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# Comparison of ammonia volatilisation rates in algae and duckweed-based waste stabilisation ponds treating domestic wastewater

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## Abstract

Quantification of ammonia volatilisation from wastewater stabilisation ponds is important in order to understand its significance for overall nitrogen removal in these widely applied low-cost treatment systems. Ammonia volatilisation rates were measured in pilot plant facilities consisting of one line of four algae-based ponds in series and a parallel line of four ponds with a floating mat of duckweed (*Lemna gibba*). Ammonia volatilisation was assessed during a period of one and a half years. The method applied is accurate, convenient and is proposed for analysis of a wide range of gasses emitted from stabilisation ponds and possibly other aquatic systems. The ammonia volatilisation rates in algae-based ponds (ABPs) were higher than in duckweed-based ponds (DBPs). This can be explained by the lower values of  $\text{NH}_3$  in DBPs due to shading and lower pH values, since the volatilisation rate highly correlated with free ammonia concentration ( $\text{NH}_3$ ) in pond water. The duckweed cover appeared not to provide a physical barrier for volatilisation of unionised ammonia, because whenever  $\text{NH}_3$  concentrations were equal in ABP and DBP also the volatilisation rates were equal. Volatilisation was in the range of  $7.2\text{--}37.4\text{ mg-N m}^{-2}\text{ d}^{-1}$  and  $6.4\text{--}31.5\text{ mg-N m}^{-2}\text{ d}^{-1}$  in the ABPs and DBPs, respectively. Average influent and effluent ammonium nitrogen measurements showed that the ammonia volatilisation during the study period in any system did not exceed 1.5% of total ammonium nitrogen removal. Therefore this study confirmed results from simultaneous experimental work in our laboratory indicating that nitrification/denitrification, rather than ammonia volatilisation, is the most important mechanism for N removal in ABPs and DBPs.

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**Keywords:** Ammonia; Volatilisation; Duckweed; *Lemna gibba*; Stabilisation ponds; Wastewater

## 1. Introduction

Waste stabilisation ponds are low-cost and efficient systems for wastewater treatment, producing high-quality effluent that enables water re-use in irrigation [1]. Recently there is a growing interest in modification of conventional algae-based ponds (ABPs) into duck-

weed ponds (DBPs). These are basically regular ponds with a floating cover of duckweed plants (*Lemnaceae*). The growing interest stems from the potential cost recovery through use of duckweed as animal feed. For both systems there is a lack of knowledge on the significance of the various nitrogen transformations and fluxes in the system. Quantification of nitrogen transformations is important for proper pond design and production of effluent suitable for disposal or crop irrigation.

Several research works on ammonia volatilisation are reported for farmlands and for dairy and animal slurry

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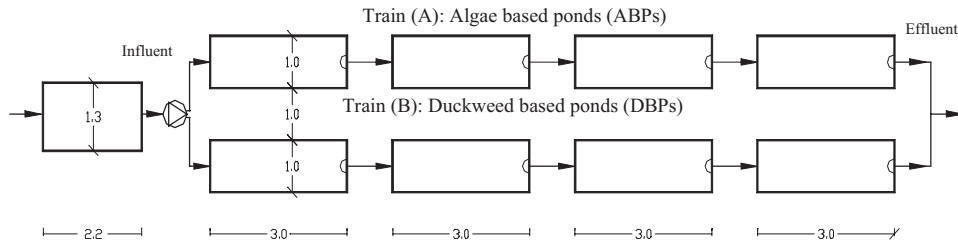


Fig. 1. Schematic presentation of the treatment pond systems consisting of 4 algae and 4 duckweed-based stabilisation ponds (HRT = 7 d each), preceded by a holding pond (HRT = 1 d).

concentrated wastewater [2–5]. This interest was triggered because of the effects of ammonia on acid rain, crops and farm workers. Unfortunately very few measurements of ammonia release have been made from wastewater stabilisation ponds.

Contradictory conclusions were drawn on the importance of ammonia volatilisation from two studies [6,7] in which data were obtained from the same stabilisation pond systems. Pano and Middlebrooks [6], in accordance with King [8], Silva et al. [9] and Soares et al. [10], argued that ammonia volatilisation largely explains total nitrogen removal from ponds. However, Ferrara and Avei [7] claimed that sedimentation of organic nitrogen is the predominant mechanism for N-removal. Unfortunately all these studies are based on theoretical assumptions and not on a complete nitrogen mass balance. Other studies on duckweed covered stabilisation ponds report incomplete mass balance equations and concluded that ammonia volatilisation is not very important in overall nitrogen removal [11–13]. These authors suggested that main N-removal mechanisms include nitrification/denitrification, duckweed uptake and harvesting. Zimmo et al. [14] found a nitrogen loss of 32% and 13% in algae and duckweed batch experiments, respectively, that was attributed to ammonia volatilisation and denitrification. The importance of ammonia volatilisation in algae and duckweed based systems was however not clarified. This paper describes the application of a simple method to assess relative contribution of ammonia volatilisation in overall N-removal from domestic waste stabilisation ponds.

