Estimating Surface-Air Gas Fluxes by Inverse Dispersion Using a Backward Lagrangian Stochastic Trajectory Model

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We review the niche for Lagrangian models on the micrometeorological scale in the context of "inverse dispersion," as applied to estimate the rate of gas transfer Q from small surface sources to the atmosphere. The backward Lagrangian stochastic (bLS) method for inverse dispersion is widely used to quantify local sources of such gases as methane and ammonia, typically stemming from the agricultural sector. Data for a particular case study are given, offering interested readers a simple case illustrating the bLS method.

1. INTRODUCTION

Suppose a well-defined area of surface is steadily emitting gas to the atmosphere at an unknown rate Q, causing the mean concentration of that gas to rise above the background value by the amount C (see Figure 1). The concentration rise is linearly proportional to the source strength, and an atmospheric dispersion model might diagnose the mean concentration rise at position $\mathbf{x}$ downwind of the source as (symbolically)

$$C(\mathbf{x}) = Qf(\mathbf{x}|u_*, \beta, L, z_0), \tag{1}$$

where the variables to the right of the "|" in the function $f()$ represent the (given) state of the atmosphere, in this case represented in terms of surface layer scaling variables, namely, the friction velocity u_* or equivalently a mean wind speed U at some arbitrary height, the mean wind direction β, the Obukhov length L, and the surface roughness length z_0 (strictly speaking, we should include the depth δ of the atmospheric boundary layer, but in cases of interest here this difficult-to-procure variable exerts only a small influence on f). For simplicity, we restrict attention to steady sources.

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It follows from equation (1) that if we were to measure C at some point $\mathbf{x_P}$ and provide to a suitable dispersion model an adequate description of atmospheric state, we could deduce the numeric value of Q, the source strength. This is an "inverse dispersion" method, with equation (1) denoting the "C-Q relationship" on the micrometeorological scale in an undisturbed (i.e., horizontally homogeneous) atmospheric surface layer. The sources are of limited streamwise extent, so that a negligible fraction of the trajectories linking the source to the detector make excursions above the surface layer, and accordingly, the dispersion model (computing f) needs to account only for the state of the surface layer, which is described by the Monin-Obukhov similarity theory. As is appropriate in a monograph concerned with Lagrangian models of the atmosphere, we focus here on the case that inverse dispersion is performed using a Lagrangian model of transport and dispersion: more specifically, we cover the so-called "backward Lagrangian stochastic (bLS)" method, meaning that the needed atmospheric dispersion model is a bLS trajectory model, computing an ensemble of random paths backward in time from the detector(s) to the source(s).

Inverse dispersion may be viewed as a form of data assimilation, in the sense that it entails the blending of measurement and theory, and many variants exist: e.g., using a Lagrangian stochastic (LS) model at the smallest scale, *Hsieh et al.* [2003] inverted vertical profiles of carbon dioxide concentration to infer the vertical profile of CO_2 source strength within a plant canopy, while *Seibert and Frank* [2004] used a large-scale LS model driven by the European

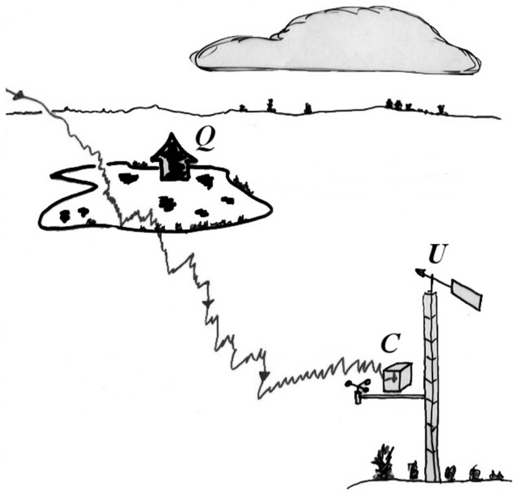

Figure 1. A known patch of the landscape is emitting gas at a steady, unknown rate Q, thereby causing a rise C in the downwind concentration that (for any particular measurement point) must depend on the state of motion of the atmospheric surface layer (i.e., mean wind speed U and mean wind direction β, Obukhov length L, and surface roughness length). The single (and schematic) trajectory that is shown has made a single touchdown on the source (close to its nearby perimeter) and several subsequent touchdowns outside the source.

Centre for Medium-Range Weather Forecasts analysis fields to establish the source-receptor relationship linking measured ^{137}Cs concentration in Stockholm with emission over Ukraine, Belorus, and Russia. It is feasible both to locate a source and to estimate its strength [e.g., *Yee et al.*, 2008], but here we address only the circumstance that one has prior (and complete) knowledge as to source location and perimeter. In terms of antecedents to the "bLS" method, *Wilson et al.* [1982] used a forward LS model to prepare a nomogram for inferring emission from small circular sources based on a single concentration measurement at a particular height ("ZINST") above the center of the plot (the "ZINST" or theoretical profile shape, "TPS" method). The bLS method generalizes this to arbitrary geometry and arbitrary atmospheric state. An earlier review of bLS is given by *Flesch and Wilson* [2005], while *Denmead* [2008] covers bLS in context with the range of other techniques for estimating gas fluxes to the atmosphere.

In section 2, we outline the types of problems for which an inverse dispersion approach is particularly suitable and the reasons for preferring a backward Lagrangian treatment. We will not document technical details of the model (which in many instances can be chosen to be *Thomson's* [1987] well-mixed, 3-D model for Gaussian inhomogeneous turbulence), but in section 3, we will convey the practicalities of implementing bLS at typical sites at which the winds can reasonably be approximated as undisturbed (for clarity, we shall often designate the procedure as "MO-bLS" because one assumes the Monin-Obukhov similarity theory satisfactorily describes the wind statistics) and give an example of the application to measure the rate of volatilization of ammonia after manure spreading (readers interested to learn the practicalities of bLS may wish to download WindTrax and analyze this case as a simple learning exercise). Section 4 will briefly summarize the advantages and disadvantages of accommodating sites that entail flow disturbances by invoking a computed 3-D field of velocity statistics to "drive" the trajectory model, an approach we label "3D-bLS," while section 5 touches on the range of applications of MO-bLS so far seen.

