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Synthetic ozone deposition and stomatal uptake at flux tower sites

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Abstract

We develop and evaluate a method to estimate O_3 deposition and stomatal O_3 uptake across networks of eddy covariance flux tower sites where O_3 concentrations and O_3 fluxes have not been measured. The method combines standard micrometeorological flux measurements, which constrain O_3 deposition velocity and stomatal conductance, with a gridded dataset of observed surface O_3 concentrations. Measurement errors are propagated through all calculations to quantify O_3 flux uncertainties. We evaluate the method at three sites with O_3 flux measurements: Harvard Forest, Blodgett Forest, and Hyytiälä Forest. The method reproduces 83% or more of the variability in daily stomatal uptake at these sites with modest mean bias (21% or less). At least 95% of daily average values agree with measurements within a factor of two and, according to the error analysis, the residual differences from measured O_3 fluxes are consistent with the uncertainty in the underlying measurements.

The product, called synthetic O_3 flux or SynFlux, includes 43 FLUXNET sites in the United States and 60 sites in Europe, totaling 926 site-years of data. This dataset, which is now public, dramatically expands the number and types of sites where O_3 fluxes can be used for ecosystem impact studies and evaluation of air quality and climate models. Across these sites, the mean stomatal conductance and O_3 deposition velocity is 0.03-1.0 cm s^{-1}. The stomatal O_3 flux during the growing season (April-September) is 0.5-11.0 nmol m^{-2} s^{-1} with a mean of 4.5 nmol m^{-2} s^{-1} and the largest fluxes generally occur where stomatal conductance is high, rather than where O_3 concentrations are high. The conductance differences across sites can be explained by atmospheric humidity, soil moisture, vegetation type, irrigation, and land management. These

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stomatal fluxes suggest that ambient O_3 degrades biomass production and CO_2 sequestration by 20-24% at crop sites, 6-29% at deciduous broadleaf forests, and 4-20% at evergreen needleleaf forests in the United States and Europe.

1 Introduction

Surface ozone (O_3) is toxic to both people and plants. Present-day and recent historical O_3 levels reduce carbon sequestration in the biosphere (Reich and Lassoie, 1984; Guidi et al., 2001; Sitch et al., 2007; Ainsworth et al., 2012), perturb the terrestrial water cycle (Lombardozzi et al., 2012, 2015), and cause around $25 billion in annual crop losses (Reich and Amundson, 1985; Van Dingenen et al., 2009; Avnery et al., 2011; Tai et al., 2014). The basic plant responses to O_3 injury are well established from controlled exposure experiments (e.g. Wittig et al., 2009; Ainsworth et al., 2005, 2012; Hoshika et al., 2015) but few datasets are available to quantify O_3 fluxes and responses for whole ecosystems or plant functional types that are represented within regional and global biosphere and climate models. The eddy covariance method has been widely used to measure land-atmosphere fluxes of carbon, water, and energy and evaluate their representation in models (Baldocchi et al., 2001; Bonan et al., 2011), but few towers measure O_3 fluxes (Munger et al., 1996; Fowler et al., 2001; Keronen et al., 2003; Gerosa et al., 2004; Lamaud et al., 2009; Fares et al., 2010; Stella et al., 2014; Zona et al., 2014). A recent review identified just 78 field measurements of O_3 fluxes over vegetation during the last 4 decades, many lasting just a few weeks (Silva and Heald, 2017). This paper demonstrates a reliable method to estimate O_3 fluxes at 103 eddy covariance flux towers spanning over two decades to enable O_3 impact studies on ecosystem scales.

The land surface is a terminal sink for atmospheric O_3 due to the reactivity of O_3 with unsaturated organic molecules and the modest solubility of O_3 in water. Surface deposition removes about 20% of tropospheric O_3, making it an important control on air pollution (Wu et al., 2007; Young et al., 2013, Kavassalis and Murphy, 2017). This O_3 deposition flux includes stomatal uptake into leaves, where O_3 can cause internal oxidative damage, and less harmful non-stomatal deposition to plant cuticles, stems, bark, soil, and standing water (Fuhrer, 2000; Zhang et al., 2002; Ainsworth et al., 2012). O_3 can also react with biogenic volatile organic compounds in the plant canopy air and this process is commonly included in non-stomatal deposition (Kurpius and Goldstein, 2003). The deposition flux (mol m^{-2} s^{-1}) can be described as:

$$F_{O_3} = v_d n(\chi - \chi_0) = v_d n\chi \quad (1)$$

where χ and χ_0 are the O_3 mole fractions (mol mol^{-1}) in the atmosphere and at the surface, respectively, n is the molar density of air (mol m^{-3}), and v_d is a deposition velocity (m s^{-1}) that expresses the net vertical O_3 transport between the height where χ is measured and the surface. F_{O_3} is defined positive for flux towards the ground. Eq. 1 reasonably assumes that $\chi_0 = 0$ because terrestrial surfaces have abundant organic compounds that react with and destroy O_3. The deposition velocity can be decomposed into resistances (s m^{-1}) for aerodynamic transport (r_a),

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diffusion in the quasi-laminar layer (r_b), stomatal uptake (r_s), and non-stomatal deposition (r_{ns}) (Wesely, 1989):

$$v_d^{-1} = r_a + r_b + (r_s^{-1} + r_{ns}^{-1})^{-1}. \qquad (2)$$

For stomatal and non-stomatal processes, the rates are often expressed as conductances (m s^{-1}), which are the inverse of the resistances: $g_s = r_s^{-1}$ and $g_{ns} = r_{ns}^{-1}$. The sum of stomatal and non-stomatal conductances is the vegetation canopy conductance, $g_c = g_s + g_{ns}$. The stomatal O_3 flux is the portion of F_{O_3} that enters the stomata, and can be described as:

$$F_{s,O_3} = F_{O_3} g_s (g_s + g_{ns})^{-1} = v_d n \chi g_s (g_s + g_{ns})^{-1}. \qquad (3)$$

To construct the synthetic O_3 flux, or SynFlux, we use measurements of O_3 concentration and standard eddy covariance flux measurements to derive nearly all of the terms in Eqs. 1-3 from surface observations, using minimal additional information from remote sensing and models. This enables the estimation of F_{O_3} and F_{s,O_3}, as described in Section 2. In Section 3 we then evaluate the method against observations at three sites that measure F_{O_3}, examine the importance of stomatal and non-stomatal deposition, and compare flux-based metrics of O_3 damage with concentration-based metrics. Finally, we discuss the strengths, limitations, and implications of our approach in Section 4.

