Reply to the comments of RC3

RC3: General comment

General comment 1: The manuscript aims to evaluate and compare four dry deposition schemes implemented in CMAQ v5.4, focusing on simulating an East Asian dust episode from January 2023. The authors then selected the scheme by Emerson et al. (2020) (E20) for further evaluation of dust and black carbon transport. The main issue with this work, in its current form, is that the conclusions, particularly given the strong tone used in parts of the text, are not well supported by the marginal differences between the four schemes. Specifically, the results in Table 4 and Figure 4 show small statistical significance between the models, so it's hard to justify the claim that the E20 scheme outperformed the others.

Response: We thank the reviewer for the comments. To re-justify the E20 scheme model performance, we have included the model evaluation over most parts of mainland China, which is identified as the dust source/near-source region. 100 observation sites have been obtained from observation sites on the Chinese air quality online monitoring analysis platform's website (www.aqistudy.cn/). The averaged observed PM_{10} and $PM_{2.5}$ concentrations from the responding dataset were used to evaluate the model performance for the latest dust emission scheme and four dry deposition schemes. Also, instead of the 10-day simulation in January 2023, we widened the simulation period by considering the multiple dust storm episodes during spring 2021 for about 40 days of simulation (12 Mar-20 Apr 2021). We added the discussion as "In addition, the hourly PM_{10} and $PM_{2.5}$ of nearly 100 sites distributed over mainland China (Fig. S1), covering the period of 22-31 January 2023 and 12 March-20 April 2021 obtained from the Chinese air quality online monitoring analysis platform's website (www.aqistudy.cn/)." **Page 8, Line 188-190**

Also, we have added the discussion as "During the spring of 2021, a series of dust storms (15 March, 27 March, and 18 April) occurred over the Gobi area, with one of the most significant dust storms in the past decade (15 March, the "3.15" dust storm hereafter) causing environmental impact over the continental (Jin et al., 2022; Gui et al., 2022; He et al., 2022; Liang et al., 2022; Tang et al., 2022). More interestingly, one of the multiple dust storm episodes reached western Pacific Ocean due to the extreme typhoon episode (Kong et al., 2024). Hence, we intend to re-emphasize the precision of various deposition schemes on the CMAQ for the recent dust storm episode over the Asian Continental highlighted by Kong et al. (2024). We evaluated the CMAQ simulations with the different dry deposition schemes for the 40-day sensitivity test on 12 March-20 April 2021 against measured PM_{10} and $PM_{2.5}$ concentrations across the observation sites in mainland China (Table 4). The observation sites used for the model comparison are marked in Fig. S1. Generally, the evaluation results for Taiwan and mainland China were consistent. During the 40 days of Spring 2021, the CMAQ PM_{10} of NMB was the highest for Off_PR11 (NMB = -79.19 %), followed by Dust_PR11 (-60.53 %). The latest inline dust emission scheme embedded with E20 dry deposition scheme for PM_{10} was well performed by NMB of -25.43 %, compared to the Dust_S22 (-45.97 %) and Dust_P22 (-59.82 %). For the $PM_{2.5}$ simulation, Dust_PR11 has been improved from Dust_Off, and Dust_S22 was slightly better than Dust_E20.

Figure 5 shows the scatter plot of simulated and observed PM across mainland China. The correlation coefficient (R), a factor of two (FAC2), and the mean observed and simulated PM are marked in Figure 5. The modeled PM_{10} without the dust scheme had the lowest correlation, followed by Dust_PR11. Among all of these simulations, Dust_E20 performed the best (R > 0.3) compared to Dust_PR11, Dust_S22 and Dust_P22. However, for $PM_{2.5}$, the correlation between the model and measured values was similar for all the dry deposition schemes. The statistical index of FAC2 was used in the present work since either low or high outliers less influence it (Chan and Hanna, 2004). The dataset is reliable for FAC2 values between 0.5 and 2.0, with the ideal model of 1.0. The simulated PM_{10} by E20 performed well, with a nearly perfect value of 1.1. Meanwhile, the $PM_{2.5}$ by S22 simulation was slightly better than E20 but much better than the other experiments." **Page 11, Line 259-284**.

