



Modeling CMAQ dry deposition treatment over Western Pacific: A distinct characteristic of mineral dust and anthropogenic aerosol

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Abstract. Dry deposition plays a vital role in the aerosol removal process from the atmosphere. However, 12 the chemical transport model (CTM) is sensitive to the dry deposition parameterization and yet remains 13 to be determined due to the limited particle deposition measurement. By utilizing the CMAQv5.4 with 14 the refined dust emission treatment (Kong et al., 2024), the East Asian dust (EAD) simulation during 15 January 2023 was constructed to evaluate the performance of dry deposition parameterizations developed 16 by PR11 (Pleim and Ran, 2011), E20 (Emerson et al., 2020), S22 (Shu et al., 2022) and P20 (Pleim et al., 17 2022), respectively. The result showed that the dry deposition parameterization could significantly 18 improve the CMAQ dust emission treatment. By implementing the E20 dry deposition scheme, the 19 CMAQ simulation performance of the surface PM_{10} has been considerably improved with the NMB of -20 41.9 %, as compared to the dry deposition proposed by PR11 (54.05 %), S22 (-47.01 %) and P22 (-53.90 21 %). The modeled PM_{10} pattern by E20 at the upper level (700 hPa) was mostly consistent with the 22 observed PM₁₀ at the Lulin Atmospheric Background Station (LABS; 23.47° N, 120.87° E; 2862 m a.s.l.) 23 where is a typical background site at Western Pacific, particularly in capturing the peak value. The high-24 altitude correlations (R) were well performed for E20 by 0.55, as compared to PR11 (0.47), S22 (0.54) 25 and P22 (0.46). Moreover, E20 improved the simulated aerosol optical depth (AOD) value during the 26 multiple dust storm in spring 2021. The noticeable reduction of the coarse mode particle's deposition 27 velocity (V_d) was responsible for resolving the PM_{10} simulation underestimation. Moreover, the 28 significant improvement of PM₁₀ was also shown by the modeled PM_{2.5} On 22-31 January 2023, the *in-*29 situ measurement of the upper level observed the possibility of natural dust and anthropogenic aerosol. 30





This is consistent with the CMAQ, which shows that both aerosol types displayed a clear "long dust-black carbon belt" along the 15°N. We proposed implementing the E20 dry deposition approach, resolving the

33 uncertainty of the CMAQ dust emission treatment.

34 1 Introduction

The chemical transport model (CTM) is a powerful tool for comprehending air pollution, encompassing 35 emission, transport, radiative impact, and removal mechanisms at various grid scales. Among 36 these, particle dry deposition, a crucial aerosol removal process, exerts a significant influence on the 37 physical and chemical aerosol properties, meteorological impact, terrain, and vegetation. The derivation 38 of the dry deposition is based on the resistance framework and electrical analogue, but its implementation 39 40 can vary across models (Wesley, 1989; Giardina and Buffa, 2018; Gaydos et al., 2007; Khan and Perlinger, 2017; Shu et al., 2017). A key challenge in dry deposition simulation is the scarcity of 41 measurement data for model verification, underscoring the necessity for further research to enhance the 42 accuracy of air quality modeling. 43

44 An immense range of dry deposition parameterization has been implanted in the model. The deposition mechanism by Slinn (1982) includes the deposition process such as turbulent transfer, 45 Brownian diffusion, impaction, interception, gravitational settling, and particle rebound, where the 46 particle grows under humid conditions. Zhang et al. (2001) suggested the dry deposition scheme is 47 sensitive to land use category and several parameters. For instance, due to the particle growth, the 48 deposition velocity (V_d) over the ocean is much higher than on another land surface, as the V_d increased 49 rapidly with the increase of particle size. However, Zhang et al. (2001) parameterization still 50 underestimated the global PM_{2.5} concentration. The latest dry deposition scheme revision by Emerson et 51 al. (2020) has resolved the problem, marking a significant step forward in our quest for more accurate air 52 53 quality modeling.

An updated deposition scheme that reduces the dependence of the deposition velocity on the aerosol mode width has been proposed (Shu et al., 2022). Indeed, the approach suggested that vegetation dependence increased the V_d for submicrons and decreased for large particles by 37 % and -66 %,





respectively. It also reduced the functional biases by 56-97 % for vegetated land-use type and equivalence performance over the water. Moreover, adding the second inertial impaction term for microscale obstacles such as leaf hairs, microscale ridges, and needle leaf edge effects managed to increase the mass dry deposition of the accumulation mode aerosols in the model (Pleim et al., 2022). These modifications reduced the averaged $PM_{2.5}$ in the atmosphere during July 2018 over the contiguous United States.

With a plethora of deposition approaches in use, it becomes paramount to comprehend their impact 62 on model performance in predicting aerosol behavior. The surface fine particle concentrations can vary 63 by 5-15 % due to the different dry deposition schemes, with the boarder extending by more than 200 % 64 of particle dry deposition due to the different algorithms (Saylor et al., 2019). A comprehensive evaluation 65 of five different parameterizations has been conducted, with the simplest and most effective deposition 66 mechanism suggested for the CTM (Khan and Perlinger, 2017). However, the model's reliance on 67 meteorological factors such as frictional velocity, relative humidity, rainfall, or wind speed, which can 68 significantly influence the model's accuracy, remains a challenge (Kong et al., 2021). 69

70 Besides the model bias on $PM_{2.5}$, the simulation of PM_{10} has been underestimated by the deposition mechanism, as the coarse mode has been generally represented, particularly over the western Pacific 71 (Kong et al., 2021). The V_d is overestimated for coarse particles, where the dry deposition velocity is too 72 high for coarse particles when the frictional velocity is large, which is why the surface PM₁₀ concentration 73 is underestimated (Ryu and Min, 2022). The model performance of PM_{10} simulation that is widely 74 influenced by the dust treatment embedded within CMAQ has been revised (Dong et al., 2016; Liu et al., 75 2021; Kong et al., 2021, 2024) and are found to effectively simulate the PM_{10} over the western Pacific 76 region such as Taiwan. However, the issue regarding the deposition algorithm's impact on the model 77 performance at the corresponding region needs to be discussed. The present research intends to evaluate 78 the CMAQ model performance due to the different deposition schemes on aerosols in the Taiwan region. 79

The model performance in Taiwan is of paramount importance in our study, as the area is equipped with a substantial number of well-maintained surface observation sites, providing comprehensive coverage.





serves as the sole background station for monitoring transboundary pollutants, further underscoring the
relevance of our research.

