A Diagnostic Intercomparison of Modeled Ozone Dry Deposition Over North America and Europe Using AQMEII4 Regional-Scale Simulations

Christian Hogrefe[1], Stefano Galmarini[2], Paul A. Makar[3], Ioannis Kioutsioukis[4], Olivia E. Clifton[5,6], Ummugulsum Alyuz[7], Jesse O. Bash[1], Roberto Bellasio[8], Roberto Bianconi[8], Tim Butler[9], Philip Cheung[3], Alma Hodzic[10], Richard Kranenburg[11], Aurelia Lupascu[12], Kester Momoh[7], Juan Luis Perez-Camanyo[13], Jonathan E. Pleim[1], Young-Hee Ryu[14], Roberto San Jose[13], Martijn Schaap[11], Donna B. Schwede[1,*], and Ranjeet Sokhi[7]

[1]Center for Environmental Measurement and Modeling, US Environmental Protection Agency, 109 T.W. Alexander Dr., P.O. Box 12055, RTP, NC 27711, USA
[2]JRC, European Commission, Ispra, Italy
[3]Environment and Climate Change Canada, Toronto, Canada
[4]Department of Physics, University of Patras, Patras, Greece
[5]Center for Climate Systems Research, Columbia University, New York, NY, USA
[6]NASA Goddard Institute for Space Studies, New York, NY, USA
[7]Centre for Climate Change Research (C3R), U. Hertfordshire, UK
[8]Enviroware srl, Concorezzo, MB, Italy
[9]Research Institute for Sustainability – Helmholtz Centre Potsdam, Germany
[10]NCAR, Boulder, CO, USA
[11]TNO, Utrecht, the Netherlands
[12]ECMWF, Bonn, Germany
[13]Technical University of Madrid (UPM), Madrid, Spain
[14]Yonsei University, Seoul, South Korea
[*]retired

Correspondence to: Christian Hogrefe (hogrefe.christian@epa.gov)

Abstract. This study analyzes ozone (O_3) dry deposition fluxes and velocities (V_d) from regional-scale simulations that were performed over North America and Europe in Phase 4 of the Air Quality Model Evaluation International Initiative (AQMEII4). AQMEII4 collected grid-aggregated and land use (LU)-specific O_3 V_d and effective conductances and fluxes for the four major dry deposition pathways. Consistent with recent findings in the AQMEII4 point model intercomparison study, analysis of the grid-aggregated fields shows that grid models with similar V_d can exhibit significant differences in the absolute and relative contributions of the different depositional pathways. Analysis of LU-specific V_d and effective conductances reveals a general increase in model spread compared to grid-aggregated values. This indicates that an analysis of only grid-aggregated deposition diagnostics can mask process-specific differences that exist between schemes. An analysis of AQMEII4 LU distributions across models revealed substantial differences in the spatial patterns and abundance of certain LU categories over both domains, especially for non-forest partially vegetated categories such as agricultural areas, shrubland, and grassland. We

demonstrate that these differences can contribute to or even drive differences in LU-specific dry deposition fluxes. Two recommendations for future deposition-focused modeling studies emerging from the AQMEII4 analyses presented here are to 1) routinely generate diagnostic outputs to advance a process-based understanding of modeled deposition and support impact analyses, and 2) recognize the importance of documenting and analyzing the representation of LU across models and work towards harmonizing this aspect when using air-quality grid models and model ensembles for deposition analyses.

1 Introduction

Dry deposition to the surface is one of the largest sinks in the budget of tropospheric ozone (O_3) (Wild, 2007) and can be a key contributor to O_3 variability (Clifton et al., 2020a; Lin et al., 2020; Baublitz et al., 2020). As summarized in Hardacre et al. (2015) based on previous global modeling studies (Stevenson et al., 2006; Wild, 2007; Ganzeveld et al., 2009; Young et al., 2013), the magnitude of this sink has been estimated to be on the order of 1000 Tg/yr, with about 2/3 of the dry deposition flux occurring over land and the remaining 1/3 occurring over oceans. Most global and regional chemistry transport models (CTMs) employ resistance frameworks to represent O_3 dry deposition, such as the frameworks introduced in Wesely (1989) and Zhang et al. (2003). In these frameworks, dry deposition fluxes are represented as the product of the O_3 mixing ratio and a deposition velocity (V_d) which in turn is represented as a combination of process-specific resistances acting in parallel or in series to impede the transfer of mass to the surface. However, the implementation of these resistance frameworks can differ significantly across models, especially with respect to representing the surface resistance and its split between the stomatal and non-stomatal components (Hardacre et al., 2015; Wu et al., 2018; Wong et al., 2019; Clifton et al., 2020b and 2023; Galmarini et al., 2021).

The ability of CTMs to provide spatially continuous fields of O_3 mixing ratios, V_d, and dry deposition fluxes has led to their use in numerous studies quantifying ecosystem damages due to O_3 dry deposition. In some studies, ecosystem impacts were calculated using O_3 mixing ratios simulated by global CTMs in conjunction with crop distribution datasets and literature-based concentration-response relationships without explicitly taking into account modeled stomatal dry deposition fluxes (Wang and Mauzerall, 2004; Van Dingenen, 2009; Avery et al., 2011). In a study focused on assessing O_3 dry deposition in global models, Hardacre et al. (2015) analyzed fields of monthly grid-aggregated O_3 dry deposition fluxes from 15 CTMs that had participated in the first Task Force on Hemispheric Transport of Air Pollution (TF HTAP) model intercomparison project (Fiore et al., 2009). Because the native land use (LU) databases used by each model were not available, the study overlaid the modeled fluxes on two independent global LU databases to estimate O_3 fluxes to specific LU types. A similar approach was taken by Schwede et al. (2018) to estimate global nitrogen deposition to forests using deposition fluxes archived from the second TF HTAP model intercomparison (Tan et al., 2018) and combining them with a global forest cover dataset independent of the models' internal LU databases. While this approach makes direct use of modeled dry deposition fluxes, potential inconsistencies between the LU datasets used in the CTMs and those used in the post-processing attribution of modeled fluxes

to specific ecosystems can introduce uncertainties. Moreover, utilizing archived grid-aggregated deposition fluxes does not account for potential subgrid variations in LU that could have a strong impact on ecosystem specific deposition. To explore the impacts of such subgrid effects, Schwede et al. (2018) also presented results from the EMEP MSC-W model (Simpson et al., 2012) which employs a mosaic approach to calculate and optionally output V_d and deposition fluxes for each LU present in a grid cell. Results showed that the total global estimate of nitrogen deposition to forest was 12% higher using forest-specific EMEP fluxes compared to grid-aggregated fluxes and that locally fluxes estimated using the two approaches could differ by more than a factor of 2 depending on the LU heterogeneity in a given grid cell (Schwede et al., 2018). A companion paper in this issue showed large land-use dependent variations in deposition fluxes for both sulfur and nitrogen species (particularly for bidirectional fluxes of ammonia gas) and noted that LU databases used for post-processing critical load analysis may also differ from those used in regional models, with harmonization of these databases recommended for future modeling studies (Makar et al., 2024).

As discussed above, CTM-based approaches to assess ecosystem impacts of air pollution can be affected by uncertainties in input datasets (emissions, meteorology, and LU) as well as the representation of a variety of chemical and physical processes. Hardacre et al. (2015) noted that advancing a process-level understanding of how uncertainties and errors in modeled dry deposition contribute to overall CTM error and variability would require the generation of detailed diagnostic model outputs at an hourly resolution, including stomatal and non-stomatal conductances and fluxes at both the grid-aggregated level and for individual LU categories. While a few global modeling studies using individual CTMs have incorporated such diagnostic fields and demonstrated their utility (Paulot et al., 2018; Clifton et al., 2020a) and prior to AQMEII-4 this practice was recommended by a recent review (Clifton et al. 2020b), to date it had not been adopted in model intercomparison studies at either the global or regional scale. Partially motivated by this research gap, the fourth phase of the Air Quality Model Evaluation International Initiative (AQMEII4) was designed around complementary grid and single point modeling activities for a diagnostic intercomparison and evaluation of simulated deposition with a specific focus on dry deposition of gaseous species (Galmarini et al., 2021; Clifton et al., 2023).

In this study conducted within the framework of AQMEII4, we apply the diagnostic approaches described in the AQMEII4 technical note (Galmarini et al., 2021) and the manuscript presenting the evaluation of single point models at eight long-term O_3 flux measurement sites (Clifton et al., 2023) to the analysis of O_3 dry deposition simulated by the AQMEII4 regional-scale models over North America (NA) and Europe (EU). Our analysis also leverages the common AQMEII4 LU types defined in Galmarini et al. (2021). As discussed in that study, assigning model native LU types to a common set of AQMEII4 LU types allows consistent LU-specific comparisons (note that some of the most common LU types, such as evergreen needleleaf forest, are present in all native model LU databases). First, we quantify temporal and spatial variations in estimated O_3 deposition by analyzing different deposition pathways and comparing grid-aggregated vs. AQMEII4 LU-specific diagnostics. We also demonstrate that the LU-specific diagnostics can be used to connect grid model results to the evaluation of single point models against long-term O_3 flux measurement sites described in Clifton et al. (2023). Next, we assess the impacts of differences in

LU between the AQMEII4 grid models on simulated fluxes to different ecosystems. Finally, we discuss the implications of our analyses to O₃ deposition impact studies, particularly impact studies based on datasets from multi-model intercomparison activities, as well as to future model development aimed at improving dry deposition process representation.

2 Datasets

105   Table 1 lists the participating regional-scale models, the institutions performing the simulations, the continent each simulation was performed for, their dry deposition schemes, and the LU classification scheme used in their dry deposition calculations. Detailed descriptions of the model configurations were provided in Makar et al. (2024) while their dry deposition algorithms were documented in Galmarini et al. (2021), Clifton et al. (2023), and Hogrefe et al. (2023). The 16 AQMEII4 LU types to which native LU types were assigned are described in Galmarini et al. (2021) and discussed in detail in section 3.3. An

110   operational and probabilistic evaluation of simulated O₃ mixing ratios against ground level network observations over NA and

Modeling System	Domain	Modeling Group	Dry Deposition Scheme	LU for Dry Deposition Scheme
WRF/CMAQ (M3Dry)	NA	U.S. EPA	M3Dry	MODIS
WRF/CMAQ (STAGE)	NA	U.S. EPA	STAGE	AQMEII4
GEM-MACH (Base)	NA	Environment and Climate Change Canada	Wesely	Makar et al., (2018)
GEM-MACH (Zhang)	NA	Environment and Climate Change Canada	Zhang	Zhang et al. (2003)
GEM-MACH (Ops)	NA	Environment and Climate Change Canada	Wesely	Makar et al., (2018)
WRF-Chem (RIFS)	NA	Research Center for Sustainability (RIFS)	Wesely	USGS24
WRF-Chem (UPM)	NA	Technical University of Madrid (UPM)	Wesely	USGS24
WRF-Chem (NCAR)	NA	National Center for Atmospheric Research / Yonsei University	Wesely	USGS24
WRF-Chem (RIFS)	EU	Research Center for Sustainability (RIFS)	Wesely	USGS24
WRF-Chem (UPM)	EU	Technical University of Madrid (UPM)	Wesely	USGS24
LOTOS/EUROS	EU	TNO	DEPAC	DEPAC
WRF/CMAQ (STAGE)	EU	University of Hertfordshire	STAGE	AQMEII4

EU is presented in Kioutsioukis et al. (2025).

Table 1: Model simulations analyzed in this study. See Makar et al. (2024) for additional details on the configuration of each simulation.

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The analysis in this manuscript utilizes dry deposition fluxes and the O_3 dry deposition diagnostic variables described in detail in Galmarini et al. (2021) and Clifton et al. (2023), namely O_3 dry deposition velocities (V_d) and effective conductances (Paulot et al., 2018; Clifton et al., 2020a) for the stomatal, cuticular, soil, and lower canopy pathways reported at both the grid-aggregated scale and for each of the 16 AQMEII4 LU categories. No effective conductances were available for

120 LOTOS/EUROS but reported grid-aggregated O3 V_d, deposition fluxes, and LU distributions are included in the analysis. For WRF-Chem (NCAR), LU-specific dry deposition calculations for its native "mixed shrubland / grassland" category were inadvertently mapped to and then reported for the AQMEII4 "savanna" category (along with its native "savanna" category) rather than the intended AQMEII4 "shrubland" category. This inadvertent mapping was corrected for the comparison of LU categories across models but could not be corrected for the analysis of LU-specific V_d and effective conductances. For all

125 models except GEM-MACH (Base) and GEM-MACH (Ops), the spatial fields of LU categories in both the land-surface model (LSM) and dry deposition code were time independent and considered the "snow and ice" category to represent permanent snow and ice. For the GEM-MACH (Base) and GEM-MACH (Ops) dry deposition calculations, the LU categories used in the dry deposition calculations and subsequently reported to AQMEII4 were modified from their time-independent LSM values to represent the time-varying effects of transient snow cover through the "snow and ice" category, and proportionally reduced

130 the fractions of all other LU categories in the grid cell if snow cover was present. To enable a comparison of these time-dependent LU fractions to the fractions reported by other models, time-independent fractions for GEM-MACH (Base) and GEM-MACH (Ops) were derived by scanning the hourly reported fields and determining the LU fractions in each grid cell when the coverage of the "snow and ice" category reached its annual minimum (zero for the vast majority of grid cells) for that grid cell. This approach allows the use of LU fractions as stratification criterion for all models in our analysis while

135 retaining the effects of transient snow cover (represented through separate snow cover fields rather than time-dependent LU fractions in other dry deposition schemes) in the reported dry deposition diagnostics.

As described in Galmarini et al. (2021), each simulation reported gridded fields of V_d and effective conductances as 288 individual values representing monthly median diurnal cycles (24 hours times 12 months) at each grid cell for each simulated year. This approach was chosen because reporting and storing these diagnostic fields for all hours in the year would have been

140 logistically impossible, and the selected approach still allows an analysis of their diurnal and seasonal variations. However, it should be noted that this approach of using monthly median values for individual diagnostics can introduce a temporal mismatch between reported V_d and effective conductance diagnostics, which in turn may cause the sum of the effective conductances on a monthly median basis to differ from monthly median V_d, even though on an hour-by-hour basis their sum equals V_d (Paulot et al., 2018; Clifton et al., 2020a). Despite this potential for deviations, the sum of the reported effective

145 conductances was found to be within 3.5% of V_d on an annual domain mean basis for all simulations. The largest deviation (3.5%) occurred for WRF/CMAQ (M3DRY) for which grid-aggregated V_d was calculated within the model while effective

conductances were estimated through post-processing (Hogrefe et al., 2023). For the remainder of the discussions in this manuscript, we therefore treat the sum of the monthly median effective conductances as equivalent to V_d.

3 Results

150 ### 3.1 Grid-Aggregated Results

Table 2 lists the annual total O_3 dry deposition fluxes estimated by the participating models over both the NA and EU domains. These totals are calculated over all non-water grid cells that were common to all simulations. The multi-model mean value is 74.3 Tg/yr over the NA domain and 67.4 Tg/yr over the EU domain. Individual model estimates range from 59.5 Tg/yr (GEM-MACH (Zhang)) to 91.4 Tg/yr (WRF-Chem (UPM)) over the NA domain and from 47.2 Tg/yr (LOTOS/EUROS) to 82.9

155 Tg/yr (WRF-Chem (UPM)) over the EU domain. For context, past global modeling studies reported *global* annual O_3 deposition totals generally ranging between 800 Tg/yr and 1200 Tg/yr (e.g. Hardacre et al., 2015; Stevenson et al., 2006; Wild 2007; Ganzeveld et al., 2009).