Table 4. CMAQ evaluation for PM_{10} and $PM_{2.5}$ against the averaged 100 observation sites across mainland China (Fig. S1) and AOD against MODIS daily observation near the dust source region (above 30°N) with Normalized Mean Bias (NMB) under the multiple simulation scenarios (Fig. S3). Spring 2021, 3.15, 3.27, and 4.18 represent the evaluation period by 12 March-20 April 2021, 14-16 March 2021, 26-28 March 2021, and 17-19 April 2021, respectively.

Parameters	Period	CMAQ-M3DRY		CMAQ-STAGE		
		Off_PR11	Dust_PR11	Dust_E20	Dust_S22	Dust_P22
PM_{10}	Spring 2021	-79.15	-60.53	-25.43	-45.97	-59.82
$PM_{2.5}$	Spring 2021	-60.94	-44.84	-37.50	-36.29	-42.47
AOD	3.15	-81.92	-49.54	-38.97	-46.41	-48.45
	3.27	-75.10	-46.12	-36.39	-41.84	-44.52
	4.18	-55.88	-16.49	-3.20	-7.83	-14.52
	Mean AOD	-70.97	-37.38	-26.19	-32.03	-35.83

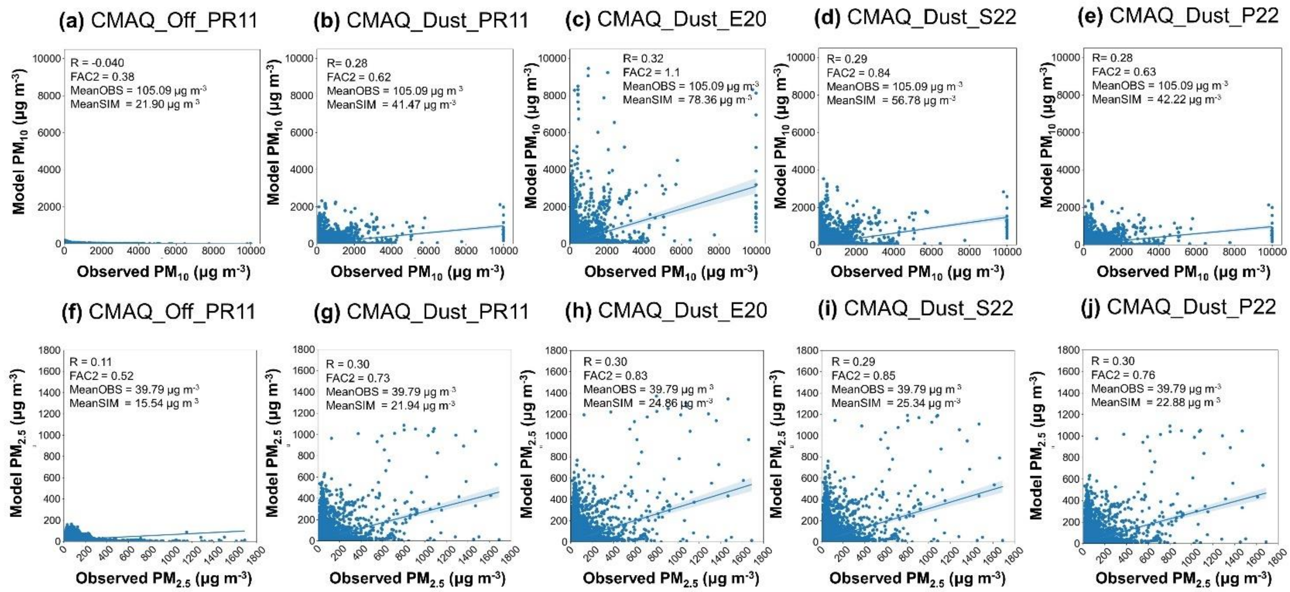

Figure 5: The scatter plot of the observed against modeled PM_{10} (a-e) and $PM_{2.5}$ (f-j) for CMAQ_Off_PR11 (a, f), CMAQ_Dust_PR11 (b, g), CMAQ_Dust_E20 (c, h), CMAQ_Dust_S22 (d, i) and CMAQ_Dust_P22 (e, j), at the 100 sites of the mainland China on 12 March-20 April 2021 (http://www.aqistudy.cn/). R is the correlation coefficient between the observation and model; FAC2 is the factor of two; MeanOBS and MeanSIM are the mean of PM from observation and model, respectively.