2. THE NICHE FOR INVERSE DISPERSION BY bLS

Figure 2 classifies a range of possible micrometeorological approaches for estimating ground-air gas transfer (for comparative perspectives on these various techniques see *Lapitan et al.* [1999], *Wilson et al.* [2001], *Pattey et al.* [2006], *Denmead* [2008], *Loubet et al.* [2010], and *Harper et al.* [2011]). Eddy covariance is generally regarded as the ground truth method and directly measures the covariance $\overline{w'c'}$ of fluctuations in vertical velocity and gas concentration at a (nominal) point $\mathbf{x_P}$ (if w, c are the total vertical velocity and total concentration, respectively, then the total mean vertical convective flux density is $\overline{wc} = WC + \overline{w'c'}$, where the component due to any apparent mean vertical motion W is usually neglected in favor of the ideal value given by *Webb et al.* [1980]). The eddy flux $\overline{w'c'}$ is (by definition) the turbulent component of the mean vertical mass flux density *at that point* $\mathbf{x_P}$. What surface area the measured point flux $\overline{w'c'}$ represents is defined by (and can be quantified by estimating) the "flux footprint" [*Schmid*, 2002; *Vesala et al.*, 2008], and suffice to say that the latter (for practical instrument heights, say 2 m or higher) covers a substantial distance of upwind surface (some 300 m or more). Then, what if the source is quite small (characteristic dimension of order, say, 1–10 m)? Eddy covariance cannot be used. The same problem arises if a flux-gradient method is contemplated.

One approach to handling small sources is the integrated horizontal flux (IHF) method [see *Denmead and Raupach*, 1993; *Denmead*, 2008], which equates the emission rate to the total *horizontal* flux across a reference plane downwind of the source, an approach that has the appeal that it does not necessitate adopting a transport model, but the disadvantage that it requires multiple wind and concentration sensors (the TPS method was conceived as a means to circumvent large errors in the IHF method, which may arise if the vertical

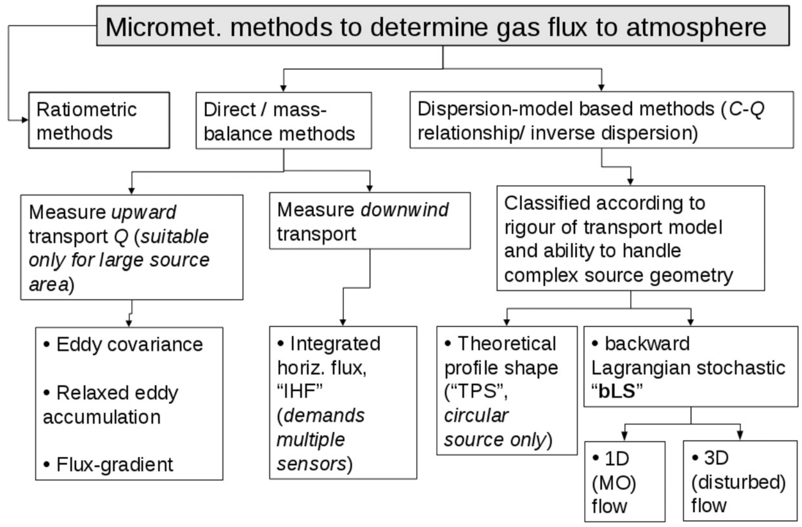

Figure 2. Classification of methods to determine gas flux between ground and atmosphere. (Not all variants of main methods are indicated. "MO" stands for a surface layer flow described by the Monin-Obukhov similarity theory.)

profile of streamwise flux is subject to large measurement errors; in effect, the profile of the horizontal flux at the middle of the circular plot is constrained to conform with theory). Inverse dispersion offers a considerably simpler approach (than IHF) for quantifying emission from small sources, and in a broader context (than "small" sources), it is appealing from the perspective of its convenience. Technically, it is often easier (or less expensive) to measure a mean absolute concentration C than the rapidly fluctuating concentration $c = c(t)$ needed for eddy covariance or the mean vertical concentration difference ΔC needed for a flux-gradient method.

2.1. Why a Lagrangian Model, and Why Backward?

The argument for using a Lagrangian stochastic model in the context of inverse dispersion rests on convenience. Ordinarily, the concentration measurement will be in the far field of the source(s) so that an Eulerian approach (e.g., eddy diffusion or higher-order closure) [e.g., *Wilson et al.*, 2001, section 3] is acceptable in principle. However, the complexity of surface layer wind statistics, even in the case of uniform flow, more or less mandates a numerical approach, i.e., analytic solutions of wide generality and flexibility as regards wind and diffusivity profiles and source distribution do not exist (*McInnes et al.* [1985] treated the special case of circular source geometry by adopting an analytical dispersion model, i.e., a solution of the advection-diffusion equation based on power law profiles of wind and diffusivity, and *Loubet et al.* [2010] took essentially the same approach, albeit for a more complex geometry). Arranging to numerically solve an advection-diffusion equation whose boundary conditions (and profiles U, K) are different for every averaging period (e.g., every 15 min) is not impossible, but it is very inconvenient. Conversely, the Lagrangian treatment, being grid-free, is very convenient and very straightforward to program. Furthermore, an LS model is superior in its ability to directly incorporate all known velocity statistics, e.g., the standard deviations of velocity (σ_u, σ_v, σ_w), which are supplied by the sonic anemometer typically used to procure wind (and temperature) statistics.

