# Application of Gauss's Theorem to quantify localized surface emissions from airborne measurements of wind and trace gases

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**Abstract**

Airborne estimates of greenhouse gas emissions are becoming more prevalent with the advent of rapid commercial development of trace gas instrumentation featuring increased measurement accuracy, precision, and frequency, and the swelling interest in the verification of current emission inventories. Multiple airborne studies have indicated that emission inventories may underestimate some hydrocarbon emission sources in U.S. oil and gas producing basins. Consequently, a proper assessment of the accuracy of these airborne methods is crucial to interpreting the meaning of such discrepancies. We present a new method of sampling surface sources of any trace gas for which fast and precise measurements can be made and apply it to methane, ethane, and carbon dioxide on spatial scales of ~1000 m, where consecutive loops are flown around a targeted source region at multiple altitudes. Using Reynolds decomposition for the scalar concentrations, along with Gauss's Theorem, we show that the method accurately accounts for the smaller scale turbulent dispersion of the local plume, which is often ignored in other average "mass balance" methods. With the help of large eddy simulations (LES) we further show how the circling radius can be optimized for the micrometeorological conditions encountered during any flight. Furthermore, by sampling controlled releases of methane and ethane on the ground we can ascertain that the accuracy of the method, in appropriate meteorological conditions, is often better than 10%, with limits of detection below 5 kg hr$^{-1}$ for both methane and ethane. Because of the FAA mandated minimum flight safe altitude of 150 m, placement of the aircraft is critical to preventing a large portion of the emission plume from flowing underneath the lowest aircraft sampling altitude, which is generally the leading source of uncertainty in these measurements. Finally, we show how the accuracy of the method is strongly dependent on the number of sampling loops, or time spent sampling the source plume.

## 1    Introduction

Accurate national inventories of greenhouse gas emissions (primarily carbon dioxide ($CO_2$), methane ($CH_4$), and nitrous oxide ($N_2O$)) is of paramount importance in developing strategies to understand global emissions. The multitude of sources, however, are so often highly variable in area, emission magnitude, height above ground, and duration that rigorous verification is exceedingly difficult. Nevertheless, measurement techniques have improved markedly in the past decade, and these are being employed to an unprecedented extent in an effort to evaluate and refine emission inventories (Nisbet and Weiss, 2010). Most so called "bottom-up" inventories are developed by aggregating statistical correlates of individual process emissions to such mapping variables as population density, energy consumption, head of cattle, etc., extrapolating to total emissions using a relatively small number of direct measurements. On the other hand, atmospheric scientists have long striven to use measurements from global surface networks, aircraft campaigns, and satellites to try to determine emissions based on the amounts and build-up rates of observed trace gases. Aircraft and satellites, the "top-down" approach, conveniently integrates the multitude of sources, but is heavily reliant on a detailed knowledge of atmospheric transport. Top-down methods also suffer from difficulties attributing sources and generalizing measurements made over a relatively short time period. Attempts to reconcile these two distinct methods on global (Muhle et al., 2010) and continental scales (Gerbig et al., 2003; Miller et al., 2013) have often indicated an apparent underestimation by the "bottom-up" methods of a factor 1.5 or more.

In principle, the aircraft top-down measurements can be conducted at all the atmospheric scales to better understand and identify the emissions at comparable scales. For long-lived greenhouse gases, which readily disperse throughout the atmosphere, the global scale is very instructive. The seminal experiment began with Keeling's acclaimed $CO_2$ curve [1960], and has continued through more contemporary techniques by Hirsch et al.(2006) and Neef et al. (2010) for $CH_4$ and $N_2O$, respectively. At progressively smaller scales more details of the source strengths and apportionment can be made: from synoptic or continental scales which can help constrain national inventories (Bergamaschi et al., 2005) or specific biogeographic regions (Gallagher et al., 1994), to mesoscale investigations that estimate emissions from urban areas (Mays et al., 2009; Turnbull et al., 2011; Wecht et al., 2014) or specific oil and gas producing fields (Karion et al., 2013; Petron et al., 2014) and even down to individual point/area sources on the order of 10-100 m size (Denmead et al., 1998; Lavoie et al., 2015; Roscioli et al., 2015).

Aircraft in-situ measurements are particularly useful for "top-down" methods at the sub-mesoscale because they can be used to measure the air both upwind and downwind of a source region. However, deployments tend to be costly and thus sporadic. As far as we know, the aircraft methods used so far can be categorized into three types. First, there is the eddy covariance technique that is carried out at low altitudes wherein the vertical fluxes of gases carried by the turbulent wind are measured by tracking rapid fluctuations of both concentrations and vertical wind  (Hiller et al., 2014; Ritter et al., 1994; Yuan et al., 2015). This method is generally thought to be the most direct, but it is limited to small footprint regions which must be repeatedly sampled for sufficient statistical confidence, requires a sophisticated vertical wind measurement, and can be subject to errors due to flux divergence between the surface and the lowest flight altitude and acceleration sensitivity of the gas sensor. The second, and by far the most common approach is what chemists usually refer to as "mass balance" and what is known in the turbulence community as a "scalar budget" technique. Many different sets of assumptions and sampling strategies are employed, but the overall goal is to sample the main dispersion routes of the surface emissions as they make their way into the overlying atmosphere after first accumulating near the surface. The scales that can be addressed by this method are from a few kilometers (Alfieri and Blanken, 2012; Hacker et al., 2016; Hiller et al., 2014; Tratt et al., 2014) to tens of kilometers (Caulton et al., 2014; Karion et al., 2013; Wratt et al., 2001) to even potentially hundreds of kilometers (Beswick et al., 1998; Chang et al., 2014), and this approach has been the focus of recent measurements in natural gas production basins. These basins present a source apportionment challenge in that emissions from multiple sources (agriculture, oil & gas wells, geologic seepage, etc.) commingle as the air mass travels across the basin. The third method of source quantification is to reference measurements of the unknown trace gas to a reference trace gas with a metered release (tracer) or otherwise known emission rate and assume that the tracer and the scalar of interest have the same diffusion characteristics.  Typically this tracer release technique is applied to small scales of tens to hundreds of meters (Czepiel et al., 1996; Lamb et al., 1995; Roscioli et al., 2015), but the principle has been attempted at the basin (Peischl et al., 2013) and continental (Miller et al., 2012) scales using a reference trace gas with a suitable known emission rate such as $CO_2$ or CO.

The airborne mass balance flight strategies can be grouped into three basic patterns: a single height transect around a source assuming a vertically uniformly mixed boundary layer (Karion et al., 2013); single height upwind/downwind (Wratt et al., 2001) or sometimes just downwind flight legs (Conley

86   et al., 2016; Hacker et al., 2016; Ryerson et al., 1998); and multiple flight legs at different altitudes,
87   (Alfieri et al., 2012; Gordon et al., 2015; Kalthoff et al., 2002); or just a 'screen' on the downwind face
88   of the box (Karion et al., 2015; Lavoie et al., 2015; Mays et al., 2009).

89   Here we describe a new airborne method borne out of a necessity to identify and quantify source
90   emissions to within 20% accuracy in a large heterogeneous field of potential sources. The novel
91   technique applies an aircraft flight pattern that circumscribes a virtual cylinder around an emission
92   source and, using only observed horizontal wind and trace gas concentrations, applies Gauss's
93   Theorem to estimate the flux divergence through that cylinder. By integrating the outward horizontal
94   fluxes at each point along the circular flight path, the flux contributions from enclosed sources can be
95   accounted for. Making an accurate estimate, however, requires the selection of an appropriate
96   circling radius based on the micrometeorological conditions inferred in flight from measurements
97   onboard the aircraft. The pattern must be far enough downstream for the plume to mix sufficiently in
98   the vertical, yet not so far that the trace gas plume enhancements do not stand out sufficiently from
99   the background concentration.

100  In this study we first present the general analytical method used to derive emission estimates using
101  airborne measurements. Next, we investigate the structure of a generalized dispersing plume using
102  large-eddy simulation (LES) to better understand the optimal sampling strategies for quantifying
103  near-surface gas sources. Because the wind fields of turbulent flows cannot be predicted in detail, we
104  do not attempt to compare specific features of our observations with specific LES results, but rather
105  we use the numerical experiments to guide the development of the observational methodology. For
106  example, by investigating the LES flux divergence profiles in the layer below the lowest flight altitude,
107  we are able to estimate the contribution of this unmeasured component to the overall source
108  strength. We then evaluate the accuracy of the approach using coordinated planned release
109  experiments and by applying the method to $CO_2$ emitted from several power plant plumes to
110  compare with reported emissions.

111

## 2   Data Collection

## 2.1   Airborne Instrumentation

114  The airborne detection system is flown on a fixed wing single-engine Mooney aircraft, extensively
115  modified for research as described in Conley et al. (2014). Ambient air is collected through ~5 m of
116  tubing (Kynar, Teflon and stainless steel) that protrudes out of backward-facing aluminum inlets
117  mounted below the right wing. In-situ $CH_4$, $CO_2$, and water vapor are measured with a Picarro 2301f
118  cavity ring down spectrometer as described by Crosson (2008), which is operated in its precision
119  mode at 1 Hz. In-situ ethane ($C_2H_6$) is measured with an Aerodyne Methane/Ethane tunable diode
120  infrared laser direct absorption spectrometer (Yacovitch et al., 2014). There is a 5-10 second time lag
121  in both analyzers that depends on the flow rate and tubing diameter. We use a 1/8" OD (3.175 mm)
122  stainless line for the Picarro (~0.2 slpm flow rate), and a 6.3 mm (¼ in) Teflon line for the $CH_4$/ $C_2H_6$
123  spectrometer (~4 slpm flow rate). This results in lag times of ~5 s for the Aerodyne and ~10 s for the
124  Picarro. The lag time for the Picarro is calculated using a "breath test", whereby we exhale into the air
125  inlet and measure the time required for the $CO_2$ measurement to peak.  The ethane lag time is

126    adjusted to maximize the correlation between the ethane and Picarro methane time series in plumes
127    where both gases are emitted. Both lag times are slightly dependent on pressure, i.e., with a typical
128    altitude change of ~1 km, the change in lag time is less than 10%, and is inconsequential when
129    applying this method within a few hundred meters from the surface. The horizontal wind speed and
130    direction, sampled at 1 Hz, is based on a standard aircraft pitot-static pressure airspeed measurement
131    and a dual GPS compass that determines aircraft heading and ground speed.  The accuracy of the
132    horizontal wind measurement is about 0.2 m s$^{-1}$ (Conley et al., 2014). The horizontal wind is
133    calibrated periodically by flying ~5 km L-shaped patterns in the free troposphere; a heading rotation
134    and airspeed adjustment is made to the wind calculation to minimize the dependence of the wind on
135    aircraft heading. These adjustments typically amount to less than 2° rotation and 3% adjustment of
136    the airspeed. In flying the tight circle patterns described below, the pilot does not adjust the rudder
137    trim to use the same calibration coefficients in the wind measurement calculation throughout the
138    flight.

