Reply to the comments of RC1

RC1: General comment

This paper describes a modeling study where four aerosol dry deposition schemes were used to model dust and BC. The Main point seems to be that one of the deposition schemes is best and improves the modeling of dust. This conclusion, however, is not well supported by the marginal improvement of some statistics relative to two measurements sites and MODIS AOD for brief periods during dust episodes. The problem is that the observed data is very meager and concluding that one scheme is best assumes that all other aspects of the modeling system are perfect, especially the dust emissions. Also, this a very specific application for short periods of time so there is no reason to think that these conclusions are generally relevant. Another problem with this study is that the relative performance of the four deposition schemes is not consistent with other modeling studies. In particular, the P22 scheme typically results in significantly greater deposition velocities than the E20 scheme. Also, Fig 5 shows that P22 is almost the same as PR11. This suggests that there were errors made in running these models. Table 2 says that CMAQv5.4 was used. If the STAGE option was used for dry deposition (which should be noted in the Table) a choice of S22, E20, and P22 are available. However, PR11 is not. How was this used?. Table 2 also states that the NOAH LSM was used in WRF. CMAQ needs several parameters from WRF that typically are output when using the PX LSM. When NOAH is used, default calculations for these parameters, which are important for deposition, are made in the Meteorology-Chemistry Interface Processor (MCIP). These calculations are not the same as in the LSM and will result in additional errors. There are many grammatical and other sloppy errors in the text. Far too many for me to correct.

Response: We greatly appreciate the referee#1 for the suggestions to improve the manuscript. All of the changes in the revised manuscript have been highlighted in yellow. Corrections (blue text) with line numbers indicated in this response document refer to the revised manuscript.

Our point-by-point responses to the reviewer's comments are given below:

General comment 1: This paper describes a modeling study where four aerosol dry deposition schemes were used to model dust and BC. The Main point seems to be that one of the deposition schemes is best and improves the modeling of dust. This conclusion, however, is not well supported by the marginal improvement of some statistics relative to two measurements sites and MODIS AOD for brief periods during dust episodes. The problem is that the observed data is very meager and concluding that one scheme is best assumes that all other aspects of the modeling system are perfect, especially the dust emissions. Also, this a very specific application for short periods of time so there is no reason to think that these conclusions are generally relevant.

Response: We thank the reviewer for the comments. For the reviewer's concern, the present study focuses on the difference of the dust model improvement using the dry deposition schemes embedded in CMAQv5.4, particularly over the western Pacific region, which used Taiwan as the primary receptor. The importance of using Taiwan's observation data for CMAQ model evaluation has been highlighted. We have mentioned as "The model performance in Taiwan is paramount in our study, as the area is equipped with a substantial number of well-maintained surface observation sites, providing comprehensive coverage. The LABS station in the high-altitude subtropical western North Pacific region serves as the sole background station for monitoring transboundary pollutants. This station is crucial in our research as it provides unique data on the long-range transport of pollutants, further underscoring the relevance of our study." **Page 3-4, Line 79-83**.

We also agree that dust emissions at the Asian continental level should be considered. We have included the model evaluation over most parts of mainland China, considering the dust source and nearby source region. 100 observation sites have been obtained from 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, as well as 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 12 March-20 April 2021, obtained from 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 correlation ($R > 0.3$) 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

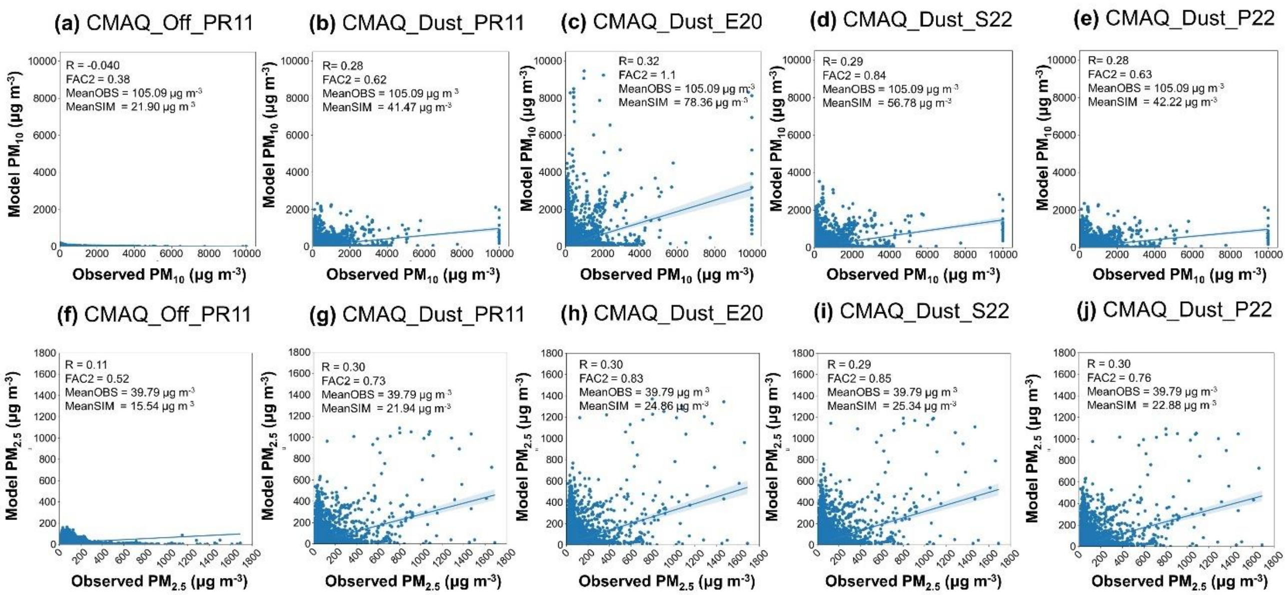

Figure 5: The scatter plot of the observed against modeled PM_{10} (a-e) and $PM_{2.5}$ (f-j) for CMAQ_Off_PR11 (a, f), CMAQ_Dust_PR11 (b, g), CMAQ_Dust_E20 (c, h), CMAQ_Dust_S22 (d, i) and CMAQ_Dust_P22 (e, j), at the 100 sites of the mainland China on 12 March-20 April 2021 (http://www.aqistudy.cn/). 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.