The argument for using a *backward* LS model is also one of convenience and computational speed [*Flesch et al.*, 1995]. In a horizontally homogeneous surface layer, backward paths from a point $\mathbf{x}_P = (x_P, y_P, z_P)$ are statistically identical to those from any other point at the same height. This means, for instance, that trajectories from a line-averaging gas detector, such devices being particularly suitable, for reasons covered below, need to be computed only once (for given atmospheric state and surface roughness length, assuming the averaging is along a *horizontal* line) and can be mapped to every other element of that detector.

2.2. Trajectory Touchdowns

An ensemble of backward trajectories originating at the concentration detector will feature a collection of "touchdowns," i.e., points of contact with the surface either within or outside the source or sources (see Figure 1). Because the surface is treated as a perfect reflector, each touchdown is a point where the trajectory is reflected, so that any one trajectory may touch down many times. Letting (x_k, y_k, w_k) label the position and vertical velocity of the kth touchdown, the C-Q relationship for a single source is given by

$$\frac{C}{Q} = \frac{1}{N_P} \sum_k \frac{2}{|w_k|}, \tag{2}$$

where the sum extends over all touchdowns within the perimeter of the source, and N_P is the total number of trajectories computed (k may exceed N_P). The factor $2/|w_k|$ originates from the fact that the residence time of a reflected trajectory within a thin ground-based layer of depth Δz is $2\Delta z/|w_k|$. For inverse dispersion, the salient feature of the backward trajectories, then, is the "touchdown cloud."

2.3. Computational Aspects of bLS

The absence of a grid makes MO-bLS models uniquely simple to implement in software. Concentration sensors and emitting sources can be positioned and designed without restriction (except that they must be close enough to meet the constraints implied by basing the wind statistics on the Monin-Obukhov theory, i.e., trajectories connecting detectors and sources must remain within the surface layer) and can be arbitrarily complex. Sources can be represented as a discontinuous collection of small domains, which will record the occurrence of all particle touchdowns within them, and then be combined to determine the total footprint. Similarly, sensors can be arbitrary collections of points at which backward moving particles are "released" (though in practice, aligned sets of evenly spaced points representing line-integrating sensors are most commonly used).

This flexibility comes at a price: LS models are computationally inefficient and are generally slower to execute than their Eulerian counterpart (though if the latter entails a higher-order closure and is implemented on a high-resolution grid, the difference may not be significant). In part, this is because numerical particles may spend a significant amount of time moving slowly through tiny distances near the surface at each time step. Simulations with large tracking distance to source-area ratios require that more particles be released (and therefore more calculations be performed) to generate meaningful estimates because a given particle is less likely to touch down within the source target. To improve their efficiency, it is important to take advantage of numerical techniques in coding LS models. For example, random number calculations generally use transcendental functions, which require far more clock cycles than addition or multiplication; precalculating a set of random numbers to be used in the motion calculations can reduce computation time by nearly half.

Although it depends on factors such as the distance between source and sensor, the number of particles released, atmospheric stability, the height of release, and the speed of the computer, a set of touchdowns from a single release location takes on the order of a few minutes to complete. Once calculated, a set of touchdowns can be rapidly reused for other release locations with the same release height. There is, therefore, a significant future computational advantage to placing line-averaging sensors as close to horizontal as is practical in the field. Typically, assuming one had already entered the experimental geometry into the diffusion model and had streamlined data acquisition, it is possible to have a first estimate of source strength Q within a few minutes of having procured the needed signals (mean concentration C and wind data), i.e., if winds are suitable, Q can be determined almost in real time.

3. MO-bLS IN UNDISTURBED FLOW

Thomson and Wilson [this volume] provide a history of LS models, and we leave it to the reader to verify that a well-mixed LS model that has been appropriately calibrated provides simulations of dispersion that are in excellent agreement with observations of vertical dispersion, over a broad range of atmospheric stability. (Modern LS models require that one specify a single dimensionless coefficient C_0 that is, in principle, universal; however, as discussed by *Thomson and Wilson* [this volume], it may have different optimal values for different regimes of turbulence.)

But note the qualification "vertical" dispersion. For reasons covered by earlier papers [e.g., *Wilson and Sawford*, 1996; *Wilson et al.*, 2009], the level of error in simulating *crosswind* dispersion is typically much larger, essentially because the variance spectrum of wind direction (or cross wind velocity) is complicated by the idiosyncratic presence of low-frequency energy, related to large quasi-horizontal boundary layer eddies and even larger scales of motion (e.g., mesoscale). Hence, in the context of inverse dispersion, it can be advantageous to use a line-averaging gas detector to integrate (quasi-) horizontally across the plume, because this desensitizes the inferred value of Q to uncertainties in representing horizontal spread. The rapid adoption of bLS, and its track record of useful accuracy, to some extent reflect the fact that IR laser gas detectors with path lengths measured in hundreds of meters have been available for some time. However, in this context we should not neglect a point raised by a reviewer, that is, that the precision and accuracy of open-path instruments (laser or broad spectrum) may be markedly inferior to those of closed-path devices: depending on the source(s) and the placement of detectors, this may compromise accuracy in measuring the needed concentration difference between locations upwind and downwind from the sources [*Laubach*, 2010]. Path integration can be achieved

