Research Paper

Model-based evaluation of ammonia removal in biological air scrubbers

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A mechanistic model for ammonia removal in a countercurrent biological air scrubber was set up. This model was used to study the effect of the influent characteristics — air temperature, ventilation rate and ammonia load, on ammonia removal efficiency. Besides mass balances of the components participating in the biological conversions, the water mass balance and the heat balance were considered. The effects of the pH and the concentration of the nitrogen components on the driving force for mass transfer were examined. The model output was compared against experimental data from a pig housing facility. Simulations were performed to assess the usefulness of pH control and to investigate the effect of inflow air conditions on the ammonia removal efficiency. The study found out that although pH control affected the nitrogen component distribution in the washing water, it hardly affected the ammonia removal efficiency. Thus, pH control for biological air scrubbers is not recommended in practice, however, an on/off pH control system adding only acid at critical moments (pH above 7.5) could be considered. The variations in the ammonia removal efficiency are mainly caused by a changing ventilation rate rather than air temperature fluctuations or ammonia load.

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1. Introduction

Biofiltration by biological air scrubbers or so-called bio-trickling filters has evolved to an off-shelf technique to treat exhaust air from industries but also from mechanically ventilated animal houses, as they have been successfully applied for large air streams with low concentrations of pollutants (Cox & Deshusses, 1998). The contaminated air is passed through a packed bed where soluble and biodegradable gases such as ammonia or volatile compounds are absorbed into a biofilm and converted to harmless forms. Despite the relatively simple operating principle of biofiltering filters, biofiltration is a complex process involving several physical, chemical and biological interactions.

In biological air scrubbers, regular discharge of the washing water is needed for proper operation. The moment of
discharge can be automatically controlled by measuring electrical conductivity (EC), as it is correlated to the total nitrogen (TN) concentration, including ammoniacal, nitrite and nitrate nitrogen (Melse, Ploegaert, & Ogink, 2012b). When the predefined EC setpoint value is reached or exceeded, a fixed volume of water is discharged from the buffer tank using a time-controlled valve and replaced by fresh water. At most installed biological air scrubbers, the discharged volume is kept relatively small. In this way the EC or nitrogen content in the buffer tank decreases only slightly and will fluctuate close around the predefined setpoint. It can be said that the EC is kept nearly constant. Additionally, pH is measured as it must remain between 6.5 and 7.5 to maintain proper operation (InfoMil, 2015; MB31/05/2011). However, biological air
scrubbers installed at pig housing facilities in Flanders (Belgium) and the Netherlands most often do not have a pH control system. Ammonia absorption and nitrification have a counteracting effect on pH, keeping the pH rather constant under normal operating conditions. In Germany, biological air scrubbers mostly have a pH dosing system, applying acid or base when necessary (DLG, 2014). In this paper, particular attention was paid to the effect of pH and the concentration of nitrogen components in the washing water on the driving force for mass transfer. The nitrification of a batch test was simulated to study the effect of the decreasing pH and the decreasing nitrogen concentrations on the driving force for mass transfer. Additionally, the total behaviour of a biological air scrubber was described through a mechanistic model. Ammonia removal, temperature and evaporation dynamics were described through mass and energy balances. Simulations were carried out, comparing open-loop operation (i.e. without pH control) to that with closed-loop operation (i.e. with pH control).