2 Data sources and methods

2.1 SynFlux: synthetic O_3 flux

The FLUXNET2015 dataset (Pastorello et al., 2017) aggregates measurements of land-atmosphere fluxes of CO_2, H_2O, momentum, and heat at sites around the world (http://fluxnet.fluxdata.org/data/fluxnet2015-dataset, accessed 24 February 2017). Measurements are made with the eddy covariance method on towers above vegetation canopies (Baldocchi et al., 2001; Anderson et al., 1984; Goldstein et al., 2000) with consistent gap-filling (Reichstein et al., 2005; Vuichard and Papale, 2015) and quality control across sites (Pastorello et al., 2014). Flux and meteorological quantities are reported in half hour intervals. We analyze data from all sites in the United States and Europe in the FLUXNET2015 Tier 1 dataset. This analysis is restricted to the US and Europe because these regions have dense O_3 monitoring networks, described below. There are 103 sites meeting these criteria, all listed in Table S1 with references to full site descriptions. Three of these sites—Blodgett Forest, Harvard Forest, and Hyytiälä Forest—measure O_3 flux with the eddy covariance method, which we will use in Sect. 3 to evaluate our methods.

SynFlux aims to tightly constrain O_3 deposition resistances using measured water, heat and momentum fluxes, in contrast to other methods that rely more heavily on atmospheric models or standard meteorology observations (Finkelstein et al., 2000; Mills et al. 2011; Schwede et al., 2011; Yue et al., 2014). From the eddy covariance measurements, we derive the resistance

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components of Eq. 2 using methods similar to past studies (Kurpius and Goldstein, 2003; Gerosa et al., 2005; Fares et al., 2010). The aerodynamic and quasi-laminar layer resistances (r_a and r_b, respectively) are derived from measured wind speed, friction velocity, and fluxes of sensible and latent heat every half hour using Monin-Obukhov similarity theory (Foken, 2017). The stomatal conductance for O_3 (g_s) is derived from the measured water vapor flux and meteorological data every half hour with the inverted Penman-Monteith equation (Monteith, 1981; Gerosa et al., 2007). Some studies instead calculate g_s from the measured gross primary productivity (GPP) (Lamaud et al., 2009; El-Madany et al., 2017). That method likely underestimates the stomatal flux, however, because the g_s/GPP ratio increases as humidity rises and because g_s remains non-zero when GPP has ceased at night (Dawson et al., 2007; Medlyn et al., 2011). Appendix A provides further details of these calculations. To avoid complications to the Penman-Monteith equation from wet canopies, we exclude times when dew may be present (RH > 80%), and days with precipitation (> 5mm). We also exclude the top and bottom 1% of g_s values, which include many unrealistic outliers (e.g. $|g_s| > 0.5$ m s^{-1}). Figure 1 shows the mean stomatal conductance during the growing season (April-September) at all sites.

The terms in Eqs. 1-3 that cannot be derived from FLUXNET2015 measurements are O_3 mole fraction and non-stomatal conductance. The O_3 mole fraction is taken from a gridded dataset of hourly O_3 measurements that spans the contiguous United States and Europe (Schnell et al., 2014). This dataset has 1° spatial resolution, so some differences from measured O_3 abundances at individual sites are inevitable. Schnell et al. (2014) estimated these errors to be 6-9 ppb (rms) or about 15% of summer mean O_3 in the US and similar in Europe. Figure 2 shows that the daytime gridded O_3 concentrations correlate well with observations at three flux tower sites where O_3 was measured (R^2 = 0.63-0.87) and have modest negative bias (5-10 ppb, –12 to –28%), consistent with the accuracy reported by Schnell et al. (2014). We use the Zhang et al. (2003) parameterization of non-stomatal conductance, which accounts for O_3 deposition to leaf cuticles and ground and has been evaluated at sites in North America. The parameterization requires leaf-area index, which we take from satellite remote sensing (Claverie et al., 2014; 2016), snow depth, which we take from MERRA2 reanalysis (GMAO, 2015; Gelaro et al., 2017), and standard meteorological data provided by FLUXNET2015.

Figure 3 shows the stomatal O_3 flux at each site calculated with Eq. 3, then averaged over the April-September growing season. Figure S1 shows the corresponding total O_3 flux (Eq. 1). We refer to these products as the "synthetic" total and stomatal O_3 fluxes (F'_{O_3} and F'_{s,O_3}, respectively) and use a prime to distinguish them from the measured O_3 fluxes (F_{O_3} and F_{s,O_3}) that are only available at a few sites. Together, we refer to F'_{O_3} and F'_{s,O_3} as SynFlux. In total, the measurements required to calculate F'_{s,O_3} are O_3 mole fraction, sensible and latent heat fluxes, friction velocity, temperature, pressure, humidity, canopy height, and leaf area index. There are 43 sites in the US and 60 sites in Europe within the FLUXNET Tier 1 database with sufficient measurements to calculate F'_{s,O_3}.

2.2 Observed O_3 flux

We evaluate SynFlux and its inputs at three sites where O_3 flux measurements are available: Harvard Forest, Massachusetts, United States (Munger et al., 1996); Blodgett Forest, California, United States (Fares et al., 2010); and Hyytiälä Forest, Finland (Keronen et al., 2003; Mammarella et al., 2007; Rannik et al., 2009). These forest sites sample a range of environmental and ecosystem conditions summarized in Table 1. All three sites have at least 6 years of half-hourly or hourly flux measurements. Two sites are evergreen needleleaf forests (Blodgett and Hyytiälä), while one is a deciduous broadleaf forest containing some scattered stands of evergreen needleleaf trees (Harvard). Climate also differs across these sites. Blodgett Forest has a Mediterranean climate with cool, wet winters and hot, dry summers. Hyytiälä and Harvard Forests have cold winters and wetter summers, with Harvard Forest being the warmer of the two.

Harvard Forest water vapor flux measurements were recalibrated for this work based on matching water vapor mixing ratio measured by the flux sensor to levels calculated from ambient relative humidity and air temperature, resulting in a 30% increase in evapotranspiration during the 1990s and no change since 2006. In addition, we remove sub-canopy evaporation from the measured water vapor flux before the Penman-Monteith calculation. Based on past measurements at these sites, the sub-canopy fraction of evapotranspiration is 20% at Hyytiälä Forest, 10% at Harvard Forest in summer, and 50% at Harvard Forest in months without leaves (Moore et al., 1996; Launiainen et al., 2005). We are unable to make this correction at all FLUXNET sites since water vapor flux is typically measured only above canopy.

