P2.12 **FETCH AND FOOTPRINT CONSIDERATIONS DURING MEASUREMENT OF TRACE GAS EMISSIONS: A REFINERY LANDFARM CASE STUDY**

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1. INTRODUCTION

In the landfarming process, degradable wastes are applied to the soil surface and cultivated into the upper soil layer. Biodegradation by indigenous soil microorganisms is considered to be the primary route of waste reduction, however, volatilization, leaching and adsorption have also been found to reduce contaminant concentrations.

Volatile hydrocarbons released from landfarm soils through volatilization can impair local air quality through the production of ground level ozone in urban areas or through the release of odourous or irritating compounds. Emissions data are required to assess the impact of landfarm facilities, however, there are few emissions data available. To this end, a field study examining CO_2 and total hydrocarbon (THC) emissions was performed at a refinery landfarm. The geometry of the site was such that there was limited fetch in some wind directions. To maximize the amount of flux data collected, both flux gradient (FG) and aerodynamic mass balance (AMB) micromet approaches were used to estimate trace gas fluxes during active landfarm operations. Through examination of the resultant THC fluxes and the use of an analytical footprint model, an analysis of fetch requirements is presented here and used to illustrate the strengths and weaknesses of the two methods.

2. FLUX MEASUREMENT METHODS

2.1 *Concentration Measurement*

With both the FG and AMB techniques, a concentration profile of the trace gases of interest is required above the soil surface; therefore data collection for both techniques can be simultaneous, if desired. A combined system is depicted in Fig. 1. The FG method sampled at two heights, while the AMB method sampled at four. Details of the FG method and instrumentation are provided in Ausma et al. (2001). High frequency sampling of the in-house constructed FID-based total hydrocarbon detector (THD) enabled resolution of very small concentration differences. The detection limit of the THD, defined by the RMS noise, was 175 µgC m^{-3}.

2.2 *Flux Gradient (FG) Method*

The FG method is well suited for situations where a large uniform fetch is available. With this method, turbulence over a surface is assumed to be horizontally homogeneous and the flux is calculated from the product of the vertical concentration gradient and the eddy diffusivity. For the FG method a vertical trace gas flux can be described by the following flux gradient relationship:

$$Flux = -K\frac{\Delta C}{\Delta z} \quad (1)$$

where K (m^2 s^{-1}) is the integrated trace gas eddy diffusivity, and $\Delta C/\Delta z$ (µgC m^{-3} m^{-1}) is the vertical concentration gradient.

The minimum resolvable THC flux for the 2-intake FG system was 3.6 µgC m^{-2} s^{-1} based on half an hour of sampling and K = 0.25 m^2 s^{-1} (range of K depends on meteorological conditions and measurement height, a typical value for our site was selected).

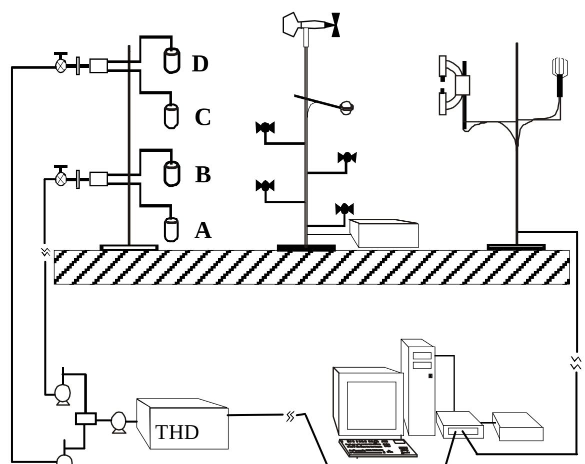

Fig. 1: Schematic of four intake flux gradient/ aerodynamic mass balance flux measurement system.

2.3 *Aerodynamic Mass Balance (AMB) Method*

The AMB method differs from the FG method in that fluxes are directly quantified using a mass balance approach. This technique is well suited for the measurement of fluxes from small finite sources that are surrounded by land that is not a source.