**2.2   Large Eddy Simulations**

140    In order to study the plume behaviour of surface emissions as it relates to sampling in the stacked
141    circles, we use the LES module of *WRF V3.6.1*. *WRF-LES* explicitly resolves the largest turbulent eddies
142    by filtering the Navier-Stokes scalar conservation equations at some scale in the inertial subrange,
143    and allowing the smaller motions beyond the cut-off to be modeled using a sub-grid (also called a
144    subfilter) scale turbulence parameterization that is based on properties of the larger-scale, resolved
145    flow. Because the aircraft data is typically sampled at 1 Hz and the true airspeed is around 70 m s$^{-1}$,
146    we use an LES horizontal grid size roughly half (40 and 50 m) the distance between aircraft data
147    samples. Because periodic lateral boundary conditions are imposed on the WRF-LES variables, care
148    must  be taken to ensure that the effluent does not reach the lateral boundaries of the simulation
149    domain. On the other hand, WRF-LES does not allow for parallelized computation, making the
150    simulations quite expensive in terms of computation time. We therefore struck a balance between a
151    large enough domain in horizontal extent (6 and 8 km) such that the effluent would not reach the
152    downwind boundary before the end of our simulation, while maintaining a grid size small enough to
153    resolve scales of the aircraft observations. The vertical domain needs to be large enough to
154    encompass a developing convective boundary layer (CBL), while at the same time containing
155    substantial free tropospheric flow above to serve as a reservoir that can feed momentum and free-
156    tropospheric scalars to the CBL. Moreover, the stable region (potential temperature lapse rate $d\theta/dz$
157    = 5 C km$^{-1}$) between the CBL inversion base and the top of the domain had to be large enough to
158    damp any wave activity before it could reflect off the upper boundary and create spurious motions
159    throughout the domain.

160    The standard *WRF-LES* module is not set up to allow for effluent release, so we implemented a
161    modified version of the *WRF* source code (*S.-H. Chen, personal communication*) that includes a
162    surface effluent release with a specified position and release rate. Three different convective
163    simulations were run with varying resultant mean wind speeds in the boundary layer, and each was
164    allowed 4-5 hours to 'spin-up' dynamically before the effluent was released at a rate of between 2.9-
165    3.5 kg hr$^{-1}$. The exact release time was selected to give reasonably stationary CBL depths and
166    turbulent kinetic energy. The conditions for the three simulations are listed in Table 1, and based on

167 the different wind speeds they span moderate to strongly convective boundary layers ($-z_i/L_{MO}$ from
168 ~50 to ~200, where $L_{MO}$ is the Monin-Obukhov length and $z_i$ is the CBL depth.)

## 3    Methods

### 3.1    Theory of Measurement using Gauss's Theorem

171 We use an integrated form of the scalar budget equation for a passive, conservative scalar in a
172 turbulent fluid to estimate the emission of a gas of interest within a cylindrical volume $V$. The volume
173 is circumscribed by a series of closed aircraft flight paths (typically circular) flown around the emission
174 source over a range of altitudes.  The altitudes encompass the lowest safe flight level (usually 75-150
175 mAGL) up to an altitude where no discernable change in the trace gas mixing ratio, $\chi$, is observed
176 around the flight loop, $z_{max}$. The scalar in our case is the mass concentration (i.e., density of a
177 chemically unreactive species in a turbulent flow field, $\boldsymbol{u}$ ($=u\boldsymbol{i}+v\boldsymbol{j}+w\boldsymbol{k}$); its Reynolds decomposition is $c$
178 $= C + c'$, where $C$ is the mean concentration around each loop and $c'$ is the departure from the loop
179 mean. Figure 1 shows an actual example of the effluent sampled by the aircraft in a sequence of
180 stacked paths $l$ that circumscribe an area, $A$, enclosing the source in a volume, $V$.  The effluent is
181 carried downwind as it mixes upward in the CBL. A virtual surface circumscribed by the circular flight
182 tracks is assumed enclosing the source and extending above the vertical extent of the plume so that
183 there is no vertical transport above that level. To estimate the source strength, we start with the
184 integral form of the continuity equation:

$$Q_c = \langle \frac{\partial m}{\partial t} \rangle + \iiint \nabla \cdot c\boldsymbol{u} \, dV \tag{1}$$

185 where $\langle \, \rangle$ denotes an average over the volume $V$, $Q_c$ is the sum of the internal sources and sinks of $c$
186 within $V$, and $m$ is the total mass of c within the volume V.  At this point, we recognize that the flux
187 divergence is composed of two terms

$$\nabla \cdot c\mathbf{u} = \boldsymbol{u} \cdot \nabla c + c\nabla \cdot \mathbf{u} \tag{2}$$

188

189 In section 3.2 we perform a scale analysis of the terms on the right-hand side (rhs) of equation 2 and
190 show that the second term, which is proportional to the horizontal wind divergence, may be
191 neglected under our normal flight protocol.  This is fortunate because of the difficulty in accurately
192 estimating the horizontal wind divergence from aircraft measurements (Lenschow et al., 2007).  The
193 vertical flux across the top of the flight cylinder is assumed to be zero and the flux from the bottom
194 (ground) is the surface source we are measuring.  This leaves us with only the horizontal flux, i.e. $c\boldsymbol{u_h}$
195 where $\boldsymbol{u_h}$ ($= u\mathbf{i}+v\mathbf{j}$).  In order to minimize the contribution from the horizontal wind divergence term,
196 we remove the loop mean concentration, C, which does not alter the first term on the rhs because
197 $\nabla C$ =0, so that equation 2 becomes

$$\boldsymbol{u}_h \cdot \nabla c + c\nabla \cdot \boldsymbol{u}_h = \boldsymbol{u}_h \cdot \nabla(c'). \tag{3}$$

198  Next, we use Gauss's Theorem to relate the volume integral to a surface integral around the volume
199  that is sampled by the aircraft flight loops:

$$Q_c = \langle\frac{\partial m}{\partial t}\rangle + \iiint \nabla \cdot (c'\boldsymbol{u}) \, dV = \left(\frac{\partial m}{\partial t}\right) + \oiint c'\boldsymbol{u} \cdot \hat{\boldsymbol{n}} \, dS \tag{4}$$

200  where $S$ is the surface enclosing $V$ and $\hat{n}$ is an outward pointing unit vector normal to the surface.

201  The surface integral can be broken into three elements: a cylinder extending from the ground up to a
202  level above significant modification by the emission, the ground surface circumscribed by a low-level
203  (virtual) circular flight path ($z = 0$), and a nominally horizontal surface circumscribed by a flight path
204  above the level modified by the source ($z = z_{max}$). We assume there is no significant flux (other than
205  the source of interest) into or out of the ground.  Next, the surface integral is estimated solely from a
206  sequence of closed path integrals measured by the aircraft at multiple flight levels to estimate the
207  right side of Eq. 5 (blue dashed lines in Fig. 1),

$$\oiint c'\boldsymbol{u} \cdot \hat{\boldsymbol{n}} \, dS = \int_0^{z_{max}} \oint c'\boldsymbol{u_h} \cdot \hat{\boldsymbol{n}} dl \, dz, \tag{5}$$

208  where $l$ is the flight path.

209

210  Combining Eqs. 4 and 5 leads to the result that is the basis for this measurement technique where a
211  series of horizontal loops at different altitudes are flown around a source region:

$$Q_c = \langle\frac{\partial m}{\partial t}\rangle + \int_0^{z_{max}} \oint c'\boldsymbol{u_h} \cdot \hat{\boldsymbol{n}} dl \, dz \tag{6}$$

212  Along each path the instantaneous outward flux is computed and summed over the loop to yield the
213  mean flux divergence via Gauss's Theorem.  A temporal trend of the total mass within the volume
214  ($\frac{\partial m}{\partial t}$) can be estimated from the flight data and added to the flux divergence integral to obtain the
215  emission rate.

216

## 3.2  Divergence Uncertainty

218   In order to estimate the relative error in the horizontal divergence term that we are eliminating, we
219  perform a scale analysis of the relative size of the two terms that make up the path integral in Eq. 2,
220  using some typical values of the CBL parameters (convective velocity scale $w_* = 1 \text{ m s}^{-1}$, boundary
221  layer depth, $z_i$ = 1,000 m), and sampling geometry (flying at a radius 1 km around a point source.)
222  Taylor's (1922) statistical theory of dispersion in a homogenous and stationary turbulent fluid
223  predicts that the root mean square lateral ($\sigma_y$) and vertical ($\sigma_z$) dispersion parameters increase
224  linearly with time, or equivalently advection distance, downwind in the near-field. Weil (1988) shows
225  several examples of the growth of both of these parameters downwind to be ~ $0.5w_*$, which we use

226 here for a rough estimate of a conical plume spreading to quantify the dilution of the source's
227 emission as it travels downwind to be intercepted by the aircraft. We use a large background mixing
228 ratio characteristic of global $CH_4$ (~1.9 ppmv), estimate the mean gradient by the plume
229 concentration divided by the distance downwind, and assume a conservatively large horizontal wind
230 divergence of $10^{-5}$ s$^{-1}$, which may in fact be typical for our small sampling region (Stull, 1988). The
231 results are shown in Figure 2 and, for all but the smallest sources of a few kg hr$^{-1}$ and wind speeds
232 below 1 m s$^{-1}$, the divergence term is at least an order of magnitude smaller than the gradient term.

233

## 3.3   Applying the Theory to the LES Results

235 We calculated a comparable estimate of $Q_c$ in the LES domain from the air density, concentration,
236 and wind along circular flight paths as a virtual aircraft would fly. Willis and Deardorff  (1976)
237 generalized results of their convection tank experiments to downwind dispersion in the convective
238 boundary layer (CBL) in terms of a dimensionless length scale $X$, the ratio of the horizontal advection
239 time to the large eddy turnover time:

$$X = \frac{x w_*}{U z_i} \qquad (7)$$

240 where $x$ is the downwind distance and $U$ is the vertically averaged mean wind speed.

