APPARATUS FOR DOWN-WELL OXYGEN TRANSFER INTO CONTAMINATED AQUIFERS

By Jae ho Bae,1 Lewis Semprini,2 and Perry L. McCarty,3 Member, ASCE

ABSTRACT: In-situ methanotrophic bioremediation of ground water contaminated with petroleum hydrocarbons and chlorinated aliphatic hydrocarbons (CAHs) often require the addition of O2 for compound oxidation. The purpose of this research was to test the feasibility of a down-well gas-transfer apparatus for adding O2 directly into recirculating ground water. At times it may be desirable to add other gases as well, such as methane for CAH cometabolism. A high downward velocity of water through the inlet tube of an inverted cone prevented upward movement of injected O2 bubbles. The addition of gas-liquid mixing cones within the transfer section of the inverted cone increased the turbulence below the inlet. Complete dissolution of gases is possible. The relationships between O2 transfer rate and system variables such as pressure loss, water flow rate, and N2 content in the influent water were evaluated. A significant factor affecting O2 mass transfer was the total dissolved gas content in the water entering the transfer device. From the results of this study, a design for practical conditions of operation was developed.

INTRODUCTION

Chlorinated aliphatic hydrocarbons (CAHs) and aromatic hydrocarbons are common ground-water contaminants (Barbash and Roberts 1986). For in-situ aerobic bioremediation, bacteria have been found capable of transforming some CAHs such as trichloroethylene (TCE), cis- and trans-1,2-dichloroethylene (c-DCE and t-DCE), or vinyl chloride (VC) through cometabolism (Semprini et al. 1991). For aerobic bacterial activity, the addition of O2 is necessary to serve as an electron acceptor. However, little attention has been paid to the difficulties in adding O2 or other possible gases to ground water contaminated with volatile compounds, which can inadvertently be transferred to the atmosphere by air sparging directly into an aquifer or within a well casing. A possible method to avoid contaminant volatilization is to add pure oxygen directly into a well where ground water is being recirculated (Herrling et al. 1991). A recirculation system for the methanotrophic cometabolic treatment of CAHs, which requires both oxygen and methane, is shown in Fig. 1 (Semprini et al. 1992). The purpose of this study was to evaluate the potential of one possible apparatus for transferring oxygen into contaminated ground water that is caused to circulate through a well in order to prevent contaminant volatilization or dilution. The apparatus for down-well gas transfer (ADGT) combines principles of downflow bubble contact aeration (DBCA), as discussed by Speece et al. (1971), and two-phase fluid mixing as described by J. S. Lang ["Method of mixing fluids in packing media for reactors." U.S. Patent No. 5,171,544 (1992)].

MATERIALS AND METHODS

The laboratory experimental system used to evaluate the ADGT is illustrated in Fig. 2. All experiments were conducted at room temperature (20°C), but the temperature of the water circulated through the ADGT was maintained at 25 ± 2°C. Water pumped from the reservoir flowed first to an air stripping tower (17 cm in diameter and 170 cm in height, filled with 1.6-cm-diameter and 1.6-cm-long flex rings), where a vacuum was applied to remove dissolved gases. In some experiments, in order to study the effect of N2 content on O2 transfer efficiency, air or N2 was added 10 cm above the water outlet of the air stripping tower at flow rates of 1.200 and 3,800 mL/min, respectively. Water leaving the tower and entering the ADGT had dissolved O2 and N2 concentrations near saturation with respect to the partial pressure applied.

The ADGT for an operating system (Fig. 2) was modified for the laboratory study as illustrated in Fig. 3. The ADGT consists of a restriction section for gas addition, and a gas-transfer section. The gas-transfer section was fabricated from a polycarbonate Iwshoff settling cone (VWR, San Francisco) cut to fit the system (Fig. 3). The height was 34 cm, and the diameter of inlet and outlet were 0.95 cm and 7.6 cm, respectively. Preliminary experiments indicated that the oxygen transfer efficiency of the ADGT could be improved by the addition of cones containing small holes through which both water and gas passed. A similar arrangement was described by Lang (1992). The effect on oxygen transfer efficiency of different numbers of cones (zero to three), and the number and size of holes in each cone, was evaluated. Here, the size and number of holes were varied while keeping the total hole area the same in an attempt to determine an optimum arrangement for design. When the cones were used, transfer occurred both above and below the cones. With a relatively high velocity of water through the restriction section, good initial mixing of water and gas occurred even above the first cone. Below each cone, water-gas jets created by the small holes created vigorous turbulent mixing and a fine bubble swirl to further enhance gas transfer. Such bubble swarms could not escape upward due to the high downward water flow velocity. The volume of space between cones was about 50 mL. The volume between the entrance and first cone was about 50 mL with more than two cones, and about 100 mL for one cone. The oxygenated water exiting the gas-transfer section flowed downward through a 100-cm-long by 7.62-cm-diameter section, and then back into the recirculation reservoir.