85 2 Data and Methodology

86 2.1 Dust emission treatment

Before delving into the details, it's important to understand the process of dust transport. Dust is primarily transported by wind through a process known as sandblasting (Kok et al., 2012). For dust to be uplifted, the horizontal wind speed must exceed a certain threshold frictional velocity $(u_{*,t})$, which is estimated by the model as follows:

91
$$u_{*,t} = u_{*,to} f_m f_r$$
 (1)

where f_m and f_r are the correction factors of soil moisture and surface roughness, respectively, and u^{*}, to is the ideal threshold friction velocity.

94 Through a collaborative effort, the windspeed, soil texture, soil moisture, and surface roughness length derived from field and laboratory studies have been integrated into the windblown dust treatment, 95 which is now a part of the Community Multiscale Air Quality (CMAQ) modeling system (Foroutan et 96 al., 2017). This model, developed and evaluated over the continental United States, has also been extended 97 to the East Asia region (Dong et al., 2016; Liu et al., 2021; Kong et al., 2021, 2024). Kong et al. (2024) 98 have proposed further improvements, including the integration of the revised soil moisture fraction, dust 99 emission speciation profile, and bulb soil density, to enhance the representation of the Asian dust 100 simulation. This ongoing collaboration is crucial for the continuous improvement of our understanding 101 and management of dust emissions. 102

103 2.2 Particle dry deposition schemes

Particle dry deposition is a complex process relating to the deposition velocity, particle size, source and
composition, land use surface, and meteorological condition. Generally, the flux of the particle mass
through the surface boundary layer is estimated as:



(2)

107
$$\mathbf{F} = \mathbf{C} \times \mathbf{V}_{\mathrm{d}}$$

where F is the deposition flux, C is the particle concentration at the surface layer, and V_d is the deposition velocity.

The difference in the particle concentration and deposition prediction among the various atmospheric chemistry models was probably due to the algorithm of the dry deposition particle. The algorithm describing particle deposition velocity as a function of particle size in almost all current air quality model systems is descended from (Slinn, 1982). The particle deposition according to vegetative canopies formulated the deposition velocity as:

115
$$\mathbf{V}_{\mathrm{d}} = \mathbf{V}_{\mathrm{g}} + \frac{1}{R_a + R_s} \tag{3}$$

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:

118
$$V_g = V_s + \frac{p_p D_p^2 g C_c}{18\eta}$$
 (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, n is the dynamic viscosity of air.

CMAQ implemented the dry deposition scheme that Pleim and Ran (2011) proposed based on 121 Slinn (1982), as shown in Table 1. Dry deposition is based on gravitational settling velocity (Vg), which 122 is the function of aerodynamic and surface resistance. According to Pleim and Ran (2011), chemical 123 surface flux modeling has become an essential process in the air quality model. For instance, the linkages 124 of ambient concentration levels to the deposition of SO_x and NO_x . The algorithm has been applied in 125 CMAQv4.5 up to CMAQv5.4. In CMAQv5.4, Surface Tiled Aerosol and Gaseous Exchange (STAGE) 126 deposition has been implemented within the model, where estimated fluxes from sub-grid cell fractional 127 land-use values, aggregates the fluxes to the model grid cell and unifies the bidirectional and 128





unidirectional deposition schemes using the resistance framework (Massad et al., 2010; Nemitz et al.,2001).

131 2.3 CMAQ model design

This study applied WRF v4.0 for the meteorological field parameters and CMAQv5.4 to simulate the 132 transboundary East Asian dust episodes on 22-31 January 2023, and the three dust storm episodes on 14-133 16 March 2021, 26-28 March 2021 and 17-19 April 2021. The modeling domain was set up to cover the 134 Taklamakan and Gobi Desert, with a resolution of 45 km, and nested towards Taiwan at a resolution of 135 15 km (d02) and 5 km (d03) (Fig .1, Table 2). Also, as Taiwan is influenced by biomass burning, the 136 domain covers up to PSEA, which will be carried out in the future (Ooi et al., 2021). The model consisted 137 of 40 vertical layers, with eight layers below~1 KM altitude, 13 layers below ~3 KM altitude, and 27 138 layers covering the upper layer to ~ 21 KM. The model's initial and lateral boundary conditions were 139 constructed using the National Centers for Environmental Prediction (NCEP) Final Analyses (FNL) 140 reanalysis dataset on a $0.5^{\circ} \times 0.5^{\circ}$ grid. The data assimilation was conducted by grid nudging in all the 141 142 domains. The CB06 gas-phase chemical mechanism and the AERO7 aerosol module model were implemented in CMAQ for the present study. 143

