Article

Measuring Trace Gas Emission from Multi-Distributed Sources Using Vertical Radial Plume Mapping (VRPM) and Backward Lagrangian Stochastic (bLS) Techniques

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Abstract: Two micrometeorological techniques for measuring trace gas emission rates from distributed area sources were evaluated using a variety of synthetic area sources. The vertical radial plume mapping (VRPM) and the backward Lagrangian stochastic (bLS) techniques with an open-path optical spectroscopic sensor were evaluated for relative accuracy for multiple emission-source and sensor configurations. The relative accuracy was calculated by dividing the measured emission rate by the actual emission rate; thus, a relative accuracy of 1.0 represents a perfect measure. For a single area emission source, the VRPM technique yielded a somewhat high relative accuracy of 1.38 ± 0.28. The bLS technique resulted in a relative accuracy close to unity, 0.98 ± 0.24. Relative accuracies for dual source emissions for the VRPM and bLS techniques were somewhat similar to single source emissions, 1.23 ± 0.17 and 0.94 ± 0.24, respectively. When the bLS technique was used with vertical point concentrations, the relative accuracy was unacceptably low, <0.62. However, when using multiple point sensors in a horizontal line, the bLS technique yielded a relative accuracy of 1.08 ± 0.44. Altogether, these results suggest that the bLS technique has significant potential for measuring trace gas emissions from distributed agricultural systems.

1. Introduction

This paper evaluates the accuracy and utility of two recently developed micrometeorological techniques for measuring trace gas emissions from distributed area sources: vertical radial plume mapping (VRPM) and the backward Lagrangian stochastic (bLS) techniques. Both techniques determine emission rates from tracer concentrations measured downwind of the source(s). The VRPM technique was developed by the United State Environmental Protection Agency (U.S. EPA) for measuring fugitive emissions from non-point sources such as closed landfills. This technique is currently listed as Other Test Method 10 (OTM 10) under the USEPA Technology Transfer Network Emission Measurement Center Category C [1]. The VRPM technique estimates the horizontal flux of gas passing downwind of the emission source. This "mass-balance" method is similar in principle to the integrated horizontal flux (IHF) and the micrometeorological mass difference (MMD) techniques. It requires knowledge of the tracer concentration and wind fields in a vertical plane downwind of the source. The innovation of the VRPM method is to use open-path optical sensors such as open-path tunable diode laser absorption spectroscopy (OP-TDLAS) or open-path Fourier transform infrared spectroscopy (OP-FTIR). These instruments measure path integrated concentration (PIC), which were used by VRPM to estimate the concentration field across the measurement plane. The VRPM procedure utilizes several downwind PICs at different heights to statistically reconstruct the concentration field [2,3]. The PIC measurements are made from a near-ground concentration sensor to reflectors positioned at different heights on a tower. This technique promises a simplified mass-balance measurement approach and a useful tool for examining emissions from area sources. However, field verification of the accuracy of VRPM is currently incomplete. Based on single trace release experiments conducted by U.S. EPA and Wastemanage, Thoma *et al.* [4] reported an overall plume capture efficiency factor of 0.81 with a standard deviation of 0.33. In a similar but limited examination of VRPM, Ro *et al.* [5] also reported relative errors ranging from −13% to 22% when calculating the emission rate from a single area source.

The bLS technique uses the backward Lagrangian stochastic dispersion model for inversely calculating emissions from distributed sources [6]. The software Wintrax 2.0, a window-based bLS computer program developed by Thunder Beach Scientific [7] was used in this study. Here one measures the concentration of tracer gas at some arbitrary position downwind of a source (or even over the source), and this concentration is used to deduce the emission rate. The relationship between the measured concentration and emission rate depends on the source configuration, wind conditions, and sensor location. In principle these relationships can be quantified with an atmospheric dispersion model called the backward Lagrangian stochastic dispersion model. The technique promises simplicity and flexibility when measuring emissions: Measurement is from a single concentration sensor, placed anywhere within the downwind emission plume. While the bLS technique was developed for idealized situations where the wind is described by simple statistical relationships (e.g., flat homogeneous

terrain), it has been shown to be robust even in non-ideal conditions [8-11]. It has even been used to measure gas emissions from farms [12], fields [11], feedlots [13], ponds [14], and pastures [15].

