| 1 | An evaluation of ozone dry deposition simulations in East Asia |
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23 Abstract

24 We use a 3-D regional atmospheric chemistry transport model (WRF-Chem) to 25 examine ozone dry deposition in East Asia, which is an important but uncertain 26 research area because of insufficient observation and numerical studies focusing on 27 East Asia. Here we compare two widely used dry deposition parameterization 28 schemes, Wesely and M3DRY, which are used in the WRF-Chem and CMAQ models, 29 respectively. Simulated ozone dry deposition velocities with the two schemes under 30 identical meteorological conditions show considerable differences (a factor of 2) 31 owing to surface resistance parameterization discrepancies. Resulting ozone 32 concentrations differ by up to 10 ppbv for a monthly mean in May when the peak 33 ozone typically occurs in East Asia. An evaluation of the simulated dry deposition 34 velocities shows that the Wesely scheme calculates values with more pronounced 35 diurnal variation than the M3DRY and results in a good agreement with the 36 observations. However, we find significant changes in simulated ozone concentrations 37 using the Wesely scheme but with different surface type datasets, indicating the high 38 sensitivity of ozone deposition calculations to the input data. The need is high for 39 observations to constrain the dry deposition parameterization and its input data to 40 improve the use of air quality models for East Asia.

41

43 1. Introduction

Ozone (O₃) is a harmful air pollutant in surface air and the primary chemical oxidation driver in the free troposphere. Tropospheric ozone concentrations are largely controlled by the balance among net chemical production, influx from the stratosphere, and physical losses (Wu et al., 2007). Dry deposition of ozone is a dominant physical loss process and accounts for approximately 25% of the total ozone lost in the troposphere (Lelieveld and Dentener, 2000).

50 In typical chemical transport models, dry deposition is calculated as a first-order 51 process that uses dry deposition velocity, which is parameterized as a function of 52 surface type and atmospheric stability conditions (Wesely, 1989). However, in models, 53 its parameterization is highly uncertain because of complexities from surface 54 conditions at sub-grid scales (Wu et al., 2011). Thus, previous studies on dry 55 deposition calculations have primarily focused on the United States and Europe, for 56 which observations on ozone fluxes or dry deposition velocities were available to 57 validate either simulated ozone losses or dry deposition velocity parameterization 58 (Rannik et al., 2012; Wu et al., 2011; Charusombat et al., 2010; Gerosa et al., 2007). 59 East Asia (China, Japan, and Korea) has recently experienced rapid economic 60 growth, during which anthropogenic emissions have increased and deteriorated air 61 quality (Ohara et al., 2007). Thus, the use of air quality models has also increased in 62 East Asia to understand the spatial and temporal distributions of air pollutants and to 63 examine the impact of the increased anthropogenic emissions on air quality 64 degradation for East Asian countries (Park and Kim, 2014). A critical role of such 65 models includes quantifying the regional air pollution sources, including trans-66 boundary transport of air pollutants and their precursors in East Asia (Ku and Park, 67 2011; Jeong et al., 2011). In this context, the dry deposition simulation is important

68 for accurately assessing the contribution from a source to regional air pollutant69 concentrations.

However, air quality model evaluations have been relatively limited because of
the lack of long-term regional observations in East Asia. In particular, evaluating
individual processes, including the dry deposition calculation, has not been rigorous
for East Asia. Several studies focusing on ozone dry deposition simulations have been
conducted for a tropical forest in Southeast Asia (Matsuda et al., 2005; Matsuda et al.,
2006), but the vegetation type differs from East Asia.

76 The purpose of this study is to evaluate the ozone dry deposition simulations 77 (schemes) in two of the most widely used regional air chemistry models in East Asia: 78 the Weather Research and Forecasting-Chemistry (WRF-Chem) and the Community 79 Multiscale Air Quality (CMAQ) models. We conducted multiple model simulations to 80 understand the differences between the two models as well as the two different dry 81 deposition schemes and factors that affect dry deposition and ozone concentrations in 82 East Asia. We also evaluated the simulated ozone concentration and dry deposition 83 velocity by comparing such results with observations. Finally, we conducted several 84 sensitivity simulations using different input datasets to demonstrate the uncertainties 85 of the dry deposition calculations, which should be considered in assessing the spatial 86 and temporal distributions of ozone and the contributions from a specific source to a 87 particular region, including the trans-boundary transport of ozone precursors in East 88 Asia.

