

Reliability Evaluation for Static Chamber Method at Landfill Sites

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Abstract: In this study, COMSOL Multiphysics was used for the reliability evaluation for static chamber method at landfill sites. Static chamber method, which measures landfill gas emission fluxes, is widely used at landfill sites for the monitoring of greenhouse gas emission such as methane and carbon dioxide. The accuracy and the reliability of static chamber method are dependent on the measuring condition and the magnitude of the gas flux emitted from a landfill site. To investigate their effects on the accuracy and reliability of static chamber method, this study proposed the simulation model for the multicomponent gas advection-diffusion-reaction model, and showed the following findings. In order to accurately measure gas flux by static chamber method, it is necessary to shorten the testing duration for static chamber method at a site where gas is remarkably emitted. And, the chamber with the long height is also effective for an accurate measurement.

Keywords: Landfill, Greenhouse effect gas, Static chamber method, Density dependent gas flow, Multicomponent gas transport

1. Introduction

Monitoring of landfill gas such as methane (CH₄) and carbon dioxide (CO₂) is important to evaluate the environmental impact on the global warming as well as the stabilization of landfill wastes. Static chamber method, which measures a gas flux, is widely used at landfill sites and swamp sites etc. because of a relatively low cost, easy installation and simple operation. Figure 1 shows a photo of static chamber method at a landfill site. The chamber is first placed on the landfill surface, then the gas concentration in the chamber is measured with time. The gas flux is calculated from the product of the chamber height and the gas concentration gradient to the elapsed time, when the gas pressure in the chamber is assumed to be constant. Therefore, the measurement by the static chamber method becomes unreliable at a landfill site where the gas flux is large and the gas pressure in the

chamber is significantly increased. To measure the gas flux with a good accuracy, the difference between the actual gas flux emitted from a landfill site and its measured flux should be investigated. Then, the measuring condition to minimize the difference must be considered through the investigation results.

In this study, COMSOL Multiphysics ver 3.5a was used for theoretically investigating the flux measurement mechanism of the static chamber method. The purpose is to evaluate the error between the actual gas flux emitted from a landfill site and its measured flux, then to clarify the measuring condition of the chamber to minimize the error.



Figure 1. Static chamber method.

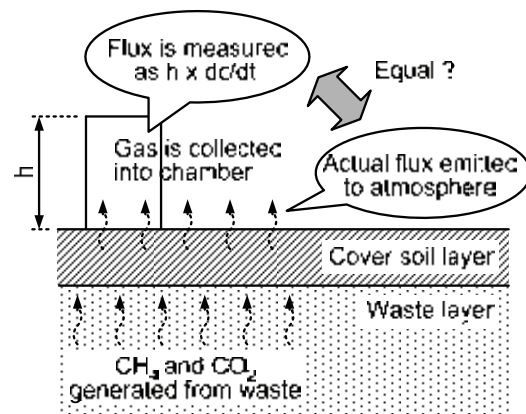
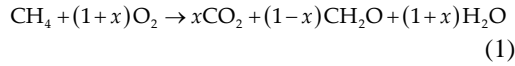


Figure 2. The way to measure the gas flux using the static chamber method, and the difference between the measured flux and the real flux.

2. Theory and Governing equations

The common structure of landfill consists of two layers. One is waste layer, and the other is soil cover layer. Landfill gas of CH₄ and CO₂ are generated by the degradation of the landfill wastes. They pass through the cover soil layer, then are emitted to atmosphere. The CH₄ gas passing through the cover soil layer is oxidized. The oxidation rate depends on both biochemical and physical processes in soil. In the presence of oxygen (O₂), the CH₄ can be degraded by methanotrophic bacteria. The process leads to generation of CO₂ and water (Christophersen, 2001; Visscher and Cleemput, 2003):



where, CH₂O represents biomass, and x is the stoichiometric factor. This study focused on four gas components of CH₄, CO₂, O₂, and nitrogen (N₂) which are mainly included in landfill, and simulated their transport and reaction. The gas temperature was assumed to be constant at 25 °C.

