $PM_{2.5}$  impacts on RF at the surface in these two regions (Figure 8a). Conversely, the NB region, with low  $PM_{2.5}$  concentrations but high RF, showed less importance of  $PM_{2.5}$  than in the BLT and SB. Differences in the importance of meteorological parameters across regions may be related to local topography and geographical location. Figure 1 indicates that the northwestern parts of the BLT and NB regions border mountainous terrain, where wind direction and speed significantly influence pollution dispersion and transport. Consequently, wind (V-wind in the BLT region, U- and V-winds in the NB region) exhibited greater importance for RF in these regions compared to the SB region. Conversely, the SB region's lower latitude and absence of nearby high mountain ranges resulted in temperature factors being more important than in the BLT and NB regions.”.

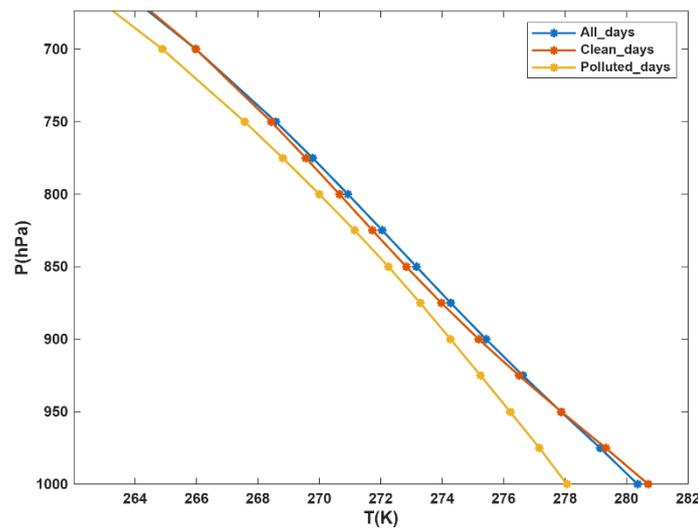
③ Regarding the land-sea differences, we demonstrated the variations in influencing factors between land and ocean at the end of Section 3.3.1. In the revised manuscript, we have expanded the analysis of influencing factors related to land-ocean differences, as “The relative importance of influencing factors between land and ocean also exhibited certain variations. Regarding the surface and atmosphere RF, the significance of  $PM_{2.5}$  over land surpassed that over the ocean, while the importance of

U and V over the ocean exceeded that over land areas (U500 in clear-sky, V1000 in all-sky). Given that land surfaces exhibited higher aerosol concentrations than oceans, the prominence of PM<sub>2.5</sub> over land was entirely justified. The heightened significance of U and V over the ocean was primarily linked to transport from land to the ocean. Based on the land-ocean distribution in Figure 1, it is evident that zonal winds (particularly westerlies) facilitate the transport of land-based pollutants towards the ocean, while meridional winds also promote this transport (both southerly and northerly winds). Conversely, transport from the ocean to land also occurred, though its impact was comparatively minor relative to the influence of high-concentration aerosols over the land.”.

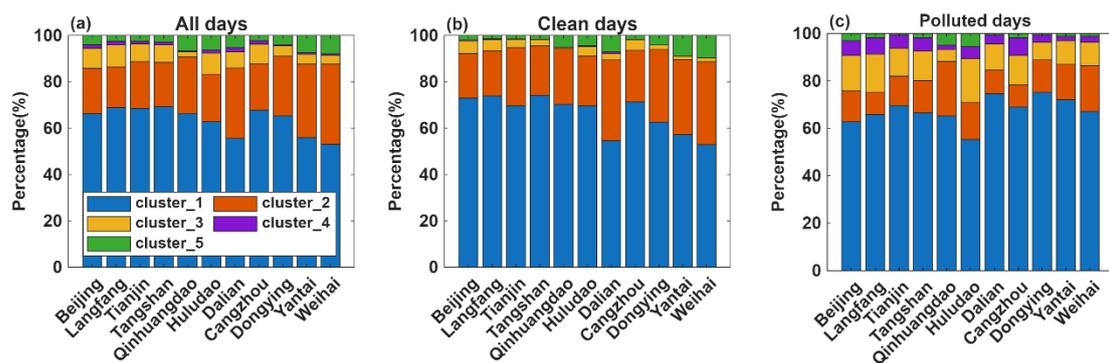
5. Another question is: what are the profiles during the rest of the analyzed time, not only during high-concentration episodes? For example, temperature profile clusters 2 to 4 exhibit inversion from around 950 to 850 hPa. The frequency of occurrence during the investigated episodes is around 30–40%. What happens during the rest of the investigated period (autumn, winter)? Another issue is how “wind clusters” are presented. Figure 5a is completely unreadable. Maybe the authors will find another way to present changes with the altitude of wind speed and direction?

