Supporting Information for "The combined impact of canopy stability and soil NOx exchange on ozone removal in a temperate deciduous forest"

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1. Text S1: Spatio-temporal context of ozone deposition

We analyze a synthetic ozone flux data set (SynFlux; Ducker et al., 2018), where the stomatal ozone flux is derived for flux tower eddy covariance measurements based on a combination of inferred stomatal conductance (by inverting the Penman-Monteith equation for flux tower measurements), a gridded dataset of surface ozone concentrations, and a parameterized non-stomatal ozone flux component. Figure 1a shows SynFlux-derived stomatal and total ozone fluxes for summer 2012 (June-August) near North Italy. To place this in a temporal context, we calculate stomatal and total ozone flux anomalies by subtracting the multi-year June-August flux from the June-August 2012 mean flux per site, depicted in Figure 1b.

The ozone flux anomalies in Figure 1b are overlaid on a Standardized Precipitation-Evaporation Index (SPEI) map for June-August 2012. SPEI is a drought index that is based on the difference between precipitation and potential evaporation (Vicente-Serrano et al., 2010). SPEI can be integrated over different timescales; we here use the 6-month SPEI to analyze water deficits occurring over a 6-month time period to capture effects from the onset of the growing season. A 6-month SPEI time series over 1989-2018 is shown in Figure S1. The negative SPEI values in Figure 1b (range: -1.17 - -0.95) indicate a water deficit in summer 2012, but this falls within the 1-σ range of North-Italian summer SPEI-values in the climatological time period. We therefore conclude that the Bosco Fontana observations in summer 2012 are likely representative for typical summer conditions in this region.

References

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Vicente-Serrano, S. M., Beguería, S., & López-Moreno, J. I. (2010). A multiscalar drought index sensitive to global warming: The standardized precipitation evapotranspiration index. *Journal of Climate*, *23*(7), 1696–1718. doi: 10.1175/2009JCLI2909.1

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Table S1. A-g_s settings used in the MLC-CHEM simulations. The first column contains C3 reference settings (Ronda et al. 2001), and the second column contains the values applied in this study.

	C3 (reference)	Bosco Fontana (this study)
$g_{m,298}$ (mm s^{-1})	7.0	1.5
f_0 (-)	0.89	0.99
g_{m,T_1} (K)	278	283
g_{m,T_2} (K)	301	306
A_{m,max,T_1} (K)	281	286

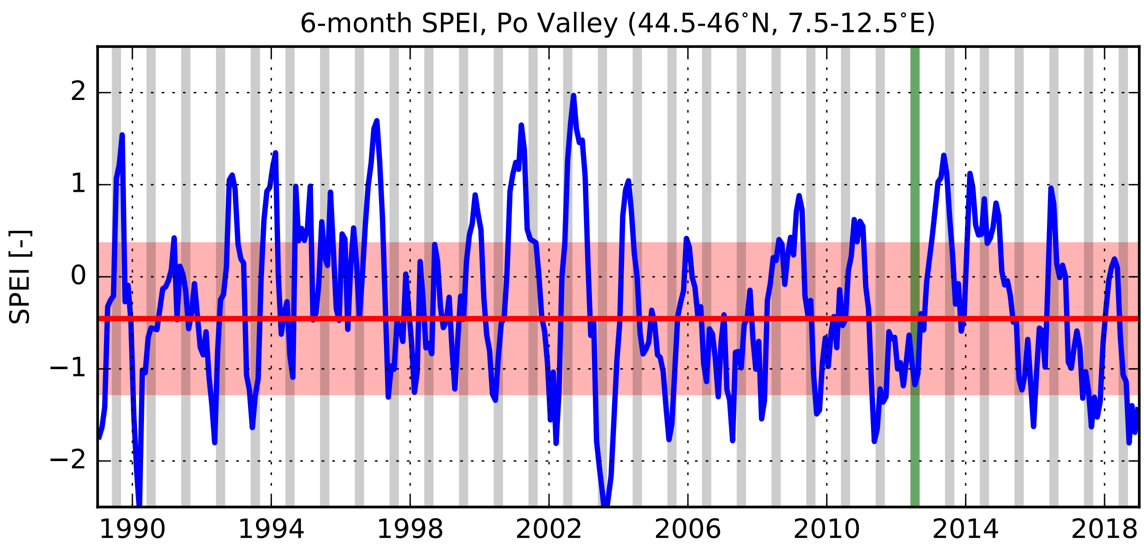

Figure S1. 30-year time series (1989-2018) of the Standardized Precipitation Evaporation Index (SPEI) integrated over the preceding 6 months for the Po Valley in North Italy. Red line and shaded area indicate the June-August mean 6-month SPEI value over the 30-year time series. The green shaded area indicates June-August 2012 when the Bosco Fontana intensive measurement campaign took place.

