Landfill Methane Oxidation: Predictive Model Development

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ABSTRACT
Methane (CH₄) emissions from landfills are continually increasing due to population growth and growing per capita waste generation. Microbial CH₄ oxidation in landfill cover soils might provide a means of controlling CH₄ emissions. This study proposes an Artificial Neural Network (ANN) approach to predict CH₄ oxidation in sandy landfill cover soils based on the soil moisture content, soil temperature and oxygen (O₂) concentration at a depth of 10 cm in the cover soil. The optimum ANN model giving the lowest Mean Square Error (MSE) was trained with the Levenberg-Marquardt algorithm and comprised of three layers, with 50 and 20 neurons at the first and the second hidden layers, respectively and a logistic sigmoid (logsig) transfer function between the hidden and output layers. This study revealed that the ANN oxidation model can predict CH₄ oxidation with a MSE of 0.001475 and a coefficient of determination (R²) between the measured and predicted outputs of up to 0.91. In conclusion, the ANN oxidation model provides an effective tool for predicting the percentage of CH₄ oxidized in landfill cover soil.

Keywords: Landfill cover soil, methane oxidation, air hitam, methane emission, neural networks

INTRODUCTION
Disposal of Municipal Solid Waste (MSW) in landfills generates large amount of greenhouse gas (GHG) emission. The decomposition of MSW under anaerobic conditions produces landfill gas (LFG) containing approximately 50-60% methane (CH₄) and 30-40% carbon dioxide (CO₂) by volume. The CH₄ has a global warming potential 25 times greater than that of CO₂ and thus CH₄ emissions to the atmosphere have adverse effects on the environment. The migration of CH₄ gas from landfills to the surrounding environment can potentially affect human life. The CH₄ emissions from landfills are continually increasing due population growth and growing per capita waste generation with landfills ranking as the third-largest anthropogenic CH₄ source after rice paddies and ruminants (Ritzkowski and Stegmann, 2007; Qingxian et al., 2007).

Surface LFG emissions vary over space and time due to biological, chemical and physical processes occurring within landfill cover soils. Microbial CH₄ oxidation in landfill cover soils might provide a means of controlling CH₄ emission. Previous studies have reported that the CH₄ oxidation process in landfill cover soil is efficient at reducing CH₄ emissions (Abushammala et al., 2013a, b; Hilger and Humer, 2003; Huber-Humer, 2004; Huber-Humer et al., 2008; Stern et al., 2007). Laboratory and field studies have shown that the CH₄ oxidation capacity of landfill cover soils varies between 0 and 100% (Abushammala et al., 2014; Jugnia et al., 2008).

Several attempts to model CH₄ oxidation in landfill cover soils have been made previously. Most of the current models are analytical and based on fundamental scientific laws governing oxidation processes (De Visscher and van Cleemput, 2003) while few models are based on empirical statistical modeling approaches (Albanna et al., 2007).
Artificial Neural Network (ANN) analysis is an empirical modeling approach that can determine relationships between different sets of data and an ANN is capable of solving problems where statistical models are inefficient (Zhou, 2005). ANNs are defined by Kneale et al. (2004) as “Structures which forecast and predict through pattern matching and comparison”. ANN analysis has been used in many fields for performing complex functions such as pattern recognition, classification, vision, control systems and prediction (Zilouchian, 2001). In prediction modeling, the ANN takes a set of inputs and produces one or more outputs. This task is accomplished by training the ANN on a set of input and output data that allows the ANN to gain knowledge of the dynamics of the system and use this knowledge to predict the output of the system for any given inputs (Zilouchian, 2001).

Several factors affect CH₄ oxidation in landfill cover soils. The most important factors reported in literature are soil texture, organic matter content, moisture content, temperature, pH, nutrient availability, O₂ concentration and CH₄ concentration (Boeckx et al., 1996; Borjesson et al., 2001; Spokas and Bogner, 2011; Wilshusen et al., 2004). However, climate and season also have an impact on oxidation capacity. Methane (CH₄) oxidation is expected to be less in the following environments: Subarctic or highland regions due to colder temperatures, subtropical regions due to the relatively dry conditions of cover soils and humid continental regions due to longer, colder winters and more dry summers (Onk, 2010).

The objective of this study is to develop an ANN model to predict CH₄ oxidation in a sandy landfill cover soil. Several variables, such as average air temperature (°C), average soil temperature (°C) at 10 cm depth of cover soil, average volumetric soil moisture content over soil depths (%v/v), CH₄ concentration underlying the bottom of soil cover (%v/v) and oxygen (O₂) concentration at 10 cm depth of cover soil (%v/v), were tested in order to select the main predictive variables.

