distributions of O_3 deposition to the major land cover classes, quantifies the contributions of stomatal versus non-stomatal pathways, identifies the regions in the world with the largest interannual variability in $V_{d,O3}$, and draws implications concerning the deployment of future measurements. **In Section 6,** we examine the influence on surface O_3 simulations from the shift of $V_{d,O3}$ from the Wesely scheme to the new scheme as implemented in GFDL LM4.0. Specifically, we leverage the Tropospheric Ozone Assessment Report (TOAR) Surface Ozone Database with vast spatial coverage around the world (Schultz et al., 2017) and a new dataset over China (http://106.37.208.233:20035/) to assess the two deposition schemes. Finally, we synthesize in **Section 7** the model strengths and limitations, discuss the implications, and make recommendations for future O_3 flux measurements.

2. Methods

2.1 Ozone dry deposition observations

[Table 1 about here]

We compile a suite of field-based $V_{d,O3}$ observations at 41 locations, obtained from 26 literature sources published between 1990 and 2018 (**Table 1**). Model evaluations are conducted on a site-by-site basis for the purpose of examining the influence from regional to local meteorology and land cover. Sites with continuous measurements for at least two years are used to evaluate the seasonal cycle of $V_{d,O3}$ (**Section 3**). To explore the influence of water availability on $V_{d,O3}$ seasonality, observations are separated into the dry and wet season for evergreen forest sites in Mediterranean Europe (Castelporziano, Italy), South Asia (Mea Moh and Datum Valley), and the Amazon. Multi-year measurements at a boreal forest in Denmark (1996-2000) and a deciduous forest in Ontario Canada (2008-2013) are analyzed for the influence of drought stress on $V_{d,O3}$ interannual variability (**Section 4**). For short-term observations, we focus on daytime average (9am-3pm) for the growing season (June-July-August) to evaluate the modeled spatial variability of $V_{d,O3}$ across North America and Europe (**Section 5**). For comparison with observations, we sample modeled $V_{d,O3}$ to the land-cover tile that best matches the observed vegetation type at individual sites (as opposed to using a grid-cell average). Given the heterogeneity of land surface properties and the uncertainty in both the land model forcing dataset and O_3 flux measurements themselves at finer temporal scales (i.e., daily to weekly), we focus on evaluating the most salient processes influencing seasonal to interannual variability in $V_{d,O3}$.

2.2 Model formulations and experiments

Paulot et al. (2018) developed an interactive dry deposition scheme in GFDL LM3.0 and evaluated the dry deposition velocities and fluxes of reactive nitrogen species. Here we evaluate and improve the dry deposition scheme for O_3 in LM3.0 and an updated version of the land model, LM4.0. LM4.0 is a new model of terrestrial water, energy, and carbon, intended for use in global hydrological analyses and as a component of GFDL earth system and physical climate models contributing to the Coupled Model Intercomparison Project, Phase 6 (CMIP6) (Zhao et al., 2018a; b). Both LM3.0 and LM4.0 include five vegetation types (C3 and C4 grasses, and temperate deciduous, tropical and cold evergreen trees) and describe small-scale heterogeneity of land surface cover in each grid cell using a mosaic approach, as a combination of sub-grid tiles in four land use categories: lands undisturbed by human activity (i.e., "primary" or "natural"), cropland, pasture, and lands harvested at least once (i.e., "secondary"), including managed forests and abandoned croplands and pastures (Shevliakova et al., 2009; Malyshev et al., 2015). Planting and harvesting dates for crops as well as pasture grazing are updated as described by *Paulot et* *al.* (2018). Neither of the land model configurations used in this study includes treatment of irrigation or of nitrogen limitation on plant growth.

LM3.0 uses a 2° latitude x 2.5° longitude grid and is configured similarly to the land component of GFDL ESM2Mb (Dunne et al., 2012; Malyshev et al., 2015), except for the updates on cropping dates and pasture grazing. LM4.0 employs a cubed-sphere grid resolution of ~100x100 km^2 and serves as the land component for the new set of GFDL AM4/CM4 models (Zhao et al., 2018a; b). Motivated by biases in LM3.0 simulations, the standard version of LM4.0 includes the following updates: (1) decreasing the cold season length threshold to better locate the cold evergreen–temperate deciduous forest boundary; (2) decreasing critical leaf temperature to better match the seasonal green-up as inferred from MODIS reflectances; (3) using a more physically based approach for drought-induced leaf drop; (4) changing soil types and parameters affecting surface albedo, plant hydraulics and biogeography (see Section 10 in Zhao et al., 2018a); (5) limiting the maximum LAI attainable by the vegetation on the basis of light availability. The aforementioned updates (1) to (3) follow parameterizations previously implemented in LM3.1, as described by *Milly et al*. (2014). In the LM4.0 experiments used in this study, soil types and soil parameter values were switched back to those used in LM3.0, which we find better simulate the observed sensitivity of $V_{d,O3}$ to drought (not shown). In **Section 3,** we evaluate how the changes in vegetation properties from LM3.0 to LM4.0 influence simulated $V_{d,O3}$.

Ozone deposition in the models is parameterized following an electrical circuit analogy as described in detail by *Paulot et al.* (2018).