Gaseous O2 was introduced from an O2 reservoir, pressurized above 1.0 atm pressure with a 3.7-m-high column of water (36.5 kPa), into the restriction area in the ADGT through a 20-gauge needle located 7.6 cm above the gas-transfer section. The O2 addition rate was controlled by a valve. The mass of O2 added into the system over a given time period

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Note. Discussion open until January 1, 1996. To extend the closing date one month, a written request must be filed with the ASCE Manager of Journals. The manuscript for this paper was submitted for review and possible publication on January 25, 1993. This paper is part of the Journal of Environmental Engineering, Vol. 121, No. 8, August, 1995. ©ASCE, ISSN 0733-9372/95/0008-0565—0570/$2.00 + $2.50 per page. Paper No. 5414.
The DO probe was standardized as described in Standard (1992). In an alternative head-space method, the O₂ and N₂ compositions of water samples taken directly from sampling ports were determined as follows: a 125-mL bottle was filled with sample (no head space) and capped with a sleeve-type septum. Ten mL of helium (He) was added with a syringe into the bottle while simultaneously taking out 10 mL of water. The bottle was mixed on a shaker table for 90 minutes at 20°C to equilibrate gases between phases. Head space was analyzed for O₂, N₂, and He with a Gas Partitioner (Fisher Scientific model 25 V, Pittsburgh). The concentration of O₂ and N₂ in the original water was calculated based on mass balances using gas partial pressures and gas solubilities reported by Stephan and Stephan (1963).

As pumping costs are directly related to pressure loss, the pressure loss caused by the gas-transfer device was measured. Here, pressure gauges were installed 18 cm above the gas introduction port on the ADGT and 11 cm below the ADGT exit. The difference between gauge readings minus the water head between the gauges was taken as the pressure loss through the ADGT. Velocity head differences were small and thus neglected in this analysis.

EXPERIMENTAL RESULTS

Factors affecting gas transfer, such as bubble size, turbulence in the mixing zone, water flow rate, and inlet dissolved gas concentration, were evaluated. In addition, the overall mass-transfer coefficient for O₂ was evaluated and compared with reported values for other types of gas-transfer equipment. Following the vigorous mixing in the transfer section, very fine gas bubbles flowed into the exit tube, and generally disappeared within the first 60 cm. For experiment 1, DO transfer was determined, first, for the case where added O₂ was completely dissolved, and second, for the non-steady-state case where complete dissolution did not occur, and bubble buildup in the gas-transfer section took place. The maximum steady-state transfer capacity of the ADGT lies somewhere between these two values. For the other experiments, the highest DO addition that could be obtained without the accumulation of bubbles in the gas-transfer section was determined, and these are the values presented in the following.

Experiment 1—Effect of Cone Number and Hole Size

Experiment 1 was conducted to evaluate the effect of the number of cones and the size of cone holes. In addition, the effect of O₂ bubble size at the gas inlet was examined (Table 1). The total cross-sectional area of the holes in a given cone was kept constant at 0.31 cm². The number of holes used was

<table>
<thead>
<tr>
<th>Experiment</th>
<th>Number of holes</th>
<th>Size of holes (mm)</th>
<th>Number of cones</th>
<th>Water flow rate (L/min)</th>
<th>Influent DO (mg/L)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1-1</td>
<td>2</td>
<td>0.44</td>
<td>0</td>
<td>3.04</td>
<td>2.7</td>
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<tr>
<td>1-2</td>
<td>2</td>
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<td>0</td>
<td>3.04</td>
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<td>2</td>
<td>0.44</td>
<td>3</td>
<td>3.04</td>
<td>2.8</td>
</tr>
<tr>
<td>1-6</td>
<td>2</td>
<td>0.28</td>
<td>2</td>
<td>3.15</td>
<td>2.7</td>
</tr>
<tr>
<td>1-7</td>
<td>2</td>
<td>0.20</td>
<td>2</td>
<td>3.10</td>
<td>2.1</td>
</tr>
</tbody>
</table>