241 Figure 3 shows the crosswind-integrated concentration profile for the plume release in the UCD50B
242 WRF-LES run as function of $X$, and normalized height, $Z = z/z_i$. Because of the time limitation due to
243 the periodic boundary conditions, the plume is averaged for only ~15 minutes of simulation time
244 which is just under a large eddy turnover time for the conditions of the run. The results displayed in
245 Figure 3 are in good qualitative agreement with the results of Willis and Deardorff (1976) and Weil et
246 al., (2012) save for the release being at the surface in our LES study, and at $Z = 0.067$ for the above
247 studies (see Fig. 1 and 2 of Weil et al., (2012)). Figure 3 shows the maximum concentration being
248 lofted near X~0.2 and leveling off near $Z \sim 0.8$ around $X \sim 0.6$; beyond $X > 1.5$ the plume is fairly well-
249 mixed throughout the extent of the boundary layer.

## 3.4   The Upwind Directed Turbulent Flux

251 Horizontal turbulent fluxes are generally ignored in boundary layer budget studies due to the fact
252 that while they are often sizeable in magnitude they do not change significantly over horizontal
253 length scales under consideration (the horizontal homogeneity assumption). In the vicinity of a point
254 source, however, this is not likely. The method outlined here estimates source emissions using a
255 measured horizontal flux that incorporates wind and scalar measurements at 1 Hz sample rate,
256 resolving scales of ~70 m (Conley et al., 2014), which should include nearly all of the turbulent
257 contributions to the horizontal flux. Here we consider the nature of this turbulent flux and the error
258 in emission estimates if only the mean transport were considered. We start with the budget equation
259 for a horizontal scalar flux in a horizontally homogeneous turbulent flow where the molecular
260 diffusive/viscous term has been neglected (Wyngaard, 2010),

261
$$\frac{d\overline{c'u'}}{dt} = -\overline{u'^2}\frac{\partial C}{\partial x} - \overline{u'w'}\frac{\partial C}{\partial z} - \overline{c'w'}\frac{\partial U}{\partial z} - \frac{\partial \overline{c'u'^2}}{\partial x} - \frac{\partial \overline{c'u'w'}}{\partial z} - \frac{1}{\rho}\overline{c'\frac{\partial p'}{\partial x}}$$ (8)

262  where $\rho$ is density and $p'$ is the pressure fluctuation. We then assume stationarity and integrate
263  across the source from a point just upwind to a point within the plume and obtain

264
$$\int_{0-}^{x}\frac{\partial \overline{c'u'}}{\partial x'}dx' = -\frac{1}{U}\int_{0-}^{x}\left[\overline{u'^2}\frac{\partial C}{\partial x'} + \overline{u'w'}\frac{\partial C}{\partial z} + \overline{c'w'}\frac{\partial U}{\partial z} + \frac{\partial \overline{c'u'^2}}{\partial x'} + \frac{\partial \overline{c'u'w'}}{\partial z} + \frac{1}{\rho}\overline{c'\frac{\partial p'}{\partial x'}}\right]dx'$$ (9)

265  We further assume that although the scalar field is not homogeneous the flow field is, and the
266  background horizontal c-flux upwind is much smaller than the flux induced by the point source. This
267  results in an equation for the in-plume flux

268
$$\overline{c'u'} = -\frac{1}{U}\left[\overline{u'^2}(C_x - C_{bckg}) + \overline{u'w'}\int_{0-}^{x}\frac{\partial C}{\partial z}dx' + \int_{0-}^{x}\overline{c'w'}\frac{\partial U}{\partial z} + \int_{0-}^{x}\left(\frac{\partial \overline{c'u'^2}}{\partial x'} + \frac{\partial \overline{c'u'w'}}{\partial z} + \frac{1}{\rho}\overline{c'\frac{\partial p'}{\partial x'}}\right)dx'\right]$$ (10)

269  The first three terms on the rhs of equation 10 are negative within the plume with their largest
270  magnitudes on the upwind side and diminishing downwind.  On the largest, boundary layer filling
271  eddy scales the mean concentration of $C$ downwind of a source is greater than in the upwind region,
272  $(C_x-C_{bckg})>0$, and therefore the first term is negative, but decreases in magnitude with distance
273  downwind. However, this term is also positive on smaller scales within the plume where the mean
274  gradient is directed upwind towards the source, and is most likely responsible for the specious
275  intuitive impression that the horizontal turbulent flux should transport the plume downwind from the
276  source along with the mean wind advection. Moreoever, the second and third terms on the rhs are
277  negative because the momentum flux, $\overline{u'w'}$, and mean vertical gradient, $\partial C/\partial z$, are negative while the
278  vertical turbulent flux, $\overline{c'w'}$, and wind shear,$\partial U/\partial z$ , are positive. Based on the vertical concentration
279  profiles shown in Weil et al. (2012) (their Figures 3 & 4) it can be inferred that the vertical
280  concentration gradient, $\partial C/\partial z$, changes from negative to positive near $X{\sim}1$ and becomes negligible for
281  X > 2-3.  Similarly, in the third term, the vertical flux, $\overline{c'w'}$, decreases with fetch. Thus the counter-
282  directed flux ($\overline{c'u'} < 0$) will fade with distance downwind. Wyngaard et al.  (1971)  have shown that
283  the third-moment, turbulent transport terms (4 and 5 on rhs of equation 10) in the horizontal heat
284  flux equation are small in the surface layer compared to the source terms, so we assume the same
285  holds for this scalar flux. Finally, the remaining pressure covariance term is believed to be the main
286  sink in the budget equation working to decorrelate the wind and the scalar as was shown in the
287  surface layer measurements of Wilczak and Bedard (2004). Therefore, the dominant production
288  terms for negative $\overline{c'u'}$ (terms 1-3 on the rhs of 10) must be balanced by the pressure-correlation
289  term leading to an upwind-directed horizontal turbulent flux within the plume that decreases in
290  magnitude in the downwind direction.

291  This conclusion is supported by several previous studies. For example, in a wind-tunnel study of flux-
292  gradient relationships Raupach & Legg (1984) reported that the mean streamwise horizontal heat flux
293  calculated by multiplying the mean wind by the mean temperature overestimates the total heat flux
294  by approximately 10%, which suggests that the turbulent component of the horizontal heat flux is
295  negative; that is, the turbulent flux is upwind, directed counter to the mean flow. Other researchers
296  have reported an even larger disparity. Field experiments by Leuning et al. (1985) indicate that the
297  horizontal turbulent flux of a trace gas is ~15% the mean flux, while Wilson and Shum (1992) suggest
298  it may be 20%. A recent LES study of particle dispersion over a plant canopy by Pan et al. (2014)

299 indicates magnitudes of 20% or more for the negative turbulent component of scalar fluxes in the
300 vicinity of the source and decreasing with downwind fetch. We therefore conclude that when
301 sampling a near-surface point source at $X$ of order unity or less, if only the mean concentration
302 difference is measured, a significant overestimate of the scalar source is likely to occur.

303 Further evidence of this is shown in the average cospectrum of the outward wind and concentration
304 fluctuation observed in the flight loops in Figure 4.  Because the integral of the cospectrum yields the
305 total flux (scalar and wind covariance), this function is useful for examining the contributions to the
306 overall flux from each of the scales of motion (represented by aircraft speed divided by frequency).
307 The results shown in Figure 4 are from a $CH_4$ point source with an estimated emission rate of 46 $\pm$7 kg
308 hr$^{-1}$ which was circled 70 times at a dimensionless radius $X$ of approximately 0.35. All cospectra of
309 sampled sources have the same structure seen in Figure 4: there is an obvious peak at the mean flight
310 loop frequency (usually ~100 s period) followed by a smaller negative dip at higher frequencies within
311 the meandering effluent plume. We believe this to be good evidence that our method captures this
312 important component of the overall flux away from the source, which cannot be obtained with a
313 traditional mean wind and an integrated concentration enhancement measurement that is so often
314 employed in airborne source estimates (Ryerson et al., 2001; White et al., 1976).

## 3.5   Choosing the Downwind Sampling Distance

316 Determining the optimal sampling distance from the targeted point source is a balance of several
317 factors. First, not surprisingly, the largest plume signal occurs closest to the source (Figure 3). Second,
318 a high degree of confidence in the results is contingent upon sampling the majority of the plume at
319 and above the lowest flight altitude, which only occurs downwind after a sufficient time has elapsed
320 to loft the initially near-surface plume. And third, an attempt is made to sample the plume before it
321 reaches the top of the boundary layer so that the vertical turbulent entrainment flux does not
322 become appreciable violating the assumption of negligible flux through the top of the volume V as
323 discussed in Equation 2. Finally, close to the source, the fluctuations in concentration will be very
324 large, intermittent, at small scales, and highly variable.

325 To gain further insight into the second feature of the dispersing plume, Figure 5 shows the average
326 horizontal flux divergence profiles derived from the three WRF-LES runs. Here we discuss a
327 dimensionless $R$, which is identical to $X$, to emphasize that this scaled downwind distance from the
328 source is a radius of a flight loop. The flux divergence values are made dimensionless by the boundary
329 layer height, $z_i$, and the source emission rate, $Q$. Very close to the source, before the plume has had a
330 chance to loft, the flux divergence profile exhibits a strong gradient below the minimum safe flight
331 altitude, making that term difficult to measure directly, as shown in Figure 5. Farther from the source,
332 the signal becomes weaker with increasing altitude and eventually becomes increasingly influenced
333 by entrainment fluxes.  We therefore seek a sampling distance that is far enough to allow sufficient
334 vertical lofting yet close enough so that plume crossings are easily observable against the background
335 variability and instrument noise, and are not yet influenced by entrainment mixing.