89

- 90 2. Model description
- 91 **2.1 General Description**

92 We used the WRF-Chem model (version 3.3) to simulate ozone in East Asia. 93 The model is a fully coupled meteorology-chemistry model, which was developed by 94 the National Center for Atmospheric Research (NCAR) (Grell et al., 2005) to account 95 for the interaction between meteorological and chemical processes at each time step 96 (Chapman et al., 2009). The model is described in detail elsewhere (Grell et al., 2005). 97 Herein we primarily describe our model simulations. 98 The model has a horizontal resolution of 45 x 45 km with 14 eta vertical grids 99 and a 50 hPa top. The model domain for our simulations is shown in Fig. 1, which 100 includes the nested grid domain that focuses on the Korean peninsula. For 101 meteorology simulations, we used physics modules in the WRF, as shown in Table 1. 102 In particular, turbulent mixing at the surface and within the planetary boundary layers 103 was calculated using schemes developed by Chen and Dudhia (2001) and Hong et al. 104 (2006), respectively. 105 We used anthropogenic emissions from the Sparse Matrix Operator Kernel 106 Emissions-Asia (SMOKE-Asia), which was developed by Woo et al. (2012) to 107 operate the CMAQ model (Byun and Ching, 1999) over East Asia. The SMOKE-Asia 108 calculates anthropogenic emissions based on the Carbon Bond 05 (CB05) chemical 109 mechanism (Appel et al., 2007), which slightly differs from the Carbon Bond 110 mechanism Z (CBMZ) used in WRF-Chem. We used the chemical mapping in Table 111 2 to match the emission species between CB05 and CBMZ. A few species do not 112 precisely correspond between the two schemes, but such species are relatively 113 unimportant for our ozone simulations below. The total NO_x, CO, and VOC emissions in the domain are 24.6 Tg yr⁻¹, 150.2 Tg yr⁻¹, and 96.0 Tg yr⁻¹, respectively. 114 115 The initial and lateral boundary conditions for the meteorology simulations were determined using a WRF preprocessing system with the NCEP Final 116

117 Operational Model Global Tropospheric Analyses data (National Centers for 118 Environmental Prediction, 2000). Climatological values were used to generate the 119 initial and boundary values for the chemical species concentrations (Grell et al., 2005). 120 We conducted WRF-Chem simulations for April-July 2004 in East Asia using 121 the two dry deposition schemes, Wesely and M3DRY. A description on the two 122 schemes is provided in Sections 2.2 and 2.3. Identical boundary and initial conditions 123 were used for the model, including species emissions, except for the dry deposition 124 scheme. Therefore, the differences in the results are entirely due to the discrepancy 125 between the two dry deposition schemes. The model simulation for April was used for 126 spin-up, and we primarily focus our analysis on the results for May when the peak 127 ozone typically occurs in East Asia. Because of summer monsoon, ozone 128 concentrations are lower in summer than in spring in East Asia (Li et al., 2007). 129

130 **2.2 Dry deposition parameterization**

131 Chemical species loss (*F*) owing to dry deposition in air chemistry models is
132 typically computed as a first-order process with the dry deposition velocity as shown
133 in equation (1).

(1)

134 $F = v_d C$

135 v_d indicates the dry deposition velocity, and *C* represents the species concentrations 136 in the lowest model layer. Therefore, the species lost through dry deposition is 137 directly proportional to the dry deposition velocity, which is parameterized in such 138 models.

139 The dry deposition velocity is computed as the reciprocal of the sum for 140 aerodynamic resistance (R_a) , quasi-laminar resistance (R_b) , and surface resistance (R_c) 141 as follows:

142
$$v_d = \frac{1}{R_a + R_b + R_c}$$
. (2)

| 144 | important factor that determines dry deposition velocity. Generally, the surface |
|-----|---|
| 145 | resistance is the largest among the three resistances, and it determines the dry |
| 146 | deposition velocity (Erisman et al., 1994); we will discuss the surface resistance |
| 147 | formulation in Section 2.3. |
| 148 | Here we compare two widely used dry deposition schemes: the Wesely and |
| 149 | M3DRY schemes. The first scheme was developed by Wesely (1989) and is used in |
| 150 | WRF-Chem as a default method (hereinafter, the Wesely). The latter scheme was |
| 151 | proposed by Pleim et al. (2001) and is used as a default scheme in CMAQ; it is a part |
| 152 | of the meteorological transport module Meteorology-Chemistry Interface Processor |
| 153 | (MCIP) version 3.3 used in CMAQ, (Otte and Pleim, 2010) (hereinafter, the M3DRY). |
| 154 | We implemented the M3DRY as part of MCIP v3.3 in WRF-Chem to examine the |
| 155 | sensitivity of ozone simulations to the two different dry deposition schemes using |
| 156 | identical input data. We found that both schemes use fairly similar parameterizations |
| 157 | for the aerodynamic and quasi-laminar resistances, but their surface resistance |
| 158 | parameterizations differ considerably, as discussed below. |

As shown in equation (2), the resistance with the largest value is the most

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160 **2.3 Surface resistance parameterization**

161 The surface resistance represents the surface uptake of chemical species and

162 depends on the surface chemical and physical characteristics. As the surface

163 resistance decreases, surface uptake of chemical species increases. The surface

164 resistance can be further classified into four specific resistances: the

165 stomata mesophyll resistance (R_{sm}), cuticle resistance (R_{cut}), in-canopy resistance

166 (R_{inc}) , and ground resistance (R_{gnd}) . The first three are related to physical and

167 chemical characteristics of vegetation, and the last resistance is related to ground
168 conditions. The four resistances combine in parallel to yield the surface resistance as
169 follows:

170
$$\frac{1}{R_c} = \frac{1}{R_{sm}} + \frac{1}{R_{cut}} + \frac{1}{R_{inc}} + \frac{1}{R_{gnd}}$$
. (3)