2.1 Governing equations

The governing equations are the gas seepage equation based on Darcy's Law, and the multi-component gas advection-diffusion equations based on Maxwell-Stefan diffusion model:

$$S_g \left(\rho_g \frac{\partial \phi}{\partial p_g} + \phi \frac{\partial \rho_g}{\partial p_g} \right) \frac{\partial p_g}{\partial t} + \nabla \cdot \left[-\rho_g \frac{k_{rg} \mathbf{K}}{\mu_g} (\nabla p_g + \rho_g g \nabla z) \right] = 0 \quad (2)$$

$$\rho_g \phi S_g \frac{\partial \omega_1}{\partial t} + \rho_g \mathbf{u}_g \nabla \omega_1 + \nabla \cdot \left[-\rho_g \omega_1 \sum_{\kappa=1}^4 \phi S_g \tau \bar{D}_{1\kappa} \left(\nabla x_\kappa + (x_\kappa - \omega_\kappa) \frac{1}{p_g} \nabla p_g \right) \right] + \gamma_1 = 0 \quad (3)$$

$$\rho_g \phi S_g \frac{\partial \omega_2}{\partial t} + \rho_g \mathbf{u}_g \nabla \omega_2 + \nabla \cdot \left[-\rho_g \omega_2 \sum_{\kappa=1}^4 \phi S_g \tau \bar{D}_{2\kappa} \left(\nabla x_\kappa + (x_\kappa - \omega_\kappa) \frac{1}{p_g} \nabla p_g \right) \right] + \gamma_2 = 0 \quad (4)$$

$$\rho_g \phi S_g \frac{\partial \omega_3}{\partial t} + \rho_g \mathbf{u}_g \nabla \omega_3 + \nabla \cdot \left[-\rho_g \omega_3 \sum_{\kappa=1}^4 \phi S_g \tau \bar{D}_{3\kappa} \left(\nabla x_\kappa + (x_\kappa - \omega_\kappa) \frac{1}{p_g} \nabla p_g \right) \right] + \gamma_3 = 0 \quad (5)$$

where, p_g (Pa) is the total gas pressure, ω_1 - ω_4 are the mass fraction of each gas component and x_1 - x_4 are the molar fraction of each gas component. The type of the each gas component is shown by the subscript of "1" to "4". "1" is for the CH₄, "2" is for the CO₂, "3" is for the O₂, and "4" is for the N₂. Total mass fraction or molar fraction must be equal to one. S_g is the degree of the gas saturation. ϕ and τ are the porosity and tortuosity of the cover soil or landfill waste, respectively. ρ_g (kg/m³) and μ_g (Pa s) are the fluid density and viscosity of the mixture gas, respectively. k_{rg} is the relative permeability and \mathbf{K} (m²) is the intrinsic permeability. The darcy velocity, \mathbf{u}_g (m/s), is calculated as follows:

$$\mathbf{u}_g = -\frac{k_{rg} \mathbf{K}}{\mu_g} (\nabla p_g + \rho_g g \nabla z) \quad (6)$$

$\bar{D}_{\kappa\iota}$ (m²/s) is the symmetric diffusivity proposed by Curtiss and Bird (1999). The symmetric diffusivity is calculated from the binary diffusion coefficient, $D_{\kappa\iota}$ (m²/s), between κ -th and ι -th gas component. This calculation is automatically conducted in COMSOL Multiphysics ver 3.5a. γ_1 - γ_4 (kg/m³/s) are the source/sink term as to CH₄ oxidation. Based on Eq.(1), each source/sink term can be defined using first-order degradation assumption:

$$\gamma_1 = \phi S_g \lambda_1 \rho_g \omega_1 \quad (7)$$

$$\gamma_2 = -x \frac{M_2}{M_1} \gamma_1 = -x \frac{M_2}{M_1} \phi S_g \lambda_1 \rho_g \omega_1 \quad (8)$$

$$\gamma_3 = (1+x) \frac{M_3}{M_1} \gamma_1 = (1+x) \frac{M_3}{M_1} \phi S_g \lambda_1 \rho_g \omega_1 \quad (9)$$

$$\gamma_4 = 0 \quad (10)$$

where, λ_1 (1/s) is the first-order methane oxidation rate, and M_1 - M_4 (kg/mol) are the molecular weight of each gas component.