336 Based on the simulation results presented in Figure 5, we see the gradient below the lowest flight
337 safe altitude typically becomes very small for R > 0.4, and therefore we attempt to target that
338 distance to minimize the extrapolation error from the flight data to the surface. We do not currently

339    measure all the necessary parameters to estimate R in-flight (primarily the surface heat flux $(\overline{w'\theta'_v})_0$
340    which is required to estimate w$_*$). Instead, we estimate $w_*$ based on the observed boundary layer
341    height, standard deviation of wind speed, and a parameterization for $w_*= \sigma_u$ */0.6 (Caughey and*
342    *Palmer, 1979).*

## 3.6   Minimum number of passes

344    The atmospheric boundary layer is a turbulent medium, meaning that two passes across a plume at
345    the same altitude and distance downwind will likely make very different measurements of the trace
346    gases of interest. A natural question arises as to how many passes are required to develop a
347    statistically sound estimate of the emission rate. We investigate the number of passes required to
348    obtain a statistically robust estimate using the WRF-LES results and a controlled release experiment.
349    By calculating the horizontal flux divergences with a virtual airplane flying through the simulated
350    tracer field, and then randomly sampling the flux divergences from each of the legs and plotting the
351    resultant estimated emission rate as a function of the number of samples used we obtain the results
352    presented in Figure 6. The gray region around the red line mean represents the standard deviation of
353    estimates based on a random set of loops. Figure 7 shows results from an analysis of actual flight data
354    from the ethane controlled release test near Denver, Colorado on November 19, 2014. It is evident
355    from both the simulation data and the field data that a statistically stable estimate seems to be
356    achieved somewhere between 20-25 loops around the source.

## 3.7   Discretization and Altitude Binning the Flux Divergence Data

358    Measurements of the relevant scalars (e.g. $CH_4$) and meteorological variables are sampled at discrete
359    time intervals.  For our analyses, we interpolate all measurements including GPS (3 Hz), methane (1
360    Hz), temperature (1 Hz) and computed variables including horizontal wind (1 Hz), and air density (1
361    Hz) onto a synchronous 1 Hz time series.  Next, we estimate the path integral for each individual loop
362    of the flux normal to the flight path by summing up the flux contributions times the sample length
363    around each loop and then summing over the height intervals,

$$Q_c = \left\langle \frac{\partial m}{\partial t} \right\rangle + \oiint \boldsymbol{F_c} \cdot \hat{\boldsymbol{n}}\, dS = \frac{\Delta m}{\Delta t} + \sum_{z=0}^{z=Z_t}\left(\sum_0^L (\rho \cdot u_n)\cdot \Delta s\right)\cdot \Delta z, \qquad (9)$$

364    where $\rho$ is the scalar air density, $u_n$ is the wind speed normal to the flight path, $\Delta s$ is the distance
365    covered during the 1 s time interval of each measurement and *L* is the distance covered in one
366    complete circuit.  The outer summation sums each of the discrete vertical laps from the bottom (*z* =
367    0) to the highest lap (*z* = $z_t$).  If all laps were sampled at equidistant altitudes, the total divergence
368    could be calculated as the average divergence of all laps multiplied by the top altitude. However,
369    because there is greater horizontal transport and variability at lower altitudes, as demonstrated by
370    the widening standard deviations approaching the surface in the theoretical flux divergence profiles
371    shown in Figure 4, more sampling laps at lower altitudes increase the statistical validity of the largest
372    horizontal transport values. To ensure that all altitudes are nearly equally weighted, we divide the
373    vertical range into six equally spaced bins, save for the lowest bin which is extended to the surface,
374    and then average the measurements from the laps within each bin. The total emission is the sum of
375    the flux times the path length in each bin multiplied by the bin width. We also performed 6 flights

376  where we sampled equally at all altitudes to allow a comparison of the direct average versus the
377  binned results, and in all of these flights the values derived by the two methods agreed to within 5%.

378  **3.8   Error Analysis**

379  Our method assumes a stationary emission source.  The leg-to-leg variability is primarily driven by the
380  stochastic nature of turbulence (e.g. we may sample the plume on one lap and miss it on another). By
381  aggregating the laps into vertical bins, we can use the standard deviation of the horizontal fluxes
382  within each bin as an estimate of the uncertainty within that bin. Then the total uncertainty in the
383  estimate of the flux divergence is simply estimated by adding up the individual bin uncertainties in
384  quadrature. The first term on the rhs of Equation 6 is the time rate of change of the scalar mass
385  within the cylindrical flight volume. This storage term is estimated by performing a least squares fit of
386  the methane density with time and altitude. The resulting uncertainty in the time rate of change is
387  then combined (summed in quadrature) with the uncertainty from the altitude bins to achieve a total
388  uncertainty in the measurement.
389
390  **4   Results and Discussion**

391

392  We use measurements from three sets of flights to characterize the accuracy of this estimation
393  method.  We flew 2 days measuring an ethane controlled release provided by Aerodyne Research,
394  Inc., 4 days measuring a natural gas controlled release provided by the Pacific Gas & Electric Company
395  (PG&E), and 6 power-plant flights where our estimates are compared with reported hourly power
396  plant $CO_2$ emissions.

397
398

399  **4.1   Ethane Controlled Releases**

400  Two experiments with known/controlled ethane releases were performed in collaboration with the
401  Aerodyne Mobile Laboratory team.  Pure ethane was released and measured with a flowmeter by the
402  Aerodyne ground crew.  The Colorado site (November, 2014) was in a remote area approximately 105
403  miles NE of Denver.  This site was chosen because of the flat terrain and lack of other nearby ethane
404  sources that could pollute the controlled release plume.  The flux profiles for both releases are shown
405  in Figure 8 and indicate that, in both cases, the aircraft successfully flew above the ethane plume
406  (measurements tend toward zero with increasing altitude).  An example of an individual lap is shown
407  in Figure 9 and indicates a clear plume signal downwind of the release.  As the aircraft climbs,
408  eventually the signal disappears, as shown in the figure.  Agreement was excellent, with the
409  estimated emission just 2% over the actual controlled release rate.   The second Aerodyne controlled
410  release in Arkansas on October 3, 2015 was performed at a site surrounded by nearby emission
411  sources and an elevation change (~70m) within the aircraft flight path. The aircraft-derived ethane
412  emission estimate was 25% higher than the controlled release rate and the calculated uncertainty
413  was significantly higher than on other sites (Table 2).

414   A significant upwind ethane source was observed during the Arkansas experiment. This source was
415   evident on roughly half of the upwind passes, suggesting that techniques which rely on a limited
416   number of upwind passes to characterize the background could have a large random error and thus
417   erroneously estimate the upwind source strength.  A similar problem would affect those techniques
418   that employ a downwind transect, using the edges of that transect lying outside the plume to
419   estimate the background concentration.  These observations demonstrate the complication (and
420   bias) that can arise from nearby sources.  Since this method integrates all the emission sources in the
421   area within the flight circle and a small distance upwind of the circle depending on the vertical
422   mixing, estimates from Gauss's method may be biased high if there are sources within that area.  The
423   average error of the two ethane releases is 13%.
424

## 4.2   Natural Gas Controlled Releases

426   In conjunction with PG&E, we performed two sets of two-day ground-level controlled release
427   experiments from existing PG&E facilities, exactly one year apart.  The first set was performed
428   southeast of Sacramento near the town of Rio Vista, CA at the Rio Vista "Y" station and the second
429   set near Bakersfield, CA.  For the Rio Vista test, the release rate was not calibrated with a flow meter
430   but, based on the size of the orifice and the upstream pressure, the release rate was estimated at
431   $15.2 \pm 1.5$ kg hr$^{-1}$. This release rate is an estimate of the total gas being released which is a
432   combination of primarily $CH_4$ and $C_2H_6$.  We use the regression fit of ethane to methane (averaging
433   0.085 by mass) to estimate the actual release rate of each scalar.

434   In comparison with the $C_2H_6$ controlled release, $CH_4$ controlled releases suffer from the effect of small
435   enhancements relative to the background concentration. During the Rio Vista release, the largest
436   enhancement that we measured was 100 ppb, with 30-40 ppb being typical.  Using a typical
437   background level of 1.9 ppm, a 40 ppb enhancement represents 2% of the background.  In contrast,
438   for ethane the enhancements are as large or larger than the background.   The results of the methane
439   controlled release tests are shown in Table 3  and indicate aircraft-derived estimates within 17% of
440   the controlled release rate.  This large background results in increased uncertainty in the emission
441   calculation.  The average difference between the estimated emission and the calculated flow rate is
442   13%.
443

## 4.3   Power Plant Flights

445   Power plants in the U.S. are required to report $CO_2$ emissions to EPA (https://ampd.epa.gov/ampd)
446   on an hourly basis. The accuracy of the reported $CO_2$ emissions has been determined to be ±10.8-
447   11.0% based on reported U.S. average differences between Energy Information Administration (EIA)
448   fuel-based estimates and EPA continuous emission monitoring-based estimates (Ackerman and
449   Sundquist, 2008; Peischl et al., 2010; Quick, 2014).  Also, Peischl et al. (2010) determined an accuracy
450   of power plants reporting $CO_2$ emissions in Texas of ±14.0% based on differences between observed
451   downwind $SO_2/CO_2$ and $NOx/CO_2$ emission ratios and those reported via EPA continuous emission
452   monitoring (Peischl et al., 2010). Here, we use the slightly larger uncertainty from Peischl et al.

453   (2010).  Power plant emissions are "hot" gases and very buoyant, in contrast to a surface emission
454   source that is typically not buoyant.  An additional uncertainty arises from temporal emission
455   variability (hourly averaged reported $CO_2$ emissions vs. <1 hr power plant flights that may cover parts
456   of two reported consecutive hourly values). We estimate the total reported uncertainty by summing
457   in quadrature the Peischl estimate and the relative difference between two reported consecutive
458   hourly $CO_2$ emission values closest to the time of the power plant sampling. The aircraft frequently
459   encountered power plants during oil & gas monitoring campaigns, but usually did not have the flight
460   time to perform a full emissions characterization of the power plant.  Here we limit our comparison
461   to days when the aircraft performed a minimum of 10 laps around the plant, thus excluding the quick
462   fly-bys where uncertainties would be unacceptably large.  The results are presented in Table 4 and
463   indicate very good agreement between Gauss's method and the reported $CO_2$ emissions with the
464   averaged difference being 10.6%.   A comparison plot of the reported versus measured $CO_2$ emissions
465   is shown in Figure 10. The average difference between the reported and measured emissions for the
466   5 power plants is 11%.