For this study, the interest was the situation that is often seen in agricultural applications, where two distinct area sources are located adjacent to one another (e.g., two constructed wetlands for wastewater treatment). Specifically, the objectives of this study were to assess the determination of trace gas emission from multi-distributed sources by (1) the VRPM and (2) the bLS techniques.

2. Methodology

2.1. Experimental Layouts

This study was conducted on a grass field at the USDA-ARS Coastal Plains Soil, Water and Plain Research Center in Florence, SC (N 34°14.741′ and W 79°48.605′) during February to April, 2009. During the study period the site was covered with short Bermudagrass typically less than 0.1 m tall. The predominant wind direction was NE-SW along the length of the field (approximately 700 m in length). Three different synthetic emission sources were employed for the study. The first source was constructed of 1.3 cm PVC pipe assembled in a 27 m square grid. The grid was setup with an "I" shaped manifold connected to a cylinder of compressed commercial grade methane. Laterals were connected at 3 m intervals along the manifold. Each lateral had 26, 1.6-mm holes drilled at 1 m intervals along the entire length. A total of 10 laterals were used. The second simulated emission source was constructed using seven soaker hoses stretching out from a gas cylinder to make a star-shaped circular emission source with a diameter of 23 m. The third simulated emission source was constructed of 1.9 cm PVC pipe assembled in a 3.1 m square grid. The grid has similar structure as that of a 27 m grid, except the laterals were connected at 0.6 m intervals; there were 54, 1.6-mm holes drilled at 0.3 m intervals along the length.

Figure 1 shows four different experimental layouts involving a single emission source using the 27 m square grid. On 2/12/2009 and 3/5/2009, the experiments were designed to measure emission rates using the bLS technique with PICs taken within and crossing the distributed emission source. On 2/26/2009 and 3/31/2009, the PICs were taken from downwind of the emission source, and the experiments were designed to measure emission rates using both bLS and VRPM techniques. They were mostly perpendicular to the mean wind directions. On 3/31/2009, additional PICs were taken almost parallel to the mean wind direction in order to assess the accuracy of bLS technique with very small footprints (The fraction of the source area where emissions were "measured" by the sensor. Its values range from 0 to 100%). Also on the single day using a photoacoustic multigas analyzer (INOVA, California Analytical Inc.), we collected vertical point concentrations at 1.1 m, 1.8 m, 3.1 m, and 6.1 m at the center of the distributed emission source for the bLS technique. The ambient concentrations, taken 30 to 60 min prior to gas release, were used as background concentrations for both VRPM and bLS techniques.

Figure 1. Layouts for single emission source experiment (☆ : 3D sonic anemometer; M: location of retroreflector).

2/12/2009
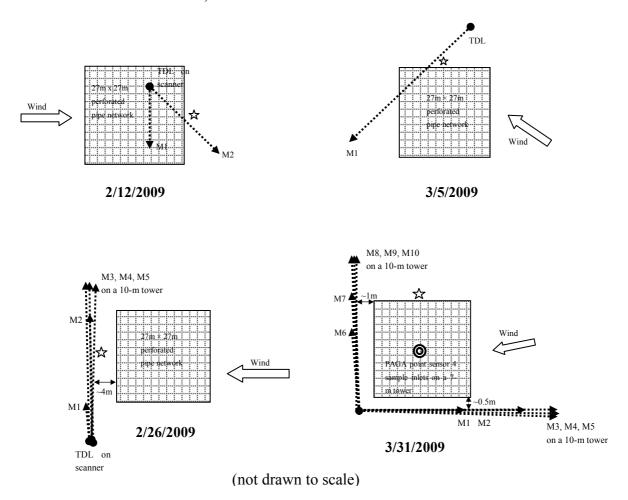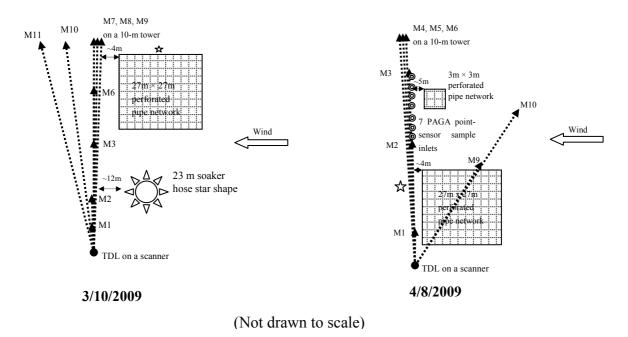
3/5/2009