2.2 Physical properties

The fluid density, ρ_g (kg/m³), of the mixture gas is calculated from ideal gas equation.

$$\rho_g = \frac{p_g M_g}{RT} = \frac{p_g}{RT} \sum_{\kappa=1}^4 M_\kappa x_\kappa \quad (11)$$

where, M_g (kg/mol) is the molecular weight of the mixture gas, R (J/K/mol) is the gas constant, T (K) is the absolute temperature. M_κ (kg/mol) and x_κ are the molecular weight and molar fraction of κ -th component gas, respectively.

The viscosity, μ_g (Pa s), of the mixture gas was estimated from Wilke equation as follows:

$$\mu_g = \sum_{\kappa=1}^4 \frac{\mu_{\kappa} x_{\kappa}}{\sum_{\iota=1}^4 \phi_{\kappa\iota} x_{\iota}} \quad (12)$$

where,

$$\phi_{\kappa\iota} = \frac{\left[1 + (\mu_{\kappa}/\mu_{\iota})^{1/2} (M_{\iota}/M_{\kappa})^{1/4}\right]^2}{\left[8(1 + M_{\kappa}/M_{\iota})\right]^{1/2}} \quad (13)$$

μ_{κ} (Pa s), M_{κ} (kg/mol), and x_{κ} are the viscosity, molecular weight, and molar fraction of κ -th gas component.

The binary diffusion coefficient, $D_{\kappa\iota}$ (m²/s), in low gas pressure range of $100 < p_g < 500$ kPa can be estimated from Chapman-Enskog theory.

$$D_{\kappa\iota} = 5.9546 \frac{\left[T^3 (1/M_{\kappa} + 1/M_{\iota})\right]^{1/2}}{p_g \sigma_{\kappa\iota}^2 \Omega_D} \times 10^{-4} \quad (14)$$

where, $D_{\kappa\iota}$ (m²/s) and $\sigma_{\kappa\iota}$ (10^{-10} m) are the binary diffusion coefficient and intermolecular force constant between κ -th and ι -th gas component, respectively. The intermolecular force constant is calculated as $\sigma_{\kappa\iota} = (\sigma_{\kappa} + \sigma_{\iota})/2$. M_{κ} (kg/mol) is the molecular weight of κ -th gas component. p_g (Pa) is the total gas pressure, and T (K) is the absolute temperature. Ω_D is represented as

$$\Omega_D = \frac{1.06036}{T_N^{0.1561}} + \frac{0.19300}{e^{0.47635T_N}} + \frac{1.03587}{e^{1.52996T_N}} + \frac{1.76474}{e^{3.89411T_N}} \quad (15)$$

where, T_N is a parameter, calculated from $T_N = kT/\varepsilon_{\kappa\iota}$. k (J/K) is the Boltzman's constant, and $\varepsilon_{\kappa\iota}$ (J) is the intermolecular force constant between κ -th and ι -th gas component. The values of these parameters are summarized in Appendix.

2.3 Analysis conditions

The analysis was conducted in two-dimensional axial symmetry domain, which consisted of 4.5 m waste layer and 0.5 m cover soil layer in thickness as shown in Figure 3. Temperature was assumed constant of 25 °C.

The purpose of the analysis is to evaluate effects of the real gas emission on the flux measured by static chamber method. The permeability and the first-order degradation rate of the cover soil layer were parametrically changed. The analysis condition was as follows: The permeability was 10^{-12} m² for the waste layer, and 10^{-15} - 10^{-12} m² for the cover soil layer. The