Spatial patterns of these annual deposition fluxes for each model as well as the multi-model mean and normalized standard deviation are displayed in Figure 1 for the NA domain and Figure 2 for the EU domain. Over the NA domain all models

160 estimate that O_3 deposition is highest over the Eastern U.S. as well as along the West Coast, consistent with our expectations

	Annual total O3 dry deposition flux (Tg)	Annual mean O3 dry deposition velocity (cm/s)
North America (2016)		
WRF/CMAQ (M3Dry)	66.1	0.28
WRF/CMAQ (STAGE)	68.3	0.3
GEM-MACH (Base)	73.9	0.37
GEM-MACH (Zhang)	59.5	0.27
GEM-MACH (Ops)	80.4	0.42
WRF-Chem (RIFS)	79.0	0.34
WRF-Chem (UPM)	91.5	0.38
WRF-Chem (NCAR)	75.5	0.28
Europe (2010)		
WRF-Chem (RIFS)	77.5	0.32
WRF-Chem (UPM)	82.9	0.37
LOTOS/EUROS	47.2	0.21
WRF/CMAQ (STAGE)	61.9	0.25

Table 2. Annual total O3 dry deposition fluxes and annual mean O3 dry deposition velocities estimated by the participating models over both the NA and EU domains. These totals are calculated over all non-water grid cells that were common to all simulations. The NA numbers are for 2016 while the EU number are for 2010.

that reflect a combination of higher O_3 mixing ratios due to higher O_3 precursor emissions in these regions and higher V_d over

165 areas with higher vegetation density. The pronounced differences in deposition fluxes over the densely vegetated Southeastern U.S. between WRF-Chem (UPM) and GEM-MACH (Zhang) suggest that differences in the methodology used to represent deposition to vegetation are a key driver of model spread. The map of the multi-model normalized standard deviation over NA also shows significant spread in deposition to land over the generally more arid regions in the Western U.S., in addition to the spread over the Southeastern U.S. Over the EU domain, the largest relative spread in deposition to land occurs over

170 Northwestern Scandinavia, Ireland, Great Britain, and Turkey, as well as the Alps and Northern Africa. Over both domains, there is a large relative spread in O_3 deposition to water, with generally higher values for WRF-Chem than all other models, but the absolute magnitude of the flux is much smaller than the flux over land.

To assess the role of variations in simulated V_d on the dry deposition fluxes discussed above, Table 2 includes the annual mean O_3 V_d over both domains alongside the annual total O_3. The relative differences between the annual mean V_d generally match

175 those between the annual total deposition fluxes with only small exceptions (e.g., WRF-Chem (RIFS) has slightly lower V_d but a slightly higher dry deposition fluxes than GEM-MACH (Base)). Examining the spatial patterns of annual mean V_d for each model and the multi-model mean and normalized standard deviation in Figures 3-4 and comparing them to the patterns of the corresponding dry deposition flux maps in Figures 1-2 shows a high level of agreement. For example, the areas over the Southeastern U.S. where GEM-MACH (Base) and GEM-MACH (Ops) have larger fluxes than other models coincide with

180 areas where these two simulations also have the highest O_3 V_d. Moreover, the areas with the highest normalized flux standard deviation discussed above for both the NA and EU domains also show the highest normalized standard deviation for V_d. In some instances, areas may have similar V_d but noticeably different modeled dry deposition fluxes (e.g. the Southeastern U.S. vs. parts of the Canadian boreal forest region for model GEM-MACH (Ops)), suggesting that factors other than V_d, such as the location and density of precursor emission sources as well as other parameterizations for regional transport and chemical

185 lifetime also impact modeled dry deposition flux patterns. GEM-MACH (Ops) and GEM-MACH (Base) share the same gas-phase deposition code, though GEM-MACH (Base) more explicitly simulates transport through the forest canopy (Makar et al., 2017), and incorporates a parameterization for the influence of vehicle motion on vertical diffusive transport of mobile emissions (Makar et al., 2021), both of which have been shown to have a substantial impact on ozone concentrations near the surface. However, despite these potentially confounding influences, the general agreement of both the model rankings for the

190 domain-wide dry deposition fluxes vs. V_d (Table 2, Figures S1 vs. S2) and their spatial patterns (Figure 1 vs. Figure 3 and Figure 2 vs. Figure 4) confirms the influence of O_3 V_d towards deposition fluxes estimated in these regional-scale simulations. This in turn establishes that a diagnostic understanding of simulated O_3 V_d can aid in interpreting model-to-model differences in dry deposition fluxes.
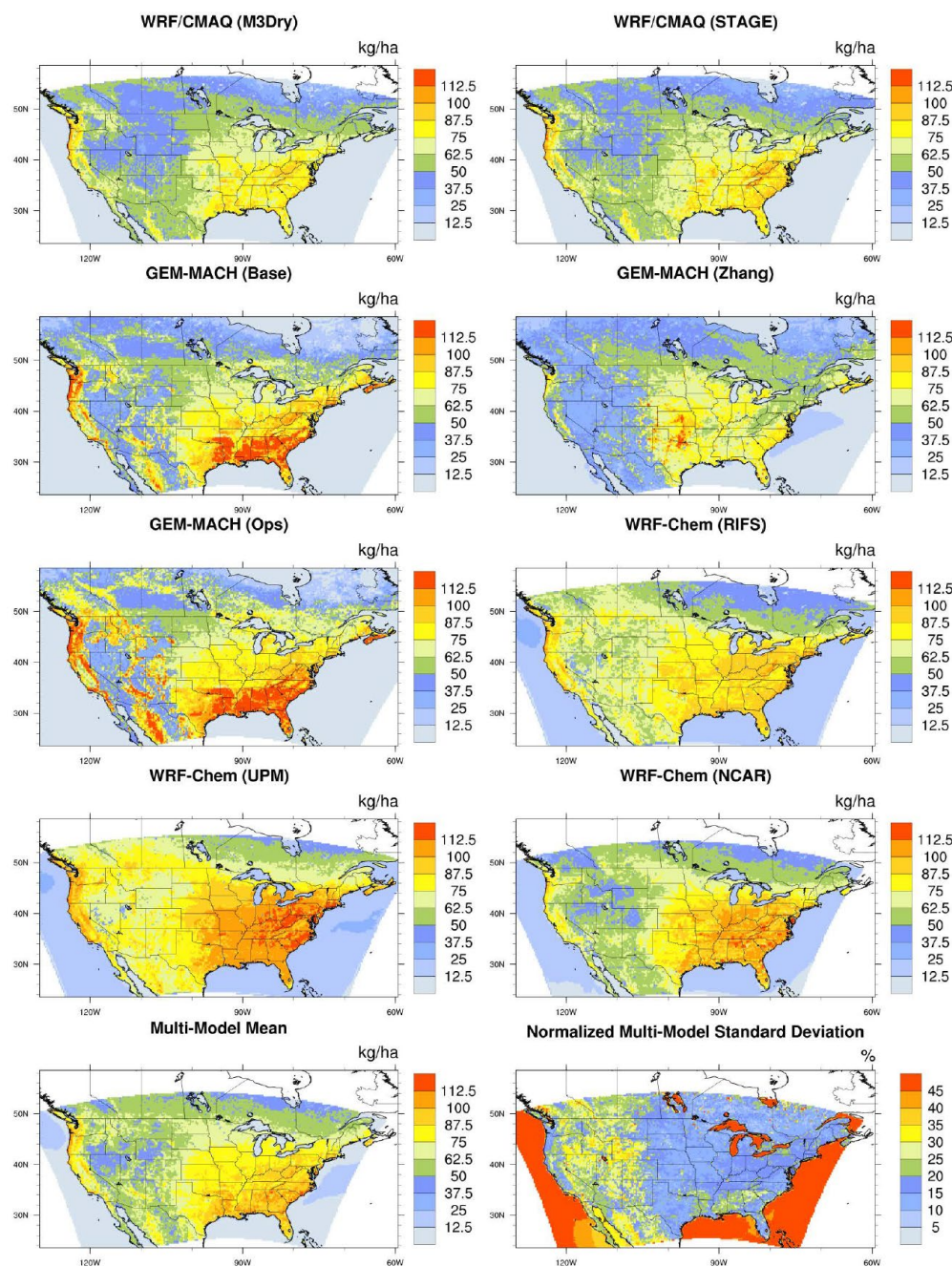
Figure 1. 2016 annual total O3 grid-scale dry deposition fluxes for each model, the multi-model mean, and the normalized multi-model standard deviation over the NA domain. Note that the plots for individual models are not clipped to the domain common to all simulations and show the maximum spatial extent submitted for each model. The multi-model mean and normalized standard deviations are calculated and shown over the common domain.
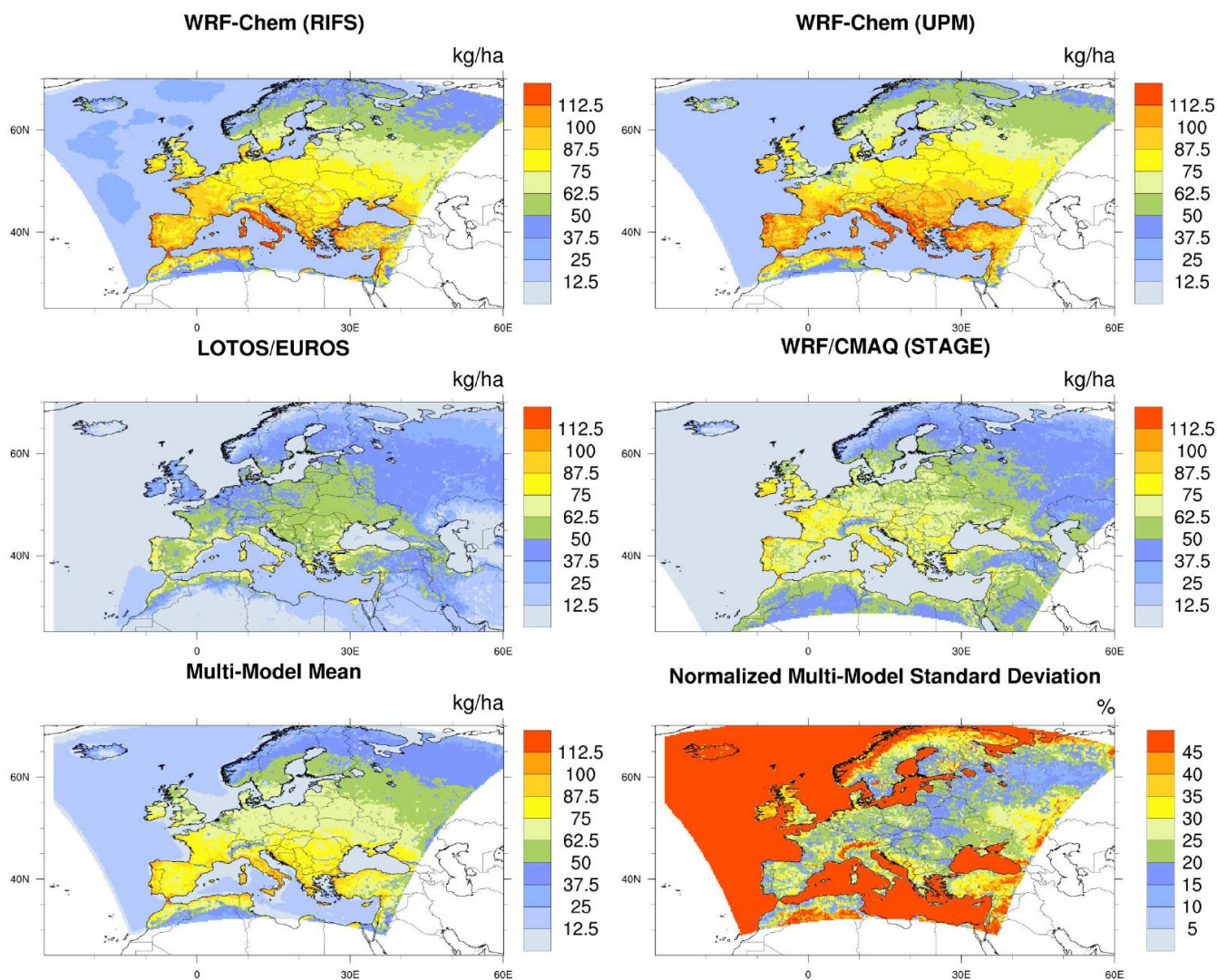
Figure 2. 2010 annual total O3 grid-scale dry deposition fluxes for each model, the multi-model mean, and the normalized multi-model standard deviation over the EU domain. Note that the plots for individual models are not clipped to the domain common to all simulations and show the maximum spatial extent submitted for each model. The multi-model mean and normalized standard deviations are calculated and shown over the common domain.
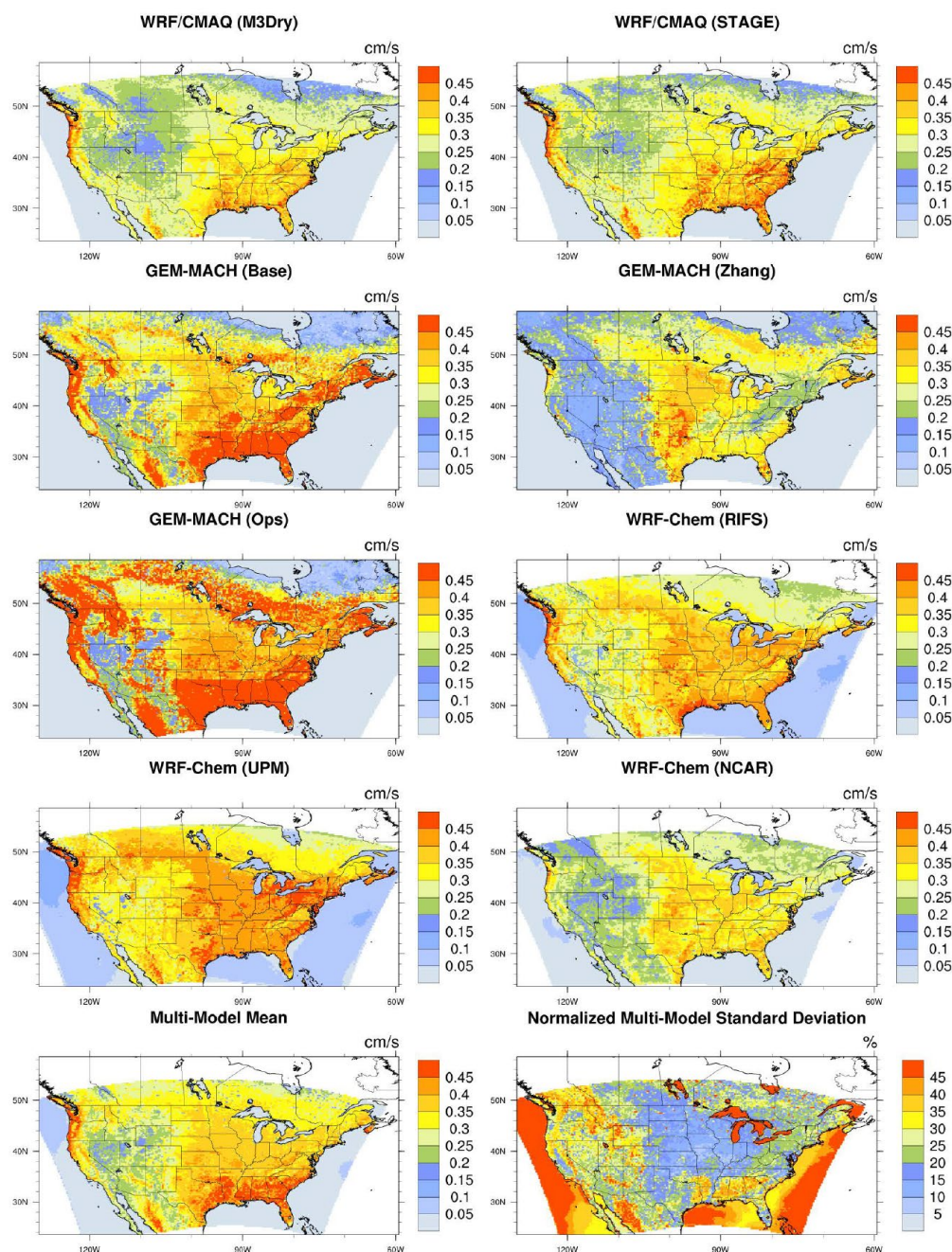
Figure 3. 2016 annual mean O3 grid-scale dry deposition velocities for each model, the multi-model mean, and the normalized multi-model standard deviation over the NA domain. Note that the plots for individual models are not clipped to the domain common to all simulations and show the maximum spatial extent submitted for each model. The multi-model mean and normalized standard deviations are calculated and shown over the common domain.

