Looking for Indirect Observations of Dry Deposition: The Ambient Ozone-VPD Correlation

Sarah C. Kavassalis and Jennifer G. Murphy
University of Toronto, Department of Chemistry, Toronto, Ontario, Canada

Background

Meteorological controls on ozone pollution are well documented [1-5] however, models disagree regionally on both the sign and magnitude of the surface ozone response to projected climate change due to contrasting responses to meteorology. Here we present observations of ozone-meteorology coupling in the United States and show that from spring to fall, day-to-day variability in afternoon surface ozone levels is strongly correlated with day-to-day variability in vapor pressure deficit (VPD), a key variable in determining stomatal conductance and thus dry deposition. Through simple modeling, we show that a plausible explanation for the heretofore unexplained ozone-humidity correlation is the VPD-dependent dry deposition sink. This analysis helps to explain the observed ozone-meteorology correlations not currently captured by models and suggests an indirect metric for ozone-deposition that can be observed in ambient datasets.

Observations

O_3 and meteorological data from 1987 and 2015 are obtained from the Environmental Protection Agency's Clean Air Status and Trends Network (CASTNet). The analysis is confined to 101 rural CASTNet stations reporting hourly O_3, temperature (T), relative humidity (RH), incoming solar radiation (SR), wind speed (WS), wind direction (WD), and soil wetness (WET) with at least 70 stations active at any given time. Measurements of hourly soil wetness stopped in 2012. VPD and water vapor pressure (P_{H2O}) were calculated from temperature and relative humidity measurements.