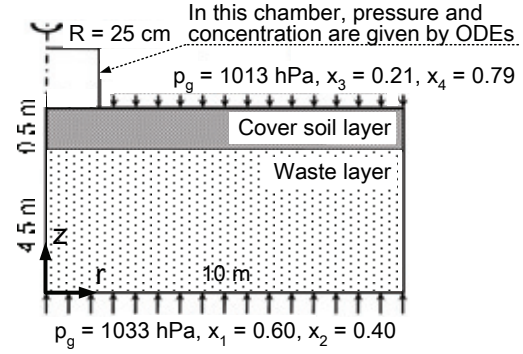


Figure 3. Analysis domain.

first-order degradation rate of the cover soil layer was $0-10^{-3}$ 1/s. The porosity and the tortuosity were 0.3 and 0.1 for each layer, respectively. The fluid density and viscosity of the mixture gas were calculated from Eq.(11) and Eq.(12), depending on the solutions of Maxwell-Stefan advection-diffusion analysis. The binary diffusion coefficients were determined from Eq.(14), depending on the solutions of Darcy Law's pressure analysis.

The left boundary was the central axis of symmetric analysis domain. The right boundary was given as zero flux. The top boundary was given as atmospheric pressure and molar fractions (Namely, $p_g = 1013$ hPa, $x_1 = 0$, $x_2 = 0$, $x_3 = 0.21$, $x_4 = 0.79$). The bottom boundary was given as $p_g = 1033$ hPa, $x_1 = 0.6$, $x_2 = 0.4$, $x_3 = 0$, $x_4 = 0$. These gas concentrations are typical values. Gas pressure and concentration on the contact face with the chamber were $p_g = p_{\text{chm}}$, $x_1 = p_{\text{chm},1}/p_{\text{chm}}$, $x_2 = p_{\text{chm},2}/p_{\text{chm}}$, $x_3 = p_{\text{chm},3}/p_{\text{chm}}$, $x_4 = p_{\text{chm},4}/p_{\text{chm}}$. p_{chm} (Pa) is the total pressure of $p_{\text{chm},1}$ to $p_{\text{chm},4}$ in the chamber. $p_{\text{chm},1}$ - $p_{\text{chm},4}$ (Pa) are the partial pressure of each gas component.

The partial pressures in the chamber, $p_{\text{chm},1}$ - $p_{\text{chm},4}$, were expressed as the following ordinary differential equations:

$$\frac{dp_{\text{chm},1}}{dt} = -\frac{Q_1 RT}{M_1 V_{\text{chm}}} \quad (16)$$

$$\frac{dp_{\text{chm},2}}{dt} = -\frac{Q_2 RT}{M_2 V_{\text{chm}}} \quad (17)$$

$$\frac{dp_{\text{chm},3}}{dt} = -\frac{Q_3 RT}{M_3 V_{\text{chm}}} \quad (18)$$

$$\frac{dp_{\text{chm},4}}{dt} = -\frac{Q_4 RT}{M_4 V_{\text{chm}}} \quad (19)$$

where, V_{chm} (m^3) is the chamber volume, and Q_1 - Q_4 (kg/s) are the mass flow into the chamber.

Initial conditions for Darcy Law's pressure analysis and Maxwell-Stefan advection-diffusion analysis were their stationary solutions without chamber. And, initial condition for the ordinary differential equations in the chamber were given as $p_{\text{chm}} = p_{\text{atm}}$, $p_{\text{chm},1} = 0$, $p_{\text{chm},2} = 0$, $p_{\text{chm},3} = 0.21 p_{\text{atm}}$, and $p_{\text{chm},4} = 0.79 p_{\text{atm}}$. These initial pressure conditions were the same as those of atmosphere.

2.4 Use of COMSOL Multiphysics

Two application modes of Earth Science Module and Chemical Engineering Module were used. One was Darcy Law's Pressure Analysis. The other was Maxwell-Stefan Diffusion and Convection Analysis with four gas components of CH_4 , CO_2 , O_2 and N_2 . These were used in the axial symmetry two dimensional analysis mode. Additional settings were required as follows.