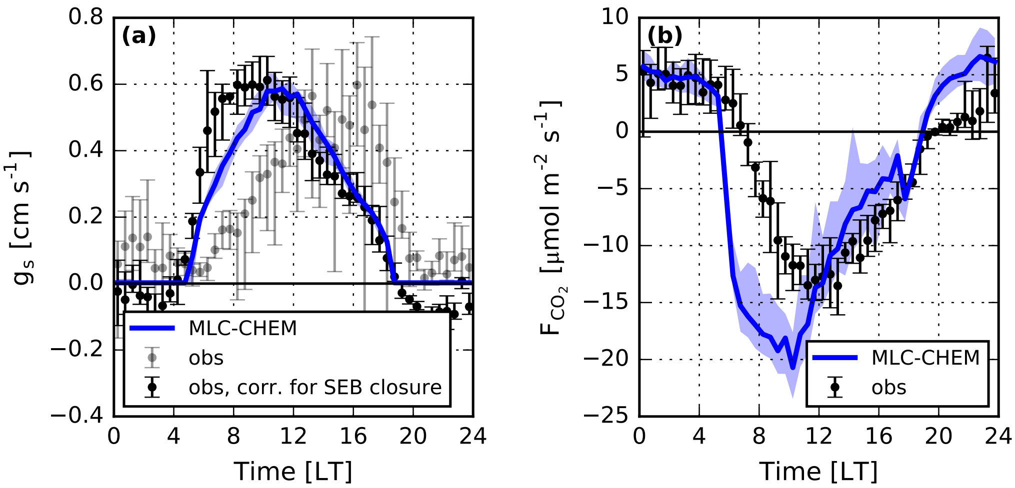

Figure S2. Diurnal averages of stomatal conductance (left panel) and the canopy-top CO_2 flux (right panel), as observed at Bosco Fontana (points and whiskers), and as simulated by MLC-CHEM with two different A-gs configurations (red and blue lines).

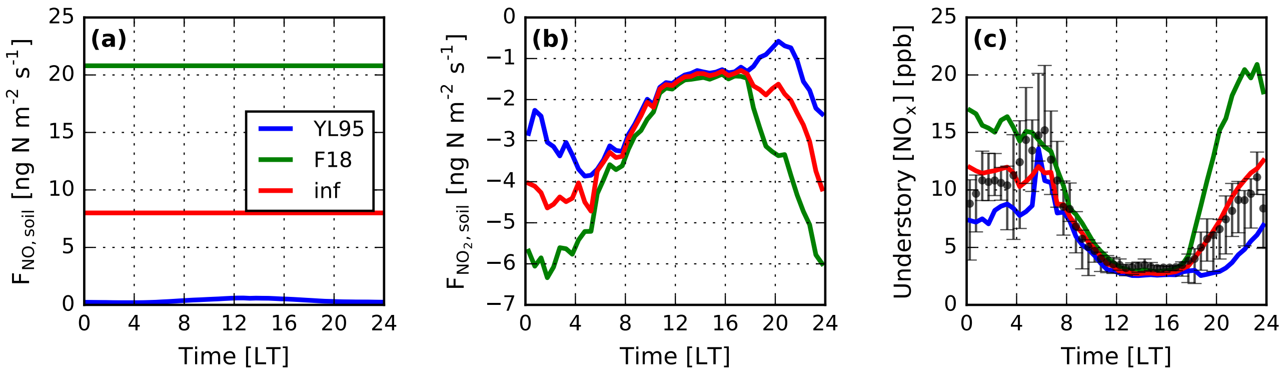

Figure S3. Diurnally averaged soil fluxes of NO (panel a) for different MLC-CHEM runs during July 2012 in Bosco Fontana, based on default MLC-CHEM emissions factors for deciduous forests (blue line; Yienger & Levy, 1995), the emission strength at Bosco Fontana derived from observations above the forest floor (green line; Finco et al., 2018) and the inferred "effective" soil NO flux representative for the soil impact on simulated mixing ratios at 6.5 m. Panels b and c show the resulting impacts in the diurnal averages of the soil NO_2 deposition flux and NO_x mixing ratios in the understory, respectively. Note that the three MLC-CHEM simulations presented in this figure have been performed with MLC-CHEM's reference parameterization of vertical exchange (REF).