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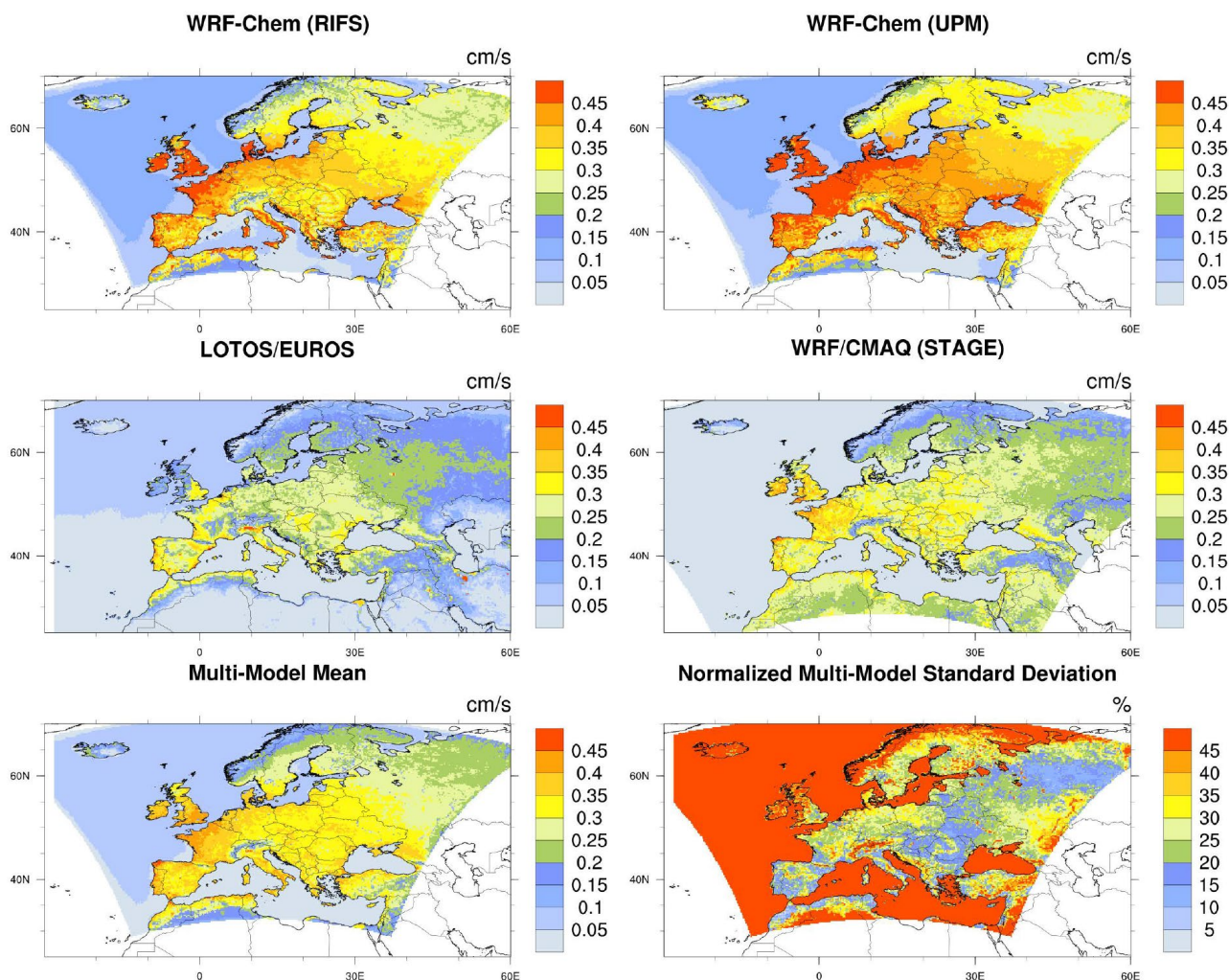
Figure 4. 2010 annual mean O3 grid-scale dry deposition velocities for each model, the multi-model mean, and the normalized multi-model standard deviation over the EU domain. Note that the plots for individual models are not clipped to the domain common to all simulations and show the maximum spatial extent submitted for each model. The multi-model mean and normalized standard deviations are calculated and shown over the common domain.

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Figures S1 and S2 provide insight into temporal differences in modeled deposition by showing domain-wide deposition fluxes and O$_3$ V$_d$ calculated for winter vs. summer and daytime vs. nighttime periods for the NA domain. The largest deposition fluxes occur during summer daytime hours while the lowest deposition fluxes occur during winter nighttime hours, a pattern also present for V$_d$. Photochemical production of O$_3$ is also higher in summer than in winter, which will also contribute to

220 higher summertime fluxes. Fluxes during winter daytime hours are generally of the same order of magnitude as fluxes during

summer nighttime hours, both times corresponding to low photochemical production or ozone destruction through titration. These figures also suggest that the low annual total deposition value of model GEM-MACH (Zhang) and the high annual total deposition value of model WRF-Chem (UPM) shown in Table 2 were driven by low and high summer daytime flux values, respectively, and/or that other terms such as photochemical production in WRF-Chem (UPM) are enhancing summer ozone concentrations, hence increasing fluxes. While the absolute model-to-model differences are smaller during winter than summer for both fluxes and V_d, the normalized standard deviation is comparable across seasons for both variables (summer daytime flux 18.2%; winter daytime flux 14.5%; summer nighttime flux 22.7%; winter nighttime flux 22.8%; summer daytime V_d 18.6%; winter daytime V_d 17.2%; summer nighttime V_d 34.2%; winter nighttime V_d 28.2%). During daytime, fluxes and V_d exhibit similar relative spread. The larger nighttime spread in V_d compared to the fluxes suggests that nighttime chemical sinks and/or a smaller ozone reservoir in the shallower nighttime mixing layer may be more important factors controlling O_3 concentrations, and hence O_3 deposition fluxes, than the magnitude of the deposition velocity itself. However, it should also be noted that Clifton et al. (2020a) found that even small differences in wintertime V_d can have substantial impacts on the tropospheric O_3 budget given the longer chemical lifetime.

Figures 5-6 depict the contributions of the four effective conductance pathways to the annual domain-average V_d for each model over both domains. The total height of the bars for each model closely corresponds to that model's V_d shown in Table 2, within the caveats discussed in Section 2. Note that some of these simulations (WRF/CMAQ (M3DRY), WRF/CMAQ (STAGE), and GEM-MACH (Zhang)) do not incorporate a separate lower canopy pathway in their dry deposition schemes, and thus only have three deposition pathways in total (Galmarini et al., 2021; Clifton et al., 2023). These Figures show that models with similar V_d (e.g. WRF/CMAQ (M3DRY), GEM-MACH (Zhang), and WRF-Chem (NCAR) over NA) can show significant differences in the absolute and relative contributions of different pathways to V_d. Moreover, these Figures also demonstrate that effective conductances allow an attribution of model differences in V_d to specific processes. For example, GEM-MACH (Base), GEM-MACH (Ops), and WRF-Chem (RIFS) all have similar soil and lower canopy effective conductances, revealing that the differences in V_d between these simulations stem from differences in the cuticular and, to a lesser extent, stomatal pathways. Specifically, both GEM-MACH (Base) and GEM-MACH (Ops) have a higher cuticular effective conductance than WRF-Chem (RIFS). For the two CMAQ simulations over NA, Figure 5 confirms the generally larger contributions from the stomatal and cuticular pathways for WRF/CMAQ (M3DRY) and the generally larger contribution from the soil pathway for WRF/CMAQ (STAGE) that was reported in Hogrefe et al. (2023). Comparing GEM-MACH (Base) with GEM-MACH (Zhang) shows lower contributions from the soil and stomatal pathways for the latter, which also does not include a separate lower canopy pathway.
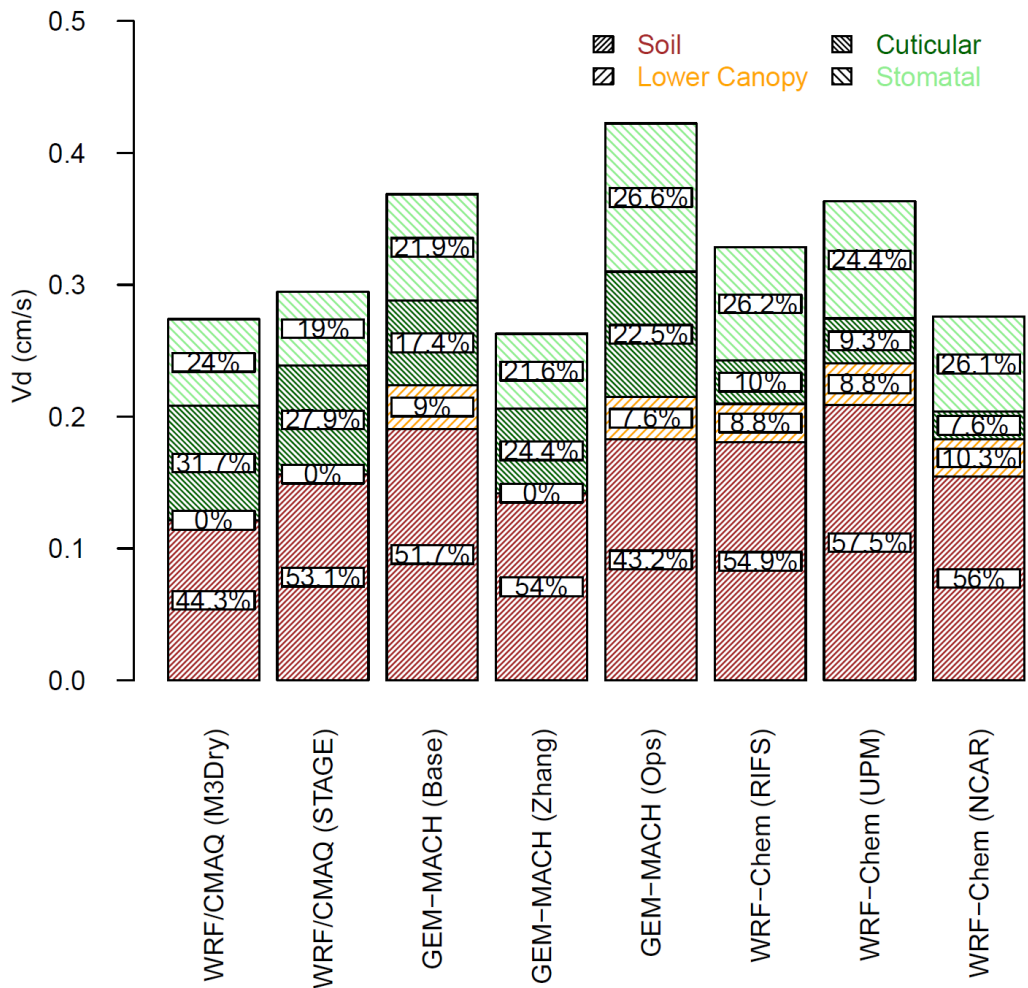
Figure 5. Annual domain-average grid-scale effective conductances and ozone deposition velocities for 2016 over the NA domain. Averages were calculated over all non-water grid cells in the portion of the analysis domain shared by all model simulations.

Comparing the two GEM-MACH simulations with the same dry deposition scheme, GEM-MACH (Base) and GEM-MACH
(Ops), reveals that differences other than the dry deposition scheme that exist between these simulations (e.g. meteorology and leaf area index (LAI)) have a noticeable impact on the magnitude of the cuticular and stomatal pathways. It should be noted that both GEM-MACH (Base) and GEM-MACH (Ops) are coupled models with chemistry residing within a weather forecast model – however, GEM-MACH (Base) is "fully coupled", i.e. includes aerosol direct and indirect feedbacks on the predicted meteorology. GEM-MACH (Base) also includes several other parameterizations affecting chemical transport as noted above. The difference in the strength of the cuticle and stomatal conductances between these two models thus reflects differences in the forecasted meteorology in turn influencing the deposition velocity components.
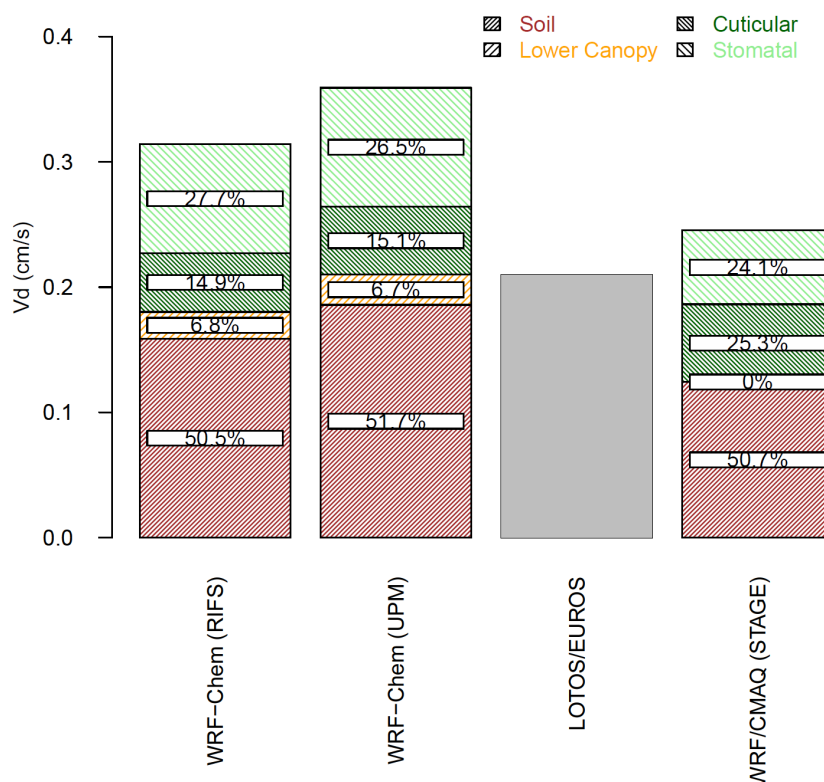
Figure 6. Annual domain-average grid-scale effective conductances and ozone deposition velocities for 2010 over the EU domain. Averages were calculated over all non-water grid cells in the portion of the analysis domain shared by all model simulations. The bar for LOTOS/EUROS shows only the deposition velocity since no effective conductances were reported.

The impact of factors other than the dry deposition scheme itself on simulated V_d and pathway contributions is further illustrated by the comparison of the three WRF-Chem simulations performed over NA and the two WRF-Chem simulations performed over EU. These simulations all use the WRF-Chem implementation of the Wesely scheme and as a result all share similar relative pathway contributions to V_d, but V_d itself shows a variation of roughly 25% over NA and 15% over EU between these simulations. All three of these models make use of feedbacks between aerosols and meteorology – and hence other parameterization differences in addition to the gas-phase deposition code influence the resulting gas-phase deposition velocities, through changes in the predicted temperature, relative humidity, and the other meteorological terms influencing deposition velocity.

Figures 7-8 depict maps of absolute pathway contributions to annual mean V_d over both domains, where the mean is from the average of the monthly median values. The maps show the lower contributions from the cuticular pathway for WRF-Chem compared to other simulations, especially over the Eastern U.S. and Central Europe. This is consistent with the domain-average contributions shown in Figure 5. For the models that include the lower canopy pathway as a separate term from the other

pathways (all WRF-Chem simulations and two of the GEM-MACH simulations), its contribution to V_d is less than 0.06 cm/s throughout both domains (10% or less of the domain-wide annual means as shown in Figures 5-6). Both CMAQ simulations
280 and the GEM-MACH (Zhang) simulation tend to have stronger longitudinal gradients over the NA domain for the soil pathway contribution compared to other simulations. The two GEM-MACH simulations using the Wesely dry deposition scheme (GEM-MACH (Base) and GEM-MACH (Ops)) and the WRF-Chem (CIFS) and WRF-Chem (UPM) simulations have noticeably higher absolute soil pathway contributions in the South-Central and Southeastern U.S. than the other simulations, indicating that the higher values in the domain-average soil pathway contributions for these four simulations shown in Figure
285 3 originate in these regions.