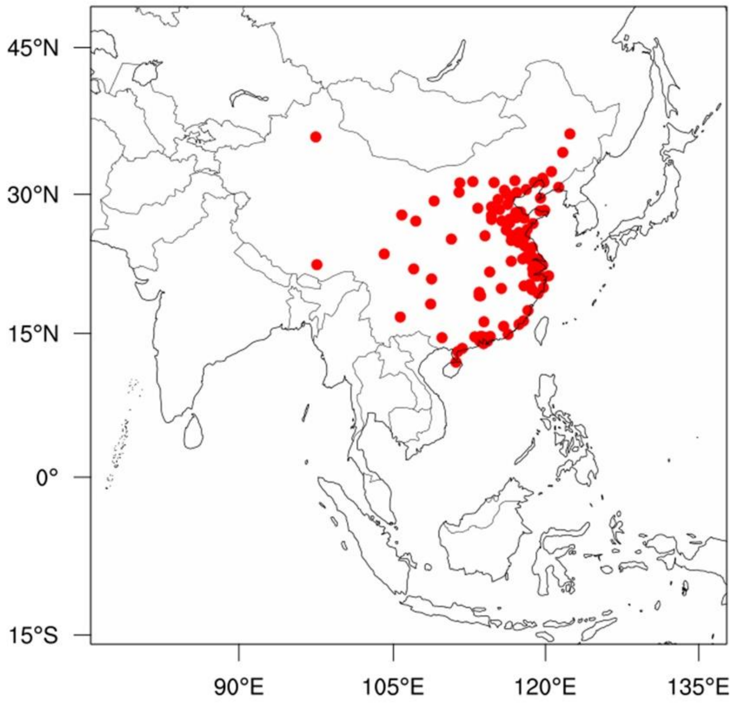

Figure S1: The location of monitoring sites over mainland China used for model evaluation (http://www.aqistudy.cn/).

General comment 2: Another problem with this study is that the relative performance of the four deposition schemes is not consistent with other modeling studies. In particular, the P22 scheme typically results in significantly greater deposition velocities than the E20 scheme. Also, Fig 5 shows that P22 is almost the same as PR11. This suggests that there were errors made in running these models.

Response:

We agree with the reviewer regarding the P22 and E20. The greater deposition velocity is by P22 than E20, particularly in the coarse mode. The STAGE and M3DRY under CMAQv5.4 have been examined over Continental U.S.A. (CONUS) during July 2016 (USEPA, 2024). 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. S5). 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.

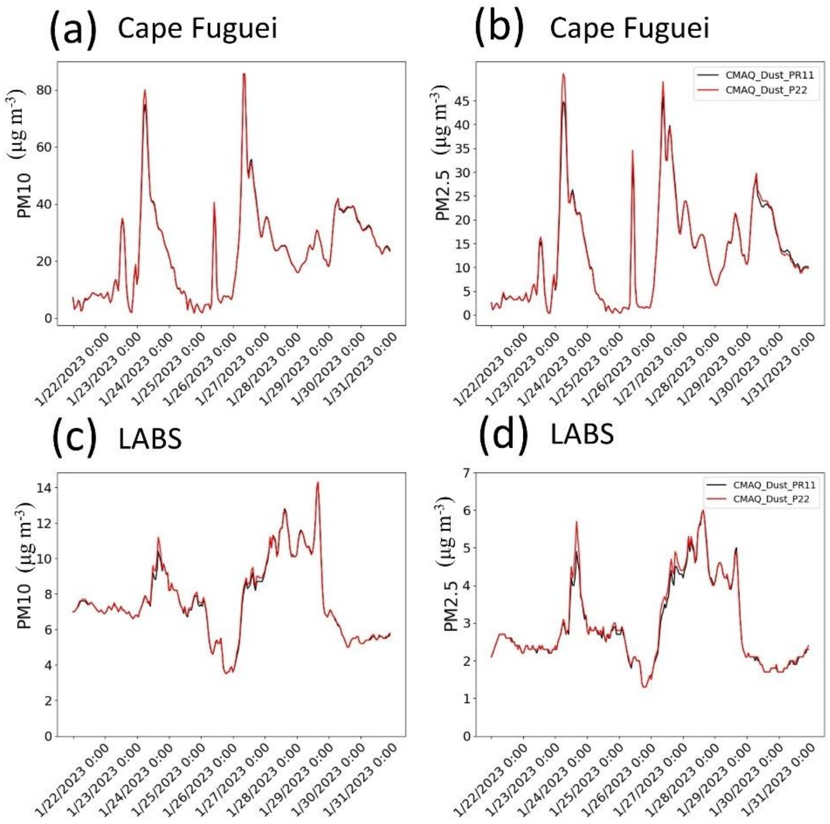

Figure S6: Time series of PM_{10} (left panel) and $PM_{2.5}$ (right panel) concentrations during 22-31 January 2023 under CMAQ_Dust_PR11 and CMAQ_Dust_P22 simulations over the Cape Fuguei (upper panel) and LABS (lower panel), representing the surface and high altitude, respectively.

Also, we have updated the STAGE dry deposition description as "Moreover, Surface Tiled Aerosol and Gaseous Exchange (STAGE) deposition has been implemented within the CMAQv5.3, where estimated fluxes from sub-grid cell fractional land-use values, aggregates the fluxes to the model grid cell and unifies the bidirectional and unidirectional deposition schemes using the resistance framework (Massad et al., 2010; Nemitz et al., 2001). The updated STAGE version in CMAQv5.4 could aggregate the grid-scale values that match the grid-scale values from most kinds of Land Surface Model of WRF (Hogrefe et al., 2023)." **Page 6, Line 146-152**.

References:

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.

Hogrefe, C., Bash, J. O., Pleim, J. E., Schwede, D. B., Gilliam, R. C., Foley, K. M., Appel, K. W., and Mathur, R.: An analysis of CMAQ gas-phase dry deposition over North America through grid-scale and land-use-specific diagnostics in the context of AQMEII4, Atmos. Chem. Phys., 23, 8119–8147, https://doi.org/10.5194/acp-23-8119-2023, 2023.

General comment 3: Table 2 says that CMAQv5.4 was used. If the STAGE option was used for dry deposition (which should be noted in the Table) a choice of S22, E20, and P22 are available. However, PR11 is not. How was this used?.

Response: We have included the dry deposition options in Table 1. Also, the corresponding changes have been used to modify the scenario description in Table 2. We change the tables as below:

Table 1. Model settings.

Model setting	Descriptions
Period	12 March-20 April 2021 and 22-31 January 2023
Domain	d01, d02, and d03 with 45 KM, 15 KM, and 5 KM of the resolutions, respectively
Boundary condition	NCEP FNL lateral boundary condition
Surface and land surface model	NOAH
Numerical weather model	WRF v40, including grid and observation nudging at d01.
Chemical transport model	CMAQ v5.4
Gas-phase chemistry and aerosol mechanism	CB06e51 + AE7
Emission Inventory	MICS-ASIA III emission in 2023, adjusted from the emission in 2017 (Zhang et al., 2018) based on the OMI-NO_x satellite (Huang et al., 2021).
Online dust treatment	The windblown dust treatment suggested by Kong et al. (2024).
Dry deposition option	M3DRY (PR11) and STAGE (E20, S22 and P22).

Table 2. Simulation scenarios used in this present study.