171 Therefore, the resistance with the smallest value largely determines the surface 172 resistance. Typically, the stomata mesophyll and ground resistances are the smallest 173 (Wu et al., 2011). The stomata mesophyll resistance is related to vegetation 174 photosynthetic activity, and thus, is a function of solar radiation. During the day, the 175 stomata mesophyll resistance substantially decreases, and it has the smallest value 176 among the four, causing it to largely determine the surface resistance. The diurnal 177 variation of the stomata-mesophyll resistance differs depending on the vegetation type. 178 However, at night, its value becomes higher than the ground resistance, which plays a 179 key role in determining surface resistance without solar radiation. In models, the four 180 resistances shown in equation (3) are calculated using complex parameterizations; a 181 detailed discussion on this subject is beyond the scope of our work. We briefly 182 discuss major differences of the stomata-mesophyll and ground resistances 183 parameterizations between the two schemes below. 184 The key part of the stomata-mesophyll resistance is the stomata resistance in 185 both of the two dry deposition schemes. In the Wesely, the stomata resistance is 186 parameterized as a function of solar radiation, surface air temperature, and surface

type; the first two determine the diurnal variation during the day. The M3DRY uses a

188 complex parameterization considering solar radiation, surface air temperature, vapor

189 pressure deficit, and water stress (Noilhan and Planton, 1989). In addition, the

190 vegetation fraction and leaf area index are used to account for the dependency of the

191 surface resistance on the surface type. We find that the assigned vegetation fraction

and leaf area index are the important factors for the stomata resistance calculation of
the M3DRY, and typically yield the resistance value of the M3DRY higher than that
of the Wesely.

The ground resistance is important at night and is calculated differently in the two schemes. We generally find that the M3DRY computes a value higher than the Wesely. For example, the former computes 1000 s m⁻¹ over cropland (the major surface type in China), whereas the latter calculates 350 s m⁻¹. This discrepancy results in a higher dry deposition velocity with the Wesely than that of the M3DRY at night.

201 The M3DRY that we implemented in WRF-Chem was a standalone package 202 that used a fixed value for a certain parameter such as water stress, depending on the 203 surface type for the stomata resistance calculation. However, the latest development 204 of the M3DRY uses the calculated stomata resistance from the Pleim-Xiu land surface 205 model in order to maintain the consistency with meteorological simulations toward an 206 online approach (Xiu and Pleim, 2001). Therefore, we also examine the effect of this 207 change (standalone versus online) on the simulated dry deposition velocities with the 208 M3DRY below. All the simulated results with the M3DRY below are from the model 209 with the standalone package except for Fig. 2, which compares the values from the 210 two applications of the M3DRY (standalone versus online).

211

212 2.4 Observations

213 We used observations from the Bio-hydro-atmosphere interactions of Energy,

214 Aerosols, Carbon, H₂O, Organics, and Nitrogen-Rocky Mountain Organic Carbon

215 Study (BEACHON-ROCS) campaign conducted at the Manitou forest observatory in

the United States by NCAR for August 7-31, 2010. Details on this campaign are at the

217 following website (<u>https://wiki.ucar.edu/display/mfo/Manitou+Forest+Observatory</u>).

- We used the gradient method from Tsai et al. (2010) to compute the measured ozone dry deposition velocity, as shown below. We first estimated ozone flux as a product of the friction velocity and the ozone eddy concentration. The ozone eddy
- 221 concentration (c^*) can be calculated using equation (4) as follows:

222
$$c^* = k\Delta c \left[\ln \left(\frac{z_0 - d_0}{z_1 - d_0} \right) - \Psi_h \left(\frac{z_2 - d_0}{L} \right) + \Psi_h \left(\frac{z_1 - d_0}{L} \right) \right]$$
(4),

223 where k is the von Karman constant, and Δc represents the ozone concentration 224 difference between two different observation levels, z_1 (12 m) and z_2 (25 m). d_0 is 225 the zero-plane displacement height, L is the Monin-Obukhov length, and integrated 226 stability function (Ψ_h) is from Businger et al. (1971). After calculating the ozone flux, 227 the dry deposition velocity was calculated by dividing the ozone flux by the ozone 228 concentration at level 2 (z_2) . Following the previous observation studies (Tsai et al., 229 2010; Matsuda et al., 2005), we used values only for a case in which 1) the ozone 230 concentration was greater than 1 ppbv, 2) the surface wind speed was greater than 1 m s^{-1} , and 3) a computed value was less than the maximum ozone dry deposition 231 velocity defined as 1.5 x $(R_a + R_b)^{-1}$. Finally the variation in zero-plane displacement 232 233 height (d_0) can generate a large uncertainty that is proportional to the vegetation 234 height (15 m at the Manitou forest observatory). We accounted for this variation by 235 applying linear coefficients that range from 0.55 to 0.78 for the vegetation height 236 (Garratt, 1994; Lovett and Reiners, 1986; Perrier, 1982). We computed a range of 237 measured dry deposition velocities with minimum and maximum linear coefficients. 238 We also used ozone dry deposition velocities directly measured using the eddy 239 covariance method at a Niwot Ridge AmeriFlux site in the Roosevelt National Forest 240 in the Rocky Mountains of Colorado for May 21-31, 2005 (Turnipseed et al., 2009). 241 Details for this site are at the following website:

242 http://ameriflux.ornl.gov/fullsiteinfo.php?sid=34.

