QBS
Vol. 37, No. 1, pp. 27-32 (2018)
https://doi.org/10.22283/qbs.2018.37.1.27
pISSN 2288-1344 eISSN 2508-7185 **Original Article**

Quantitative Bio-Science
Official Journal of the Institute of Natural Science at Keimyung University
Available Online at https://qbs.kmu.ac.kr:442/

Mathematical Modeling for Gas Removal in a Fixed-film Bio-scrubber Process

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(Received April 9, 2018; Revised May 16, 2018; Accepted May 19, 2018)

ABSTRACT

Bio-scrubbers include fixed-film or suspended microorganisms and can be used to remove gas and liquid contaminants. They often require reliable modeling due to their highly complex mechanisms. However, only a few studies have conducted mathematical modeling for bio-scrubbing processes, particularly, gas treatment using fixed-film microorganisms in bio-scrubbers. This study applied a simple mathematical model to predicting the removal efficiencies of ammonia, formaldehyde, toluene, butanol, acetone, and ethylene gases. The model presumed a steady-state condition and combined various physicochemical and microbial factors to obtain a series of closed-form solutions from equations. In the model, the gaseous compounds were absorbed into liquid phase and degraded by fixed-film microbes attached to the scrubber media. As a result, removals of strongly hydrophilic or hydrophobic gases were relatively invariant along the media depth. The gas and water flow rates highly affected scrubber performances for the gaseous compounds with moderate or low water solubility. In contrast, the removals of the highly soluble gaseous compounds, that is, formaldehyde and butanol, were not sensitive to variations in gas or water flow rates. Water flow at under 100 L/h produced limited wetting efficiencies ($<20\%$) of the scrubber media (600 m^2/m^3). Henry constants and microbial parameters of maximum microbial degradation rates and half saturation coefficients had significant effects on gas removal, while media properties, such as specific surface areas and surface tensions, had less significant effects.

Key words : Bio-scrubber, Gas removal, Mathematical model

Introduction

Since mixtures of gas phase contaminants would be generated during various industrial or other activities, air cleaning systems are often needed to remove the contaminants from the air. Physico-chemical methods (activated charcoal, lithium hydroxide, or catalytic converter) are currently used dominantly. This study focuses on the potential of integrated biological techniques to relieve or share the burden with physicochemical methods [1]. Thus, this study is developing a mathematical model for a bio-scrubber to remove the gas contaminants. Both processes have been known to be effective in removing organic and some inorganic gas pollutants. However, the mechanisms of bio-scrubbers are so complex that development of a theoretical model is necessary prior to experimentation to understand the mechanisms.

Bio-scrubber processes generally consist of water, solid media, and the microbial mixed culture of suspended in liquid or attached onto the solid media. Bio-scrubbers absorb gas contaminants in free water before biodegradation by mostly fixed or partly suspended microorganisms. The free water phase in bio-scrubbers can provide a continuous supply of nutrition and removal of toxic materials. Except for the presence of the free water phase, the overall mechanisms of bio-

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scrubbing are similar to those of a biofiltration. However, the bio-scrubber is considered better than conventional biofiltration for removing contaminants that are difficult to biodegradable [2].

Development of biological gas treatment model started in the 1980s from Ottengraf's works on simulation of submerged biofilms. His model has been commonly referred in other similar process (i.e. biofiltration) models [2-4]. Diks and Ottengraf simulated the reaction in fixed film scrubbers based on the assumption of zero-order reaction in microbial degradation [5,6]. Alonso et al. suggested a dynamic mathematical model for the biodegradation of toluene in biotrickling filtration [7], which contains lower liquid flow rates than bio-scrubbers. Their model incorporates the equations of free water flow and biomass accumulation on filter media as well as toluene removal.

This study suggested the simulation data using equations revised from the model developed by Ockeloen et al. [8]. Their model had been developed to simulate a typical fixed-film bio-scrubber process. It is advantageous that the model is simple and fast in calculation, and includes an air stripping model equation conventionally used in modeling chemical plants, so many kinds of mass transfer parameters or functions developed earlier may be used. Also, the model contains the parameters about the free water phase in a bio-scrubber. In Ockeloen et al.'s research, the co-current water flow direction, instead of the counter-current flow, was strongly recommended to treat the gas contaminant with a high Henry's law coefficient value [8]. Also, they insisted that decrease water input increase biodegradability by increasing water retention time. However, they could not find more of the role of free water phase in the bio-scrubbing should be considered further. For example, there is a lack of wetting of media surface if injected water amount is too low.