At these three sites, observed v_d, g_{ns}, and F_{s,O_3} can be derived from the F_{O_3} measurements with methods that differ slightly from Sect. 2.1. O_3 deposition velocity is inferred from measurements of O_3 concentration and flux via $v_d = F_{O_3}(n\chi)^{-1}$. Resistance or conductance terms r_a, r_b, and g_s are calculated as described in Sect. 2.1, then both canopy and non-stomatal conductance are derived from observations via $g_c = (v_d^{-1} - r_a - r_b)^{-1}$ and $g_{ns} = g_c - g_s$, respectively. With those values, Eq. 3 gives the observed stomatal O_3 flux. The half-hourly or hourly measured and synthetic flux still have some outliers (Fig. S2), but the error analysis reveals that most of the outlying points have large uncertainties.

2.3 Gap filling for friction velocity

The FLUXNET2015 dataset uses gap filling for most flux and meteorological measurements (Vuichard and Papale, 2015), but not for friction velocity (u_*), which is required to calculate v_d

and F'_{s,O_3}. Filling this one variable would significantly reduce the fraction of missing data in our analysis. Monin-Obukhov similarity theory predicts that friction velocity is proportional to wind speed in the surface layer, for a given roughness length and stability regime (Foken, 2017). On this basis, we regress the available friction velocity measurements against wind speed and net radiation (a proxy for stability) separately for each site and month (a proxy for vegetation roughness). This gap filling was possible at 91 sites that report net radiation measurements.

The predicted friction velocities from the regression model are highly correlated with available observations ($R^2 > 0.7$) and have minimal mean bias (±10%) at 68 out of 91 eligible sites (Fig. S3). At the remaining 23 sites, frequent stagnant and stable conditions ($u_* \lesssim 0.5$ m s^{-1}) degrade the regression performance. We used the regression model to fill missing friction velocity measurements and were thus able to increase the number of F'_{s,O_3} estimates by 1-20%. The differences between monthly mean F'_{s,O_3} with and without gap filling are 10% (rms), so although the u_* gap filling is a potential source of uncertainty, the F'_{s,O_3} estimates are robust.

2.4 Error analysis, averaging, and numerical methods

We quantify the errors in F'_{O_3}, F'_{s,O_3}, and all other calculated variables from the measurement uncertainties using standard techniques for propagation of errors through all equations (see Appendix B). This method provides the uncertainty, quantified as standard deviation, of each variable in each half hour interval. The error analysis reveals that F'_{s,O_3} and other derived quantities have uncertainties that change from hour to hour by two orders of magnitude (Fig. S2). In addition, many extreme values of F'_{s,O_3}, g_s, and other variables have very large uncertainties. We retain these outliers in our analysis and use the error analysis to appropriately reduce their influence on averages and other statistics, as described below, without discarding data.

The FLUXNET2015 dataset contains error estimates for sensible and latent heat measurements. We use these reported values in the error analysis. Where uncertainties in these fluxes are missing, we fill the gaps using a linear regression of available flux errors against flux values for that site. For friction velocity, the uncertainty is the prediction error in the linear model used for gap filling (Sect. 2.3). Based on expert judgment, the standard deviation of O_3 mole fraction is set to 20%, pressure to 0.5 hPa, temperature to 0.5 K, relative humidity to 5%, and canopy height to the lesser of 15% or 2 m. For leaf area index, we use reported uncertainties in the remote sensing for each plant functional type (Claverie et al., 2013; 2016). The Zhang et al. (2003) g_{ns} parameterization has 5 vegetation-specific parameters and all are assigned 50% standard deviation. Zero error is assumed for the flux tower height. Based on these inputs, the median relative uncertainty in F'_{s,O_3} is 44%, but it rises to several hundred percent for some half-hour intervals. The error analysis shows that most of the uncertainty in F'_{s,O_3} derives from uncertainty in the latent heat flux measurement.

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Daily and monthly averages of F'_{s,O_3} and other quantities are constructed in stages. We first calculate a mean diurnal cycle for the day or month by pooling measurements during each hour in a maximum likelihood estimate, a weighted average that accounts for the uncertainty in each measurement. The maximum likelihood estimate is appropriate when combining values from the same distribution, which is expected to apply for measurements within a particular hour, but not across hours of the day. We then average across hours with an unweighted mean to calculate the daily or monthly value. Seasonal values are the unweighted mean of the months they contain. Uncertainties are propagated through each stage of these averages, as detailed in Appendix B. Our discussion focuses on daily averages of daytime data when the sun is at least 4° above the horizon.

Analyses are performed in Python 3.5 with NumPy, Pandas, PySolar, and Statsmodels (Reda and Andreas, 2005; Van Der Walt et al., 2006; McKinney, 2010; Seabold et al., 2010). We quantify the slope and strength of linear relationships between variables using standard major axis fitting (SMA, Warton et al., 2006), the non-parametric Thiel-Sen slope (Sen, 1968), and coefficient of determination (R^2).

2.5 Data availability

The SynFlux dataset produced in this work is available in the supplementary information for download and use. The dataset includes synthetic stomatal and total O_3 fluxes, O_3 concentrations, O_3 deposition velocity, canopy conductance, stomatal conductance, and all of their propagated uncertainties. Monthly mean values are provided, with and without u_* gap filling, for 103 sites totaling 926 site-years.

3 Results and discussion

3.1 Evaluation of synthetic fluxes

Figure 4 compares daily daytime averages of synthetic F'_{s,O_3} to measured F_{s,O_3}. At all three sites, F'_{s,O_3} is strongly correlated with measured values (R^2 = 0.83-0.93). The mean and median biases are –16 to –21% and at least 95% of F'_{s,O_3} values agree with measurements within a factor of 2. The majority of F'_{s,O_3} values lie near the 1:1 line with F_{s,O_3} and the slopes (0.71 to 0.85) reflect this. For 98% of points, the differences between F'_{s,O_3} and F_{s,O_3} are less than the 95% confidence interval derived from the error analysis (two-sided t test). Thus, the errors in F'_{s,O_3} are consistent with the uncertainty in the observations. The half hourly F'_{s,O_3} values perform similarly well against observations (Fig. S4), but our analysis focuses on averages. Overall, these results

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suggest that synthetic F'_{s,O_3} is a reliable estimate of stomatal O_3 uptake into plants that can be used at eddy covariance sites without O_3 measurements.

The measurements enable us to evaluate synthetic total deposition, F'_{O_3}, as well, although this is less relevant to ecosystem impacts than stomatal uptake, F'_{s,O_3}. Figure S5 shows that bias (–13 to +65%), slope (0.3-1.4), and R^2 (0.05-0.43) for F'_{O_3} are all worse than for F'_{s,O_3}. The reasons can be derived from Eq. 3. The canopy resistance for O_3 is normally much greater than the quasi-laminar layer and aerodynamic resistances, meaning $r_c \gg r_a$ and $r_c \gg r_b$, often by a factor of 3-10. Therefore, the O_3 deposition velocity is approximately $v_d \approx r_c^{-1} = g_c$. Under these conditions, Eq. 1 simplifies to $F_{O_3} \approx n\chi(g_s + g_{ns})$ and Eq. 3 simplifies to $F_{s,O_3} \approx n\chi g_s$. While g_s is calculated from measured H_2O fluxes, g_{ns} comes from a parameterization, which inevitably introduces error into g_{ns} and F'_{O_3}. Since F'_{s,O_3} has little sensitivity to g_{ns} or its errors, it can be calculated more accurately than F'_{O_3}, as seen when comparing Figures 4 and S4. Despite these larger errors, the F'_{O_3} mean is within 50% of the observed value at two sites and within a factor of 2 at all, which may be useful for some applications, given the paucity of prior F_{O_3} measurements.