Figures S3 – S4 show the relative, rather than absolute, contribution of the four pathways to annual mean V_d over both domains while corresponding maps for summer and winter over NA are included as Figures S5 – S6. These figures show that the WRF-Chem simulations tend to have a weaker spatial variation than the WRF/CMAQ and GEM-MACH simulations in the split between different pathways over both domains, especially for the relative contribution of the soil pathway. For example, the
290 dominance of the stomatal and cuticular pathways over the soil pathway over the Eastern U.S., and of the soil pathway over the stomatal and cuticular pathways in the Western U.S. that is simulated by WRF/CMAQ and GEM-MACH is less pronounced in the WRF-Chem simulations. Over the EU domain, the WRF-Chem simulations show a larger relative contribution of the soil pathway than the WRF/CMAQ simulation throughout much of Central Europe. For the NA domain, the relative contributions of the cuticular component is highest for WRF/CMAQ over the Northeastern U.S., followed by
295 GEM-MACH and then WRF-Chem. These features are especially pronounced during summer (Figure S5). For the EU domain, the relative contribution of the stomatal component is highest for WRF/CMAQ over eastern Central Europe and exceeds that of the WRF-Chem simulations.
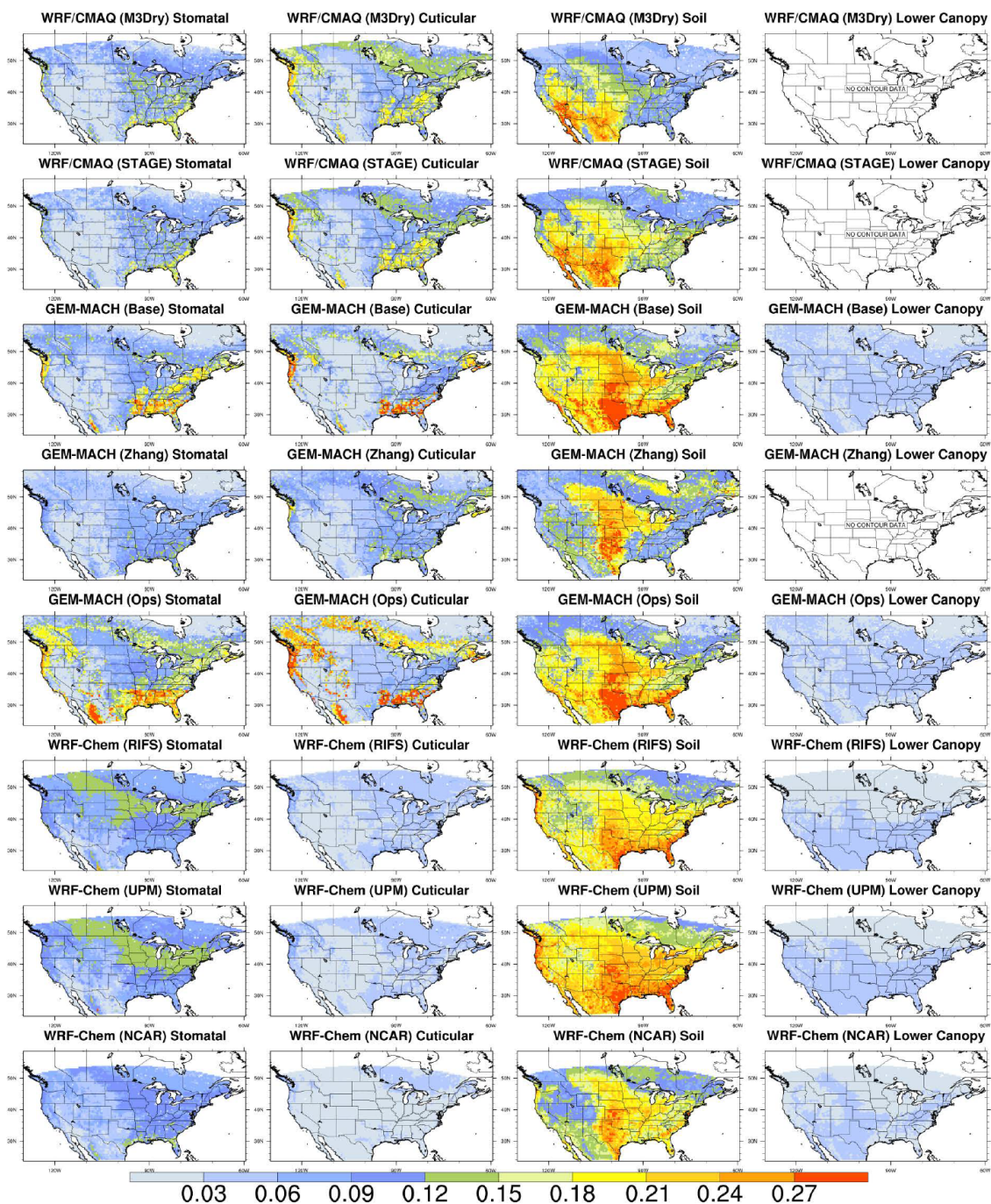
Figure 7. Absolute grid-scale ozone effective conductances (cm/s), averaged over the entire year. Results are for the NA domain during 2016. Note that these maps are not clipped to the domain common to all simulations and show the maximum spatial extent of non-water cells submitted for each model.
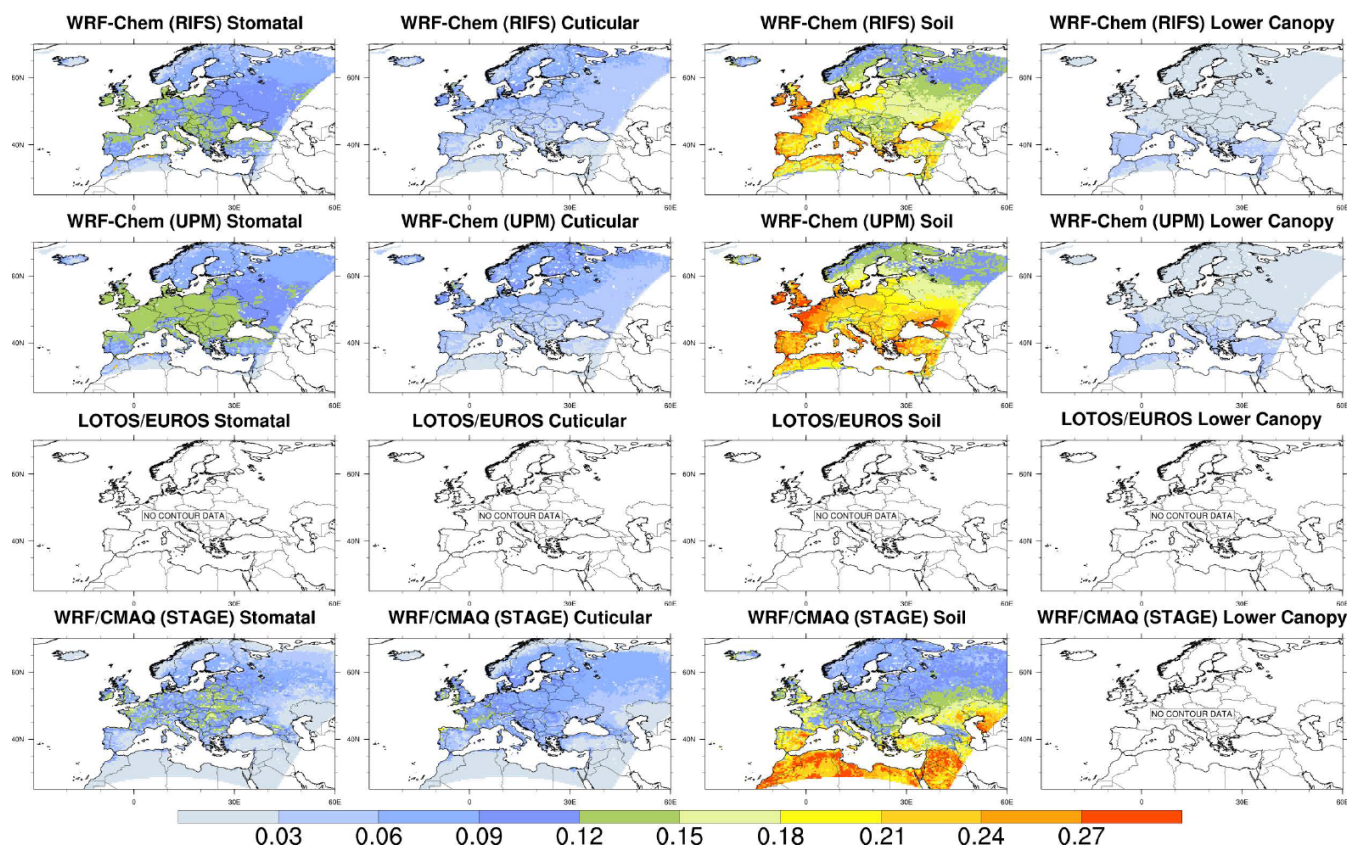
Figure 8: Absolute grid-scale ozone effective conductances (cm/s), averaged over the entire year. Results are for the NA domain during 2016. Note that these maps are not clipped to the domain common to all simulations and show the maximum spatial extent of non-water cells submitted for each model.

3.2 LU-Specific Results

Because parameter choices in dry deposition schemes are often tied to LU, the model-to-model differences in domain-average grid-aggregated V_d and pathway contributions may result not only from different process representations, but also different LU spatial distributions and/or LU-dependent parameter choices. To investigate these effects, AQMEII4 collected V_d and effective conductances for 16 standardized LU categories in addition to the grid-aggregated values analyzed above. Figure 9 shows the contributions of the four effective conductance pathways to the annual mean V_d for evergreen needleleaf forest for each model over the NA domain, averaged over only those grid cells were a given model had 85% coverage of evergreen needleleaf forest (note that the number of such grid cells differed across models, as discussed in greater detail in Section 3.3).
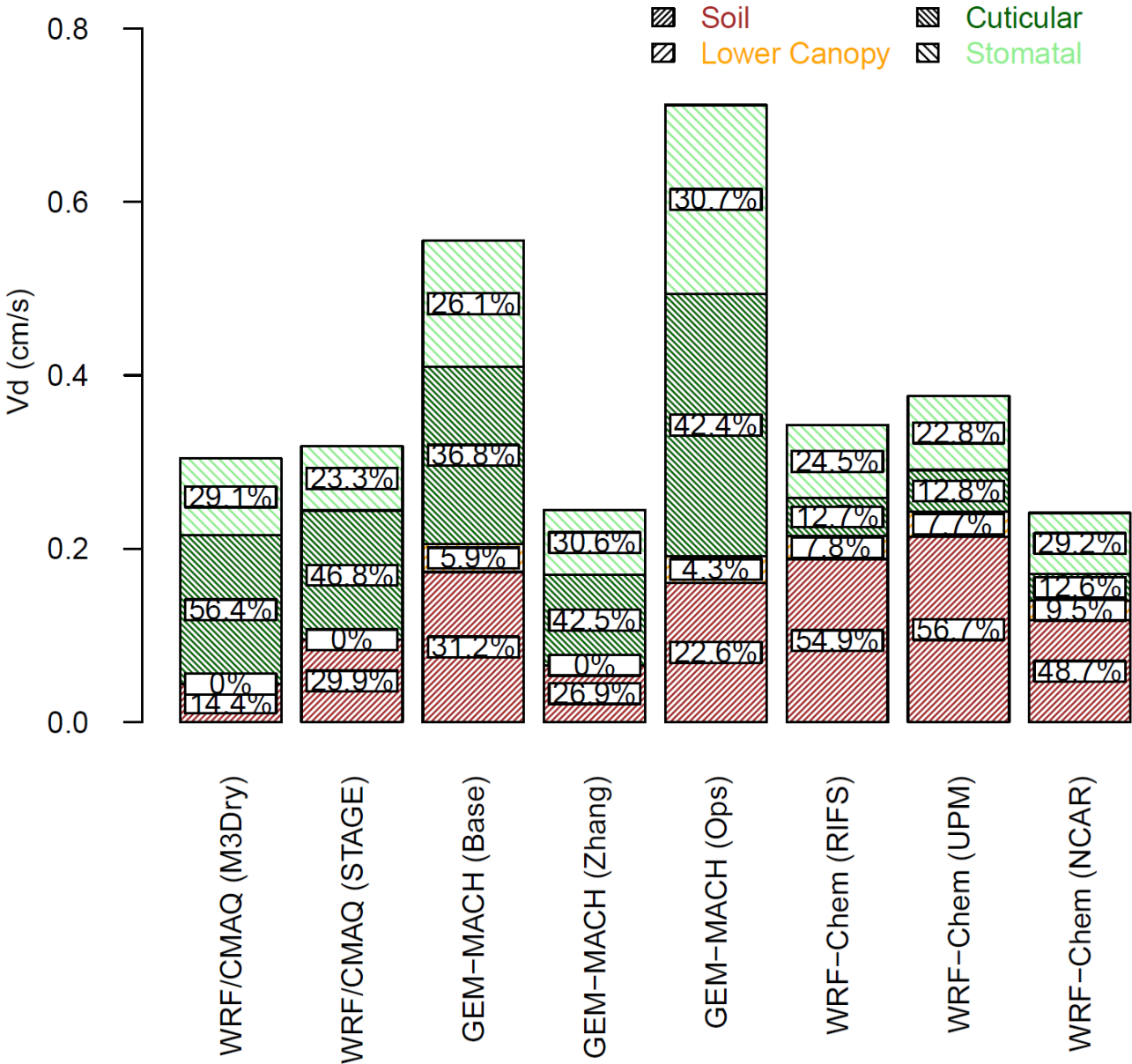
Figure 9. 2016 annual domain-average LU-specific effective conductances and ozone deposition velocities for evergreen needleleaf forest over the NA domain. Averages were calculated over all grid cells in the portion of the analysis domain shared by all model simulations for which a given model had coverage of at least 85% for the evergreen needleleaf forest LU category.

Comparing this Figure to the equivalent grid-aggregated results shown in Figure 5 when all common non-water grid cells were analyzed shows that considering instead only the model grid cells containing a specific LU type can increase model spread. For example, annual mean grid-aggregated V_d ranges from 0.26 - 0.42 cm/s averaged over all common non-water grid cells

over NA (Figure 5) while the range is 0.24 - 0.71 for V_d for grid cells dominated by the evergreen needleleaf forest category (Figure 9). Similar increases can also be seen in the ranges of absolute pathway contributions (e.g. soil effective conductance from 0.12 – 0.21 cm/s in Figure 5 to 0.04 – 0.21 cm/s in Figure 9) and relative pathway contributions (e.g. cuticular conductance from 7.6% to 31.7% in Figure 5 to 12.6% – 56.4% in Figure 9). Results of individual and summed pathways contributions for a total of eight LU categories over both domains are shown in Tables 3a-3e. These results confirm the general increase in model spread of both V_d and pathway contributions when considering LU-specific diagnostics, especially for V_d and the cuticular and stomatal effective conductances over forested and agricultural LU types. Such increased heterogeneity of process-level diagnostics when considering locations corresponding to specific LU categories compared to locations representing a mix of LU categories is also reported in Kioutsioukis et al. (2025). This finding suggests that LU-dependent parameter choices

	Gridscale	URB	BAR	ENF	DBF	MF	SHR	AGR	GRA
WRF/CMAQ (M3Dry)	0.28	0.21	**0.25**	0.3	0.37	0.27	*0.27*	*0.28*	*0.21*
WRF/CMAQ (STAGE)	0.3	**0.27**	0.24	0.32	0.42	0.31	*0.27*	0.3	0.25
GEM-MACH (Base)	0.36	0.26	0.21	0.55	**0.48**	0.39	0.35	0.4	0.36
GEM-MACH (Zhang)	*0.26*	*0.17*	0.14	*0.24*	*0.21*	0.27	0.34	0.33	0.34
GEM-MACH (Ops)	**0.41**	0.26	0.21	**0.71**	0.45	**0.49**	**0.41**	**0.42**	**0.36**
WRF-Chem (RIFS)	0.33	0.18	0.13	0.34	0.37	0.28	0.28	0.37	0.32
WRF-Chem (UPM)	0.36	0.2	0.16	0.38	0.42	0.31	0.31	0.41	0.34
WRF-Chem (NCAR)	0.27	0.2	*0.12*	*0.24*	0.32	*0.26*		0.35	0.27
Range NA	0.15	0.1	0.13	0.47	0.27	0.23	0.14	0.14	0.15
WRF-Chem (RIFS)	0.32	*0.18*	*0.14*	0.31	0.38	0.26	**0.3**	0.36	0.3
WRF-Chem (UPM)	**0.36**	0.19	0.17	**0.33**	**0.42**	**0.29**	0.29	**0.42**	**0.31**
LOTOS/EUROS									
WRF/CMAQ (STAGE)	*0.24*	**0.26**	**0.25**	*0.22*	*0.29*	*0.24*	*0.22*	*0.29*	*0.2*
Range EU	0.12	0.08	0.11	0.11	0.13	0.05	0.08	0.13	0.11

Table 3a. Annual mean grid-scale and LU-specific sum of effective conductances (cm/s) for individual simulations as well as the range of values across all simulations. For a given column and domain, the maximum values are shown in bold and the minimum values shown in italics. Blank cells indicate that data for a given model, pathway, and/or LU was not available. The grid-scale results reflect spatial averages over all non-water grid cells in the common analysis domain. The LU-specific results reflect spatial averages over all cells for which that LU fraction exceeded 85% for a given model (note that the number of such cells can vary across models). The LU categories are abbreviated as URB (urban), BAR (barren), ENF (evergreen needleleaf forest), DBF (deciduous broadleaf forest), MF (mixed forest), SHR (shrubland), AGR (planted / cultivated), and GRA (grassland).