243 As mentioned above, observed ozone dry deposition fluxes or ozone dry 244 deposition velocities are very limited in East Asia. Matsuda et al. (2005) provided the 245 observed ozone dry deposition velocities at a site (Mae Moh) in northern Thailand for 246 January-April 2002 based on their ozone flux measurements. Although the 247 measurements were made above a tropical forest that differed from the major surface 248 type of East Asia, we used their observations to evaluate simulated dry deposition 249 velocities in Section 3.

250 In addition, we used ozone concentrations in surface air observed at sites from 251 the National Institute of Environmental Research (NIER, http://www.nier.go.kr) in 252 Korea and from the Acid Deposition Monitoring Network in East Asia (EANET, 253 http://www.eanet.cc). The Korean sites are primarily located in polluted urban regions, 254 including Seoul, the capital of South Korea, and Pusan, the second largest city in 255 South Korea, whereas the EANET sites are primarily in islands, rural regions, and 256 mountains to avoid the direct influence from local pollution (Fig. 3). Ozone

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observations in China are not available to the public, which limits our discussion on

258 observed ozone spatial patterns. Therefore, we primarily focused on the downwind

259 regions of the continental pollution outflow, which was successfully used in the

260 previous analysis during the TRACE-P campaign to chemically characterize East

261 Asian environments (Jacob et al., 2003). The observations were averaged over the

262 model grid boxes for comparison with the model.

263

264 3. Ozone dry deposition velocity

265 Figure 1 compares the calculated monthly mean ozone dry deposition velocities 266 for May from the WRF-Chem simulations with the Wesely and M3DRY schemes for

East Asia. The values are typically high on the continent relative to the ocean, which reflects the decrease in the surface resistance owing to vegetation. However, as shown in Fig. 1c, we found substantial differences in calculated dry deposition velocities between the two schemes. The Wesely typically yields higher values compared with the M3DRY because of the lower surface resistances in the Wesely. The domain mean of the Wesely is 0.24 cm s⁻¹ and is by a factor of 2.4 higher than that of the M3DRY (0.10 cm s⁻¹), implying a more rapid ozone loss with the Wesely.

274 We evaluate the dry deposition velocities calculated using the two schemes by 275 comparing such values with the observations and primarily focusing on the diurnal 276 variability. The observations were acquired from the BEACHON ROCS and Niwot 277 Ridge AmeriFlux sites in Colorado, USA, and from the Mae Moh site in northern 278 Thailand. For this comparison, we additionally conducted WRF-Chem dry deposition 279 calculations with the two schemes at each observation site to obtain the simulated 280 ozone dry deposition velocities for the corresponding observation periods. The model 281 classifies surface types of the corresponding model grids to observation sites as shrub 282 land (BEACHON), evergreen needle leaf (Niwot Ridge), and cropland/pasture (Mae 283 Moh).

284 Figure 2 compares the hourly measured and simulated ozone dry deposition 285 velocities averaged for the observation periods at the BEACHON and the Niwot 286 Ridge sites in the United States and at the Mae Moh site in northern Thailand. The 287 measured values at the BEACHON ROCS site are high in the early morning and 288 decrease toward the afternoon, which reflects the friction velocity diurnal variation 289 that depends on solar radiation. The measured values from the AmeriFlux site also 290 show similar diurnal variation with a broad maximum during the daytime; the greatest 291 value is found in the afternoon. Compared to the values at the two US sites, the

observations in tropical northern Thailand show relatively sharp daytime variation
such that the peak appears in the early morning and a rapid decrease occurs afterward.
The different observation periods and vegetation types may contribute to the

295 dissimilar diurnal variation of the observations among the sites.

296 Figure 2 also presents the simulated results with the Wesely and the M3DRY. 297 The former appears to calculate values higher than the latter, particularly during the 298 day, and shows a larger diurnal variation. The large diurnal variation is a pronounced 299 observed feature at all three sites and is well captured by the Wesely, whereas the 300 M3DRY significantly underestimates the observations especially during the day. The 301 stomata resistance is the most dominant factor for determining the dry deposition 302 velocity during the day and is certainly better resolved in the Wesely than in the 303 M3DRY. Moreover, the underestimates of daytime values are consistently shown in 304 the two different M3DRY applications: standalone and online. In fact, the online 305 approach that uses the stomata resistance directly from the land surface model 306 performs slightly better than the standalone M3DRY for reproducing the daytime 307 values. Understanding this discrepancy is also important but beyond the scope of our 308 present work. We plan to examine this issue in the future study.

309 The largest discrepancy between the Wesely and the observation occurs at the 310 Mae Moh site where the model cannot capture the peak in the morning and 311 overestimates the observed values at night. As discussed above, the Mae Moh site is 312 located in the tropical forest (Matsuda et al., 2005), but the model grid corresponding 313 to the Mae Moh site is assigned as a cropland/pasture. We believe that the model 314 horizontal resolution is too coarse to properly represent the observation site in 315 northern Thailand and is likely the cause for the discrepancy between the model and 316 the observations.