**MATERIALS AND METHODS**

**Site description:** The Air Hitam sanitary landfill is located southwest of the city of Kuala Lumpur, approximately 18 km from the city center, at 3°00'05"N and 101°39'37"E (Fig. 1). The landfill has a total area of 42 ha, divided into seven phases while in operation, it received approximately 1,636 t of domestic waste per day. The landfill began receiving waste in December 1995 and was closed in December 2006. The total amount of waste in the landfill is estimated at 6,527,640 t. The Air Hitam landfill is the first landfill included in the LFG renewable energy project in Malaysia.

In this study, measurements were taken in an inactive part of the landfill (phase six) (Fig. 1). The total area of this phase is 47,500 m². It contains waste ranging from two to seven years in age. The phase began receiving waste in September 2003 and closed in January 2006. The average waste depth of the phase is approximately 30 m. The phase was covered by approximately 30-40 cm of poorly graded sand with an interim soil layer of gravel. In certain sections of the phase, vegetation was not well established during the period in which measurements were taken, while other sections contained grasses and herbs. At the time of the study, the phase did not have a gas collection system and thus the LFGs that were not oxidized escaped through the cover soil and were released to the atmosphere.

**Experimental data:** Four monitoring stations were randomly located within a 42×42 m area overlying the sixth phase of the landfill (Fig. 1) to investigate the CH₄ oxidation capacity of the landfill’s cover soil. Prior to the field investigation, disturbed and undisturbed soil samples were collected from each monitoring station and were transported to the laboratory of the Civil and Structural Engineering Department at the Universiti Kebangsaan Malaysia (UKM) for investigation of soil properties. Samples were sieved and analyzed for particle density, moisture content, bulk density, pH and organic matter. The measurements were conducted following the methods of Head (1992) and were previously presented by Abushammala et al. (2013a).

The CH₄ and CO₂ emissions at the soil surface and the soil gas concentration profiles (at depths of 10, 20 and 30 cm) at the monitoring stations were measured twice per month from October to December, 2010. All measurements were taken between 9.00 am and 11.00 pm. A square static flux chamber was constructed and used to measure CH₄ and CO₂ emissions.
following the methods described by Abushammala et al. (2012). Soil gas was trapped in pre-installed stainless steel tubes constructed as described by Kiese and Butterbach-Bahl (2002). The gas was collected using 10 mL gas-tight syringes for direct analysis. The concentrations of three main soil gases were measured: CH₄, CO₂, and O₂. To monitor the temporal variation in soil gas concentrations, two of the four monitoring stations were randomly selected for sampling once a month during the morning (8:00-10:00), midday (12:00-14:00) and afternoon (15:00-17:00) from October to December, 2010. At each monitoring station, the air temperature and soil temperature were measured at a depth of 10 cm, as well as atmospheric pressure, volumetric soil moisture and soil gas emissions and concentration profiles. The combination of surface CH₄ and CO₂ emissions and soil gas concentration profiles was used to estimate the percentage of CH₄ oxidized in the soil cover, in accordance with the method by Christophersen et al. (2001). The gas analyses were performed on a Varian Micro-GC (CP-4900) gas chromatograph (Abushammala et al., 2012).

Air temperature was measured using Skymaster, SM-28 and Speedtech instruments. The soil temperature at a depth of 10 cm was measured using a Dual Channel Type K Monarch 306 connected to two K wire thermocouple probes (Monarch Instrument, Columbia, USA). An integrated profile probe (PR2) connected to a moisture meter (H112) was used to measure the soil volumetric moisture content profile. The PR2 probe was inserted into pre-installed access tubes and measurements were taken at different depths.

**ANN modeling:** The ANN oxidation model was developed using 46 experimental data sets obtained from previous field investigations of the Air Hitam landfill (Abushammala et al., 2013a). The data sets were first normalized and then divided into an input matrix and target matrix. Methane (CH₄) oxidation (%) was used as a target. To find the appropriate input parameters for developing the ANN oxidation model, an initial ANN structure with two hidden layers comprised of 50 and 30 neurons for the first and the second layers, respectively, was used to evaluate the performance of combinations of measured variables (Karacan, 2008). The input variables of the model that provided the smallest Mean Square Error (MSE) and greatest coefficient of determination (R²) were selected as the effective input variables for predicting CH₄ oxidation (%). The measured variables were soil temperature (p₁), air temperature (p₂), soil moisture content (p₃), CH₄ concentration at the bottom of the soil cover (p₄) and O₂ concentration at a depth of 10 cm within the soil cover (p₅). The entire dataset was first randomized to prevent bias and to construct representative sections of the dataset to train and test the ANN. Subsequently, the whole dataset was divided into 34 exemplars (74%) for training data and 12 exemplars (26%) for testing.

The initial ANN type was Feed-Forward. Supervised back-propagation is the most popular training method (Rene and Saidutta, 2008) and therefore, was used for training the initial ANN using the Levenberg-Marquardt training algorithm (trainlm). The training process was repeated and the internal connection weights were modified in response to the computed error until the performance error decreased to an acceptable level. For each iteration, the error was calculated and the summed errors from overall neurons were divided by the size of the data used in the training process to estimate the average MSE. The logistic sigmoid transfer function (logsig) was selected at the hidden and output layers, as it is the most common type of the transfer function used to construct neural networks (Mjalli et al., 2007) and is commonly used in back-propagation networks (Bishop, 1995). The momentum factor of 0.92 was constant.