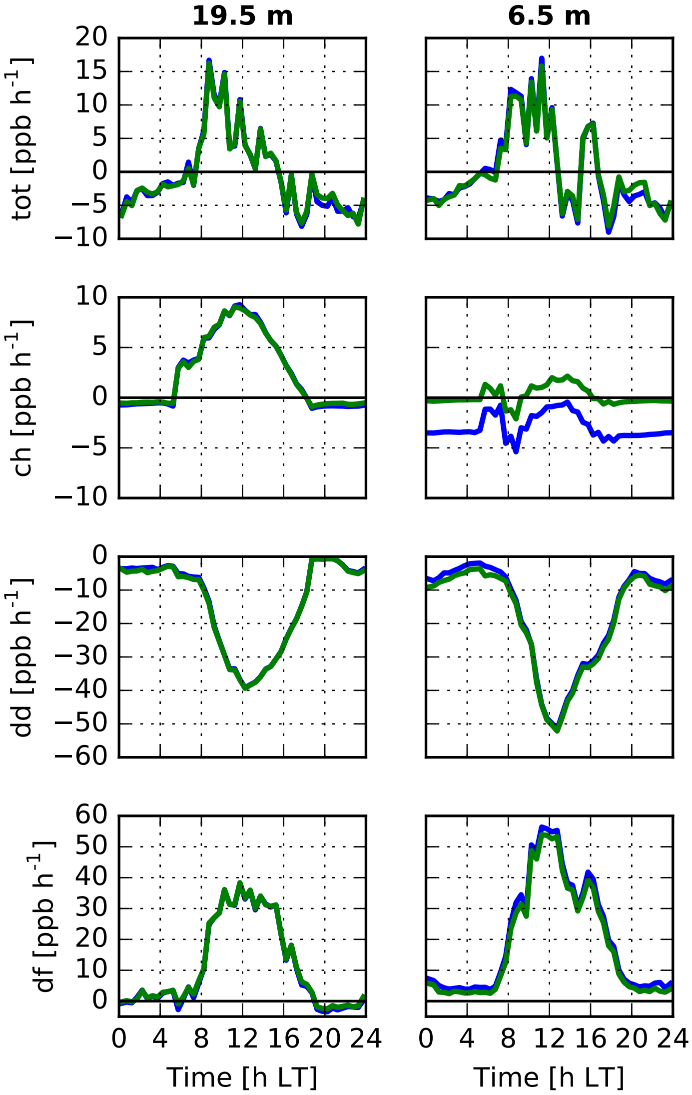

Figure S4. Mean diurnal variation in MLC-CHEM-simulated process tendencies in the upper canopy (19.5 m) and the lower canopy (6.5 m) for the simulations with and without soil NO_x exchange (simulations 4 and 7 in Table 1 in the main text). Tendencies from the following processes are shown: vertical exchange (df), dry deposition (dd), chemistry (ch), and total (tot, i.e., the sum of the previous three tendencies).

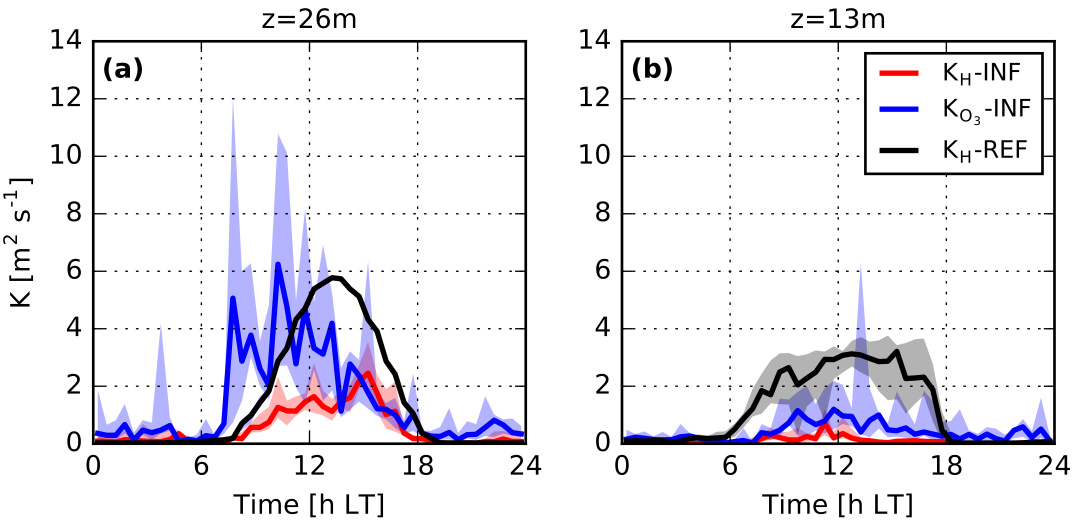

Figure S5. Diurnal variation in vertical diffusivity derived from MLC-CHEM's reference simulation (black line), inferred from sensible heat flux and potential temperature observations (red line), and from vertical profile measurements of the ozone flux and ozone mixing ratios (blue line; obtained by applying Eqn. 2 in the main text for the observed ozone flux and vertical gradient). Solid lines display the campaign median, and shaded areas indicate the inter-quartile range.