2/26/2009

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(not drawn to scale)

Figure 2 shows two different experimental layouts for the double emission sources. Both experimental layouts were designed to measure emission rates from the two emission sources. In the case of the VRPM technique which usually requires five downwind PICs perpendicular to mean wind direction, a range of three to seven PICs was used. In the case of the bLS technique which requires two downwind PICs, a range of two to three PICs was used. These bLS PICs were nested within the total PICs used in the VRPM technique. There were also seven ground-level (1 m height) point concentration measurements made using a photoacoustic multigas analyzer. These were along a path length of 27 m approximately perpendicular to mean wind direction. The five points in the middle of the path were 3 m apart, and the two end points were 6 m from the adjacent points. These point concentration data were used to estimate the emission source from the 3.1 m square grid using the bLS technique.

Figure 2. Layout for double emission source experiments (☆: 3D sonic anemometer).

3/10/2009

4/8/2009

(Not drawn to scale)

2.2. Path Integrated Concentration (PIC) Data Collection

The PIC data were taken with the open-path tunable diode laser absorption spectroscopy (OP-TDLAS) system. The OP-TDLAS system consisted of: (1) an open-path tunable diode laser (GasFinder2.0 for CH_4, Boreal Laser Inc., Spruce Grove, Canada) mounted on an automatic positioning device (Model 20 Servo, Sagebrush Technology, Inc.); and (2) Five to eight ground-level retroreflectors at a height of 1 m. In addition, there were 4 retroreflectors mounted on two, 10-m weather station masts (at two heights, 5 m and 10 m). A 3-D sonic anemometer (CSAT, Campbell Scientific, Logan, UT) was mounted on a tripod at a height of 1.77 m. Two cup anemometers (CS800-L Climatronics Wind Speed and Direction Sensor, Campbell Scientific, Logan, UT) were mounted on the 10-m weather station mast (at two heights, 2 m and 10 m).

The communication between the cup wind sensors and the base computer was achieved through the RF401 spread spectrum data radio/modems (Campbell Scientific, Logan, UT). The distance from the GasFinder2.0 to each retroreflector was measured with a 100-m tape ruler. These retroreflectors were positioned in such a way that the vertical plane containing the reflectors was approximately perpendicular to the mean wind direction. The automatic positioning device sequentially directed the infrared beam of the GasFinder2.0 to each retroreflector. At each retroreflector, the GasFinder2.0 collected about 12–15 downwind PIC data sets before moving to next position. The coordinates of the retroreflectors were entered into the VRPM software (Arcadis Inc., NC). Then the PIC data for each retroreflector were saved for reconstructing plane-integrated concentration maps. One or more PICs described above were also used to estimate emission rates using the bLS technique. The automatic positioning device eliminated the need for multiple OP-TDLAS systems. Thus, the bLS technique substantially reduced the total costs.

2.3. Multiple Point Concentration Data Collection

Multiple vertical and horizontal point concentrations of methane were measured using a photoacoustic multi-gas monitor and a multi-channel sampler (California Analytical). The monitor and sampler both collected and analyzed these collected gas samples. The sampling network was through 1/8″ i.d. PTFE tubing. The sample points were measured sequentially.

2.4. Post Data Processing

As recommended by the USEPA, the 3-cycle moving average VRPM data were filtered with the following criteria; (1) concordance correlation factor (CCF) > 0.8 and (2) the mean wind direction between −10° and +25° from perpendicular to the optical vertical plane [1,5].

The software WindTrax 2.0 combines an interface where sources and sensors are mapped with the bLS model [6]. For each study configurations, a map of sources and sensors was created. WindTrax also inputs the time series of concentration and wind measurements to deduce a time series of emissions. For each measurement period, the bLS model calculated thousands of trajectories (5000 in this study) upwind of the concentration sensor in order to determine the relationship between downwind concentration and emissions.