The anthropogenic emission inventories in East Asia, crucial for our research, were obtained from 144 the MICS-Asia (Model Inter-Comparison Study for Asia) Phase III emission inventory (Li et al., 2017). 145 The emissions of SO₂, NOx, NMVOC, NH₃, CO, PM₁₀, PM_{2.5}, BC, OC and CO₂ has been meticulously 146 modified, taking into account of the relative changes in China's anthropogenic emissions between 2010 147 and 2017 (Zheng et al., 2018). Additionally, the modified emission of NO₂ was adjusted further by the 148 satellite imagery OMI-NO₂ in January 2023 (Huang et al., 2021). Biogenic emissions for Taiwan were 149 prepared by the Biogenic Emission Inventory System version 3.09 (BEIS3, Vukovich and Pierce, 2002) 150 and, for regions outside Taiwan, by the Model of Emissions of Gases and Aerosols from Nature v2.1 151 (MEGAN, Guenther et al., 2012). TEDS 10.0 (Taiwan Emission Database System, TWEPA, 2011; 152 https://erdb.epa.gov.tw/,last access: 18 January 2024) was used for domain 3 (d03). 153

To ensure the precision of the multiple dry deposition parameterizations, the present research conducted six simulation scenarios, namely CMAQ_Off_PR11, CMAQ_Dust_PR11,





CMAQ Dust PR11, CMAQ Dust E20, CMAQ Dust S22 The CMAQ Dust P22. and 156 CMAQ_Off_PR11 scenario did not include the inline dust calculation (Table 3). Meanwhile, the latest 157 refined integrated dust treatment was implemented in the CMAQ_Dust_PR11 scenario (Kong et al., 158 2024). Indeed, both CMAQ Off PR11 and CMAQ Dust PR11 used the dry deposition mechanism by 159 Pleim and Ran (2011). The dry deposition mechanism of Emerson et al. (2020), Shu et al. (2022), and 160 Pleim et al. (2022) were implemented in CMAQ_Dust_E20, CMAQ_Dust_S22, and CMAQ_Dust_P22 161 scenarios, respectively. 162

163 2.4 Ancillary dataset

 PM_{10} (particulate matter $\leq 10 \ \mu m$ in aerodynamic diameter) and $PM_{2.5}$ (particulate matter $\leq 2.5 \ \mu m$ in 164 aerodynamic diameter) concentrations during the dust events in January 2023 were obtained from Lulin 165 Atmospheric Background Station (LABS; 23.47° N, 120.87° E, 2862 m MSL) and Cape Fuguei (25.30° 166 N, 121.54° E, 10 m MSL). The Modern Era Retrospective-analysis for Research and Application version 167 2 (MERRA-2) reanalysis data was used to demonstrate the spatiotemporal distribution of dust without the 168 influence of clouds. MERRA-2 (Gelaro et al., 2017) is a NASA reanalysis utilizing Goddard Earth 169 Observing System Data Assimilation System Version 5 (GEOS-5) and covering remotely sensed data at 170 a native spatial resolution of $0.5 \circ \times 0.625 \circ$. Also, Moderate Resolution Imaging Spectroradiometer 171 (MODIS) Terra satellite images and the level-3 MODIS AOD at 550 nm (MYD08) were obtained from 172 the U.S. National Aeronautics and Space Administration (https://worldview.earthdata.nasa.gov/). 173

174 **3 Results and Discussion**

175 **3.1 Observed air quality and weather conditions**

Figure 2 shows the dust outbreak over East Asia, displayed by the MODIS Terra sensor and MODIS AOD at 550 nm from 22-31 January 2023. The satellite image showed the dust claw pattern over East Asia on 24 and 25 January (Fig. a3, a4). The next day, the same region was covered by a thick cloud, then another dust plume again widely distributed during 27-30 January 2023. Using MODIS AOD to verify the dust plume (Han et al., 2012; Kong et al., 2021), the dust plume was distributed in Central China and northern Taiwan on 24 January 2023. Moreover, the most intense dust plume in the eastern China and





East China Sea region was observed on 27 January. Fig. S1 shows the synoptic weather map across the study domain. On 22-23 January, the southward high-pressure system was responsible for pushing the pollutant across the Asian Continent, which is consistent with Chuang et al. (2018) and Kong et al. (2021, 2022, 2024) (Fig. S1a-b). The high-pressure system that moved southward will then move eastward toward the Western Pacific Ocean (Fig S1c-d). Meanwhile, the high-pressure system on the northwest side again expands in the southeast direction. The second high-pressure system again pushed the pollutant for the second time and caused the high pollutant problem on 27 January.

The impact of East Asian dust on the air quality over the high-altitude western Pacific region was 189 widely discussed (Kong et al., 2022). Two interesting high pollution events at Mt. Lulin (2862 m above 190 sea level) during 24-26 Jan and 27-30 January, respectively, are shown in Fig. 3. The latter event was 191 more intense compared to the earlier one, where the maximum PM_{10} concentration can reach up to 35 µg 192 m⁻³. Moreover, it was observed that the black carbon concentrations could reach up to a maximum of 400 193 ng m⁻³. Based on the *in-situ* measurement, it was interesting to find the mixing state between dust, black 194 carbon, and brown carbon (Fig. 3c). Different from what has been discussed by Kong et al. (2022), the 195 long-range transport air pollution at the high-altitude not just merely EAD, but also included the 196 anthropogenic pollutant from mainland China. 197

198 3.2 Evaluation of CMAQ dust emission and dry deposition parameterizations

Table 3 shows the statistical analysis of PM_{10} and $PM_{2.5}$ concentrations over Cape Fuguei (northern 199 Taiwan) from 22-31 January under the multiple deposition mechanisms. $CMAQ_Off_PR11$, the PM_{10} 200 simulation presented without the inline dust calculation, recorded the normalized mean bias (NMB) of 201 -57.59 %. CMAQ_Dust_PR11 improved the simulation over Cape Fuguei (northern Taiwan) by -54.05 202 % as we included the refined dust treatment (Kong et al., 2024). However, the improvement is 203 insignificant due to the weak intensity dust episodes and the limitation due to the excessive deposition 204 mechanism within the model (Kong et al., 2021). Hence, we expanded the sensitivity simulation to 205 examine the impact of the deposition algorithm on the aerosol prediction. CMAQ_Dust_E20 simulations 206 utilizing the Emerson et al. (2020) approach increased the modeled PM₁₀ simulation by NMB of -41.9 %. 207





In addition, the deposition algorithm proposed by CMAQ_Dust_S22 (Shu et al., 2022) and CMAQ_Dust_P22 (Pleim et al., 2020) has reduced the NMB by -47.01 % and -53.90 %, respectively.

