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Reducing the discrepancies between the Aerodynamic Gradient Method and other micrometeorological approaches for measuring fumigant emissions☆

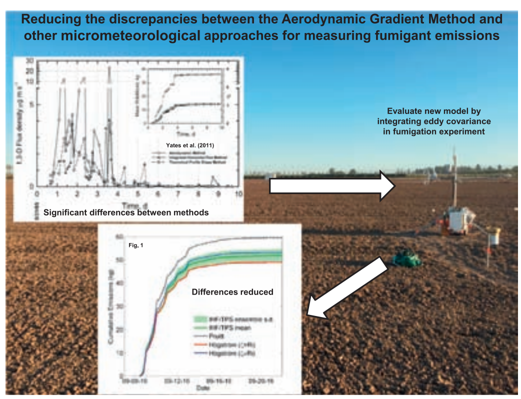
Ray G. Anderson [a,]*, Scott R. Yates [a], Daniel J. Ashworth [b,a], Dennise L. Jenkins [a], Qiaoping Zhang [a]

[a] US Department of Agriculture – Agricultural Research Service, US Salinity Laboratory, Riverside, CA 92507, USA
[b] University of California, Riverside, Department of Environmental Sciences, Riverside, CA 92521, USA

HIGHLIGHTS

- Accurate observations of fumigant emissions are needed for protecting environmental health.
- The Aerodynamic Gradient Method has consistently observed higher emissions than other methods.
- We correct the aerodynamic method using a new transport function and compare to eddy covariance.
- New function results in greater concurrence of aerodynamic observations with other approaches.

GRAPHICAL ABSTRACT



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ABSTRACT

Observations of fumigant and pesticide emissions are needed for multiple public health and environmental protection mandates. The aerodynamic gradient method (ADM) is commonly used to measure fumigant and pesticide emissions. However, the ADM may over estimate emissions compared to other micrometeorological and modeling approaches, which would increase uncertainty over the true flux estimate. Different studies with ADM have also used multiple differing transport functions that relate concentration gradients to emissions. Therefore, we tested different and more recent transport functions to try to correct the anticipated observed higher values with ADM using observations from two sites in California, USA. We evaluated different transport functions against eddy covariance observations and found that using the functions developed by Högström (1996) corrected the ADM values to be in line with other observational methods. For the Fresno experiment,

Abbreviations: 1,3-D, 1,3-dichloropropene; ADM, Aerodynamic Gradient Method; EC, Eddy Covariance Method; IHF, Integrated Horizontal Flux Method; MOST, Monin-Obukhov Similarity Theory; TPS, Theoretical Profile Shape Method.

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* Corresponding author at: USDA-ARS-USSL, George E. Brown Jr. Salinity Laboratory, 450 W. Big Springs Rd., Riverside, CA 92507-4617, USA.
E-mail addresses: ray.anderson@usda.gov (R.G. Anderson), daniel.ashworth@usda.gov (D.J. Ashworth), dennise.jenkins@usda.gov (D.L. Jenkins), qiaoping.zhang@usda.gov (Q. Zhang).

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cumulative emission masses from the ADM- Högström functions were within 7% of other approaches while the Pruitt function was >15% higher. Applying the Högström functions to a series of previous fumigation experiments in California saw reductions in the ADM observations of >25% for cumulative mass emissions. The results indicate that the Högström functions should be used for future ADM experiments in the absence of more robust transport factors for local meteorological conditions. The results also illustrate how previous ADM observations could be corrected to reduce uncertainty in flux emissions estimates.

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1. Introduction

Fumigants are commonly applied gaseous compounds that control soil borne pests. Over 44 million kilograms of fumigants were used in the United States in the late 2000s (Leahy, 2013), with 26 million kilograms used in California in 2015 (California Department of Pesticide Regulation, 2017). Ideally, fumigants stay in the soil to kill target pests and chemically degrade prior to being released via atmospheric volatilization. However, a major portion of fumigants are often released to the atmosphere prior to degradation (Wang et al., 2001). These emissions reduce the efficacy of application (Fennimore and Ajwa, 2011), accumulate in significant atmospheric concentrations near agricultural fields (van Wesenbeeck et al., 2011), may pose a significant human and environmental health threat near agricultural fields (Lee et al., 2002; O'Malley et al., 2005) and can have global environmental effects (Porter et al., 2009).

Numerous application techniques (Ajwa et al., 2002; Gan et al., 1998c), soil amendments (Ashworth and Yates, 2007; Graber et al., 2011; Gan et al., 1998a, 1998b), and soil covers or mulches (Gao and Trout, 2007; Ou et al., 2007; Papiernik et al., 2001) have been evaluated and implemented to reduce relative fumigant emissions. These techniques need robust estimates of their efficacy to assess their effectiveness in reducing fumigant emissions. This is particularly important as the amount of fumigant applications are often regulated by geographic region, time of application and/or mitigation measures to reduce emissions (Carpenter et al., 2001).

Multiple atmospheric techniques exist to measure fumigant emissions at field scale (for regulatory purposes) and for assessing the impact of mitigation measures to reduce fumigant emissions. These include the Aerodynamic Gradient Method (ADM) (Majewski et al., 1995; Parmele et al., 1972; van Wesenbeeck et al., 2007), Integrated Horizontal Flux (IHF) (Denmead et al., 1977; Sullivan and Ajwa, 2011), and Theoretical Profile Shape (TPS) (Wilson et al., 1982). In addition, Gaussian dispersion models such as CALPUFF (Scire et al., 2000), the AMS/EPA Regulatory Model (AERMOD) (US EPA, 2004) and the Industrial Source Complex-Short Term, Version 3 (ISCST3) (Atkinson et al., 1997) can be used to calculate emissions based on atmospheric concentrations; these models are widely used for regulatory purposes (Wang et al., 2006). While AERMOD has been adopted for US federal use, ISCST3 is still preferred in California due to its extensive historical use and basis for decision making by the California Department of Pesticide Regulation (Barry, 2008).