*For Experiment 1-1, O₂ was injected with a cylindrical Hastaalloy C Mobil Phase Filter with pore size of 2 μm (Altech, Deerfield, Ill.) installed in the gas-transfer device. A 20-gauge needle was used for the rest of experiment 1. The absolute pressure applied to the air stripping tower ranged from 0.26 to 0.32 atm.
two, five, and 10, for which the diameters used were 0.44, 0.28, and 0.20 cm, respectively. The bubble size at the gas inlet was varied using addition either through a 20-gauge needle or through a porous filter with 2 μm pore size (Table I).

Fig. 4 illustrates the effects of O₂ bubble size, the number of cones, and size of cone holes on effluent DO for steady-state DO addition. Comparison between results of experiments 1-1 and 1-2 reveals that O₂ addition with the needle was better than with the porous filter. Gas bubbles grew at the surface of the filter, and when released, were bigger than those generated with the needle. This resulted in incomplete oxygen uptake by the water, and gas accumulated in the ADGT. The addition of one cone increased effluent DO from 6.5 mg/L to 10.5 mg/L (experiments 1-2 and 1-3). Considering the difference between effluent DO (Fig. 4) and influent DO (Table I), DO increase with a single cone was twice that without it. Additional cones provided little additional advantage, but increased the pressure loss. Increasing the number of holes from two to five, with a two-cone system, did not have a significant effect on effluent DO, but, with 10 holes, a significantly higher DO resulted. This, however, was accompanied by a significant increase in pressure loss (41 kPa versus 6–13 kPa with fewer holes).

The Fig. 4 results indicate that mass transfer was always somewhat greater when gas bubbles were allowed to accumulate in the ADGT, but gas accumulation is not desirable for continuous operation. Thus, it should be noted that the desired operation without gas accumulation does not actually provide maximum gas-transfer conditions. From a mass balance, the measured DO increase was calculated to equal from 90 to 110% of the O₂ added as a gas. Thus, good mass balances were achieved in the experiments.

**Experiment 2—Effect of Water Flow Rate**

The effect of water flow rate on effluent DO concentration was evaluated with two-hole cones. Here, the absolute pressure maintained at the air stripping tower was about 0.26–0.32 atm, and the stripping-tower effluent DO was about 2.5–3 mg/L.

Generally, as water flow rate increased, a higher transfer-derived effluent DO was achieved [Fig. 5(a)]. This probably resulted from the higher velocity associated with the higher flow rates, which created greater turbulence in the mixing zone and greater energy loss. The net result was a linear increase in the O₂ addition rate with flow rate [Fig. 5(b)]. A change in the number of cones did not significantly affect the O₂-addition rate.

**FIG. 5. Effect of Water Flow Rate on O₂ Addition: (a) Water Flow Rate versus Effluent DO; and (b) Water Flow Rate versus O₂ Addition Rate**

Pressure loss is a function of flow rate, so DO change should also be related to pressure loss. Fig. 6(a) shows the relationship between the pressure loss and the flow rate for the two-hole cone system. An increase in the number of cones from two to three did not increase pressure loss significantly, nor did it increase the DO addition rate. Fig. 6(b) illustrates that as total pressure loss increased, O₂ addition rate also increased almost linearly for the two-hole systems. This effect is addressed more later.

**Experiment 3—Effect of Dissolved Gases**

An important factor affecting the ability to add O₂ is the dissolved gas content of the water. At equilibrium, the sum total partial pressures of the dissolved gases should equal the pressure on the system. Thus, theoretically, when water is less saturated with other gaseous components, more O₂ can be added. To illustrate this, the dissolved gas content in the influent water to a two-hole one-cone system was controlled by varying pressure and gas composition (N₂ or air) in the air stripping tower as summarized in Table 2. Dissolved nitrogen concentrations were thus varied in the experiments from 4.7 to 16.5 mg/L.