Continuous short-term and long-term ammonia measurements at biological air scrubbers applied in pig housing facilities showed a daily and seasonal pattern in the ammonia removal performance (Melse, Hofschreuder, & Ogink, 2012a; Van der Heyden, Brusselman, Volcke, & Demeyer, 2016a). The electrical conductivity was kept nearly constant. These changes could be attributed to a fluctuating ventilation rate, ammonia loading rate or air temperature. Because of the interrelations between ventilation rate, air inlet temperature and loading rate, it is not fully clear which are the primary variables responsible for the variation in biotrickling filter performances. The ventilation rate in a pig housing facility determines the air flow rate through the air scrubber and is controlled depending on the air temperature inside the housing facility (Van Gansbeke, Van den Bogaert, & Vettenburg, 2009). Besides, an increased ventilation rate results in a lower ammonia inlet concentration due to dilution if the emission rate is kept constant. Nevertheless, it is possible that with an increased ventilation rate, more ammonia is extracted from the building due to an increased ammonia emission by emitting surfaces or due to increased defecation and urination (Melse et al., 2012b). The air flow through an air scrubber is inversely proportional to the empty bed residence time (EBRT), which reflects the contact time through the air scrubber, and thus may negatively affect the ammonia removal efficiency. At higher air temperatures, the water temperature will increase, making ammonia less soluble (higher H) and in turn reducing the driving force to the liquid phase. Melse et al. (2012b) performed a long term measurement campaign on a biological air scrubber for a pig housing facility and hypothesized that air temperature is the main influencing disturbance variable, rather than the changing ventilation rate and ammonia load. However, it is difficult to test the relative importance of these variables in practice as they are interrelated. By using the biological air scrubber model, these variables can be tested independently. In this paper, the biological air scrubber model, with pH control, was used to study the effect of the influential characteristics — air temperature, ventilation rate and ammonia load, on ammonia removal efficiency.

2. Theory

2.1. Operating principles of a biological air scrubber

The physicochemical and biological conversions associated with a biological air scrubber for ammonia removal are schematically presented in Fig. 1. Ammonia is absorbed from the gas phase into the liquid phase, where a chemical equilibrium with ammonium is established based on the pH. The total ammoniacal nitrogen (TAN; ammonia and ammonium) concentration is reduced by nitrification, i.e. biological conversion of ammonia to nitrite and subsequently to nitrate. The total nitrogen (TN; ammonia, ammonium, nitrite and nitrate) content in the washing water is controlled by discharge to keep a nearly constant concentration. Sometimes the pH is controlled by dosing acid and/or base to keep a constant pH value.

The essential concepts to describe the gas–liquid mass transfer are Henry’s law and the two-film model (Lewis & Whitman, 1924). The driving force (DF) for ammonia mass transfer from the gas to the liquid phase is determined by the difference between the equilibrium ammonia concentration in the liquid phase (\(C_{\text{L}} \)), and the actual ammonia concentration in the liquid phase (\(C_{L,NH_3}\)). The former is determined by dividing the ammonia concentration in the gas phase (\(C_{G,NH_3}\)) by the Henry’s law volatility constant (K_H). The latter is a function of the total ammoniacal nitrogen (TAN) concentration (\(C_{L,TAN}\)), the pH and the acid dissociation constant, as described in Eq. (1):

\[
DF = C_{L,NH_3} - C_{L,NH_3} = \frac{C_{G,NH_3}}{K_H(T)} - C_{L,NH_3} \\
= \frac{C_{G,NH_3}}{K_H(T)} - C_{L,TAN} \left(1 - \frac{C_{G,NH_3}}{C_{H^+} + K_a[NH_3](T)}\right)
\]

where \(C_{L,NH_3}\) is the free ammonia concentration in the liquid phase, \(C_{G,NH_3}\) is the ammonia concentration in the gas phase,

![Fig. 1 – Physicochemical and biological conversions in a biotrickling filter for ammonia removal. Nitrification by AOB (ammonia-oxidizing bacteria) and NOB (nitrite-oxidizing bacteria) results in a reduction of the ammonia concentration (A) as well as in a pH decrease (B). Absorption of ammonia from the gas into the liquid phase results in a pH increase (C).](image-url)
Fig. 1), additionally increasing the driving force for ammonia mass transfer from the gas phase to following equation:

\[ Q_d = \frac{C_{LN}^{\text{in}} \cdot Q_{\text{vent}} \cdot \eta}{100} \]  

where \( Q_{\text{vent}} \) is the ventilation rate and \( \eta \) is the ammonia removal efficiency.

