

Responses to the Comments by J. Pleim

(1) This manuscript has some serious problems that lead to very misleading conclusions. The manuscript reports that the dry deposition model in WRF-Chem based on Wesely (1989) produces more accurate ozone dry deposition velocities than the M3Dry model that is used in the CMAQ model. This conclusion is based solely on Figure 2 that shows hourly measured ozone dry deposition velocities from two field studies in Colorado and model results from Wesely and M3Dry. There is extremely little explanation of this comparison. Are these results averaged over many days or weeks? For what time periods?

→ We obtain the hourly results by averaging the data for the observation period. The simulated and observed hourly mean concentrations at the BEACHON site are for Aug. 07-31, 2010, and the values at the Niwot Ridge Ameriflux site are for May 21-31, 2005, and the values at a site in northern Thailand are for Jan-Apr, 2002.

(2) Are the time periods the same for both sites? How were the model results produced?

→ The periods differ at different sites as shown above. We conducted a separate WRF-Chem dry deposition calculation at each site.

(3) The only modeling discussed is for WRF-Chem applications in Asia. Are the results shown in Fig 2 from WRF-Chem in Colorado? Are they box-model results using measured inputs, if so, from which site? **These sites are about 100 km apart and are presented as if they are directly comparable.**

→ We used the simulations for each observation site. We show a comparison at each site in the different panel in the revised manuscript.

(4) All these questions need to be answered if this Figure is to be taken as evidence of model performance.

→ Yes we modified our manuscript to answer to all the questions that you raised as follows:

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

Figure 2 compares the hourly measured and simulated ozone dry deposition velocities averaged for the observation periods at the BEACHON and the Niwot Ridge sites in the United States and at the Mae Moh site in northern Thailand. The measured values at the BEACHON_ROCS site are high in the early morning and decrease toward the afternoon, which reflects the friction velocity diurnal variation that depends on solar radiation. The measured values from the AmeriFlux site also show similar diurnal variation with a broad

maximum during the daytime; the greatest 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 dissimilar diurnal variation of the observations among the sites.

(5) Another major problem is the lack of explanation about how the M3Dry is implemented in WRF-Chem. The article cited for M3Dry (Pleim et al 2001) presents the dry deposition model as part of a coupled land surface model (LSM) for meteorology and chemistry. This article, and a more recent article by Pleim and Ran (2011), both explain that a key advantage of M3Dry over stand-alone dry deposition models such as the Wesely model is that the stomatal conductance and several other parameters are used directly from the LSM in the meteorology model. In this way, the stomatal pathway for dry deposition is proportional (scaled by the ratio of the chemical diffusivity to the diffusivity of water) to the stomatal conductance used to compute transpiration for surface moisture flux in the meteorology model. In the most recent model versions, **dry deposition velocities are computed in CMAQ (M3Dry was removed from MCIP in 2011) using stomatal conductance that is output from the meteorology model.** Thus the stomatal pathway for dry deposition in M3Dry is as good or bad as the stomatal pathway for evapotranspiration. If the stomatal conductance is not realistic, the meteorology simulation will not be accurate.

→ Yes we added a following paragraph in Section 2.3 to explain how we implemented M3DRY in the WRF-Chem simulations as follows:

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

(6) Prior to 2012, MCIP did include an option for a standalone stomatal conductance calculation for use if this parameter was not available from the meteorology model output. This option was never used at the USEPA (the developers of CMAQ) since stomatal conductance is a standard output from WRF when using the Pleim-Xiu LSM. The stomatal conductance from other LSM options in WRF can also be used. Since this manuscript does not explain how the stomatal conductance was computed for the WRF-Chem modeling presented, I guess that the alternate standalone stomatal calculation was probably used. This is not the preferred way to apply M3Dry and is in fact no longer available in recent CMAQ versions. I strongly suggest that the M3Dry be applied as intended using the stomatal conductance and other parameters such as aerodynamic conductance from the LSM in WRF. **This should not be difficult since WRF-Chem is an online met-chem model.** The results shown here provide no valid basis for concluding that either M3Dry or Wesely are better for calculating ozone dry deposition velocity. If, as I suspect, the stomatal conductance for the M3Dry model runs did not use the stomatal conductance from the WRF LSM (in this case Noah), the results are not representative of the model's typical performance since it used an

option that is not recommended, never used in EPA applications of CMAQ, and is no longer supported or available.

