

## Data filtering for inverse dispersion emission calculations



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### ABSTRACT

Inverse dispersion techniques are used to infer the emission rate of gas sources from concentration measurements and dispersion model calculations. Criteria for the selection of measurement intervals having wind conditions conducive to technique accuracy are examined on the basis of a short range tracer experiment. By introducing a supplementary condition that the measured vertical temperature gradient be quantitatively compatible with Monin–Obukhov similarity theory, it was possible to use a less stringent threshold for the friction velocity than has previously been used ( $u_* \geq 0.05 \text{ m s}^{-1}$  instead of  $\geq 0.15 \text{ m s}^{-1}$ ). Under the new criteria a larger proportion of measurement intervals are retained (76% versus 49%), while the ratio of inferred to actual emission rate  $Q_{IS}/Q$  exhibits negligible bias (average  $Q_{IS}/Q = 1.00$ ) and an acceptably small level of random error (interval-to-interval standard deviation  $\sigma_{Q/Q} = 0.25$ ).

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### 1. Introduction

“Inverse dispersion” refers to the practice of inferring the atmospheric emission rate ( $Q$ ) of localized gas sources from the excess concentration ( $C$ ) they cause, by modelling the  $C$ – $Q$  relationship under the existing meteorological state. The technique has proven particularly successful for calculating emissions from discrete ground level sources using nearby concentration measurements (e.g., Ferrara et al., 2014). This micrometeorological scale problem requires only an upwind and downwind gas concentration, with substantial freedom to choose convenient measurement locations. The technique does have the disadvantage that, in its most practical form, it entails idealizations that may compromise accuracy in some circumstances. Chief among these is the assumption that a dispersion model predicated on horizontally-homogeneous winds will suffice for the inversion, obviating what is (otherwise) a burdensome computation. While strictly unobstructed wind fields are the exception, fortunately a number of studies in disturbed winds have indicated that the technique can be quite robust (e.g., Wilson et al., 2010).

Different types of dispersion models can be used for inverse calculations. A common implementation combines a Lagrangian

stochastic (LS) model with a Monin–Obukhov (MO) similarity theory description of the wind (Wilson et al., 2012), and to that end specialized “MO–LS” software has evolved<sup>1</sup>. In an agricultural context MO–LS has been used to calculate emissions from barns (Harper et al., 2010), fields (Sanz et al., 2010), cattle feedlots (Todd et al., 2011), waste storage ponds (Flesch et al., 2013), grazing cattle (McGinn et al., 2011), and many other variations of source and environment.

An MO–LS emission calculation presumes MO accurately describes the vertical profiles of the average wind and turbulent statistics. The theory posits that these properties are characterized by the friction velocity  $u_*$  and Obukhov length  $L$  (in conjunction with the surface roughness length  $z_0$ ). Light winds and extreme atmospheric stratification, associated with small magnitudes of  $u_*$  and  $L$ , limit the applicability of MO. Several studies show a deterioration in the accuracy of MO–LS emission calculations as  $u_*$  and  $|L|$  decrease (e.g., Flesch et al., 2004; Gao et al., 2009), which has led to the introduction of filtering criteria to remove periods when  $u_*$  and  $|L|$  fall below threshold values. For example, Flesch et al. (2005) used thresholds of  $u_{*thres} = 0.15 \text{ m s}^{-1}$  and  $|L|_{thres} = 10 \text{ m}$ , McBain

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<sup>1</sup> The MO–LS designation can include backward Lagrangian stochastic (bLS) calculations for area sources, or a corresponding forward calculation (fLS) for point sources.

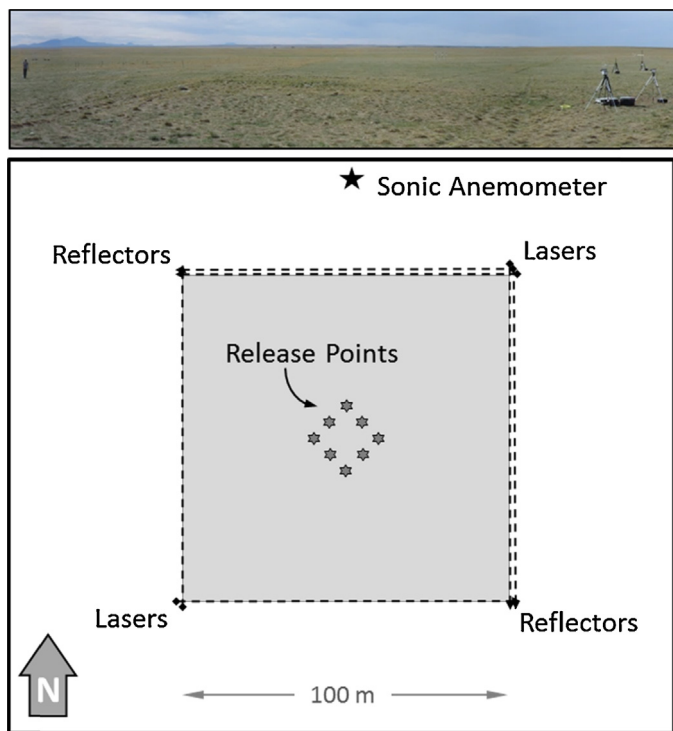


Fig. 1. Field site (top) and equipment and gas release configuration (bottom).