Therefore, using the proposed model, the influences of physical, chemical, and biological parameters on removal of mixtures of gaseous contaminants were estimated using the modified model. This study chose six representative gaseous compounds (ammonia, formaldehyde, toluene, n-butanol, acetone and ethylene) for the model test due to their significance in air pollution and toxicity. Also by incorporating Onda's methods, the study described the overall mass transfer functions including not only water flow but also wetting in media surfaces [9]. Consequently, this model may describe optimal points in water loading. Then, this study made a better recommendation in engineering design of a better bio-scrubber, prior to experimentation.

Model Development

On a basis of the assumption of plug flow conditions of water/gas gas co-current flow and convective transport of contaminants, the material balance of gas phase substrate concentration S_G is described as follows;

$$dS_G/dz = -[K_L\, a_w (S_L/H - S_L)] / u_G \quad (1)$$

where

z = biofilter depth (m)

u_G = superficial gas velocity (m/s)

S_L = water phase substrate concentration (mg/L)

H = Henry's law coefficient (−)

K_L = Overall mass transfer coefficient (m/s)

a_w = wetted specific surface area of media (m^2/m^3)

Also, the material balance of the downward flow of water is given by

$$dS_L/dz = -[K_L\, a_w (S_L/H - S_L)] / u_L - Ja_w/u_L \quad (2)$$

where

u_L = superficial water velocity (m/s)

J = substrate flux to biofilm (mg/s)

According to the study of Ockeloen et al. [8], the substrate flux equation into the biofilm is given by

$$J = [2\, D_f\, Ks\, q\, X_f]^{1/2}\, [S_L/Ks - \ln(1 + S_L/Ks)]^{1/2} \quad (3)$$

where

D_f = diffusivity into biofilm (m^2/s)

Ks = Half-saturation coefficient (mg/L)

q = maximum specific substrate utilization rate (1/s)

Xf = biomass density (mg/L)

For the maximized flux of substrate (J_{max}), a zero order equation at maximum flux was suggested when the biofilm is fully penetrated and contaminant concentration in biofilm (L_f in depth (m)) is higher than Ks as follows:

$$J_{max} = q\, X_f\, L_f \quad (4)$$

In addition to a series of the equations above, this model incorporated the overall mass transfer and wetted surface area of media, in which necessary equations were suggested by Ondo et al. [9]. When using the gas and water phase transfer

Table 1. Input gaseous compounds into the hypothetical bio-scrubber of this study

	Generation (mg/day)	Risks (mg/m^3)	Input (mg/m^3)	Henry's law constant (−)	M.W (g/mol)	D_{fo} (m^2/h)	D_g (m^2/h)
Ammonia	2115	7.0	600	0.01	17	1.16E-05	0.098
Formaldehyde	3	0.1	50	0.00001	30	6.48E-06	0.061
Toluene	257	10	50	0.1	28	7.67E-06	0.068
N-butanol	356	121.0	50	0.0001	74	3.15E-06	0.032
Acetone	384.0	712.5	300	0.001	58	3.60E-06	0.040
Ethylene	0	344.1	100	1	26	5.11E-06	0.054

equations, this study calibrated these parameters in order to obtain the same resultant data as the previous results of Ockeloen et al. [8]. Consequently, this proposed model produces the same data if it uses same input values of Ockeloen et al.

There are several assumptions for simplification before simulation of biotrickling filter processes, summarized as follows. First, the model only considers a steady-state condition, and microbial growth occurs. Also, concentration gradients are generated only along plug flow, not along biofilm phase. Neither oxygen nor nutrient limitation is assumed. Water used in the biotrickling filtration may be recycled without further treatment.

The contaminant chemicals are assumed to have the same microbial degradation parameters, because such microbial parameters are largely unknown or uncertain. This study compensates this unrealistic assumption by sensitivity analysis with variation of microbial parameters.