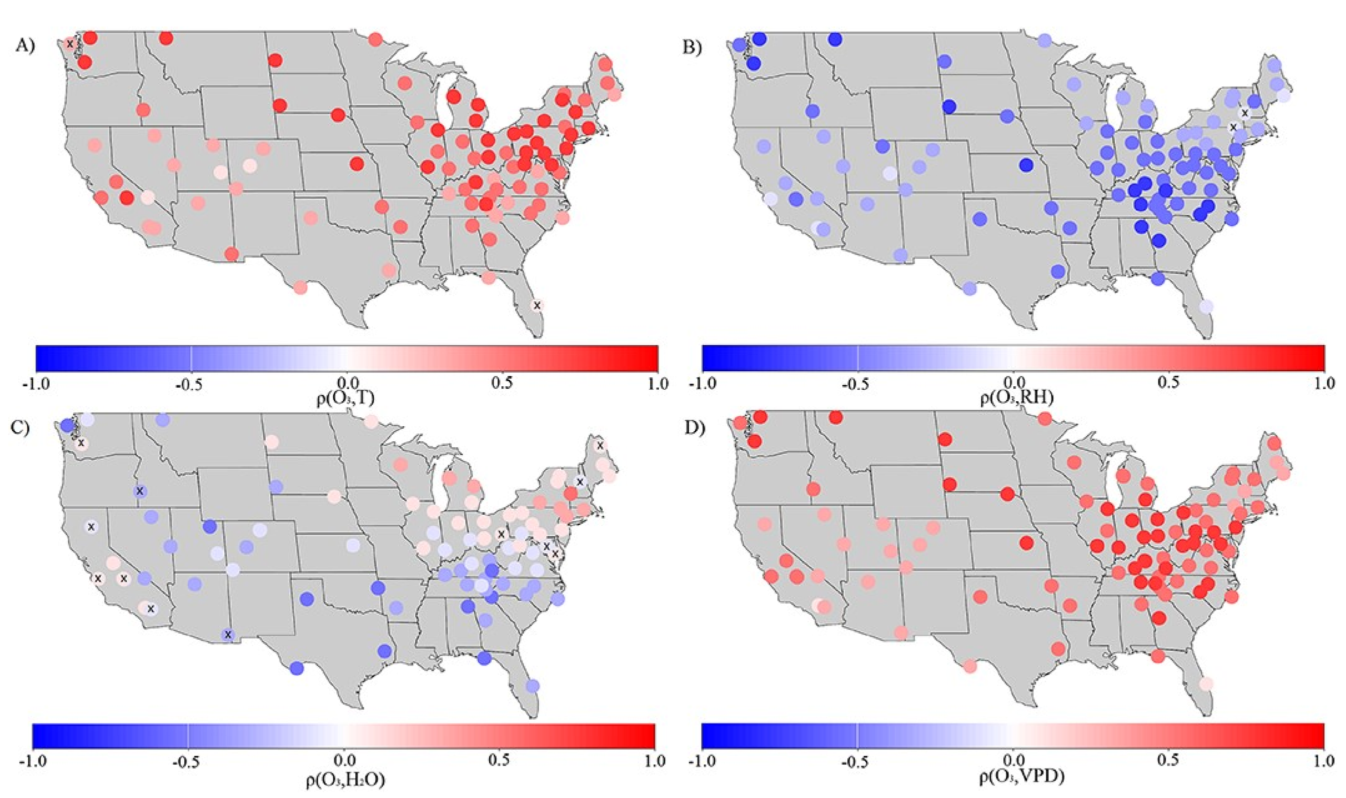

Fig 1. Summer observed midday (12–4 P.M.) average Pearson's correlation coefficient of ozone (a) temperature, (b) relative humidity, (c) P_{H2O}, and (d) VPD (Stations for which correlations were not significant ($p \geq 0.01$) are noted with an x.