and Desjardins (2005) suggested  $u_{*thres} = 0.19 \text{ m s}^{-1}$ , while Laubach et al. (2008) used  $0.12 \text{ m s}^{-1}$ .

A consequence of filtering is the loss of potentially valuable data. With  $u_{*thres} = 0.15 \text{ m s}^{-1}$  our experience has been of data loss rates of 40 to 50%, and sometimes more than 75% in long term rural studies (clearly this loss depends on the regional climate and the weather encountered during a campaign). The loss of low wind speed data hinders efforts to characterize sources having an emission rate that correlates with wind speed (e.g., ammonia from waste ponds), since emissions during light winds are unresolved. And because filtering preferentially removes light wind nighttime data, a daytime-biased dataset is created. This complicates the calculation of average emissions from diurnally varying sources, such as animals or industrial sites having a daily activity pattern.

The focus of this study is the filtering criteria used in MO–LS calculations. Our motivation comes from the perspective of animal studies, where the loss of nighttime data is a significant problem for characterizing emissions. We will examine the potential of  $u_*$ ,  $L$ , wind speed and air temperature as filtering criteria, and consider how these affect MO–LS accuracy and the rate of data retention (particularly at night). A tracer release study, designed to mimic the configuration of a small herd of cattle, provides a large dataset for this evaluation.

## 2. Methods

### 2.1. Tracer release experiment

A tracer release experiment was conducted in September 2012 at Onefour, Alberta, Canada ( $49^{\circ}06'N$ ;  $110^{\circ}30'W$ ; elevation 925 m). This semi-arid grassland site was selected because of its extensive short grass terrain and the absence of nearby gas sources (Fig. 1). The study was designed to mimic the configuration of a small herd of cattle. Eight release points were clustered near the centre of an imaginary  $100 \times 100 \text{ m}$  square paddock (Fig. 1), at a height of 0.5 m above ground. Methane was released at a known rate using

a mass flow controller (GFC47, AALBORG, Orangeburg, NY, USA) and verified by weighing of gas cylinders. The gas was directed to a manifold and distributed to the release points. The combination of a large diameter manifold and long and equal length tubing was assumed to give equal emission rates from each release point. The total release rate was either  $0.46$  or  $0.92 \text{ kg CH}_4 \text{ h}^{-1}$  (equivalent to 50 to 150 cattle: a large release rate chosen to minimize concentration measurement errors). Releases took place intermittently from September 3 to 10, with a total of 215 15-min release periods. Of these, 144 occurred during the night (sunset to sunrise).

### 2.2. Concentration and wind measurements

Open-path  $\text{CH}_4$  lasers (GasFinder 2, Boreal Laser Inc., Edmonton, Canada) measured the average gas concentration along the four sides of the simulated paddock. There were four stand-alone lasers and one laser on a pan-tilt head that scanned between two retro-reflectors (DSM; PTU D300, FLIR Motion Control Systems, Burlingame, CA, USA). This setup gave measurement duplication on two of the four paddock sides. The average path heights of the four laser lines varied from 1.5 to 2.15 m due to gentle terrain undulations.

Laser calibration was completed after the study. Recently, GasFinder lasers were found to have a previously unaccounted for temperature and pressure dependence (discussed by Laubach et al., 2013). This has been addressed by Boreal Laser Inc. in a new calibration procedure that gives temperature and pressure correction factors, and our lasers were re-calibrated after the study and these corrections were applied retroactively. Laser calibrations were also adjusted to match on-site measurements from a gas chromatograph (GC). Air samples were collected during a single 15-min interval between release periods, and laser-specific radiometric correction factors were applied to force agreement between the lasers and the GC measured concentration ( $1.77 \text{ ppm}_v$ ).

Laser measurements were processed to give 15-min average concentrations. Concentrations were converted from the reported  $\text{ppm}_v$  to  $\text{g m}^{-3}$  using measured air pressure and temperature. Laser observations were not used if a 15-min period did not include more than 25% good data (i.e., light levels  $> 2000$  units and  $R^2 > 96$ : quality parameters reported by the laser).