Input contaminants and their physico-chemical parameters are listed in Table 1. Seven kinds of chemicals are chosen as typical representative contaminants, in which the selection is based on generation rate, risk and comparison to literature data [10,11]. Although not predicted exactly, the concentrations of the contaminants were roughly determined with arbitrary values for the first stage of experiments. In spite of the negligible generation rate, ethylene was involved, because of its high risk. Methane was excluded because its modeling results may be similar to those of ethylene. Henry's law constants were cited from references [2], and guessed using solubility when there is no information for corresponded chemicals. Molecular diffusivity constants in biofilm (D_{fo}) and air phases (D_g) for each chemical were assumed using molecular weight or molar volume, if there is no information in references [2]. Tolerance values which are shown in the Table 1 means the maximum allowable concentrations in a cabin space with confined indoor air and internal re-circulation. The default input data are shown in Table 2.

Table 2. Default input design and operation parameters for this study

Media depth	m	1
Sectional Dia.	m	0.1
Media Bulk Dia.	m	0.01
Air flow rate	L/min	7
Water flow rate	L/h	10
Porosity	−	0.9
Specific surface area of media	m^2/m^3	600
q*Xf	Subst. g COD/h/m^3	5.00E+05~5.00E+7
Lf	m	1.00E-04
Ks	mg/m^3	10000

Results

The left plot Fig. 1 describes the effects of the change of gaseous contaminant concentration along biofilter depth. Gas removal is highly dependent of Henry's law coefficients so that absorption effects may be the most significant. The right plot of Fig. 1 shows the similar effects onto the dissolved contaminant concentration along biofilter depth. Since water is recycled, the contaminant concentrations at the bottom and at the top of the reactor are same each other. If water is not recycled and clean water is continuously loaded, gas contaminant removed more highly. The data in Fig. 1 are for the bio-scrubber with water recycled.

As water flow rate increases up to 1-100 L/hr, the gaseous contaminant removal increases as shown in the left plot of Fig. 2 but water contaminant concentration decreases (data are not shown). This result is different to that of previous research, because the previous research utilized a constant overall mass transfer and specific surface area values [8]. They referred that even a large variation of overall mass transfer coefficients results in only small variation of removal effects for the contaminants of Henry's law constants below 0.1.

However, this study shows that, under considering wetting phenomena, decreased water flow may decrease the wetted surface area as shown in the right plot of Fig. 2, so gas removal effects decreased. Under the whole wetting condition regard-

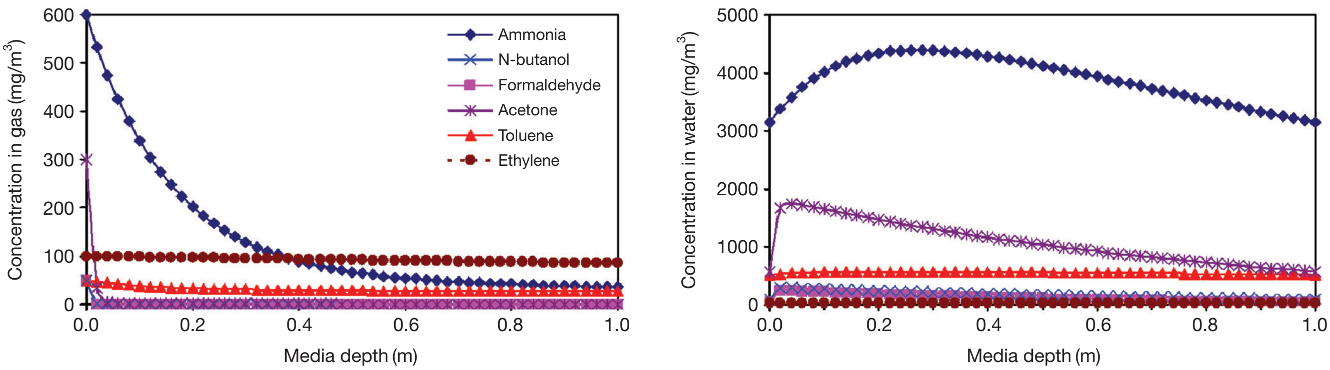

Fig. 1. The concentrations of gaseous compounds (Left) and their dissolved ones (Right) as a function of media depth when gas flow and re-circulated water flow rates are equal to 7 L/min and 10 L/h, respectively.