The back-propagation training algorithm and number of neurons in the hidden layers were investigated in order to optimize the performance of the ANN model in predicting CH₄ oxidation. The MSE and R² values were noted in the test phase to select the best predictive model.

**RESULTS AND DISCUSSION**

**Effective model factors:** Table 1 shows the performance of various models with various sets of inputs. The MSE in the testing stage for model 4 was lower than other models’ MSE, indicating that p₁, p₃, and p₅ were the most important factors affecting CH₄ oxidation. Therefore, these factors were selected as the input variables for predicting CH₄ oxidation.

**Final network parameters and structure:** Levenberg-Marquardt (trainlm) was the best back-propagation training algorithm, producing the lowest MSE and the highest R² value in the testing phase. Using 50 neurons in the first hidden layer and 20 neurons in the second hidden layer provided the minimum MSE in the testing phase and improved the performance of the neural network when dealing with new data. Figure 2 shows the optimized neural network structure.

<table>
<thead>
<tr>
<th>Model</th>
<th>Input variables</th>
<th>MSE training</th>
<th>MSE testing</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>p₁, p₃, p₄, p₅</td>
<td>9.4968×10⁻⁵</td>
<td>0.001450</td>
</tr>
<tr>
<td>2</td>
<td>p₁, p₃, p₅</td>
<td>9.3634×10⁻⁵</td>
<td>0.001626</td>
</tr>
<tr>
<td>3</td>
<td>p₁, p₄, p₅</td>
<td>9.0036×10⁻⁵</td>
<td>0.001608</td>
</tr>
<tr>
<td>4</td>
<td>p₁, p₃, p₅</td>
<td>9.5717×10⁻⁵</td>
<td>0.001475</td>
</tr>
<tr>
<td>5</td>
<td>p₁, p₄, p₅</td>
<td>9.7013×10⁻⁵</td>
<td>0.001538</td>
</tr>
<tr>
<td>6</td>
<td>p₁, p₃, p₅</td>
<td>9.9480×10⁻⁵</td>
<td>0.001803</td>
</tr>
<tr>
<td>7</td>
<td>p₁, p₃, p₅</td>
<td>9.5715×10⁻⁵</td>
<td>0.001537</td>
</tr>
<tr>
<td>8</td>
<td>p₁, p₃, p₅</td>
<td>9.9162×10⁻⁵</td>
<td>0.001640</td>
</tr>
</tbody>
</table>

Table 1: MSE of training and testing data with different set of inputs

![Fig. 2: Optimal ANN structure](image-url)

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Fig. 3(a-b): Data set distributions (a) Before and (b) After training the ANN model

Fig. 4(a-b): Iteration curve and linear regression analysis between the network outputs and the corresponding targets during the training stage

**Training and testing of the model:** The best MSE achieved during the training and testing phases were $9.9885 \times 10^{-5}$ and $0.001475$, respectively. Figure 3 shows the data distribution before and after the training phase. The network training process took 176 epochs to reach the error goal ($10^{-4}$), as shown in Fig. 4. Figure 4 also shows the linear regression between the network outputs and the corresponding targets during the training phase.

Figure 5 compares the measured oxidation in the testing data set with that predicted using the proposed ANN model and presents the error associated with each predicted value. Although the target data exhibited large variations, the ANN model outputs were reasonable and mimicked the target data. The individual errors of the proposed ANN ranged from 2.2-25% with an average of 13.15%. The prediction error was estimated using Eq. 1. The model exhibited an ability to predict the testing data with an $R^2$ of 0.91 (Fig. 6):

$$\text{Error (\%)} = \frac{\text{Actual} - \text{Predicted}}{\text{Actual}} \times 100$$  \hspace{1cm} (1)

The $R^2$ value found in this study was higher than those reported in the literature: 0.50 for the prediction of the CH$_4$ fraction of LFG (Ozkaya et al., 2007), 0.90 for the prediction of the quantitative characteristics of water bodies (Palani et al., 2008) and 0.90 for the prediction of performance of a wastewater treatment plant (Nasr et al., 2012). However, some previous studies have achieved $R^2$ values of up to 0.99
CONCLUSION

This study proposed an ANN approach to predict CH₄ oxidation in a sandy landfill cover soil in relation to soil moisture content, soil temperature, and O₂ concentration at a depth of 10 cm in cover soil. The results indicated a small MSE (0.001475) and relatively high R² (up to 0.91) between the measured and predicted values. In conclusion, the ANN oxidation model provides an effective tool for predicting the amount of CH₄ oxidation in landfill cover soils. However, the model cannot be used with different cover soil properties or input values outside the range of the data used to train the model.

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