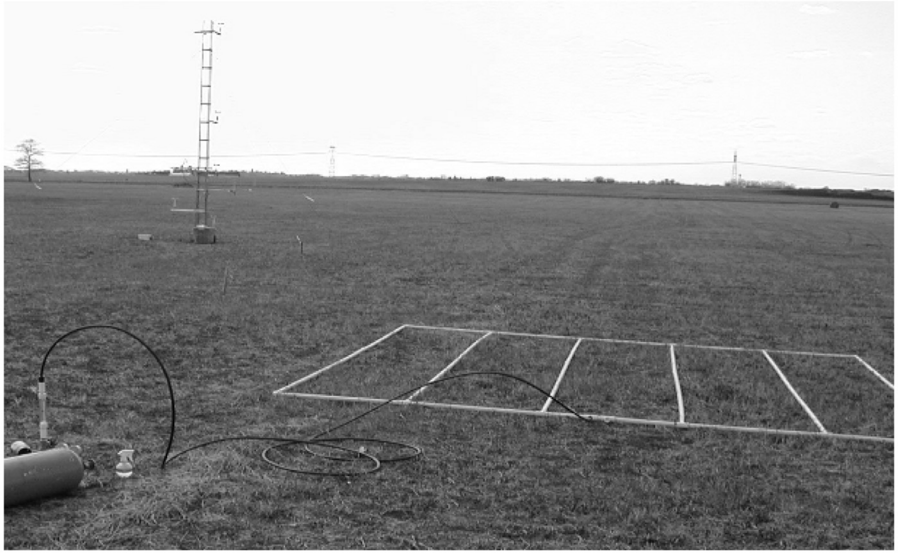

Figure 3. Setup for test of backward Lagrangian stochastic (bLS) method for inverse dispersion [*Flesch et al.* 2004]: a synthetic methane source (6 m × 6 m) on flat, open land at Ellerslie, Alberta.

with point sensors, if wished, by setting up line-averaging intakes [e.g., *Loh et al.*, 2009].

How is a bLS measurement carried out? At some convenient location one will erect a mast and run whatever instruments have been elected to quantify the state of the atmosphere. One will place a concentration detector at some arbitrary location in the downwind plume off the source. If background concentration is nonzero (as is the case, e.g., for methane) and unknown, one will require, also, an upwind detector or one may choose to occasionally move a single detector upwind of the source, or rely on changing wind direction to effect that alteration. It is important to minimize any systematic erroneous offset between upwind (background) and downwind measurements, to ensure a suitably small fractional error in the concentration difference. The downwind detector needs to be close enough to the source so as to meaningfully measure concentration rise and preferably should lie well within, rather than toward the edges of the envelope of the emitted gas plume. Finally, one will record the perimeter of the source and the location of the detector (in the case of a laser, the end points of the light path), typically using a handheld GPS. One will gather the measurements, organized into suitable averaging intervals (typically 15 to 60 min). One will exploit the *C*-*Q* relationship (equation (1), the function *f* being quantified by the Lagrangian model) to obtain a time series of *Q*, perhaps using purpose-designed software (e.g., the freely available "WindTrax," www.thunderbeachscientific.com).

Figure 4. Line-averaging laser concentration detector and synthetic methane source at Ellerslie, Alberta [*Flesch et al.*, 2004].

The experiment described by *Flesch et al.* [2004], a trial of MO-bLS by the controlled release of tracer methane at known rate from a 6 m × 6 m area source on level terrain (see Figures 3, 4), gives an indication of the expected level of uncertainty in MO-bLS estimates of source strength in the simplest circumstances. Others [e.g., *McBain and Desjardin*, 2005] have performed similar tests, and *Harper et al.* [2010a, Tables A1, A2] tabulate the performance of bLS (relative to independently estimated emission rates) for some 20 experiments [see also *Harper et al.*, 2011]. As stressed by one of the reviewers of this present article, the early tracer-release experiments were necessary proof of concept, undertaken in near-ideal, obstacle-free flows, whereas for practical application it is invaluable to have comparisons of bLS with other, corroborating, methods. Such comparisons have been done, in real-world farming situations, e.g., by *Sommer et al.* [2004, 2005] for manure pile emissions, by *Laubach et al.* [2008] for grazing cattle and by *Sintermann et al.* [2011] for manure slurry spreading.

3.1. Example: Ammonia Volatilization From Manure

As an exercise for readers who may wish to learn how a bLS analysis is performed by working through a simple case, we here document all needed information from an experiment that determined the rate of loss of gaseous ammonia (NH_3) from manure spread as a springtime crop fertilizer (Wetaskiwin, Alberta, May 1998). Ammonia volatilization from manure is a process of interest to agronomists, for nitrogen emitted to the atmosphere as NH_3 represents a loss of valuable plant nutrients (there is also the environmental concern of adding reactive N to the atmosphere). Swine manure was sprayed from a truck over a level field of bare, cultivated soil, creating a rectangular NH_3 source 50 m × 100 m in size (Figure 5). With that well-defined source boundary, and in the absence of obstacles near enough to disturb the wind, this experiment represents an ideal application of MO-bLS methodology.

A single open path IR laser NH_3 detector (Gas Finder, Boreal Laser Inc., Edmonton, Canada) provided the line-integrated concentration (C_L) between the laser source and a reflector (Figure 5), the light path running a distance of 161 m at a height of 1 m above ground along the western boundary of the manure source; of course, in this configuration, the bLS technique could be used to calculate emissions only for broadly easterly winds. Five-level wind speed and temperature profiles were measured on a 3 m mast in the field near the manure source, and mean wind direction (β) was measured by a wind vane (β is stated in the compass convention, i.e., β = 90° for an easterly wind). Friction velocity (u_*) and the Obukhov length (L) were calculated from the profiles by best-fitting Monin-Obukhov profiles [e.g., *Argete and Wilson*, 1989], taking the roughness length (z_0, determined from the average of a set of near-neutral wind profiles) as being constant over the observation period. Manure spreading began at 12:15 on 13 May (noon air temperature being about 20°C), and was completed within 15 min. Measurement of ammonia concentration began at 12:00 just before manure application and continued for 31.5 h (until the wind shifted to the west). For bLS calculations, the background ammonia concentration (C_b) was assigned a constant value (about 0.07 ppm) derived from the measured C_L signal prior to manure spreading, i.e., the value at 12:00 (in view of the large concentration rise observed, this approximation should be adequate). The meteorological and concentration time series for the first 9 h of the study are given in Table 1.