The AMB method measures the horizontal flux from an emission area using the vertically integrated product of wind speed (u [m s^{-1}]) and the concentration difference between upwind and downwind (ΔC [µg m^{-3}]) divided by the fetch length (d [m]):

$$Flux = \frac{\sum u\Delta C\Delta z}{d} \quad (2)$$

where Δz (m) is the thickness of the air layer that the

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mass balance is applied to. This method assumes that the volume defined by the mass balance is well mixed and that the system is at steady state. It also assumes that the measured trace gases are conservative. ΔC is measured between a sampling point above the soil surface (height = z) and the background. The background can either be measured off-site, or on-site at a sufficiently high height to represent background concentration levels. The AMB method was applied periodically during the field study when fetch was limited in several wind directions. Trace gas concentration was measured at 4 heights, with the uppermost height selected to represent the concentration of the incoming ambient air.

d was calculated with the aid of a vane anemometer and the dimensions of the field. Since wind angles are half-hour averages some error is introduced by calculating d using this method. However, the component of this error due to wind variation is for the most part small depending on field geometry. Usually variations in wind angle are not large in stationary conditions (i.e., 10 to 15 degrees).

The minimum resolvable flux using the AMB method depends upon the wind-speed profile and the fetch. Using a typical profile of 4.5, 6.1 and 6.9 m s^{-1} and a fetch of 35 m, the minimum resolvable THC flux would be 3.7 μgC m^{-2} s^{-1}. This is based upon the RMS system noise and the number of concentration measurements used in each ΔC calculation.

2.4 *Site and Field Study Description*

During the fall of 1999, a field study was performed at a refinery landfarm in south western Ontario. The goal of this study was to obtain a time series of THC fluxes from an intensively used refinery landfarm. Data were collected between 25 October (DOY 298) and 3 November 1999 (DOY 307). The available spreading area was 4.4 hectares composed of 8 flat fields of variable dimensions laid out in a north/south direction (Fig. 2). Flux measurements were performed on a 30 by 140 m field in the northwest corner of the facility. A field of the same size and which experienced the same treatment schedule as the monitored field was situated to the south.

Three masts supporting intakes and micromet instrumentation were placed 3.5 to 6 m from the eastern edge of the field and 30 m from the northern edge. Intakes were mounted 0.24 (height A), 0.56 (height B), 0.83 (height C) and 1.1 (height D) m above the soil surface (Fig. 1). The soil surface had the roughness of a frequently cultivated field (z_0 = 3 mm).

An oily liquid waste pond was part of the landfarm facility and was located approximately 100 m to the southwest of the measurement field. The facility was in an open area surrounded by low-lying vegetation. The surface of the landfarm was flat, offering ideal micromet conditions. Refinery operations were located about 1 km to the northeast of the landfarm.

Due to odour complaints, no waste was applied during August and September. Intensive daily waste application and site cultivation resumed on 22 October 1999 (DOY 295). During the study, oily liquid wastes were applied by subsurface injection to a depth of approximately 15 cm at a daily rate of 0.9 kg m^{-2} between Oct 22 (DOY 295) and Oct 30 (DOY 303), and 1.3 kg m^{-2} on Oct 31 (DOY 304) and Nov 1 (DOY 305). Digester waste was applied to the soil surface to supply microbes at a daily application rate of 0.8 kg m^{-2}.