(1) Modifications of equation system

To consider the density dependent gas flow and Maxwell-Stefan diffusion in porous media, the default equation system should be changed, as shown in Eqs.(2)-(5). The fluid density, ρ_g , of the mixture gas was added to the Damping/Mass term and the Conservative flux source term in the default equation system of the Darcy Law's Pressure Analysis. And, the porosity, ϕ , the gas saturation, S_g , and the tortuosity, τ , were added to the Damping/Mass term and the Conservative flux source term in the default equation system of the Maxwell-Stefan Diffusion and Convection Analysis.

(2) Settings of chemical reaction terms

Although chemical reaction rates in Eqs.(7)-(10) can be given to the reaction rate terms in the Subdomain Settings in the Maxwell-Stefan Diffusion and Convection Analysis, it should be noted that their reactions are caused only when the oxygen concentration is more than zero. Their reactions are stopped when the oxygen concentration reaches zero. The switch which depends on the oxygen concentration can be carried out by giving the reaction rates shown in Eqs.(7)-(10) to the reaction rate terms with Heaviside function. The Heaviside function had the argument of the oxygen mass fraction, ω_3 .

(3) Settings of ordinary differential equations

Gas pressure and concentration on the contact face with the chamber were defined by the ordinary differential equations shown in Eqs.(16)-(19). COMSOL Multiphysics ver 3.5a can easily define them from Global Equations in Physics menu. Here, each mass flow into the chamber, Q_1 - Q_4 , was the integral value of the mass flux of each gas component which passes through the contact face with the chamber. COMSOL can also calculate each value from Integration Coupling Variables in Option menu.

(4) Settings of constants and expressions

Constants, expressions and functions used in the analysis were summarized in Appendix.

3. Results and Considerations

3.1 Concentration profiles in landfill

Figure 4 shows the stationary solutions for the analysis domain without the chamber. It is the mass fraction profiles in the vertical cross-section. The effects of the methane oxidation reaction shown in Eq.(1) appeared in the profiles. The solutions with the methane oxidation effect had a lower CH_4 concentration in the cover soil layer than those without the methane oxidation effect. That CO_2 concentration was increased and O_2 concentration was decreased according to this methane oxidation effect is also simulated.

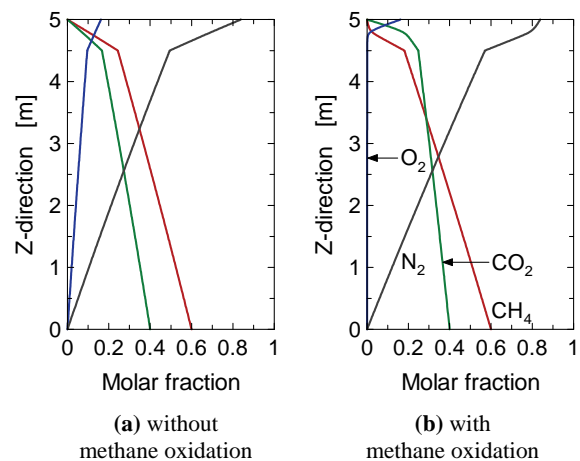


Figure 4. Simulated stationary concentration profiles in landfill sites; the $0 < z < 4.5$ m range is for waste layer and the $4.5 < z < 5$ m range is for cover soil layer.

To enhance the stabilization of the landfill wastes, it is necessary to provide oxygen into the waste layer. This simulation model showed that most of the oxygen in the atmosphere was consumed in the cover soil layer for the methane oxidation. It was impossible for oxygen to pass through the cover soil layer and to diffuse into the waste layer. The COMSOL simulation concluded that the oxygen for the landfill waste stabilization should be directly provided into the waste layer by injection wells and so on.