Instead of PM_{10} simulation, the present study found that the inline dust treatment and deposition algorithms could influence $PM_{2.5}$ simulation performances. For instance, the modeled $PM_{2.5}$ improved from -19.55 % (CMAQ_Off_PR11) to -16.53 % (CMAQ_Dust_PR11). Meanwhile, the deposition algorithm embedded in CMAQv5.4 has further enhanced the modeled $PM_{2.5}$ by -10.65 %, -15.22 %, and -8.84 % under CMAQ_Dust_E20, CMAQ_Dust_S22 and CMAQ_Dust_P22, respectively. This incident suggested that the East Asian dust from northwest China transported to the Western Pacific Ocean could also carry the anthropogenic emission of East China.

Figure 4 shows the time series of hourly PM_{10} and $PM_{2.5}$ concentrations over Cape Fuguei (northern Taiwan) and Mt. Lulin (high altitude region) from 22-31 January under the multiple deposition mechanisms. Generally, all the patterns of PM_{10} simulations were consistent with the observed PM_{10} , especially in capturing the peak value. For instance, the maximum observed (CMAQ_Dust_E20) PM_{10} concentrations at the surface during Jan 24 and Jan 27 were 141 (102.6) µg m⁻³ and 114 (163.2) µg m⁻³, respectively. A similar time-series pattern was found for the $PM_{2.5}$ simulation (Fig. 4b).

223 The CMAQ model performance over the high-altitude region needed to be carried out and discussed. The biomass-burning episode of the north peninsula of Southeast Asia over Mt. Lulin has been 224 225 finely correlated by plume rise injection (Chuang et al., 2016; Ooi et al., 2021). Meanwhile, the modeled EAD episodes over Mt. Lulin were due to the convergent pattern over the Asian Continental, which 226 correlated well with the MERRA2 (Kong et al., 2022). From Fig. 4c, the modeled PM₁₀ pattern for 227 CMAQ Dust Off could not correlate well with observed PM_{10} over Mt. Lulin, with a poor correlation of 228 0.17. The correlation was increased for CMAQ_Dust_PRll (0.47), CMAQ_Dust_S22 (0.54), 229 CMAQ Dust P22 (0.46), and primarily well performed for CMAQ Dust E20 (0.55). The modeled result 230 was somehow consistent with the surface PM_{10} simulation at Cape Fuguei. The high observed PM_{10} 231 episodes during 27-28 January with a maximum of 34.5 µg m⁻³ was only 53.3 % higher than 232





233 CMAQ_Dust_E20 of 22.5 μ g m⁻³. For the CMAQ PM_{2.5}, the simulation generally underestimated the 234 observed PM_{2.5}.

The present work is consistent with the dust scheme in the WRF-Chem, where the dust loading is 235 236 very sensitive to the dry deposition schemes and dust emission schemes, especially over the downwind region (Zeng et al., 2020). Fig. 5 shows the CMAQ estimated ten days averaged mean PM_{10} and $PM_{2.5}$ 237 for the PR11 deposition scheme and its corresponding change by E20, S22, and P22, 238 respectively. Generally, the spatial distribution of the high PM_{10} concentrations by > 80 µg m⁻³ was 239 distributed over northwest China, which is the dust source region's location, consistent with the simulation 240 suggested by Kong et al. (2021, 2022, 2024). Such high particulate matter dissipated to east China, 241 indicating the transport pathway in the southeastern direction towards the western Pacific (Fig. 5a). The 242 difference between E20 and PR11 suggested the high $PM_{10} > 50 \ \mu g \ m^{-3}$ distributed over northwest China. 243 meaning E20 successfully increased the PM₁₀ concentrations. As compared to the S22 deposition scheme, 244 it also increased the PM_{10} over northwest China by around 30 µg m⁻³ but was not as intense as E20. For 245 P22, the difference of PM₁₀ between PR11 and P22 was less than 10 μ g m⁻³, indicating less efficiency of 246 P22 in improving the PM_{10} . Another fascinating fact about E20 was that the PM_{10} increased over the 247 southern South China Sea. For the modeled $PM_{2.5}$ concentrations, the high concentration was 248 distributed over the Asian Continental. Under the E20 mechanism, the modeled $PM_{2.5}$ has been 249 increased over PSEA. For S22, such improvement of PM2.5 was more intense over a similar region. 250 Meanwhile, the PM_{2.5} simulated by P22 didn't have much difference compared to PR11, which showed 251 consistency as the PM_{10} simulation. 252

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 WPO due to the extreme typhoon episode (Kong et al., 2024). Hence, we intend to reemphasize the precision of various deposition schemes on the CMAQ for the recent dust storm episode highlighted by Kong et al. (2024). As a result, we specially conducted the East Asia region simulation





that is d01, for the 3-day averaged sensitivity test for three dust storm episodes: 14-16 March 2021 ("3.15" 260 dust storm), 26-28 March 2021 2021 ("3.27" dust storm), and 17-19 April 2021 2021 ("4.18" dust storm) 261 (Table 5). Overall, CMAQ Dust_E20 above 30°N has evaluated well the MODIS AOD by NMB of -26.2 262 %, as compared to PR11 (-37.4 %), S22 (-32.0 %) and P22 (-35.8 %). The CMAQ AOD by E20 during 263 the most intense SDS in 3.15 has significantly improved over northern China, the dust source region, as 264 shown in the red dash rectangular box (Fig. S2). Additionally, the modeled AOD by E20 over WPO 265 (shown in red dash rectangular box) increased in episode 4.18, reporting a value of 0.7 compared to 0.4 266 by PR11. Importantly, the E20 deposition scheme has primarily enhanced the PM_{10} prediction over the 267 marine boundary layer, addressing the model uncertainty due to the typhoon mentioned by Kong et al. 268 (2024) and demonstrating the practical implications of our research. 269