467

## 5      Conclusion

469   This technique was developed out of the necessity to identify and quantify individual well pads in an
470   extensive oil and gas production field. Consequently the frequent tracking of the upwind and
471   downwind side of the source provides a very accurate determination of the location and magnitude
472   of a given emission site. The main uncertainty arises from the effluent below the lowest flight
473   altitude, but this is minimized by targeting a downwind distance determined by LES studies to provide
474   very little change in the plume flux divergence from the lowest loop to the ground. In addition to the
475   controlled release experiments, hundreds of sites have been measured using this technique with
476   varying levels of success.  Ideal conditions include flat terrain, ample sunlight to promote vertical
477   mixing, consistent winds, and no nearby competing sources.  Under optimal conditions we have
478   demonstrated that measurement uncertainties are quite low, often better than 10%.  As the
479   conditions deteriorate from the ideal to situations involving complex terrain, variable winds or nearby
480   upwind sources, measured uncertainties can increase to be as large or larger than the emission
481   estimates themselves. In the worst case of stably stratified conditions (winter or night time), for
482   instance, the lack of vertical mixing may preclude the trace gases emitted at the surface from
483   reaching the minimum safe flight altitude. Complex terrain provides a challenge to the method
484   because the aircraft is unable to maintain a constant altitude above the ground.  A possible future
485   refinement of this technique to be applied in complex terrain would be to fit the measurements of
486   both wind and mixing ratio to a uniform 3-dimensional surface surrounding the source, where the
487   grid passes through the terrain and then integrate the flux normal to this irregular virtual flight path.
488   This would not assume level loop flight legs and would, in principle, account for individual loops being
489   flown at differing altitudes and thus more closely track mass continuity near the terrain elevation.

490

| Simulation | $\Delta x$, $\Delta y$ (m) | Lx, Ly (km) | $\Delta Z$ (km) | $\Delta t$ (s) | $\Delta z$ (m) | CBL Depth (m) | CBL mean wind (ms$^{-1}$) | $w_*$ (ms$^{-1}$) | $-z_i/L_{MO}$ | $X_{max}$ |
|---|---|---|---|---|---|---|---|---|---|---|
| **UCD50A** | 50 | 8 | 2.5 | 0.30 | 8 | 750 | 2 | 0.92 | 210 | 4.5 |
| **UCD50B** | 50 | 8 | 2.5 | 0.30 | 8 | 600 | 3.8 | 0.86 | 73 | 3.6 |
| **UCD40** | 40 | 6 | 2.5 | 0.24 | 10 | 850 | 4.5 | 0.96 | 53 | 2.4 |

Table 1 - Domain and micrometeorological parameters for the three WRF-LES experiments in this study. *L* represents the Monin-Obukhov length.

| Experiment Location | Date | Laps | Released $CH_4$ kg hr$^{-1}$ | Estimated $CH_4$ kg hr$^{-1}$ | Released $C_2H_6$ kg hr$^{-1}$ | Estimated $C_2H_6$ kg hr$^{-1}$ | Ethane Difference |
|---|---|---|---|---|---|---|---|
| Colorado | 2014-Nov-19 | 50 | 0.0 | -0.1±0.3 | 5.5±0.5 | 5.6±2.9 | +2% |
| Arkansas | 2015-Oct-03 | 19 | 0.0 | -3.4±12.3 | 8.1±0.8 | 10.0±6.1 | +24% |

Table 2 - Ethane controlled releases.

| Experiment Location | Date | Laps | Released $CH_4$ kg hr$^{-1}$ | Estimated $CH_4$ kg hr$^{-1}$ | Released $C_2H_6$ kg hr$^{-1}$ | Estimated $C_2H_6$ kg hr$^{-1}$ | Methane Difference |
|---|---|---|---|---|---|---|---|
| Rio Vista | 2014-Nov-03 | 37 | 13.9±2.8 | 12.8±8.5 | 1.2±0.5 | 0.6±0.4 | -8% |
| Rio Vista | 2014-Nov-04 | 27 | 13.9±2.8 | 11.5±3.2 | 1.2±0.5 | 0.5±0.3 | -17% |

Table 3 - Natural Gas controlled release

| Power Plant | Date | Hour UTC | Laps | Reported $CO_2$ T hr$^{-1}$ | Estimated $CO_2$ T hr$^{-1}$ | Difference |
|---|---|---|---|---|---|---|
| Rocky Mountain Energy | 2014-Oct-06 | 20 | 19 | 99±14 | 111±24 | 13% |
| Saint Vrain | 2014-Oct-04 | 19 | 21 | 124±17 | 122±41 | -1% |
| Pawnee | 2014-Nov-19 | 20 | 14 | 575±81 | 555±160 | -3% |
| Saint Vrain | 2015-Sep-17 | 20 | 14 | 361±54 | 280±115 | -23% |
| Four Corners Power Plant | 2015-Apr-11 | 18 | 12 | 1289±387 | 1119±343 | -13% |

Table 4 - Power Plant estimates. The mid-point of the measurements (hours UTC) is indicated in the third column (Hour). The reported emissions from the hour before to the hour after that time were averaged to derive the "Reported" emissions in column 5. Emissions are reported in units of metric tons (T) per hour.