A 3-D sonic anemometer (CSAT-3, Campbell Scientific, Logan, UT, USA) provided the wind information for our calculations: friction velocity  $u_*$ , Obukhov length  $L$ , surface roughness length  $z_0$ , and wind direction  $\beta$  (calculated as described in Flesch et al., 2004). The anemometer was positioned just north of the release site at a height of 2.0 m above ground. Wind statistics were calculated for 15-min intervals matching the  $\text{CH}_4$  concentration record.

### 2.3. MO–LS emission calculations

Emission rates were calculated using the freely available WindTrax software. The software combines the MO–LS model described by Flesch et al. (2004) with mapping capabilities. Dispersion from each release point was simulated with a forward LS model using 10,000 trajectories. Identical emission rates were assigned to the eight release points in the calculation. From each set of 15-min laser concentrations, WindTrax calculated the total emission rate  $Q_{15}$  ( $\text{kg CH}_4 \text{ h}^{-1}$ ) and the corresponding background concentration  $C_b$ . This problem is mathematically over-determined (e.g., six concentrations used to solve for two unknowns) and a best-fit procedure was used in the WindTrax calculation.

In the following analysis 19 out of the 214 release periods (15-min each) are not used. Fifteen had a calculated  $z_0$  greater than the source height of 0.5 m; one period violated the Cauchy–Schwarz inequality (error in calculated wind statistics that can occur in light winds); in one extremely stable nighttime period the calculated

gas plumes did not reach the height of the laser paths; and in two periods a critical downwind laser observation was missing.

#### 2.4. Temperature and wind speed profile

Air temperature ( $T$ ) and wind speed ( $U$ ) were measured with sonic anemometers (CSAT-3) at 1.0 and 2.9 m above ground. These instruments were in addition to our primary anemometer placed at a height of 2 m. Sonic anemometers measure acoustic temperature, which is related to the actual air temperature as (Kaimal and Gaynor, 1991):

$$T_{\text{son}} = T \left( 1 + \frac{0.32e}{p} \right), \quad (1)$$

where  $e$  is the water vapor pressure and  $p$  is the absolute air pressure<sup>2</sup>. In this study our interest is the height gradient in  $T$ . Differentiating Eq. (1) gives:

$$\frac{\partial T_{\text{son}}}{\partial z} = \frac{\partial T}{\partial z} \left( 1 + \frac{0.32e}{p} \right) + 0.32T \left( \frac{1}{p} \frac{\partial e}{\partial z} - \frac{e}{p^2} \frac{\partial p}{\partial z} \right). \quad (2)$$

For realistic values of  $p$  and  $e$  at our site (e.g., gradients consistent with latent heat fluxes  $< 50 \text{ W m}^{-2}$ ) the difference between the height gradients of  $T_{\text{son}}$  and  $T$  is  $< 0.05 \text{ K m}^{-1}$ . This is much smaller than the magnitude of the temperature gradients we will be concerned with, and we thus assume a height difference  $\Delta T_{\text{son}}$  equals the difference in actual temperature  $\Delta T$ .

It is known that  $T_{\text{son}}$  can have an instrument specific bias due to uncertainties in the sonic path length. In their study of CSAT-3 anemometers, Burns et al. (2012) used an offset to correct for this. For moderate winds and neutral stratification we expect  $T_{\text{son}}$  to be nearly identical between our anemometers. In these conditions two anemometers were in very good agreement. The third was systematically cooler, and an offset of 0.25 K was applied.

### 3. Results

In the analysis that follows, each emission rate calculation  $Q_{\text{LS}}$  is expressed as a ratio of the actual emission rate  $Q$ , with  $Q_{\text{LS}}/Q = 1$  being a perfectly accurate calculation. A good filtering procedure is judged to be one that results in a set of MO–LS observations having an average  $Q_{\text{LS}}/Q$  near 1 (good overall accuracy), a small standard deviation  $\sigma_{Q/Q}$  (good period-to-period fidelity), and minimal data loss. We assume the tracer release rate, gas concentrations, wind statistics, and the location of lasers and sources are measured accurately, and that the MO–LS model describes turbulent dispersion during winds that are consistent with MO theory. This assumption of “no measurement error” is clearly false, but we believe these errors are small enough so that errors associated with an inaccurate MO description of the wind will be revealed over our large dataset.