Scenarios	Descriptions
CMAQ_Off_PR11	Without in-line dust calculation, with the M3DRY dry deposition algorithm by Pleim and Ran (2011).
CMAQ_Dust_PR11	Implement the latest refined dust treatment proposed by Kong et al. (2024), with the M3DRY dry deposition algorithm by Pleim and Ran (2011).
CMAQ_Dust_E20	Same as CMAQ_Dust_PR11, but with the STAGE dry deposition algorithm by Emerson et al. (2020).
CMAQ_Dust_S22	Same as CMAQ_Dust_PR11, but with the STAGE dry deposition algorithm by Shu et al. (2022).
CMAQ_Dust_P22	Same as CMAQ_Dust_PR11, but with the STAGE dry deposition algorithm by Pleim et al. (2022).

General comment 4: Table 2 also states that the NOAH LSM was used in WRF. CMAQ needs several parameters from WRF that typically are output when using the PX LSM. When NOAH is used, default calculations for these parameters, which are important for deposition, are made in the Meteorology-Chemistry Interface Processor (MCIP). These calculations are not the same as in the LSM and will result in additional errors.

Response: We greatly appreciate the reviewer's comments. The reason for using PXLSM is to get the look-up table directly from WRF PXLSM, which would be readable by the MCIP calculation needed for the deposition algorithm. However, under the STAGE option, the mapping by source code in ASX_DATA_MOD.F could be set up to simulate most kinds of LSM, including NOAH and CLM. With NOAH, the parameterizations, including Z0, VEG, and LAI, were directly taken from the WRF look-up table. In addition, the NOAH-LSM contains additional LU importance in simulating dust deserts such as playa and white sand, which is not included in PXLSM. So, applying NOAH-LSM over East Asia, which experienced frequent dust episodes, could be more representative. However, we do realize that due to the difference in LU categories and soil type of both PXLSM and NOAH, the impact on aerosol (PM10/PM25) emission and deposition can vary, and we shall propose this idea as our future model testing.

General comment 5: There are many grammatical and other sloppy errors in the text. Far too many for me to correct.

Response: We thank the reviewer for the suggestion. The grammar of the manuscript has been proofread by Grammarly (version Premium) online text editor.

RC1: Specific comments and responses

Comment 1: Table 1 has many errors. For example, the equations for PR11 are all for gasses not aerosols. Please remove Table 1.

Response: We thank the reviewer for the suggestion. Table 1 has been removed.

Comment 2: Line 17. P20 should be P22.

Response: We have revised the term P20 to P22. **Page 1, Line 20**.

Comment 3: L19. This sentence implies that dry deposition directly affects dust emissions.

Response: The sentence has been revised. We corrected the sentence as "The result showed that the dry deposition parameterization could significantly improve the CMAQ in simulating PM_{10} and $PM_{2.5}$ concentrations." **Page 1, Line 17-18**.

Comment 4: L30. Sentence does not make sense.

Response: The sentence emphasized the LABS measurement located at 2862 m.s.l. showed the mixing of dust and black carbon from 22-31 January 2023. We corrected the sentence as "On 22-31 January 2023, the *in-situ* measurement of the upper level observed the coexistence of natural dust and anthropogenic aerosol." **Page 1, Line 29-30**.

Comment 5: L32-33. This sentence "resolving the uncertainty of the CMAQ dust emission treatment" is a gross overstatement.

Response: Agree. We have revised the sentence as "We proposed implementing the E20 dry deposition approach, narrowing the uncertainty of the CMAQ dust emission treatment." **Page 1, Line 31-33**.

Comment 6: L38. Does not make sense.

Response: The sentence has been revised. We modified the sentence as "Among these, particle dry deposition is a crucial aerosol removal process and an important sink for particles in the model." **Page 1, Line 36-38**.

Comment 7: L51. Again, a gross overstatement: "Emerson et al. (2020) has resolved the problem."

Response: Agree. We have revised the sentence as "The latest dry deposition scheme revision by Emerson et al. (2020) has reduced the uncertainty, marking a significant step forward in our quest for more accurate air quality modeling." **Page 2, Line 50-52**.

Comment 8: L64. What "boarder"?

Response: We have revised the sentence as "The surface fine particle concentrations can vary up to 5-15 %, and the particle dry deposition has more than 200 % discrepancy due to the different dry deposition schemes. (Saylor et al., 2019)." **Page 3, Line 62-64**.

Comment 9: L70-72. Does not make sense.

Response: The statement mentioned the simulated PM_{10} underestimation caused by the uncertainty of the deposition mechanism. We changed the sentence as "Besides the model bias on $PM_{2.5}$, the simulation of PM_{10} has been underestimated due to the uncertainty of the deposition mechanism, particularly over the western Pacific." **Page 3, Line 69-70**.

Comment 10: L92-93. Fix notation.

Response: The notation has been fixed. We modified the notation as "

$$u_{*,t} = u_{*,to} f_m f_r \qquad (1)$$

Where $u_{*,to}$ is the ideal threshold friction velocity, while f_m and f_r are the correction factors of soil moisture and surface roughness, respectively." **Page 5, Line 113-115**.

Comment 11: L100. Bulb?

Response: The word has been corrected as "…and bulk soil density…". **Page 5, Line 122.**

Comment 12: L115-118. This is very sloppy. Please fix Vs and Vg

Response: The physical formulation and notation have been corrected as below: "

$$V_d = V_s + \frac{1}{R_a + R_s} \qquad (3)$$

where V_s is the gravitation settling velocity, R_a is the resistivity aerodynamic and R_s is the surface resistivity. The V_s is calculated according to Stokes's Law as:

$$V_s = \frac{p_p D_p^2 g C_c}{18\eta} \qquad (4)$$

where, ρ_p is the density of the particle; D_p is the diameter of the particle; g is gravitational acceleration; C_c is the Cunningham correction factor for small particles; and, η is the dynamic viscosity of air." **Page 6, Line 137-142**.

Comment 13: L122-123. This is not true: "Dry deposition is based on gravitational settling velocity (Vg), which is the function of aerodynamic and surface resistance."

Response: Agree. The statement has been removed.

Comment 14: L126. Should note that STAGE is one of two options, the other being M3Dry.

Response: Agree. The methodology has been revised as "CMAQ is embedded with M3Dry dry deposition calculation that implements the scheme of Pleim and Ran (2011), which is based on Slinn (1982). As noted by Pleim and Ran (2011), chemical surface flux modeling has become an essential process in the air quality model. For instance, the linkages of ambient concentration levels to the deposition of SO_x and NO_x. Moreover, Surface Tiled Aerosol and Gaseous Exchange (STAGE) deposition has been implemented within the CMAQv5.3, where estimated fluxes from sub-grid cell fractional land-use values, aggregate the fluxes to the model grid cell and unifies the bidirectional and unidirectional deposition schemes using the resistance framework (Massad et al., 2010; Nemitz et al., 2001). The updated STAGE version in CMAQv5.4 could aggregate the grid-scale values that match the grid-scale values from most kind of Land Surface Model of WRF (Hogrefe et al., 2023)." **Page 6, Line 143-152**.