	Gridscale	URB	BAR	ENF	DBF	MF	SHR	AGR	GRA
WRF/CMAQ (M3Dry)	0.07	*0*	0	0.09	0.13	0.09	0.01	0.09	*0.02*
WRF/CMAQ (STAGE)	*0.06*	0	0	*0.07*	0.1	0.08	*0*	*0.07*	*0.02*
GEM-MACH (Base)	0.08	**0.01**	0	0.15	**0.18**	0.1	0.05	*0.07*	0.04
GEM-MACH (Zhang)	*0.06*	0	0	*0.07*	*0.08*	*0.07*	0.05	*0.07*	**0.13**
GEM-MACH (Ops)	**0.11**	**0.01**	0	**0.22**	0.16	**0.15**	**0.1**	0.09	0.04
WRF-Chem (RIFS)	0.09	*0*	0	0.08	0.12	0.09	0.05	0.11	0.06
WRF-Chem (UPM)	0.09	*0*	0	0.09	0.13	0.1	0.04	**0.12**	0.05
WRF-Chem (NCAR)	0.07	*0*	0	*0.07*	*0.09*	0.09		0.1	0.05
Range NA	0.05	0.01	0	0.15	0.1	0.08	0.1	0.05	0.11
WRF-Chem (RIFS)	0.09	*0*	0	**0.07**	0.12	0.08	**0.04**	**0.12**	**0.06**
WRF-Chem (UPM)	**0.1**	*0*	0	**0.07**	**0.13**	**0.09**	**0.04**	**0.12**	**0.06**
LOTOS/EUROS									
WRF/CMAQ (STAGE)	*0.06*	**0.01**	0	*0.06*	*0.05*	*0.07*	*0.01*	*0.09*	*0.01*
Range EU	0.04	0.01	0	0.01	0.08	0.02	0.03	0.03	0.05

Table 3b. As in Table 3a but for the stomatal effective conductances (cm/s).

	Gridscale	URB	BAR	ENF	DBF	MF	SHR	AGR	GRA
WRF/CMAQ (M3Dry)	**0.09**	*0*	0	0.17	**0.19**	0.14	*0.01*	**0.07**	*0.02*
WRF/CMAQ (STAGE)	0.08	0	0	0.15	**0.19**	0.13	*0.01*	**0.07**	*0.02*
GEM-MACH (Base)	0.06	0	0	0.2	0.1	0.12	0.02	0.03	0.04
GEM-MACH (Zhang)	0.06	**0.02**	0	0.1	0.06	0.09	**0.07**	0.05	**0.05**
GEM-MACH (Ops)	**0.09**	0	0	**0.3**	0.09	**0.19**	0.05	0.04	**0.05**
WRF-Chem (RIFS)	0.03	*0*	0	0.04	0.03	0.04	0.02	0.03	0.03
WRF-Chem (UPM)	0.03	*0*	0	0.05	0.03	0.04	0.02	0.03	0.03
WRF-Chem (NCAR)	*0.02*	*0*	0	*0.03*	*0.02*	*0.03*		*0.02*	*0.02*
Range NA	0.07	0.02	0	0.27	0.17	0.16	0.06	0.05	0.03
WRF-Chem (RIFS)	*0.05*	**0.02**	**0.01**	*0.07*	*0.05*	*0.05*	0.03	*0.04*	**0.04**
WRF-Chem (UPM)	*0.05*	**0.02**	*0*	**0.08**	*0.05*	0.06	**0.04**	0.05	**0.04**
LOTOS/EUROS									
WRF/CMAQ (STAGE)	**0.06**	*0.01*	*0*	**0.08**	**0.07**	**0.08**	*0.01*	**0.06**	*0.01*
Range EU	0.01	0.01	0.01	0.01	0.02	0.03	0.03	0.02	0.03

340   **Table 3c.** As in Table 3a but for the cuticular effective conductances (cm/s).

	Gridscale	URB	BAR	ENF	DBF	MF	SHR	AGR	GRA
WRF/CMAQ (M3Dry)	*0.12*	0.21	**0.25**	*0.04*	*0.05*	*0.04*	0.25	*0.12*	0.17
WRF/CMAQ (STAGE)	0.16	**0.27**	0.24	0.1	0.13	0.1	**0.26**	0.16	0.21
GEM-MACH (Base)	0.19	0.24	0.2	0.17	0.15	0.12	0.23	**0.26**	**0.23**
GEM-MACH (Zhang)	0.14	*0.15*	0.14	0.07	0.07	0.11	0.22	0.21	*0.16*
GEM-MACH (Ops)	0.18	0.24	0.2	0.16	0.15	0.11	0.22	0.25	**0.23**
WRF-Chem (RIFS)	0.18	0.18	0.13	0.19	0.19	0.13	*0.17*	0.2	0.2
WRF-Chem (UPM)	**0.21**	0.2	0.16	**0.21**	**0.22**	**0.14**	0.21	0.23	0.22
WRF-Chem (NCAR)	0.15	0.2	*0.12*	0.12	0.17	0.11		0.2	0.17
Range NA	0.09	0.12	0.13	0.17	0.17	0.1	0.09	0.14	0.07
WRF-Chem (RIFS)	0.16	*0.16*	*0.13*	0.15	0.18	0.11	0.19	0.18	*0.17*
WRF-Chem (UPM)	**0.19**	0.17	0.17	**0.16**	**0.21**	**0.12**	*0.18*	**0.22**	**0.18**
LOTOS/EUROS									
WRF/CMAQ (STAGE)	*0.12*	**0.24**	**0.25**	*0.08*	*0.17*	*0.09*	**0.2**	*0.14*	**0.18**
Range EU	0.07	0.08	0.12	0.08	0.04	0.03	0.02	0.08	0.01

Table 3d. As in Table 3a but for the soil effective conductances (cm/s).

	Gridscale	URB	BAR	ENF	DBF	MF	SHR	AGR	GRA
WRF/CMAQ (M3Dry)									
WRF/CMAQ (STAGE)									
GEM-MACH (Base)	**0.03**	**0.01**	**0.01**	**0.03**	**0.05**	**0.05**	**0.05**	**0.04**	**0.05**
GEM-MACH (Zhang)									
GEM-MACH (Ops)	**0.03**	**0.01**	**0.01**	**0.03**	**0.05**	0.04	*0.04*	**0.04**	0.04
WRF-Chem (RIFS)	**0.03**	*0*	*0*	**0.03**	*0.03*	*0.02*	0.04	*0.03*	*0.03*
WRF-Chem (UPM)	**0.03**	*0*	*0*	**0.03**	0.04	0.03	*0.04*	*0.03*	0.04
WRF-Chem (NCAR)	**0.03**	*0*	*0*	*0.02*	0.04	0.03		*0.03*	*0.03*
Range NA	0	0.01	0.01	0.01	0.02	0.03	0.01	0.01	0.02
WRF-Chem (RIFS)	**0.02**	*0*	*0*	**0.02**	**0.03**	**0.02**	**0.04**	*0.02*	**0.03**
WRF-Chem (UPM)	**0.02**	*0*	*0*	**0.02**	**0.03**	**0.02**	*0.03*	**0.03**	**0.03**
LOTOS/EUROS									
WRF/CMAQ (STAGE)									
Range EU	0	0	0	0	0	0	**0.01**	0.01	0

Table 3e. As in Table 3a but for the lower canopy effective conductances (cm/s).

and the representation of vegetation effects on O_3 dry deposition (e.g. whether or not a given scheme accounts for soil moisture effects on stomatal conductance) are a significant source of variability in grid-aggregated V_d while also indicating that an

345   analysis of only grid-aggregated deposition diagnostics may partially mask the effects of process-specific differences that exist between schemes. However, the attribution of LU-specific diagnostics to LU-dependent parameter choices is complicated by potential model-to-model differences in LU distribution – for example, differences in the number and location of grid cells with a given LU type in a given modeling system may cause differences in macro-scale meteorological variables like wind speed and solar radiation that affect the deposition calculations in this LU-specific analysis. Differences in LU distributions

350   between models are analyzed in Section 3.3.

The availability of LU-specific dry deposition diagnostics from the AQMEII4 grid models also provides an opportunity to compare these diagnostics to the results from the point model intercomparison study by Clifton et al. (2023). At each of the eight O_3 flux measurement sites examined in Clifton et al. (2023), the point model simulations were constrained to use a common set of site-specific variables like LAI, roughness length, and soil moisture. Figures 10 and S6 – S8 show examples

355   of this comparison. Figure 10 compares winter and summer average V_d and effective conductances simulated by the grid models for grid cells with mixed forest coverage greater than 85% against the corresponding results for point models at the two mixed forest sites analyzed in Clifton et al. (2023), i.e. Borden Forest and Harvard Forest. The point model results are identical to those shown in Figure 5 of Clifton et al. (2023), but while 18 point simulations were included in that Figure, only the five simulations corresponding to the schemes appearing in Clifton et al (2023) that were also implemented in the AQMEII4

360   grid models (CMAQ M3Dry, CMAQ STAGE, GEM-MACH Wesely, GEM-MACH Zhang, and WRF-Chem Wesely) are reproduced here. Also note that while the seasonal grid model values shown in Figure 10 are derived for a single year (2016) but averaged over all grid cells for which a given model had a fractional coverage of mixed forest exceeding 85%, the point model values at Borden Forest and Harvard Forest are multi-year means at single sites. The motivation for performing this comparison despite these differences in spatio-temporal aggregation is to assess to which extent the conclusions of a single-

365   point modeling study are consistent with results obtained from grid model deposition diagnostics and could therefore inform grid model development by providing process-level insights.

A comparison of the summertime grid model and point model results in Figure 10 leads to similar conclusions regarding the magnitudes of simulated V_d and pathway contributions across models. For example, both grid model and point model V_d range between 0.4 and 1.0 cm s^{-1} across models. The lowest V_d for the point models is simulated by GEM-MACH Zhang while the

370   highest V_d is simulated by GEM-MACH Wesely. This is consistent with the grid model results, with GEM-MACH (Zhang) showing lower V_d than all other grid models and GEM-MACH (Base) and GEM-MACH (Ops) showing higher V_d than all other grid models. Grid and point models also agree that CMAQ M3Dry has a much smaller contribution of the soil effective conductance to V_d compared to all other models and that WRF-Chem Wesely has a smaller contribution of the cuticular effective conductance to V_d compared to all other models.
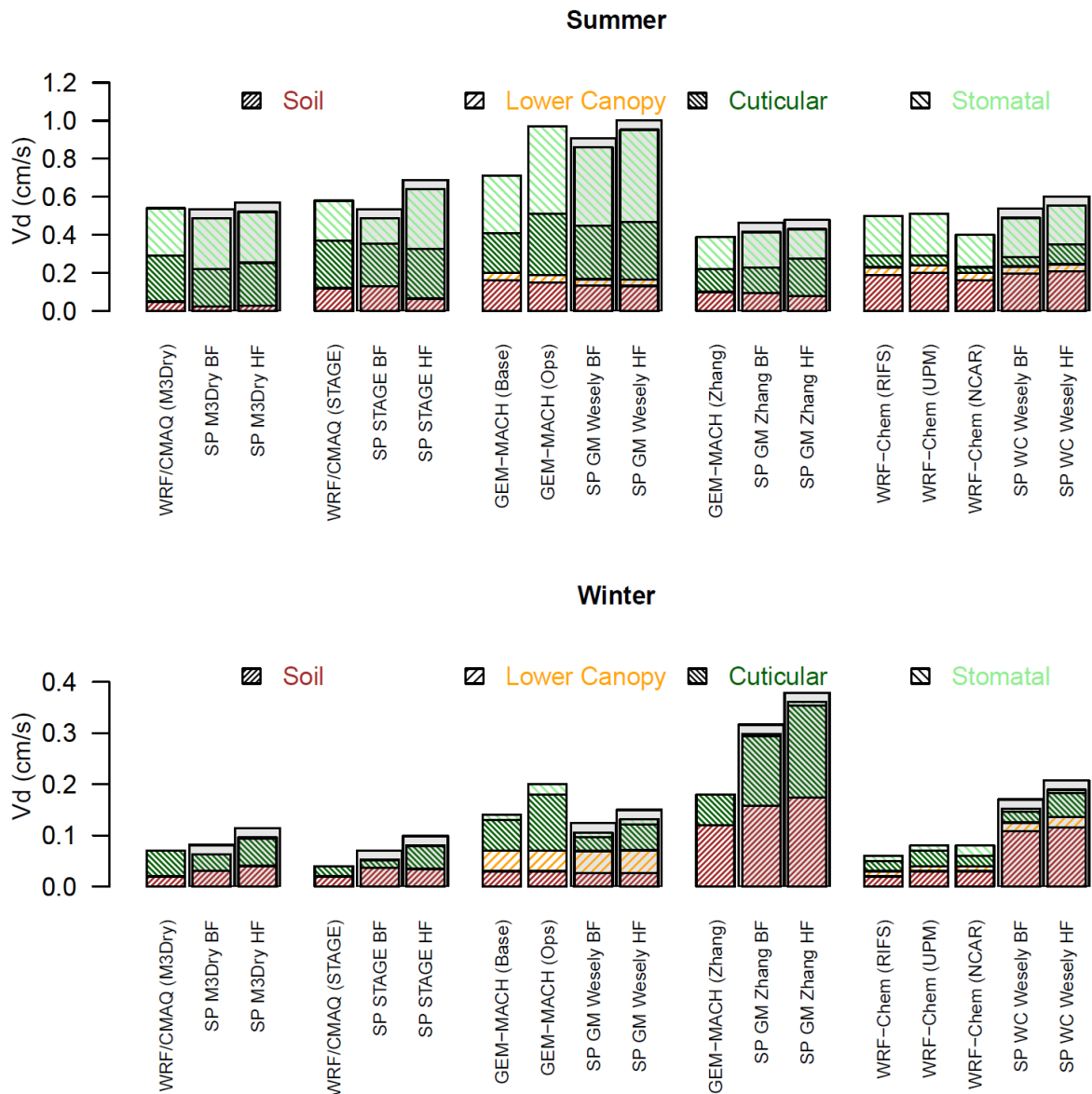
Figure 10. Summer and winter effective conductances and ozone deposition velocities calculated by the grid models for mixed forest grid cells and calculated by the corresponding subset of single point (SP) models analyzed in Clifton et al. (2023) at the Borden Forest (BF) and Harvard Forest (HF) sites. The bars for the SP models are overlayed on grey boxes to visually distinguish them from the bars representing grid models. In the x-axis labels, results for the SP GEM-MACH Wesely simulations are shown as "SP GM Wesely" while results for the SP WRF-Chem Wesely simulations are shown as "SP WC Wesely". The mixed forest grid cells selected for this analysis are those in which a given model had at least 85% coverage for this LU category. The number of these grid cells differs across models due to underlying differences in LU (see Section 3.3).