270 3.3 Impact on the CMAQ ambient particle concentrations

Figure 6 shows the boxplot of the 10-day averaged simulated V_d for the Aitken, accumulation, and coarse 271 particles modes under multiple deposition schemes, namely PR11 (Pleim and Ran, 2011), E20 (Emerson 272 et al., 2020), S22 (Shu et al., 2022) and P22 (Pleim et al., 2022). The different types of dry deposition 273 treatments substantially impact aerosol profile, altering the ambient total dry deposition regionally. As 274 shown in the figure, E20, S22, and P22 increased the deposition velocity of the Aitken (accumulation) 275 modes particle as compared to PR11 by 22.56 (11.32) %, 117.76 (86.43) % and 2.5 (7.52) % respectively. 276 The simulation suggested that S22 contained the highest V_d compared to E20 and P22. For coarse-mode 277 particles, the P22 simulation median V_d increased by 14.36 % compared to PR11. On the other hand, E20 278 and S22 showed a different simulation by the median V_d reduction by -9.1 % and -12.1 %, respectively. 279 Also, the 75^{th} percentile V_d has been significantly reduced by -50.57 % and -58.27 %, respectively. The 280 result suggested that the reduced V_d of the coarse mode particle was responsible for resolving the PM_{10} 281 simulation underestimation of PR11, consistent with the simulation by Shu et al. (2022) and Ryu and Min 282 (2022). The slow V_d means the total loss of aerosol to the surface has been minimized, leading to 283 increased aerosol concentration. 284

We estimated the CMAQ averaged particle modes for the PR11 dry deposition scheme and the corresponding percentage changes using E20, S22, and P22 (Fig. 7). By using E20 and S22, we found





that the V_d corresponding to the Aitken and accumulation modes has been increased by >100 % over 287 most of the CMAQ domain, which was most obvious over Asian continent. Meanwhile, the variation of 288 V_d distribution was insignificant for P22. For the coarse mode particles, the V_d has been tremendously 289 reduced for E20 and S22 compared to PR11. However, for S22, the V_d has increased by >100 % over 290 northwest China, which is the dust source region. This leads to a significant deposition over the desert 291 before transporting it to the downwind region, causing less PM_{10} simulated by S22 than E20. A previous 292 study proposed the V_d for the aerosol at the water surface was associated with the CTM uncertainly at the 293 downwind region (Kong et al., 2021, 2024; Ryu and Min, 2022). The V_d of Aitken and accumulation 294 modes at land and water surfaces increased generally, except E20 at the water surface. Interestingly, the 295 coarse mode V_d at the water surface for E20 and S22 decreased significantly by -44.65 % and -21.44 %, 296 respectively, suggesting that both deposition schemes, particularly E20, could resolve the excessive 297 deposition over the marine boundary layer, as mentioned by Kong et al. (2021) (Table 6). 298

299 **3.4 CMAQ of dust and black carbon synoptic pattern at the upper level**

Black carbon, known as elementary carbon, released from the biofuels, fossil fuels and biomass burning, 300 has been proven to impact the radiative budget and regional climate (Ramanthan, V and Carmicheal, 301 2008; Pani et al., 2016, 2020). In the meantime, China has been a significant contributor to global 302 anthropogenic black carbon emission, particularly in the cities of the northern part (Xiao et al., 2023; 303 Wang et al., 2024). During the severe dust episodes in the spring of 2023, the contribution of black carbon 304 brought by EAD was captured in North China (Wang et al., 2024). As depicted in Fig. 2, the 305 transboundary episode observed in the upper level of Taiwan during this event could be the mixing of the 306 natural dust and anthropogenic haze episodes, which demonstrates the consistency. Additionally, 307 308 blending mineral dust with anthropogenic transport due to the north easterly wind, a wind that blows from the northeast, has been a subject of extensive discussion (Lin et al., 2007, 2012; Li et al., 2012). During 309 the EAD, the dust from the Gobi Desert that was transported towards the western Pacific region could 310 also carry anthropogenic aerosol, contributing to different levels of pollutant concentration. However, the 311 distinct transport pathway at the high altitude between both aerosol types is a topic that has received less 312 attention but is of significant importance to our understanding of atmospheric dynamics. 313





Figure 8 illustrates mineral dust concentration's spatial and temporal distribution under the 314 CMAQ_Dust_E20 scenario at 700 hPa from 24-30 January. The model reveals a high proportion of 315 modeled dust aerosol (red dash circle) at the source region, indicating an uplift from the surface to 700 316 hPa (Fig. 8a, b). This uplift, driven by the strong pressure gradient at the surface and the 'eastward moving 317 trough system' at the upper level (700 hPa), is a key factor in the eastward and southward transfer of the 318 dust (Fig. 8c-d). The high dust fraction reappears at the source region (Fig. 8e, f) and is transported 319 eastwardly by the similar upper-level trough (Fig. 8g-j), causing a long dust belt at 15°N, distributing 320 over central Asia continental, Taiwan Straits, Taiwan and large part of western Pacific Ocean. (Fig. 8i, j). 321 On 29 January, the model of E20 clearly predicted that the dust plume moved in the southward direction 322 toward the South China Sea (Fig. 8k, l). The dust aerosol was left distributed at a certain part of the 323 northern South China Sea and the Philippine Sea until it totally dissipated (Fig. 8m, n). This interesting 324 result suggests the possible EAD at the longer distance at the upper level, which is a topic for further 325 investigation. 326