ADM is one of the oldest and most established meteorological approaches for measuring fumigant fluxes (Parmele et al., 1972; Pruitt et al., 1973). The approach assumes that Monin-Obukhov Similarity Theory (MOST) is observed in the surface atmospheric layer (Foken, 2006) in order to parameterize atmospheric transport functions to relate concentration gradients to fluxes. However, ADM has been observed to overestimate meteorological fluxes compared to other micrometeorological approaches, ADM has shown higher and more variable emissions with fumigants (Majewski et al., 1993; van Wesenbeeck et al., 2007; Yates et al., 2011, 2016), other pesticides (Majewski et al., 1990), and other trace gas fluxes such as NH_4, NO_2 and dry deposition velocities for O_3 (Hensen et al., 2009; Loubet et al., 2013; Stella et al., 2012; Wu et al., 2015). Previous studies have highlighted the sensitivity of ADM to Monin-Obukhov transport functions (Luhar et al., 2009; Stella et al., 2012). Multiple coefficients of the transport function have been proposed (Foken, 2006; Högström, 1996; Pruitt et al., 1973) with the largest divergence between these functions observed under stable nighttime conditions, suggesting this is an area where future refinement can have maximum benefit.

Early MOST transport functions were developed under relatively limited data sets that were focused on periods with generally good micrometeorological conditions and often collected over grass or other short vegetation. However, ADM observations of fumigant emissions should work reasonably well under all meteorological conditions to assess both cumulative emissions and peak concentrations and fluxes. In recent years, three-dimensional sonic anemometry and eddy covariance (EC) have become commonly used tools to assess atmospheric turbulence and mass and energy fluxes from the land surface, and sonic anemometry has been used to calculate transport functions for recent studies using ADM (Glenn et al., 2010; Loubet et al., 2013; Maas et al., 2013). EC can provide independent evaluations of alternate MOST transport functions. To investigate the discrepancies between ADM and other measurement and modeling approaches, we deployed an EC tower near an ADM tower in two field experiments in California, USA to test alternate MOST transport function parameterizations and discrepancies between ADM and other observational and modeling approaches.

2. Methods

2.1. Eddy covariance and aerodynamic method intercomparison experiments

We conducted two intercomparison experiments to test alternate MOST transport functions and reanalyzed data from five previous fumigant emission flux studies. Meteorological instrumentation and observation heights for both EC and ADM masts were identical for both experiments (SI: Table S1). The first experiment was performed at the University of California, Riverside (hereafter referred to as "Riverside") research farm (SI: Table S2) from 16 October 2014 to 1 December 2014. There was no fumigation component in this experiment, and we did not compare fluxes in experiment 1. The primary purpose experiment 1 was to assess ADM transport functions (Section 2.2) using EC and ADM tower mast data over surfaces with similar physical characteristics as fumigated fields and to provide preliminary data and insight for experiment 2, which was much more logistically demanding. This field is representative of the fetch limited conditions often found in coastal California, with a maximum fetch of 110 m and substantial surface changes (buildings and an orchard) to the north and east, respectively. The field was bare and plowed prior to represent typical fumigation conditions but was not irrigated prior to the experiment. EC and ADM data were analyzed for 30-min periods. More details about the Riverside field location can be found in (Luo et al., 2013).

The second intercomparison experiment occurred between 8 September 2016 and 22 September 2016 at the Western Research Center of Dow AgroSciences in Fresno, California. Details on this experiment are reported in detail in Ashworth et al. (2018) and are summarized here. The fumigated area was 128 m × 128 m (1.66 ha.) with an application rate of 1,3-dichloropronene (Telone II,[1] Dow AgroSciences, Indianapolis, Indiana, USA) of 133 kg ha^{-1} (212 kg total application). The field was plowed and irrigated prior to fumigation, and fumigation was by shank injection with a target depth of 0.46 m. The ADM mast was set up in the middle of the fumigated area while the EC instrumentation was set up about 20 m to the southeast (downwind) of the ADM mast. As discussed in (Ashworth et al., 2018a) and following previous experiments' protocols (Yates et al., 2011, 2016), wind speeds and temperature were observed at 0.2, 0.4, 0.6, and 1.5 m above ground with an additional anemometer placed at 2.4 m height. Charcoal tubes for fumigant sampling were changed about every 2 h initially with a 10-h sampling overnight due to logistical considerations. As the experiment progressed, sampling intervals were lengthened to ensure sufficient fumigant concentrations for analytical analysis. There were no substantial topographic obstructions around the fumigated area for over 200 m.

In addition to the Fresno fumigation experiment, we reanalyzed data from five previous fumigation experiments conducted near Buttonwillow, California in 2005 and 2007 that tested various emission control strategies at field scale (SI: Table S2). Details about these experiments are extensively reported by Yates et al. (2008, 2011, 2015, 2016, 2017) and are very briefly discussed here. These experiments tested the impact of different treatments on flux loss for fumigants, including deep injection, ammonium thiosulfate (ATS), organic amendments, irrigation post fumigation, and no treatment (standard practice). The sampling heights and equipment for the ADM method was the same as the 2016 Fresno experiment (Ashworth et al., 2018). Like the Fresno experiment, both the 2005 and 2007 Buttonwillow experiments occurred during September and had similar climatic conditions with extensive fetch. The only parameters that were changed for the Buttonwillow reanalysis were the MOST transport functions that were altered to test the potential to correct other ADM fumigant studies.