General comment 2: Additionally, results presented in various figures seem inconsistent. For instance, Table 4 suggests a clear difference between the PR11 and P22 schemes, but Figure 5 shows minimal differences between the two (see panels d & h).

Response: We thank the reviewer for the comments. For the reviewer's understanding, we have referred to the modeling evaluation result between STAGE and M3DRY under CMAQv5.4 conducted by USEPA (2024). According to the report, the difference in modeled particulate matter between PR11 and P22 dry deposition was within 5 %, and the time series trend was similar. To justify the present model result of PR11 and P22, we purposely replot the corresponding PM10 and PM25 for a clear comparison (Fig. S6). Moreover, the STAGE by E20 simulated a higher $PM_{2.5}$ compared to STAGE P22 (USEPA, 2024). This shows that the model testing results over the CONUS were consistent with the East Asia domain demonstrated in the present study.

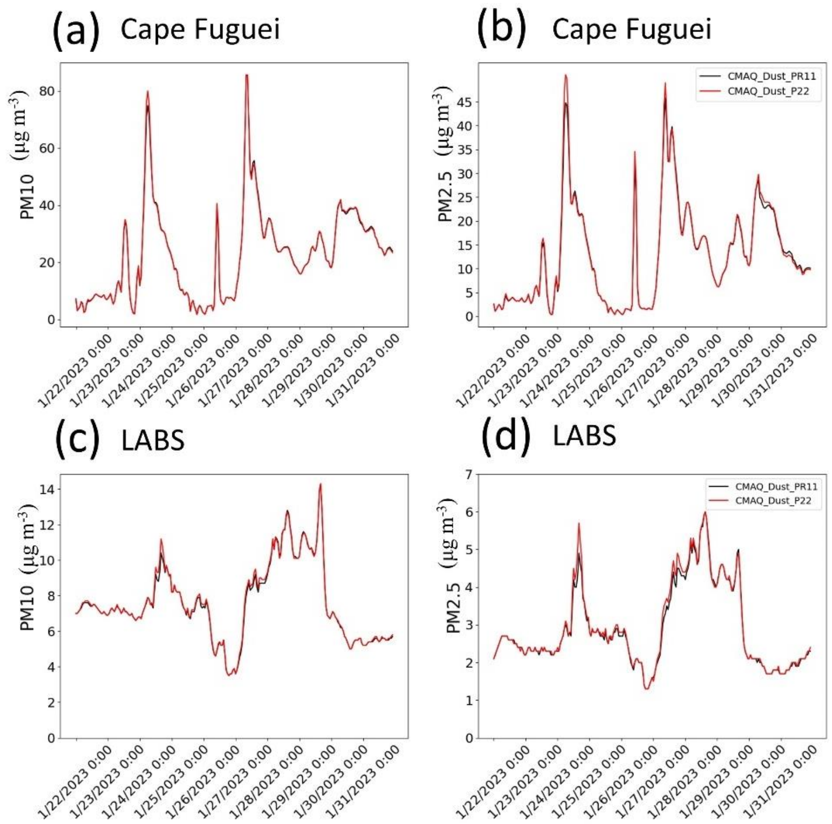

Figure S6: Time series of PM_{10} (left panel) and $PM_{2.5}$ (right panel) concentrations during 22-31 January 2023 under CMAQ_Dust_PR11 and CMAQ_Dust_P22 simulations over the Cape Fuguei (upper panel) and LABS (lower panel), representing the surface and high altitude, respectively.

