

AIR POLLUTION-ECOSYSTEM FEEDBACKS: OBSERVATIONAL CONSTRAINTS ON OZONE-FOREST INTERACTIONS FROM AN UNMANNED AERIAL VEHICLE

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Abstract

Tropospheric ozone (O_3) is an air pollutant and key atmospheric oxidant. Global chemical transport models suggest 15–30% of tropospheric O_3 is lost by deposition to the Earth's surface annually. We require knowledge of O_3 losses to predict O_3 variability in the troposphere; however, there are few direct measurements of the atmosphere-surface loss rate, known as the deposition velocity. Here, we describe a method for observationally-deriving the O_3 deposition velocity using a flux-gradient approach combined with a light-weight low-power measurement payload onboard an unmanned aerial vehicle (UAV). Using indoor and outdoor test flight data, we demonstrate that combined uncertainties in O_3 concentration and horizontal wind measurements are small enough to derive O_3 deposition velocities on the order of those observed over a variety of vegetated landscapes. Here, we present UAV payload design, the derivation for deposition velocity, and preliminary flight data.

Introduction

Tropospheric ozone (O_3) is a pollutant that is harmful to humans (Cohen et al., 2017; Jerrett et al., 2009) and plants (e.g., Ashmore, 2005; Reich, 1987). O_3 plant damage has been observed in forests (Ainsworth et al., 2012; references therein) and linked to decreases in global crop production (Avnery et al., 2011a, 2011b; Wilkinson et al., 2011), with the severity of O_3 effects varying as a function of plant species and development stage (Bergman et al., 1995; Fuhrer et al., 2003; Prozherina et al., 2003). O_3 is a greenhouse gas that also

affects climate indirectly (IPCC, 2013), altering ecosystem gross and net primary productivity (Fares et al., 2013; Paoletti and Grulke, 2010; Tai et al., 2014; Wittig et al., 2007, 2009) and contributing to the production of secondary organic aerosol (Jokinen et al., 2015; Liu et al., 2011).

Global chemical transport models suggest 15–30% of tropospheric O_3 is lost by deposition to the Earth's surface annually (Wild, 2007; Young et al., 2013), with deposition to plant canopies via stomatal uptake being generally the dominant loss process over terrestrial ecosystems (30–90%) (e.g., Cieslik, 2004; Clifton et al., 2017; Fares et al., 2014; Fowler et al., 2001, 2009). The O_3 velocity of deposition (v_d) is highly variable with vegetation, land cover type, and environmental conditions that influence the rate of stomatal conductance (g_s). O_3 v_d over vegetation typically ranges from 0 to 1 $cm\ s^{-1}$, with higher deposition velocities over forests and croplands and lower values over grasslands and tundra (Hardacre et al., 2015). O_3 v_d is both over and under predicted by models by a factor of two, with measurement-model discrepancies varying with vegetation type (Clifton et al., 2017; Fowler et al., 2009; Hardacre et al., 2015; Val Martin et al., 2014). While knowledge of O_3 v_d is important to the global O_3 budget, O_3 deposition rates are among the most uncertain and poorly constrained terms (Wild, 2007). Uncertainties are due in part to lack of O_3 v_d observations over the range of vegetative canopy types, with many global ecosystems under or unsampled (Young et al., 2018).

O_3 v_d over vegetative canopies can be empirically determined from measurements of

the O_3 flux (F_{O_3}) using the eddy-covariance technique (Eq. 1). Eddy-covariance is a micrometeorological method based on principles of mass and momentum conservation, in which F_{O_3} is equivalent to the turbulence-driven O_3 vertical mass transport through a reference layer, and computed as the deviation from the mean covariance of measured O_3 concentrations and vertical winds (w) (e.g., Baldocchi et al., 1988; Foken, 2006; McMillen, 1988) (Eq. 2).

$$O_3 v_d = -F_{O_3} / [O_3] \quad (1)$$

$$F_{O_3} = \int_{t_0}^t w' [O_3]' dt = \frac{1}{n} \sum_{i=1}^n (w_i - \bar{w}) \left([O_3]_i - \overline{[O_3]} \right) = \overline{w' [O_3]'} \quad (2)$$

In practice, these observations are collected from atop stationary research towers, representing homogenous plant canopies, with fast (generally 10 s^{-1}) 3-dimensional anemometers and high-precision sub-second time response chemical analyzers to capture higher-frequency eddy flux contributions. As a result, infrastructure investments are typically required, which may be substantial for taller plant canopies and difficult to maintain in remote areas. Moreover, there are few off-the-shelf commercially-available O_3 instruments that meet the analytical requirements of eddy-covariance.