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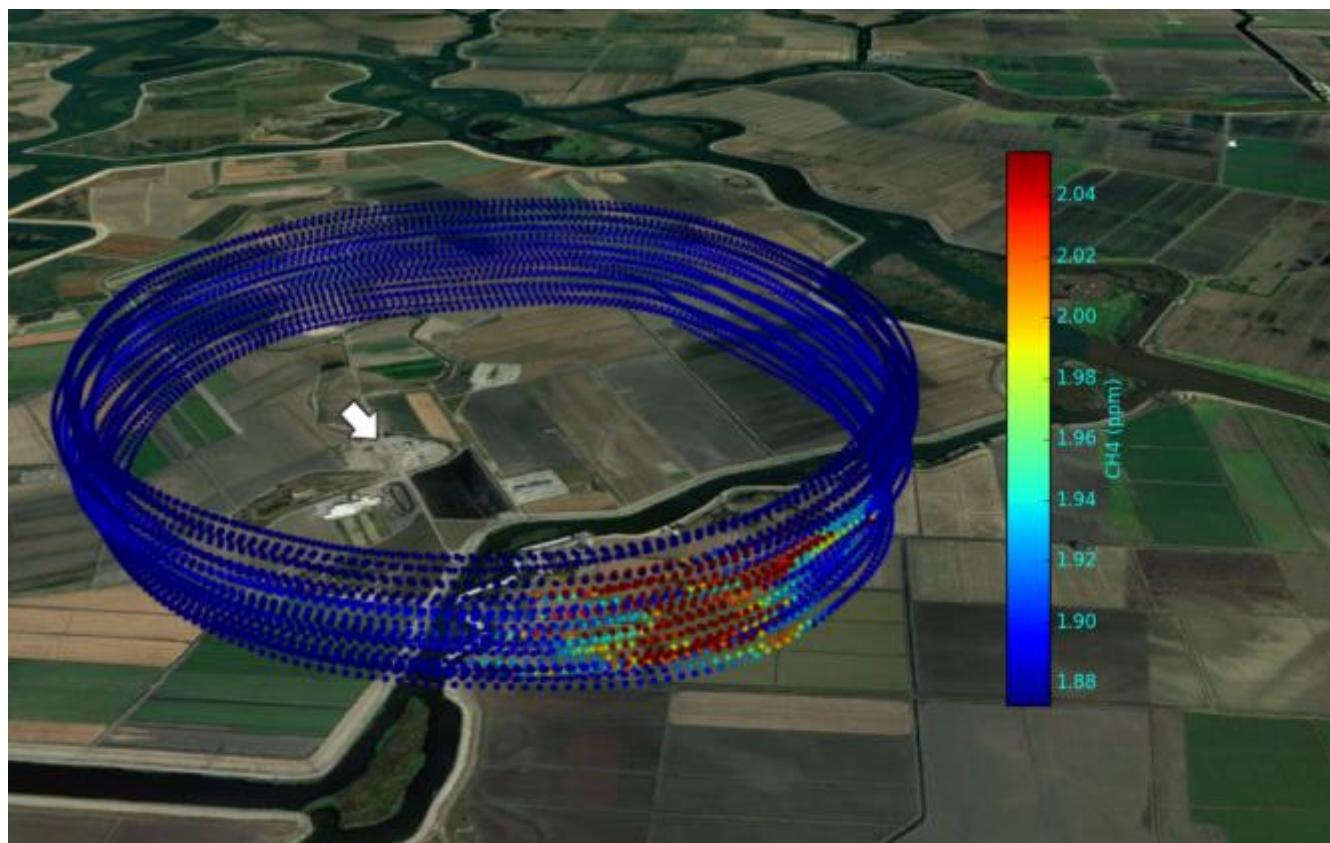
**Figure 1 - Map of the airplane flight pattern sampling a methane plume emanating from an underground storage facility. Wind direction is indicated by the white arrow and the methane mixing ratio is given by the color bar to the right. This flight was conducted on June 28, 2016 and took place between 12:46PM and 1:52PM LT at altitudes ranging from 91 m to 560 m with a loop diameter of approximately 3 km. The measured methane emission rate was 763±127 kg hr$^{-1}$.**
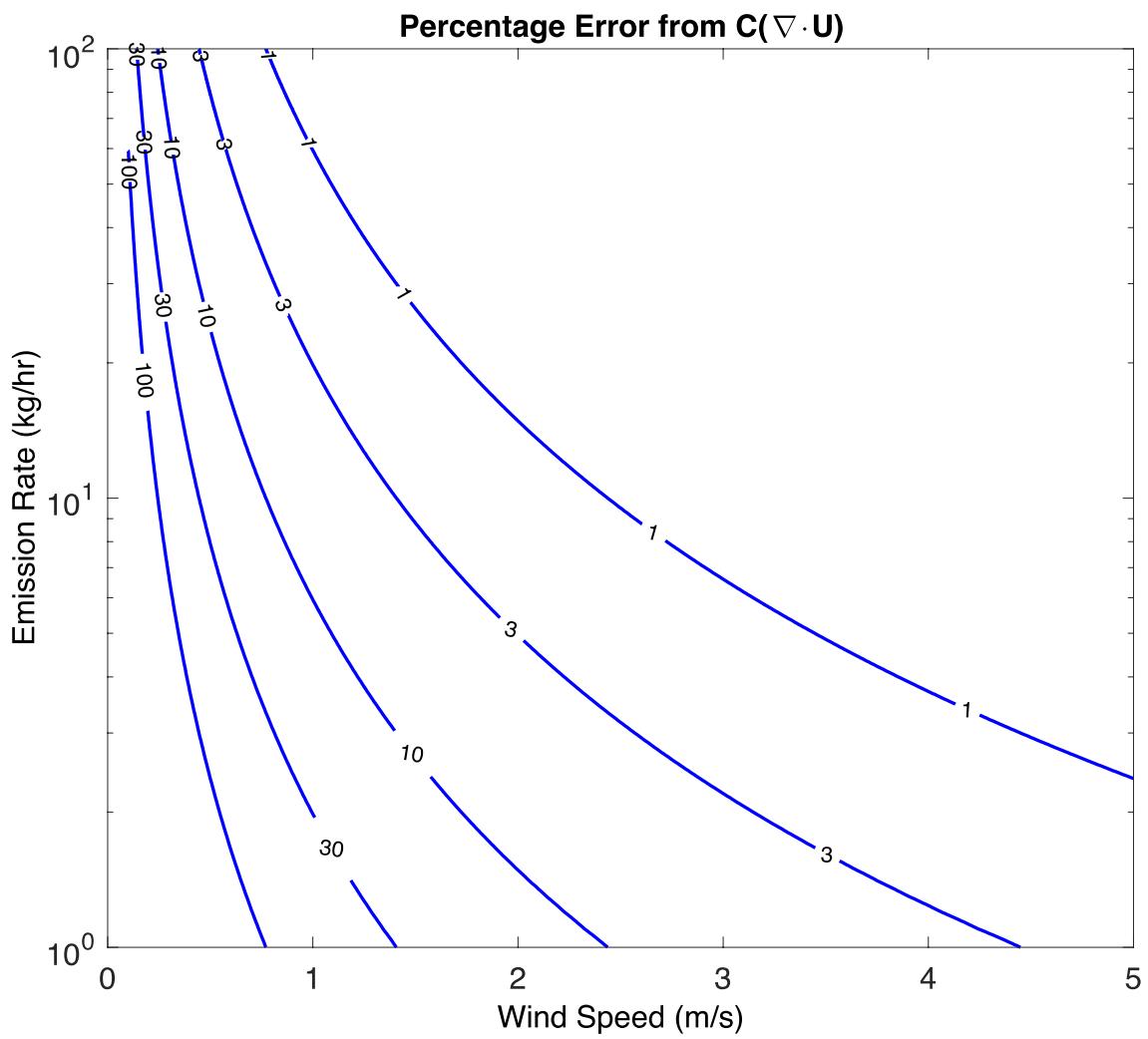
**Figure 2** - Graphical representation of the relative magnitude (%) of the contribution of the horizontal wind divergence to the horizontal advective terms in Equation 3, as a function of wind speed and source magnitude for methane, using a typical global background of 1.9 ppm and divergence of $10^{-5}$ s$^{-1}$.
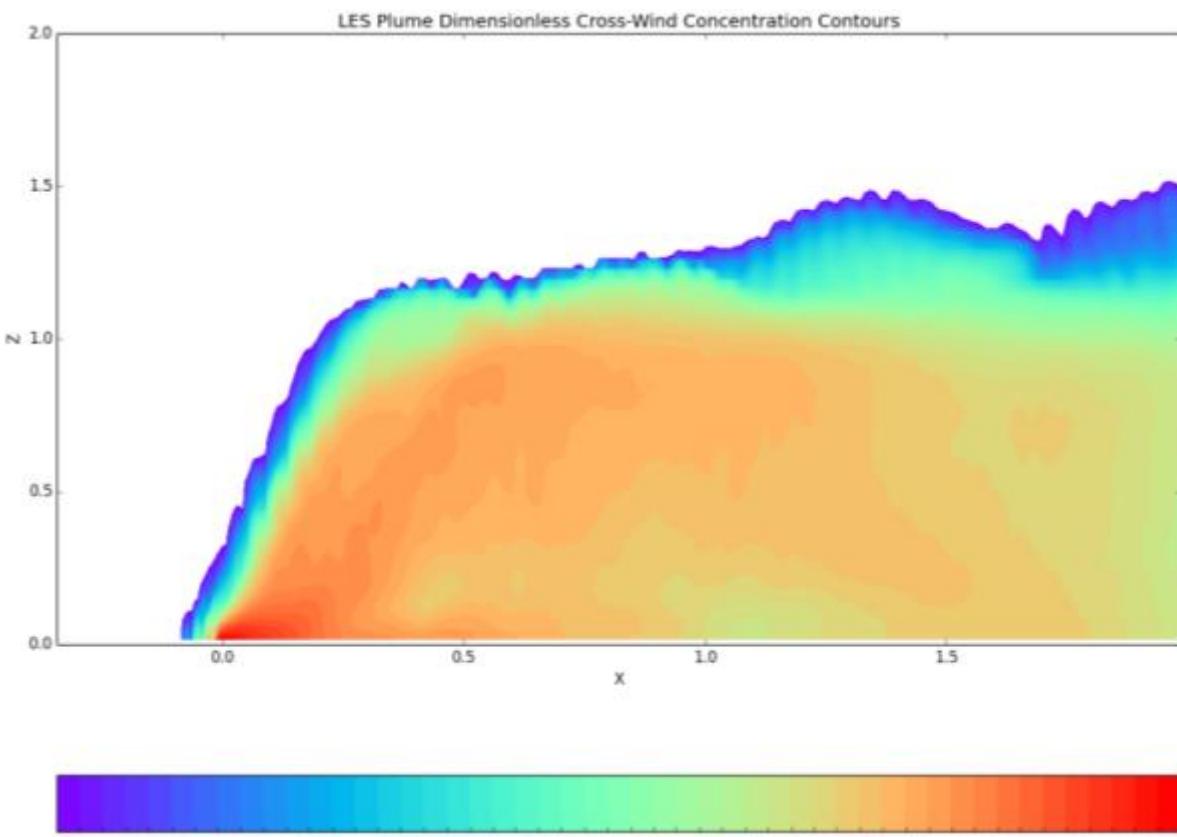
**Figure 3 Relative cross wind integrated concentrations of an effluent plume released at the surface in the UCD50B simulation. The data are averaged over 15 minutes of simulation time.**
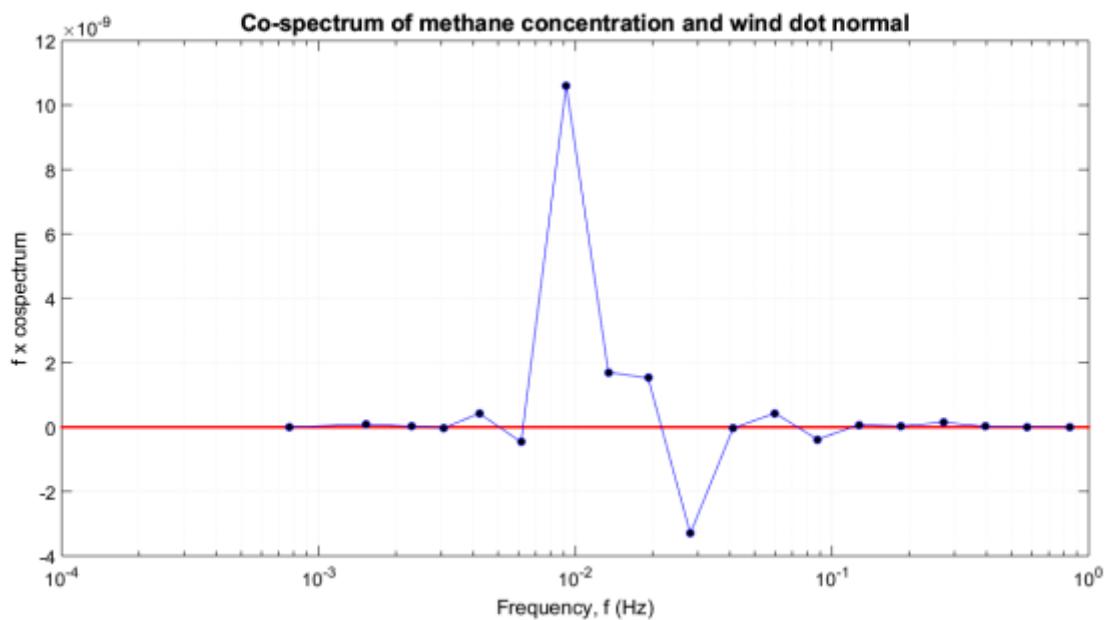