3.2 Change of gas pressure and concentration profiles during static chamber method

Figures 5 and 6 show the time-dependent simulation results for the analysis domain with the chamber. The initial conditions were the stationary solutions for the analysis domain without chamber (namely, concentration profile shown in Figure 4). Figure 5 is the distribution of the total gas pressure around the chamber. The r -axis (m) is the radius vector, and the z -axis (m) is the elevation vector. $0 < z < 4.5$ m is for the waste layer domain and $4.5 < z < 5$ m is for the cover soil layer domain. The gas pressure on the contact face with the chamber, which is in the range of $0 < r < 0.25$ m and on $z = 5$ m, was gradually increased with time so that it became difficult for gas to flow into the chamber (see the CH_4 flux vectors in Figure 5). Figure 6 shows the changes of the molar fraction of each gas component and the total gas pressure in the chamber with time. The initial molar fraction immediately after the chamber was placed on the cover soil layer was the same as that of the atmosphere. The molar fraction of CH_4 and CO_2 gas were increased with time, because the CH_4 and CO_2 gas in the cover soil layer and the waste layer flowed into the chamber. However, when the elapsed time became so long, the gas cannot flow into the chamber as shown in Figure 5(b). It was because the gas in the cover soil layer and waste layer was collected and the gas pressure in the chamber was increased. The increased gas pressure in the chamber made it difficult for the gas in the cover soil and waste layer to flow into the chamber. As the result, the increase of the gas concentration in the chamber stopped. That the gas concentration in the chamber reaches such saturation level sometimes deteriorates the gas flux estimation accuracy.

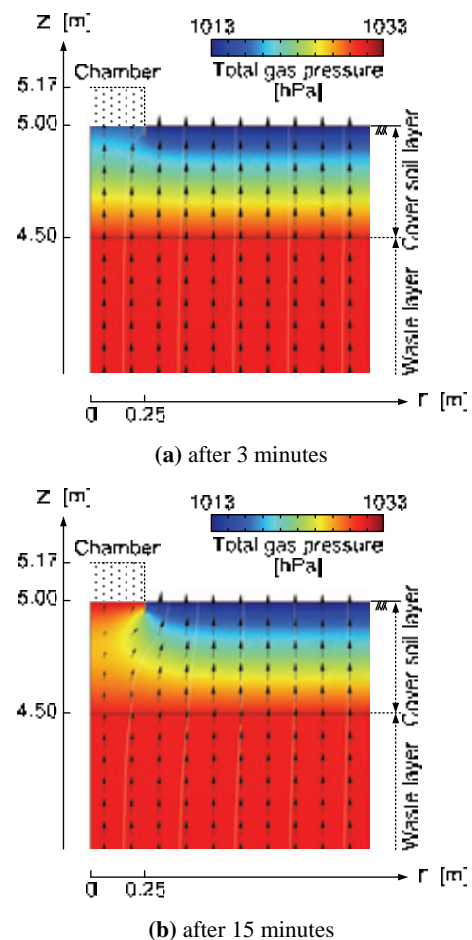


Figure 5. Gas pressure distribution and the total CH_4 flux vectors after the static chamber method starts: The analysis conditions are that the gas permeability is 10^{-12} m^2 for waste layer, and $2 \times 10^{-14} \text{ m}^2$ for cover soil layer. The methane oxidation is neglected.

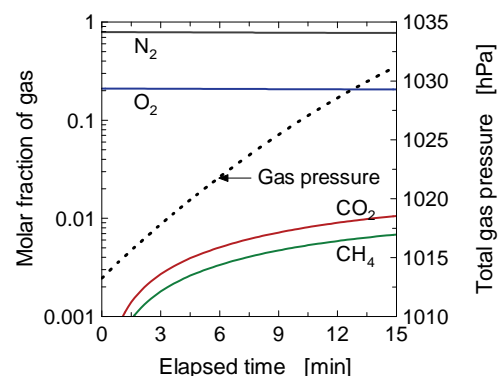


Figure 6. Gas pressure and concentration in chamber during static chamber method: The analysis condition is the same as shown in the caption in Figure 5.