3.2 Stomatal and non-stomatal deposition

Figure 5 shows the seasonal cycles of observed O_3 deposition velocity and its important components at the three study sites with O_3 flux measurements. For low or moderately reactive gases like O_3, canopy resistance is typically greater than aerodynamic or quasi-laminar layer resistance, so it controls the overall deposition velocity. At these three sites, deposition velocity is lowest in winter (0.1-0.2 cm s^{-1}) and highest in summer (0.5–0.6 cm s^{-1}). Stomatal conductance, which peaks when weather conditions favor growth, explains most of this seasonal variation, except at Blodgett Forest as discussed below. Stomatal conductance is generally thought to exceed non-stomatal conductance during the growing season at most vegetated sites (Wesely, 1989; Zhang et al., 2003). At both Harvard and Hyytiälä Forests, the mean stomatal conductance (0.2-0.6 cm s^{-1}) is 1.5-6 times larger than non-stomatal conductance (0.08-0.2 cm s^{-1}) during the growing season, so about 60-90% of O_3 deposition occurs through stomatal uptake. In winter at these sites, the calculated stomatal conductance can exceed canopy conductance, which is not possible, but is likely an artifact of evaporation from soil or snow exceeding leaf transpiration at that time of year. At Blodgett, non-stomatal conductance slightly exceeds stomatal conductance in summer (0.4 vs. 0.3 cm s^{-1}). The fast non-stomatal deposition is explained by O_3 reacting with biogenic terpenoid emissions below the flux measurement height (Kurpius and Goldstein, 2003; Fares et al., 2010). These biogenic emissions depend strongly on temperature and light and have a large seasonal cycle with maxima in summer and minima in winter, so stomatal uptake is generally < 50% of O_3 deposition at Blodgett in the summer but > 70% in winter.

A recent analysis of O_3 flux measurements at Harvard Forest suggests that non-stomatal deposition averages 40% of daytime O_3 deposition during summer months, with a range of 10-60% across years (Clifton et al., 2017). Our analysis of the same site, using re-calibrated latent heat flux measurements, does not support such a large role for non-stomatal deposition at this site in summer. As seen in Fig. 5, only 15% of O_3 deposition is non-stomatal during these months, with a range of 4-32% across years. At Hyytiälä Forest, our results are consistent with prior work that found that the non-stomatal deposition is 26% to 44% of daytime O_3 deposition during the growing season (Rannik et al., 2012). Nevertheless, non-stomatal deposition equals or exceeds stomatal uptake where there are large terpene emissions (e.g. Blodgett) and at some other temperate sites that probably lack large biogenic emissions (Fowler et al., 2001; Cieslik, 2004; Lamaud et al., 2009; Stella et al., 2011; El-Madany et al., 2017).

At Harvard and Hyytiälä Forests, the parameterized g_{ns} has a similar mean to measurements during summer, with discrepancies less than a factor of two (Fig. 5). The observed day-to-day variability in g_{ns} is as large as the variability in g_s at Harvard and Hyytiälä Forests and the calculated g_{ns} does not reproduce it ($R^2 < 0.09$), so an important but undetermined non-stomatal process is missing from the parameterization. At Blodgett Forest, the parameterized g_{ns} is one-third of measured g_{ns} in summer, but this is not surprising since the parameterization does not account for O_3 reactions with biogenic volatile organic compounds (BVOC), which are known to be important at this site (Fares et al., 2010). We attempted, unsuccessfully, to use BVOC emissions from the MEGAN biogenic emission model (Guenther et al., 2012) to improve the g_{ns} parameterization, but the correlations between measured g_{ns} and compounds that react fastest with O_3 (monoterpenes and sesquiterpenes) were poor ($R^2 \leq 0.15$). On that basis, synthetic F'_{O_3} may also underestimate total O_3 deposition at other sites with high monoterpene and sesquiterpene emissions, such as warm-weather pine forests, but synthetic F'_{s,O_3} should retain its quality everywhere.

3.3 Spatial patterns of synthetic fluxes

Across the 43 sites in the US shown in Fig. 3, mean F'_{s,O_3} during the growing season ranges from 0.5 to 11.0 nmol m^{-2} s^{-1} with an average of 4.4 nmol m^{-2} s^{-1}. The highest F'_{s,O_3} generally occurs in the Midwest (5-9 nmol m^{-2} s^{-1} in Wisconsin, Michigan, Nebraska, Ohio) due to its moderate O_3 concentrations (Fig. S6) and moisture levels, which promotes stomatal conductance (Fig. 1). The Western US has higher average O_3 concentrations, but generally lower moisture and stomatal conductance, especially the Southwest US, so F'_{s,O_3} (0-4 nmol m^{-2} s^{-1}) is mostly lower than the Midwest. Land cover, land management, and plant types drive large differences in F'_{s,O_3} between nearby sites, even when O_3 concentrations and meteorology are similar. For example, three

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Nebraska sites are all crop fields and O_3 concentrations are nearly identical, but two irrigated fields have higher stomatal conductance and higher F'_{s,O_3} than the nearby rainfed field (6.2 vs. 4.8 nmol m^{-2} s^{-1}). Two sites in central California have high g_s and F'_{s,O_3} compared to surrounding sites due to irrigation and naturally wet soil in the California Delta. A combination of topography and climate is also an important factor in California: forest sites in the Sierra Nevada mountains have lower g_s and F'_{s,O_3} than the lowland crops and wetland grasses. In Oregon, an evergreen needleleaf site regrowing after a fire has higher g_s and F'_{s,O_3} than two older forest stands nearby. The differences between 9 Wisconsin forest sites, however, are mostly due to different years of data at each site combined with interannual variability in F'_{s,O_3}; fluxes at these sites are similar in overlapping years.