The gas stripper transfer efficiency was first determined by comparing gas concentrations in the gas stripper effluent (headspace method) with equilibrium values. In addition, DO determination by the head-space method was found to compare favorably with that by the probe method. For all cases stripping tower effluent DO and N₂ concentrations were close to the equilibrium values, within error ranges of ±10% (data not shown).

Fig. 7 illustrates the effect of N₂ concentration on DO concentration increase through the transfer device. As N₂ concentration increased, less DO was added (experiments 3-2 to 3-6). This occurs because dissolved N₂ exchanges with
gaseous O<sub>2</sub> in the mixing device, and the sum of the partial pressures of all dissolved gases cannot exceed the total pressure within the transfer device, which in this case was 1 atm. In the extreme case (experiment 3-6), with an influent equivalent to 0.9 atm partial pressure of N<sub>2</sub>, only about 1.8 mg/L of DO could be added. When air instead of N<sub>2</sub> was introduced into the air stripping tower at given absolute pressures, the DO increase was the same (compare experiment 3-1 with 3-2, and 3-3 with 3-4). This indicates that it is the total equivalent partial pressure of gases in the influent water rather than gas composition per se that governs the capacity for DO increase.

Fig. 7 also illustrates the relationship between DO increase and the theoretical maximum DO increase (TMDO) for all cases in experiment 3. Here, TMDO is assumed to equal \( (1 - P_{N_2} / P_N) C_s \), where \( P_N \) is the partial pressure of N<sub>2</sub> (atm) in the influent, and \( C_s \) is the saturation concentration for DO at the pressure in the transfer device (1 atm for this experiment). As shown, the DO added represented about 44–53% of TMDO. Thus, about a 50% transfer efficiency was achieved by the ADGT over a range of operating conditions.

### Mass Transfer and Energy Requirements

The overall mass-transfer coefficient for O<sub>2</sub> was estimated and compared with values for other aeration systems reported by Munz and Roberts (1982) and Roberts and Dandliker (1983). Assuming the transfer section to operate as a completely mixed reactor, a mass balance for O<sub>2</sub> at steady-state yields

\[
K_{t,a} = \frac{Q (C_{out} - C_{in})}{V (C_s^a - C_s)}
\]

where \( K_{t,a} \) = overall O<sub>2</sub> mass transfer coefficient (s<sup>-1</sup>); \( C_s^a = DO \) in the transfer section (mg/L); \( C_s^a = DO \) in equilibrium with the gas phase partial pressure (mg/L) \( C_s^a = P_{N_2}/H \), where \( H \) is Henry's constant; \( Q = \) flow rate (m<sup>3</sup>/s); \( V = \) volume of the transfer section (m<sup>3</sup>); \( C_{out} = DO \) in the influent (mg/L); and \( C_{in} = DO \) in the effluent (mg/L). For (1), some of the variables were estimated by indirect means as they were unknown. For example, the effective \( C_{eff} \) would range between \( C_{in} \) and \( C_{s,k} \), and the effluent \( P_{O_2} \), would range between (1 - \( P_{N_2} \)) and 1 atm. For modeling, however, it was assumed \( C_{eff} \) equaled \( C_{in} \) and \( P_{O_2} \), equaled (1 - \( P_{N_2} \)). Another unknown is \( V \), the volume of the reactor. Two values of V were considered, 0.5 and 3.2 L. The former represents the volume of the transfer section itself, and the latter represents the total volume of the system where dissolution of O<sub>2</sub> occurred (the transfer section plus the downward pipe volume). A smaller assumed mixing volume yields larger gas transfer coefficients \( K_{t,a} \).

The power required to overcome the transfer device pressure loss was estimated as follows:

\[
P_c = \frac{Q \Delta P}{P_c}
\]

where \( P_c = \) gas transfer power requirement (W); \( Q = \) flow rate (m<sup>3</sup>/s); and \( \Delta P = \) pressure loss through transfer device (Pa).

Fig. 8 illustrates the calculated \( K_{t,a} \) value for the two-hole one-cone ADGT used in experiments 2 and 3. The \( K_{t,a} \) is plotted versus the specific power dissipation (W/m<sup>3</sup>), which represents the power divided by the volume of the gas-transfer section (\( P_c/V \)), as described by Roberts and Dandliker (1983) and Roberts (1984). Here, the increased \( K_{t,a} \) was associated with an increase in the specific power dissipation, up to a maximum of 1,200 W/m<sup>3</sup>. The maximum \( K_{t,a} \), depending on assumptions made, varied between 0.02 and 0.2 s<sup>-1</sup>. The minimum and maximum values were calculated for transfer.