For all 195 release periods (unfiltered) the average  $Q_{\text{LS}}/Q$  is 1.27 and  $\sigma_{Q/Q} = 2.00$ ; meaning calculated emissions are 27% higher than the actual rate on average, and there is poor period-to-period fidelity. This is a poorer result than reported by Gao et al. (2009) for their unfiltered data ( $Q_{\text{LS}}/Q = 1.04$ ,  $\sigma_{Q/Q} = 0.55$ ), but almost identical to the unfiltered results reported by Flesch et al. (2004);  $Q_{\text{LS}}/Q = 1.27$ ,  $\sigma_{Q/Q} = 2.07$ . Plotting  $Q_{\text{LS}}/Q$  with  $u_*$  and  $1/L$  (Fig. 2) shows the expected trend toward less accurate inversions in light winds and stable conditions—typical nighttime conditions. Also notice that our dataset includes more stable than unstable periods, reflecting our interest in nighttime conditions,

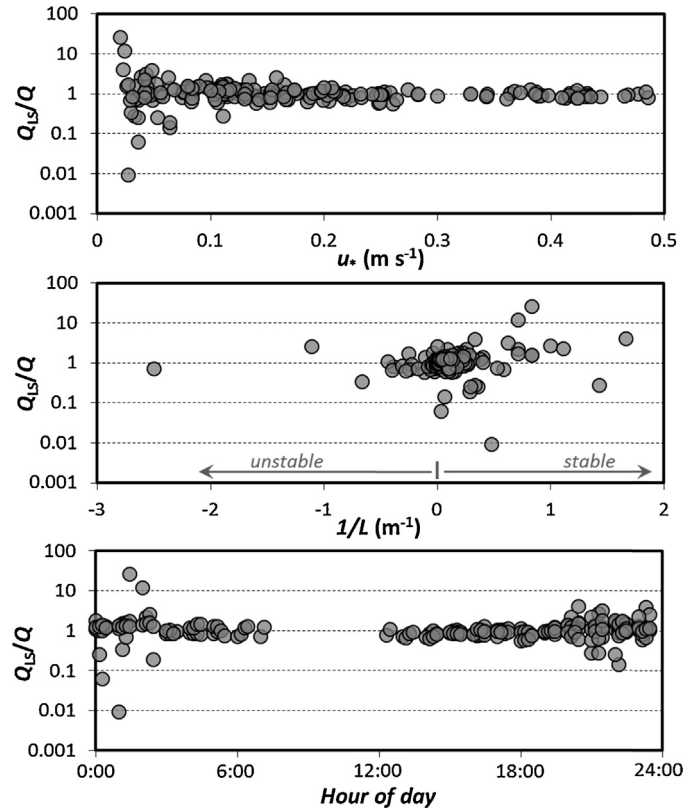


Fig. 2. Calculated emission rate ( $Q_{\text{LS}}/Q$ ) plotted with friction velocity ( $u_*$ ), the reciprocal of the Obukhov length ( $1/L$ ), and hour of day (12:00 is local noon). A  $Q_{\text{LS}}/Q = 1$  is perfect accuracy (notice the logarithmic scale).

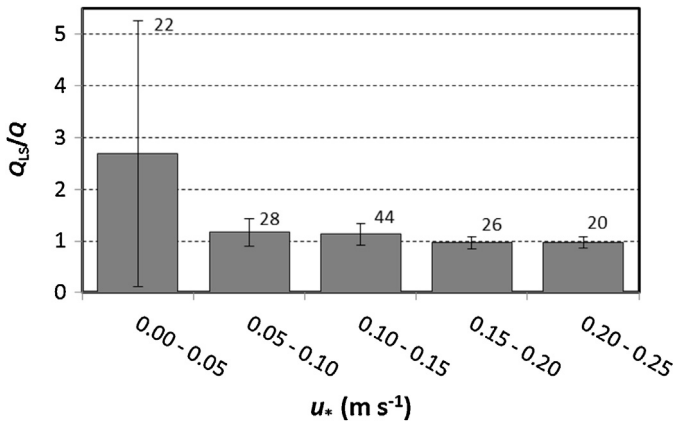
which limits our ability to assess filtering during unstable daytime conditions.

#### 3.1. Filtering with $u_*$

Flesch et al. (2004) reported that  $u_* \leq 0.15 \text{ m s}^{-1}$  was the single best diagnostic for identifying inaccurate  $Q_{\text{LS}}$  in their tracer experiment. In the present context, using a  $u_{*\text{thres}} = 0.15 \text{ m s}^{-1}$  filter leads to a substantial improvement in accuracy, with the average  $Q_{\text{LS}}/Q$  falling from 1.27 to 0.96, and  $\sigma_{Q/Q}$  dropping from 2.00 to 0.19. This confirms what has been reported in many studies, that MO–LS is accurate in moderate to strong wind conditions. However, the cost of filtering is the loss of 50% of our data, and almost 75% of our nighttime data. Can  $u_{*\text{thres}}$  be reduced from  $0.15 \text{ m s}^{-1}$  in order to increase data retention while still maintaining  $Q_{\text{LS}}$  accuracy?