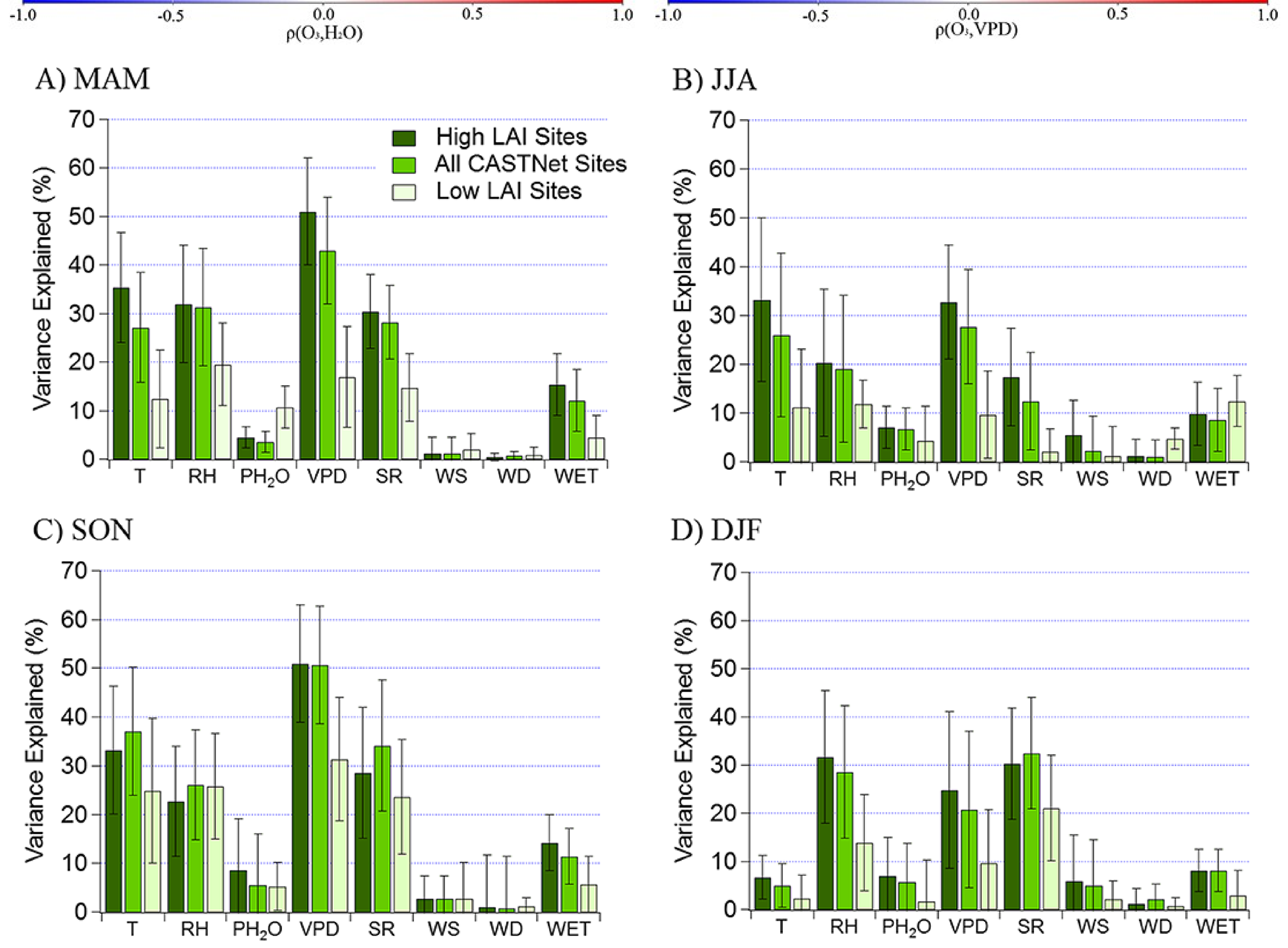

Fig 2. Midday daily ozone variance attributed to midday surface meteorology for (a) spring, (b) summer, (c) fall, and (d) winter. The error bars represent one standard deviation from the mean. High LAI sites are those with summer LAI > 3.8, and low LAI sites are those with summer LAI < 1.7.

A Role for Dry-Deposition?

Dry deposition is thought to be the dominant sink for ozone in the summertime planetary boundary layer over the continental US [6] with 30-90% of that dry deposition sink attributable to stomatal uptake [7-8]. Stomatal resistance is controlled by response to sunlight, CO_2, temperature, soil water potential, and VPD. **VPD is the difference between ambient vapour pressure, vp_{air}, and saturated vapour pressure, vp_{sat}** and is thus coupled with humidity and temperature.

$$\mathrm{VPD} = P_{H_2O,sat}(T) - P_{H_2O} = \left(1 - \frac{RH}{100}\right) P_{H_2O,sat}(T)$$

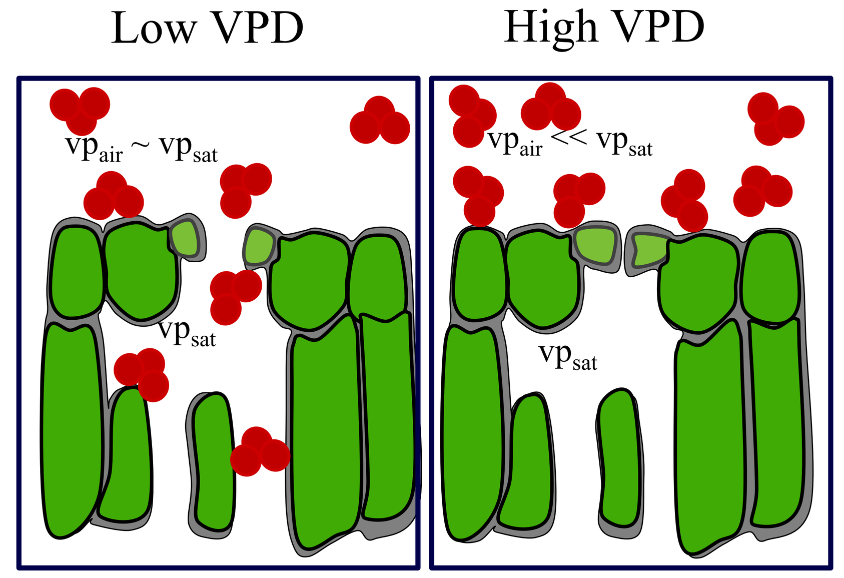

Fig. 3. Representation of uptake of ozone by vegetation through stomata. High VPD means stomata close, removing a large ozone sink.