2.2. Data processing and intercomparison with eddy covariance

The ADM equation for fumigant flux emissions is as follows (Rosenberg et al., 1983):

$$F(t) = -k^2 \frac{\left(\overline{C_2(t)} - \overline{C_1(t)}\right)\left(\overline{u_2(t)} - \overline{u_1(t)}\right)}{\Phi_m(t)\Phi_c(t)\left(\ln\left({}^{z_2}/_{z_1}\right)\right)^2} \tag{1}$$

where F(t) is the flux at time t, k is the Von Karmin constant (k = 0.41), C_2 and C_1 are the atmospheric fumigant concentrations at heights 2 and 1, u_2 and u_1 wind speeds at heights 2 and 1, the overbars denote period means, $\Phi_m(t)$ and $\Phi_c(t)$ are the MOST transport functions for momentum and concentration (Pruitt et al., 1973), and z_2 and z_1 are the measurement heights. Atmospheric data processing for the ADM parameters, such as the gradient Richardson number and the wind speed ($\delta u/\delta z$) and temperature ($\delta T/\delta z$) gradients, followed previous studies (Yates et al., 2008, 2011, 2015, 2016, 2017).

We used selected data from the EC tower to independently calculate MOST transport functions in conjunction with ADM observations of $\delta u/\delta z$ and $\delta T/\delta z$ derived following Prueger and Kustas (2005):

$$\Phi_{mEC} = \frac{kz}{u_*}\frac{\delta u}{\delta z} \tag{2}$$

$$\Phi_{hEC} = -\frac{kz}{T_*}\frac{\delta T}{\delta z} \tag{3}$$

where Φ_{mEC} and Φ_{hEC} are the MOST transport functions from combining EC and gradient data, z is the EC measurement height, u_* is the friction velocity (m s^{-1}) from the EC tower, and T_* is the scaling temperature (K) also from the EC tower. The form of Eqs. (2) and (3) came from Eqs. 19 and 20 in Prueger and Kustas (2005). For this study, we assumed that $\Phi_h = \Phi_c$; this assumption is reasonable given the multiple past studies that have reported identical functions for Φ_h and Φ_c under both unstable and stable cases (Prueger and Kustas, 2005). To evaluate the impact of the choice of transport function on flux calculations, we compare the inverse product (IPTF) of the transport functions ($IPTF = {}^1/_{(\Phi_m\Phi_h)}$) from the EC and ADM approaches. We further evaluated the functions for periods with good micrometeorological conditions [i.e. u* > 0.1 m s^{-1}, which is a common threshold for EC data processing (Baker and Griffis, 2005)] and all conditions. The Riverside experiment had an additional constraint of winds between 225° and 360° to ensure a minimum fetch of 70 m.

High frequency sonic anemometer, fine-wire thermocouple, and infrared gas analyzer observations were recorded at 20 Hz for the EC tower. We processed high-frequency observations using the EddyPro Program (V6.2, Licor Inc.). Details about EddyPro can be found in (Fratini and Mauder, 2014). Default settings for flux calculations and corrections were used: double block coordinate rotation, block average detrending, covariance maximization for correcting time lags, turbulent density corrections (Webb et al., 1980), quality control tests and flags (Mauder and Foken, 2011), and footprint length estimation (Kljun et al., 2004).

2.3. Alternate MOST transport functions and post hoc correction of previous flux studies

Previous flux studies have used the MOST transport functions of (Pruitt et al., 1973) to relate the gradient Richardson number to Φ_m and Φ_c across a wide variety of sites and trace gases (Caro et al., 1977; Li et al., 2008; Majewski et al., 1991; Sandy et al., 2012; Yates et al., 2008) as follows:

$$\Phi_m = (1-16Ri)^{-1/3} \text{ when } Ri \le 0 \tag{4}$$

$$\Phi_h = [0.885(1-22Ri)]^{-2/5} \text{ when } Ri \le 0 \tag{5}$$

$$\Phi_m = (1+16Ri)^{1/3} \text{ when } Ri > 0 \tag{6}$$

$$\Phi_h = [0.885(1+34Ri)]^{2/5} \text{ when } Ri > 0 \tag{7}$$

Based on the subsequent review of (Foken, 2006) and the need for improved method performance during stable period, we have decided to test the alternative MOST transport functions of Högström (1996) as follows:

$$\Phi_m = (1-19\zeta)^{-1/34} \text{ when } \zeta \le 0 \tag{8}$$

$$\Phi_h = [0.95(1-11.6\zeta)]^{-1/2} \text{ when } \zeta \le 0 \tag{9}$$

$$\Phi_m = (1+5.3\zeta) \text{ when } \zeta > 0 \tag{10}$$

$$\Phi_h = (1+8\zeta) \text{ when } \zeta > 0 \tag{11}$$

where ζ is the Monin-Obukhov stability parameter. Unlike Ri, ζ cannot be calculated directly from ADM mast data. Prueger and Kustas (2005) suggest that $\zeta \cong Ri$ for moderately stable and unstable conditions. Högström (1996) proposes that $\zeta = 1.5Ri$ when $Ri < 0$ and $\zeta = Ri$ when $Ri \ge 0$, and we test all three MOST transport functions (Pruitt, Högström when $\zeta = Ri$ for unstable conditions, and Högström when $\zeta \ne Ri$ for unstable conditions) in the ADM method and compare to two other observational methods, Theoretical Profile Shape (TPS) and Integrated Horizontal Flux (IHF), and two atmospheric dispersion models, CALPUFF and ISCST3. To obtain the flux rate using atmospheric

[1] Mention of trade names or commercial products in this publication is solely for the purpose of providing specific information and does not imply recommendation or endorsement by the U.S. Department of Agriculture.

dispersion models, a back-calculation procedure was used. This involved comparing observed fumigant concentrations in the atmosphere surrounding a field to concentrations obtained using CALPUFF or ISCST3. An optimization procedure was used whereby the model's emission rate was adjusting to produce the best fit to the measured concentration values.