Fig. 3 shows  $Q_{\text{LS}}$  accuracy for different  $u_*$  ranges. When  $u_* < 0.05 \text{ m s}^{-1}$  the quality of  $Q_{\text{LS}}$  is very poor (average  $Q_{\text{LS}}/Q = 2.69$ ,  $\sigma_{Q/Q} = 5.15$ ) and most observations are in error by more than 50%. We conclude there is little potential for “recovering” accurate  $Q_{\text{LS}}$  at these low values of  $u_*$ . The situation improves for  $u_*$  between 0.05 and  $0.15 \text{ m s}^{-1}$  (average  $Q_{\text{LS}}/Q = 1.14$ ,  $\sigma_{Q/Q} = 0.47$ ). This is encouraging, as this  $u_*$  group accounts for nearly 40% of our data. Because the overall accuracy of this low  $u_*$  group is not particularly bad, one might simply include these data in an emission analysis. However, more than 20% of the observations in this group are outliers having  $0.5 > Q_{\text{LS}}/Q > 1.5$ . Outliers can be a problem with a small population of observations, e.g., a dataset divided in order to calculate hourly emission rates. Below we describe our attempts to discriminate the accurate and inaccurate  $Q_{\text{LS}}$  in this group having  $0.05 \leq u_* < 0.15 \text{ m s}^{-1}$ .

<sup>2</sup> In applying Eq. (1) to average temperatures (via Reynolds’s averaging) we assume the fluctuating term  $eT$  is negligible, and that pressure fluctuations are a very small fraction of the average pressure and can be ignored.



**Fig. 3.** Accuracy of the calculated emission rate ( $Q_{LS}/Q$ ) grouped by friction velocity ( $u_*$ ). The height of the “error bars” represents  $\sigma_{Q/Q}$  for each group. Values above each column give the number of observations in the group.

### 3.2. Stability as a filtering criterion

Some studies have used atmospheric stability as a filtering criterion for MO–LS (Flesch et al., 2005; McGinn et al., 2011); rejecting observations having an Obukhov length  $L$  below a threshold  $|L|_{\text{thres}}$ . Lowering  $|L|_{\text{thres}}$  results in the inclusion of periods with increasingly stable or unstable stratification. In Fig. 4 we see the result of imposing different  $|L|_{\text{thres}}$  values on the target  $0.05 \leq u_* < 0.15 \text{ m s}^{-1}$  group. No substantial improvement in the accuracy of the inversion  $Q_{LS}$  is observed using a stability filter.

### 3.3. Temperature gradient as a filtering criterion

A comparison between the measured and MO predicted temperature gradient may indicate an inaccurate MO description of the atmosphere, and inaccurate MO–LS calculations. The difference in  $T$  between heights  $z_1$  and  $z_2$  is given by the MO formula<sup>3</sup> (Garratt, 1992):

$$\Delta T_{MO} = \frac{u_*^2 T_{\text{ave}}}{k_v^2 g L} \left[ \ln \left( \frac{z_2}{z_1} \right) - \Psi_H(z_2) + \Psi_H(z_1) \right] \quad (3)$$

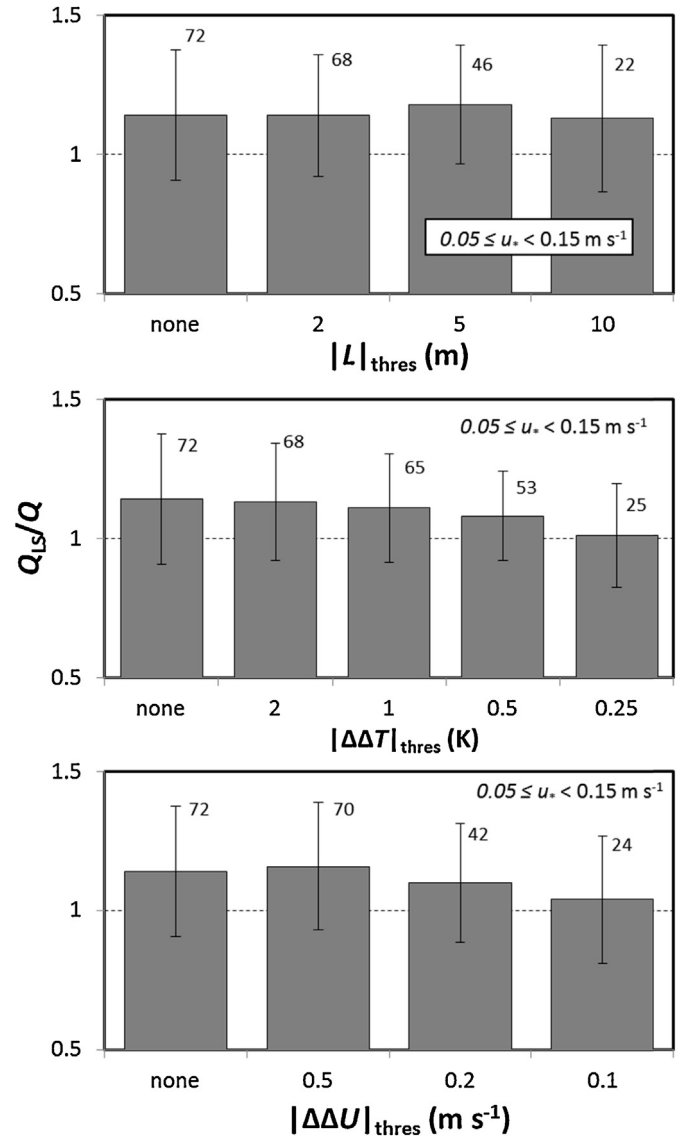
where  $T_{\text{ave}}$  is the average air temperature (K, here taken at  $z = 2 \text{ m}$ ),  $k_v$  is von Karman’s constant (0.4),  $g$  is the gravitational acceleration ( $9.81 \text{ m s}^{-2}$ ), and  $\Psi_H$  is a stability correction function:

$$\begin{cases} \Psi_H = 2 \ln[(1 + \sqrt{1 - 16z/L})/2] & , \text{ (for } L < 0) \\ = -5z/L & , \text{ (for } L > 0) \end{cases} \quad (4)$$

For each measurement period we calculate the difference between the measured and MO-calculated gradient ( $\Delta T - \Delta T_{MO}$ ) evaluated at  $z = 1$  and  $2.9 \text{ m}$ . Hereafter, we refer to  $(\Delta T - \Delta T_{MO})$  as  $\Delta \Delta T$ . A filter is created to reject periods when  $|\Delta \Delta T|$  is greater than a threshold  $|\Delta \Delta T|_{\text{thres}}$ .

Fig. 4 shows the impact of the  $\Delta \Delta T$  filter on the target  $0.05 \leq u_* < 0.15 \text{ m s}^{-1}$  group. As  $|\Delta \Delta T|_{\text{thres}}$  decreases from 2.0 to 0.25 K we see improvement in the average  $Q_{LS}/Q$  from 1.13 to 1.01, with a slight improvement in  $\sigma_{Q/Q}$  from 0.24 to 0.18. The choice  $|\Delta \Delta T|_{\text{thres}} = 0.5 \text{ K}$  eliminates the worst  $Q_{LS}$  outliers and retains 74% of this  $u_*$  group.

<sup>3</sup> This formula is for potential temperature. Over a small height difference it can be applied to actual temperature.



**Fig. 4.** Accuracy of the calculated emission rate ( $Q_{LS}/Q$ ) as affected by filtering criteria based on the Obukhov length  $L$ , temperature errors  $\Delta \Delta T$ , and wind speed errors  $\Delta \Delta U$ . The height of the “error bars” represents  $\sigma_{Q/Q}$  for each group, and the values above each column give the number of observations. These results are for periods having  $0.05 \leq u_* < 0.15 \text{ m s}^{-1}$ .

### 3.4. Wind speed as a filtering criterion

Along the same lines as the temperature gradient, a comparison between predicted and measured wind speed ( $U$ ) may provide an alternative indication of MO failure. The difference in  $U$  between heights  $z_1$  and  $z_2$  is given by the MO formula (Garratt, 1992):

$$\Delta U_{MO} = \frac{u_*}{k_v} \left[ \ln \left( \frac{z_2}{z_1} \right) - \Psi_M(z_2) + \Psi_M(z_1) \right] \quad (5)$$

where  $\Psi_M$  is a stability correction function. We use:

$$\begin{cases} \Psi_M(z) = 2 \ln[(1+x)/2] + \ln[(1+x^2)/2] - 2 \tan^{-1} x + \pi/2, & \text{(for } L < 0) \\ = -5z/L & , \text{(for } L > 0) \end{cases} \quad (6)$$

where  $x = (1 - 16z/L)^{1/4}$ . With Eq. (5) we compare the difference between measured and calculated  $\Delta U$  for  $z = 1$  and  $2.9 \text{ m}$  ( $\Delta U - \Delta U_{MO}$ ), hereafter referred to as  $\Delta \Delta U$ . A filter is used to reject periods when  $|\Delta \Delta U|$  is greater than a threshold  $|\Delta \Delta U|_{\text{thres}}$ .