→ We appreciated very much for your constructive comments. In our manuscript that you reviewed, we used an option for a standalone stomatal conductance calculation, which is a function of the surface type. The Wesely method also uses its own standalone parameterization of stomatal conductance. The difference between these two standalone parameterizations is the key for the discrepancy of simulated O₃ dry deposition velocities and O₃ concentrations. But as you pointed out that the preferred way of applying M3DRY is to use stomatal conductance from LSM models. Therefore, following your suggestion we modified the WRF-Chem code and used the calculated stomatal conductance from the Pleim-Xiu LSM model in the M3DRY. We discuss this in the revised manuscript as follows:

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

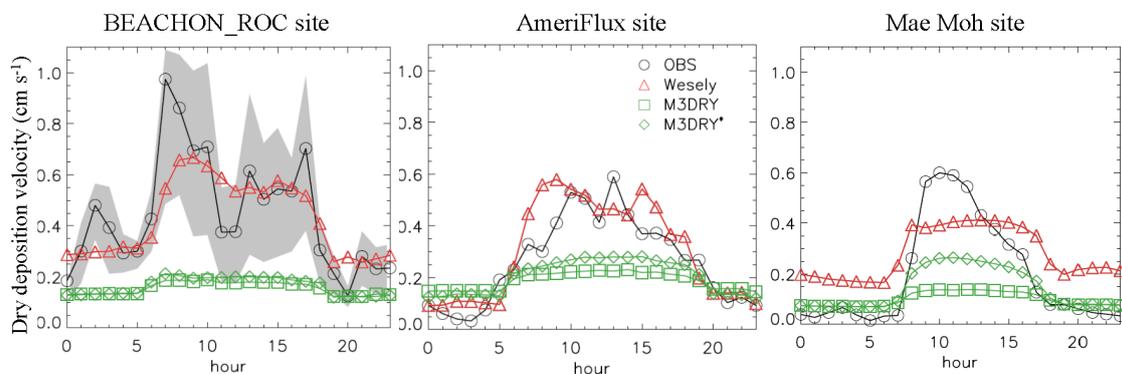


Figure 2. A comparison of the simulated and observed hourly mean O₃ dry deposition velocities from the BEACHON-ROCS campaign at the Manitou forest observatory for Aug. 07-31, 2010 (left panel), at the Niwot Ridge AmeriFlux site in the Roosevelt National Forest in the Rocky Mountains of Colorado for May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

Responses to the Comments by anonymous reviewer #1

(1) In-situ ozone dry deposition velocity measurements in Asia: The authors state that their evaluation is limited due to the lack of observations of ozone deposition velocity in East Asia. I would generally agree with this statement. However, Matsuda et al (2005) (as referenced by the authors) presented measurement of ozone dry deposition in deciduous (teak) forest in northern Thailand. The observed diurnal cycle of dry deposition velocity is shown in Figure 3. I wonder why this data are not included in Figure 2. I would suggest the authors to consider this observation in comparison with their model, and modify the sentences accordingly.

→ Thanks for the constructive comment. We added a comparison at the Mae Moh site in northern Thailand in the revised manuscript as shown below.