3. Results

3.1. Experiment 1 – Riverside

Meteorological data from the Riverside experiment showed largely typical fall/early winter patterns (SI: Fig. S1), though data from 31 October 6 November were lost due to a data card failure. Turbulent fluxes (SI: Fig. S1b) and air temperature (SI: Fig. S1c) showed decreasing trends into winter, but air temperature had a variable diurnal temperature range depending upon the strength of on-shore or off-shore wind flows. Maximum and minimum observed daily air temperatures were 33.5 °C and 6.2 °C, respectively. Atmospheric turbulence, as assessed u*, showed typical daytime/nighttime patterns (SI: Fig. S1a), with most nights having very low friction velocity ($u* < 0.05$ m s^{-1}). A few days had vigorous midday turbulence ($u* > 0.4$ m s^{-1}), but most days had maximum u* of ~ 0.2 m s^{-1}. Similarly, atmospheric stability, as assessed with the Monin-Obukhov stability parameter (ζ), remained moderately unstable to stable, with 56% of observed periods having ζ between −0.50 to 0.25 (SI: Fig. S1d).

With respect to the inverse product of the transport functions (IPTF), the Pruitt functions overestimated IPTF compared to the eddy covariance based IPTF for both all meteorological conditions and good meteorological conditions ($u* > 0.1$ m s^{-1} and winds between winds between 225° and 360°), with an overestimation >30%, as assessed by the slope of the regression, during good conditions (SI: Fig. S2 and Table S3). For all conditions, the Högström function with $\zeta = \mathrm{Ri}$ underestimated the EC IPTF but was non-significantly different ($p > 0.05$) during good conditions. This function also had the lowest error as assessed by the root mean squared error (RMSE). The Högström function with $\zeta \neq \mathrm{Ri}$ overestimated EC IPTF, but to a smaller degree than the Pruitt functions and had intermediate RMSE.

3.2. Experiment 2 – Fresno – transport functions

Turbulent fluxes and atmospheric turbulence were higher in Fresno than Riverside with a larger percentage of periods (21%) having $u* > 0.2$ (SI: Fig. S3). Air temperature had less diurnal variation than in Riverside. Finally, atmospheric stability was more moderate than Riverside, with 66% of observed periods having ζ between −0.50 to 0.25 (SI: Fig. S3d). Additional meteorological details are reported in Ashworth et al. (2018).

With respect to the IPTF, both Högström functions and the Pruitt function were relatively lower compared to the EC IPTF versus the Riverside experiment (SI: Fig. S4 and Table S3). There was much less variation in the functions' performance between good and all meteorological conditions. Despite being closer to unity, the slopes for the Pruitt function were still significantly higher than 1, indicating an overestimation of transport with Pruitt compared to the EC parameterization. The Högström function ($\zeta \neq \mathrm{Ri}$) was the only one with a slope that was not significantly different from 1. When assessed on longer flux averaging intervals that are characteristic for ADM fumigant observations, the Högström function ($\zeta = \mathrm{Ri}$) had significantly lower IPTF for most measurement intervals (SI: Fig. S5). Not unexpectedly, all functions had IPTF below 1 for the longer measurement periods that correspond to stable nighttime intervals, when sampling is limited due to logistical issues. At the typical nighttime interval (720 min), Högström ($\zeta = \mathrm{Ri}$ and $\zeta \neq \mathrm{Ri}$) IPTF was substantially lower (mean value of 0.35) than for Pruitt (mean value of 0.45).

3.3. Experiment 2 – Fresno – fumigant emissions

The choice of function shows clear impacts when we compare the ADM flux fumigant emissions using the various IPTF. With respect to cumulative emissions (Fig. 1), the Pruitt IPTF yields the highest emissions (59.5 kg) of any approach. Both Högström functions ($\zeta = \mathrm{Ri}$ and $\zeta \neq \mathrm{Ri}$) are both lower (53.1 and 48.4 kg, respectively) and within the standard deviation envelope of the IHF/TPS mean (51.8 kg). The flux emission rates, expressed both per second (Fig. 2) and measurement interval between changes in charcoal tubes (Fig. 3), also show the higher values with the Pruitt function with maximum fluxes of 18.2 μg m^{-2} s^{-1} and 9.9 kg measurement period^{-1}, respectively before rapidly decreasing after ~4 days.