Fig. 4 shows the impact of the  $\Delta \Delta U$  filter on the  $0.05 \leq u_* < 0.15 \text{ m s}^{-1}$  group. There is modest improvement in

**Table 1**  
Effect of filtering strategies on MO–LS results.

	No filter	Filtering options			
		$u^*_{\text{thres}} = 0.15 \text{ m s}^{-1}$	$u^*_{\text{thres}} = 0.05 \text{ m s}^{-1}$	$ \Delta\Delta T _{\text{thres}} = 0.5 \text{ K}$	$u^*_{\text{thres}} = 0.05 \text{ m s}^{-1},  \Delta\Delta T _{\text{thres}} = 0.5 \text{ K}$
<b>Average <math>Q_{\text{LS}}/Q(\sigma_{Q/Q})</math></b>					
Total	1.27 (2.00)	0.96 (0.19)	1.04 (0.35)	1.02 (0.30)	1.00 (0.25)
Daytime	0.90 (0.16)	0.89 (0.14)	0.90 (0.16)	0.90 (0.16)	0.90 (0.16)
Nighttime	1.47 (2.47)	1.09 (0.18)	1.14 (0.41)	1.13 (0.35)	1.09 (0.28)
<b>Observations</b>					
Total	195	96	168	153	149
Daytime	70	63	70	70	70
Nighttime	125	33	98	83	79
<b>Data Retention</b>					
Total	100%	49%	86%	78%	76%
Daytime	100%	90%	100%	100%	100%
Nighttime	100%	26%	78%	66%	63%

the average accuracy of  $Q_{\text{LS}}/Q$  as the threshold declines from 0.5 to  $0.1 \text{ m s}^{-1}$ . There is no improvement in  $\sigma_{Q/Q}$  however, and the data loss with the  $\Delta\Delta U$  filter is greater than with the  $\Delta\Delta T$  filter for a similar improvement in  $Q_{\text{LS}}$  accuracy. We conclude that  $\Delta\Delta U$  is not as successful as a criterion based on  $\Delta\Delta T$ .

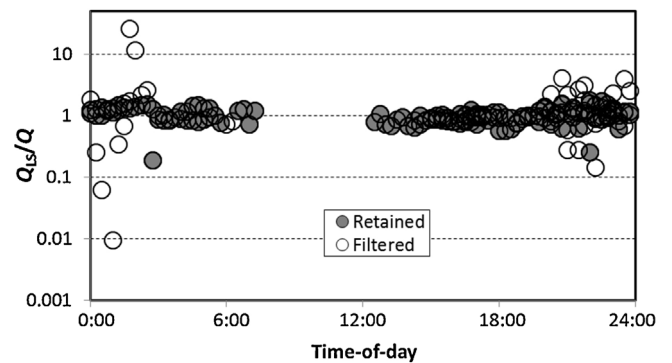
#### 4. Discussion

Our study corroborates earlier work showing good accuracy in  $Q_{\text{LS}}$  during higher wind speeds ( $u^* \geq 0.15 \text{ m s}^{-1}$ ). However, in many experiments a significant portion of observations occur at low wind speeds, and increasing data retention requires identifying low wind periods when accurate inversions are likely to occur. At the lowest wind speeds, when  $u^* < 0.05 \text{ m s}^{-1}$ , the possibility of an accurate calculation appears low. But with  $0.05 \leq u^* < 0.15 \text{ m s}^{-1}$ , we found observations having a mix of accurate and inaccurate inversions. A measurement of  $\Delta\Delta T$  helped to discriminate those two outcomes.

We now consider four filtering strategies applied to our full dataset (including high wind speed periods). The first uses only the stringent  $u^*_{\text{thres}} = 0.15 \text{ m s}^{-1}$  filter, used in previous MO–LS studies. The second relaxes this to  $u^*_{\text{thres}} = 0.05 \text{ m s}^{-1}$ . The filter  $|\Delta\Delta T|_{\text{thres}} = 0.5 \text{ K}$  acts alone in our third strategy. And finally, we combine  $u^*_{\text{thres}} = 0.05 \text{ m s}^{-1}$  and  $|\Delta\Delta T|_{\text{thres}} = 0.5 \text{ K}$ . The impact of these filters is summarized in Table 1. Several things to note:

- Filtering has little effect on daytime results. Even the most restrictive filter ( $u^*_{\text{thres}} = 0.15 \text{ m s}^{-1}$ ) results in a small daytime data loss. While we see no benefit in daytime filtering in our dataset, with only seven periods having very unstable stratification ( $|L| < 5 \text{ m}$ ) we are limited in assessing filtering in unstable daytime conditions.
- Filtering has a large impact at night. Each filter improves the average  $Q_{\text{LS}}/Q$  over the unfiltered data, although the difference in average accuracy between the four filters is small. The greater difference is in the data rejection rate and  $\sigma_{Q/Q}$ . The combination  $u^*_{\text{thres}} = 0.05 \text{ m s}^{-1}$  and  $|\Delta\Delta T|_{\text{thres}} = 0.5 \text{ K}$  is a good compromise of accuracy, period-to-period precision, and high data retention.
- $Q_{\text{LS}}/Q$  is negatively biased during the day, and positively biased at night (statistically different from 1.0 in all cases:  $t$ -test with  $P=0.9$ ). This is consistent with earlier MO–LS observations of a negative/positive bias in unstable/stable stratification (Flesch et al., 2004; Gao et al., 2009). We note the possibility that this bias is an artifact of laser temperature sensitivity. The  $Q_{\text{LS}}/Q$  is correlated with  $T$  ( $r=-0.41$ ), and this is consistent with a greater sensitivity to  $T$  than indicated by the factory calibration<sup>4</sup>.