317 Nevertheless, we find that the Wesely successfully reproduces the observed 318 diurnal variation and the daytime values and performs better than the M3DRY 319 particularly at the two US sites. We acknowledge that our evaluation is still too 320 limited to be applied for East Asia. However, the Manitou forest observatory is a 321 ponderosa pine plantation in the middle of shrub land (Kim et al., 2010), which is 322 prevalent in East Asia, especially in the middle of China (Fig. 5a). Therefore, our 323 evaluation provides limited but valid guidance of how the two dry deposition schemes 324 perform over the majority of the East Asian land. We emphasize here that our 325 evaluation does not represent East Asia in its entirety, and in-situ ozone dry 326 deposition velocity measurements thus are critical and necessary for enhancing our 327 understanding of ozone loss and modeling capability for East Asia.

328

329 4. Simulated ozone concentrations in East Asia

Figure 3 shows the observed and simulated monthly mean ozone concentrations in surface air over East Asia for May 2004. The observations show a spatial gradient in which the values at polluted urban sites in Korea are lower than those at clean rural sites in Japan. Ozone losses by the titration of high NO in large megacities explain this observed spatial pattern with low values in Korea.

The simulated ozone concentrations with the two schemes also show a similar spatial gradient, which is high over the downwind ocean and relatively low over the continent. The model generally captures the observed spatial pattern, but the simulated pattern is not as clear as the observation because the model spatial resolution is not fine enough to capture concentrated pollution plumes at urban sites in Korea and to delineate sharp coastline variation in Japan.

341 However, the most striking feature is that the simulated ozone concentrations 342 differ considerably between the two schemes such that the Wesely values are 343 significantly lower than those of the M3DRY. The simulated ozone difference 344 between the two schemes is up to 10 ppbv for the monthly mean and is 4.7 ppbv for 345 the domain mean (Table 3). The largest differences occur in the Yellow Sea and 346 northwestern Pacific. We find that the simulated ozone differences are spatially 347 inconsistent with the differences of the simulated dry deposition velocities between 348 the two schemes. As shown in Fig. 1, the largest difference of the simulated dry 349 deposition velocity appears on the continents, but the ozone concentrations difference 350 is the greatest over the downwind ocean. We think that this feature is caused by the 351 efficient ozone export from the polluted continent to the downwind oceans where 352 ozone accumulates (Goldberg et al., 2014). In addition, the ozone differences over the 353 ocean may significantly be attributed to excessively high surface water resistance 354 (low deposition loss) in the M3DRY relative to the Wesely. This issue is further 355 discussed in Section 5. The export of ozone precursors also contributes to high ozone 356 over the oceans, but is relatively minor compared with the direct ozone export.

357 Table 3 summarizes the simulated surface ozone concentration and ozone dry 358 deposition velocity averaged over the domain for May and June 2004, respectively, to 359 examine their seasonal variation from spring to summer. We do not find considerable 360 change in the simulated values between the two months except that the ozone dry 361 deposition velocity with the M3DRY slightly increases in June relative to May 362 because of the increase of the vegetation cover. However, the ozone concentration 363 remains the same in June compared with May because an increased ozone production 364 offsets the increased ozone loss through dry deposition.

Figure 4 shows the hourly mean observed and simulated ozone concentrations averaged at the NIER sites in Korea and EANET sites in Japan for May 2004. The simulated values are sampled from the corresponding model grids to the observation sites for this comparison. The diurnal variation differs between the two networks such that the observed ozone concentrations in Korea show a strong diurnal variation, a peak in the afternoon and a minimum at night, which reflects a direct influence from local pollution.

372 The model generally captures the observed diurnal variation, but also shows 373 considerable discrepancies from the observations (Fig. 4). For example, at the NIER 374 sites in Korea, the M3DRY overestimates the observations by 4.4-17.1 ppbv. This 375 high bias is reduced when we use the Wesely although the model still cannot capture 376 the lowest ozone concentration in the early morning, caused by the NO titration 377 during the rush hour traffic. We further examine this issue in Section 5. 378 On the other hand, the simulated ozone concentrations are lower than the 379 observations at the EANET sites. This low bias is consistently shown in the model 380 with both the Wesely and the M3DRY. The ozone differences between the two 381 methods are 4.6-5.1 ppbv, smaller than 5.4-7.4 ppbv at the NIER sites. Although the 382 M3DRY shows smaller biases than the Wesely, it is difficult to validate the dry 383 deposition simulation alone because the EANET sites are primarily located at the 384 coast where the ocean heavily influences the observed ozone concentrations. It is 385 known that the model and observation discrepancies at the coastal sites are caused by 386 the model's inability to simulate steep sub-grid land-to-sea gradients at a mixing

depth (Gao and Wesely, 1994; Loughner et al., 2011) that is shallower over the ocean

388 compared with the continent. Our model with 45 x 45 km spatial resolution may not

adequately represent the shallow mixing depth at the EANET sites.

Although the model reproduces the certain observed features as shown in the comparisons in Figs. 3 and 4, it is difficult to determine the scheme with the best performance for the observed ozone concentrations in East Asia. However, as discussed in Section 3, the model with the Wesely reproduced the observed dry deposition velocities better than the M3DRY. Therefore, we use the Wesely results for our subsequent analysis below, where we examine the simulated sensitivity to other input parameters.