During winter, the point and grid model results show consistency in terms of relative model rankings (GEM-MACH Zhang
and the corresponding grid model simulations GEM-MACH (Base) and GEM-MACH (Ops) have the highest V_d while the
385 CMAQ STAGE point and grid model simulations have the lowest V_d), the magnitude of V_d (ranging from about 0.1 to about
0.35 cm s^{-1}), and in terms of model-to-model variations in pathway contributions. However, the grid model results show
generally lower V_d and a different ranking than the point model results, with GEM-MACH Wesely V_d roughly equal to GEM-
MACH Zhang in the point model comparison rather than significantly lower when comparing the corresponding grid model
simulations GEM-MACH (Base), GEM-MACH (Ops), and GEM-MACH (Zhang). Spatial variations in snow cover across the
390 mixed forest grid cells in the grid models as well as interannual variability in snow cover at the flux measurement sites may
play a role in causing these wintertime differences, although Clifton et al. (2023) found that the point models results were not
very sensitive to snow cover. The results of the grid model and point model analysis agree on the non-negligible wintertime
contribution of the lower canopy pathway for the models that consider it, i.e. the GEM-MACH Wesely (20-40% across the
corresponding grid and point models) and WRF-Chem Wesely (10-14% across the corresponding grid and point models) point
395 models and their corresponding grid model implementations.

Figures S7, S8, and S9 show corresponding results for evergreen needleleaf forest, broadleaf deciduous forest, and grassland,
respectively. For each of these LU cases, the grid model results reflect simulated LU-specific V_d and effective conductances
averaged over all grid cells with coverage greater than 85% for that LU category for a given model. The point model results
adapted from Figure 5 of Clifton et al. (2023) are for Hyytiälä (evergreen needleleaf forest), Ispra (deciduous broadleaf forest),
400 and Bugacpuszta and Easter Bush (both grassland)[1]. The results for evergreen needleleaf and deciduous broadleaf forest in
Figures S7 and S8 are broadly consistent with those discussed above for mixed forest. In particular, agreement between grid
and point modeling results is generally better during summer than winter, especially in terms of the GEM-MACH Wesely vs.
GEM-MACH Zhang comparison. The non-negligible contribution of the wintertime lower canopy effective conductance
simulated by GEM-MACH Wesely and, to a lesser extent, WRF-Chem Wesely discussed above for mixed forest is also visible
405 in the deciduous broadleaf forest results, for both grid and point models, while it is less pronounced for evergreen needleleaf
forest. The model-to-model comparisons for grassland show consistent behavior between the grid model and point model
analyses for both winter and summer. During winter GEM-MACH Zhang and WRF-Chem Wesely show the highest V_d while

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[1] We note that the LU present at these observation sites may not necessarily represent the dominant land use at the broader
grid scale used in this analysis, and this may account for some of the residual differences between the grid model results
extracted for specific LU classes and the point model results at these sites. Ispra flux tower (45.812495N, 8.634771E) is within
a forest of about 300m extent surrounded by the Joint Research Centre and the town of Ispra, and Easter Bush Field site
(55.865N, 3.206W) is located 200m from the University of Edinburgh's Easter Bush Campus. The Hyytiälä site is more
representative of the evergreen needleleaf forest on the grid cell scale, with the nearest changes in LU occurring ~800m from
that site, and Bugacpuszta is also more representative of the larger region, which contains largely farmland/grassland with
some trees.

GEM-MACH Wesely shows the lowest V_d. During summer, GEM-MACH Wesely V_d exceeds GEM-MACH Zhang V_d for both the grid model results and both grassland point intercomparison sites, though summertime GEM-MACH Zhang V_d for
410 grassland is not as low relative to other models as for the different forest LU categories discussed above. Moreover, both grid model and point model results for summertime also agree that GEM-MACH Zhang has the largest relative contribution of the stomatal effective conductance to V_d for grassland.

The results presented above demonstrate that despite differences in LU in the region of the observation sites at the scale of model grid cells, the analysis of O_3 dry deposition schemes implemented in grid models can successfully be linked to the
415 detailed point model evaluation using long-term O_3 flux measurements presented in Clifton et al. (2023) when generating LU-specific diagnostic information and limiting the analysis to specific LU categories. This in turn allows the developers of grid models to leverage process-level insights gained from point intercomparison studies (Clifton et al., 2023; Khan et al., 2024) for improving the representation of dry deposition in their modeling systems. However, as also discussed in the first part of this section, differences in LU characterizations between models can complicate a process-level attribution of differences
420 across models to differences in deposition schemes, especially when considering LU-specific dry deposition fluxes. Similarly, observation sites with a variety of LU within a short distance of the measurement location itself may be less useful for evaluating LU-specific aspects of deposition algorithms. Model differences in LU distributions and their effects on deposition fluxes are analyzed in the next section.

3.3 The Influence of Land Use Data on Ozone Deposition

425 Table 4 shows the native LU categories used in each model's dry deposition calculations and how these categories were mapped to the 16 common AQMEII4 LU categories defined in Galmarini et al. (2021) when reporting LU-specific diagnostics. For some models (WRF/CMAQ (M3DRY), GEM-MACH (Zhang), WRF-Chem (RIFS), and WRF-Chem (UPM)), the native LU categories used in the dry deposition calculations are identical to those used in the land-surface model (LSM) of the driving meteorological model. For the other models, the LU classification scheme differed between the LSM in the driving
430 meteorological model and the dry deposition calculations, and Tables S1 – S5 provide details on the internal mapping between the LSM and dry deposition calculations implemented in these models. We note that several AQMEII4 LU categories are held in common with most model native LU categories (e.g. "evergreen needleleaf trees") while others were less direct, requiring assignment into the nearest AQMEII4 LU category as documented in these Tables.

Figures 11 and 12 show bar charts of the distribution of all 16 AQMEII4 LU categories for each model across all common
435 grid cells that aren't dominated by water for the NA and EU domains. These charts reveal that 7 of the 16 categories (barren, evergreen needleleaf forest, deciduous broadleaf forest, mixed forest, shrubland, planted/cultivated land, and grassland) account for roughly 90% of all LU over both NA and EU. Over NA, the fractional coverage of some categories like evergreen needleleaf forest are similar across models, as might be expected from the commonality of this LU within the native LU categories of Table 4. On the other hand, there is considerable disagreement between models for the fractional coverage of

AQMEII4	WRF/CMAQ (M3Dry)	WRF/CMAQ (STAGE)	GEM-MACH (BASE) and GEM-MACH (Ops)	GEM-MACH (Zhang)	WRF-Chem (RIFS), WRF-Chem (UPM), and WRF-Chem (NCAR)	LOTOS/EUROS
1: Water	17: water	1: Water	13: Inland water; 14: Ocean	1: Water (Ocean); 3: Inland Lake (Fresh)	16: Water Bodies	09: Inland Water; 10: Sea Water
2: Developed / Urban	13: Urban and Built-up	2: Developed / Urban	15: Urban	21: Urban	1: Urban and Built-Up Land	08: Urban
3: Barren	16: Barren or Sparsely Vegetated	3: Barren	8: Desert	24: Desert	19: Barren or Sparsely Vegetated; 23: Bare Ground Tundra	05: Bare rock
4: Evergreen needleleaf forest	1: Evergreen Needleleaf Forest	4: Evergreen needleleaf forest	1: Evergreen needle-leaf forest	4: Evergreen needleleaf trees	14: Evergreen Needleleaf Forest	02: Coniferous forest
5: Deciduous needleleaf forest	3: Deciduous Needleleaf Forest	5: Deciduous needleleaf forest	3: Deciduous needle-leaf forest	6: Deciduous needleleaf trees	12: Deciduous Needleleaf Forest	N/A
6: Evergreen broadleaf forest	2: Evergreen Broadleaf Forest	6: Evergreen broadleaf forest	2: Evergreen broadleaf forest	5: Evergreen broadleaf trees; 8: Tropical broadleaf trees	13: Evergreen Broadleaf Forest	N/A
7: Deciduous broadleaf forest	4: Deciduous Broadleaf Forest	7: Deciduous broadleaf forest	4: Deciduous broadleaf forest	7: Deciduous broadleaf trees; 9: Drought deciduous trees	11: Deciduous Broadleaf Forest	04: Deciduous forest
8: Mixed forest	5: Mixed Forest	8: Mixed forest	5: Mixed Forest	25: Mixed Wood Forest	15: Mixed Forest	N/A
9: Shrubland	6: Closed Shrublands; 7: Open Shrublands	9: Shrubland	10: Dwarf trees, shrubs	10: Evergreen broadleaf shrubs; 11: Deciduous shrubs; 12: Thorn shrubs; 26: Mixed shrubs	8: Shrubland; 9: Mixed Shrubland/Grassland [WRF-Chem(NCAR) mapping]	N/A
10: Herbaceous	N/A	10: Herbaceous	N/A	13: Short grass and forbs	N/A	N/A
11: Planted / Cultivated	12: Croplands; 14: Cropland-Natural Vegetation Mosaic	11: Planted / Cultivated	7: Crops, mixed farming	15: Crops; 16: Rice; 17: Sugar; 18: Maize; 19: Cotton; 20: Irrigated crops	2: Dryland Cropland and Pasture; 3: Irrigated Cropland and Pasture; 4: Mixed Dryland/Irrigated Cropland and Pasture; 5: Cropland / Grassland Mosaic; 6: Cropland / Woodland Mosaic	01: Arable land; 03: Permanent crops
12: Grassland	10: Grasslands	12: Grassland	6: Grassland	14: Long grass	7: Grassland	06: Grassland
13: Savanna	8: Woody Savanna; 9: Savanna	13: Savanna	N/A	N/A	9: Mixed Shrubland / Grassland [WRF-Chem(RIFS) and WRF-Chem(UPM) mapping]; 10: Savanna	11: Semi-natural vegetation
14: Wetlands	11: Permanent Wetlands	14: Wetlands	11: Wetland with plants	22: Swamp	17: Herbaceous Wetland; 18: Wooded Wetland	
15: Tundra	18: Wooded Tundra; 19: Mixed Tundra; 20: Barren Tundra	15: Tundra	9: Tundra	22: Tundra	20: Herbaceous Tundra; 21: Wooded Tundra; 22: Mixed Tundra	
16: Snow and Ice	15: Snow and Ice	16: Snow and Ice	12: Ice caps and glaciers	2: Ice	24 Snow or Ice	07: Ice

Table 4. Mapping of LU classes from the categories used in the models' dry deposition code to the 16 AQMEII4 categories.

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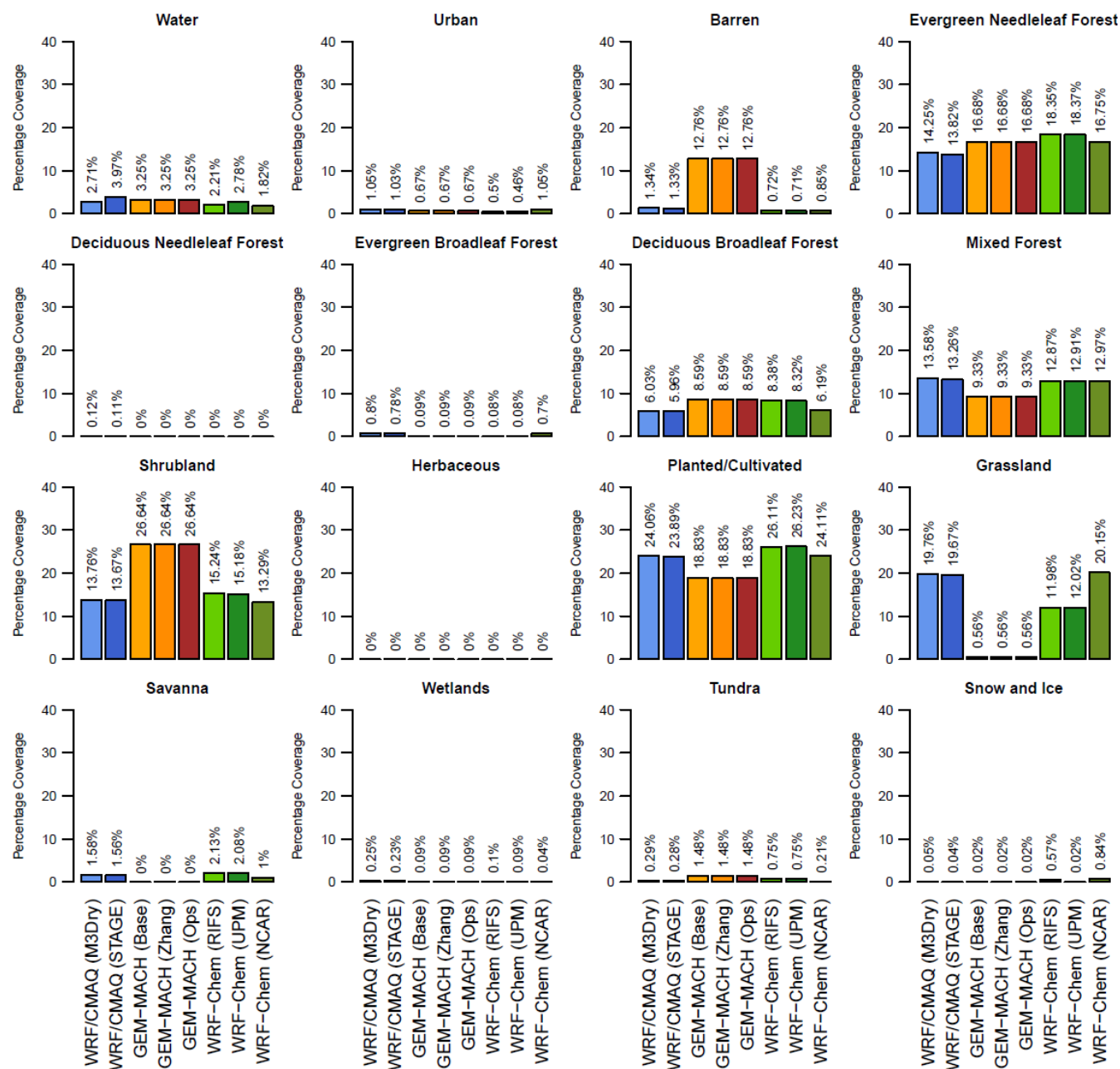
Figure 11. Fraction of the NA domain common to all models covered by each AQMEII4 LU category, excluding grid cells dominated by water by each model.
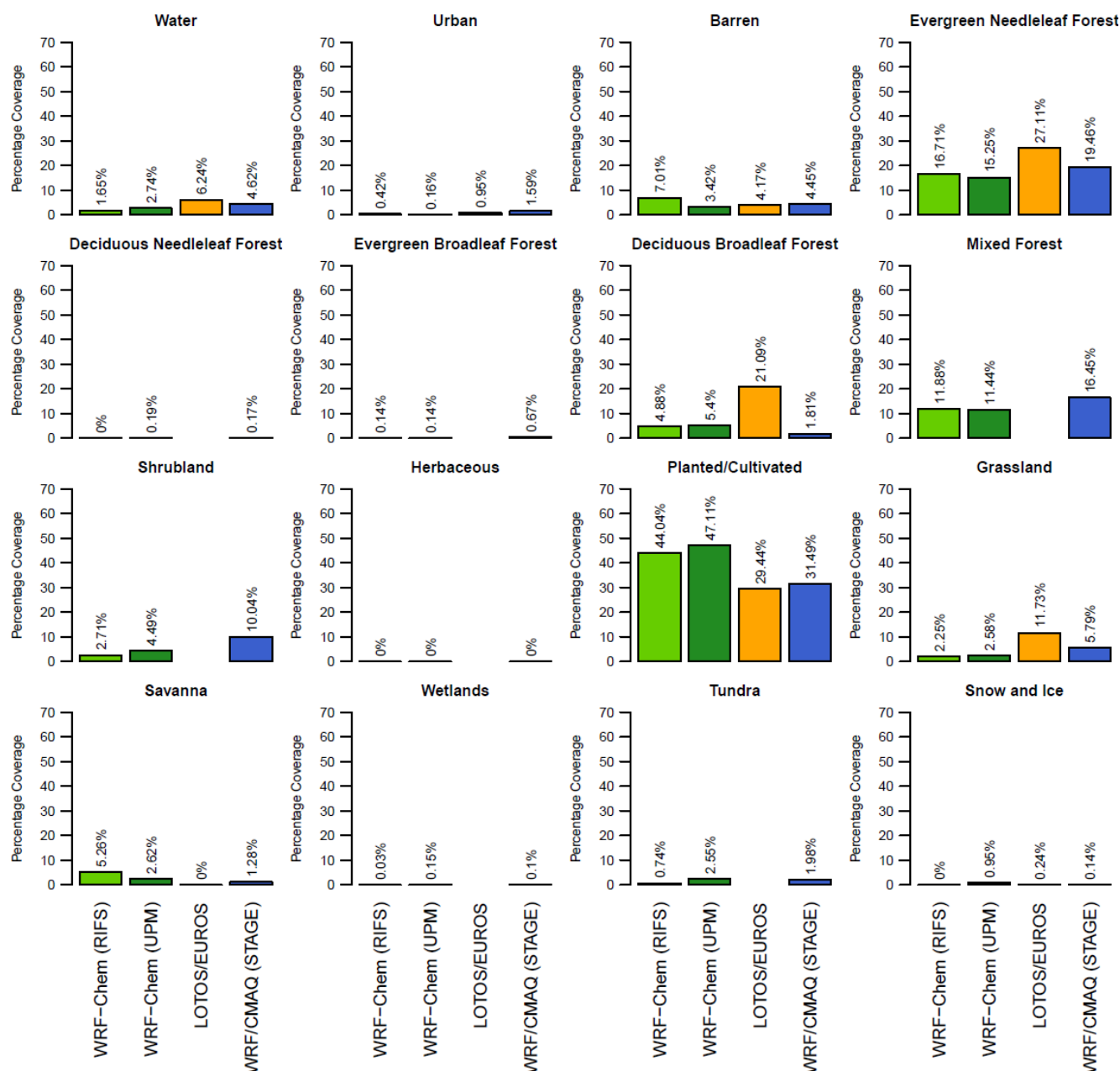
Figure 12. Fraction of the EU domain common to all models covered by each AQMEII4 LU category, excluding grid cells dominated by water by each model.