Comment 15: L137. Here, and many other places, abbreviations such as PSEA are used without defining. Also, SDS, WPO.

Response: The abbreviations have been defined as "…peninsular Southeast Asia (PSEA)…". **Page 7, Line 159**.

The abbreviation SDS is removed and replaced with the full defining terms. We modified it to "Super Dust Storm." **Page 11, Line 289**.

The abbreviation WPO is removed and replaced with the full defining terms. We modified it to "western Pacific Ocean." **Page 9, Line 210-211**.

Comment 16: L139. What are the chemical LBCs?

Response: The chemical LBC was generated by a time-invariant set of predefined, vertical concentration profiles. For nested simulations, the dynamic boundary conditions are extracted from CCTM output from a coarse-grid simulation (USEPA, 2010).

Reference:

U.S. Environmental Protection Agency: Operational Guidance for the Community Multiscale Air Quality (CMAQ) Modeling System Version 4.7.1, https://www.cmascenter.org/cmaq/documentation/4.7.1/Operational_Guidance_Document.pdf, 2010.

Comment 17: L156. CMAQ_Dust_PR11 is repeated.

Response: We thank the reviewer for the comment. The additional "CMAQ_Dust_PR11" is removed.

Comment 18: L177. Don't see dust claw.

Response: The statement has been revised. We modified the sentence as "The satellite image showed dust induced by a high-pressure system on 24-25 January (Fig. 2a3, 2a4). The next day, the same region was covered by a thick cloud, and dust was again widely distributed from 27-30 January 2023." **Page 8, Line 202-204**.

Comment 19: L199. Table 3 should be 4.

Response: Table 3 remains. **Page 9, Line 224**.

Comment 20: L209. Numbers are reversed.

Response: We thank the reviewer for the comment. The numbers in the text are correct as Table 3 has been modified between the columns of Dust_S22 and Dust_P22. We have corrected the table as below:

Table 3. Statistical evaluation for PM_{10} and $PM_{2.5}$ concentrations during 22-31 January 2023 for Cape Fuguei under the multiple simulation scenarios.

	Benchmark	CMAQ-M3DRY		CMAQ-STAGE		
		Off PR11	Dust_PR11	Dust_E20	Dust_S22	Dust_P22
PM_{10}						
MeanObs		49.97	49.97	49.97	49.97	49.97
MeanMod		21.19	22.97	29.04	26.48	23.04
NMSE		0.82	0.71	0.49	0.56	0.71
NMB	± 85%	-57.59	-54.05	-41.90	-47.01	-53.90
Corr	> 0.35	0.41	0.44	0.52	0.46	0.42
NMBF		-1.36	-1.18	-0.72	-0.89	-1.17
$PM_{2.5}$						
MeanObs		15.52	15.52	15.52	15.52	15.52
MeanMod		12.48	12.95	13.86	14.15	13.16
NMSE		0.31	0.29	0.29	0.30	0.31
NMB	± 85%	-19.55	-16.53	-10.65	-8.84	-15.22
Corr	> 0.35	0.52	0.55	0.53	0.53	0.52
NMBF		-0.24	-0.20	-0.12	-0.10	-0.18

Comment 21: Fig 4. Hard to tell the different model runs apart especially for the PM25. It would help to expand the scale on the PM25 plots.

Response: The scale on the PM2.5 plots in Fig.4 has been expanded. We modified the figure as:

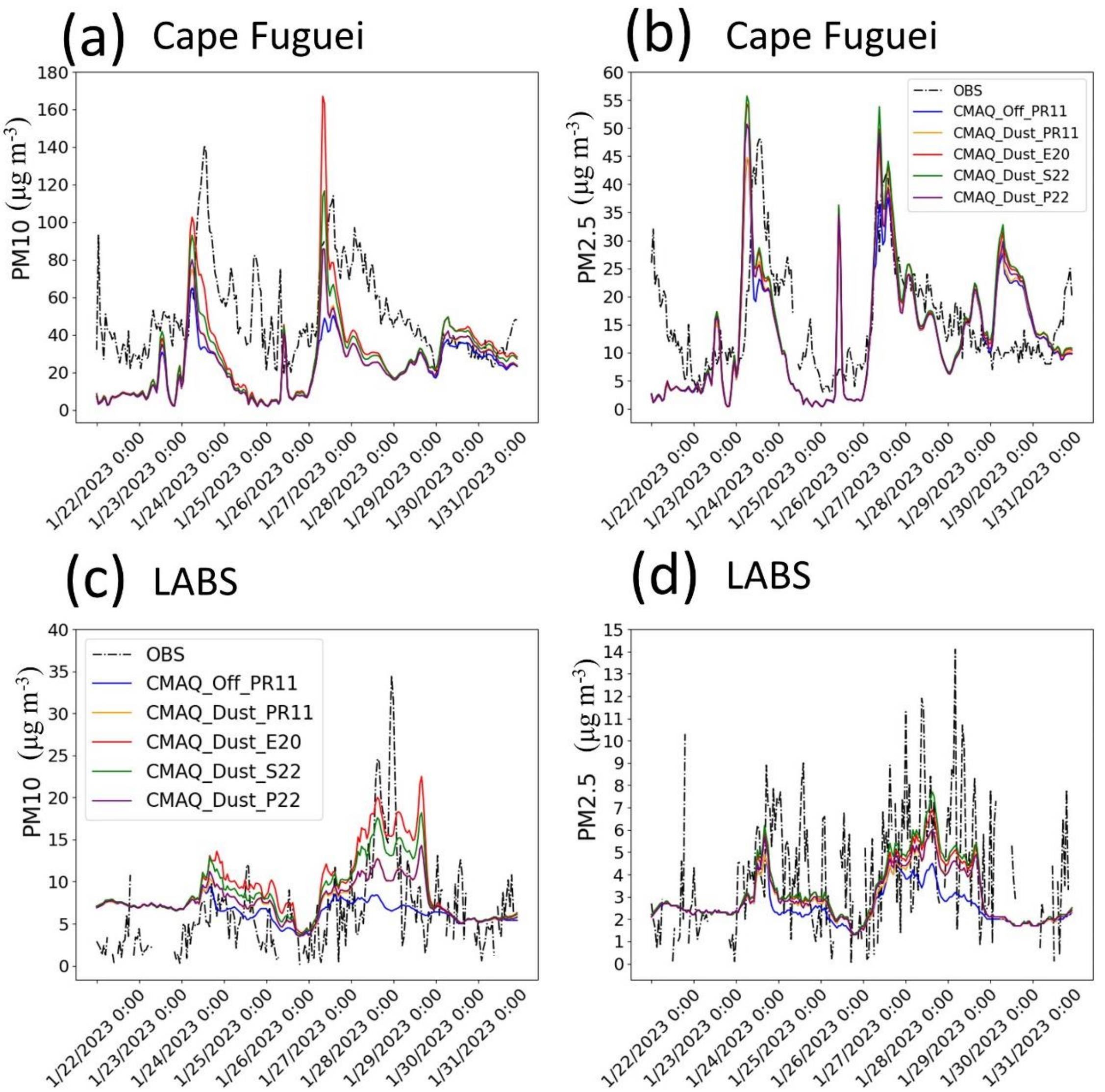

Figure 4: Time series of PM_{10} (left panel) and $PM_{2.5}$ (right panel) concentrations during 22-31 January 2023 under multiple deposition schemes over the Cape Fuguei (upper panel) and LABS (lower panel), representing the surface and high altitude, respectively.