Non-stomatal resistance for O_3, which includes in-canopy aerodynamic, cuticular, stem, and ground resistances, is parameterized as a function of friction velocity, LAI, and canopy wetness (Paulot et al., 2018). In this study, the input parameters for non-stomatal deposition are modified to simulate more realistic $V_{d,O3}$ and surface O_3 over snow-cover landscapes and under cold temperatures (see Supplemental **Text S1 and Figs. S1-S2**). The updates implemented by Clifton O.E. (2018) are not included here. For stomatal deposition, we incorporate leaf physiology by combining models of stomatal behavior and photosynthesis, as an alternative approach to modelling stomatal behavior only in terms of physical variables with a Javis (1976) type function. The equations for photosynthesis and stomatal conductance are described in detail in Appendices B3 and B4 of *Weng et al.* (2015), and are briefly summarized here.

Non–water limited stomatal conductance $\bar{g}_s$ (mol H_2O m^{-2} s^{-1}) averaged over the entire canopy depth is calculated as:

$$\bar{g}_s = max\left(\frac{m\bar{A}_n}{(C_i-\Gamma_*)(1+D_s/D_0)}, g_{s,min}\right) \quad \textbf{(1)}$$

Where $\bar{A}_n$ is the net photosynthesis rate (mol CO_2 m^{-2} s^{-1}) for a well-watered plant averaged over the entire canopy depth, m is an empirical coefficient which represents the species-specific sensitivity of stomatal conductance to photosynthesis, D_s is canopy air water vapor deficit (kg H_2O kg^{-1} air, D_0 is a reference value), C_i is intercellular concentration of CO_2 (mol CO_2 mol^{-1} air), Γ_* is the CO_2 compensation point (mol CO_2 mol^{-1} air), and $g_{s,min} = 0.01$ mol H_2O m^{-2} s^{-1} is the minimum stomatal conductance for water vapor allowed in the model. Increasing atmospheric water vapor deficits and CO_2 concentrations both cause $\bar{g}_s$ to decrease. A thermal inhibition factor $f(T)$ is applied to photosynthesis, affecting carbon acquisition and respiration equally:

$$f(T) = \left(1 + e^{0.4(T_1 - T_v)}\right)^{-1}\left(1 + e^{0.4(T_v - T_2)}\right)^{-1} \quad \textbf{(2)}$$

Where T_v is leaf temperature, and $T_1 = 5°C, T_2 = 45°C.$ This factor causes stomatal conductance to decrease rapidly when temperature is outside of the $[T_1, T_2]$ range.

After the non–water limited photosynthesis and stomatal conductance are determined, the model applies corrections to account for limitations imposed by soil water availability (ψ_w) and by canopy wetness (ψ_i):

$$\overline{\overline{g_s}} = \psi_w \psi_i \overline{g_s} \quad \textbf{(3)}$$

$$\psi_w = min(U_{max}/U_d, 1) \quad \textbf{(4)}$$

where U_{max} is the maximum plant water uptake rate ("water supply"), defined as the uptake rate when root water potential is at the plant permanent wilting point; U_d is "water demand", calculated as transpiration rate at non–water limited stomatal conductance. Calculation of vegetation water uptake (Milly et al., 2014; Weng et al., 2015) considers the vertical distribution of soil water, the vertical distribution of fine roots, and their biomass simulated by the LM3.0/LM4.0 vegetation dynamics (Shevliakova et al., 2009). In each soil layer, roots are represented as cylinders of small radius, and the difference between bulk water potential of the soil and water potential at the soil-root interface for this layer is determined by the near-field steady-state solution of the flow equation (Gardner, 1960), with xylem of the plant-root system providing the connection across layers (Weng et al., 2015).

Downregulation of photosynthesis due to water interception is

$$\psi_i = 1 - (f_s + f_l)\alpha_{wet} \quad \textbf{(5)}$$

where f_s and f_l are the fractions of canopy covered by snow and liquid water, respectively; α_{wet} is the down-regulation coefficient, assumed to be 0.3 (i.e., photosynthesis of leaves fully covered by water or snow is reduced by 30% compared to dry leaves).

We conduct a suite of approximately 600-yr simulations with LM3.0 and LM4.0. The experiments consist of a 300-yr potential vegetation spin-up phase (undisturbed by human activity), an intermediate land-use spin-up phase (1700-1860; Hurtt et al., 2011), and a historical phase (1861-2014) with varying CO_2 and land use (See Text S2). The dry deposition simulations are initialized from the 1948 conditions in the historical phase and continue through 2014, driven by observation-based meteorological forcings (Sheffield et al., 2006) (3-hourly precipitation, humidity, pressure, downward short and longwave radiation, near-surface temperatures and winds; available at http://hydrology.princeton.edu/data.php). These standalone land model hindcast simulations driven by observationally-based atmospheric forcings (here after "LM3.0" or "LM4.0") allow us to first investigate uncertainties in $V_{d,O3}$ parameterizations. Then we couple the land model to an atmospheric model ("AM3_LM3"; starting from the same 1948 initial land conditions as in LM3.0) to investigate the influence on simulated $V_{d,O3}$ from uncertainties in model atmospheric forcings, particularly precipitation (**Section 4**). To examine the influence of changes in $V_{d,O3}$ on surface O_3, we also conduct a simulation with a prototype version of the new GFDL AM4