397

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5. Effect of surface-type uncertainty on ozone concentrations

399 The spatial distribution of the dry deposition velocity closely resembles that of 400 the land-use data, implying that the dry deposition simulation may be highly sensitive 401 to the use of the land-use data. The WRF-Chem typically employs the land-use data 402 from the United States Geological Survey (USGS) as a default option (Table 4). Here 403 we explore the model sensitivity to the land-use data using the USGS and the MODIS 404 land-use data (Friedl et al., 2002), which are widely used in meteorological research. 405 In order to use the MODIS data, we developed a mapping table between the two datasets (Table 5), which was used to implement the MODIS land-use data in the 406 407 WRF-Chem simulations below. 408 Figure 5 shows the USGS and the MODIS land-use data. In general, 409 vegetation types identified by the two datasets are generally consistent for East Asia, 410 but we find certain differences as well, especially for south China. One notable 411 difference is that the USGS classifies the Korean peninsula as savanna, which differs 412 from the MODIS classification (mixed forest). The different surface-type 413 classifications affect ozone dry deposition calculations in the model as discussed 414 below.

415 Figure 6 shows the differences of dry deposition velocities and ozone 416 concentrations in the model using the two land-use datasets: MODIS and USGS. Here 417 we use the Wesely of which the simulated dry deposition velocities were consistent 418 with the observations and were more sensitive to surface types than the M3DRY. The 419 simulated differences of the dry deposition velocities reflect the different surface-type 420 classifications between the two datasets. We find lower dry deposition velocities for 421 East Asia using the MODIS compared with values with the USGS. The largest 422 discrepancy occurs in southern China where the surface type was changed from 423 cropland/pasture, cropland/grassland mosaic, shrubland, and savanna to mixed forest 424 (Fig. 5). This surface-type change increased the surface resistances and thus decreased 425 the dry deposition velocity. On the other hand, the calculations in Manchuria and 426 Republic of the Union of Myanmar showed increased dry deposition velocities 427 because the surface types there were changed from mixed forest to cropland/pasture 428 or evergreen broadleaf.

429 The change of the land-use data from the USGS to the MODIS results in an 430 increase of the monthly mean ozone concentration by 10.2 ppbv in southern China 431 and the downwind regions, including Korea, Japan and the north Pacific for May. The 432 average ozone concentration over the domain is increased with the MODIS land-use 433 data by 1.3 % compared with the USGS data. This change seems negligible, but in the 434 urban and industrialized regions the ozone increase with the MODIS data is much 435 greater by 5.1 ppbv (13 %) compared with the USGS data, indicating the considerable 436 sensitivity of ozone simulations to the surface-type classification.

The simulated sensitivity is also shown in the comparison of the hourly mean
ozone concentrations at the NIER sites in Korea (Fig. 7). We find an increase of
ozone concentrations averaged at all the sites by 3.9 ppbv simply by changing the

surface type from savanna to mixed forest, urban and built-up land. The model with
the MODIS performs slightly worse than that with the USGS, but the model spatial
resolution was still too coarse to represent surface-type inhomogeneity at the sites in
Korea, which are primarily in urban regions. The surface-type sub-grid scale
variability may also be a potentially important source for model uncertainty. On the
other hand, the model shows minimal changes in ozone at the EANET sites located
near the sea.

447 We further examine the sensitivity of the simulated ozone to the different surface water resistances in the dry deposition schemes. The Weselv used 2000 s m⁻¹ 448 for the water resistance, which was lower than the value of the M3DRY $(10^5 \sim 10^6 \text{ sm}^-)$ 449 450 ¹). We conduct a model simulation using the Wesely by switching the water surface 451 resistance from the Wesley to the M3DRY values. Figure 8 shows the resulting 452 differences of the ozone dry deposition velocities and ozone concentrations. The dry deposition velocity largely increases up to 0.043 cm s⁻¹ and causes an ozone decrease 453 454 as low as 8.7 ppbv over the ocean. This change explains 76% of the previous overall 455 ozone concentration difference between the two schemes over the ocean. Although 456 the ozone dry deposition loss is lower over the ocean compared with the continent, 457 this result indicates that the model is highly sensitive to the water surface resistance, 458 which has an important implication for estimating long-range ozone transport from a 459 source to a downwind region.

Finally, we conduct a nested model simulation using a finer spatial resolution (15 km) focusing on the Korean peninsula to examine the effect of NO titration on ozone concentrations in polluted urban cities. Figure 9 compares the simulated ozone concentrations from the nested model with the observations at the NIER sites in Korea. With the finer spatial resolution, the nested model yields lower ozone

465 concentrations by the enhanced NO titration because the concentrated NO emissions 466 are better represented in the nested model compared with the coarse model. We find 467 that the greatest reduction occurs in the early morning when the NO emission from 468 the rush hour traffic is the greatest. However, the high bias for the early morning 469 remains in the model, suggesting that the 15 km resolution is still too coarse to 470 represent the concentrated plume from traffic.

471

472 6. Conclusions

473 We used the WRF-Chem model with the two widely used dry deposition 474 schemes (Wesely and M3DRY) to evaluate the dry deposition simulations and to 475 examine the sensitivity of the simulated surface air ozone concentrations to dry 476 deposition calculations for East Asia. We found significant differences in ozone 477 concentrations up to 10 ppbv for the monthly mean, primarily driven by the dry 478 deposition velocity differences between the two schemes. The Weselv generates two-479 fold greater dry deposition velocity compared with the M3DRY under identical 480 meteorological conditions because of the discrepancies in the surface resistance 481 parameterization.