In a biotrickling filter, the bacteria carrying out the biological conversions are mostly attached to the packing material. The retention of bacteria on the biotrickling filter packing was modelled by including a retention factor in the biomass material. The applicability of this type of zero-dimensional models for the description of a moving bed biofilm reactor has been demonstrated before (Plattes, Henry, & Schosseler, 2008). Two types of bacteria were considered in the biofilm and in the liquid phase, namely ammonia-oxidizing bacteria \( (X_{\text{AOB}}) \) and nitrite-oxidizing bacteria \( (X_{\text{NOB}}) \).
The individual biomass mass balance in cell \( i \) was implemented as
\[
\frac{dX_i}{dt} = \frac{r Q_i}{V_{L,cell}} \cdot (X^{i-1} - X^i) + r_b
\]  
(3)
where \( r \) is the biomass retention factor, \( Q_i \) is the liquid flow rate and \( V_{L,cell} \) is the volume of each cell. \( r_b \) denotes the conversion rate of component \( b \) (Eq. (4)), which is calculated from the process rates and the stoichiometric matrix (Supplementary Information A – SA3, Table S1)
\[
r_b = \sum A_{bi} \cdot R_j
\]  
(4)
where \( A_{bi} \) is the stoichiometric component of component \( b \) and process \( j \), and \( R_j \) is the reaction rate for process \( j \).

The process rates are expressed by a Monod-equation taking into account the affinity for \( O_2 \) and \( NH_3 \) during nitrification and for \( O_2 \) and \( NO_2 \) during nitratation. Inhibition by free ammonia (FA) and free nitrous acid (FNA) was assumed to be negligible in this study as nitrite was not found to accumulate in the study of Melse et al. (2012b). For biomass decay, the decay rate is directly proportional with the biomass concentration. The stoichiometric and kinetic parameters related to AOB and NOB growth are summarized in Supplementary Information A – SA3 (Table S2). The effect of temperature (\( T \)) on the maximum specific growth rate and decay rate (\( r_c \)) is taken into account, considering the Arrhenius equation:
\[
r_T = r_{293} \cdot \exp \left( \frac{-E_{act} \cdot (293 - T)}{R \cdot 293 \cdot T} \right)
\]  
(5)
where \( r_{293} \) is specific growth/decay rate at 20 °C, \( E_{act} \) is the activation energy, \( R \) is the gas constant and \( T \) is the temperature.

Absorption of ammonia and the biological conversion reactions that involve proton consumption or production affect the pH of the medium. Vice versa, pH influences the biological conversion rates both through a direct effect on the maximum growth rates (Van Hulle et al., 2007) and indirectly through the chemical equilibria of the species involved. pH was therefore added in the model as a state variable and calculated by chemical equilibria of the species involved. pH was therefore also considered, containing a total nitrogen content of 3.2 gN.L\(^{-1}\) (i.e. the maximum value allowed in Flanders) (MB31/05/2011), distributed as 50% TAN and 50% nitrate. An ammonia gas concentration of 20 ppm was assumed; the corresponding equilibrium liquid concentration was considered in the calculation of the driving force for ammonia mass transfer (Eq. (1)). A temperature of 20 °C was assumed. The change in pH was iteratively calculated through the charge balance method (Supplementary Information A – SA4, Eq. S9), considering also the bicarbonate/carbon dioxide equilibrium. A total inorganic carbon (TIC) concentration of 500 gC/m\(^3\) was considered, assuming that the washing water is saturated with CO\(_2\). The washing water then has a large buffer capacity resulting only in a pH change due to nitrification and not by CO\(_2\) absorption.

Subsequently, the biological air scrubber model was used to simulate some scenarios. A reference case was defined based on the biotrickling filter studied by Melse et al. (2012b). It consists of a countercurrent biotrickling filter used to treat the exhaust gas of a pig housing facility of 600 sows, with a maximum of 40000 m\(^3\).h\(^{-1}\) ventilation air. Discharge was automatically controlled using an electrical conductivity (EC) setpoint around 16 mS.cm\(^{-1}\) to limit the total nitrogen content in the washing water, which varied between 1775 and 4817 gN.m\(^{-3}\). The discharge rate was on average 0.33 m\(^3\).d\(^{-1}\). The inlet conditions were set according to constant values, representing the average values during the 141 days measurement period of Melse et al. (2012b) (Table 1). In the model, the maximum allowed total nitrogen content in the washing water \((C_{BT,TN}^{\text{max}})\) was set at 3200 gN.m\(^{-3}\), the setpoint in Flanders (MB31/05/2011). Additionally, this value is comparable to the total nitrogen concentrations of the washing water in the study of Melse et al. (2012b). The minimal total nitrogen concentration in the buffer tank after discharge \((C_{BT,TN}^{\text{min}})\) was set to 3150 gN.m\(^{-3}\). The discharge rate to keep the total nitrogen concentration below this setpoint amounts to 0.80 m\(^3\).d\(^{-1}\) (Eq. (2)). The initial conditions were set according to the values in Table 2. The initial values of AOB and NOB were estimated based on prevailing concentrations of bacteria in activated sludge systems.