$O_3 v_d$ can also be measured using the flux-gradient method with light-weight, low-power, commercially-available equipment. Flux-gradient methods quantify F_{O_3} as a function of the vertical atmospheric concentration and horizontal wind speed (u) gradients, and the Von Kármán constant (k), a measure of the turbulent fluid flow gradient at a boundary (Eq. 3 & 4) (Keenan et al., 2012).

The flux-gradient method has been implemented to determine $O_3 v_d$ from multiple platforms, including tethered balloons (Broder et al., 1981), aircrafts using O_3 columns measured with LIDAR systems (Couach et al., 2003), as well as modeled (Kramm et al., 1995;

Sitch et al., 2007), but is most commonly measured from stationary research towers or ship masts (Bocquet et al., 2011; Colbeck and Harrison, 1985; Droppo Jr, 1985; Duyzer et al., 1983; Duyzer and Westrate, 1995; Enders, 1992; Fontan et al., 1992; Galbally and Allison, 1972; Garland and Derwent, 1978; Hargreaves et al., 1992; Hole et al., 2004; Horvath et al., 1998; Kramm et al., 1991; Matsuda et al., 2005; Mikkelsen et al., 2000; Muller et al., 2009; Neiryck et al., 2012; Zapletal et al., 2011). Agreement between flux-gradient and eddy-covariance derived $O_3 v_d$ and flux-gradient and modeled $O_3 v_d$ have been reported (Bocquet et al., 2011; Droppo Jr, 1985; Duyzer and Westrate, 1995; Hole et al., 2004; Mikkelsen et al., 2000; Muller et al., 2009; Wohlfahrt et al., 2009).

Unmanned aerial vehicles (UAVs) have increasingly become a platform for atmospheric sampling, including the deployment of instrumentation to measure greenhouse gases (Berman et al., 2012; Brody et al., 2017; Illingworth et al., 2014; Li et al., 2017; McGonigle et al., 2008; Rüdiger et al., 2018; Thomas et al., 2012; Watai et al., 2006), O_3 concentrations (Illingworth et al., 2014; Li et al., 2017), pollen (Crazzolara et al., 2018), and reactive organic gas collection for off-line analysis (McKinney et al., 2018). The quick to deploy mobile sampling has proven advantageous for research over things such as volcanic plumes (Diaz et al., 2015; McGonigle et al., 2008; Mori et al., 2016; Rüdiger et al., 2018).

In this paper, we describe a novel application of the flux-gradient method to determine $O_3 v_d$ from onboard an unmanned aerial vehicle (UAV). The UAV-based system can be easily deployed to ecosystems not readily sampled with eddy-covariance or tower-based flux-gradient approaches. First, we describe the flux-gradient method. Second, we describe the UAV system. Third, we describe UAV flight planning to derive vertical O_3 and u gradients.

Finally, we describe the study site where future science flights will take place.

Flux-gradient method

An alternative method to observationally determining O_3 v_d , known as the flux-gradient method, using slower time response O_3 instrumentation has found wide application. Here, F_{O_3} can be inferred from the vertical concentration gradient scaled by the eddy diffusivity constant (K), where K accounts for the rate of vertical mixing (Businger et al., 1971; Dyer and Hicks, 1970; Webb, 1970) (Eq. 3). The flux-gradient method is based off of micrometeorological similarity theory, wherein turbulent eddies carry all atmospheric constituents equally (e.g., O_3 and CO_2), such that K determined for one constituent represents all constituents (i.e. $K_{O_3} = K_{CO_2}$) (Oke, 1987). Eq. 3 is an analog of Fick's Law of Diffusion, with vertical transport due to the random movement of air parcels by turbulent diffusion along the concentration gradient (Denmead, 2008; Denmead and Bradley, 1985). As a result, molecular movement of a given trace gas follows the direction of high to low concentration of that species.