Comment 22: L224. Where is "the north peninsula of Southeast Asia"?

Response: The term has been revised. We have modified the term as "northern PSEA". **Page 10, Line 250.**

Comment 23: L251. There is something wrong here. P22 and PR11 should be very different.

Response: Please refer to the response of **General Comment 2**.

Comment 24: L275-276. These results seem contrary to other modeling studies where P22 generally has greater much Vd for Accumulation mode than E20. Maybe put these numbers in a table.

Response: Agree. The statement in the text was referring to the median of deposition velocity, which indicates that S22 was the highest median value among all. For the deposition velocity at the 75th percentile, 75 % of the V_d for P22 was higher than E20. We have modified the sentence as "As shown in the figure, the median of E20, S22, and P22 increased the deposition velocity of the Aitken (accumulation) modes particle as compared to PR11 by 22.56 (11.32) %, 117.76 (86.43) % and 2.5 (7.52) % respectively." **Page 13, Line 318-320**.

Also, the V_d values of the four dry deposition schemes has been included in supplementary. We added the Table S1 as below:

Table S1: V_d percentiles (cm s^{-1}) of atiken, accumulation and coarse particle modes by the four dry deposition schemes.

Dry deposition schemes	Percentiles	Aitken	Accumulation	Coarse
PR11	25th	0.033	0.020	0.046
	50th	0.056	0.036	0.059
	75th	0.086	0.066	0.186
E20	25th	0.043	0.025	0.046
	50th	0.068	0.040	0.053
	75th	0.102	0.066	0.092
S22	25th	0.076	0.041	0.043
	50th	0.122	0.068	0.052
	75th	0.201	0.114	0.077
P22	25th	0.034	0.021	0.048
	50th	0.057	0.039	0.067
	75th	0.089	0.073	0.208

Comment 25: L286 (Fig.7). Why not show dry deposition velocity for each model rather that difference from PR11?

Response: We thank the reviewer for the question. The primary goal of our study is to enhance the performance of the latest dust model by implementing STAGE deposition schemes in contrast to the M3DRY scheme. Simply showing the difference from PR11 is most significant as it provides a clear overview of how the model has been enhanced, thereby advancing our understanding of dust modeling behavior. The dry deposition velocity for each model is also displayed at the supplementary document. We added Fig. S4 as:

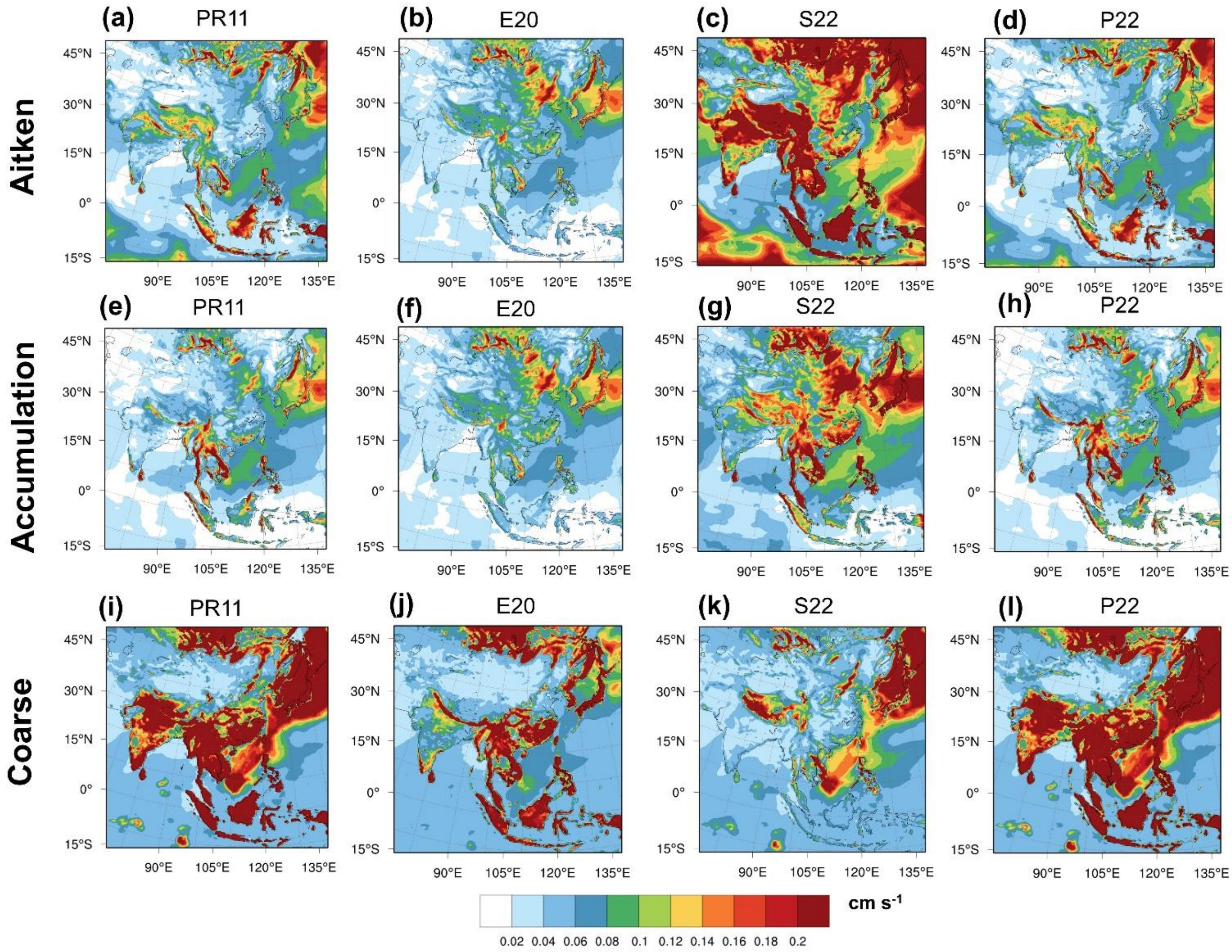

Figure S4: CMAQ estimated 10 days (22-31 January 2023) averaged for the (a-d) Aitken, (e-h) accumulation, and (i-l) coarse particle modes for (a, e, i) PR11, (b, f, j) E20, (c, g, k) S22 and (d, h, l) P22 dry deposition schemes

Comment 26: L300. Elemental

Response: The term has been revised. We modified the sentence as "Black carbon, often known as elemental carbon, released from the biofuels, fossil fuels and biomass burning, has been proven to impact the radiative budget and regional climate". **Page 14, Line 346-347**.