In site investigations, the gas in the chamber is sampled with time. For example, the gas was sampled to each gas bag at 1, 7, and 15 minutes after static chamber method starts. The CH₄ and CO₂ concentration in gas bags can be measured by the gas chromatography and so on. The gas flux can be calculated from the product of the chamber height and the gradient of the relation between the measured gas concentration and the elapsed time:

$$F_{1,est} = h \frac{d\rho_1}{dt}, \quad F_{2,est} = h \frac{d\rho_2}{dt} \quad (20)$$

$F_{1,est}$ - $F_{2,est}$ (kg/m²/s) are the estimated fluxes of the CH₄ or CO₂ gas, and ρ_1 - ρ_2 (kg/m³) are the measured concentrations of the CH₄ or CO₂ gas in the chamber, and h (m) is the chamber height. The gradient shown in Eq.(20) is evaluated by fitting the relation between the measured gas concentration and the time, which the gas was sampled at, to a liner function. But, when the gas concentration in the chamber reaches the saturation level and has a non-linear relation to the time, the gas flux cannot be estimated from Eq.(20). For this case, static chamber method should be finished before the gas concentration in the chamber reaches such a saturation level.

3.3 Reliability of static chamber method

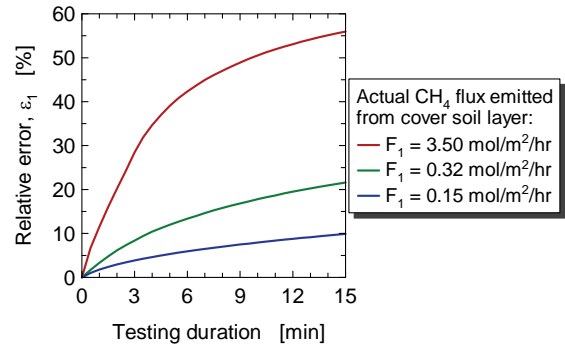
In site, however, it is very difficult to judge whether the gas concentration in the chamber reaches the saturation level or not, because the sampled gas concentration cannot be measured immediately. Thus, it is significantly important to clarify the error which is included in the measured value of gas flux, when static chamber method is conducted under a certain condition (chamber height, interval time for gas sampling, and duration for static chamber method).

Figure 7 shows the effects of the magnitude of the actual gas flux and the testing duration on the relative error of its measured gas flux. The relative error, ε_1 - ε_2 , are defined as follows:

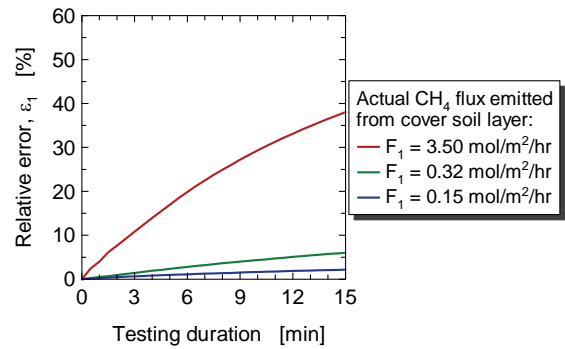
$$\varepsilon_1 = \frac{F_{1,est} - F_1}{F_1}, \quad \varepsilon_2 = \frac{F_{2,est} - F_2}{F_2} \quad (21)$$

where, F_1 - F_2 (kg/m²/s) are the actual CH₄ or CO₂ gas flux emitted from a site. They were the gas fluxes emitted from the cover soil layer surface, and were evaluated from the stationary solutions for the analysis domain without chamber. The

$F_{1,est}$ - $F_{2,est}$ were calculated from Eq.(20) using the gas concentration profiles in the chamber (e.g. Figure 6). For this calculation, the concentration profiles from start to end of the testing duration were approximated with a linear function. The simulation results showed that the relative error was increased with the testing duration and the emitted gas flux. Therefore, it is necessary to shorten the testing duration for static chamber method at a site where gas is remarkably emitted. The long chamber is also effective to decrease the relative error. The relationships as shown in Figure 7 will be effective to evaluate the reliability of measured gas flux data and to correct them according to the testing condition for static chamber method.



(a) chamber height = 0.17 m



(b) chamber height = 1.00 m

Figure 7. Effects of the actual gas flux emitted from a site and the testing duration for static chamber method on the relative error in the measured gas flux. This figure is only for CH₄. The figure for CO₂ is almost the same as this.