We evaluated the impact of various meteorological controls on the differences between ADM approaches and the mean of the TPS and IHF methods (TPS_IHF). With respect to IPTF values, the largest differences between ADM and TPS_IHF occurred when IPTF was below 0.6 (Fig. 4), and the Högström IPTF had consistently smaller positive and larger negative differences than TPS_IHF. Substantial differences (>0.25 kg period^{-1}) also occurred at larger (>3) IPTF values. Unsurprisingly, the largest ADM-TPS_IHF differences occurred when there were larger fumigant gradients (Fig. 5), with most larger differences (>0.50 kg period^{-1}) occurring when the concentration gradients were below −75 μg/m^3 per m above the surface. Flux discrepancies were also strongly linked to positive temperature gradients (Fig. 6), with large differences occurring during inversions. All non-inversion periods (decreasing temperature with increasing height above the surface) had absolute differences <0.5 kg period^{-1}. Similar relationships were observed with atmospheric stability and flux differences (Fig. 7), with all larger differences (both positive and negative) occurring when the Richardson gradient number was positive. These variables (Figs. 4–7) are all strongly correlated with stable, nighttime periods where a stable atmosphere results in a shallow planetary boundary layer that can cause temperature inversions and strong trace gas concentration gradients between the surface and the lower atmosphere. However, wind gradients did not appear to have a significant relationship with ADM-TPS_IHF differences (Fig. S6), with the largest differences occurring at intermediate gradients rather than periods with little to no vertical

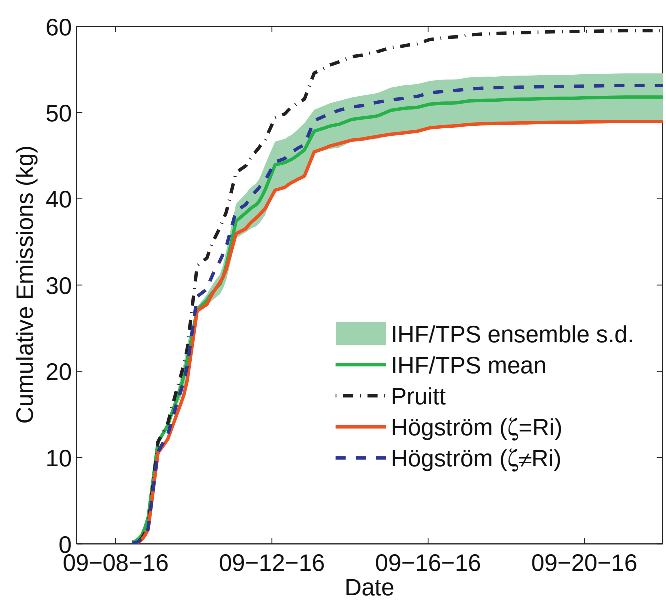

Fig. 1. Time series of cumulative emissions from the Fresno experiment showing the ADM parameterizations compared to the mean and standard deviations (s.d.) of the IHF and TPS methods.

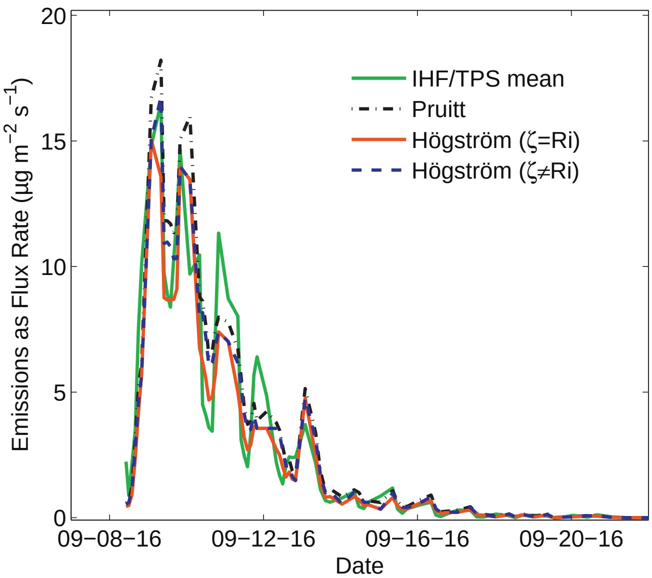

Fig. 2. Time series of emission rates from the Fresno experiment showing the ADM parameterizations compared to the mean and s.d. of the IHF and TPS methods.

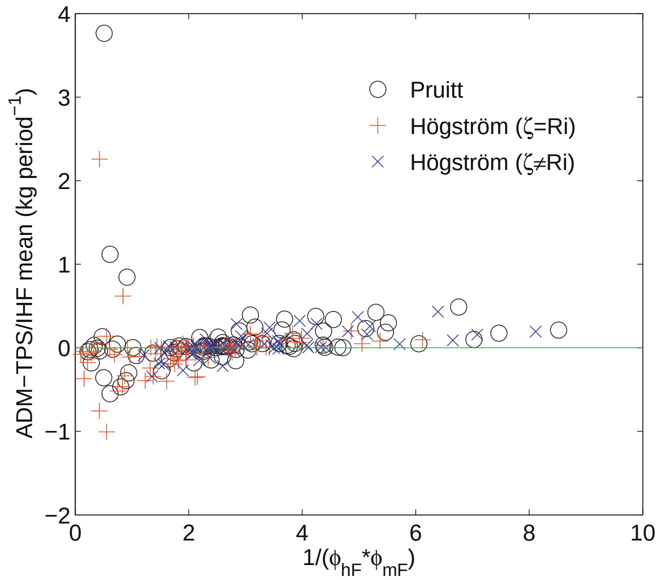

Fig. 4. Relationship between the different ADM approaches and TPS_IHF and mean IPTF for the Fresno site. Values for the Högström function ($\zeta \neq Ri$) are not plotted when $\zeta > 0$ as both Högström functions are identical at this point.