The ammonia emission rate was inferred from the observations using WindTrax (see Figure 6), whose user interface greatly facilitates (and largely automates) MO-bLS. WindTrax imports files allowing to define sources of arbitrary perimeter (e.g., as derived from GPS data) and will (if wished) overlay the source(s) and detector(s) on an aerial or satellite image of the site. A built-in "ruler" can be calibrated to the distance between known reference points (e.g., on a satellite photograph), and the coordinate origin can be located wherever is convenient. Column-organized input and output files are flexibly linked by the user within WindTrax, which, once its own diagnostics are satisfied that all necessary information has been provided, computes an ensemble of trajectories and outputs Q. As indicated earlier, the C-Q relationship involves a summation over all trajectory touchdowns on the source (see Figure 6) of the reciprocal of the vertical velocity upon contact with ground.

The ammonia emission rate Q behaved over the 2 days generally as expected (Figure 7). Emissions began promptly after manure application and, after 90 min, peaked at about 400 μg m^{-2} s^{-1}. A high emission rate was sustained for approximately 3 h, after which the rate began to rapidly decline. During the evening following application, Q fell to about 10% of its peak rate. On the day after spreading, the

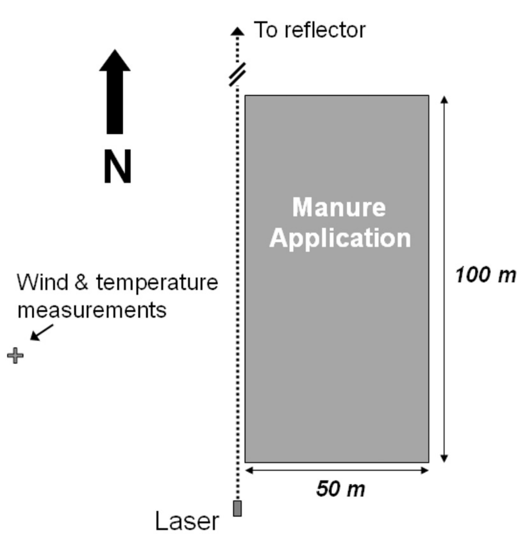

Figure 5. Manure spreading trial, Wetaskiwin, Alberta. Map of the manure source and of detector location. The laser light path length (one-way) was 161 m.

Table 1. Observations From the Manure-Spreading Experiment[a]

Time	u_* (ms^{-1})	L (m)	z_0 (m)	β (°)	C_b (μgm^{-3})	C_L (μgm^{-3})	Q ($\mu gm^{-2}s^{-1}$)
12:00	0.35	−20	0.003	121	43	43	0
12:30	0.37	−20	0.003	122	43	123	32
13:00	0.39	−25	0.003	127	43	748	275
13:30	0.37	−20	0.003	130	43	1083	393
14:00	0.35	−20	0.003	130	43	1226	423
14:30	0.34	−15	0.003	138	43	1262	384
15:00	0.33	−15	0.003	137	43	1340	399
15:30	0.34	−15	0.003	135	43	1299	404
16:00	0.32	−15	0.003	132	43	1184	353
16:30	0.30	−15	0.003	122	43	993	296
17:00	0.27	−10	0.003	153	43	826	196
17:30	0.24	−10	0.003	133	43	870	194
18:00	0.30	−25	0.003	137	43	745	194
18:30	0.30	−40	0.003	135	43	653	168
19:00	0.30	−80	0.003	122	43	517	147
19:30	0.31	−300	0.003	124	43	423	118
20:00	0.28	150	0.003	120	43	363	94
20:30	0.21	50	0.003	137	43	344	55
21:00	0.16	20	0.003	132	43	303	39

[a]The 30 min averages end at the indicated local time. Manure spreading began at 12:15 and was completed by 12:30. The line-averaged NH_3 concentration (C_L) was measured with the laser, with background concentration (C_b) given by measured C_L prior to manure spreading (i.e., the value at 12:00). The NH_3 emission rate (Q) was calculated using backward Lagrangian stochastic method (bLS).

emissions stayed low until about 2 h after sunrise and then began to rise rapidly. The emissions peaked in the early afternoon at about half the rate of the previous day and then declined again into the evening. The total loss of ammonia in 31.5 h after spreading was 12.6 g m^{-2}. The loss the first day was 7.6 g NH_3 m^{-2} and, on the second, 5.0 g NH_3 m^{-2}. This 2 day loss corresponds to 100 kg ha^{-1} of N, a large nitrogen loss to the atmosphere.

It is important to recognize that not all field observations provide good bLS emission estimates and that identifying lesser-quality data is crucial (this is true for any micro-meteorological method). Accurate MO-bLS calculations hinge on the validity of the Monin-Obukhov similarity theory (MOST) as description of the atmosphere. Periods of rapid atmospheric change, or of extreme stability, typically are not well described by MOST and render the calculations suspect. Following *Flesch et al.* [2005b], in this present study we ignored (removed) observation intervals when u_* < 0.15 ms^{-1} or $|L|$ < 10 m. Those filtering criteria eliminated 30% of the measurements, almost exclusively nighttime observations (due to light winds). In order to estimate total N losses, one must estimate emissions during these data gaps. In this case, we can safely assume that nighttime emission rates are small compared to daytime rates. One might assume *zero* emissions during the missing nighttime gaps; however, instead, we chose to accept the error-prone bLS calculations during these periods, acknowledging that they may be associated with large errors but knowing that any errors will be irrelevant given the small emission rates that occur during these periods. In general, there are many other applications where this would not be an advisable option, and thus, the issue of "gap-filling" is an important consideration for many bLS applications.