Reference:
U.S. Environmental Protection Agency: https://github.com/USEPA/CMAQ/wiki/CMAQ-Release-Notes:-Dry-Deposition-Air-Surface-Exchange:-Surface-Tiled-Aerosol-and-Gaseous-Exchange-(STAGE), last access 15 October 2024.

General comment 3: Moreover, the connection between the first part of the paper, which compares four schemes, and the second part, which focuses on dust and black carbon transport, is unclear. This makes the paper feel disjointed. The same applies to the mention of the three dust storms from 2021; it's unclear how they tie into the paper's objectives.

Response: The connection between the first and second part of the paper has been included in the introduction section. We added the paragraphs as "The transboundary pollutants mechanisms have been widely discussed through LABS measurements, cooperating with the backward trajectory, reanalysis dataset, and modeling approach. Previous research reveals that LABS pollutants could be associated with severe fire emissions from northern Peninsular Southeast Asia (Huang et al., 2020; Ooi et al., 2021) and Indonesia (Ravindra Babu et al., 2023). Moreover, the intense wind speed in northwest China could transport the mineral dust through the surface and high-altitude layer detected at LABS (Kong et al., 2021; Kong et al., 2022). Additionally, the transport process of East Asian haze due to the cold surge from the Asian Continental industrial region towards Taiwan has been widely discussed (Chuang et al., 2020). Instead of pure aerosol, the coexistence of dust and biomass burning over Taiwan, a condition discovered in previous research, has significant implications for the regional climate (Dong et al., 2018; Dong et al., 2019). However, the high-altitude synoptic pattern associated with the coexistence between natural dust and anthropogenic pollutants remains unknown due to lack of observations at the upper layers.

This study used the chemical transport model to investigate the long-range transport of East Asian dust (EAD) that occurred on 22-31 January 2023 and 12 March-20 April 2021. Due to the limitation of the dust model, the CMAQ version 5.4, embedded with four types of dry deposition schemes, was implemented to justify the effectiveness of improving our latest refined dust model (Kong et al., 2024). LABS detected the recent transboundary episode in January 2023 as a mixing aerosol type (see Section 3.1), which has not been widely discussed, and the multiple dust storm episodes mentioned by Kong et al. (2024) provide an opportunity to model the EAD over the downwind region. Recognizing the significant transboundary events detected through Taiwan's observations, the improvement of the CMAQ dust model by the dry deposition schemes, and its application in characterizing the transport mechanism can be vital. The paper is organized as follows. The model setup and ancillary datasets are discussed in Sect. 2. The results and discussion are presented in Sect. 3, followed by the conclusions in Sect. 4." **Page 11, Line 259-284**.

General comment 4: Overall, the manuscript appears rushed. The figures are not properly discussed, and the text contains several grammatical and stylistic errors. It would significantly improve the paper if the authors dig deeper into the mechanistic differences between the schemes and how these differences impact the simulations.

Response: We thank the reviewer for the comments and suggestions. The main objectives of the research are to improve the dust model performance by using the latest deposition schemes from CMAQv5.4. This research has the potential to significantly advance our understanding of atmospheric processes. Also, the variation of the aerosol particle mode caused by the difference in dry depositions schemes was the main concern. The mechanics relating to V_d and aerosol profile has been revised in Section 3.3 as "We estimated the CMAQ averaged particle modes for the PR11 dry deposition scheme and the corresponding percentage changes using E20, S22, and P22 (Fig. 8). By using E20 and S22, we found that the V_d corresponding to the Aitken and accumulation modes has been increased by >100 % over most of the CMAQ domain, which was most obvious over Asian continent (Fig 8b, c, f, g). Meanwhile, the variation of V_d