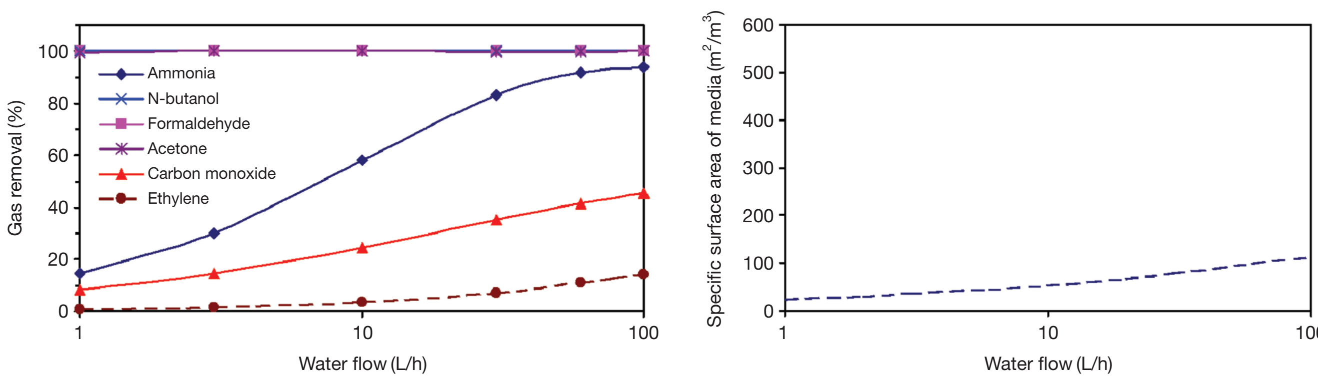

Fig. 2. Water flow effects to gas removals (Left) with the assumably full wetting condition and wet specific surface areas.

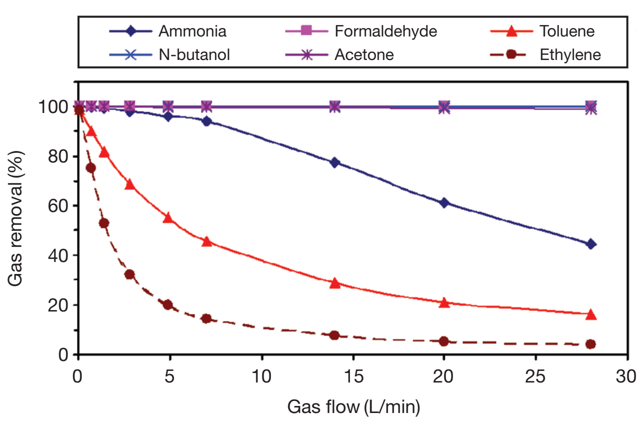

Fig. 3. The effects of gas flow rates to gas removals.

less of the water flow rate, water flow may be reduced to achieve the same gas removal effects which are seen in the left plot of Fig. 2. Consequently, homogenization in wetting can save water consumption about 1/5∼1/10. Fig. 3 shows the effects of the changing gas flow rate to gas removal along the full media bed depth. The resultant effects are as significant and apparent as those of water flow rate. The insoluble gaseous removals highly increase with decreasing gas flow rate. It is expect that the contaminants difficult to degrade may be effectively removed by decreasing gas flow rate.

Fig. 4 describes the effects of three microbial parameters considered in this study, maximum specific utilization rate and half-saturation coefficient. Other parameters such as biofilm thickness and diffusivity coefficients were assumed to be invariant and not considered in this study, because their effects should be compounded with the two parameters considered in the proposed model. For efficiency in a graph presentation, maximum specific utilization rate and biomass density were unified into one parameter. Generally, the influence of microbial parameters looks so huge (although the horizontal axes are log scale). This study selected toluene as the target chemical due to its moderate Henry's law coefficient shows rather higher variation. Similarly, a small change of half-saturation coefficient or large value of the other combined parameters produced significant influences on the gas removal effects.