Variability across the 60 sites in Europe is controlled by similar factors. Stomatal uptake ranges from 1.4 to 9.6 nmol m^{-2} s^{-1}, with an average of 4.7 nmol m^{-2} s^{-1} (Fig. 3). The Mediterranean region has high O_3 concentrations (Fig. S6), but generally low stomatal conductance due to the dry climate (Fig. 1). Within this region, vegetation type explains broad patterns. Shrub sites in Spain, France, and Sardinia have very low g_s (~0.15 cm s^{-1}) so F'_{s,O_3} is low (1-3 nmol m^{-2} s^{-1}), while the most of the sites in mainland Italy are broadleaf and evergreen forests that have slightly greater g_s (~0.2-0.4 cm s^{-1}) and F'_{s,O_3} (3-6 nmol m^{-2} s^{-1}), despite similar climate and O_3. In central and northern Europe, temperate climate promotes higher stomatal conductance while O_3 concentrations remain modest throughout the growing season. The largest F'_{s,O_3} is 9.8 nmol m^{-2} s^{-1} at a deciduous broadleaf forest in Switzerland, while nearby evergreen forests, cereal crops, and grasslands all have lower fluxes (6-8 nmol m^{-2} s^{-1}). While Finland has generally low F'_{s,O_3} of 2-5 nmol m^{-2} s^{-1}, the high end of this range is similar to rural sites in Germany, illustrating that O_3 can impact ecosystems with low O_3 concentrations far from major industrial emissions.

Table 2 quantifies SynFlux, O_3 deposition velocity, and conductance for each plant functional type. Wetlands, crops, and forests have the highest average F'_{s,O_3}, which is about two times higher than woody savanna or shrublands, the vegetation types with the lowest F'_{s,O_3}. The vegetation types rank in the same order for stomatal conductance, again showing stomata as the main control on O_3 deposition and uptake. Stomatal uptake exceeds non-stomatal uptake for all plant functional types except woody savanna and shrubland. O_3 deposition velocities reported in the table fall within the ranges of past literature, as reviewed by Silva and Heald (2017). However, while Silva and Heald found that the mean deposition velocity was greater over deciduous forests than coniferous forests, crops, or grass, we do not. Rather, we find that variability between sites within each of these categories is large, having a standard deviation about 30% of the multi-site mean.

3.4 Metrics for O_3 damage to plants

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Since O_3 injures plants mainly by internal oxidative damage after entering the leaves through stomata, the most physiological predictor of plant injuries is the cumulative uptake of O_3 (CUO, Reich, 1987; Fuhrer, 2000; Karlsson et al., 2004; Cieslik, 2004; Matyssek et al., 2007). CUO is defined as the cumulative stomatal O_3 flux exceeding a threshold flux Y that can be detoxified by the plant, integrated over a period of time:

$$\mathrm{CUOY} = \sum_i H(F_{s,\mathrm{O_3},i} - Y)(F_{s,\mathrm{O_3},i} - Y)\,\Delta t_i.$$

Here, $H(x)$ is the Heaviside step function and Δt_i is the time elapsed during measurement of $F_{s,\mathrm{O_3},i}$. The sum is carried out over time *i* in the growing season, which we take to be April to September. The detoxification threshold varies across vegetation types, even among related species (Karlsson et al., 2004, Büker et al., 2015), and thresholds for specific FLUXNET sites are generally unknown. As a compromise, we calculate CUO, with Y=0, and also CUO3, with Y = 3 nmol m^{-2} s^{-1}, which has been suggested as a reasonable generic threshold (Mills et al., 2011). CUO is always greater than CUO3, but the sites with high CUO tend to also have high CUO3, so their spatial patterns are similar (Fig. S7).

While CUO is a physiological dose, concentration-based metrics remain common for assessing ozone impacts because they are easier to measure. Concentration-based metrics quantify O_3 in ambient air irrespective of whether that O_3 enters leaves. These metrics follow the general form

$$M = \sum_i w(\chi_i)\,(\chi_i - \chi_c)\,\Delta t_i$$

where $w(\chi)$ is a weighting function applied to the O_3 mole fraction χ, and χ_c is a constant. Like CUO, the sum is usually over time *i* during the growing season. Three of the most common concentration-based O_3 metrics are the mean O_3 concentration, the accumulated concentration over a threshold of 40 ppb (AOT40; UNECE, 2004), and the sigmoidal-weighted index (W126; Lefohn and Runeckles, 1987). For mean, $w(\chi) = (\sum \Delta t_i)^{-1}$ and $\chi_c = 0$. For AOT40, $w(\chi) = H(\chi - \chi_c)$ and χ_c = 40 ppb. For W126, $w(\chi) = (1 + 4403\exp(-(126\ \mathrm{ppb}^{-1})\chi))^{-1}$ and χ_c = 0. Both AOT40 and W126 use only daytime (8am-8pm) measurements and W126 also takes the maximum value over a 3-month period. The weighting functions for AOT40 and W126 give little or no weight to O_3 concentrations below 40 ppb. In addition, W126 gives increasing weight to concentrations up to about 110 ppb and full weight for higher concentrations based on the understanding that exposure to high O_3 concentrations is more injurious than moderate or low concentrations. Other concentration-based metrics (e.g. SUM60) use other thresholds or weighting functions, but many are strongly correlated with AOT40 or W126 or otherwise qualitatively similar (Paoletti et al., 2007).

The spatial patterns of AOT40 and W126 closely resemble that of mean O_3 concentration in the US and Europe despite their different weighting functions (Fig. S7). The CUO and CUO3 spatial patterns, however, are similar to $F'_{s,\mathrm{O_3}}$ and distinct from the concentration-based metrics. This illustrates that locations with high AOT40 or W126, like the Southwest US or Mediterranean Europe, can have low CUO.

Even though concentration-based metrics do not measure the physiological O_3 dose to plants, they can be useful if the metric is proportional to the flux-based dose and injuries. Indeed, many controlled experiments and observational studies have documented correlations between both AOT40 and W126 and either uptake or plant injuries (e.g. Fuhrer et al., 1997; Cieslik, 2004; Musselman et al., 2006; Matyssek et al., 2010). However, many of these studies were carried out at a single site or under conditions where stomatal conductance was relatively steady while O_3 concentrations varied, for example by maintaining well-watered soil. When stomatal conductance varies widely, such as between arid and humid climates or seasons, concentration-based metrics may not correlate with stomatal O_3 flux (Mills et al., 2011).

Figure 6 shows that all of the concentration-based metrics are poorly correlated with CUO across the sites (AOT40: $R^2 = 0.05$, W126: $R^2 = 0.03$, mean O_3: $R^2 = 0.04$). Humidity helps explain some of the scatter in Figure 6. The sites with high concentration-based metrics and low CUO have high vapor pressure deficit (VPD), low stomatal conductance, and are mostly in the western US and Mediterranean Europe. Restricting the analysis to humid sites (VPD < 1.5 kPa) does not improve the correlation ($R^2 \approx 0.05$) and at the arid sites (VPD > 1.6 kPa) the concentration-based metrics are modestly anti-correlated with CUO (AOT40: $R^2 = 0.19$, W126: $R^2 = 0.05$, mean O_3: $R^2 = 0.37$). This result reinforces that concentration-based metrics can misrepresent CUO and plant injuries (Mills et al., 2011).