$$F_{O_3} = -K \frac{\partial[\overline{O_3}]}{\partial z} \quad (3)$$

Numerous equations have been developed to calculate K from measurements, such as momentum flux, sensible heat flux, friction velocity, and horizontal wind gradient (Denmead and Bradley, 1985; Droppo Jr, 1985; Goldstein et al., 1996; Meredith et al., 2014). In the latter case, K can be defined as a function of the horizontal wind speed (u), von Kármán constant (k), and measurement height (z) (Eq. 4) (Oke, 1987).

$$K = k^2 \left(\frac{z_2 - z_1}{\ln\left(\frac{z_2}{z_1}\right)} \right)^2 \left(\frac{\partial \bar{u}}{\partial z} \right) \quad (4)$$

Flux-gradient theory is best applied to species that are unreactive on timescales of turbulent transport and only valid under neutral

atmospheric stability conditions, when wind speeds and radiation fluxes are constant over the measurement period (Oke, 1987), and using measurements collected above the roughness sublayer (Raupach and Legg, 1984).

Under neutral conditions, buoyancy effects have little impact on measured concentration and wind gradients (Oke, 1987). Atmospheric stability can be determined using the Richardson number (Ri), which is a function of u , z , potential temperature (θ), and acceleration due to gravity (g) (Bocquet et al., 2011; Kaimal and Finnigan, 1994) (Eq. 5). When $Ri = 0 \pm 0.01$, conditions are neutral; when conditions are near-unstable ($-0.1 < Ri < -0.01$) or near-stable ($0.01 < Ri < 0.1$), an empirical correction using the value of Ri is applied to Eq. 3 (Bocquet et al., 2011; Oke, 1987).

$$Ri = \frac{g}{\theta} \left(\frac{\Delta \bar{\theta}}{\Delta z_{\theta}} \right) / \left(\frac{\Delta \bar{u}}{\Delta z_u} \right)^2 \quad (5)$$

Because near the canopy surface, turbulence is influenced by the roughness of the canopy top, creating wake turbulence and altering the vertical concentration and wind gradients of the atmosphere (Baldocchi et al., 1988; Neirynek et al., 2012; Oke, 1987), measurements should be collected above the so-called roughness sublayer. The roughness sublayer is approximately $h + 1.5L_t$, where h is the canopy height and L_t is the width of an average tree crown (Baldocchi et al., 1988; Raupach et al., 1980). Above the roughness sublayer, vertical flux divergence or convergence is less than 10 percent of the flux, making it an approximately constant flux layer (Oke, 1987).

We combine O_3 concentration and horizontal wind speed data to derive F_{O_3} using the flux-gradient method (e.g., Bocquet et al., 2011).

UAV System

A hex-rotor UAV (DJI M600 Pro) has been modified to support a 6 kg, 300 W payload measuring O_3 concentration (205 O_3 analyzer, 2B Technologies); and horizontal wind speeds



Fig. 1. Modified hex-rotor UAV with mounted O₃ analyzer and anemometer.

(DS-2 Sonic Anemometer, Decagon Devices) (Fig. 1). The O₃ analyzer is mounted underneath the UAV body. The anemometer is mounted atop a 0.5 m pole affixed to the UAV body. Additionally, the UAV contains a flight computer and batteries for the instrumentation and computer, both mounted to the body of the UAV.

Study Site

Test flights were conducted at the UVA JPJ Basketball Arena and the Milton Field Airport, near Charlottesville, Va. Subsequent science flights will take place at the Virginia Forest Laboratory (VFL). The VFL is located in the forested Piedmont region of Virginia at the eastern base of the Blue Ridge Mountains ((37.92°N, 78.27°W). The canopy is 24 meters tall, comprised of mixed deciduous trees with patches of conifer, representative of second-growth forests throughout the region.