### Table 2: Experimental Conditions for Experiment 3

<table>
<thead>
<tr>
<th>Parameter</th>
<th>3-1</th>
<th>3-2</th>
<th>3-3</th>
<th>3-4</th>
<th>3-5</th>
<th>3-6</th>
</tr>
</thead>
<tbody>
<tr>
<td>Water flow rate (L/min)</td>
<td>4.9</td>
<td>4.9</td>
<td>4.9</td>
<td>4.9</td>
<td>5.0</td>
<td>5.0</td>
</tr>
<tr>
<td>Pressure in tower (atm)</td>
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<td>0.34</td>
<td>0.68</td>
<td>0.68</td>
<td>0.8</td>
<td>0.9</td>
</tr>
<tr>
<td>Gas to tower</td>
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<td>Air</td>
<td>Air</td>
<td>N&lt;sub&gt;2&lt;/sub&gt;</td>
<td>N&lt;sub&gt;2&lt;/sub&gt;</td>
<td>N&lt;sub&gt;2&lt;/sub&gt;</td>
</tr>
<tr>
<td>DO influent (mg/L)</td>
<td>2.5</td>
<td>0</td>
<td>5.1</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>N&lt;sub&gt;2&lt;/sub&gt; influent (mg/L)</td>
<td>4.7</td>
<td>5.4</td>
<td>9.2</td>
<td>12.0</td>
<td>14.7</td>
<td>16.5</td>
</tr>
</tbody>
</table>

### Fig. 7: Effect of Dissolved Gas Content: DO versus TMDO (Percent Represents Ratio of DO Added to TMDO)

![Graph showing the relationship between DO and TMDO for different conditions](image-url)
section volumes of 3.2 and 0.5 L, respectively. Experiment 3 results agreed with those from experiment 2, suggesting $K_a$ was not a function of dissolved gas composition, but of power supplied. For comparison, $K_a$ values between 0.0008 and 0.006 s$^{-1}$ were reported by Roberts and Dandliker (1984) for surface aeration and 0.002 to 0.01 s$^{-1}$ by Munz and Roberts (1982) for bubble-column aeration.

Also shown in Fig. 8 are surface aeration values versus specific power inputs (Roberts and Dandliker 1984). The higher $K_a$ values for the ADGT at the higher levels of specific power dissipation are consistent with that for surface aeration. Roberts (1984) found the $K_a$ increased linearly with the specific power dissipation. The ADGT results follow a similar trend and are in the range of values expected based on extrapolation from the surface aeration correlation. The results indicate that the ADGT is an effective small-volume gas-transfer device.

The mechanical power required for mixing is often estimated from the mean velocity gradient, or $G$ value, which is estimated as follows:

$$G = \left( \frac{P}{\mu h} \right)^{1/2}$$

where $G$ = mean velocity gradient (s$^{-1}$); and $\mu$ = viscosity of water (kg m$^{-1}$ s$^{-1}$).

The $G$ values estimated for the ADGT ranged from 350 to 2,800 s$^{-1}$, with contact times ranging from 5 to 80 s. Gemmell (1971) suggested that backmix reactors that are designed for 10-30 s contact time have $G$ values between 700 and 1,000 s$^{-1}$; while Amirtharajah and O’Melia (1990) indicated that in-line blenders with contact times of 0.5 to 1.0 s have $G$ values between 3,000 and 5,000 s$^{-1}$. The ADGT thus has mechanical mixing power input that is consistent with that of other mass-transfer devices.

Some observations might be made on the nature of the pressure loss through the ADGT based on the results summarized in Fig. 6(a). It is apparent here that the pressure loss was greater through the first cone than through the second, and greater through the second cone than through the third, even when the orifice sizes and liquid flow rates are the same through each cone. This suggests that the gas portion of the two-phase flow had a major influence on the pressure loss. The gas flow rate through the first cone would be greater than through the second, since a portion of the gas dissolves between cones, and the bubble size through the first cone was larger as well. The relative significance of gas flow rate and bubble size on the pressure loss generated could not be determined from the experiments conducted as they both decreased with passage through the ADGT. Pressure loss with one-phase water flow through a rounded submerged orifice (with coefficient of 1.0) and with diameter of 0.15 cm$^2$ and one-half the flow rate of 5.7 L/min was calculated by the standard orifice equation to equal 5 kPa. This is about the difference in pressure loss found between the three-cone and two-cone system at liquid flow rate of 5.7 L/min. Thus, pressure loss due to two-phase flow appears dominant with the first two cones, but pressure loss due to single-phase liquid flow appears to have been dominant through the third cone.