As pH was not controlled in the study of Melse et al. (2012b), the reference case behaviour was therefore simulated with the model including pH dynamics. The initial pH was set to 6.6, which is the average pH value measured (Melse et al., 2012b). The simulation results of the reference case were compared to the measurement results of Melse et al. (2012b). As the measurement results were only used as input for the model simulation and were not used for parameter estimation, only an intuitive qualitative comparison was made between the model and the measurement results. The
4. Results and discussion

4.1. Effect of pH and TAN concentration on mass transfer driving force — simple representation

The total nitrogen content in the washing water is set through the discharge setpoint and corresponds with a certain TAN concentration in the washing water, low enough. The lower the pH, the higher the total nitrogen concentration allowed in the washing water to still keep a high driving force for ammonia transfer. At a pH of 4 or lower, as applied in a chemical air scrubber, all total ammoniacal nitrogen is in the form of ammonium. Therefore, the total allowed nitrogen content in the washing water may be significantly higher than in biological air scrubbers (58 gN.L\(^{-1}\); MB31/05/2011), reducing the discharge rate significantly (Eq. (2)). In a biological air scrubber, such high total nitrogen concentrations, and thus total ammoniacal nitrogen concentrations, cannot be allowed. As the pH lies only around 7, a slight increase can immediately result in a strongly reduced driving force.

4.2. Effect of nitrification on mass transfer driving force — batch test

Figure 3 represents the nitrification of a batch of washing water with a total ammoniacal nitrogen concentration of 1600 gN.m\(^{-3}\). The total effect of nitrification concerns both a decreased total ammonium concentration and a decreased pH.
The driving force for ammonia transfer (DF) increases with increasing ammonium conversion, until it reaches the maximum value equalling $C^*_{\text{L,NH}_3}$; at this point, no free ammonia (FA) is present in the liquid as the pH is low ($\text{pH} < 4$) and the ammonia/ammonium equilibrium is shifted to ammonium ($pK_{\alpha,\text{NH}_3} = 9.4$ at 20 °C).

The effect of nitrification on the driving force for ammonia mass transfer (DF) was also studied for a constant pH, considering only the decreased total ammonium concentration (dashed lines). In this case, the driving force increases linearly with an increasing ammonium conversion, at a gradual manner (i.e. small slope). Comparing the two scenarios, it is clear that the effect of nitrification on the driving force for ammonia mass transfer is mainly due the associated pH decrease rather than due to the reduction of the total ammonium concentration in the liquid.
The effect of pH will even be more pronounced when the initial pH of the liquid batch is higher because the initial driving force is smaller. If 1% is nitrified at an initial pH of 7.5, this induces a decrease of pH by only 0.2, but this doubles the initial driving force. In practice, the pH will not drop below 4 as nitrification has an optimum pH and is inhibited at low pH (Van Hulle et al., 2007). These results only represent a batch test. During normal operation of an air scrubber, absorption of ammonia from the gas to the liquid phase will again increase the pH of the washing water.

4.3. Reference case — open-loop operation

The scenario without pH control resulted in a steady state ammonia removal efficiency of 92%. The total ammoniacal nitrogen, total nitrite and nitrate concentrations were 1.75, 0.001 and 1.45 gN.L$^{-1}$, respectively, corresponding with a total nitrogen concentration of 3.2 gN.L$^{-1}$ (Van der Heyden et al., 2016b). The steady state simulation results agree well with the measurement data of Melse et al. (2012b) (Fig. 4, Table 3). The slightly higher simulated ammonia removal, compared to the average measured value over the entire measurement period, can be attributed to the effect of pH, as explained in Section 3.1. As nitrification produces two moles of protons, while absorption only consumes one mole of protons, the bacteria can only convert half of the absorbed ammonia. At steady state, an equilibrium between ammonia absorption and nitrification is reached at a pH of 6.2.