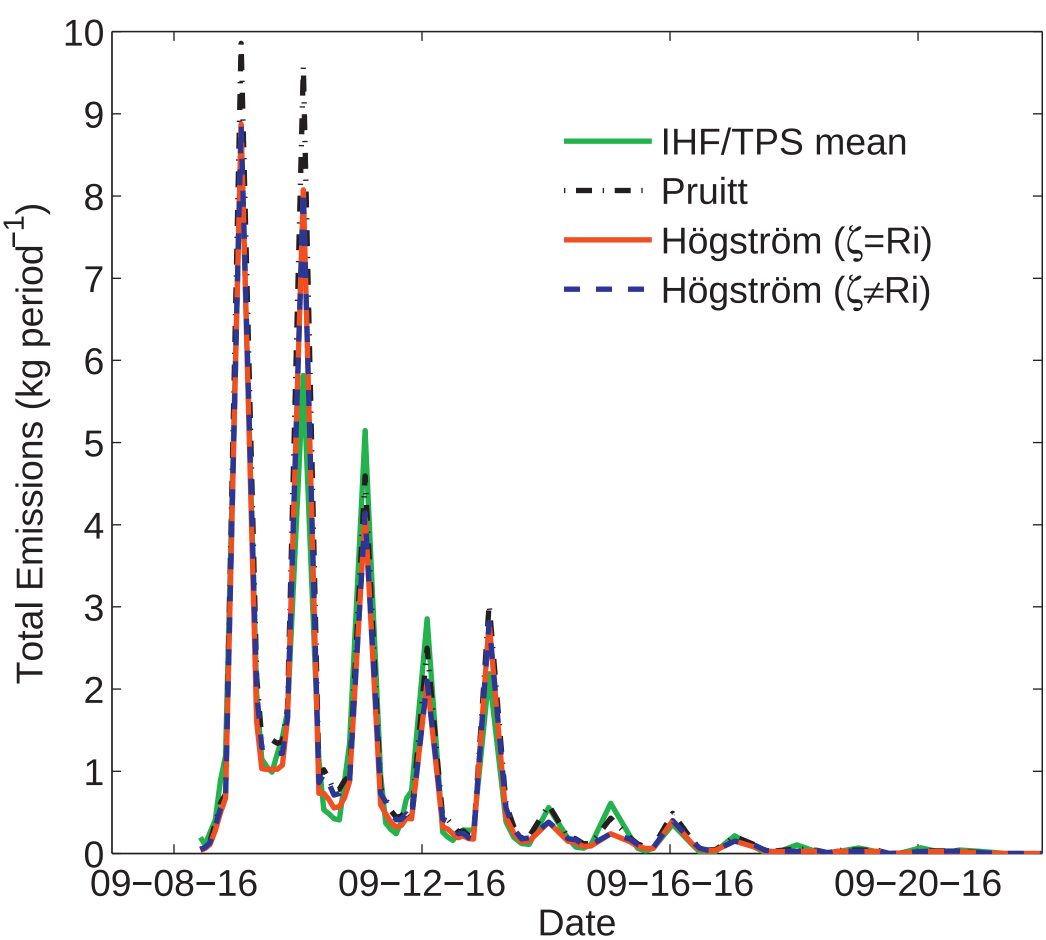

Fig. 3. Time series of emissions per measurement period from the Fresno experiment showing the ADM parameterizations compared to the mean and s.d. of the IHF and TPS methods. Periodic spikes are the longer nighttime observation periods.

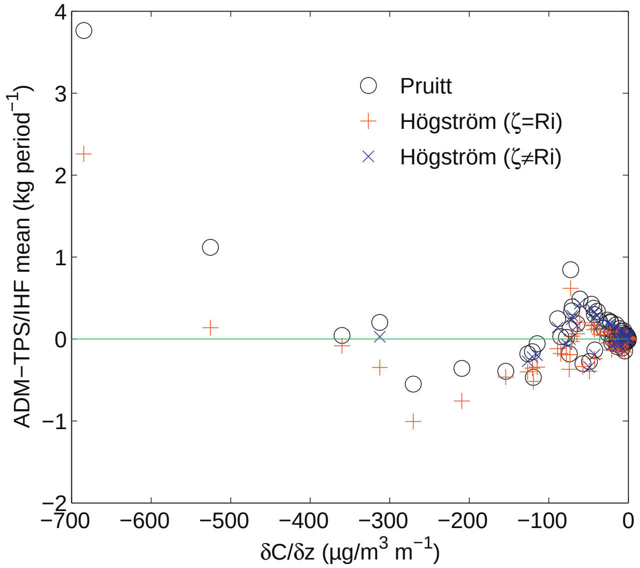

Fig. 5. Relationship between the different ADM approaches and TPS_IHF and mean fumigant concentration gradient, Fresno site.

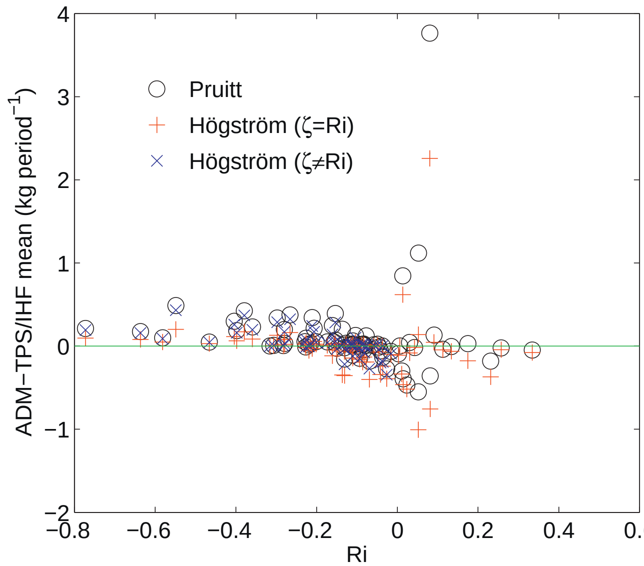

Fig. 7. Relationship between the difference between the different ADM approaches and TPS_IHF and stability as assessed by the Richardson Gradient number (Ri).

gradient in windspeed. This suggests that temperature induced stability may be more important than a lack of turbulence due to calm winds in causing differences between micrometeorological approaches for assessing fumigant emissions.