<sup>4</sup> Laubach et al. (2013) found greater  $T$  sensitivity in their Boreal laser than reported in our factory calibrations.



**Fig. 5.** The  $Q_{\text{LS}}/Q$  plotted with time-of-day (12:00=local noon). The open circles are data that would be removed by filtering with  $u^*_{\text{thres}} = 0.05 \text{ m s}^{-1}$  and  $|\Delta\Delta T|_{\text{thres}} = 0.5 \text{ K}$ .

Fig. 5 shows the effect of the ( $u^*_{\text{thres}} = 0.05 \text{ m s}^{-1}, |\Delta\Delta T|_{\text{thres}} = 0.5 \text{ K}$ ) filter on our full dataset. This filter combination eliminates most of the  $Q_{\text{LS}}$  outliers while retaining 76% of the data. In particular, this filter more than doubles the nighttime data retention of the  $u^*_{\text{thres}} = 0.15 \text{ m s}^{-1}$  filter.

From Table 1 we could conclude that adding  $\Delta\Delta T$  as a filtering criterion, in addition to a relaxed  $u^*_{\text{thres}} = 0.05 \text{ m s}^{-1}$  criterion, results in only a minor improvement in MO–LS accuracy (e.g., the nighttime average  $Q_{\text{LS}}/Q$  improved slightly from 1.14 to 1.09). However, this may underestimate the potential benefit of  $\Delta\Delta T$  in discriminating  $Q_{\text{LS}}$  outliers. In animal studies for example, hourly emission rates may be needed to establish a diurnal emission relationship (e.g. McGinn et al., 2011). Even a large dataset may have a small number of data points in a given hourly interval, and the presence of an outlier can have a large impact. For example, over the 02:00–03:00 interval of our dataset, the  $\Delta\Delta T$  filter reduces  $Q_{\text{LS}}/Q$  from 1.38 to 1.06 compared with the single  $u^*_{\text{thres}} = 0.05 \text{ m s}^{-1}$  criterion. Another benefit with the  $\Delta\Delta T$  filter is a reduction in the random period-to-period error as reflected in  $\sigma_{Q/Q}$ . Adding the supplementary  $\Delta\Delta T$  filter reduces  $\sigma_{Q/Q}$  by approximately 30%. Statistically, this reduction increases the ability to detect emission differences between populations or treatments.

#### 5. Conclusions

A filtering strategy with  $u^*_{\text{thres}} = 0.05 \text{ m s}^{-1}$  and  $|\Delta\Delta T|_{\text{thres}} = 0.5 \text{ K}$  gave a good combination of  $Q_{\text{LS}}$  accuracy and high data retention rates. The value of adding  $\Delta\Delta T$  as part of a refined filtering strategy ultimately depends on the importance of accurate nighttime data. For some types of emission sources the retention of nighttime data is unimportant. For example,  $\text{CH}_4$  produced from

decomposition in deep ponds may have neither a diurnal trend nor wind speed correlation (i.e., emissions are a function of a slowly varying deep bottom temperature). In this case daytime measurements may adequately establish the average emission rate, and applying a larger  $u_{\text{thres}}$  filter to liberally eliminate potential errors is a wise choice. But if emissions vary diurnally (e.g., animals) or correlate with wind speed (e.g., ammonia from soils), retaining light wind nighttime data is important.

The use of this  $|\Delta\Delta T|_{\text{thres}} = 0.5$  K criterion is specific to our measurement heights of 1 and 3 m (nominally). Ideally we would prefer a non-dimensional measure of temperature error, such as the fractional error  $\Delta T/\Delta T_{\text{MO}}$ . However, as  $\Delta T_{\text{MO}}$  is often near zero, large fractional errors occur even when the absolute accuracy is good. Alternatively, one might express the  $\Delta T$  error in gradient form as  $|(\Delta T/\Delta z) - (\Delta T/\Delta z)_{\text{MO}}|$  to increase generality across a range of measurement heights. However, the non-linearity of the temperature profile near ground means this will also be a height dependent criterion.

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