**Response:** ①As for this question, we compared the average meteorological profiles during the study period between all days, regional clean days, and regional heavy pollution days across the region (see Figure 5 below). It can be observed that the average profiles showed subtle differences between heavy polluted and clean days, failing to reflect the presence of temperature inversions. Consequently, we clustered temperature profiles from all stations during autumn and winter into also five categories (decreasing, decreasing with isotherms, low-level inversion, mid-level inversion, and upper-level inversion). We then calculated the proportion of each profile type across all days, regional clean days, and regional heavy pollution days, as shown in Figure 6. The comparison clearly demonstrates that the proportion of inversion conditions on regional heavy pollution days is significantly higher than that on regional clean days, and also exceeds the proportion across all days, indicating the crucial role of inversions during heavy pollution episodes. This section has been placed in the supplementary file (Figure S5) and we added the comparison in section 3.2.1 as “The comparison of proportions of different temperature profile types on all days, regional clean days, and regional

heavy pollution days in the study period (Figure S5) showed clearly that the proportion of temperature inversions occurring on regional heavy pollution days was significantly higher than that on regional clean days, and also exceeded the proportion across all days, indicating the significant role of temperature inversions on regional heavy pollution days.” .



**Figure 5.** Average temperature profiles for all days, regional clean days, and regional heavy pollution days during autumn and winter at monitoring stations within the study region.



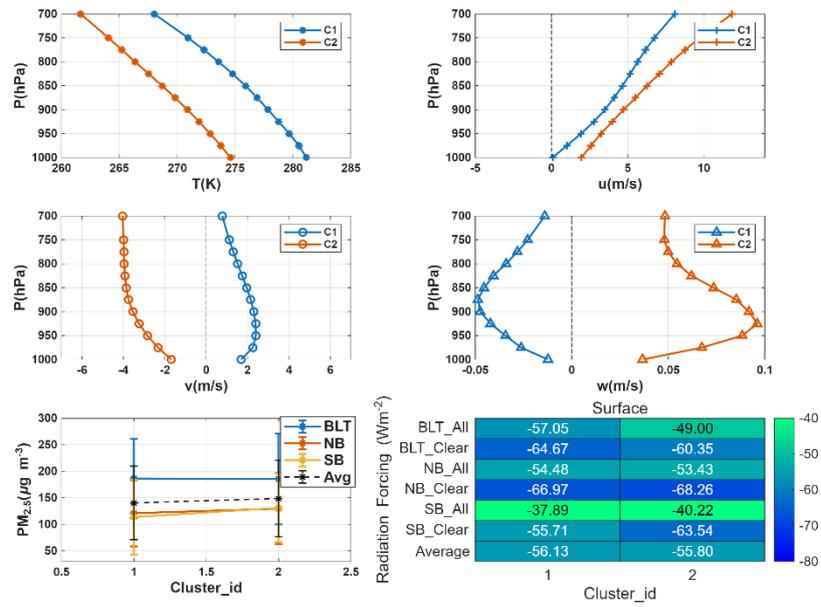
**Figure 6.** Proportions of different temperature profile types (C1: decreasing, C2: decreasing with isotherms, C3: low-altitude inversion, C4: mid-altitude inversion, and C5: upper-altitude inversion) on all days (a), regional clean days (b), and regional heavy polluted days (c) at each station during autumn and winter 2014-2023.