3.4. Post hoc correction of previous flux studies

Table 1 shows the results of the alternate MOST functions on flux calculations from the Buttonwillow experiments. The Högström function ($\zeta = \mathrm{Ri}$) had lowest peak fluxes and cumulative fluxes of the three ADM parameterizations, with the Pruitt function having the highest values and the Högström function ($\zeta \neq \mathrm{Ri}$) having intermediate values. However, unlike the Fresno experiment, the lowest ADM value (Högström $\zeta = \mathrm{Ri}$) exceeded the mean of the IHF and TPS methods. Relative differences were larger for cumulative fluxes compared to peak fluxes, with the ADM approaches often having cumulative differences of over 30%. The Högström function ($\zeta = \mathrm{Ri}$) resulted in the least differences with IHF and TPS and lowered the mean and standard deviation of emissions from the observational approaches (Table 1). While not calculated into the average, the modeling approaches (CALPUFF and ISCST3) had the least differences with the Högström function ($\zeta = \mathrm{Ri}$) as well. Slight differences in the relative performance of the varying MOST functions are observed between the approaches for the cis and trans isomers of 1,3-D due to fluctuating variations in concentration gradients covaried with meteorological differences.

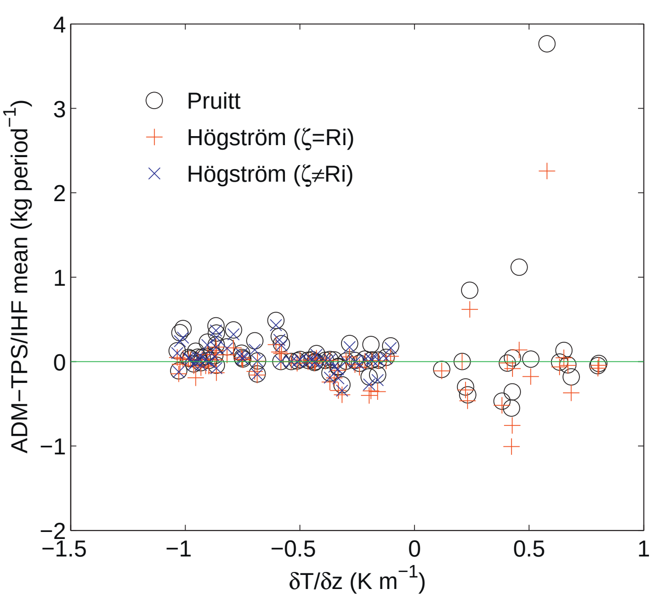

Fig. 6. Relationship between the different ADM approaches and TPS_IHF and mean temperature gradient, Fresno site.

4. Discussion and concluding remarks

4.1. Implications for interpreting previous ADM observations of fumigant and pesticide emissions

The results show that ADM using the Pruitt MOST formulae consistently overestimate flux emission rates and cumulative fluxes compared to ADM using other MOST functions, other meteorological approaches (IHF and TPS), and modeling approaches. While differences occurred at more unstable conditions, the bulk of the differences between ADM and IHF/TPS occurred with relatively few nighttime periods with atmospheric inversions. Four measurement intervals large differences (>0.5 kg period^{-1}) contributed 5.2 kg (68%) of the total 7.7 kg discrepancy between ADM using Pruitt and the IHF/TPS mean. These differences occur during times when observational conditions are challenging for most meteorological approaches. While some researchers recommend excluding fluxes observed during these periods (Van Den Berg et al., 1999), the fluxes during these periods are sufficiently large that the data cannot be excluded or interpolated in a meaningful way without creating major impacts on the data. The largest differences in IPTF values between the Pruitt and Högström functions occur during stable atmospheric periods (Ri and ζ both >0), and future ADM development work should focus on improving MOST transport functions during nighttime periods with moderate stability values characteristic of longer measurement periods at night.

Regulatory approaches that incorporate the ADM method may be more conservative than necessary for public and environmental health goals. Studies where the underlying field data are still available should be reanalyzed with alternate functions to assess the impact on the flux calculations. Along with reanalyzing previous data, future studies

Table 1
Cumulative flux fumigant emissions from the Buttonwillow experiments. Note averages are only for observational approaches, thus no average is calculated for the standard practice experiment (Yates et al., 2015).