Test Flights

First, to characterize rotor interference on the anemometer data, test flights were completed indoors at the UVA JPJ Basketball Arena, a near-windless environment, and no difference was found in the anemometer measurements with the rotors on and with the rotors off, indicating that the rotors of the UAV do not interfere with the wind speed measurements (Fig. 2).

Combined uncertainties in O₃ concentration and horizontal wind velocities are small enough to sample O₃ deposition velocities of 0.02 cm s⁻¹ at 10 second averaging times at S/N = 3. This detection limit is low enough to capture previously observed variability over a variety of

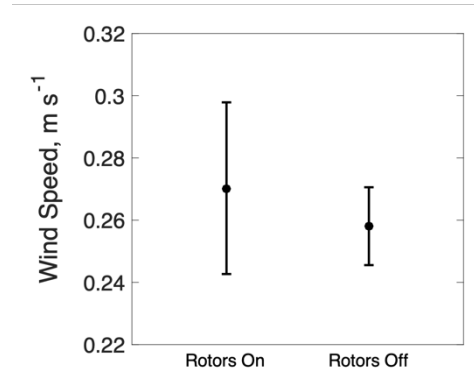


Fig. 2. Wind speed and standard error with UAV rotors off and with rotors on.

vegetated landscapes of 0.1–0.5 cm s⁻¹ in months when photosynthetic activity is high (Hardacre et al., 2015). The UAV is flown in a three-phase square wave (Fig. 3). Sampling occurs ahead of UAV movement to minimize disruption of vertical atmospheric structuring. The UAV is flown above the roughness sublayer in the constant flux layer.

The instrumentation on the UAV is able to capture the vertical variability in the O₃ mixing ratio and the wind speed (Fig. 4 & 5).

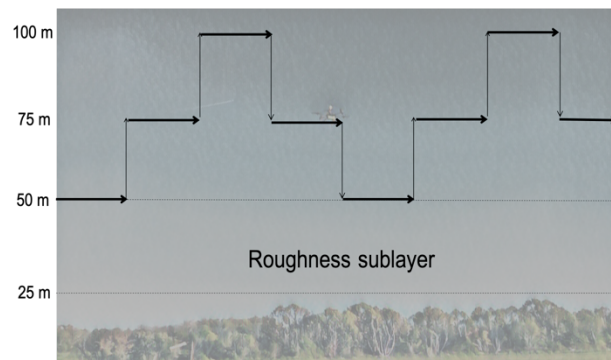


Fig. 3. Flight path sampling in square wave over a forest canopy.

Future Work

Spring–summertime observations will be made from above the forest at the VFL. We will compare the UAV-based O₃ v_d to O₃ v_d derived using eddy-covariance O₃ flux measured at the VFL research tower with in the UAV footprint. We will qualify and

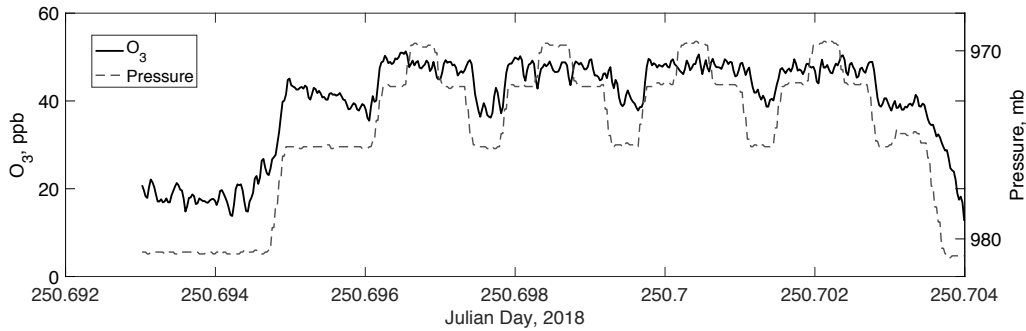


Fig. 4. O₃ mixing ratio in ppb (solid line) and cell pressure, a proxy for altitude (dashed line) at Milton Field Airport on September 7, 2018.