The nitrate concentration is high (44.7% of the total nitrogen concentration) while the nitrite concentration in the washing water is very low (0.8% of the total nitrogen concentration), indicating that almost all nitrite, which is formed by the AOB, is immediately converted by NOB. The AOB concentration is higher than the NOB concentration, as AOB have a higher specific yield than NOB (Supplementary material, Figure S1).

The gas temperature inside the air scrubber decreased from 20°C at the inlet to 15°C at the outlet, thereby heating up the colder washing water to 15°C. The relative humidity increased from 65% to almost 100%. These temperature and relative humidity profiles are similar to the ones for the chemical air scrubber model (Van der Heyden et al., 2016b).

Table 3 — Comparison between measured data (Melse et al., 2012b) and steady-state simulation results for the reference case without pH control and with pH control.

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Unit</th>
<th>Measured (Melse et al., 2012b)</th>
<th>Simulated without pH control</th>
<th>Simulated with pH control</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\eta$</td>
<td>%</td>
<td>82</td>
<td>92</td>
<td>94</td>
</tr>
<tr>
<td>$C_{\text{NH}_3}$</td>
<td>ppm</td>
<td>2.4</td>
<td>1.0</td>
<td>0.82</td>
</tr>
<tr>
<td>$Q_{\text{D}}$</td>
<td>m$^3$.d$^{-1}$</td>
<td>0.33</td>
<td>0.87</td>
<td>0.80</td>
</tr>
<tr>
<td>$R_{\text{HG}}$</td>
<td>%</td>
<td>&gt;95</td>
<td>99.2</td>
<td>99.2</td>
</tr>
<tr>
<td>$T_{\text{G}}$</td>
<td>°C</td>
<td>16</td>
<td>15</td>
<td>15</td>
</tr>
<tr>
<td>$T_{\text{BLT}}$</td>
<td>°C</td>
<td>16</td>
<td>15</td>
<td>15</td>
</tr>
<tr>
<td>pH</td>
<td>pH in water buffer tank</td>
<td>6.6</td>
<td>6.2</td>
<td>6.6</td>
</tr>
</tbody>
</table>
period (efficiency: 92% simulated versus 82% measured), could be attributed to dynamics in the operation of the air scrubber of Melse et al. (2012b). For instance, the total nitrogen content was not constant during the measurement campaign. Therefore, the average total nitrogen content was also slightly higher than the used value in the simulations (Fig. 4). The distribution of the nitrogen components in the water was similar, with only half of the total nitrogen that could be converted to nitrate. This was also observed during other measurement campaign (Melse & Mosquera, 2014; Mosquera et al., 2011; Ottosen et al., 2011; Van der Heyden et al., 2016a). The simulated results thus agree well with the experimental data from a conventional pig housing facility.

### 4.4. Effect of pH control — closed-loop operation

When the pH is controlled at a constant value, almost all (99%) absorbed ammonia is converted to nitrate (Fig. 4), instead of only 44.7% when pH is not controlled. With pH control, nitrification is not inhibited by the decreasing pH, resulting in full conversion of ammonium to nitrate. Through dosing of a base, the pH is kept around 7, thereby allowing nitrification to exceed ammonia absorption. Because of this, the biomass can have higher growth compared to the case without pH control, resulting in a higher AOB and NOB concentration (Supplementary Information B — Figure S1). Additionally, at the same pH, the driving force will be higher compared to the situation without regulated pH, as the TAN concentration is much lower (Fig. 4). However, only a slight increase in the ammonia removal efficiency is observed overall (94% compared to 92%).

In a lab scale study by Blázquez et al. (2017), it was shown that a biotrickling filter treating ammonia gas concentration of 100 ppm could reach 100% ammonium conversion to nitrate (i.e. complete nitrification) by keeping the pH constant around 7. An on/off pH control system based on the addition of NaOH (0.5M) or HCl (0.5M) was used. For this high ammonia gas concentration, this type of control system could be beneficial.