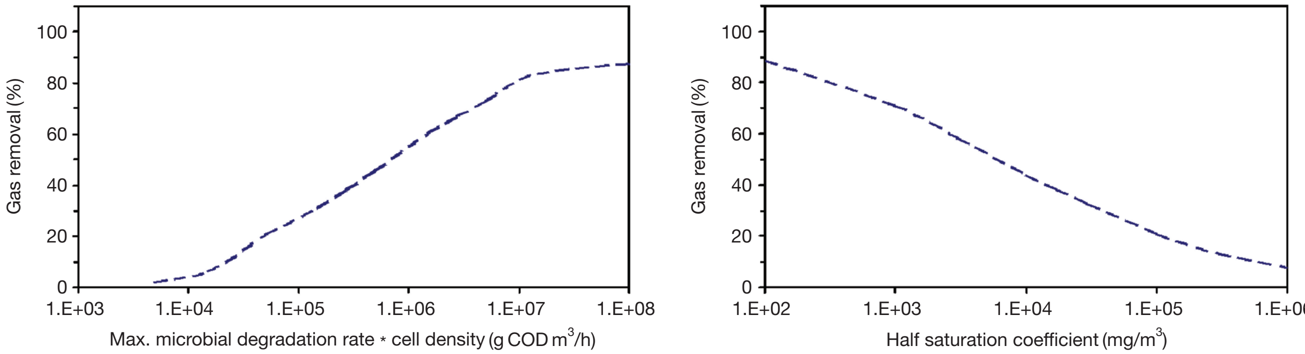

Fig. 4. Toluene gas removals as a function of microbial degradation rates (Left) and half saturation coefficients (Right) at 7 L/min of gas flow.

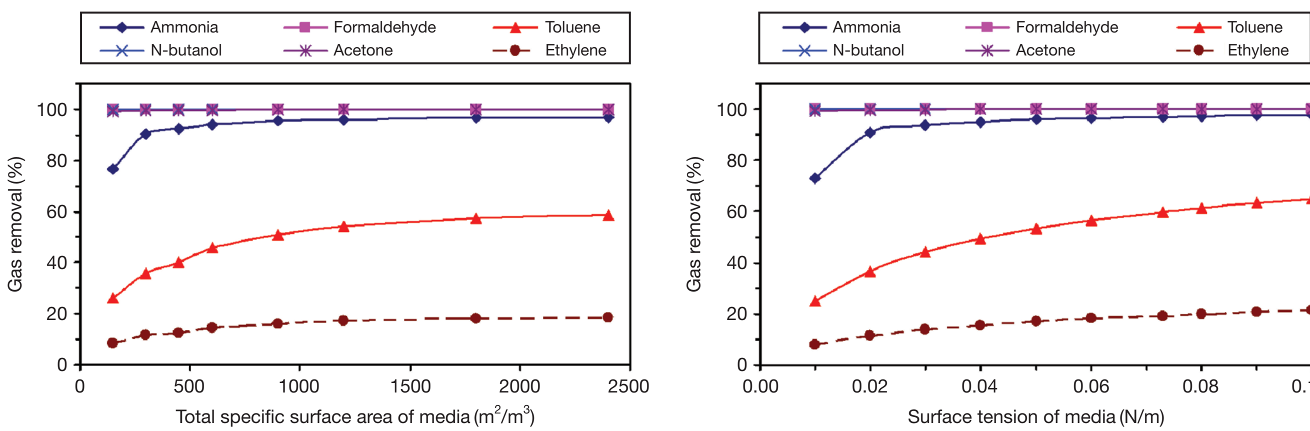

Fig. 5. Gas removal as a function of total specific surface areas (Left) and media surface tension (Right).

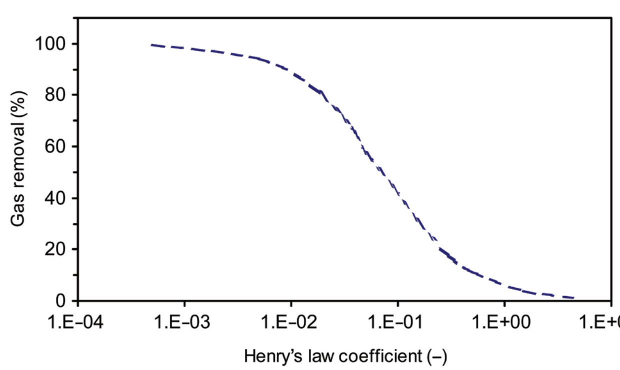

Fig. 6. Gas removals with variation of Henry's constants at 10 L/h in water flow and 7 L/min in gas flow.

The left plot of Fig. 5 describes an increasing tendency of gas removals as total media surface areas increase, but the increasing rate is reduced as surface area increases. Even if this figure uses the wet surface area, the model can calculate wetted surface area. The right plot of Fig. 5 shows the effects of media surface tension effects to the gas removal. Since water surface tension is about 0.77 N/m^2, wetting ability decreases as surface tension decreases. In this graph, surface tension effects are not so significant if tension value is not too low. Finally, Fig. 6 ensures the significance of Henry's constant of toluene with respect to removal efficiencies. As the constant values decrease, removal effects highly increase. Therefore, it is found that the absorption effects incorporating Henry's law constant may be one of the most significant to determine the gas removal.