3.2. Handling Multiple Sources

Sometimes one may wish to simultaneously determine the emission rates from two or more sources, by making several concentration measurements at suitable points. Suppose there are M sources and one has made N measurements of the concentration rise above a known background concentration C_b. Let Q_j (j = 1. . .M) label the unknown source strengths and C_i (i = 1. . .N) the measured concentrations. If $N < M$, the problem is "underdetermined" and cannot be solved. If $N > M$, the problem is "overdetermined" and can be solved to obtain several independent estimates of the Q_j; for example, if $M = 1$ but $N = 2$, one can obtain two estimates of the Q, depending on which of the two C_j one has dropped (ignored). From the two estimates, one could (for example) take an average. Some subjectivity is entailed in choosing the solution to an overdetermined problem (the most objective approach adopts the Bayesian inference process, but this

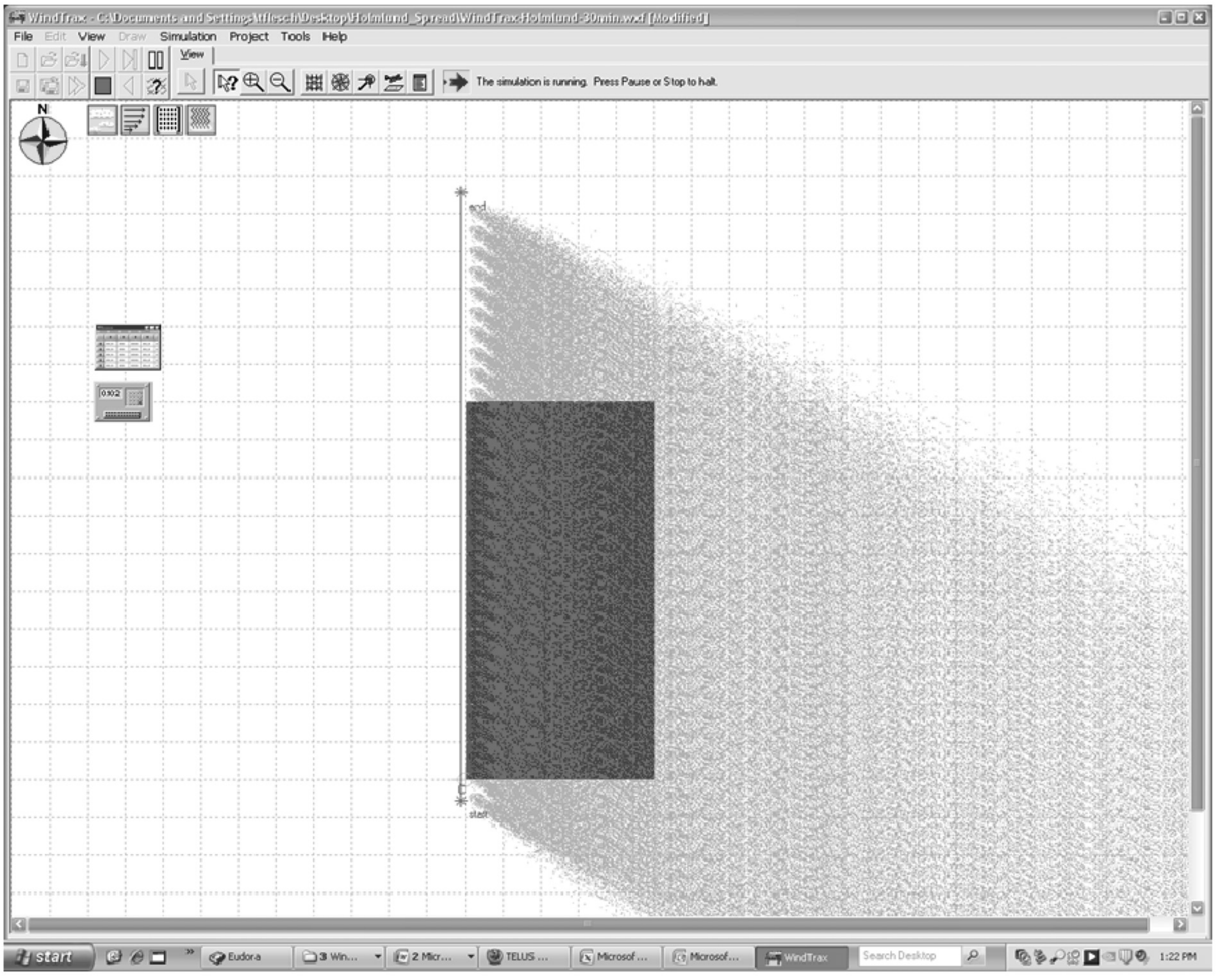

Figure 6. Screenshot of WindTrax. The mean wind direction for this 15 min interval was southeasterly, carrying ammonia from the source (rectangle, 50 m × 100 m) to the laser gas detector (line with endpoint stars denoting laser and reflector, path length 161 m). Computed backward trajectories originate at a single segment of the laser line, and their points of contact with the ground map out a "touchdown cloud." Because of the prevailing horizontal symmetry, the computed touchdown cloud from that one segment of the laser can be mapped to all (in this case, 29) other segments. The laser was 1 m above the ground; thus, the closest touchdowns occurred a short distance upwind of the light path. During this interval, almost the entire source area contributed to the concentration signal detected by the laser.

entails attributing quantitative uncertainties not only to the measurements but also to the modeled Qs).

In the case that $M = N$, the inverse dispersion problem, neither overdetermined nor underdetermined, can be stated as the matrix problem

$$a_{ij} Q_j + C_b = C_i, \tag{3}$$

where summation over j is implied. The solution is

$$Q_j = a_{ij}^{-1} (C_i - C_b). \tag{4}$$

It would appear, then, that one can easily handle multiple sources by including the same number of measurements of concentration rise. In reality, there can be problems in that, depending on the wind direction and the relative sizes, strengths, and positions of these sources, the matrix a_{ij}, whose inverse is (in effect) needed to procure the solution, can be "ill conditioned," meaning there is a large level of uncertainty in the solution.