Comment 27: L330. Trans what boundary?

Response: The term has been modified as **"…** the long-range transport of modeled black carbon …"**. Page 15, Line 376**.

Comment 28: L350-351. The modeled BC in Fig 11h seems to end at Taiwan. Also, there is no Fig 11i.

Response: Fig. 11h is replaced by Fig. 12d; Fig. 11i has been removed and changed as Fig. 12e. We fixed the figure number as **"**As shown in Fig. 12d, the modeled black carbon was found distributed at the western Pacific Ocean. In Fig. 12e, a clear black carbon dome was distributed along 700 hPa, showing a similar pattern as dust." **Page 15-16, Line 396-399**.

Comment 29: L353. This phrase makes no sense: "as the coarse particles could comprise of fine particles".

Response: The phrase has been removed. We change the sentence as "This simulation proposes the consistency of the "double dome" mechanism of Asian dust and biomass burning episodes" **Page 16, Line 398-399**.

Comment 30: L373. "vastly" is again an overstatement.

Response: The term has been removed. We change the sentence as "…surface PM_{10} has been improved by ..." **Page 16, Line 416**.

Reply to the comments of RC2

RC2: General comment

This study describes an improvement to the CMAQ dry deposition over Western Pacific using various dry deposition schemes. It also addressed the performance of a model-simulated long dust-black carbon belt along 15N. The study indicated improvements in the results, but it is unclear how the improvements are statistically relevant. The impact of other processes could affect the dry deposition scheme but was not addressed. A statistical rather than a visual comparison between CMAQ and satellite/assimilated AOD would be more convincing to demonstrate modeling performances. Also, the manuscript contains numerous grammatical and technical errors that require thorough proofreading before resubmission.

General Comment Response: The authors wish to thank the reviewer for the constructive comments on our work. The present research purposely investigates how the various types of dry deposition schemes embedded within CMAQv5.4 could help improve the latest refined dust model proposed by Kong et al. (2024). The other processes that could impact the dry deposition scheme efficiency, such as resistance, land surface, roughness length …, which is not our research scope. However, we thank the reviewer for pointing out the possible research question. We will consider the idea proposed to minimize the research gap. The manuscript has been proofread through Grammarly (version Premium) online text editor

All of the changes in the revised manuscript have been highlighted in yellow. Corrections (blue text) with line numbers indicated in this response document refer to the revised manuscript.

RC2: Specific comments and responses

Comment 1: The abstract should not include references.

Response: The references has been removed. We modified the sentence as "By utilizing the CMAQv5.4 with the refined dust emission treatment, the East Asian dust (EAD) simulation during January 2023 was constructed to evaluate the performance of four dry deposition parameterizations, namely PR11, E20, S22, and P22." **Page 1, Line 14-17**.

Comment 2: L 82. What is "LABS"?

Response: "LABS" refers to "Lulin Atmospheric Background Station." The abbreviation has been included on **Page 1, Lines 22**.

Comment 3: L 115. Where is Vs in the equation?

Response: V_s as one of the functions in the physical formulation of V_d. We corrected the formula as below: "

$$V_d = V_s + \frac{1}{R_a + R_s} \tag{3}$$

where V_s is the gravitation settling velocity, R_a is the resistivity aerodynamic and R_s is the surface resistivity. The V_s is calculated according to Stokes's Law as:

$$V_s = \frac{p_p\, D_p^2\, g C_c}{18\eta} \tag{4}$$

where, p_p is the density of the particle; D_p is the diameter of the particle; g is gravitational acceleration; C_c is the Cunningham correction factor for small particles; and, η is the dynamic viscosity of air." **Page 6, Line 137-142**.

Comment 4: L 168. The sentence is unclear. Clouds always induce biases in modeled and assimilated aerosols.

Response: The sentence has been revised. We modified the sentence as "The Modern Era Retrospective-analysis for Research and Application version 2 (MERRA-2) reanalysis data was used to demonstrate the spatiotemporal distribution of dust, compared with the air quality model, irrespective of the influence of clouds." **Page 8, Line 190-193.**

Comment 5: L 170. MERRA-2 is a data-assimilated system rather than a remotely sensed data.

Response: The sentence has been revised. We changed the sentence to "MERRA-2 (Gelaro et al., 2017) is a NASA reanalysis product utilizing Goddard Earth Observing System Data Assimilation System Version 5 (GEOS-5) and covering the data-assimilated system at a native spatial resolution of 0.5 ∘ × 0.625 ∘." **Page 8, Line 193-195.**

Comment 6: L 227. MERRA-2 is a data-assimilated product rather than a pure observational product. It's unclear how this sentence fits in with Figure 4.

Response: The sentence has been removed.

Comment 7: Fig S1. The link in the caption does not show these synoptic maps.

Response: The link in the caption has been revised. We corrected the caption as "Figure S2: Surface weather maps for the weather pattern obtained by Taiwan Central Weather Bureau (https://www.cwa.gov.tw/)." **Supplementary, Page 3, Line 33.**

Comment 8 Fig S2. A statistical comparison between collocated CMAQ and MODIS AOD with a scatterplot is needed to quantify their agreements.

Response: We thank the reviewer for the comment. MODIS AOD retrieved consisted of the missing value due to the cloud cover. Hence, the visualized qualitive comparison between CMAQ and MODIS can be more appropriate instead of a statistical comparison. However, we agree that using a scatter plot is needed for the evaluation quantification. A detailed model evaluation between CMAQ and the observed dataset over mainland China has been delivered to carry out the statistical comparison, which is more reliable in testing the model efficiency (Table 5). We added the discussion as "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 275-284.**

Comment 9: Fig S3. A statistical comparison is also needed by using the MERRA-2 AOD as well, not just the dust column. MERRA-2 provides AOD for each species.

Response: We thank the reviewer for the comment. Fig S5 aims to demonstrate the consistency of the transport pattern between dust and black carbon over the western Pacific Ocean, as shown by MERRA-2. Please see **Comment 8** for the detailed scatter plot analysis.