Nonetheless, not all observation periods allowed for good bLS emission measurements: Dispersion models are known to be inaccurate in some conditions. Three criteria were used to evaluate potential inaccuracy [8]:

1. Wind Speed: Wind speed conditions were indicated by the friction velocity (u *) measured by the sonic anemometer. In light wind conditions, dispersion models are known to be error prone. Flesch *et al.* [6] suggested using $u * \geq 0.15$ m s^{-1} as a bLS filtering criteria to improve accuracy.

2. Atmospheric Stability: Atmospheric stratification was indicated by the Obukhov stability length (L). In strongly stable or unstable stratification (e.g., |L| < 2 m), dispersion models can be inaccurate. The |L| threshold value, below which the dispersion model accuracy will deteriorate, should depend on the distance between the source and sensors [6]. Although |L| ≥ 2 m was used as a filtering criteria for a short range study [6], |L| ≥ 10 m was used for a longer range study [8]).

3. Footprint: For some wind directions the emission plume may only "glance" the path of concentration sensors. This leads to uncertain emission estimates. This was determined in WindTrax by the calculation of the sensor "footprint": The fraction of the source area where emissions were "measured" by the sensor.

The relative accuracy of VRPM or bLS technique was calculated as:

$$\text{relative accuracy} = Q_{VRPM} \text{ or } Q_{bLS}/Q \tag{1}$$

where Q = actual emission (g/s), Q_{VRPM} or Q_{bLS} = calculated emission via VRPM or bLS technique (g/s).

The central tendency and its precision of the relative accuracy were represented with arithmetic averages and standard deviations.

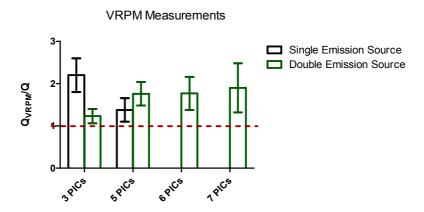## 3. Results and Discussion

3.1. Accuracy of VRPM Technique

3.1.1. Single Emission Source

The relative accuracy of the VRPM technique was evaluated with the experiments conducted on 2/26/2009, 3/31/2009, 3/10/2009, and 4/8/2009. For the single emission source experiments conducted on 2/26/2009 and 3/31/2009, two PICs at 5 and 10m above the ground and three PICs on the ground (M1,2,3,4,5 for 2/26/2009 and M6,7,8,9,10 for 3/31/2009) were used to run the VRPM program. Because of the wind directions were mostly non-perpendicular to the vertical optical plane, only one valid VRPM data set out of 15 datasets was obtained from the 2/26/2009 experiment. Combining with the 12 data sets obtained on 3/31/2009, the relative accuracy of the VRPM technique with single source emission became 1.46 ± 0.26. If VRPM data previously collected by the authors [5] were added to our dataset, the accuracy of the VRPM technique improved slightly to 1.38 ± 0.28 (Figure 3 and Table 1).

Table 1. Relative Accuracies of VRPM and bLS Techniques.

Method	Emission Source	No. of Datasets	Rel. Accuracy
VRPM	single	17	1.38 ± 0.28
VRPM *	double	21	1.23 ± 0.17
bLS [@]	single	10	0.98 ± 0.24
bLS [#]	double	10	0.94 ± 0.24

* total VRPM emission rate using three PICs; [@] bLS emission rate using one PIC data with filtering criteria of $u* \geq 0.15$ m s^{-1}, $|L| \geq 10$ m, and footprint > 30%; [#] average of the combined pool of bLS data using two PICs with filtering criteria of $u* \geq 0.15$ m s^{-1}, $|L| \geq 10$ m, and footprint > 50%.

Figure 3. Accuracy of measured emission Q_{VRPM}/Q from double sources *vs.* number of path integrated concentration data us.