Simple numerical experiments support the intuitive result that sensitivity to error is minimized when, as far as possible, each of the measured concentrations responds uniquely to a single one of the sources [*Crenna et al.*, 2008]. Attempting to

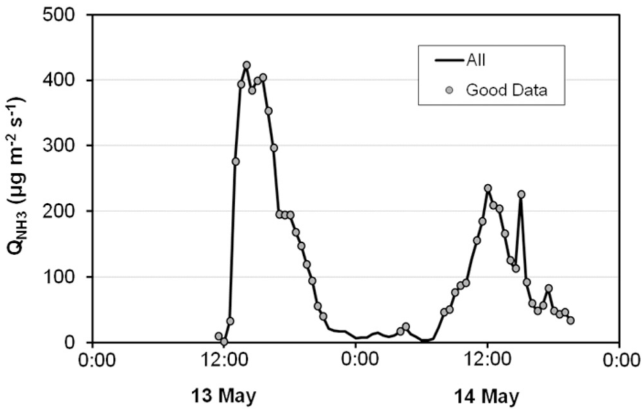

Figure 7. Time series of ammonia emission rate from a manure slurry application. Manure spreading began at 12:15 on 13 May. The black line represents all the bLS emission data, and the circles ("good data") represent data meeting the meteorological threshold requirements.

estimate emission rates from multiple sources on the basis of nearly colocated downstream concentration observations is certain to fail; most likely to succeed are those situations (as defined by their experimental geometry and the orientation of the mean wind) that entail measurements made close to individual sources and having minimal influence from any other sources: ideally, each row of the a_{ij} matrix would have only a single nonzero entry or one entry that is overwhelmingly dominant. Similarly, where the background concentration is significant and unknown, it is best to measure the concentration upstream and downstream of even a single source, rather than attempting to infer both the unknown background and emission rate from multiple downstream measurements alone.

4. INVERSE DISPERSION IN STRONGLY DISTURBED FLOWS: 3D-bLS

In many applications of bLS, one is faced with a site that causes disturbance to the flow, e.g., the terrain may not be perfectly level, there may be buildings or trees, the source, itself (e.g., a lagoon surrounded by a berm) may disturb the flow directly or indirectly (e.g., altered surface temperature). Sometimes the impact of unwanted obstacles can be avoided by limiting the bLS measurements to a suitably restricted range of mean wind directions. Alternatively, it may be possible to place the concentration detector(s) sufficiently far from the location of flow disturbance as to plausibly permit the neglect of such complications, at the expense of a reduction in the magnitude of the concentration rise $C - C_b$ caused by the source, increasing the uncertainty of that key measurement. Trace gas trials with an artificial source surrounded by a windbreak [*Flesch et al.*, 2005a] support this approach. *Wilson et al.* [2001], comparing the several methods one might contemplate using to deduce surface-atmosphere gas fluxes from small surface sources in disturbed flow, found inverse dispersion using MO-bLS second in accuracy only to the integrated horizontal flux method (which makes no direct assumptions regarding the flow). However, subjectivity surrounds the qualifiers "suitably" and "plausibly," and disturbances to the flow field can result in ambiguity. Here it is pertinent to recall that, typically, one lacks the luxury of having an alternative and perfect measurement technique.

Wilson et al. [2010, hereafter WFB] examined whether it might be advantageous (in terms of accuracy of inferred Q) to invoke multidimensional (computed) wind statistics and a more complex LS model ("driven" by gridded 2- or 3-D wind fields). If the concentration detector is located in a region of very disturbed winds, then (as one would expect, and as WFB demonstrated) 3D-bLS does indeed provide a markedly better inference of Q than does MO-bLS. However, this approach to accommodating an imperfect site for bLS very much complicates the inverse dispersion approach, owing to the necessity to perform a flow calculation. Perhaps the salient conclusion of WFB is that their bLS inferences, whether by 3D-bLS or MO-bLS, were *always* within a factor of two of the actual source strength, irrespective of the location of the detector and irrespective of the strongly disturbed wind field.

5. APPLICATIONS OF MO-bLS

Most applications of MO-bLS published to date have addressed agricultural sources, most often of methane or ammonia (for a compilation of examples, see *Harper et al.* [2010a] or *Harper et al.* [2011]). Typically, one seeks to estimate aggregate emissions across distinct types of sources, e.g., animal housing (barns or stockyards), manure storage areas, waste lagoons, and sometimes groups of unconfined animals (sheep or cattle) whose locations might be tracked (by GPS). Generally, it is desirable to resolve the annual cycle in emission rates, and the daily cycle, too, can be quite revealing (as it reflects the cycle in animal activity and farm management). Distinct types of source at a given facility or farm can be quantified individually by appropriately positioning sensors and/or exploiting variations in wind direction. It goes without saying that in these circumstances, simplification and approximation are at the forefront of things. For example, a single effective source with a single effective (mean) emission rate Q may be taken in the analysis to represent what is (in reality, and perhaps even visibly) a patchy landscape and/or a landscape liable to be nonuniform in its emissions. This (with due caution) is justifiable because numerical sensitivity studies [*Flesch et al.*, 2009] have shown that, provided the detector is sufficiently far distant from a nonuniform source, bLS provides a useful estimate of the aggregate (mean) emission rate.

We shall not attempt to convey in detail the literature on applications of bLS (see www.thunderbeachscientific.com for a comprehensive, if not exhaustive, list). Examples from North America include emissions from dairy farms [e.g., *Harper et al.*, 2009; *Leytem et al.*, 2011; *McGinn and Beauchemin*, 2012; *Gao et al.*, 2011], from cattle feedlots [*Todd et al.*, 2011], from hog farms [*Flesch et al.*, 2005b; *Harper et al.*, 2010a], and from poultry barns [*Harper et al.*, 2010b]. Examples from other countries include the works of *Denmead et al.* [2008] in Australia, *Laubach and Kelliher* [2005] and *Laubach et al.* [2008, 2010, 2012] in New Zealand, *Sintermann et al.* [2011] in Switzerland, and *Sanz et al.* [2010] in Spain. Finally, as one example of bLS applied in a very different context, *Loh et al.* [2009] examined the capability of inverse dispersion for monitoring leakage from sites of geosequestration of carbon dioxide and methane.