The question arises to which extent the operation of a biological air scrubber with constant pH is different from a chemical air scrubber and if it is worth applying in practice. In a biological air scrubber the pH is only controlled around 7 compared to 4 in a chemical air scrubber. For each ammonia molecule that is absorbed, a net 1 mol of proton is formed after complete nitrification, which must be neutralised with a base (Supplementary Information C). This means that the base consumption in a biological air scrubber is as high as the acid consumption in a chemical air scrubber, increasing the operational costs substantially. Additionally, the ammonia removal efficiency only increases slightly using the base dosing system in biological air scrubbers (94% compared to 92%). In a chemical air scrubber, at a pH of 4, all total ammoniacal nitrogen is in the form of ammonium. Therefore, the total allowed nitrogen content in the washing water may be significantly higher than in biological air scrubbers (58 gN.L⁻¹ compared to 32 gN.L⁻¹) (MB31/05/2011), reducing the discharge rate significantly (Eq. (2)). In a biological air scrubber, these high total nitrogen concentrations cannot be allowed as this would inhibit the bacteria. As the pH lies only around 7, a slight increase can immediately result in a reduced driving force. Controlling the pH at a constant value around 7 by neutralising all formed protons, thus seems not a viable option to implement in practice in pig housing facilities.

However, an on/off pH control system adding only acid at critical moments (pH above 7.5) could be considered in practice. The system then only serves as a back-up if the pH increases too far. Only a high pH has a direct negative impact on the ammonia removal efficiency (Fig. 2).

### 4.5. Effect of inlet conditions

Figure 5 shows the influence of the inlet conditions: air temperature (°C), ventilation rate (m³.h⁻¹) and ammonia load (gN.h⁻¹) on the ammonia removal efficiency.

Figure 5B shows a decreased ammonia removal efficiency resulting in a higher concentration of ammonia compared to ammonium. This results in a decreased driving force as well as increased ammonia availability for the biomass. Besides, the specific growth rate and decay rate of the biomass increases (higher $\mu_{\text{max}}$ and $b$). Additionally, the mass transfer coefficient will increase (higher $K_{\text{L}}$), implying that more ammonia will be transferred from the gas phase to the liquid phase which has a positive effect on the removal efficiency. As a result of all the different effects of temperature, the total effect on the ammonia removal efficiency is less than 1% over the considered temperature range.

Figure 5C shows a decreased ammonia removal efficiency at an increased ventilation rate. At a higher ventilation rate or...
air flow rate through the air scrubber, the incoming ammonia load increases but the EBRT (empty bed residence time = contact time) decreases. The net effect of both is a decreased ammonia removal efficiency. When keeping a constant ventilation rate but increasing the inlet ammonia load by increasing the inlet ammonia concentration (Fig. 5C), the outlet ammonia concentration increases but the removal efficiency remains constant.

The effects of air temperature, ventilation rate and load on the ammonia removal efficiency are similar to those observed for a chemical air scrubber (Van der Heyden et al., 2016b). Overall, considering these simulation results, it is the ventilation rate rather than the inlet air temperature or influent load that influences the ammonia removal efficiency in biological air scrubbers.

5. Conclusions

- A mechanistic model for a biotrickling filter for ammonia removal was set up, taking into account temperature, relative humidity and pH dynamics. The simulation results were in agreement with experimental data from a conventional pig housing facility.
- The positive impact of nitrification on the driving force for mass ammonia transfer was due to the associated pH decrease rather than to the reduction of the total ammoniacal nitrogen content as such. The total nitrogen concentration in the washing water still needs to be kept sufficiently low to sustain the driving force for ammonia from the gas phase to the liquid phase.
- The application of pH control affected the nitrogen component distribution in the washing water. Without pH control, only half of the ammonia absorbed in the washing water could be oxidized by the biomass. When the pH was controlled at a constant value which is sufficiently low to allow ammonia mass transfer to the liquid phase but still high enough for bacterial growth, almost complete ammonium conversion could be achieved.
- The ammonia removal efficiency as such was hardly affected by pH control, while operational costs for base dosing are substantial. Applying pH control at biological air scrubbers in practice is therefore not recommended. However, an on/off pH control system adding only acid at critical moments (pH above 7.5) could be considered in practice.
- Simulation results clearly demonstrated that variations in the ammonia removal efficiency are mainly caused by a changing ventilation rate rather than by fluctuations in air temperature or ammonia load.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.biosystemseng.2019.12.011.

References


Author contribution

All authors have participated in (a) conception and design, or analysis and interpretation of the data; (b) drafting the article or revising it critically for important intellectual content; and (c) approval of the final version.