Discussion

Since Henry's coefficient of a gas contaminant has the highest effect on the gas removal, operator should be careful in the input of chemical with a Henry's coefficient value higher than 0.1. Especially, hydrocarbon chemicals such as methane and ethylene are expected to have low absorption effects at the normal operation condition, but fortunately they allow quiet low risks in air systems. Formaldehyde or N-butanol showed a high removal effects with high absorption due to low Henry's law coefficient values, but ammonia, acetone, and toluene showed variable removal effects under changing physico-chemical reaction conditions, which is however not sure because microbial parameters are unknown or uncertain in many cases.

Decreasing airflow rate may be one of the easiest ways if overall gas removal effect is too low. Even filter media contain a high specific surface area, homogenized wetting on the whole surface sites of media is also important to induce favorable absorption and microbial reaction. To increase wetted portion in a media, one may increase water flow rate. However, it may disturb the contaminant removal efficiency and decrease stability in reaction, because of decrease of water retention time and possible generation of biofilm shear detachment in a reactor. A better way to avoid these troubles may improve water injection method (i.e. application of rotating water nozzle). Also, filter media should not have too low surface tension to attract neighboring water molecules. According to the previous study of Ockeloen et al. [8], counter-current flow is not recommended because of lower removal effects. In future, the model will be verified with experimental data and, if needed, proper estimation of uncertain parameters with recent statistical models [12].

Conclusion

This study utilized a simple mathematical model to predict removals of gases (ammonia, formaldehyde, toluene, butanol, acetone and ethylene) through degradation by fixed-film microorganisms in bio-scrubbers. In spite of the simplicity and use of lumped parameters, the model results produced significant results in that moderately soluble compounds, ammonia or acetone, exhibited highly differentiated patterns of removals according to changes of media depth, gas/liquid flow rates. The significance of Henry constants in bio-scrubber performance should result from the strong dominance of absorption effects onto overall performances. In addition, strong gas removal dependency on microbial parameters implies requirement of enhancement in microbial degradation of the compounds. Intriguingly, the hypothetical media properties in this model study showed relative low influences on gas removals probably due to expectation of low wetting efficiencies at the given ranges of water flow rates.

Acknowledgements

This research was supported by Dongil Cultural Scholarship Foundation.

References

1. Rene ER, Veiga MC, Kennes C. Combined biological and physicochemical waste-gas cleaning techniques. J Environ Sci Health A Tox Hazard Subst Environ Eng 2012;47:920-939.
2. Devinny JS, Deshusses MA, Webster TS. Biofiltration for air pollution control. Boca Raton: CRC Press; 1999.
3. Ottengraf SPP, Konings JHG. Emission of microorganisms from biofilters. Bioprocess Eng 1991;7:89-96.
4. Ottengraf SPP. Exhaust gas purification in Biotechnology. In: Rehm HJ, Reeds G, Editors. Weinheim: CH Verlagsgesellsschaft; 1986.
5. Diks RMM, Ottengraf SPP. Verification studies of a simplified model for the removal of dichloromethane from waste gases using a biological trickling filter (Part I). Bioprocess Eng 1991; 6:93-99.
6. Diks RMM, Ottengraf SPP. Verification studies of a simplified model for the removal of dichloromethane from waste gases using a biological trickling filter (Part II). Bioprocess Eng 1991; 6:131-140.
7. Alonso C, Suidan MT, Kim BR, Kim BJ. Dynamic mathematical model for the biodegradation of VOCs in a biofilter: Biomass accumulation study. Environ Sci Technol 1998;32:3118-3123.
8. Ockeloen HF, Overcamp TJ, Grady Jr CPL. Engineering model for fixed film bioscrubbers. J Environ Eng 1996;122:191-198.
9. Onda KH, Takeuchi H, Okumoto Y. Mass transfer coefficients between gas and liquid phases in packed columns. J Chem Eng Jpn 1968;1:56-62.
10. Perry JI. Trace chemical contaminant generation rates for spacecraft contamination control system design. NASA Technical Memorandum 108497; 1995.
11. NASA. Baseline values and assumption document. NASA CR-2004-208941; 2004.
12. Kahng M-W. Influence and leverage measures in nonlinear regression. QBS 2017;36:123-126.