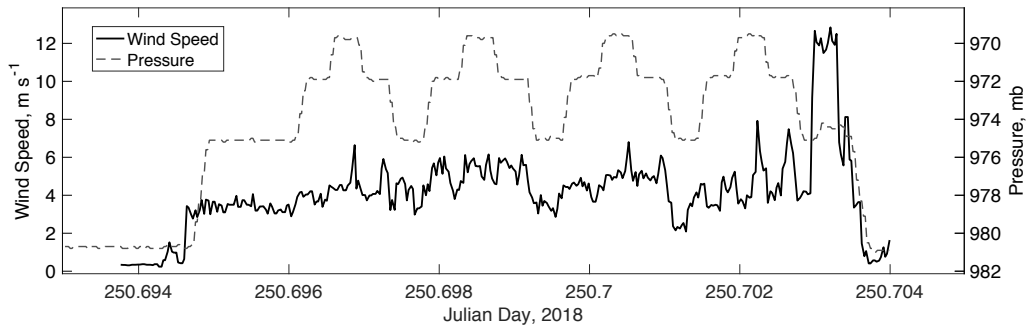


Fig. 5. Wind speed in m s⁻¹ (solid line) and cell pressure, a proxy for altitude (dashed line) at Milton Field Airport on September 7, 2018.

quantify uncertainties associated with the UAV-based O₃ v_d measurements.

Summary

This work describes a novel methodology for observationally-deriving the O₃ deposition velocity. Using a flux-gradient approach combined with a light-weight low-power measurement payload, O₃ and horizontal wind speed can be measured onboard an unmanned aerial vehicle (UAV) at multiple altitudes. The test flight data demonstrates that the rotors do not interfere with the wind speed measurements and that combined uncertainties in O₃ concentration and horizontal wind measurements are small enough to derive O₃ v_d on the order of previously modeled and observed values.

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References

- Ainsworth, E. A., et al., *Annu. Rev. Plant Biol.*, 63, 637–661, doi:10.1146/annurev-arplant-042110-103829, 2012.
- Ashmore, M. R., *Plant, Cell Environ.*, 28, 949–964, 2005.
- Avnery, S., et al., *Atmos. Environ.*, 45, 2284–2296, doi:10.1016/j.atmosenv.2010.11.045, 2011a.
- Avnery, S., et al., *Atmos. Environ.*, 45, 2297–2309, doi:10.1016/j.atmosenv.2011.01.002, 2011b.