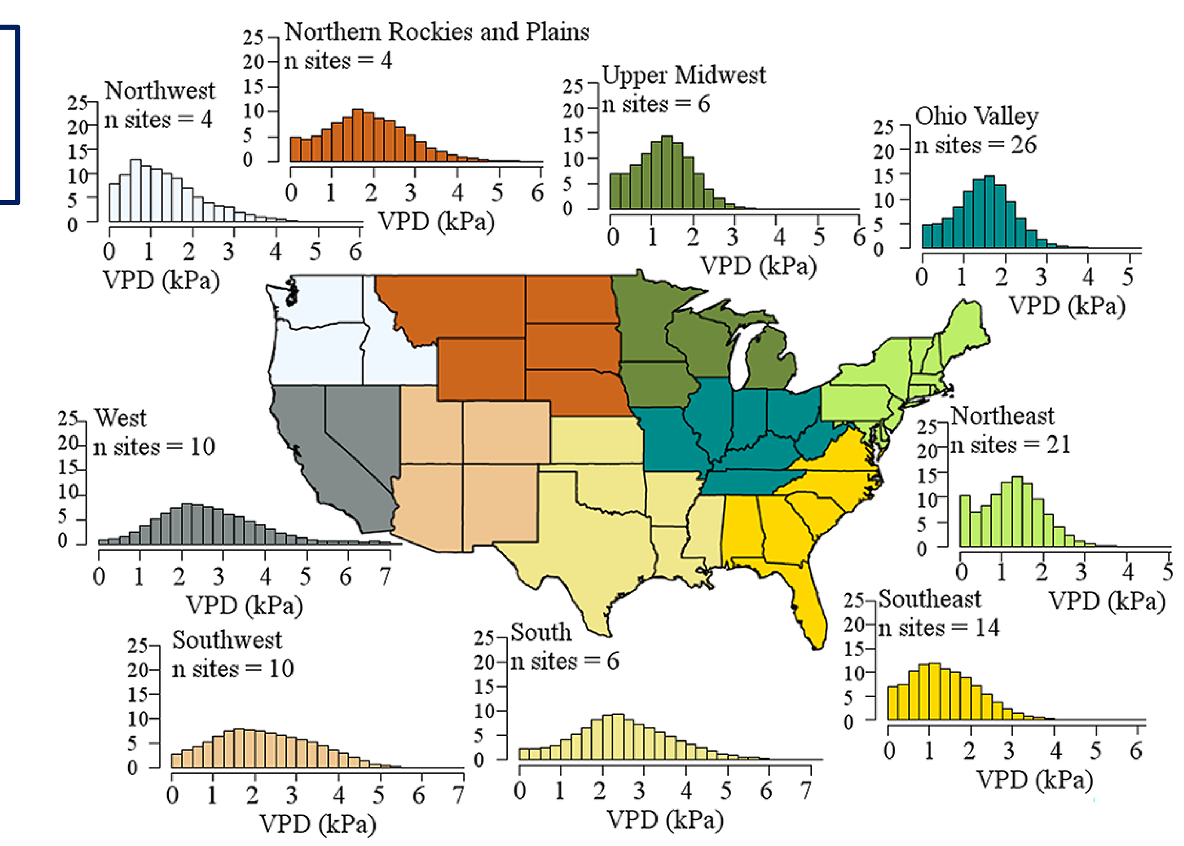

Fig. 4. Percentage relative frequency histograms of hourly VPD for midday summer at CASTNet stations.

Above a species-dependent optimal vapor pressure, stomatal conductance is regulated by VPD. This threshold value has been estimated to be from 0.31 to 1.8 kPa depending on plant type [9]. Fig 4 shows that VPD is highly variable, with values above 1.8 kPa occurring frequently in all climate regions.

If stomatal uptake of ozone is a major sink in the continental surface layer and is strongly dependent on VPD, this could be an important explanatory factor for observed O_3-RH/VPD correlations.

A Simple Model

To investigate the meteorological sensitivity of ozone resulting from VPD-dependent deposition, we used a simple photochemical model with a resistance-based deposition scheme (Fig. 5). Figure 6 shows that only by including VPD-dependent stomatal conductance in the deposition scheme can we produce a strong RH/VPD dependence in the net ozone production.