many other categories over NA, especially non-forest categories such as barren, shrubland, planted/cultivated land, and grassland. Much of this disagreement is caused by the GEM-MACH simulations having larger fractions of the barren and shrubland categories and smaller fractions for the planted/cultivated and grassland categories than either the WRF/CMAQ or

450   WRF-Chem simulations, pointing to ambiguities in classifying non-forest partially vegetated areas[2]. Over EU, the primary driver of model variability in the LU used for deposition calculation is LOTOS/EUROS which in its DEPAC dry deposition module splits mixed forest from the driving meteorological model equally between coniferous and deciduous forest and includes shrubland in the category of semi-natural vegetation and as a result shows larger coverage than WRF/CMAQ and WRF-Chem for evergreen needleleaf forest, deciduous broadleaf forest, and grassland. In addition, while all models agree that

455   planted/cultivated is the dominant category over EU, the lowest coverage (29.44%, LOTOS/EUROS) and highest coverages (47.11%, WRF-Chem (UPM)) differ by a factor of 1.6. Here it should be noted that LOTOS/EUROS is the only model using the official LU data of the European Union (in comparison to USGS for the WRF-CHEM simulations).

    Even for categories for which models show relatively close agreement of the total domain-wide coverage in Figures 11 and 12, spatial patterns of these coverages may still differ between models. To illustrate this, Figures S10 and S11 show maps of

460   the fractional coverage of the evergreen needleleaf forest category for each model over NA and EU. One fundamental difference between the WRF-Chem simulations and all other simulations is that the WRF-Chem simulations employed a dominant LU category approach in their LSM and dry deposition calculations (that is, only the LU with the largest LU fraction within a grid cell is used to represent that grid cell's LU for deposition calculations) while all other simulations accounted for subgrid variations in LU by employing a fractional LU category approach. Therefore, Figures S10 and S11 show evergreen

465   needleleaf forest fractions of either 0 or 1 for the WRF-Chem simulations and fractions between 0 and 1 for all other simulations. Both figures reveal that, despite all native LU databases including evergreen needleleaf forest as an explicit LU category, the coverage for this LU can vary substantially between models, e.g. over the Southeastern U.S. in the NA domain (Figure S10) and Central Europe and the Iberian Peninsula in the EU domain (Figure S11).

    To analyze the level of agreement in spatial coverage across models for all LU categories, while taking into account that the

470   WRF-Chem simulations used a dominant LU category approach, we applied two metrics to assess model-to-model agreement for a given LU category and grid cell. The first metric simply determines whether all models agree that the LU category being assessed is the category with the highest fractional coverage (i.e. the dominant category) compared to all other categories in that grid cell, regardless of the actual fractional coverage for that dominant category for those simulations that use a fractional coverage approach. The second metric builds upon the first metric by not only determining whether the LU category being

475   assessed is the dominant category in that grid cell, but also whether all models agree that its fractional coverage is at least 85%. By definition, all grid cells meeting the more stringent second metric also meet the first metric. Identifying grid cells meeting

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[2] A subsequent investigation of the GEM-MACH land use database suggested that this difference is due to an underlying dataset which included "short grass and forbs" being aggregated to "Dwarf trees and shrubs", in turn increasing the relative assignment to AQMEII4 shrubland, and decreasing the assignment to AQMEII4 grassland categories. This misassignment has been corrected in implementations of GEM-MACH subsequent to that used in AQMEII4.

	NA, # of common cells with "dominant coverage" for LU category for all models	NA, # of common cells with LU category coverage > 85% for all models	EU, # of common cells with "dominant coverage" for LU category for all models	EU, # of common cells with LU category coverage > 85% for all models
Water	27,925 (25.84%)	26,949 (24.94%)	81,423 (46.00%)	78,186 (44.17%)
Developed-Urban	161 (0.15%)	32 (0.03%)	63 (0.04%)	4 (0%)
Barren	112 (0.10%)	14 (0.01%)	2,203 (1.25%)	1,310 (0.74%)
Evergreen needleleaf forest	7,111 (6.58%)	1,544 (1.43%)	9,270 (5.237%)	2,531 (1.43%)
Deciduous needleleaf forest	0 (0%)	0 (0%)	0 (0%)	0 (0%)
Evergreen broadleaf forest	1 (0%)	0 (0%)	0 (0%)	0 (0%)
Deciduous broadleaf forest	2,663 (2.46%)	581 (0.54%)	362 (0.21%)	5 (0%)
Mixed forest	3,968 (3.67%)	705 (0.65%)	0 (0%)	0 (0%)
Shrubland	757 (0.70%)	43 (0.04%)	0 (0%)	0 (0%)
Herbaceous	0 (0%)	0 (0%)	0 (0%)	0 (0%)
Planted/Cultivated	10,640 (9.85%)	6,130 (5.67%)	24,627 (13.91%)	6,108 (3.45%)
Grassland	164 (0.15%)	52 (0.05%)	147 (0.08%)	0 (0%)
Savanna	0 (0%)	0 (0%)	0 (0%)	0 (0%)
Wetlands	1 (0%)	0 (0%)	0 (0%)	0 (0%)
Tundra	83 (0.08%)	0 (0%)	0 (0%)	0 (0%)
Snow and Ice	1 (0%)	0 (0%)	0 (0%)	0 (0%)
Total of grid cells with common LU (incl. water cells)	53,587 (49.59%)	36,050 (33.36%)	118,095 (66.72%)	88,144 (49.80%)
Total of grid cells with diverging LU (incl. water cells)	54,471 (50.41%)	72,008 (66.64%)	58,899 (33.28%)	88,850 (50.20%)
Total of grid cells with common LU (excl. water cells)	25,662 (32.02%)	9,101 (11.22%)	36,672 (38.37%)	9,958 (10.08%)
Total of grid cells with diverging LU (excl. water cells)	54,471 (67.98%)	72,008 (88.78%)	58,899 (61.63%)	88,850 (89.92%)

Table 5. Number and percentage of grid cells within the common NA and EU analysis domains where all models agree on the LULC category for that grid cell. Two metrics are used to assess agreement: 1) all models agree that the LULC category being assessed is the category with the highest fractional coverage compared to all other categories in that grid cell ("dominant coverage"), and 2) in addition to meeting metric 1, all models also agree that a given grid cell has at least 85% coverage for the LULC category being assessed. The last four rows summarize the level of agreement across either all 16 LULC categories (including water) or all 15 non-water LULC categories. The percentages shown in the rows corresponding to individual LULC categories are calculated with respect to all grid cells in the common analysis domains (108,058 for NA and 176,994 for EU).

485

the first metric can be thought of as a way to assess agreement in LU categories across models if all simulations (not only WRF-Chem) had used a dominant LU category approach. The subset of grid cells identified by metric 1 for a given LU category that also satisfies the >85% criterion defined for metric 2 can be thought of as the common "dominant" cells for that LU category in which even the computations performed by the models using a fractional LU category approach were mostly

490   impacted by the physical characteristics of that LU category, with only minor impacts from other LU categories possibly also present in the grid cell. This subset of grid cells was used in Section 3.2 when comparing V_d between the point model simulations at specific flux measurement sites and the grid model simulations.

The results of applying metrics 1 and 2 to all LU categories and models over both NA and EU are shown in Figures S12-S13 and Table 5 which lists the number and percentage of grid cells within the common NA and EU analysis meeting each metric.

495   The first sixteen rows of Table 5 contain results for each of the AQMEII4 LU categories. The percentages shown in these rows are calculated with respect to the total number of grid cells in the common analysis domains (108,058 for NA and 176,994 for EU), including water grid cells. Water is by far the category with the highest level of agreement over both NA and EU with only minor differences between the two metrics, indicating that most grid cells dominated by water across all models are almost fully (> 85%) or often fully covered by water, reflecting the oceans and open waters present in both analysis domains.

500   The LU categories with the second- and third-largest amount of agreement between models are planted/cultivated land and evergreen needleleaf forest over both domains. Over NA, the deciduous broadleaf and mixed forest LU categories also have about 600 – 4,000 [0.5% - 4%] common grid cells depending on metric and category. For the remaining LU categories, the number and percentage of grid cells matching across models is very low, especially for the more stringent metric 2. While to some extent this is expected given the low overall domain coverage of some of these categories by all models (Figures 11-12),

505   this is also the case for shrubland and grassland which has substantial domain-wide coverage over NA (Figure 11).

The last four rows of Table 5 summarize the level of agreement across either all 16 LU categories (including water) or the 15 non-water LU categories. Figures 13a-d depict the location of common grid cells for both metrics and continents. In these figures, grid cells meeting the metric for any LU category are colored in dark red while grid cells not meeting it for any LU category (i.e. grid cells without common LU category as measured by the metric) are colored in white. These figures along

510   with Table 5 illustrate that even for the less restrictive metric 1, 68% (62%) of non-water grid cells over NA (EU) do not share a common dominant LU category across models. Over NA, many of these non-matching grid cells are located in the southern and western portions of the domain while over EU, they are most prevalent in the western, northeastern, and southeastern portions of the domain. When considering the more stringent metric 2, i.e. grid cells in which the common category has at least 85% coverage for all models, this number of grid cells with diverging LU categories increases to roughly 90% of non-

515   water grid cells over both domains. The only areas with significant contiguous clusters of such common cells are the agricultural regions in the north-central NA domain and portions of central Europe and Sweden.
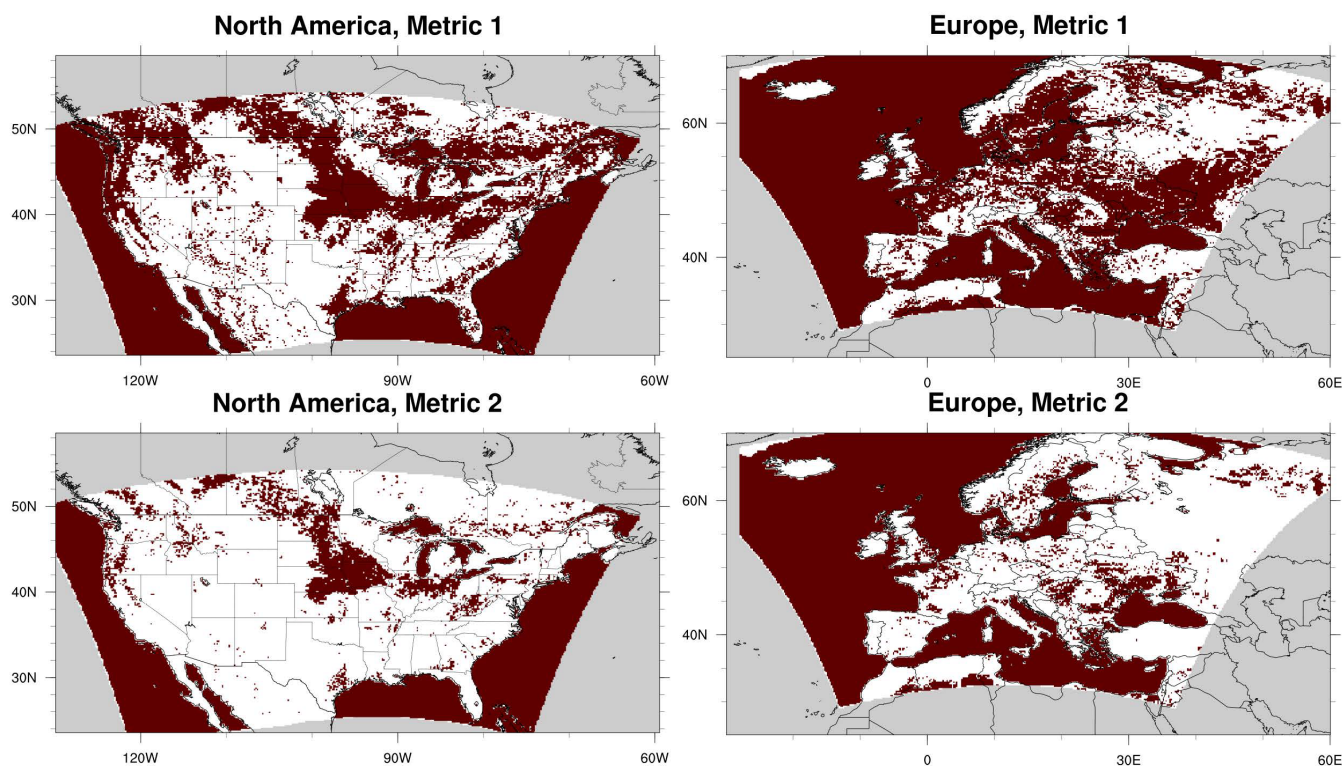
Figure 13. Red areas indicate grid cells meeting metrics 1 and 2 used to assess LU communality across models as described in the text.