3.1.2. Two Emission Sources

Although the VRPM technique cannot calculate individual emission rates from different emission sources, it was used to estimate the total combined emission rate from the two emission sources

together (if the PIC lines span both sources). The VRPM accuracy using five PICs (M3,6,7,8,9 for 3/10/2009 and M2,3,4,5,6 for 4/8/2009) was only 1.76 ± 0.28. However, due to the mostly non-perpendicular wind direction on 4/8/2009, only one VRPM data was used in this analysis. The impact of using more PICs on the VRPM accuracy was also tested. In a result that contradicted our intuition, use of more ground PICs did not improve the VRPM accuracy for double emission sources: The average VRPM accuracies,1.77 ± 0.39 with six PICs and 1.90 ± 0.58 with seven PICs, were not significantly different from that with five PICs ($P = 0.525$). Unexpectedly, the use of three PICs (one on the ground and two on the mast at 5 m and 10m heights; M7,8,9 for 3/10/2009 and M4,5,6 for 4/8/2009) significantly improved the VRPM accuracy (1.23 ± 0.17) as shown in Figure 3 and Table 1. The use of three heights for the VRPM under two sources probably improved the accuracy because the model reverted to a single Gaussian concentration field. It did not try to make a two source plume into a one-plume source, and, thereby, increasing the expected concentration between the plumes to unreal levels. It appears that the bivariate Gaussian smooth basis function minimization (SBFM) approach to reconstruct the crosswind-smoothed mass-equivalent concentration map was not able to handle multiple emission sources as occurred in this study.
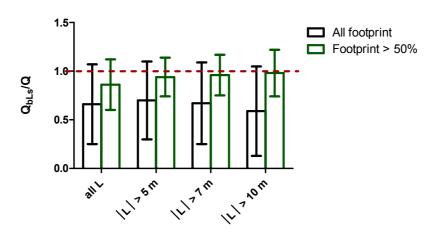
3.2. Accuracy of the bLS Technique

3.2.1. Single Emission Source

The accuracy of the bLS technique was evaluated by dividing the calculated emission rate (Q_{bLS}) by the actual emission rate of methane (Q). Figure 4 shows the accuracy (Q_{bLS}/Q) for the single emission source configuration. It reveals the effect of different filter criteria for atmospheric stability (L). Without any data filtering for stability, the bLS method measured emission rates with an accuracy of 0.66 ± 0.41. Even after removing those data associated with extreme stability (*i.e.*, u * ≤ 0.15 m s^{-1} and |L| ≤ 2 m [6], the accuracy of the calculated emission rates did not change (0.64 ± 0.42). In fact there was no sensitivity in bLS accuracy to stability, even to a threshold of |L| > 10 m.

Figure 4. Accuracy of measured emission Q_{bLS}/Q from single source *vs.* absolute Obukhove length |L| and footprint.
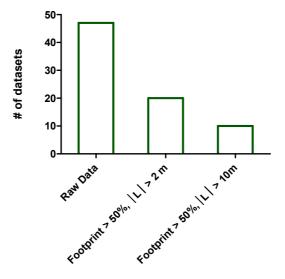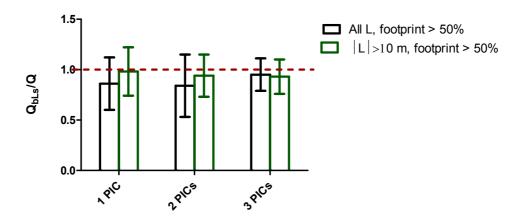
However, there was a large sensitivity in bLS accuracy to degree of sensor footprint coverage over the source. When the footprint coverage was greater than 50%, bLS accuracy significantly improved. The highest accuracy ($Q_{bLS}/Q = 0.98 \pm 0.24$) was obtained when the data were filtered according to the most stringent criteria of $u* \geq 0.15$ m s^{-1}, $|L| \geq 10$ m, and footprint > 50% (Table 1). However, the quest for the higher accuracy resulted in loss of number of viable data points from 47 to mere 10 data points after filtering the data (Figure 5). If the $|L|$ threshold was relaxed to 2 m, the number of dataset increased to 19 with a reduced accuracy of $Q_{bLS}/Q = 0.92 \pm 0.22$.

The impact of using more than one PIC dataset on the accuracy of the bLS technique was also tested. The WindTrax software allows multiple concentration sensors to be used in the emission calculation (results in an average of the emission rates calculated by the different sensors). As shown in Figure 6, the bLS accuracy significantly improved with the increase in number of PIC data with footprint > 50% for all $|L|$ (Q_{bLS}/Q: 0.86 ± 0.26 for one PIC and 0.95 ± 0.16 for three PICs). However, when the data were filtered with the stability and footprint criteria, the accuracy slightly decreased from 0.98 ± 0.24 with one PIC to 0.93 ± 17 with three PICs (Figure 6).