From the CUO values in Table 2, we can estimate the range of O_3 impacts on biomass production at the FLUXNET sites. Although species vary in their sensitivity to O_3, several studies suggest that the biomass production of broadleaf and needleleaf trees decreases 0.2 to 1% per mmol m^{-2} of CUO (Karlsson et al., 2004; Wittig et al., 2007; Hoshika et al., 2015). Combining the mean CUO for each plant functional type (Table 2) with these sensitivities, our work implies that O_3 reduces the biomass production at these FLUXNET sites by 6-29% for deciduous broadleaf forests and 4-20% for needleleaf forests. The range represents the spread of reported dose-response sensitivities within each plant type, meaning the least and most O_3-sensitive species. Lombardozzi et al. (2013) caution that species-specific responses to O_3 may not generalize to plant functional types, but the biomass reductions calculated here still indicate the general magnitude of expected O_3 damages. Several broadleaf crops are more sensitive to O_3, with biomass reductions of 1.3-1.6% per mmol m^{-2} of CUO3 (Mills et al., 2011). That sensitivity implies 20-24% drop in biomass production at FLUXNET crop sites. Some studies have quantified O_3 dose-response relationships with other thresholds Y = 1.6 to 6 nmol m^{-2} s^{-1} (e.g. Karlsson et al., 2007; Pleijel et al., 2004, 2014), but the sensitivities have similar magnitude. Fares et al. (2013) also demonstrated 12-19% reduction in gross primary production due to O_3 at some of the same crop and forest FLUXNET sites. Using prognostic models of O_3

concentrations and stomatal uptake, several past studies have also suggested that O_3 reduces biomass production and CO_2 sequestration by 4-20% in the US and Europe (Sitch et al., 2007; Wittig et al., 2007; Mills et al., 2011; Yue et al., 2014, 2016; Lombardozzi et al., 2015). Our results support this range of impacts, although some FLUXNET sites and species likely experience greater O_3 injury, but here the CUO is highly constrained from observations and therefore avoids the additional uncertainties of atmosphere-biosphere models.

4 Conclusions

We have demonstrated a method to estimate O_3 fluxes and stomatal O_3 uptake at eddy covariance flux towers wherever regional O_3 monitors exist. The method, called SynFlux, derives stomatal conductance and O_3 deposition velocity from standard eddy covariance measurements and combines them with gridded O_3 concentrations from air quality monitoring networks. We apply this method to the FLUXNET2015 dataset and derive synthetic flux estimates at 43 sites in the United States and 60 sites in Europe, totaling 926 site-years of observations. O_3 deposition measurements have previously only been sporadically available for a few sites around the world, so this work dramatically increases the flux data available for understanding O_3 impacts on vegetation and for evaluating air quality and climate models.

Three sites with long-term O_3 flux measurements provide an independent test of SynFlux. These comparisons show that daily averages of synthetic stomatal F'_{s,O_3} correlate well with measured F_{s,O_3} (R^2 = 0.83-0.93) and have a mean bias under 22% at all sites. At all three sites 95% of the synthetic F'_{s,O_3} values differ from measurements by a factor of 2 or less. The differences between F'_{s,O_3} and F_{s,O_3} are also consistent with propagated uncertainty in the underlying measurements. Synthetic total deposition, F'_{O_3}, is sensitive to errors in the parameterized non-stomatal conductance, but mean values are still with a factor of 2 of observations. The errors in this dataset are modest compared with differences between observations and regional and global atmospheric chemistry models that are frequently a factor of 2 or more (Zhang et al., 2003; Hardacre et al., 2015; Clifton et al., 2017; Silva and Heald, 2017), illustrating the utility of this dataset for evaluating models and O_3 impacts.

Across flux tower sites in the US and Europe, F'_{s,O_3} ranges from 0.5 to 11.0 nmol m^{-2} s^{-1} during the summer growing season. The spatial pattern of F'_{s,O_3} is mainly controlled by stomatal conductance rather than O_3 concentration. Patterns of stomatal conductance and F'_{s,O_3} in turn are explained by climate, especially atmospheric and soil moisture, vegetation types, and land management, such as irrigation. O_3 concentration-based metrics (AOT40, W126, mean O_3) have been widely used to evaluate O_3 damages to plants because they are easier and cheaper to measure than the cumulative uptake of O_3 (CUO) into leaves. However, these metrics have very little correlation with CUO ($R^2 \leq 0.05$) across FLUXNET sites. Using dose-response relationships between CUO and biomass reduction, we estimate that O_3 reduces biomass production and carbon uptake by 4-29%, depending on the site and plant type. Unlike most past estimates, which have used prognostic models of O_3 uptake, our assessment of biomass reduction is based on O_3 fluxes that are tightly constrained by observations. To promote further applications in ecosystem monitoring and modeling, the SynFlux dataset is publicly available in the supplement as monthly averages of F'_{s,O_3}, F'_{O_3}, O_3 deposition velocity, stomatal conductance, and related variables.

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Table 1. Description of sites that measure O_3 flux and their daytime growing season (April-September) conditions [a]

	Blodgett Forest, California, USA	Hyytiälä Forest, Finland	Harvard Forest, Massachusetts, USA
Latitude, Longitude	38.8953, –120.6328	61.8475, 24.2950	42.5378, –72.1715
Plant functional type	Evergreen needleleaf	Evergreen needleleaf	Deciduous broadleaf
Years of data	2001-2007	2007-2012	1993-1999
Days of observations	1281	1098	1281
Canopy height, m	8	15	24
GPP, μmol $m^{-2} s^{-1}$	9.22 ± 3.55	11.1 ± 5.02	12.4 ± 7.62
ET, mmol $m^{-2} s^{-1}$	3.25 ± 1.23	1.71 ± 0.82	2.95 ± 1.70
PAR, μmol $m^{-2} s^{-1}$	875 ± 149	690 ± 203	876 ± 222
Air Temperature, °C	19.1 ± 5.36	13.3 ± 5.99	17.65± 5.75
VPD, kPa	1.51 ± 0.61	0.73 ± 0.32	0.90 ± 0.34
O_3, ppb	55.4 ± 13.4	32.2 ± 8.68	48.8 ± 15.8
F_{s,O_3}, nmol $m^{-2} s^{-1}$	5.18 ± 2.11	4.35 ± 1.66	7.23 ± 4.87
Precipitation, mm day^{-1}	0.09 ± 0.49	0.42 ± 0.89	0.28 ± 0.82

[a] Values are mean ± standard deviation of daily averages, using daytime observations only. GPP is gross primary productivity. ET is evapotranspiration. PAR is photosynthetically active radiation. VPD is vapor pressure deficit. F_{s,O_3} is observed stomatal O_3 flux.