As noted earlier and in Fig. 6(b), pressure loss and oxygen transfer rate were found linearly related within the constraints applied to the system studied. These constraints are a gas flow rate selected to achieve the maximum gas-transfer rate possible while obtaining 100% dissolution of the added gas. For this system, a third cone produced less pressure drop, but it also produced little gas transfer, and thus a third cone offered no significant benefit for this system.

**CONSIDERATIONS FOR FIELD APPLICATION**

With the ADGT, $O_2$ injected into the system can be completely dissolved into recirculating ground water up until the point where the addition rate exceeds the mass-transfer rate. Under these constraints, maximum steady-state gas transfer can be achieved. However, with these conditions, only about 50% of theoretical saturation was achieved. No doubt higher percentages of saturation could be obtained under other operational conditions, but such conditions were not explored here. Complete gas dissolution is an advantageous characteristic of the ADGT. Accomplishing gas transfer down well in this manner avoids volatilization of contaminants and the use of surface gas-transfer devices. Another possible advantage is that the ADGT can operate at the prevailing pressure in a well casing where it may be placed, pressures that could be considerably above atmospheric, and thus making possible much higher oxygen transfer.

A disadvantage is that other gases dissolved in ground water hinder the rate and extent of oxygen addition. Thus, one of the most important factors governing gas transfer with the ADGT is the partial pressure of other dissolved gases in the ground water in relation to the total pressure on the transfer device. Possible gaseous components in ground water that might be considered are $N_2$, $O_2$, $CH_4$, and $CO_2$, some of which may be produced from biological activity in the ground water. The head-space method for gas analysis used here might be useful in such determinations. For the device to be useful, it is necessary that the target ground water contains a relatively low total partial pressure of dissolved gases relative to ground water pressure. This problem might be reduced by installing the apparatus at depth where ground water pressures are higher. An additional consideration here with in-situ organic biodegradation might be that this would cause the concentration of dissolved $CO_2$ to increase, but this should have only a small impact on the efficiency of gas transfer with the ADGT, since $CO_2$ is highly soluble in water and thus would exert only a small partial pressure.

As the ADGT has several advantages, as discussed, it may be useful for purposes other than in-situ ground-water bio-remediation. If enough installing depth is provided, it might be used for aeration of systems such as the hypolimnion of lakes or in U-tube aeration systems. Also, when the total partial pressures of dissolved gases are less than 1 atm, this device might be placed directly in a vertical water pipeline for aeration.

**ACKNOWLEDGMENTS**

This research was supported by the Gas Research Institute through a subcontract with Radian Corporation, the EPA’s Biosystems Program.
APPENDIX I. REFERENCES


APPENDIX II. NOTATIONS

The following symbols are used in this paper:

- $C_{eff} = \text{DO in effluent (mg/L)}$
- $C_{inf} = \text{DO in influent (mg/L)}$
- $C_T = \text{DO in transfer section (mg/L)}$
- $C_{eq} = \text{DO in equilibrium with gas phase partial pressure (mg/L)}$
- $C_r = \text{saturation concentration for dissolved oxygen in transfer device (atm)}$
- $G = \text{mean velocity gradient (s}^{-1})$
- $H = \text{Henry’s constant (mg L}^{-1} \text{ atm}^{-1})$
- $K_{oa} = \text{overall O, mass transfer coefficient (s}^{-1})$
- $P_T = \text{gas transfer power requirement (W)}$
- $P_{Na} = \text{partial pressure of N}_2 \text{ (atm)}$
- $Q = \text{flow rate (m}^3\text{s}^{-1})$
- $V = \text{volume of transfer section (m}^3)$
- $\Delta P = \text{pressure loss through transfer device (Pa)}$
- $\mu = \text{viscosity of water (kg m}^{-1} \text{ s}^{-1})$