482 We compared the simulated dry deposition velocities with the observations 483 from the BEACHON-ROCS campaign and the Niwot Ridge Ameriflux sites in the 484 U.S. and from the Mae Moh site in northern Thailand. The Wesely generally 485 computed dry deposition velocities higher than the M3DRY and successfully 486 reproduced the observed diurnal variation. The Wesely also reproduced the observed 487 ozone concentrations at the polluted urban sites in Korea, but failed to capture the 488 observations at the clean sites in Japan, indicating the existence of other important 489 factors for background ozone simulations in East Asia.

| 490 | We conducted several sensitivity simulations using the different land-use |
|-----|--|
| 491 | datasets, water surface resistances, and model spatial resolutions to examine the |
| 492 | uncertainty of ozone simulations for East Asia. The model results showed |
| 493 | considerable changes in the simulated ozone concentrations, which suggested that the |
| 494 | model was highly sensitive to such input data and the model resolution. The need for |
| 495 | in-situ observations is high to constrain the dry deposition parameterization and its |
| 496 | input data to improve the use of air quality models for East Asia. |
| 497 | The roles of vegetation have primarily been discussed for reactive biogenic |
| 498 | volatile organic compounds (BVOCs) emissions and tropospheric photochemistry that |
| 499 | enhances ozone production in East Asia (Kim et al., 2013; Bao et al., 2010; Ran et al., |
| 500 | 2011; Tie et al., 2013). The comprehensive evaluation of dry deposition schemes |
| 501 | herein clearly indicates that deposition is also a critical physical process, which must |
| 502 | be precisely constrained in regional and global air quality assessments because ozone |
| 503 | has tremendous implications for public health (Levy et al., 2001) and climate change. |
| 504 | In addition, a number of experimental studies have clearly suggested that a substantial |
| 505 | level of unknown/unobserved reactive BVOCs may enhance non-stomatal ozone dry |
| 506 | deposition rates (Kurpius and Goldstein, 2003; Hogg et al., 2007), which should be |
| 507 | further examined using an improved modeling and extensive observations. |
| | |

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- 666 667

Tables

Table 1. Model set-up for the WRF-Chem simulations

| | Feature | | Selected configur | ration | | |
|-----|----------------|---------------------------|--------------------------------|---|--|--|
| | Domain | | East Asia on 45 k | m grid with 14 layers | | |
| | Domain top | | 50 hPa | e s | | |
| | Emission | | SMOKE-ASA (Only anthropogenic) | | | |
| | Longwave rad | liation | RRTM | RRTM | | |
| | Shortwave rad | diation | Goddard | | | |
| | Microphysics | | Lin (Purdue) | | | |
| | Cumulus para | meterization | Grell-Devenyi | | | |
| | Vertical diffu | sion | Eddy | | | |
| | Chemical mee | chanism | CBMZ | | | |
| | Surface layer | physics | Monin-Obukhov | | | |
| | Land surface | model | Noah | Noah | | |
| | Planetary bou | ndary layer | YSU | YSU | | |
| | Photolysis | | Fast-J | | | |
| 671 | | | | | | |
| 672 | | | | | | |
| 673 | Table 2. Spec | ies mapping between the | CB05 and CBMZ cher | nical schemes | | |
| | CBMZ (WRF | CB05 | CBMZ | CB05 | | |
| | Chem) | | | | | |
| | E ALD | ALD2+ALDX | E TOL | TOL | | |
| | ECO | СО | EXYL | XYL | | |
| | EOL2 | ETH | EETH | ETHA | | |
| | E_HCHO | FORM | E_C2H5OH | ЕТОН | | |
| | E_ISOP | ISOP | E_OLI | IOLE | | |
| | E_NH3 | NH3 | E_CH3OH | MEOH | | |
| | E_NO | NO | | NASN | | |
| | E_NO2 | NO2 | | TERP | | |
| | E_OLE | OLE | E_KET | | | |
| | E_PAR | PAR | E_ORA2 | | | |
| | E_SO2 | SO2 | E_CLS | | | |
| 674 | * NASN, TEI | RP, E KET, E ORA2, an | d E CLS have no corr | esponding species. | | |
| 675 | , | , <u> </u> | - | | | |
| 676 | | | | | | |
| 677 | | | | | | |
| 678 | | | | | | |
| 679 | Table 3. Surfa | ace ozone concentration (| ppbv) and ozone dry de | eposition velocity (m s ⁻¹ , | | |
| 680 | value in parer | theses) in May and June | 2004 | | | |
| | W | esely M3DRY | | | | |
| | May 31.4 | (0.24) 36.1 (0.10) | | | | |
| | June 32.2 | (0.24) 36.1 (0.12) | | | | |
| | | ()() | | | | |