②To address the presentation of wind clustering, we have redrawn the figures,

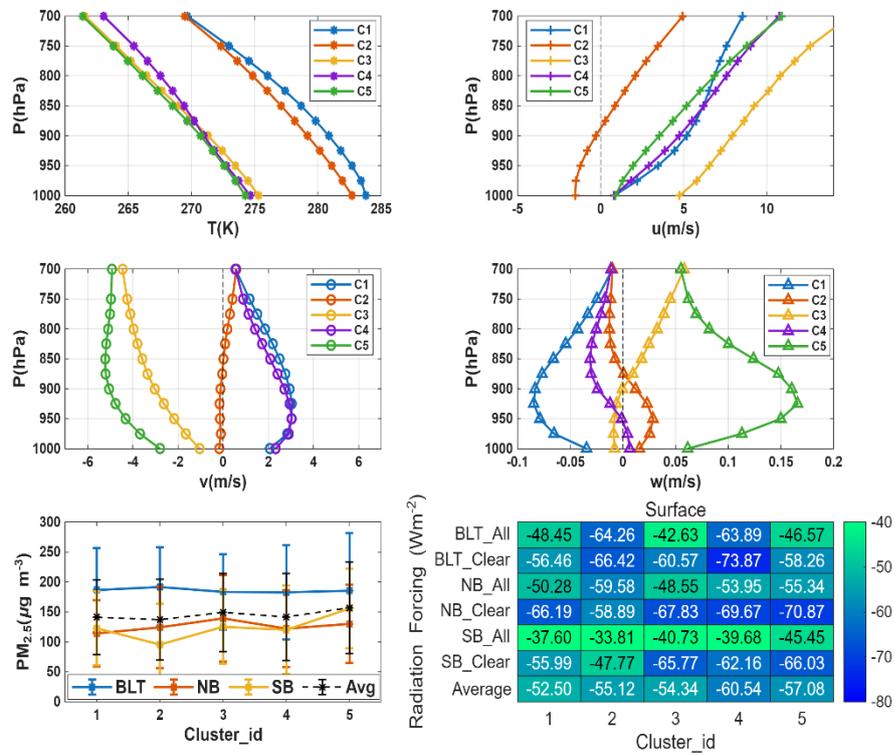
separating the u and v components in Figure 5a to enhance clarity (Figure 2 in this response document). The frequency statistics and RF\_TOA and RF\_Atmos subplots have been placed in the supplementary file (Figure S6 in the new supplementary file).

6. Regarding mean clustered profiles, it would be interesting to connect temperature profiles with wind profiles. I suggest performing multidimensional cluster analysis to find meteorological situations favoring large PM<sub>2.5</sub> concentrations—for example, inversion and low wind speed near the ground.

**Response:** We agree with your proposal. The k-means clustering method we employed is a multidimensional clustering technique characterized by high computational efficiency. Following your recommendation, we applied k-means clustering to the combined temperature and wind field profiles, utilizing the elbow method to determine the number of cluster (2 clusters) (results shown in Figure 7) or the same 5 clusters as in the original manuscript (results shown in Figure 8). We can find that these classification approaches failed to capture variations in the height of temperature inversion layers (our attempts with clusters 2–10 all proved incapable of reflecting inversion layer height changes). It also cannot distinguish which types of conditions favors increased PM<sub>2.5</sub> concentrations. Furthermore, this combined clustering fails to reflect the relative importance of temperature and wind profile variations on PM<sub>2.5</sub> and RF. Therefore, although clustering temperature and wind profiles together is meaningful, this combined approach struggles to identify meteorological conditions conducive to elevated PM<sub>2.5</sub> concentrations (such as temperature inversions and low near-surface wind speeds). Consequently, our work employed separate clustering methods for temperature and wind profiles. The results of this joint clustering analysis have been added to the supplementary file and were described in the Methods section 2.4.3 of the article, as “For the k-means clustering, we clustered the T, horizontal, and vertical winds separately to capture variations in the height of temperature inversion layers and to reveal the relative importance of temperature and wind profile variations below 850 hPa on PM<sub>2.5</sub> and RF. The clustering of combined T and winds by the k-means method failed to capture the temperature inversion layers, and could not reveal the relative importance (see Figure S1 and S2).”



