



# Site specific diel methane emission mechanisms in landfills: A field validated process based on vegetation and climate factors<sup>☆</sup>



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## ABSTRACT

Diel methane emission fluxes from a landfill that was covered by vegetation were investigated to reveal the methane emission mechanisms based on the interaction of vegetation characteristics and climate factors. The methane emissions showed large variation between daytime and nighttime, and the trend of methane emissions exhibited clear bimodal patterns from both *Setaria viridis*- and *Neyraudia reynaudiana*-covered areas. Plants play an important role in methane transportation as well as methane oxidation. The notable decrease in methane emissions after plants were cut suggests that methane transportation via plants is the primary way of methane emissions in the vegetated areas of landfill. Within plants, the methane emission fluxes were enhanced due to a convection mechanism. Given that the methane emission flux is highly correlated with the solar radiation during daytime, the convection mechanism could be attributed to the increase in solar radiation. Whereas the methane emission flux is affected by a combined impact of the wind speed and pedosphere characteristics during nighttime. An improved understanding of the methane emission mechanisms in vegetated landfills is expected to develop a reliable model for landfill methane emissions and to attenuate greenhouse gas emissions from landfills.

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## 1. Introduction

As an important greenhouse gas (GHG) with a global annual emission of 645 million tons (Carmichael et al., 2014; Miller et al., 2013), methane (CH<sub>4</sub>) accounts for 14.3% of the global anthropogenic GHG emissions. Landfills were ranked as the third largest anthropogenic CH<sub>4</sub> emission source, and CH<sub>4</sub> emissions from the waste sector are expected to reach 790 to 800 MtCO<sub>2</sub>-eq in 2015 (IPCC, 2014).

CH<sub>4</sub> emissions from landfills rapidly increase in developing countries because of rapid population growth and accelerated urbanization process (IPCC, 2014). In China, landfilling is the primary disposal method for solid waste and more than 70% of the municipal solid waste end up in landfills. Until 2014, there were 604 sanitary landfills operated in China with a daily treatment capacity of 335,316 tons (Ma, 2015). The annual landfill CH<sub>4</sub> production was estimated to be approximately 2.12 billion cubic meters (Cai et al., 2013), which

would contribute to 11%–13% of the total CH<sub>4</sub> emission in China (Lu et al., 2011). Although some large and modern landfills have installed gas collection systems, the notably low landfill gas (LFG) collection efficiency of 30% still lead to a large amount of fugitive CH<sub>4</sub> emitted into the atmosphere (Chai et al., 2011; Li et al., 2014; Zhang et al., 2010). Moreover, almost no small- and medium-sized landfills installed any LFG collection systems that CH<sub>4</sub> emissions from landfills remain unquantifiable in China.

CH<sub>4</sub> oxidation in cover soil has been well known as an efficient solution to control and mitigate CH<sub>4</sub> in the absence of LFG collection systems (Bogner et al., 1995; IPCC, 2014; Pratt et al., 2013). CH<sub>4</sub> oxidation by methanotrophs in the cover soil has attracted attention for several decades (Bogner et al., 1997; Czepiel et al., 1996; Gebert et al., 2003; Schuetz et al., 2003; Scheutz et al., 2009). CH<sub>4</sub> oxidation in landfill cover soil is mainly affected by the activity and community structure of methanotrophs, the depth and characteristics of cover soil, and climate conditions (Schuetz et al., 2003; Serrano-Silva et al., 2014). Depending on the characteristics of the cover soil and climate conditions, the CH<sub>4</sub> oxidation rate can range from negligible to 100% of CH<sub>4</sub> emissions (Gebert et al., 2011; Schuetz et al., 2003), and the steady CH<sub>4</sub> oxidation capacity can be as high as 200–250 g/m<sup>2</sup>·day (Scheutz et al., 2009).

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It is common that native plants begin to cover the landfill surface after landfills are closed for 0.5–1 year, and the coverage rates may reach 60%–70% when a landfill has been closed for four years (Chai et al., 2010). Previous reports showed that vegetation served as an important CH<sub>4</sub> emission source, which annually contributes 32 to 143 million tons of CH<sub>4</sub> to the global GHG that represents up to 22% of the annual CH<sub>4</sub> emissions (Carmichael et al., 2014). In wetlands, as one of the largest CH<sub>4</sub> emission sources generally with high vegetation coverage, the role of vegetation in CH<sub>4</sub> generation, transportation and oxidation has been widely investigated (Ding et al., 2004; Kim et al., 1998; Kaki et al., 2001; Van der Nat et al., 1998; Wang and Han, 2005; Xu et al., 2007). The diel variation of CH<sub>4</sub> emission in plants generally show the one peak pattern (Ding et al., 2004; Van der Nat et al., 1998; Wang and Han, 2005). On one hand, plants can accelerate the CH<sub>4</sub> oxidation because the plant can transport O<sub>2</sub> into the pedosphere. On the other hand, plants can stimulate CH<sub>4</sub> emissions via their gas conduits, which are mainly aerenchymatous structures. In addition, plants are the key player to result in varied diel CH<sub>4</sub> emissions and can transport 29%–90% CH<sub>4</sub>, depending on the plant species and growth environments (Ding et al., 2004; Kim et al., 1998; Kaki et al., 2001; Van der Nat et al., 1998). Some plant species, such as *Phragmites australis* and *Typha domingensis*, which generally display more obvious diel variations in CH<sub>4</sub> emissions, can use convective gas flow in addition to diffusive flow (Kim et al., 1998; Van der Nat et al., 1998). Moreover, it was shown that vegetated areas have lower internal CH<sub>4</sub> concentration than non-vegetated areas of a landfill (Chai et al., 2010), and the physico-chemical properties of the cover soil appear to be altered by the vegetation growth. Advanced biotic CH<sub>4</sub> mitigations in cover soil can be expected. The above facts indicate that the effect of vegetation on CH<sub>4</sub> oxidation and emission should be well clarified (Abichou et al., 2015; Huber-Humer et al., 2008; Ndanga et al., 2015).