- Baldocchi, D. D., et al., *Ecology*, 69(5), 1331–1340, 1988.
- Bergman, E., et al., *Water, Air Soil Pollut.*, 85, 1437–1442, 1995.
- Berman, E. S. F., et al., *Sensors Actuators B. Chem.*, 169, 128–135, doi:10.1016/j.snb.2012.04.036, 2012.
- Bocquet, F., et al., *Atmos. Meas. Tech.*, 4, 2305–2321, doi:10.5194/amt-4-2305-2011, 2011.
- Broder, B., Dutsch, et al., *Atmos. Environ.*, 15(7), 1195–1199, 1981.
- Brosy, C., et al., *Atmos. Meas. Tech.*, 10, 2773–2784, 2017.
- Businger, J. A., et al., *J. Atmos. Sci.*, 28, 181–189, 1971.
- Cieslik, S. A., *Atmos. Environ.*, 38, 2409–2420, doi:10.1016/j.atmosenv.2003.10.063, 2004.
- Clifton, O. E., et al., *Geophys. Res. Lett.*, 44, 542–552, doi:10.1002/2016GL070923, 2017.
- Cohen, A. J., et al., *Lancet*, 389(10082), 1907–1918, doi:10.1016/S0140-6736(17)30505-6, 2017.
- Colbeck, I. and Harrison, R. M., *Atmos. Environ.*, 19(11), 1807–1818, 1985.
- Couach, O., et al., *Atmos. Chem. Phys.*, 3, 549–562, 2003.
- Crazzolara, C., et al., *Atmos. Meas. Tech. Discuss.*, <https://doi.org/10.5194/amt-2018-305>, in review, 2018.
- Denmead, O. T., *Plant Soil*, 309, 5–24, doi:10.1007/s11104-008-9599-z, 2008.
- Denmead, O. T. and Bradley, E. F., B.A. Hutchinson B.B. Hicks (Eds), *For. Interact. Reidel, Dordr.*, 421–442, 1985.
- Diaz, J. A., et al., *Am. Soc. Mass Spectrom.*, 26(2), 292–304, doi:10.1007/s13361-014-1058-x, 2015.
- Droppo Jr, J. G., *J. Geophys. Res.*, 90, 2111–2118, 1985.
- Duyzer, J. and Westrate, H., *Stud. Environ.*, 64, 21–30, 1995.
- Duyzer, J. H., et al., *Atmos. Environ.*, 17(10), 2117–2120, 1983.
- Dyer, A. J. and Hicks, B. B., *Q. J. R. Meteorol. Soc.*, 98, 206–212, 1970.
- Enders, G., *Environ. Pollut.*, 75, 61–67, 1992.
- Fares, S., et al., *Glob. Chang. Biol.*, 19, 2427–2443, doi:10.1111/gcb.12222, 2013.
- Fares, S., et al., *Agric. For. Meteorol.*, 198–199, 181–191, doi:10.1016/j.agrformet.2014.08.014, 2014.
- Foken, T., *Boundary-Layer Meteorol.*, 119, 431–447, doi:10.1007/s10546-006-9048-6, 2006.
- Fontan, J., et al., *Atmos. Environ.*, 26A(5), 863–869, 1992.
- Fowler, D., et al., *Plant Soil*, 228, 117–129, 2001.
- Fowler, D., et al., *Atmos. Environ.*, 43, 5193–5267, doi:10.1016/j.atmosenv.2009.07.068, 2009.
- Fuhrer, J., et al., *Establ. Ozone Crit. Levels II* (eds P.E. Karlsson, G. Sellden H. Pleijel), 183–198, IVL, Stockholm, Sweden, 2003.
- Galbally, I. and Allison, I., *J. Geophys. Res.*, 77(21), 3946–3949, 1972.
- Garland, J. A. and Derwent, R. G., *Q. J. R. Meteorol. Soc.*, 105(443), 169–183, 1978.
- Goldstein, A. H., et al., *J. Geophys. Res.*, 101, 9149–9157, 1996.
- Hardacre, C., et al., *Atmos. Chem. Phys.*, 15, 6419–6436, doi:10.5194/acp-15-6419-2015, 2015.
- Hargreaves, K. J., et al., *Environ. Pollut.*, 75, 53–59, 1992.
- Hole, L. R., et al., *Atmos. Environ.*, 38, 2217–2223, doi:10.1016/j.atmosenv.2003.11.042, 2004.
- Horvath, L., et al., *Atmos. Environ.*, 32(7), 1317–1322, 1998.
- Illingworth, S., et al., *Atmos. Sci. Lett.*, 15, 252–258, doi:10.1002/asl2.496, 2014.