**Figure 4 Average cospectrum of the outward directed component of the observed wind and the methane concentration from 70 laps around a point source near San Antonio, Texas. The peak at $10^{-2}$ Hz corresponds to the period of the circle.**
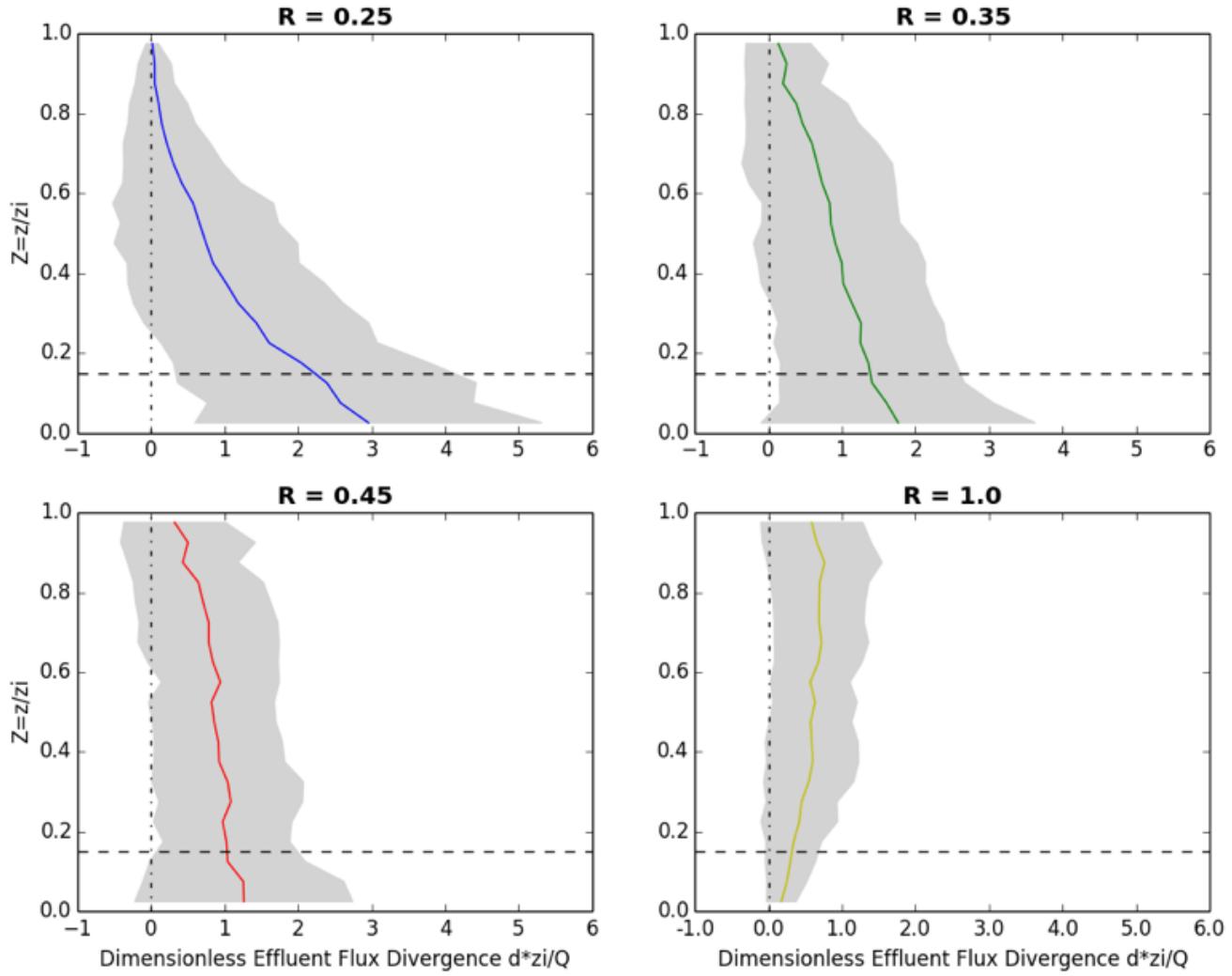
**Figure 5 Dimensionless flux divergence profiles generated from averaging over 3 different WRF-LES runs using 30 time steps for each one. The horizontal flux per unit altitude $(d = F/\Delta z)$ is normalized by the boundary layer height, $z_i$, and source strength, $Q$. The colored profiles are averages at various dimensionless distances, $R$=0.1, 0.2, 0.3, and 0.4 and the gray areas represent one standard deviation about the mean. The horizontal dashed lines are the approximate lowest safe flight altitude.**
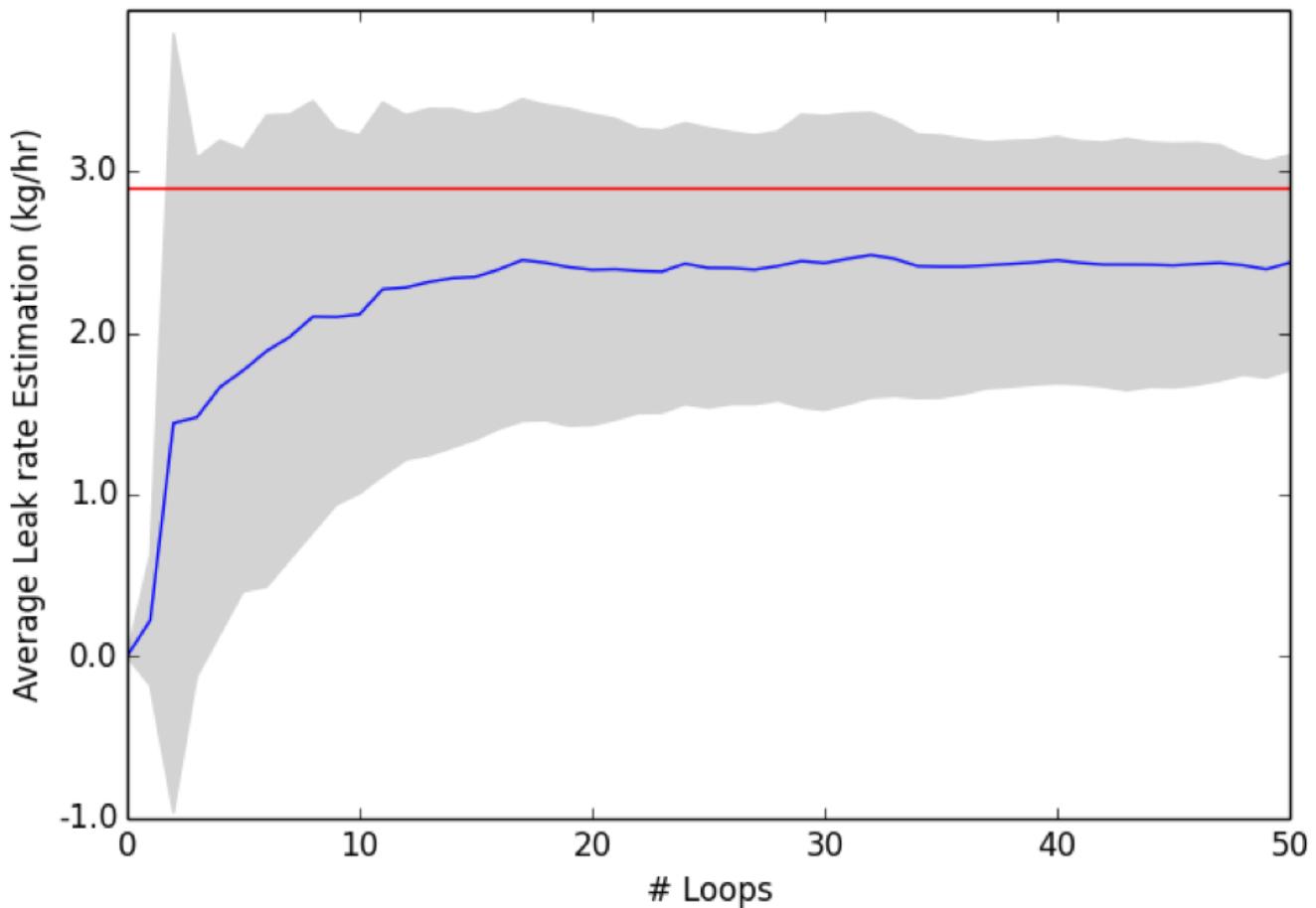
**Figure 6 Rate of convergence toward the final leak rate estimation as a function of the number of loops for LES CASE UCD508. By 15 laps, the emissions estimate (blue line) has stabilized to 2.5 kg hr$^{-1}$ compared to the actual leak rate (red line) of 2.9 kg hr$^{-1}$. Dimensionless distance R = 0.25, 50 realizations. Grey area represents 1 standard deviation.**