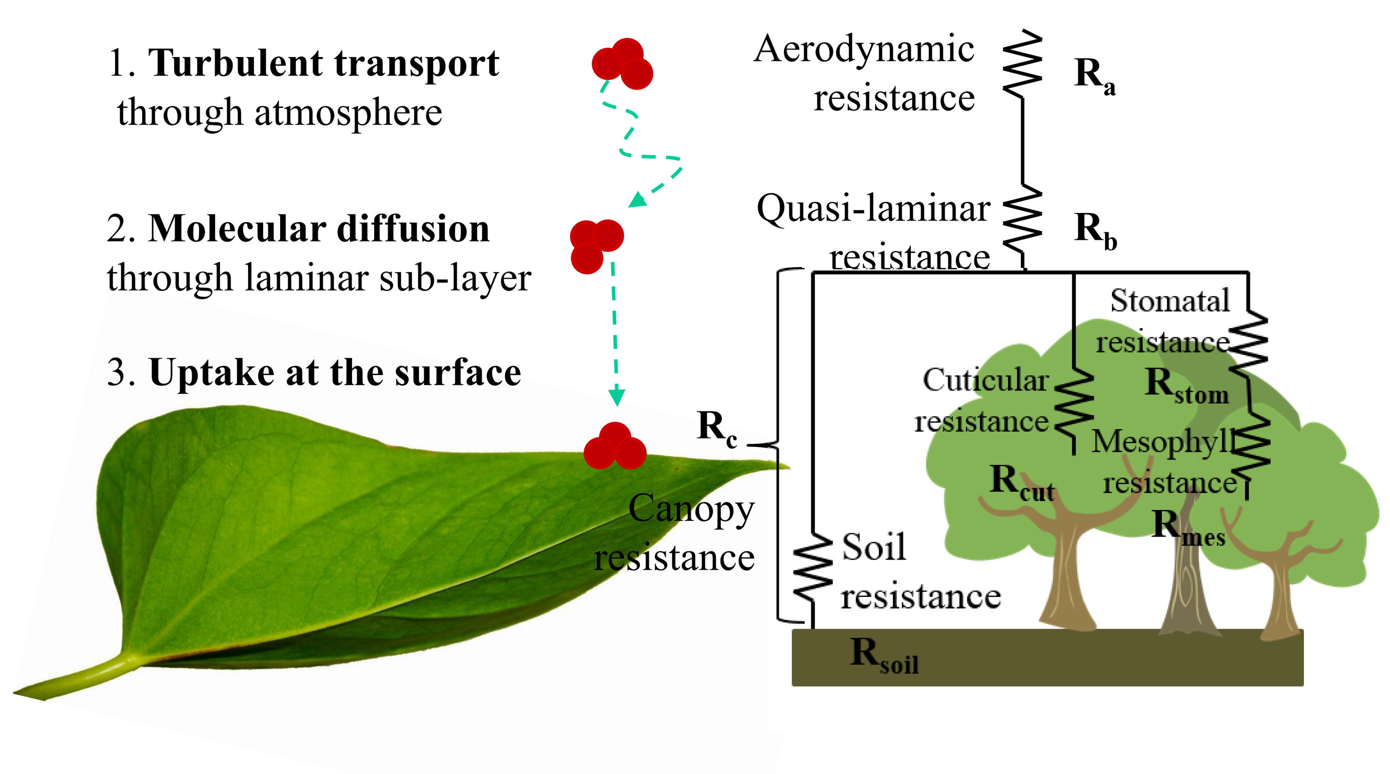

Fig. 5. Schematic of Wesely-based deposition scheme [10]