An explanation regarding the transport pattern consistency is included. We added the sentence as “Such consistency has been verified by the MERRA-2 dust and black carbon mass column over the region (red dash rectangular in Fig. S5).” **Page 15, Line 383-384.**

Also, Fig. S5 has been modified to emphasize the transboundary over the western Pacific Ocean. We change the figure as:

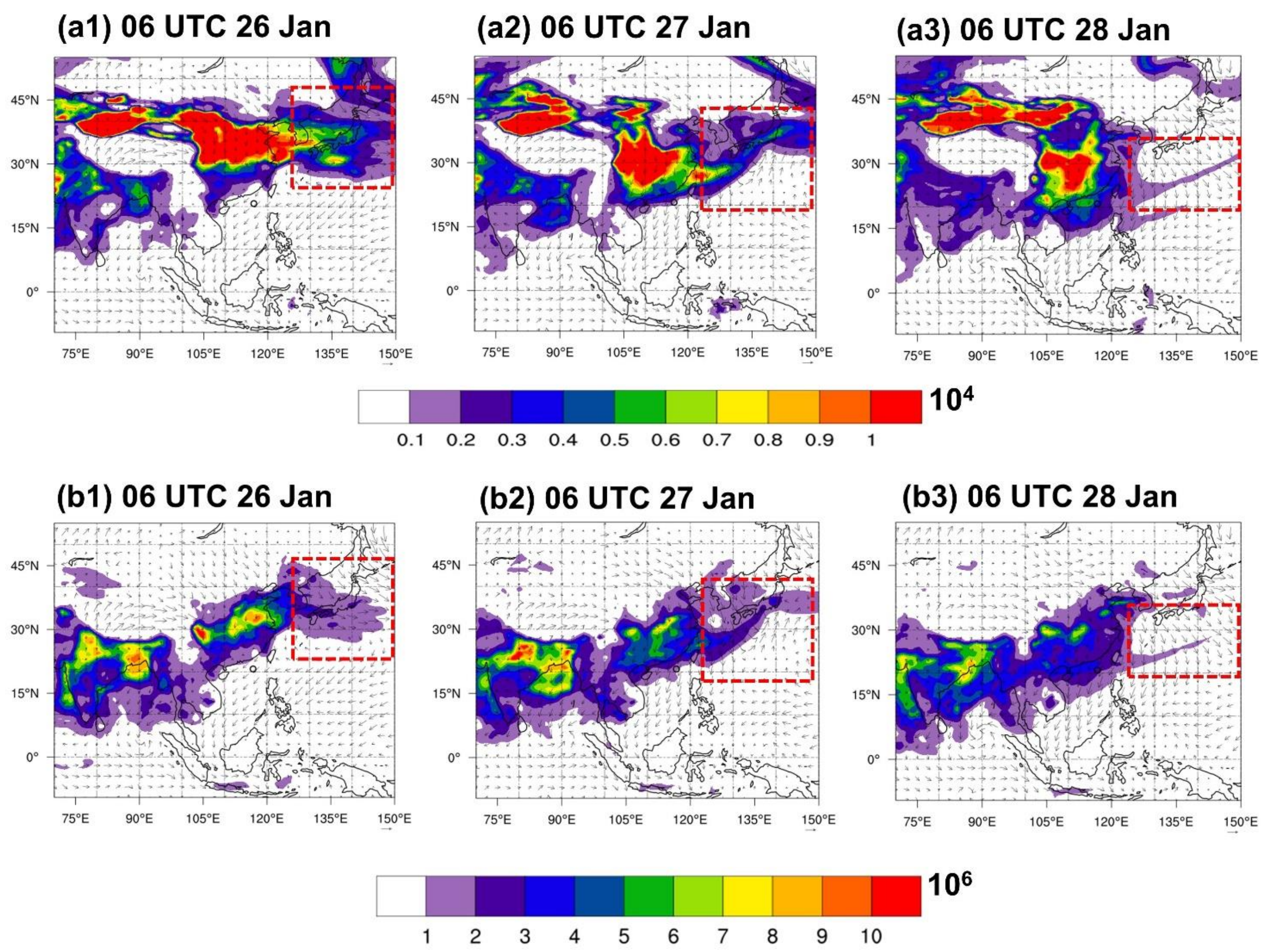

Figure S5: MERRA2 dust mass column (a1-a3) and black carbon mass column (b1-b3) during 06 UTC (a1, b1) 26 January, (a2, b2) 27 January and (a3, b3) 28 January 2023.

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

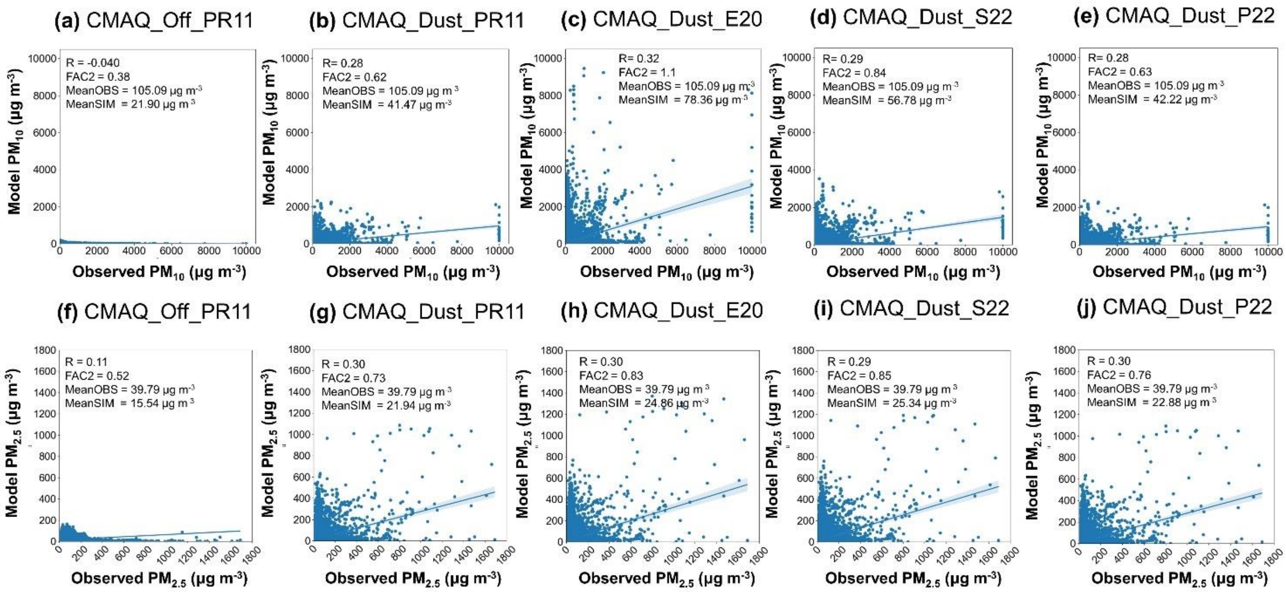

Figure 5: The scatter plot of the observed against modeled PM_{10} (a-e) and $PM_{2.5}$ (f-j) for CMAQ_Off_PR11 (a, f), CMAQ_Dust_PR11 (b, g), CMAQ_Dust_E20 (c, h), CMAQ_Dust_S22 (d, i) and CMAQ_Dust_P22 (e, j), at the 100 sites of the mainland China on 12 March-20 April 2021 (http://www.aqistudy.cn/). 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.