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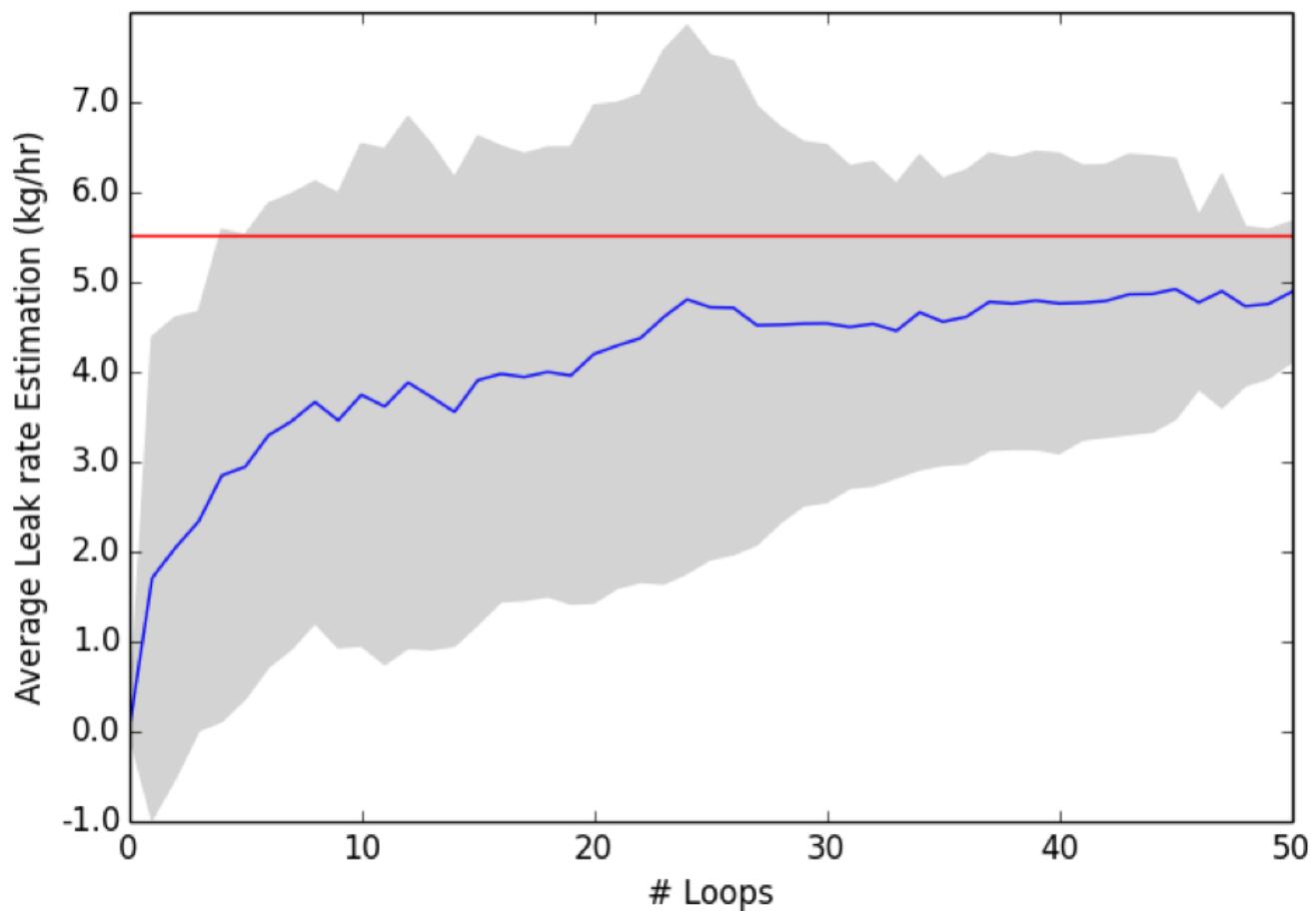
**Figure 7** **Averaged LES estimates for the Aerodyne case. This leak shows a slightly higher number of laps before convergence (~25 laps). This simulation was performed using the conditions for the Aerodyne controlled release near Denver, Colorado on November 19, 2014.**
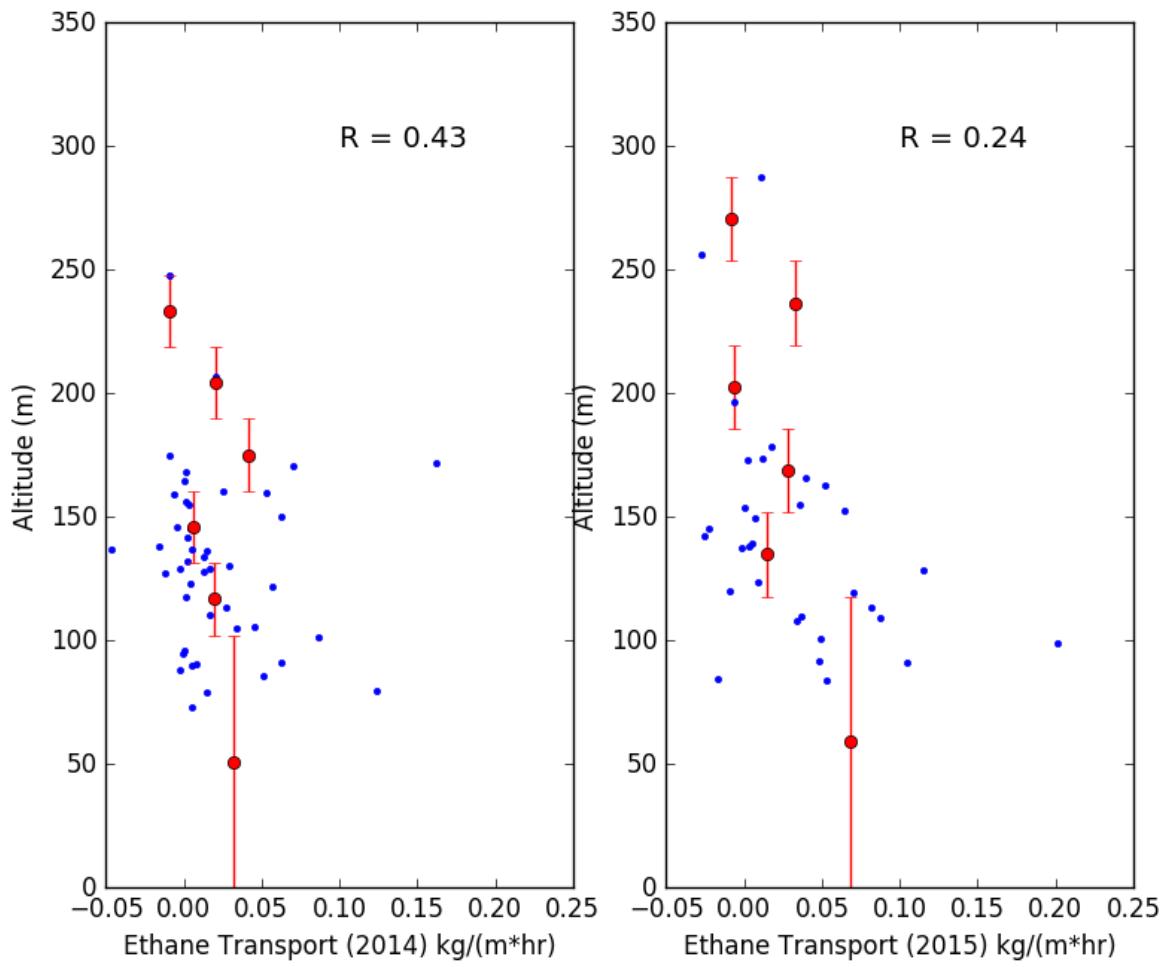
**Figure 8 - Ethane horizontal transport profiles for the Aerodyne controlled releases near Denver, Colorado on November 19, 2014 (left) and in Bee Branch, Arkansas on October 3, 2015 (right). Blue dots represent individual flight loop measurements and the red circles represent the bin average values for altitude intervals represented by the red bars.**
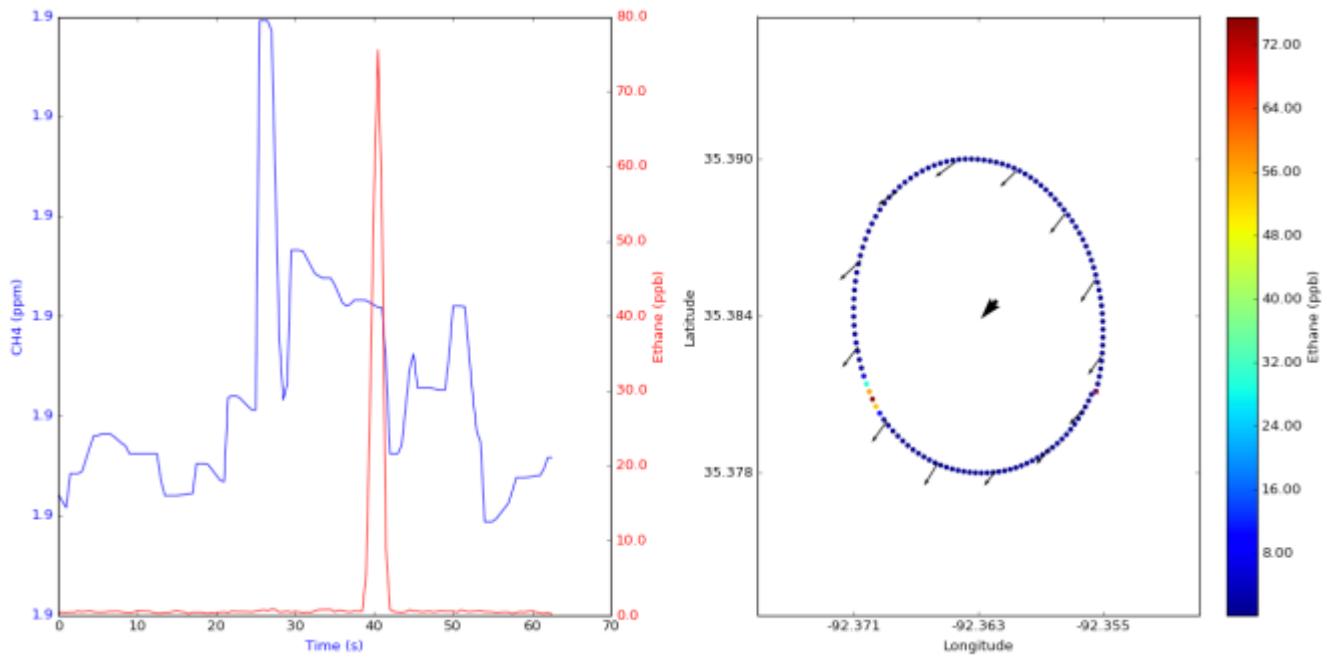
**Figure 9 (Left plot) Time series of methane (blue) and ethane (red) along with (right plot) the geographic distribution of ethane (colorbar) and instantaneous winds (arrows) from a single flight loop during the second ethane controlled release.**
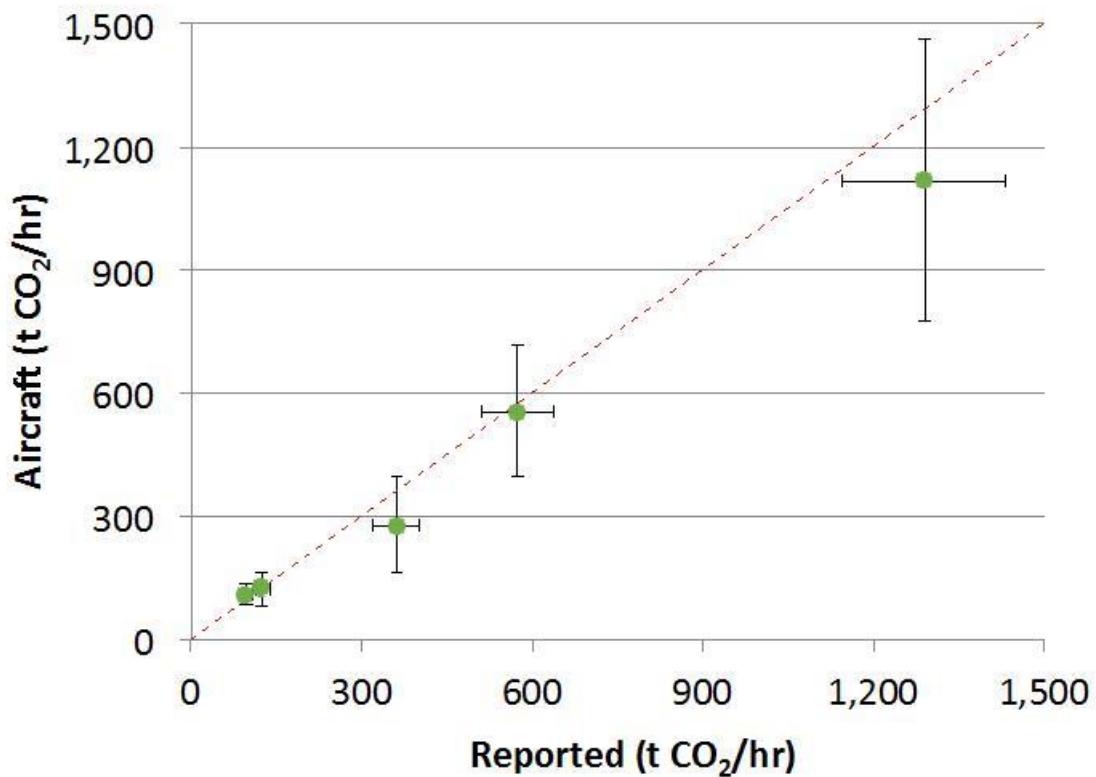
**Figure 10 Comparison of aircraft versus reported power plant emissions.**

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