Method	1,3-D (cis)		1,3-D (trans)		All
	Peak emissions (μg m^{-2} s^{-1})	Percent of applied mass lost	Peak emissions (μg m^{-2} s^{-1})	Percent of applied mass lost	Percent of applied mass lost
Standard practice (Yates et al., 2015)					
ADM (Pruitt)	17.6	42.4	9.3	28.3	35.4
ADM (ζ = Ri Högström)	13.5	30.6	7.9	20.7	25.7
ADM ($\zeta \neq$ Ri Högström)	16.6	31.8	8.6	23.4	29.2
CALPUFF	18.0	31.8	11.9	22.6	27.2
ISCST3	7.2	19.0	4.4	13.5	16.2
Irrigation treatment (Yates et al., 2008)					
ADM (Pruitt)	30.9	17.5	28.9	13.1	15.3
ADM (ζ = Ri Högström)	25.3	12.3	23.7	9.4	10.9
ADM ($\zeta \neq$ Ri Högström)	27.2	13.3	25.5	10.3	11.8
IHF	12.8	11.5	9.8	8.1	9.8
TPS	9.7	12.1	8.1	8.6	10.3
CALPUFF	5.6	12.2	3.9	8.6	10.0
ISCST3	4.3	7.8	2.8	5.4	6.6
Avg. ± std. (Pruitt)	12.7 ± 10.7	12.2 ± 3.5	10.7 ± 10.6	8.8 ± 2.8	10.4 ± 3.1
Avg. ± std. (ζ = Ri Högström)	11.5 ± 8.4	11.2 ± 1.9	9.7 ± 8.4	8 ± 1.6	9.5 ± 1.7
Avg. ± std. ($\zeta \neq$ Ri Högström)	11.9 ± 9.2	11.4 ± 2.1	10 ± 9.1	8.2 ± 1.8	9.7 ± 1.9
Deep Injection treatment (Yates et al., 2016)					
ADM (Pruitt)	18.5	31.6	13.8	21.8	26.7
ADM (ζ = Ri Högström)	13.6	21.1	10.1	14.8	18.0
ADM ($\zeta \neq$ Ri Högström)	17.1	23.8	12.8	16.7	20.2
IHF	8.6	22.1	6.3	15.4	18.8
TPS	8.8	17.1	8.7	13.1	15.1
CALPUFF	19.0	30.3	14.4	21.9	26.1
ISCST3	8.2	18.3	5.0	13.4	15.8
Avg. ± std. (Pruitt)	12.6 ± 5.6	23.9 ± 6.7	9.6 ± 4.3	17.1 ± 4.4	20.5 ± 5.6
Avg. ± std. (ζ = Ri Högström)	11.6 ± 4.7	21.8 ± 5.2	8.9 ± 3.7	15.7 ± 3.6	18.8 ± 4.4
Avg. ± std. ($\zeta \neq$ Ri Högström)	12.3 ± 5.3	22.3 ± 5.2	9.4 ± 4	15.4 ± 5.4	19.2 ± 4.4
ATS spray treatment (Yates et al., 2017)					
ADM (Pruitt)	9.6	28.5	9.9	21.6	25.0
ADM (ζ = Ri Högström)	7.0	18.0	7.2	13.9	15.9
ADM ($\zeta \neq$ Ri Högström)	8.9	22.4	9.1	17.3	19.9
IHF	4.8	14.5	3.0	10.5	12.5
TPS	4.8	13.9	3.3	11.2	12.5
CALPUFF	10.9	29.0	9.1	23.6	26.3
ISCST3	9.2	17.6	6.4	14.2	15.9
Avg. ± std. (Pruitt)	7.9 ± 2.9	20.7 ± 7.5	6.3 ± 3.2	16.2 ± 6	18.4 ± 6.7
Avg. ± std. (ζ = Ri Högström)	7.3 ± 2.7	18.6 ± 6.1	5.8 ± 2.6	14.7 ± 5.3	16.6 ± 5.7
Avg. ± std. ($\zeta \neq$ Ri Högström)	7.7 ± 2.8	19.5 ± 6.3	6.2 ± 3	15.4 ± 5.4	17.4 ± 5.8
Organic treatment (Yates et al., 2011)					
ADM (Pruitt)	10.0	9.7	13.2	6.6	8.2
ADM (ζ = Ri Högström)	7.3	5.7	9.6	4.3	5.0
ADM ($\zeta \neq$ Ri Högström)	9.3	6.7	12.4	5.0	5.9
IHF	3.1	3.9	2.3	2.9	3.4
TPS	2.6	3.8	1.9	2.8	3.3
CALPUFF	4.1	4.7	2.8	3.2	4.0
ISCST3	2.9	3.0	1.4	1.8	2.5
Avg. ± std. (Pruitt)	4.5 ± 3.1	5 ± 2.7	4.3 ± 5	3.4 ± 1.9	5.0 ± 2.8
Avg. ± std. (ζ = Ri Högström)	4 ± 1.9	4.2 ± 1.0	3.6 ± 3.4	3 ± 0.9	3.6 ± 0.9
Avg. ± std. ($\zeta \neq$ Ri Högström)	4.4 ± 2.8	4.4 ± 1.4	4.1 ± 4.6	3.1 ± 1.2	4.2 ± 1.5

could incorporate more robust MOST functions if they use the ADM method. Higher temporal resolution sampling should also be considered. While increased sample frequency (particularly at night) creates additional logistical challenges, it likely reduces the impact of any one measurement interval on cumulative emissions. Increased sampling frequency also avoids other averaging errors that can degrade flux observations (Majewski, 1996). Other micrometeorological approaches (Majewski et al., 1993; Pattey et al., 1995; Wu et al., 2015) can complement ADM, potentially increasing the robustness of flux observation estimates. Further evaluation work is needed, particularly with well-designed chamber systems (Gao and Yates, 1998).

4.2. Concluding remarks

In this study we evaluated alternate parameterizations of the Monin-Obukhov stability functions against eddy covariance observations to test the observed higher fumigant flux estimates using the Aerodynamic Gradient Method. The results showed that using functions that were less sensitive during stable nighttime periods resulted in flux calculations that had better agreement with other observational and modeling approaches. We could apply the revised functions to correct flux calculations from previous fumigation experiments, thus demonstrating the potential to reduce flux emission uncertainty with historic data. However, the largest corrections were to only a small number of nighttime periods with longer measurement intervals. Therefore, sampling intervals should be kept as short as possible given the analytical precision of laboratory instrumentation for the chemical of concern to try to minimize the impact of atmospheric parameterization during any one measurement interval.

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Appendix A. Supplementary data

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