Figure 5. Reduction of number of data with footprint and atmospheric stability requirements.
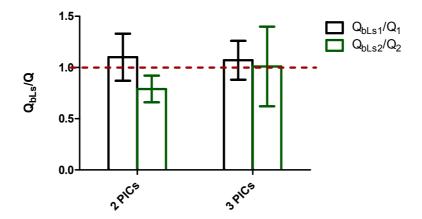

Figure 6. Accuracy of measured emission Q_{bLS}/Q from single source *vs.* number of path integrated concentration data used.

3.2.2. Two Emission Sources

In this section, the accuracy of bLS in deducing the emission rates of two different sources is considered (this is not possible in the VRPM technique). To make this calculation for two sources requires at least two concentration sensors [16]. In principle it is possible to deduce the emission rate from n sources if there are at least n sensors. In addition, sensors must be positioned so that one sensor is in the gas plume of each source. Here emissions are calculated using different combinations of PIC pairs from the data pool of M3, M7, M10, and M11 on 3/10/2009 and M2 and M4, on 4/8/2009 (Figure 2). After filtering the paired PIC data according to the three criteria; $u* > 0.15$ m s^{-1}, $|L| > 10$ m, and footprint > 50%, the accuracies of the calculated emission rate from the two sources were 1.10 ± 0.23 and 0.79 ± 0.13 (Figure 7). The average accuracy of the combined pool of data was 0.94 ± 0.24 (Table 1).

Figure 7. Accuracy of measured emission Q_{bLS}/Q from double sources *vs.* number of path integrated concentration data used.

The good accuracy of the emission rate calculations is somewhat surprising. The work of Crenna *et al.* [16] and Flesch *et al.* [17] showed the tendency of "multi-source" problems like the ones of this study to be ill-conditioned and prone to large errors depending on the geometry of the source and sensor layout. The degree of ill-conditioning can be quantified by the condition number (κ). In our calculations with the PIC pairs, κ ranged from 1.5 to 150. While Flesch *et al.* [17] concluded that accurate emission inferences required κ below approximately 20, in this study two emission rates were calculated to within 35% of their correct value even with κ greater than 100.
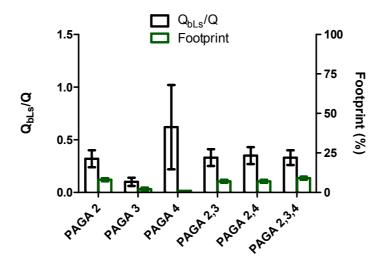
In calculations using PIC pairs the WindTrax model gives the exact solution for two emission rates, *i.e.*, it solves two equations having two unknowns. WindTrax allows more concentration information to be used in an inference. However, the calculation becomes a statistical best-fit problem, *i.e.*, the equation set is over-determined [17]. A three-PIC dataset was created to calculate the two emission rates using the same earlier data pool for 3/10/2009; additionally, M9 and M10 PIC data were added for 4/8/2009. With three PICs observations the bLS accuracy appeared to increase slightly from those using the pair PIC data: 1.07 ± 0.19 and 1.01 ± 0.39. However, these accuracies were not statistically different at $P = 0.05$. Just as with the calculations using data pairs, there was no evidence that

ill-conditioning was a problem in this study. Even with very large κ values approaching 200 in this dataset, there were no large errors in emission rate calculations as documented by Flesch *et al.* [17].
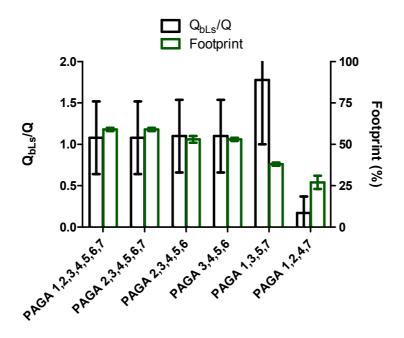
3.3. bLS with Point Concentration Data