520 This low number of grid cells with a common LU category strongly suggests that differences in LU coverage can contribute to or even drive differences in LU-specific dry deposition fluxes, in addition to any differences in process representation that exist between different models. To investigate this, Figure 14 compares LU-specific dry deposition fluxes, V_d, and LU fractions over NA for 7 selected LU categories. The LU-specific dry deposition fluxes (V_d) represent annual totals (means) over all grid cells in which the LU category being assessed has a fractional coverage of at least 85% for a given model. For some LU
525 categories with relatively similar total coverage across the domain (e.g. evergreen needleleaf forest and deciduous broadleaf forest), differences in total O_3 dry deposition flux to that LU category closely mirror differences in LU-specific average V_d. For mixed forest, the lower fractional coverage for GEM-MACH (Base) and GEM-MACH (Ops) leads to a below-average total deposition flux to that category despite V_d being the highest. For the grassland and barren categories, differences in their dry deposition fluxes are almost entirely driven by differences in LU fractional coverage rather than differences in V_d.
530 Corresponding results for the EU domain are shown in Figure S14 and confirm that differences in both V_d and LU coverage contribute to differences in LU-specific dry deposition fluxes.
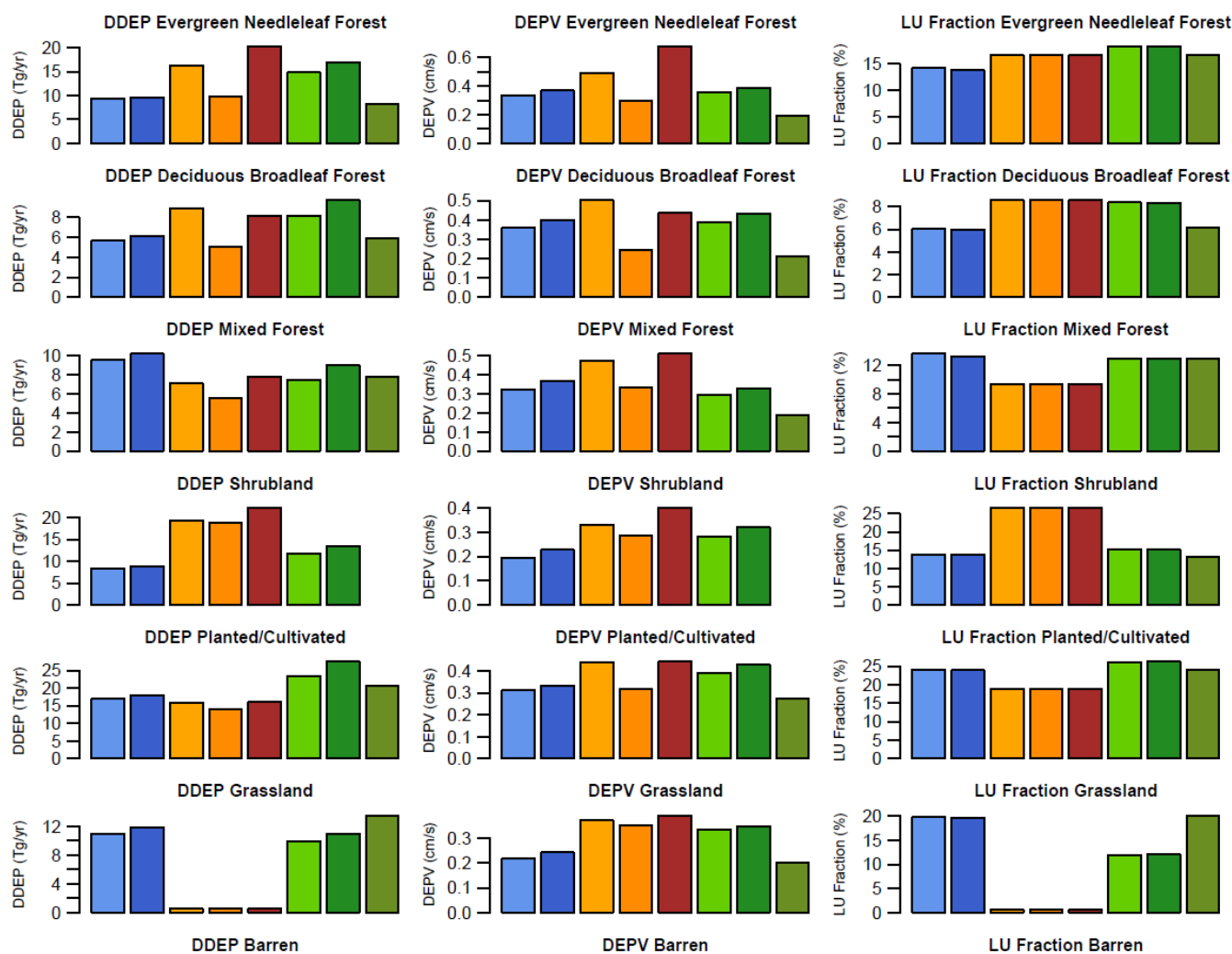
Figure 14. LU-specific annual domain-total dry deposition fluxes (Tg), LU-specific annual mean dry deposition velocity (cm/s), and percentage LU category domain coverage (excluding water grid cells) for seven selected LU categories over the NA domain. For each LU category and model, the analysis considered grid cells in the analysis domain common to all models in which a given model had at least 85% coverage for this LU category. The number of these grid cells differs across models due to underlying differences in LU (see Section 3.3).
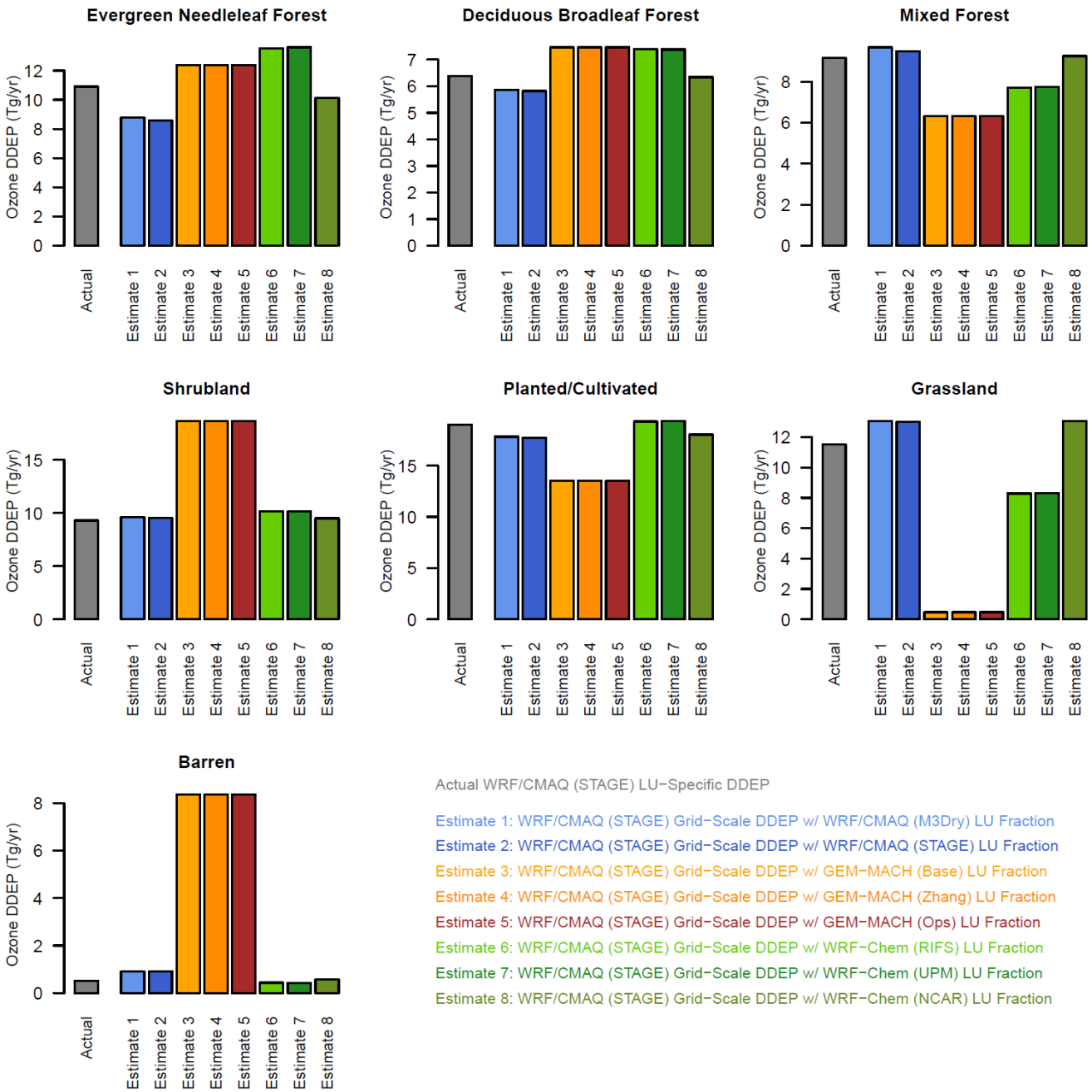
Figure 15. 2016 NA annual domain-wide total ozone dry deposition fluxes (Tg/yr) for selected LU categories. The grey bar shows LU-specific fluxes calculated by WRF/CMAQ (STAGE) while the colored bars show LU-specific fluxes estimated by combining WRF/CMAQ (STAGE) grid-scale fluxes with LU fractions from other models as described in the text.

The results shown in Figure 14 and S14 have important implications for computing estimates of deposition fluxes to specific
545   ecosystems. In past model intercomparison studies such as those performed under the umbrella of the Task Force on
Hemispheric Transport of Air Pollution (TF-HTAP, http://www.htap.org), such estimates have often been computed through
post-processing by apportioning archived modeled grid-aggregated dry deposition fluxes to specific LU categories using a
fixed LU database (e.g. Hardacre et al., 2015; Schwede et al., 2018). While this approach makes use of actual modeled dry
deposition fluxes rather than using modeled concentrations as inputs to offline dry deposition calculations (e.g. Van Dingenen
550   et al., 2009; Avnery et al., 2011), it may still be subject to uncertainties arising from differences between grid-aggregated vs.
LU-specific dry deposition fluxes as well as differences in LU categorization – a flux associated with a model LU category for
model-internal deposition may be aggregated under a different LU category in post-processing using a different post-
processing database. The diagnostic information collected for AQMEII4 and analyzed in this manuscript allows us to illustrate
and quantify both uncertainties.Figure 15 compares the LU-specific annual total dry deposition fluxes to seven LU categories
555   calculated by model WRF/CMAQ (STAGE) (gray bars) to estimates derived by combining grid-aggregated dry deposition
fluxes from the same model with LU fractions from all models (colored bars). Comparing the gray bars to the dark blue bars
shows the impact of computing actual LU-specific deposition fluxes within the model simulation vs. estimating them by
linearly scaling grid-aggregated values using the model's own LU fractions. This effect is relatively small for most LU
categories but reaches about 20% for evergreen needleleaf forest and 40% for barren. Comparing the grey bars to all the other
560   colored bars (except dark blue) shows the combined uncertainty of using a linear scaling of grid-aggregated deposition values
and using different LU datasets than the one used within the model to calculate the LU-specific value. These differences are
generally much larger and reflect the substantial divergence in LU categories across models, due to the variety of native-mode
LU databases in use between different models. This example demonstrates the advantage of computing and collecting LU-
specific diagnostic dry deposition information in model intercomparison studies so that model-to-model differences in
565   deposition estimates can be tied to differences in model-specific process representation and LU datasets. Potential future work
to harmonize LU datasets across models would allow further constraining differences to process representations only. This in
turn would make it easier to leverage insights gained from point intercomparison studies (Wu et al., 2018; Clifton et al., 2023;
Khan et al., submitted) for improving the representation of dry deposition in grid models.

4 Summary

570   This study presented a diagnostic analysis of ozone dry deposition from annual AQMEII4 photochemical grid modeling
simulations performed over NA and EU. Simulated annual O_3 dry deposition fluxes ranged from 59.5 Tg/yr to 91.4 Tg/yr over
the NA domain and from 47.2 Tg/yr to 82.9 Tg/yr over the EU domain. Analysis of grid-aggregated effective conductances
showed that models with similar V_d can exhibit significant differences in the absolute and relative contributions of different
pathways to V_d. The availability of effective conductances also allowed an attribution of model differences in V_d to specific
575   processes. For example, GEM-MACH (Base), GEM-MACH (Ops), and WRF-Chem (RIFS) all have similar soil and lower

canopy effective conductances, revealing that the differences in V_d between these simulations stem from differences in the cuticular and, to a lesser extent, stomatal pathways.

Analysis of LU-specific V_d and effective conductances revealed a general increase in model spread compared to analyzing grid-aggregated values, especially for V_d and the cuticular and stomatal effective conductances over forested and agricultural
580   LU types. This finding suggests that LU-dependent parameter choices and the representation of vegetation effects on O_3 dry deposition are a significant source of variability in grid-aggregated V_d while also indicating that an analysis of only grid-aggregated deposition diagnostics can mask process-specific differences that exist between schemes. Utilizing the LU-specific diagnostics available from the grid model simulations also provided an opportunity to compare these diagnostics to the results from the single-point model intercomparison study by Clifton et al. (2023). Results showed good agreement between the
585   behavior of different dry deposition schemes in single-point models at individual sites vs. their implementation in grid models in aggregate across either the NA or EU domain for a given LU type. For example, both grid model and point model V_d ranged between 0.4 and 1.0 cm s-1 across models for mixed forest grid model results and the corresponding point model results at Borden Forest and Harvard Forest, with the lowest V_d associated with GEM-MACH Zhang and the highest V_d associated with GEM-MACH Wesely in both comparisons. Grid and point models also agreed that WRF/CMAQ M3Dry had a much smaller
590   contribution of the soil effective conductance to V_d compared to all other models and that WRF-Chem Wesely had a smaller contribution of the cuticular effective conductance to V_d compared to all other models. This demonstrates that the analysis of O_3 dry deposition schemes implemented in grid models can successfully be connected to detailed point model evaluation studies using long-term O_3 flux measurements when generating LU-specific diagnostic information and limiting the analysis to specific LU categories. This in turn allows the developers of grid models to leverage process-level insights gained from
595   point intercomparison studies for improving the representation of dry deposition in their modeling systems. The importance of observation sites being representative of a single LU classification for this purpose has also been noted in this work.

This study also presented a detailed analysis of different models' LU distributions that revealed substantial differences in the spatial patterns and sometimes also the domain-wide coverage of certain LU categories over both domains, especially non-forest partially vegetated categories such as agricultural areas, shrubland, and grassland. Overall, 68% (62%) of non-water grid
600   cells over NA (EU) were found not to share a common dominant LU category across models. When considering only grid cells in which the common dominant category has at least 85% coverage for all models, this number of grid cells with diverging LU categories increased to roughly 90% of non-water grid cells over both domains. By comparing LU distributions, LU-specific V_d, and LU-specific dry deposition fluxes, we demonstrate that differences in LU coverage can contribute to or even drive differences in LU-specific dry deposition fluxes, in addition to any differences in process representation that exist
605   between different models. For mixed forest, the lower fractional coverage for GEM-MACH (Base) and GEM-MACH (Ops) leads to a below-average total deposition flux to that category despite V_d being the highest. Differences in the grassland and barren dry deposition fluxes over the NA domain are almost entirely driven by differences in LU fractional coverage rather than differences in V_d. On the other hand, for LU categories that have similar total domain-wide coverage across models (e.g.

evergreen needleleaf forest and deciduous broadleaf forest), differences in total O_3 dry deposition fluxes to that LU category
610   closely mirror differences in LU-specific average V_d, indicating that in this case differences in process representation drive
differences in fluxes.

Finally, we contrasted the LU-specific dry deposition fluxes simulated in AQMEII4 to estimates in which grid-aggregated
fluxes were linearly apportioned to specific LU categories based on LU fractions. When using a consistent set of LU
information between the direct simulation of LU-specific fluxes and the apportionment of grid-aggregated fluxes to LU
615   categories based on LU fractions, differences in domain-wide LU-specific deposition estimated by both approaches were
generally within 10% but could be as high as 40% for a given LU category. These variations are caused by the non-linear
effects of subgrid LU variability on V_d and deposition fluxes. When allowing for differences in LU distributions between those
used in the grid model simulations generating the LU-specific results and those used to apportion grid-aggregated results to
specific LU categories, domain-wide LU-specific deposition estimates differed by 50% or more for a number of categories.
620   This illustrates that inconsistencies in LU representation between the model simulations and the post-processing apportionment
approach can introduce substantial uncertainties in estimates of fluxes to specific ecosystems.

Overall, the results in this study demonstrate that the generation of grid-aggregated and LU-specific diagnostic outputs in
model application and intercomparison studies can provide valuable information from both a model development point of view
(to better understand drivers of model variability and identify priorities for model improvement) and an impacts analysis point
625   of view (to distinguish between stomatal vs. non-stomatal deposition and quantify deposition to specific LU types). However,
it should also be acknowledged that designing, implementing, collecting, and analyzing these diagnostics in AQMEII4 was
resource-intensive, required a high level of coordination across research groups, and resulted in several iterations before the
analyses presented in this and other AQMEII4 manuscript could be completed. Finally, the results of the LU analysis highlight
the importance of documenting and analyzing the representation of LU across models. Future work should be aimed at
630   harmonizing this aspect when using CTMs for deposition analyses, in addition to the current standard practice of harmonizing
emissions and boundary conditions in intercomparison studies.

Data availability

All data used in this work are available upon request from the authors. Please contact the corresponding author to request any
data related to this work.

Author contributions

CH led the development of this manuscript and conducted most of the analyses presented in Section 3. CH, SG, PM, IK, OEC, and DBS helped conceptualize both the AQMEII4 model intercomparison framework and the analyses presented in this study. OEC conducted the analysis of point model simulations. UA, JOB, TB, PC, AH, CH, RK, AL, PM, KM, JLPC, JEP, YHR, RSJ, and RS conducted grid model simulations. JOB, PC, PM, JEP, and RSJ conducted point model simulations. SG, Roberto Bellasio, and Roberto Bianconi developed and maintained infrastructure for data exchange and storage. All authors contributed to the editing of the manuscript.

Competing Interests

One of the co-authors (Stefano Galmarini) is an ACP editor.

Disclaimer

The views expressed in this article are those of the authors and do not necessarily represent the views or policies of the U.S. Environmental Protection Agency.

Special issue statement

This manuscript is part of the special issue "AQMEII4: a detailed assessment of atmospheric deposition processes from point models to regional-scale models".

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