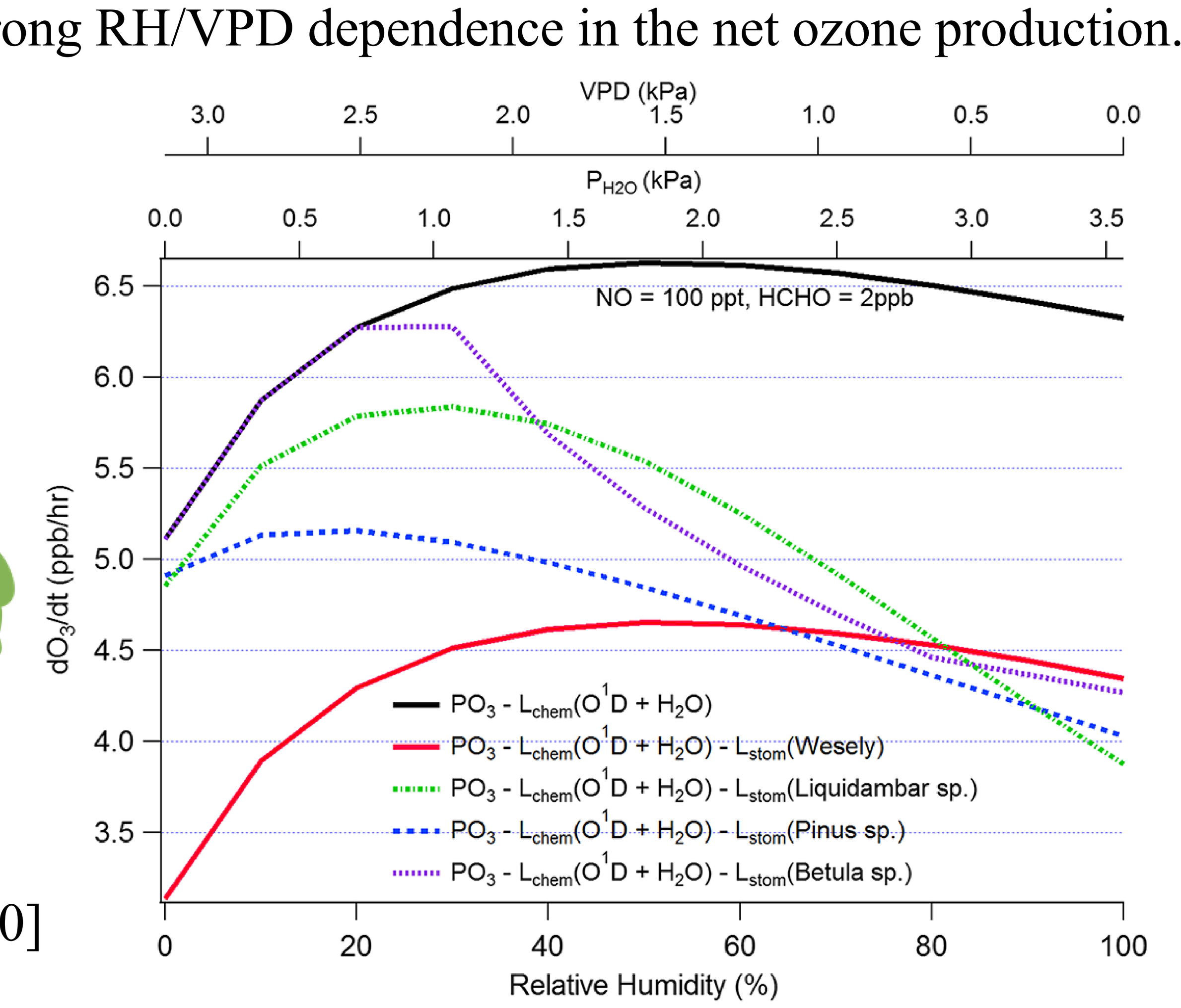

Fig. 6. Instantaneous ozone rate of change (PO_3-LO_3) under simulated summer conditions. Stomatal loss is parameterized by using the Wesely scheme (red) and then for three species-specific empirical parametrizations (sequential dashed lines) [11].

Model details and conditions can be found in *Kavassalis and Murphy (2017), Geophys. Res. Lett., 44, 2922–2931.*

References and Acknowledgements

[1] Jacob et al., JGR, 1993. [2] Cox et al., Atmos. Environ., 1993. [3] Camalier., Atmos. Environ., 2007. [4] Tawfik et al., Atmos. Environ., 2013. [5] Davis et al., Atmos. Environ., 2011. [6] Racherla and Adams, ACP, 2008. [7] Fowler et al., Water Air Soil Poll., 2001. [8] Hardacre et al., ACP, 2015. [9] Simpson et al., *EMEP MSC-W Note.* 2003. [10] Wesely, Atmos. Environ., 1989 [11] Kavassalis & Murphy, GRL, 2017.

This research was supported by NSERC CREATE IACPES (Integrating Atmospheric Chemistry And Physics From Earth To Space). Ground station data was made available by the U.S. EPA Clean Air Markets Division.

skavassa@chem.utoronto.ca