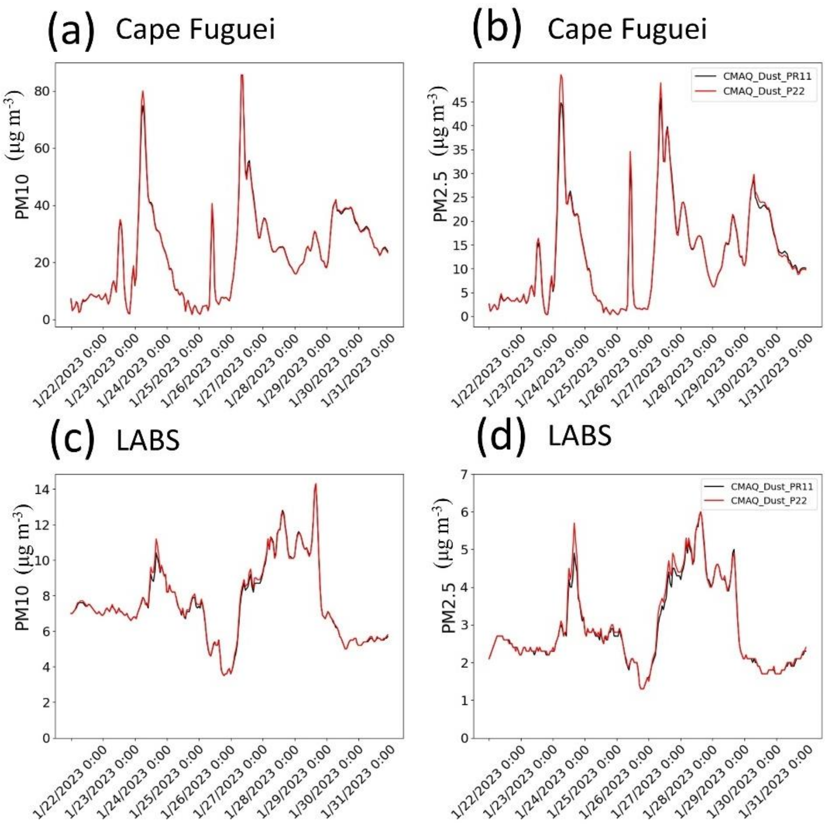

Figure S6: Time series of PM_{10} (left panel) and $PM_{2.5}$ (right panel) concentrations during 22-31 January 2023 under CMAQ_Dust_PR11 and CMAQ_Dust_P22 simulations over the Cape Fuguei (upper panel) and LABS (lower panel), representing the surface and high altitude, respectively.

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.

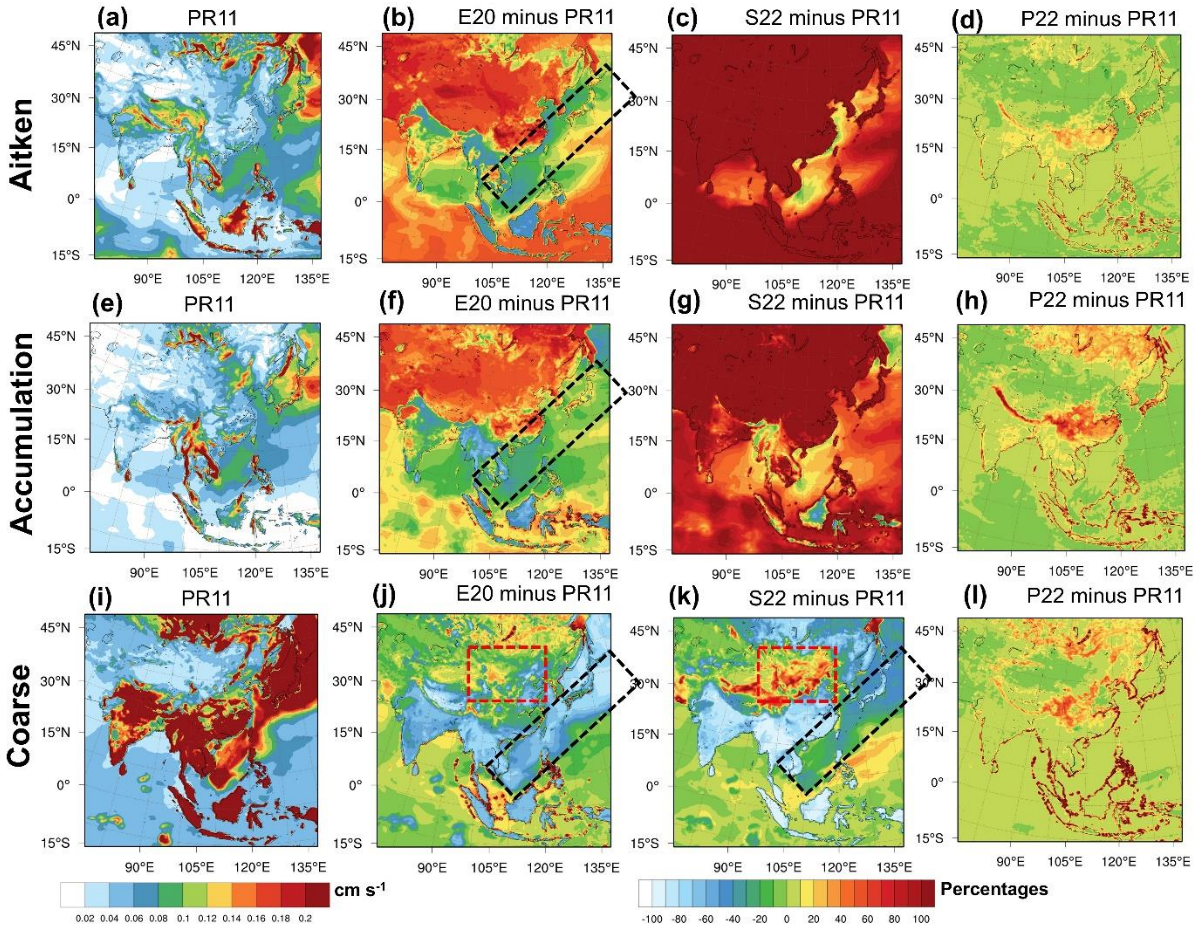

Figure 8: CMAQ estimated 10 days (22-31 January 2023) averaged for the (a-d) Aitken, (e-h) accumulation, and (i-l) coarse particle modes for PR11 dry deposition scheme (a, e, i) and the corresponding concentration percentage changes (%) using (b, f, j) E20, (c, g, k) S22 and (d, h, l) P22 schemes. Red-dash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

RC3: Specific comments and responses

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.