It has been widely reported that various climate conditions such as solar radiation (SR), temperature and wind speed affect both vegetation growth and CH<sub>4</sub> emissions (Kim et al., 1999, 1998; Nakano et al., 2000; Wang and Han, 2005). CH<sub>4</sub> emissions are enhanced with increasing SR by changing the transport mechanism from density-driven to pressure-driven, particularly in vegetated areas (Van der Nat et al., 1998). Efficient pressure-driven CH<sub>4</sub> emissions, namely convection, can be attributed to the changing pressure, soil water content and wind speed. Therefore, it is incomplete to clarify the CH<sub>4</sub> emission mechanism in vegetated landfills without considering the climate conditions because of the close relationship between the plant physiology and climate conditions.

While vegetation plays an unneglectable role on CH<sub>4</sub> transportation and oxidation in landfills, to date, the diel CH<sub>4</sub> emission patterns in vegetated landfills have not been well investigated, and less effort has been made to clarify the effect of plants on CH<sub>4</sub> emissions in landfills. The objective of this study is to investigate the combined effects of vegetation characteristics and climate conditions on CH<sub>4</sub> emissions, and to clarify the diel CH<sub>4</sub> emission mechanisms based on the interaction between vegetation characteristics and climate conditions. An improved understanding of the CH<sub>4</sub> emission mechanisms in vegetated landfills will be meaningful for developing a CH<sub>4</sub> emission model and for attenuating GHG emissions in landfills.

## 2. Materials and methods

### 2.1. Site description

The experiments were conducted in a demonstration area at the Shanghai Laogang Landfill (N31°03'13.4", E121°53'52.9"), which is one of the largest landfills in China with a daily capacity of 6000–8000 tons. The demonstration area, an anaerobic cell, had

been closed for approximately 3.5 years, and was equipped with a passive LFG collection system. The depth of the cover soil in the area was approximately 0.4 m, which was served as a final cover. The total area of the investigated area was 50,000 m<sup>2</sup>, and the surfaces were partially covered by vegetation, where five plant species had naturally grown. According to phyto-sociological methods of the Braun-Blanquet School, only two species, *Setaria viridis* (*S. viridis*) and *Neyraudia reynaudiana* (*N. reynaudiana*), were considered as the dominant species and investigated in this study (Chai et al., 2011).

### 2.2. LFG flux

The LFG flux was measured using the static chamber method in vegetated areas of landfill (Rolston, 1986). The chambers were placed in areas that were covered by *S. viridis* and *N. reynaudiana*. LFG flux measurements in *S. viridis*- and *N. reynaudiana*-covered areas were conducted over an entire day (24-h period). To evaluate the effect of vegetation on the LFG flux, the plants were cut and then the comparison experiments were conducted over the next day (24-h period) at the same site. The plants that had been cut were taken back into the chamber to avoid the difference due to the light conditions, and the cut sections of the plants were sealed with Vaseline to prevent gas emissions via the plants (Fig. 1).

The chambers used in this study are 1-m-diameter and 0.50-m-high cylindrical Plexiglas. The upper surface of the chamber has two holes with 0.03-m-inner-diameter. One hole connected to sampling syringe and another connected to a 2-L-bag that was equipped to keep pressure balance in chamber during a measurement. In addition, a fan was used inside the chamber circulating gas to keep well-mixed conditions. The LFGs were sampled every 20 min using a 30 mL syringe, and five samples were taken as one set for calculating an LFG flux. After each flux measurement (one set of sampling), the chamber was evacuated for the next measurement. CH<sub>4</sub> and O<sub>2</sub> concentrations were later analysed in the laboratory by injecting sample gas into the gas chromatograph that was equipped with a TCD detector and a molecular sieve column (Micro GC 3000, Agilent, USA). The concentration of CO<sub>2</sub> was separated with a Plot U column. Each sample was measured at least twice, and the average value of these measurements was used to calculate the LFG flux. The LFG flux was calculated using linear regression based on the concentration change as a function of time for the five samples. The LFG flux (mol/m<sup>2</sup>·h) was calculated as  $A \times B$ , where A is the height of the chamber (m) and B is the slope of the LFG concentration (mol/m<sup>3</sup>) vs. time (h).

### 2.3. Climate conditions

The climate conditions, which include SR, atmospheric temperature (AT), soil temperature (ST) at 15 cm depth under the soil cover surface, and wind speed, were simultaneously recorded with the LFG flux measurements by setting up a portable weather station (Global Water IIIB, A Xylem brand, USA).

### 2.4. Statistical analysis

Correlation analyses were performed using the Pearson method in GraphPad Prism 5.0 (GraphPad Software, Inc., USA). In this analysis, the tested factor was considered statistically significant if  $p < 0.01$  in a two-tail analysis.

## 3. Results

### 3.1. Diel CH<sub>4</sub> emissions in vegetated landfills

Diel CH<sub>4</sub> emissions from both *S. viridis*- and *N. reynaudiana*-

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