The southward high-pressure system responsible for the long-range transport haze episode has 327 been widely discussed (Chuang et al., 2008; Kong et al., 2021)—however, the upper-level transboundary 328 transport needs to be addressed more. While focusing on CMAQ Dust E20, we attempted to characterize 329 the transboundary of modeled black carbon at the upper level (700 hPa) (Fig. 9). As shown in Fig. 9(c, 330 e), the modeled black carbon concentration is shown to be significantly distributed at central China. The 331 black carbon transport pattern followed the eastward-moving trough system as the plume moved eastward 332 and southward (Fig. 9g-l). Interestingly, the long black carbon belt is consistent with the long dust belt, 333 as shown in Fig. 8. For instance, both modeled dust and black carbon were distributed at the South China 334 Sea (Fig. 8m, n; Fig 9m, n). This means that the black carbon due to the anthropogenic emission and the 335 natural EAD shared a similar transport pattern at the upper level, driven by the trough system. Such 336 consistency has been verify by the AOD simulated by CMAQ, along with the MERRA-2 dust and black 337 carbon mass column over the region (Fig. S3). 338

Dust aerosol vertical profiles (Fig. 10) show the large dust fraction was distributed over the Asian Continent (Fig. 10a, b), according to the transect drawn as a red-dash line in Fig. 1. Due to the westerly





winds as illustrated in Fig. 8, the aerosol plume transported at the eastward direction toward the western 341 Pacific Ocean, that vastly accumulated along the 700 hPa altitude. Another plume was found across the 342 ocean on the east side of Taiwan Island (Fig. 10b). The plume was moved eastward (Fig. 10c-f). During 343 00 UTC on 27 January, another large fraction of dust covered the Asian Continent (Fig. 10g); in the next 344 12 hours, the model showed an apparent dust plume located in the Western Pacific Ocean, with much 345 higher dust concentrations compared to Fig. 10b. The plume again distributed eastward showed a clear 346 dust dome (Fig. 10i-j). Then, due to the westerly airflow, the dust aerosol slowly dissipated at the upper 347 layer of the western Pacific. 348

The vertical profile of the modeled black carbon has a similar transport pattern as mineral dust 349 (Fig. 11). As shown in Fig. 11h, the modeled black carbon was found distributed at the western Pacific 350 Ocean. In Fig. 11i, a clear black carbon dome was distributed along 700 hPa, showing a similar pattern 351 as dust. This simulation proposes the consistency of the "double dome" mechanism of Asian dust and 352 biomass burning episodes, as the coarse particles could comprise of fine particles (Dong et al., 2018; 353 Huang and Fu, 2019). The issue regarding whether or not such a mechanism could cause the warming 354 effect can be considered as a future study. However, the difference is that the dust dome contains a higher 355 fraction of concentrations as compared to the black carbon dome. Considering the maximum height, the 356 present simulation suggests the dust aerosol can reach up to 500 hPa, which is consistent with Kong et al. 357 (2021). Contrary, the black carbon plume was slightly lower with approximately 600 hPa of the maximum 358 height under the same meteorological condition. As this section essentially discusses the similarity and 359 distinctiveness of natural dust and anthropogenic aerosol at the upper level, we are interested in 360 characterizing the synoptic pattern. Hence, the present simulation did not consider the two-way coupling 361 model, and it is strongly suggested for future study. 362

363 4.0 Summary and Conclusions

The chemical transport model is considered sensitive to the dry deposition parameterization besides the dust emission treatment. The present study demonstrates the impact of the four dry deposition parameterizations on aerosol performance in East Asia. It provides a significant analysis of the transboundary transport of East Asian Dust to Taiwan from a case study of 22-31 January 2023. The





incorporation of the latest dust emission treatment with PR11 (Pleim and Ran, 2011; Kong et al., 2024) 368 to the CMAQ slightly improved the model performance to -54.05 % from -57.59 %. While this 369 improvement may seem insignificant, it underscores our commitment to accuracy in aerosol modeling 370 and the potential for further advancements in the field. By implementing the E20 dry deposition scheme, 371 characterized by adding the collection efficiency by interception across the land surface, the CMAQ 372 simulation of the surface PM₁₀ has been vastly improved by NMB of -41.9 %, as compared to the dry 373 deposition proposed by S22 (-47.01 %) and P22 (-53.90 %). Moreover, the modeled PM_{10} pattern by 374 CMAQ_Dust_E20 at the upper level (700 hPa) was mostly consistent with the observed PM_{10} , especially 375 in capturing the peak value. The dry deposition of E20 was correlated well with the high altitude in-situ 376 by 0.55, as compared to PR11 (0.47), S22 (0.54) and P22 (0.46). Such a significant difference in PM_{10} 377 improvement has also been shown by modeled PM2.5. The simulation of surface PM2.5 by PR11 has been 378 improved to -16.53 % from -19.55 %, after using the latest dust treatment, and further enhanced by E20 379 (-10.65 %), S22 (-15.22 %) and P22 (-8.84 %). Additionally, the CMAQ_Dust_E20 treatment provided 380 an optimized AOD simulation value for the multiple dust storm episodes in spring 2021. 381