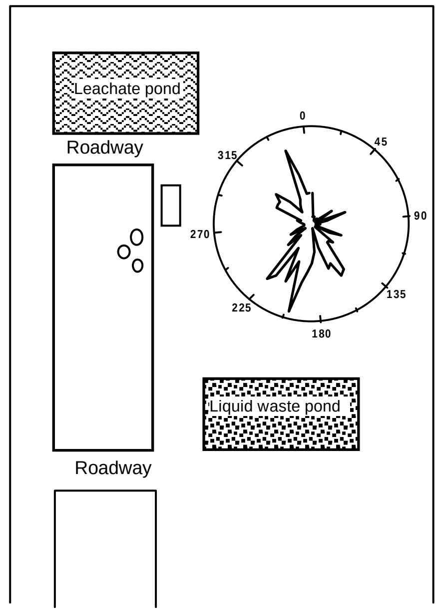

Fig. 2: Layout of the refinery landfarm. Small circles indicate mast locations. Superimposed to right of field is a plot showing predominant wind directions during the study.

Daily, between DOY 295 through 305, 3700 to 5600 kg of waste were applied to the soil subsurface, the field was cultivated 3 to 4 times, and 3400 kg of digester waste were applied. There was no landfarm activity on DOY 306 and 307 due to moderate rainfall. Flux measurements were concluded on DOY 307. Temperatures were warm until DOY 306 when they plummeted (Fig. 3).

3. RESULTS

Fig. 3 presents THC flux estimates using both the FG and AMB methods. Flux values are reported as half-hour averages in THC flux units of μgC m^{-2} s^{-1}. Positive fluxes represent emissions from the soil surface to the atmosphere.

The presence of the oily liquid storage pond, an advective source which could influence flux measurements, limited wind directions with usable data. There was also limited fetch in several other wind directions. Data were filtered to only include measurements between 180 and 360° where the fetch was adequate. FG measurements were taken between two different levels (Fig. 3). Between DOY 298 and 302 flux calculations were primarily performed between intakes B and C (+ symbol). The intake support mast during this time was situated on soil that had not been spread with waste for several months. The concentration measured by intake A was influenced by the low-emitting soil nearby and for up to 5 m of the fetch in some wind directions: using intakes A and B in this scenario would have resulted in an underestimation of the flux. On DOY 302, the intake mast was relocated several meters westward and placed on soil which had received intensive waste application for several days. Flux measurements were taken primarily between intakes A and B (x symbol) from DOY 302 onwards. AMB calculations were performed on data collected between DOY 298 and 302 when the 4-intake system was operational, and for a short period of time on DOY 305. Daily THC fluxes peaked between 45 and 300 μgC m^{-2} s^{-1} on spreading days. During periods of non-activity (i.e., early morning of DOY 300 and 304 and night of DOY 299 and 305) fluxes declined from peak daytime values to levels between 1 and 20 μgC m^{-2} s^{-1}. THC fluxes dropped late on DOY 306 to levels fluctuating between –10 and 10 μgC m^{-2} s^{-1} when the weather was rainy and temperatures dropped to under 5 °C.

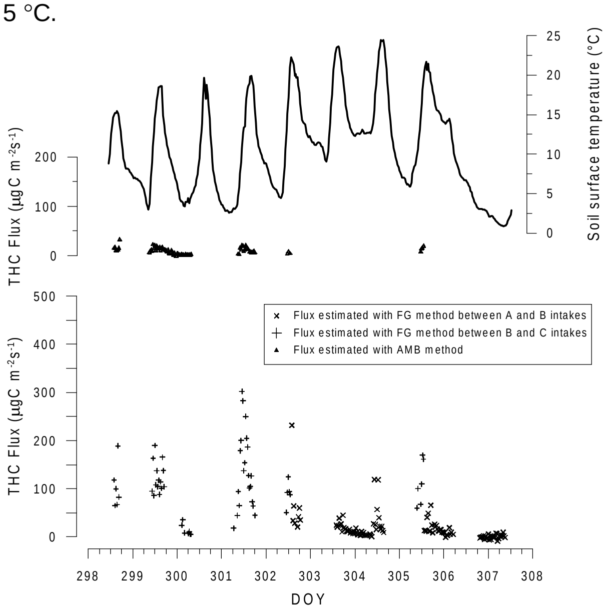

Fig. 3: Fluxes calculated using FG (lower) and AMB (upper) methods. + measurements between B and C intakes. x measurements between A and B intakes. Temperature superimposed on upper scale.