distribution was insignificant for P22 (Fig. 8d, h, i). For the coarse mode particles, the V_d has been tremendously reduced for E20 and S22 compared to PR11. However, for S22, the V_d has increased by >100 % over northwest China, which is the dust source region (Fig. 8k). This leads to a significant deposition over the desert before transporting it to the downwind region, causing less PM_{10} simulated by S22 than E20. A previous study proposed the V_d for the aerosol at the water surface was associated with the CTM uncertainly at the downwind region (Kong et al., 2021, 2024; Ryu and Min, 2022). The V_d of Aitken and accumulation modes at land and water surfaces increased generally, except E20 at the water surface. Interestingly, the coarse mode V_d at the water surface for E20 and S22 decreased significantly by -44.65 % and -21.44 %, respectively, suggesting that both deposition schemes, particularly E20, could resolve the excessive deposition over the marine boundary layer (Table 5). Such minimal deposition velocity distributing over a large part of the western Pacific Ocean, including the Sea of Japan, Yellow Sea, East China Sea, and South China Sea, might be responsible for reducing the modeled PM_{10} underestimation over Taiwan (Fig.8j, k), as mentioned by Kong et al. (2021)." **Page 13-14, Line 328-344**.

Also, we have revised Table 5 and Figure 8 as below:

Table 5. Average deposition velocity and the percentage change by PR11 corresponding to E20, S22, and P22, for Aitken, Accumulation, and Coarse modes over land and ocean boundary layer, respectively.

Dry deposition schemes	Aitken		Accumulation		Coarse	
	Land	Ocean	Land	Ocean	Land	Ocean
PR11 (cm s^{-1})	0.080	0.062	0.061	0.042	0.264	0.109
E20 (cm s^{-1})	0.090	0.074	0.065	0.040	0.139	0.060
S22 (cm s^{-1})	0.219	0.117	0.120	0.064	0.078	0.085
P22 (cm s^{-1})	0.085	0.062	0.072	0.043	0.290	0.116
ΔE20 (%)	12.66	20.06	5.43	-5.19	-47.10	-44.65
ΔS22 (%)	173.74	89.45	96.52	52.35	-70.29	-21.44
ΔP22 (%)	6.10	1.37	17.66	1.52	10.06	6.86

Note: Δ representing the percentage change by PR11 as relative to E20, S22 and P22.