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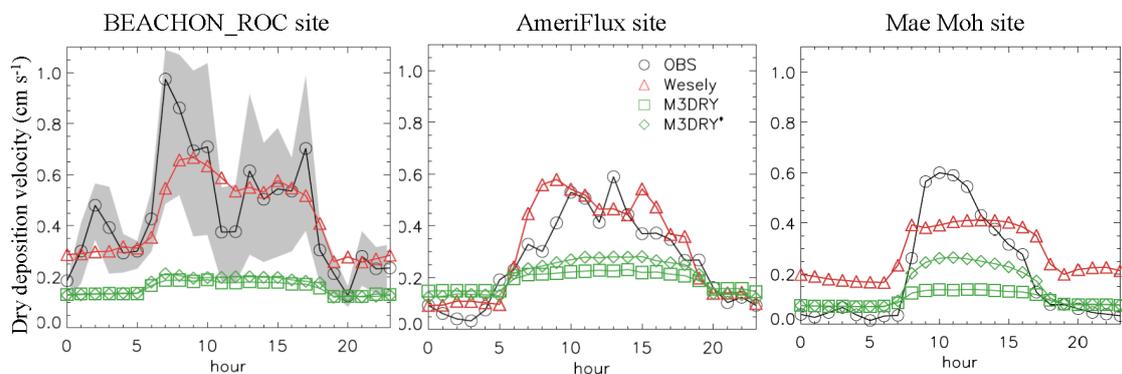


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(2) Seasonal variations of ozone dry deposition: The authors mainly discuss deposition in spring with an example in May 2004. Why is it? Do the authors have any specific reasons?

→ *Yes as we wrote in the manuscript, May is the ozone peak month in East Asia.*

As it noted in the manuscript, it seems that stomata plays a dominant role in determining the total deposition velocity, particularly during photosynthetically active period. I presume that the dry deposition would have seasonal variations. Is the impact by dry deposition greater in warm seasons than in cold ones? Is the impact greater in summer than in spring? Well, greater but not much, maybe? I am interested in how dry deposition perturbs the balance of photochemical production and long-range transport from spring to summer seasons, since these two are competing factors in controlling distributions of surface ozone in East Asia. For example, photochemical production is stronger in summer than in spring, while transport from Asian continent to downwind happens more efficiently in spring than in summer. I would like to see some analysis for the seasonal variations of ozone dry deposition and its roles in seasonal cycles of ozone in East Asia.

→ *Following your suggestion, we examined the monthly change of dry deposition velocity and ozone concentrations from May to June. After June, the Asian summer monsoon significantly affects ozone in East Asia so we did not focus our analysis on summer. The discussion of the monthly change is added in the revised manuscript as follows:*

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

Table 3. Surface ozone concentration (ppbv) and ozone dry deposition velocity (m s^{-1} , value in parentheses) in May and June 2004

	Wesely	M3DRY
May	31.4 (0.24)	36.1 (0.10)
June	32.2 (0.24)	36.1 (0.12)

L17: observations of “flux” or “dry deposition”?

→ *We clarified it as “ozone flux observations”.*

Figures 3 and 4: NIER and EANET sites. Are these plots averages at several sites or at specific single sites? Many of the EANET sites are located at island but the characteristics are greatly different between north and south (Tanimoto et al., GRL, 2005). Diurnal cycles are observed in summer at some sites, for example, at Rishiri. Please be more specific to the locations and seasons of the observations. Also, any references or websites for the NIER sites?

→ *We clarified our discussion on the surface O₃ sites and how we perform the comparison in*

the revised manuscript as follows:

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.

Page 929, L9-12: How large is the difference between the two schemes?

→ We revised the paragraph as follows:

The model generally captures the observed diurnal variation, but also shows considerable discrepancies from the observations (Fig. 4). For example, at the NIER sites in Korea, the M3DRY overestimates the observations by 4.4-17.1 ppbv. This high bias is reduced when we use the Wesely although the model still cannot capture the lowest ozone concentration in the early morning, caused by the NO titration during the rush hour traffic. We further examine this issue in Section 5.