The measurements presented in Fig. 3 were collected continuously without interfering with routine facility operations. Thus, measurements were made during activities that resulted in maximum fluxes of trace gases from the landfarm surface. The FG and AMB methods provided a time series of fluxes which were used to monitor changes in emissions as a result of landfarm manipulations such as waste application and cultivation.

3.1 *FG and AMB Methods Comparison*

The FG and AMB methods provided different estimates for the THC fluxes. The two methods were simultaneously applied between DOY 298 and 302, and for a few hours on DOY 305. Typically, the AMB THC flux estimates were at least 50% smaller than the FG estimates. The two techniques have different fetch requirements: the AMB method requires a limited and well-defined fetch while the FG method requires an extensive fetch. If one examines the theoretical footprints and the actual available fetch then perhaps the observed differences in flux measurements can be explained.

The footprint of emissions observed by each intake depends not only on the sampling height but also on atmospheric conditions and site geometry. Fig. 4 shows the actual fetch along with the theoretical footprint (*x*) responsible for 85% of the fluxes (*CF*) observed at each height as determined through a stability corrected analytical footprint model described in Wong (1999):

$$CF = \exp^{\frac{-Uz_*}{u_* kx}} \qquad (3)$$

where U is the average windspeed, z_* is the effective measuring height, k is von Karmen's constant and u_* is the friction velocity.

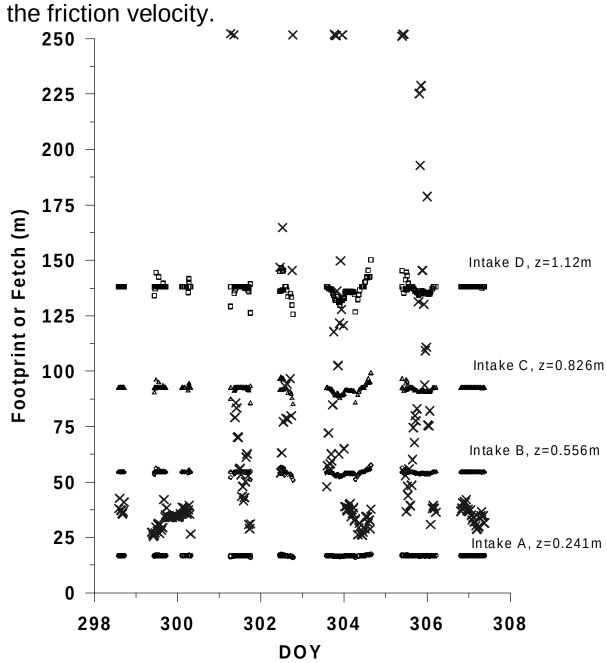

Fig. 4: Footprint of each intake (A, B, C, D) superimposed by actual fetch (x) as calculated using vane anemometer and field dimensions.

If the flux measurement scenario was one in which the fetch was shorter than the footprint, an overestimation of the flux would have occurred when using the FG method since the landfarm was surrounded by a non-emitter: the air sampled by the higher intake would have been more representative of conditions upwind of the landfarm. In the case of THC emissions, which were estimated by measuring *ΔC* between intakes B and C, this effect was observed. The footprint observed by intake C was approximately 85 m in length while the actual available landfarm surface for emissions between DOY 298 and 301 was 25 to 40 m in length. The THCs measured at this intake would have been strongly influenced by emissions from beyond the landfarm area, i.e., uncontaminated soil. This would lead to an overestimation of the THC FG flux. Under this scenario, the AMB method would provide a better estimate of the flux.