382 The previous CMAO model, modulated by Kong et al. (2021; 2024), showed excessive deposition at the marine boundary layer, leading to an underestimation of the modeled surface PM_{10} . However, our 383 updated model, using the E20 and S22 schemes over the entire model domain, has not just reduced, but 384 significantly reduced the 75th percentile of V_d by -50.57 % and -58.27 %, respectively. This precise 385 reduction of V_d of the coarse mode particle, responsible for resolving the PM_{10} simulation 386 underestimation, has not just minimized, but effectively minimized the total loss of aerosol to the surface, 387 leading to a concentration increment. Furthermore, the intense decrease of modeled V_d across the water 388 surface by E20 could play a crucial role in resolving the excessive aerosol deposition over the ocean 389 layer. These precise adjustments in the model's parameters demonstrate our commitment to improving 390 the accuracy of aerosol modeling and should inspire confidence in the results of our research. 391

In addition, the updated CMAQ was used to investigate the synoptic pattern at the upper level. The transboundary transport of EAD from the Asian Continent towards the western Pacific Ocean at the upper level was associated with the eastward moving trough system. Such transport mechanism is





found to bring along the black carbon aerosol, which is primarily the main element of China's humanmade emissions. More interestingly, both aerosol profiles created a "long dust-black carbon belt" along the 15°N. The 'double dome mechanism ', a concept proposed by Huang et al. (2019) that depicts the superposition of the two types of aerosol, was also simulated in the present study. However, since the present work did not consider two-way radiative impact, the issue of warming/cooling is proposed in the future study.

401 Data Availability

MERRA-2 data are available online through the NASA Goddard Earth Sciences Data Information Services Center (GES DISC; <u>https://disc.gsfc.nasa.gov</u>; last access: 01 August 2024). MODIS data used in this study are available at <u>https://asdc.larc.nasa.gov/</u>(last access: 01 August 2024). The observational data at LABS can be ordered by contacting corresponding authors.

406 Author Contribution

407 Steven Soon-Kai Kong: Conceptualization; Data curation; Formal analysis; Investigation; Methodology;
408 Software; Validation; Visualization; Writing – original draft; Writing – review and editing.

Software; Validation; Visualization; Writing – original draft; Writing – review and editing.
Joshua S. Fu: Conceptualization; Investigation; Methodology; Formal analysis; Writing – review and

410 editing.

411 Neng-Huei Lin: Conceptualization; Visualization; Supervision; Funding acquisition; Resources; Writing

- 412 review and editing.
- 413 Guey-Rong Sheu: Funding acquisition; Resources.
- 414 Wei-Syun Huang: Data curation; Software.

415 **Competing Interests**

416 Some authors are members of the editorial board of journal ACP.





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Mechanisms	V_d	R_a	R_b	R_s
Pleim and Ran (2011)	$\frac{1}{R_a + R_b + R_s}$	$\frac{\ln\left(\frac{ZR}{ZO}\right) - \psi H}{K^{2}}$	$\frac{B^{-1}}{u_*} \left(\frac{Sc}{Pr}\right)^{2/3}$	$\left(\frac{1}{R_{st}} + \frac{1}{R_w} + \frac{1}{R_q}\right)^{-1}$
Emerson et al. (2020)	$V_g + \frac{1}{R_a + R_s}$	$\frac{\ln\left(\frac{ZR}{ZO}\right) - \psi H}{\ln\left(\frac{ZR}{ZO}\right) - \psi H}$	0	$\frac{1}{\overline{\mathcal{E}}_{0} u_{*} (E_{h} + E_{lm} + E_{in})R}$
Pleim et al. (2022)	$\frac{V_g}{1 - \exp(-V_g(R_+ + R))}$	$\ln\left(\frac{ZR}{ZO}\right) - \psi H$	V_g	0
Shu et al. (2022)	$1 - \exp(-v_g(R_a + R_b))$ V_g	$\frac{\kappa u_*}{\ln\left(\frac{ZR}{2}\right)} - \psi H$	$LAI. u_*(E_B + E_{im} + E_{in})R$	1
	$\overline{1 - \exp(-V_g(R_a + R_s))}$	$\frac{\operatorname{III}\left(\overline{ZO}\right) - \psi \Pi}{\kappa u_*}$		$(1 + f_{veg}(max(LAI - 1, 0))F_f u_*(E_b + E_{Im}))$
585 $V_d = depo$	sition velocity			
586 $V_g = settli$	ng velocity			
587 $R_a = aerod$	ynamic resistance			
588 $R_b = quasi$	-laminar boundary layer			
589 $R_s = surfactors$	e resistance			
590 $R_{st} = stoma$	atal resistance			
591 $R_w = cutics$	ular resistance			
592 $R_g = \text{groun}$	d resistance			
593 $\mathbf{B}^{-1} = $ inver	se Stanton number			
594 $u_* = \text{Ir1ct10}$	nal velocity			
595 SC = SCHIII	ndt number			
596 FI = allalo	gous qualitity for fleat			
597 LAI – Lea 598 E _p – Brow	nian collection efficiency			
598 $E_B = Drow$ 500 $E_{cm} = Impa$	intercence of the section of the sec			
$E_{im} = Interconstructure F_{im} = Interconst$	ception efficiency			
$601 \mathbf{R} = \text{rebout}$	nd factor			
$602 E_0 = depose$	sition velocity			
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Table 1. Detailed mechanism expression relating dry deposition velocity and resistance parameters.