6. CONCLUSION

We would not want to represent inverse dispersion by MO-bLS as universally the best approach to determining gas emission from small sources, and we have indicated that, where there are disturbances to the wind field, there is inescapably an element of subjectivity in assessing whether a site is suitable. In such circumstances other techniques, such as a ratiometric method or the integrated horizontal flux method, offer the advantage that they circumvent such complications, albeit at the cost of being more complex to implement. (A ratiometric method entails releasing artificial tracer gas at a known rate Q_{tr}, the tracer source distribution being arranged, to the extent possible, to coincide with that of the unknown source of interest; a measured concentration ratio C/C_{tr} determines the unknown source strength Q because $Q/Q_{tr} = C/C_{tr}$).

On the other hand, sufficient evidence has accumulated (see references cited above) to justify the statement that bLS on the micrometeorological scale is a robust technique, with the following important provisos. First, like any other technique, bLS demands user judgment as to site suitability, instrument placement, and quality control (e.g., data filtering to eliminate periods of unsuitably oriented or very light winds). Second, just as the man who wears three unsynchronized watches will always be unsure of the exact time, the user of bLS (or any other technique) who has an overdetermined experimental regime must expect to be offered differing solutions for Q. This is arguably not a flaw of the method. It is the *simplicity* and the *parsimony* of bLS that are its foremost virtues, and if one can afford a method based on more complete measurements, then the latter may indeed (and in principle *should*) outperform bLS, or else the extra information has not been effectively exploited (so there was no point in providing it). Third, one must understand that every atmospheric experiment provides (only) one realization from an ensemble of possible outcomes, while an atmospheric model approximates the ensemble mean outcome: thus, there is a statistical fluctuation in the period-by-period estimates of Q, which may be handled by aggregating estimates (or using longer averaging intervals, within the constraints implied by the assumption of stationarity). Applied at suitable sites by a trained and knowledgeable user, bLS provides aggregate estimates whose level of uncertainty can be conservatively characterized as 10% (as opposed to 1% or 100%).

What future developments may be expected, in terms of refining bLS? Several experimenters have reported that bLS-computed emission rates vary systematically with the positioning of the concentration detector [*McBain and Desjardins*, 2005; *Laubach*, 2010; *Sintermann et al.*, 2011]. Such an outcome might be expected wherever the experimental regime deviates sufficiently from the ideals encapsulated in the MO-bLS inverse dispersion methodology, but should it occur at an ideal site (horizontally homogeneous flow) in an ideal experimental regime (sources of known location/perimeter and uniform strength), then it implies either a bias in the underlying turbulent transport model (in the case of WindTrax, *Thomson's* [1987] multidimensional LS model for Gaussian inhomogeneous turbulence) or the provision to that model (either by WindTrax or by the user) of an imperfect representation of one or more of the needed turbulence statistics. It would be excessively bold to suggest that MO-bLS in its present guise (and as implemented in WindTrax) is not subject to either type of imperfection.

LS models for the atmosphere underwent rapid development in the 1980s and (for the time being) appear to have "plateaued" in terms of their demonstrable fidelity to the "real" atmosphere. Let us take specifically the *Thomson* [1987] model, and let us (hypothetically) adopt MOST parameterizations and field inputs (e.g., u_*,L,β,z_0) that result in "true" (correct) profiles of all needed velocity statistics, namely, velocity means, variances, and covariances, and the turbulent kinetic energy dissipation rate ε. Even given these hypothetical correct profiles, "hypothetical" applies because it is difficult and perhaps even impossible to establish what are truly *the* universal MOST profiles, particularly since the MOST paradigm itself is presumably only an approximation to a more complex truth [*Wilson*, 2008, and references therein]; an unbiased alignment of the model with field measurements still depends on one's "correct" specification of the coefficient C_0, which originates from the Kolmogorov-Obukhov idealization regarding the universal character across turbulent flows of the small scales of motion. Yet the optimal value of C_0 remains somewhat mysterious (for more details, see *Thomson and Wilson* [this volume] on the history of Lagrangian stochastic models).

"Pure" atmospheric dispersion experiments that approach the ideals envisaged in theoretical dispersion models are expensive, thus rare: tuning of LS models (specification of C_0) hinges in most cases on a comparison of the model with the Project Prairie Grass experiment of the 1950s. Furthermore, the tuning of one parameter (say, C_0) may (unwittingly) compensate some other weakness of a model (e.g., a poor choice in the parameterization of ε or the reality that surface layer turbulence is not, in truth, Gaussian). Significant refinement of MO-bLS (as opposed to a shuffling from one case to another of the chosen MOST profiles) seems likely to require a solidification of our knowledge of surface layer flow and of atmospheric dispersion, including new experiments of the utmost rigor that could permit to unambiguously test the significance of what (for the present) are regarded as details that one may ignore (in the spirit of approximation), for instance, the reality that surface layer turbulence is not Gaussian or (speculating) a possible height and/or stability variation of the Kolmogorov coefficient C_0, etc.

As a more modest goal than the "perfection" of bLS, one could contemplate extending the addressable *scale* of application by using a dispersion model that parameterizes wind statistics above the surface layer; the difficulty here is the nonuniversality of the profiles of mean wind direction and speed in the bulk of the atmospheric boundary layer [*Wilson and Flesch*, 2004]. In principle, it should be possible to use additional statistics of the measured concentration signal (e.g., its standard deviation) to refine the estimation of some types of sources, namely, those whose output is fixed (i.e., unaffected by the state and content of the atmosphere overhead), but modeling the standard deviation of a line-averaged concentration is a burdensome computation. Perhaps the most helpful and most practical short-term development would be a "protocol" for bLS that would address the subjective elements, i.e., site suitability and detector placement.

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