**Figure 7.** The results of clustering of combined T and winds by the k-means algorithm using the elbow method to determine the number of clusters of 2 (this clustering of 2 is distinct for temperature and winds, but failed to capture the temperature inversion layers). (Figure S1 in the supplementary file)



**Figure 8.** The results of clustering of combined T and winds by the k-means algorithm using the same number of clusters in the manuscript of 5 (this clustering of 5 also failed to capture the temperature inversion layers). (Figure S2 in the supplementary file)

7. One major weakness of the manuscript is the insufficient discussion of the Random Forest analysis. The authors should elaborate on why individual parameters influence radiative forcing, separately for the clear-sky and all-sky cases. What are the potential mechanisms? What is the influence of clouds? Furthermore, the land-sea aspect requires a more in-depth analysis, particularly regarding aerosol transport between sea and land—for example, the influence of wind speed and direction.

**Response:** In accordance with the proposed suggestions, we have expanded the analysis and discussion concerning the random forest results in section 3.3.1. The additional content includes:

① The mechanisms underlying differences in influencing factors between clear-sky and all-sky conditions. In clear-sky, the direct radiative effect of high-concentration surface aerosols dominates the radiative forcing at the surface and in the atmosphere. In all-sky, cloud radiative effects become significant, with cloud-radiation interactions influenced jointly by aerosols and meteorological conditions, where meteorological factors may play a more prominent role. The added content is as follows: “The importance of factors to the pollution RF in clear-sky and all-sky showed a distinct difference: Surface PM<sub>2.5</sub> was the most important factor to surface and atmosphere RF in clear-sky, but V wind in high level (500 hPa) in all-sky. In clear-sky, high-concentration surface aerosols directly interact with shortwave radiation (through scattering and absorption), becoming the most important factors of pollution RF at the surface and in the atmosphere. Other meteorological factors may indirectly influence RF by altering aerosol distribution (including horizontal and vertical distribution) and aerosol chemistry formation to affect the RF indirectly, but their effect is far less pronounced than the direct interaction between high-concentration surface aerosols and radiation. In the all-sky, however, the pollution RF is primarily influenced by cloud-radiation interactions. Cloud radiative forcing is affected by cloud characteristics (cloud cover, cloud height, cloud type, etc.), which in turn are influenced by both aerosols and meteorological conditions, with the latter potentially playing a more significant role.

The critical role of upper-level (500 hPa) V-wind may be attributed to the strong association between weather systems over the Bohai Rim region and upper-level zonal winds.”.

② Analysis of differences in influencing factors between land and sea. These differences primarily stem from the high importance of PM<sub>2.5</sub> over land and the relatively high importance of u and v factors over the ocean. We have linked these differences in influencing factors to pollutant transport between land and sea. The added content is as follows: “The relative importance of influencing factors between land and ocean also exhibited certain variations. Regarding the surface and atmosphere RF, the significance of PM<sub>2.5</sub> over land surpassed that over the ocean, while the importance of U and V over the ocean exceeded that over land areas (U500 in clear-sky, V1000 in all-sky). Given that land surfaces exhibited higher aerosol concentrations than oceans, the prominence of PM<sub>2.5</sub> over land was entirely justified. The heightened significance of U and V over the ocean was primarily linked to transport from land to the ocean. Based on the land-ocean distribution in Figure 1, it is evident that zonal winds (particularly westerlies) facilitate the transport of land-based pollutants towards the ocean, while meridional winds also promote this transport (both southerly and northerly winds). Conversely, transport from the ocean to land also occurred, though its impact was comparatively minor relative to the influence of high-concentration aerosols over the land.”.