- IPCC: Climate Change 2013, Cambridge, United Kingdom New York, NY, USA, doi:10.1017/CBO9781107415324, 2013.
- Jerrett, M., et al., *N. Engl. J. Med.*, 360(11), 1085–1095, 2009.
- Jokinen, T., et al., *Proc. Natl. Acad. Sci.*, 112(23), 7123–7128, doi:10.1073/pnas.1423977112, 2015.
- Kaimal, J. C. and Finnigan, J. J., Oxford Univ. Press. New York, Oxford, 289 pp., 1994.
- Keenan, T. F., et al., *Glob. Chang. Biol.*, 18(8), 2555–2569, doi:10.1111/j.1365-2486.2012.02684.x, 2012.
- Kramm, G., et al., *J. Atmos. Chem.*, 13, 265–288, 1991.
- Kramm, G., et al., *Atmos. Environ.*, 29(21), 3209–3231, 1995.
- Li, X.-B., et al., *Environ. Pollut.*, 224, 107–116, doi:10.1016/j.envpol.2017.01.064, 2017.
- Liu, S., et al., *Atmos. Chem. Phys.*, 11, 8321–8341, doi:10.5194/acp-11-8321-2011, 2011.
- Matsuda, K., et al., *Atmos. Environ.*, 39, 2571–2577, doi:10.1016/j.atmosenv.2005.01.011, 2005.
- McGonigle, A. J. S., et al., *Geophys. Res. Lett.*, 35, L06303, doi:10.1029/2007GL032508, 2008.
- McKinney, K. A., et al., *Atmos. Meas. Tech. Discuss.*, <https://doi.org/10.5194/amt-2018-277>, in review, 2018.
- McMillen, R. T., *Boundary-Layer Meteorol.*, 43(3), 231–245, 1988.
- Meredith, L. K., et al., *Atmos. Meas. Tech.*, 7, 2787–2805, doi:10.5194/amt-7-2787-2014, 2014.
- Mikkelsen, T. N., et al., *Environ. Pollut.*, 109, 423–429, 2000.
- Mori, T., et al., *Earth, Planets Sp.*, 68:49, doi:10.1186/s40623-016-0418-0, 2016.
- Muller, J. B. A., et al., *Atmos. Sci. Lett.*, 169, 164–169, doi:10.1002/asl, 2009.
- Neiryneck, J., et al., *J. Environ. Monit.*, 14, 1684–1695, doi:10.1039/c2em10937a, 2012.
- Oke, T. R.: *Boundary Layer Climates*, 2nd Edn, Univeristy Press. Cambridge, 435pp, 1987.
- Paoletti, E. and Grulke, N. E., *Environ. Pollut.*, 158, 2664–2671, doi:10.1016/j.envpol.2010.04.024, 2010.
- Prozherina, N., et al., *New Phytol.*, 159, 623–636, doi:10.1046/j.1469-8137.2003.00828.x, 2003.
- Raupach, M. R. and Legg, B. J., *Agric. Water Manag.*, 8, 119–131, 1984.
- Raupach, R., et al., *Bound. Layer Meteorol.*, 18, 373–338, 1980.
- Reich, P. B., *Tree Physiol.*, 3(1), 63–91, doi:10.1093/treephys/3.1.63, 1987.
- Rüdiger, J., et al., *Atmos. Meas. Tech.*, 11, 2441–2457, 2018.
- Sitch, S., et al., *Nature*, 448, 791–794, doi:10.1038/nature06059, 2007.
- Tai, A. P. K., et al., *Nat. Clim. Chang. Lett.*, 4, 817–821, doi:10.1038/NCLIMATE2317, 2014.
- Thomas, R. M., et al., *Atmos. Meas. Tech.*, 5, 243–257, doi:10.5194/amt-5-243-2012, 2012.
- Val Martin, M., et al., *Geophys. Res. Lett.*, 41, 2988–2996, doi:10.1002/2014GL059651.Received, 2014.
- Watai, T., et al., *J. Atmos. Ocean. Technol.*, 23, 700–710, 2006.
- Webb, E. K., *Q. J. R. Meteorol. Soc.*, 96, 67–90, 1970.
- Wild, O., *Atmos. Chem. Phys.*, 7, 2643–2660, 2007.
- Wilkinson, S., et al., *J. Exp. Bot.*, 63(2), 527–536, doi:10.1093/jxb/err317, 2011.
- Wittig, V. E., et al., *Plant, Cell Environ.*, 30, 1150–1162, doi:10.1111/j.1365-3040.2007.01717.x, 2007.

- Wittig, V. E., et al., *Glob. Chang. Biol.*, 15, 396–424, doi:10.1111/j.1365-2486.2008.01774.x, 2009.
- Wohlfahrt, G., et al., *Atmos. Environ.*, 43, 4570–4576, doi:10.1016/j.atmosenv.2009.06.031, 2009.
- Young, P. J., et al., *Atmos. Chem. Phys.*, (13), 2063–2090, doi:10.5194/acp-13-2063-2013, 2013.
- Young, P. J., et al., *Elem. Sci. Anthr.*, 6(10), doi:http://doi.org/10.1525/elementa.265, 2018.
- Zapletal, M., et al., *Environ. Pollut.*, 159, 1024–1034, doi:10.1016/j.envpol.2010.11.037, 2011.