Table 4. USGS 24 land-use data categories. Land Use Category Land Use Description Urban and Built-up Land Dryland Cropland and Pasture Irrigated Cropland and Pasture Mixed Dryland/Irrigated Cropland and Pasture Cropland/Grassland Mosaic Cropland/Woodland Mosaic Grassland Shrubland Mixed Shrubland/Grassland Savanna Deciduous Broadleaf Forest Deciduous Needleleaf Forest **Evergreen Broadleaf** Evergreen Needleleaf Mixed Forest Water Bodies Herbaceous Wetland Wooden Wetland Barren or Sparsely Vegetated Herbaceous Tundra Wooded Tundra Mixed Tundra Bare Ground Tundra Snow or Ice

Table 5. Land-use mapping between the 20-category IGBP-Modified MODIS and 24-

709 category USGS schemes

| MODIS | USGS | MODIS | USGS |
|------------------------------|--------------------------------|-------|------|
| Evergreen Needeleleaf Forest | Evergreen Needleleaf | 1 | 14 |
| Evergreen Broadleaf Forest | Evergreen Broadleaf | 2 | 13 |
| Deciduous Needleleaf Forest | Deciduous Needleleaf Forest | 3 | 12 |
| Deciduous broadleaf Forest | Deciduous Broadleaf Forest | 4 | 11 |
| Mixed Forest | Mixed Forest | 5 | 15 |
| Closed Shrubland | Shrubland | 6 | 8 |
| Open Shrubland | Mixed Shrubland/Grassland | 7 | 9 |
| Woody Savanna | Savanna | 8 | 10 |
| Savanna | Savanna | 9 | 10 |
| Grassland | Grassland | 10 | 7 |
| Permanents Wetland | Herbaceous Wetland | 11 | 17 |
| Cropland | Irrigated Cropland and Pasture | 12 | 3 |
| Urban and Built-up | Urban and Built-up Land | 13 | 1 |
| Cropland /Natural Mosaic | Cropland/Grassland Mosaic | 14 | 5 |
| Snow and Ice | Snow or Ice | 15 | 24 |
| Barren or Sparsely Vegetated | Barren or Sparsely Vegetated | 16 | 19 |
| Water | Water Bodies | 17 | 16 |
| Wooded Tundra | Wooded Tundra | 18 | 21 |
| Mixed Tundra | Mixed Tundra | 19 | 22 |
| Barren Tundra | Bare Ground Tundra | 20 | 23 |

730 Figure Captions

731

Figure 1. Monthly mean O₃ dry deposition velocities in East Asia for May 2004 from
WRF-Chem using the Wesely (left) and M3DRY (middle). The differences between
the two simulations are shown in the right panel.

735

736 Figure 2. A comparison of the simulated and observed hourly mean O₃ dry deposition velocities from the BEACHON-ROCS campaign at the Manitou forest observatory 737 738 for Aug. 07-31, 2010 (left panel), at the Niwot Ridge AmeriFlux site in the Roosevelt 739 National Forest in the Rocky Mountains of Colorado for May 21-31, 2005 (middle 740 panel) in the United States, and at Mae Moh site in Northern Thailand for Jan-Apr 741 2002 (right panel). The circles show observed values. The triangles, squares, and 742 diamonds show the simulated values using the Wesely, the M3DRY with standalone 743 stomata resistance, and the M3DRY with stomata resistance of the Pleim-Xiu land 744 surface model, respectively. The shaded area indicates the observed dry deposition 745 velocity range for the various zero-plane displacement heights (d_0) in equation 4 from 746 the BEACHON-ROCS campaign.

747

Figure 3. Monthly mean O₃ concentrations in surface air over East Asia for May 2004.
The left and middle panels show results from the WRF-Chem model using identical
emissions and meteorological input data but different dry deposition schemes, (a)
Wesely and (b) M3DRY. Observations from the NIER and EANET sites are denoted
with colored closed circles. The O₃ concentration differences between the two
simulations are shown in the right panel (c).

754

Figure 4. Hourly mean O₃ concentrations averaged over (a) the NIER sites (left) and (b) EANET sites (right) for May 2004. The simulated values were sampled from the model grids that correspond to the site locations. The observations are denoted with open circles, and the simulated values with the Wesely and the M3DRY are shown using pluses and triangles, respectively.

760

Figure 5. Land-use data from the USGS (left) and MODIS datasets (right). The colorcoding scheme used to denote the different surface types are consistent for the
datasets and follow the USGS dataset coloring (Table 4). We used the mapping
information (Table 5) to illustrate the MODIS data.

765

Figure 6. Differences in dry deposition velocity (left) and monthly mean O₃
concentration in the surface air (right) between the MODIS and USGS land-use data
using the Wesely scheme for May 2004.

769

Figure 7. Same as in Figure 4 but the simulated O₃ concentrations were generated
using the USGS (pluses) and MODIS land-use data (diamonds) with the Wesely

- scheme.
- 773

- Figure 8. Differences in monthly mean O₃ dry deposition velocities (left) and monthly
- mean O₃ concentrations in surface air (right) between the default and sensitivity
- simulations. The sensitivity simulation was conducted using the Wesely scheme and
- replacing the ocean surface resistance with the values from the M3DRY scheme for
- 778 May 2004.
- 779
- Figure 9. Hourly mean O₃ concentrations averaged over the NIER sites (left) for May
- 781 2004. The pluses and squares indicate results from the default (45 x 45 km) and
- nested models (15 x 15 km), respectively. The observations are denoted with the open
- circles. The differences between the two models are shown in the right panel.