617	Table 2.	Model	settings.
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	Model setting		Descriptions				
	Period		14-16 March 2021, 26-28 March 2021, 17-19 April 2021 and				
			22-31 January 2023				
	Domain	d01, d02 and d03 with 45 KM, 15 KM and 5 KM of the resolut					
			respectively				
	Boundary condition		NCEP FNL lateral boundary condition				
	Surface and land	surface	NOAH				
	model						
	Numerical weather m	odel	WRF v40, including grid and observation nudging at d01.				
	Chemical transport m	odel	CMAQ v5.4				
	Gas-phase chemistr	ry and	CB06e51 + AE7				
aerosol mechanism							
	Emission Inventory		MICS-ASIA III emission on 2023, adjusted from the emission 2017				
			(Zhang et al., 2018) based on OMI-NO _x satellite (Huang et al., 2021).				
	Online dust treatment	ţ	The windblown dust treatment suggested by Kong et al. (2024).				
618							
619	Table 3. Simulation sc	cenarios u	sed in this present study.				
	Scenarios	Descript	ions				
	CMAQ_Off_PR11	Without	in-line calculation of dust, with the dry deposition algorithm by Pleim				
		and Ran	(2011).				
	CMAQ_Dust_PR11	Impleme	ent the latest refined dust treatment proposed by Kong et al. (2024), with				
		the dry o	leposition algorithm by Pleim and Ran (2011).				
	CMAQ_Dust_E20	Same as	CMAQ_Dust_PR11, but with the dry deposition algorithm by Emerson				
		et al. (20)20).				
	CMAQ_Dust_S22	Same as	CMAQ_Dust_PR11, but with the dry deposition algorithm by Shu et				
		al. (2022	2).				
	CMAQ_Dust_P22	Same as	CMAQ_Dust_PR11, but with the dry deposition algorithm by Pleim et				
		al. (2022	2).				
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Table 4. Statistical evaluation for PM_{10} and $PM_{2.5}$ concentrations during 22-31 January 2023 for Cape Fuguei under the multiple simulation scenarios.

	Benchmark	CN	MAQ	CMAQ			
		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	23.04	26.48	
NMSE		0.82	0.71	0.49	0.71	0.56	
NMB	$\pm 85\%$	-57.59	-54.05	-41.90	-53.90	-47.01	
Corr	> 0.35	0.41	0.44	0.52	0.42	0.46	
NMBF		-1.36	-1.18	-0.72	-1.17	-0.89	
			PM _{2.5}				
MeanObs		15.52	15.52	15.52	15.52	15.52	
MeanMod		12.48	12.95	13.86	13.16	14.15	
NMSE		0.31	0.29	0.29	0.31	0.30	
NMB	$\pm 85\%$	-19.55	-16.53	-10.65	-15.22	-8.84	
Corr	> 0.35	0.52	0.55	0.53	0.52	0.53	
NMBF		-0.24	-0.20	-0.12	-0.18	-0.10	

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Table 5. CMAQ AOD evaluation against MODIS daily observation nearby the dust source region (above 30°N) with Normalized Mean Bias (NMB) for the multiple simulation scenarios during the dust storm episode of 3.15 (14-16 March 2021), 3.27 (26-28 March 2021) and 4.18 (17-19 April 2021).

<u>episode of 5.15 (14-10 N</u>	$(1a1011 \pm 0.021), 5.27$	(20-20 Watch 2021)	and 4.10 (17-17 Api	11 2021).
Cases	3.15	3.27	4.18	Mean
CMAQ_Off_PR11	-81.92	-75.10	-55.88	-70.97
CMAQ_Dust_PR11	-49.54	-46.12	-16.49	-37.38
CMAQ_Dust_E20	-38.97	-36.39	-3.20	-26.19
CMAQ_Dust_S22	-46.41	-41.84	-7.83	-32.03
CMAQ_Dust_P22	-48.45	-44.52	-14.52	-35.83

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Table 6. Percentage change by PR11 corresponding to E20, S22 and P22, for Aitken, accumulation and coarse modes over land and ocean boundary layer.

Changes	Aitken		Accumulation		Coarse	
(%)	Land	Ocean	Land	Ocean	Land	Ocean
E20	12.66	20.06	5.43	-5.19	-47.10	-44.65
S22	173.74	89.45	96.52	52.35	-70.29	-21.44
P22	6.10	1.37	17.66	1.52	10.06	6.86

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Figure 1: Modeling domain configuration in East Asia. Ground-based air quality stations in Taiwan at Cape Fuguei and Lulin Atmospheric Background Station (LABS) are shown in the zoomed panel. The red dash line ($A \rightarrow B$) represents the transects that the aerosol plumes traveled along in this study and that are discussed in Section 3.4;

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Figure 2: MODIS Terra images (a1-a10) and MODIS aerosol optical depth AOD at 550 nm (b1-b10) showing dust outbreak across East Asia during 22-31 January 2023. Red Rectangular, triangle, star and circle indicate Lanzhou, Beijing, Shanghai and Taiwan. The red circle with dash line indicates the dust plume.







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Figure 3: (a) Time series of observed pollutants over LABS during January 2023. The aerosol radiation
properties during (b) 24-26 January and (c) 27-30 January 2023.





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







Figure 5: CMAQ estimated 10 days (22-31 January 2023) averaged mean (a-d) PM₁₀ and (e-h) PM_{2.5} for
(a, e) PR11 dry deposition scheme and the corresponding concentration changes using (b, f) E20, (c, g)
S22 and (d, h) P22 schemes.

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Figure 6: 10-days averaged dry V_d predicted by CMAQv5.4 for the Aitken, accumulation, and coarse particle modes using the PR11(blue), E20(red), S22(green) and P22(purple) particle dry deposition schemes. The variability illustrated by the boxes and whiskers corresponds to spatial variability in annually averaged values throughout the CMAQ domain.

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Figure 7: 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.

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Figure 8: CMAQ mineral dust aerosol concentration at the 700 hPa during 24-30 January 2023. The
yellow arrows highlight the trough moving direction. The dash-black rectangular box highlights the dust
belt.







Figure 9: CMAQ black carbon concentration at the 700 hPa during 24-30 January 2023. The yellow
arrows highlight the trough moving direction. The dash-black rectangular box highlights the dust belt.







- 710 Figure 10: Vertical profile of simulated dust aerosol for the CMAQ simulation during 24-30 January
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- Figure 11: Vertical profile of simulated black carbon aerosol for the CMAQ simulation during 26-29January 2023.

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