Table 2. Mean O_3 SynFlux, deposition velocity and its conductance components during daytime in the growing season (April-September), grouped by plant functional type (PFT).[a]

PFT[b]	Sites	Site-Years	g_s	g_{ns}	g_c	v_d	F'_{O_3}	F'_{s,O_3}	CUO	CUO3
CRO	18	148	0.42±0.17	0.28±0.09	0.68±0.18	0.53±0.12	7.66±1.96	4.77±1.52	24.8±12.4	14.9±9.3
ENF	25	254	0.37±0.10	0.25±0.06	0.60±0.11	0.54±0.10	7.37±1.33	4.61±1.16	20.0±5.69	11.9±6.30
EBF	3	31	0.21±0.02	0.15±0.02	0.36±0.03	0.33±0.03	5.02±0.65	2.90±0.28	12.1±0.81	5.12±0.45
DBF	16	158	0.41±0.14	0.20±0.09	0.60±0.18	0.53±0.15	7.87±2.28	5.37±1.69	28.6±13.8	15.7±6.66
MF	5	83	0.44±0.17	0.19±0.01	0.62±0.15	0.56±0.14	7.82±1.91	5.53±2.15	24.9±10.5	15.9±8.90
WSA	2	25	0.10±0.02	0.31±0.06	0.39±0.04	0.36±0.04	6.14±0.20	1.47±0.31	6.46±1.43	2.54±1.72
OSH	4	14	0.19±0.07	0.29±0.10	0.47±0.10	0.41±0.09	5.69±1.33	2.23±0.87	8.60±3.27	2.27±1.54
CSH	2	15	0.27±0.11	0.29±0.01	0.57±0.09	0.49±0.05	6.78±0.95	3.34±1.24	14.3±5.30	7.62±5.49
GRA	18	136	0.40±0.30	0.24±0.11	0.64±0.26	0.47±0.15	7.04±7.04	4.12±2.45	18.3±10.7	9.90±6.98
WET	10	53	0.48±0.16	0.27±0.09	0.74±0.21	0.58±0.14	8.80±2.74	5.77±2.08	25.1±9.65	19.4±15.6

[a] Values are the mean ± standard deviation across sites within each PFT. Units for g_s, g_{ns}, g_c, and v_d are cm s^{-1}. Units for F'_{O_3} and F'_{s,O_3} are nmol $m^{-2} s^{-1}$.
[b] CRO = crop, ENF = evergreen needleleaf forest, EBF = evergreen broadleaf forest, DBF = deciduous broadleaf forest, MF = mixed forest, WSA = woody savanna, OSH = open shrubland, CSH = closed shrubland, GRA = grassland, WET = wetland

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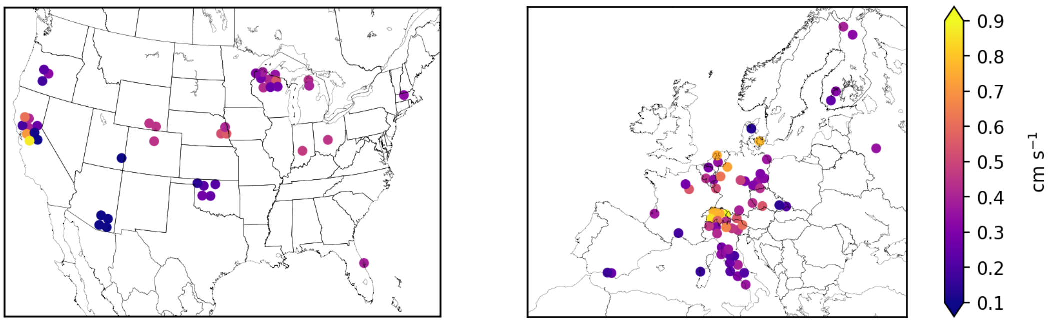

Figure 1. Mean stomatal conductance for O_3 (g_s) during daytime in the growing season (April-September) at FLUXNET2015 sites in the United States and Europe. Symbols of some sites have been moved slightly to reduce overlap and improve legibility.

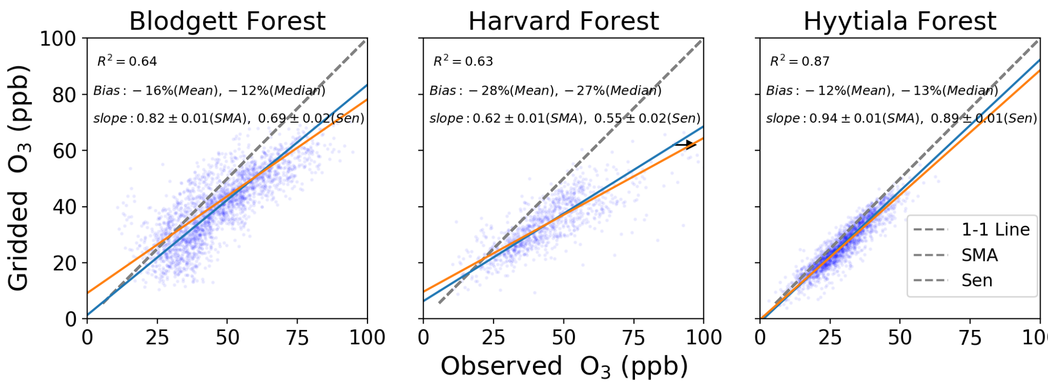

Figure 2. Gridded and observed daily daytime O_3 concentrations at Blodgett, Harvard, and Hyytiälä Forests. Inset numbers provide the coefficient of determination (R^2), mean and median bias, the standard major axis (SMA) slope, the Thiel-Sen (Sen) slope, and the 68% confidence interval of the slopes. Black arrow points towards outliers that are not shown.

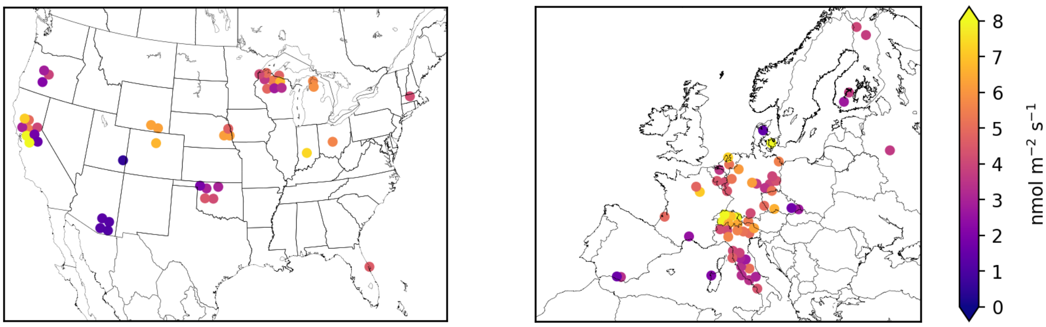

Figure 3. Mean synthetic stomatal O_3 flux (F'_{s,O_3}, Sect. 2.1) during the daytime growing season (April-September) at FLUXNET2015 sites in the United States and Europe. Symbols of some sites have been moved slightly to reduce overlap and improve legibility.