4. Conclusions

This study used COMSOL Multiphysics for the reliability evaluation for static chamber method to measure the fluxes of the methane and carbon dioxide emitted from landfill sites. The simulation model for the multicomponent gas advection-diffusion-reaction model, considering the density dependent flow, counter diffusion phenomena, and methane oxidation reaction, was proposed. The reliability of the static chamber method was discussed using this model, and the following findings were obtained.

(1) In static chamber method, the relative error was increased with the testing duration. In particular, the relative error became so large for the gas flux measurement at a site where gas is remarkably emitted. Thus, the testing duration for static chamber method should be shortened when emitted gas flux is considered large.

(2) The short chamber had larger relative error in flux measurement than the long one. In the measurement for gas flux with less than 1 mol/m²/hr, the short chamber with height of 0.17

m showed two times or more as much relative error as the long chamber with height of 1.00 m. Therefore, the long chamber is also effective to decrease the relative error.

5. References

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6. Appendix

Figure 8 summarized the analysis conditions.

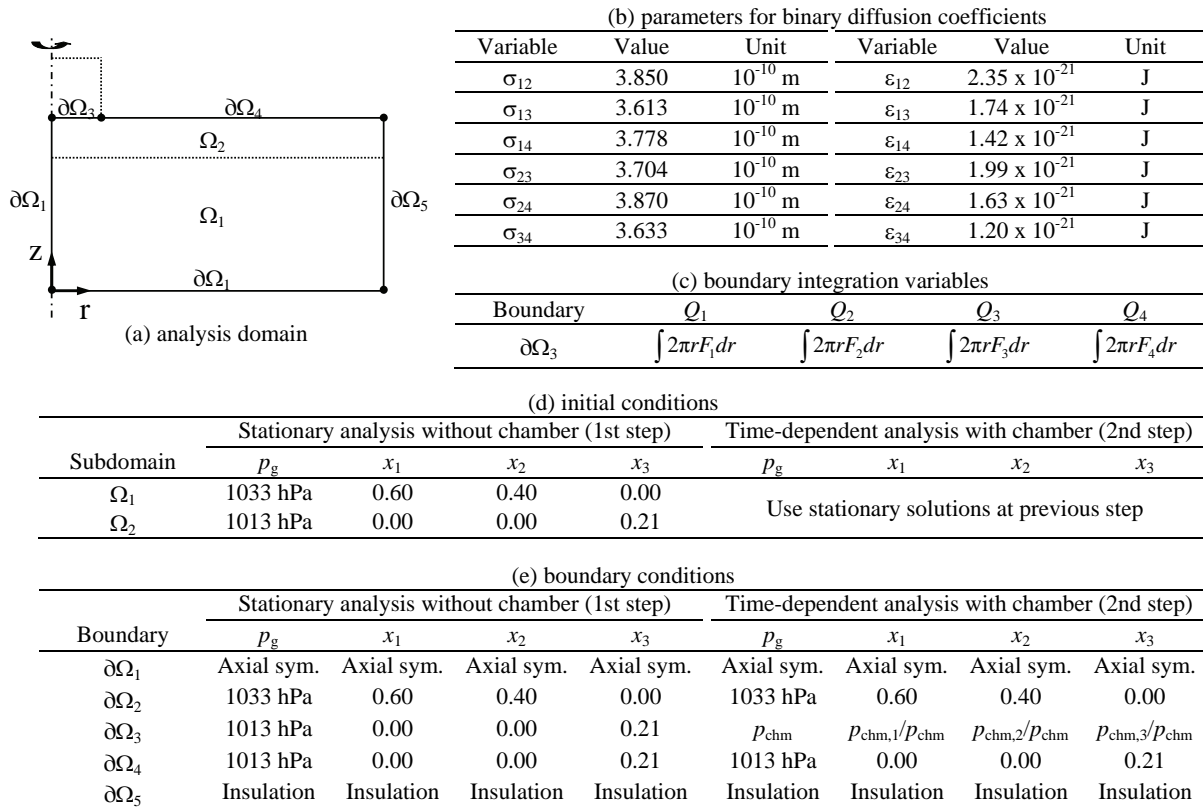


Figure 8. Main analysis conditions in this study