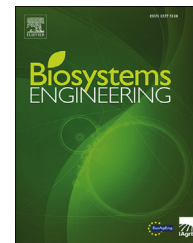




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Research Paper

Development of passive flux samplers based on adsorption to estimate greenhouse gas emissions from agricultural sources



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Passive flux samplers (PFSs) packed with adsorbents are used to estimate gaseous emissions. A key condition of their use is maintaining a linear relationship between internal and external air velocities. The performance of PFSs designs depends on the characteristics of the adsorption bed and on the sampler design. The parameters required to enable PFSs to estimate greenhouse (GHG) emissions from agricultural sources were studied. The effect of the particle size of the adsorbent used as collector medium was analyzed theoretically using the Ergun equation. Three orifice plates with 0.5, 0.7 and 1 mm bore diameter were evaluated in order to determine the most appropriate diameter to control air flow through a new passive flux sampler (PFS) prototype while maintaining adequate linearity between internal and external air velocity. The effect of the adsorbent bed thickness (19, 50, 100 and 200 mm) on the internal-external air velocity relationship in the PFS was evaluated. The best performance was obtained using the 0.7 mm orifice plate and an adsorbent bed thickness of 50 mm. Spherical adsorbents with high adsorption capacity are recommended in order to decrease the adsorbent bed thickness and improve sampling performance. A series of experiments showed that the estimated mass flow obtained by the developed PFS was close to the confidence interval of values obtained by direct detection. Thus, the developed PFS can be used as a tool for the estimating of GHG emissions from agricultural sources.

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Nomenclature

Symbols

L	Adsorbent bed thickness (m or mm)
ρ	Air density (kg m^{-3})
Q_{in}	Airflow rate through the passive flux sampler (ml min^{-1})
v_o	Air velocity in the orifice plate (m s^{-1})
$A_{orifice}$	Area of the orifice plate (m^2)
\mathcal{E}	Bed porosity
D	Diameter of the pipe (mm)
C	Discharge coefficient dependent on Reynolds number
C_D	Drag coefficient
F_D	Drag force on the passive flux sampler (N)
μ	Dynamic viscosity of the fluid ($\text{kg m}^{-1} \text{s}^{-1}$)
Y	Expansion factor
V_{ex}	External velocity (m s^{-1})
K	Internal-external velocity ratio or sampler proportionality constant
V_o	Internal velocity (m s^{-1})
F	Mass flux ($\text{g m}^{-2} \text{s}^{-1}$)
m	Mass of the target gas collected in the passive flux sampler (g)
C_o	Orifice plate constant
d	Orifice plate diameter (mm)
D_p	Particle diameter (m)
ΔP	Pressure drop (Pa)
ΔP_o	Pressure drop across the orifice plate (Pa)
ΔP_D	Pressure drop downstream of the passive flux sampler (Pa)
A_p	Projected area of the passive flux sampler with respect to the air flow direction (m^2)
r	Radius of the internal orifice (m)
β	Ratio between orifice and tube diameter (d/D)
Δt	Sampling time (s)
v_s	Superficial velocity or velocity in the empty tube (m s^{-1})

Abbreviations

NH_3	Ammonia
GHG	Greenhouse gas
N_2O	Nitrous oxide
CH_4	Methane
PFS	Passive flux sampler
PFSs	Passive flux samplers
PVC	Polyvinyl chloride

order to estimate emissions accurately, it is necessary to use precise, reproducible, robust and economic methods (Berndt & Tomkins, 2013; FAO, 2014). In this context, passive flux sampling has been recommended as a robust technique for measuring gaseous emissions because it needs few operational requirements and needs low capital investment (Godbout, Phillips, & Sneath, 2006; Mosquera, 2003; Scholtens, Hol, Wagemans, & Phillips, 2003). This technique has been applied to estimate NH_3 emissions by using sampler made with glass tubes coated with oxalic acid which allowed absorption of the target gas (Leuning, Freney, Denmead, & Simpson, 1985; Schjoerring, Sommer, & Ferm, 1992). For GHG emissions, the use of passive flux samplers (PFSs) has high potential, but studies on their application are rare. The PFSs normally used for estimating emissions consist of a packed tube containing an adsorbent as a collector medium. This is based on two principles: 1) the aerodynamic behavior which requires that the air velocity inside the PFS is proportional to the velocity of the air surrounding the sampler, and 2) the adsorption capacity of the collector medium to capture the gas sample. The adsorption capacity generally depends on the air flow rate passing through the sampler, the gas concentration in the inlet air, and the mass of the adsorbent used (Godbout et al., 2006; Larios et al., 2017). In previous research, several adsorbents used to capture N_2O and CH_4 were evaluated. For example, Godbout et al. (2006) found that zeolite 5A in powder form was found the best adsorbent to collect N_2O compared to activated carbon Carboxen 1018 and Carboxen 1021. Results for the collection of CH_4 on these three molecular sieves showed low adsorption capacity and rapid saturation of the bed. Some PFSs prototypes were developed but limitations related to the air flow through PFSs, the adsorbent mass contained in the PFSs and manufacturing costs were identified (Gaudet et al., 2005; Godbout et al., 2006; Palacios, 2010).

Depending on the adsorption capacity of the adsorbent and sampling requirements, PFSs may require long adsorbent beds to enable representative sampling times. Adsorbent beds with smaller particle size have a higher surface area relative to equivalent to the mass of adsorbents with a larger particle size (Zielinski & Kettle, 2013). However, long adsorbent beds and small particle size may generate substantial energy losses in the air flow (Wilkes, 2006) because the adsorbent bed reduces the air velocity inside the PFSs (Liu, Afacan, & Masliyah, 1994), and this consequently affects the internal (V_o), the external velocity (V_{ex}) and the ratio between the two velocities, known as the K-factor (Scholtens et al., 2003). Thus, the pressure drop (ΔP) of the air flow through a packed bed must be experimentally or theoretically evaluated. The Ergun equation is frequently used to predict ΔP . This equation expresses that ΔP is given by the sum of viscous and kinetic energy loss as a function of fluid and bed characteristics (Niven, 2002; Xu & Jiang, 2008).

However, the value of the K-factor value changes as a function of sampling design. This includes the diameter of the orifice plate placed inside the PFS, the orifice plate diameter/diameter of the pipe (d/D) ratio (β) and the adsorbent bed thickness (L). In this case, the K-factor needs to be determined experimentally. In this context, the objective of this work was to study and define the parameters determining the development of PFSs based on adsorption to estimate GHG emissions from agricultural sources. The GHG used in this study as a reference was N_2O

1. Introduction

Estimating greenhouse gas (GHG) emissions from agricultural sources encounters several obstacles. For example, there are substantial spatial and temporal variations among sources, and most of the methodologies available require complex and expensive equipment making it difficult to sample a significant number of farms (Larios et al., 2016; Wang et al., 2016). In

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