On the other hand, the simulated ozone concentrations are lower than the observations at the EANET sites. This low bias is consistently shown in the model with both the Wesely and the M3DRY. The ozone differences between the two methods are 4.6-5.1 ppbv, smaller than 5.4-7.4 ppbv at the NIER sites. Although the M3DRY shows smaller biases than the Wesely, it is difficult to validate the dry deposition simulation alone because the EANET sites are primarily located at the coast where the ocean heavily influences the observed ozone concentrations. It is known that the model and observation discrepancies at the coastal sites are caused by 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 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.

Figure 8: Why is the difference larger in marginal sea off the coast of Asian continent, compared to open oceans? Any comments?

→ We added the explanation as follows:

As shown in Fig. 1, the largest difference of the simulated dry deposition velocity appears on the continents, but the ozone concentrations difference is the greatest over the downwind ocean. We think that this feature is caused by the efficient ozone export from the polluted continent to the downwind oceans where ozone accumulates because of inefficient dry depositional loss (Goldberg et al., 2014). The export of ozone precursors also contributes to high ozone over the oceans, but is relatively minor compared with the direct ozone export. In addition, the ozone differences up to 8.7 ppbv over the ocean may partially be attributed to excessively high surface water resistance (low deposition loss) in the M3DRY relative to the Wesely, which is not clearly shown in Fig. 1. This issue is discussed in Section 5.

Responses to the Comments by anonymous reviewer #2

(1) Review of Park et al., An evaluation of O₃ dry deposition simulations in East Asia

This study evaluates and compares two dry deposition schemes used for model simulations of atmospheric ozone. Considering that dry deposition is a very important process affecting ozone levels and also the fact that relatively little studies in the literature have focused on ozone dry deposition, I think this is a topic suitable for publication at ACP. I would recommend publication of this MS after the authors address some relatively minor issues – 1. L194-196, “Herein, we only considered the measured dry deposition velocities in the range 0 to 2.0 cm s⁻¹, which is a typical O₃ dry deposition velocity range in the literature (Padro, 1996).”

So do you mean that the measurement data for dry deposition velocities outside of this range is unrealistic? I think better clarification/justification is needed here.

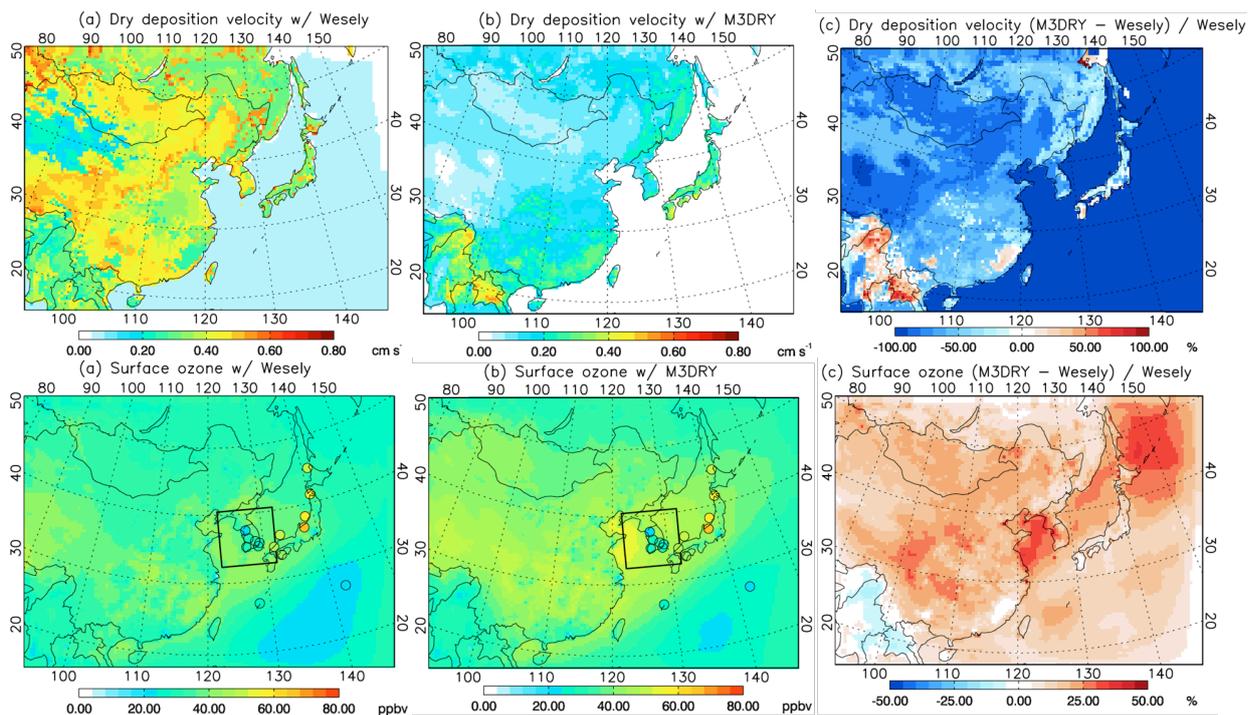
→ We adopt the new criteria to select the observed values based on the previous literature as follows:

Following the previous observation studies (Matsuda et al., 2005; Tsai et al., 2010), we used values only for a case in which 1) the ozone 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 velocity defined as $1.5 \times (R_a + R_b)^{-1}$.

(2) L275-282 on the discussion of the feather that the largest differences in O₃ dry deposition velocity over the continents do not result in the largest differences in ozone concentration there but rather over the downwind ocean, the authors attribute this to the efficient export of O₃ from the polluted continent. I think it would be more clear if the authors can show the ratio plots for O₃ concentrations as well as the dry deposition velocities (i.e. similar to Fig. 1c and 3c, but shown as ratios instead of differences). In addition, I'm thinking perhaps the changes in O₃ precursors (I assume the two dry dep. schemes were applied to not only O₃ but other species in the model) also contribute to this?

→ Following the suggestion, we made ratio plots as shown below, but decided to keep the difference plot in the manuscript because the ratio plot shows too much saturation over the ocean owing to the low values in the denominator. In addition, the absolute change of ozone can provide quantitative information for the ozone air quality criteria and thus be more useful we believe.

→ As to the export of ozone precursors, their contributions are relatively minor compared with the direct ozone export and we added this sentence in the revised manuscript.



(3) L37-38, “the model was considerably sensitive to the input parameters, which indicates a high uncertainty for such O3 dry deposition simulations.” –It is unclear to me how can we reach this conclusion, please clarify here.

→ We rewrote the sentence for clarity as follows:

However, we find significant changes in simulated ozone concentrations using the Wesely scheme but with different surface type datasets, indicating the high sensitivity of ozone deposition calculations to the input data.

(4) Some edits/proofreading is needed for this MS; I only tried on the 1st few pages as shown below for examples, but I think the authors need to check throughout their MS –

- 1). L45 O3 tropospheric => tropospheric O3
- 2). L65 air quality sources => air pollution sources
- 3). L76 in the most widely => in two of the most widely
- 4). L79 as well as two => as well as the two
- 5). L85 assessing the spatial and temporal distribution of O3 and the contribution from a specific source
- 6). L233-235, “The dry deposition velocity domain mean difference between the two methods is 0.14 cms-1, which is 1.4 greater than the M3DRY method domain mean dry deposition velocity (0.10 cm s-1)”
This sentence is very confusing and need rewriting.
- 7). L261. 4. O3 concentration spatial and diurnal patterns in East Asia => spatial and diurnal patterns of ozone concentrations in East Asia
- 8). L557 Table 1 lists not only “Physics” parameters but also Chemical mechanisms, so I would suggest changing to “Model set up for the WRF-Chem simulations”

→ We rewrote all the misused words and had a native speaker check out our manuscript to improve the use of words and to correct grammatical errors.