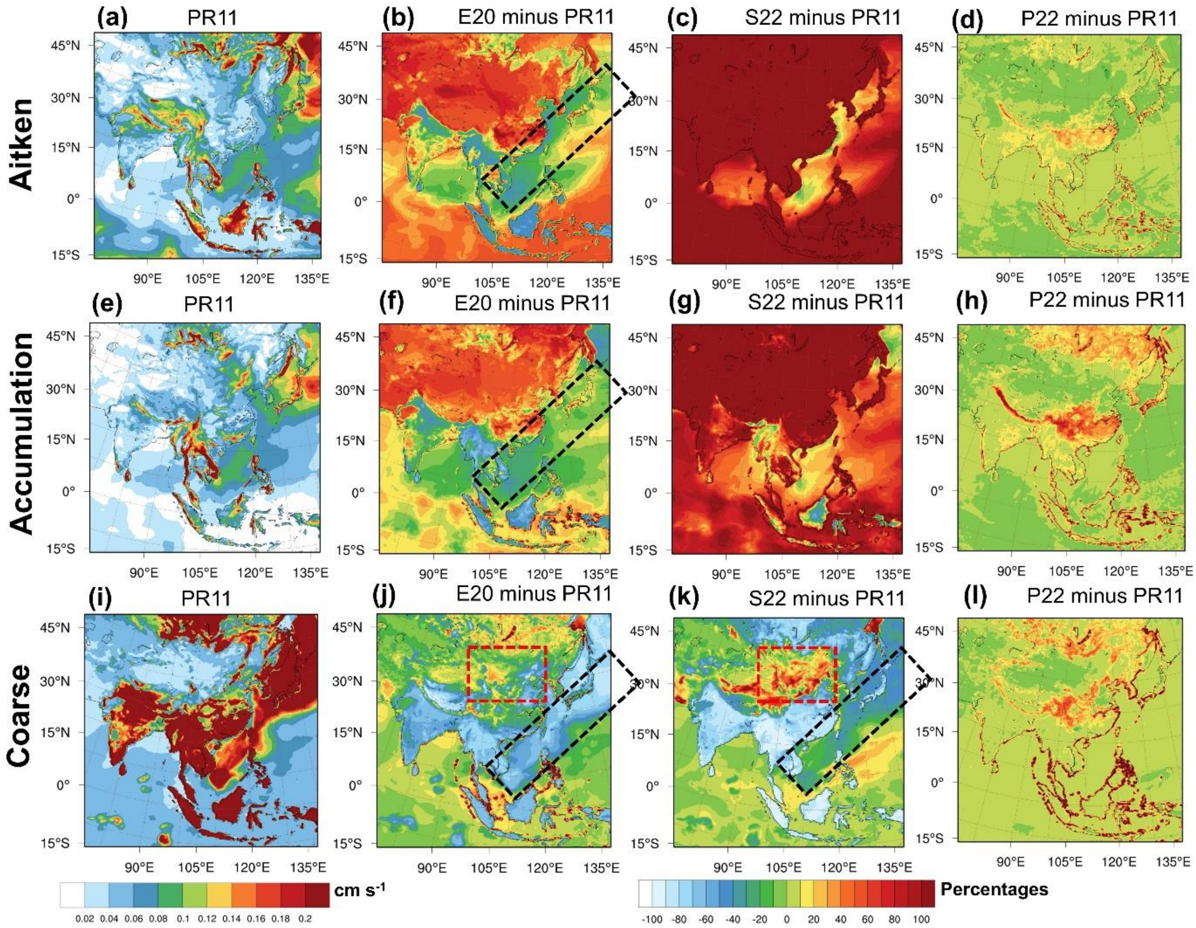

Figure 8: CMAQ estimated 10 days (22-31 January 2023) averaged for the (a-d) Aitken, (e-h) accumulation, and (i-l) coarse particle modes for PR11 dry deposition scheme (a, e, i) and the corresponding concentration percentage changes (%) using (b, f, j) E20, (c, g, k) S22 and (d, h, l) P22 schemes. Red-dash rectangular indicates the region across northwest China; Black-dash rectangular indicates the marine boundary layer.

<u>RC3: Specific comments and responses</u>

Comment 1: What's the significance of including Equations 1-4? They are all pretty standard and known to the community, and also were not referred to later in the text.

Response: We thank the reviewer for the question. Equations 1-4 represent the model algorithms used to calculate the dust emission and dry deposition. Even though the equations are not referred to the later text, the physical formulations shall be mentioned in the very beginning to introduce the calculations, which we do not treat the model a black box.

Comment 2: Figure 3 is barely discussed in the text.

Response: Figure 3 is the observation data retrieved from LABS, and has been discussed in the manuscript. We have included as "Two interesting high pollution events at Mt. Lulin (2,862 m above sea level) during 24-26 Jan and 27-30 January, respectively, are shown in Fig. 3. The latter event was more intense compared to the earlier one, where the maximum PM_{10} concentration can reach up to 35 μg m^{-3}. Moreover, it was observed that the black carbon concentrations could reach up to a maximum of 400 ng m^{-3}. Based on the *in-situ* measurement, it was interesting to find the mixing state between dust, black carbon, and brown carbon (Fig.

3c). Different from what has been discussed by Kong et al. (2022), the long-range transport air pollution at the high-altitude not just merely EAD, but also included the anthropogenic pollutant from mainland China." **Page 9, Line 215-222.**

Comment 3: Lines 380-381: The meaning here is unclear.

Response: The statement has been revised. We modified the sentence as **"**Additionally, the simulation of the multiple dust episodes in spring 2021 were re-constructed to evaluate the CMAQ performance over the Asian Continental. The E20 dry deposition scheme outperformed the other schemes with the lowest NMB value in simulating PM_{10} (-25.4 %) and AOD (-26.2%). For the modeled $PM_{2.5}$, S22 performed slightly better than E20, with NMB of -36.29 % and -37.5 %, respectively.**" Page 17, Line 423-427**.

Comment 4: Lines 369-371 and 390-391: Highly subjective statements.

Response: The sentences have been removed.

Comment 5: The first part of the conclusion section repeats previously discussed results.

Response: We thank the reviewer for the comment. The main modeling results were repeated in conclusion to summarize the whole result and discussion.