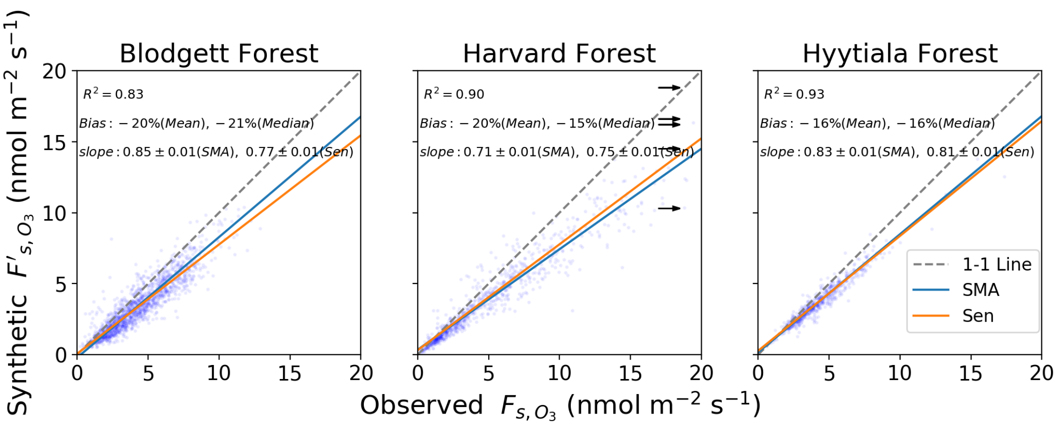

Figure 4. Synthetic and observed daily daytime stomatal O_3 flux. See Sect. 2.1 for definition of F'_{s,O_3} and Fig. 2 for explanation of lines and inset text.

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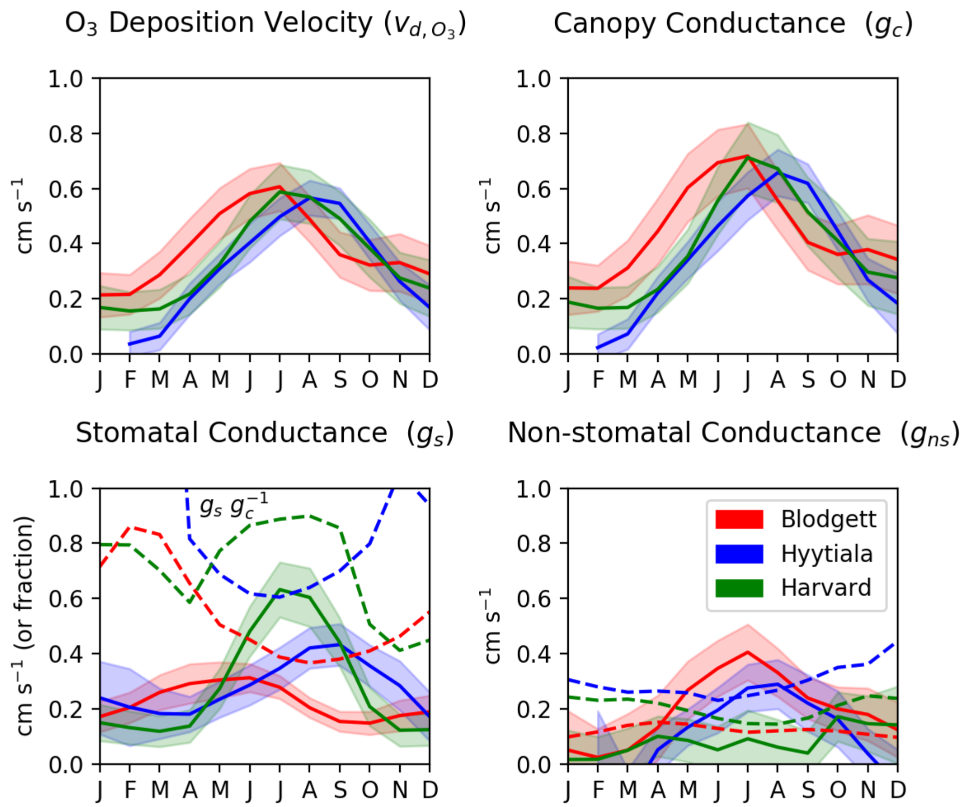

1294
1295 Figure 5. Observed O_3 deposition velocity and its in-canopy components at sites with O_3 flux
1296 measurements. Lines show means and shaded regions show standard deviation of daily values
1297 for each month. Dashed lines on the stomatal conductance panel show the stomatal fraction of
1298 total canopy conductance ($g_s\ g_c^{-1}$) and dashed lines on the non-stomatal conductance panel show
1299 the parameterized value.

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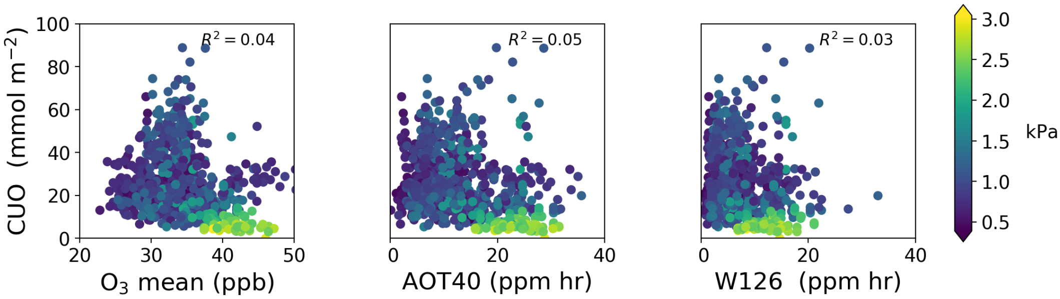

1300
1301 Figure 6. Comparison of cumulative uptake of O_3 (CUO) to concentration-based metrics of O_3
1302 exposure during the daytime growing season (April-September) at 103 sites: mean O_3
1303 concentration (left), AOT40 (center), and W126 (right). There is one value (dot) per site per
1304 year. Colors show mean vapor pressure deficit during the growing season.
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1306