Alternatively, the FG method provides better flux estimates when the fetch is longer. For the AMB method to provide reliable estimates of the flux, the concentration measured by the upper intake must be representative of background levels. If we examine the theoretical footprint observed by intake D (Fig. 4), assigned to supply the representative background concentration, it is noted that frequently a large portion of the flux, as denoted by the fetch, was derived from the landfarm. In these cases, the AMB method would have underestimated the flux and the FG method provided better estimates since the source of hydrocarbons measured at both intakes was largely emitted by the landfarm. This is evident on DOY 302 and 305 when fetches were long and the AMB flux estimates were at least 50% less than the FG estimates.

An assumption made with the AMB application here, is that air sampled by the uppermost intake was reflective of downwind conditions. This assumption is dependent upon wind direction; if the fetch was too long, the uppermost intake sampled air influenced by landfarm emissions. This can be checked by examining ΔC between the uppermost pair of intakes (C and D) and comparing it to ΔC measured by intakes B and C. If the magnitude of ΔC between intakes C and D was small relative to the ΔC between B and C, then it can be stated with confidence that the air sampled by intake D was reflective of downwind conditions. For the AMB results, the ΔC between intakes C and D was an average of 30% to 60% of ΔC between intakes B and C. This suggests that the air sampled at level D was at times influenced by landfarm emissions. This can be avoided by sampling ambient air at a higher height or downwind of the landfarm.

The graphical presentation of fetch and footprint data together, as in Fig. 4, is a simple visual technique to quickly assess if measurements are influenced significantly by the available fetch. From the data presented in Fig. 3, Fig. 4 can be used to create a single set of THC flux values using the most appropriate micromet method (FG or AMB) for the fetch conditions during the measurement period. Fig. 5 contains a single time series of flux data that was ultimately used to evaluate emissions from the landfarm.

One of the issues dealt with during each field study is the appropriate placement of sampling intakes and instrumentation. Since the flux observed by the intakes is entirely dependent on the wind direction and the available fetch, poor intake siting when there is adequate fetch only in limited wind directions can result in the loss of significant data if the wind direction is not ideal. A flux measurement system that incorporates both the FG and AMB techniques would provide the largest time series of flux estimates. Minimal data would be discarded due to poor wind direction.

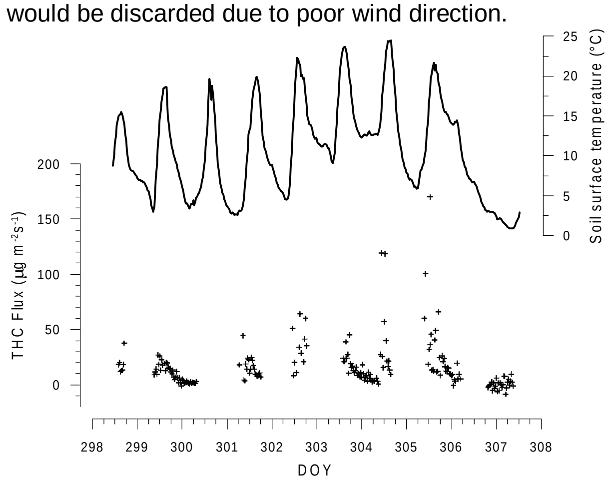

Fig. 5: Final version of flux time series created from time series in Fig. 3 and using data in Fig. 4 to optimize.

4. CONCLUSIONS

Flux gradient (FG) and aerodynamic mass balance (AMB) micromet techniques were successfully used to measure THC fluxes at a refinery landfarm during daily refinery operations. Data were collected continuously without interfering with routine facility operations, allowing measurements to be made during activities that resulted in maximum fluxes of trace gases from the landfarm surfaces.

Flux estimates from the FG and AMB methods were compared and their strengths and weaknesses were illustrated using footprint analysis. The site geometry typically encountered at refinery landfarms, i.e., limited fetch in some wind directions, leads to the recommendation that a combined system using both FG and AMB flux calculations would result in a maximized time series at facilities such as this. By measuring a 4-level concentration gradient, in conjunction with wind speed and turbulence measurements, both methods can be applied and fluxes from wind angles without advective sources can be utilized.

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