The bLS technique is flexible in that either PIC or point concentration measurements can be used to calculate emissions. The accuracy of the bLS technique using vertical and horizontal point concentration data was evaluated on 3/31/2009 and 4/8/2009, respectively. Four vertical point concentrations at the center of the synthetic emission source were taken at the following heights (using the photoacoustic analyzer); PAGA 2 at 1.1 m, PAGA 3 at 1.8 m, PAGA 4 at 3.1 m, and PAGA 5 at 6.1 m. Only the data under atmospheric stability of $|L| > 10$ m and $u * > 0.15$ m were used in this study. The bLS accuracy was very low with the vertical point concentration profile data. The Q_{bLS}/Q ranged from 0.1 to 0.62 depending on the combination of point sensors. The footprint from the point sensors was also very low ranging from 0 to 9% (Figure 8). On 4/8/2009 the concentration sensors were deployed in a horizontal line (at height 1 m above ground) downwind of one of the synthetic sources. Altogether, these sensors broadly represent a PIC sensor ("poor man's" PIC). It is not surprising that when using information from all the point sensors, the bLS results are similar to the earlier PIC based results: $Q_{bLS}/Q = 1.08 \pm 0.44$. As various sensors from the bLS analysis were removed, the accuracy of the emission calculation eventually declined. Insightfully, the decline in accuracy depended on which sensors were removed. For example, removing three particular sensors gave $Q_{bLS}/Q = 1.78$, while removing a different three gave $Q_{bLS}/Q = 0.17$, and a different three again gave $Q_{bLS}/Q = 1.10$. This result can be explained by footprint coverage of the different sensors: Whether they "see" significant parts of the source. When the combination of point sensors had a combined footprint above 50% the bLS accuracy was good. These results highlight the benefit of using line-average concentrations in a bLS analysis, as one can more assuredly acquire good footprint coverage over the source. Ideally this would be done with PIC sensors, but an arrangement of point sensors could accomplish similar desired coverage.

Figure 8. Accuracy of measured emission Q_{bLS}/Q from single source *vs.* different combinations of vertical point concentration data.

Figure 9. Accuracy of measured emission Q_{bLS}/Q from single source *vs.* different combinations of horizontal point concentration data.

4. Conclusions

The accuracy of two recently developed micrometeorological techniques was evaluated using PIC and point concentration data taken from multiple emission sources. Commercial windows-based softwares are available for both techniques; Windtrax for the bLS method and VRPM for the USEPA OTM-10 method.

The VRPM technique yielded an accuracy of 1.38 ± 0.28 with five PICs taken from single emission sources. In contrast to the bLS technique, the VRPM technique could only measure the total combined emission rate from the dual emission sources. The VRPM accuracy did not improve with use of more PICs from dual emission sources. Interestingly, the best VRPM accuracy (1.23 ± 0.17) was obtained with only three PICs for the dual emission sources.

For estimating emission rates from single emission sources, the bLS technique yielded an accuracy as high as 0.98 ± 0.24 after filtering the PIC data with the stringent criteria of $u* \leq 0.15$ m s^{-1}, $|L| \leq 10$ m, and footprint > 50%. A particularly important finding of this study was insensitivity to the problem of ill-conditioning in dual source calculations. Earlier work by Flesch *et al.* [17] suggested ill-conditioning was a serious concern in multi-source applications. However, our results suggest greater robustness in bLS for these applications (at least for two-source problems). For dual emission sources, the bLS technique with two PICs predicted each individual emission rates with accuracies of 1.08 ± 0.22 and 0.80 ± 0.18. Use of three PICs for the dual emission sources did not significantly improve its accuracy.

The bLS technique was also able to use point concentration data to calculate emission rates. The bLS technique with the vertical concentration data at the center of the emission source did not predict emission rates with acceptable accuracies. However, use of the line-horizontal point concentration data taken from downwind of emission source resulted in emission rates with accuracies up to 1.08 ± 0.48.

In addition, the few number of PIC data required by the bLS technique significantly reduced the difficulty of collecting valid data. This, along with the other relatively easily met requirements of the bLS, make the method amenable to many multiple emission-source assessments. Accordingly the bLS technique has significant potential for measuring trace gas emission from distributed environmental and agricultural systems.

Acknowledgements

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