## 2. Materials and methods

### 2.1. Layout and operation of the pilot plant

This study was carried out in two pilot-scale parallel systems of duckweed and algae-based domestic wastewater stabilisation ponds at the campus of Birzeit University (BZU), 26 km north of Jerusalem (750 m a.s.l.). Each treatment system consists of a series of four equal ponds with a total hydraulic retention time of 28

days. The layout of the two treatment pond systems (ABPs and DBPs) is shown in Fig. 1. A detailed description of the systems was presented elsewhere [15]. The flow through each system was  $0.38 \text{ m}^3 \text{ d}^{-1}$ . Duckweed biomass was harvested every fifth day to restore the density to its initial value of  $700 \text{ g fresh weight m}^{-2}$ . Algae were not harvested. The maximum duckweed density observed at the best growing conditions was  $2300 \text{ g fresh weight m}^{-2}$ . The system has been in operation since December 1998. The pilot plant was operated under two different conditions: From December 1998 till the middle of July 2000 wastewater from Birzeit University was used (average influent  $\text{NH}_4^+ \text{-N} = 60 \text{ mg l}^{-1}$ ). From the middle of July 2000–February 2001 wastewater from Al-Bireh city was used (average influent  $\text{NH}_4^+ \text{-N} = 110 \text{ mg l}^{-1}$ ).

### 2.2. Ammonia volatilisation, pH and water temperature measurements

From October 1999 measurements of ammonia volatilisation were carried out and each measurement covered a 24 h period. Ammonia volatilisation was measured in all four ponds of each treatment system (one pond each day; no simultaneous measurements from the two systems were made) by placing a Plexiglas transparent box ( $0.53 \times 0.24 \times 0.35 \text{ m}$ ) on the pond surface area. The box was kept on the surface by an iron frame connected to the pond walls. One side of the box was open and this side was placed on the pond surface. A constant airflow of  $1.5 \text{ L min}^{-1}$  that represents a wind velocity of  $1.1 \text{ m h}^{-1}$  was maintained through the box using an aquarium air pump. At the gas outlet from the Plexiglas box, ammonia gas was trapped in two flasks in sequence, each containing 90 ml 2% boric acid solutions. The tube in the trap flasks was maintained at approximately 1 cm below the acid surface (Fig. 2). Boric acid from the traps was collected and analysed for ammonium concentration. The accumulated ammonia was expressed in  $\text{mg day}^{-1}$  and divided by the area covered by the Plexiglas box to arrive at emissions expressed in  $\text{mg m}^{-2} \text{ day}^{-1}$ . No attempt was made to assess spatial variation in emissions from the ponds, the box was always

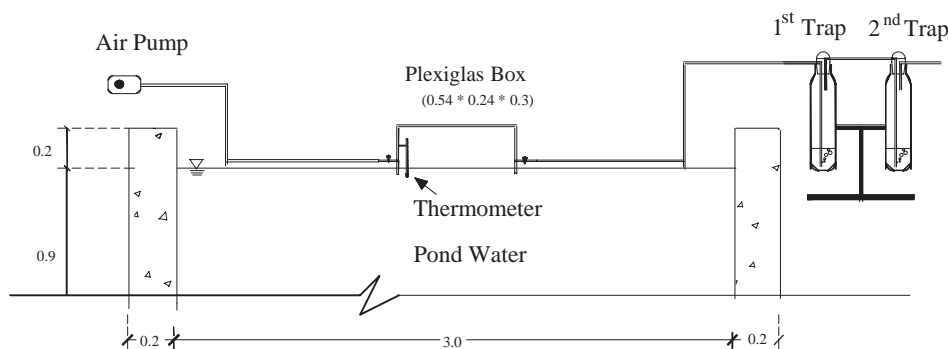


Fig. 2. Schematic diagram of experimental set-up to establish ammonia volatilisation rates.

positioned in the middle of a pond. Ammonia volatilisation measurements were performed over a period of one and a half year in order to assess seasonal variations. Ammonia volatilisation rates from the total pond surface area were extrapolated from the measured values. A blank control was used to account for ammonia interference from the surrounding air. At the end of each volatilisation measurement, a grab sample from the water column below the box was collected and analysed for ammonium concentration. Pond water temperature and pH underneath the Plexiglas box were measured three times throughout the daytime. Pond water pH and temperature were also measured over a 24 h cycle at 2 h intervals once every month during the study period.

### 2.3. Analytical methods

Dissolved oxygen (DO) was measured with a DO175m (HACH), pH with an EC10 pH meter (HACH) and temperature with a mercury thermometer. Ammonium concentration was measured by the Nessler colorimetric method according to the Standard Methods [16].

### 2.4. Theoretical considerations

The proportions of ammonium ion ( $\text{NH}_4^+$ ) and free ammonia ( $\text{NH}_3$ ) in solution are pH and temperature dependent [17]. The unionised ammonia fractions ( $\alpha$ ) in the pond water were calculated using Eq. (1) by Clement and Merlin [18]:

$$\alpha = \% \text{ Unionised } \text{NH}_3 = \frac{100}{1 + 10^{(\text{p}K_a - \text{pH})}} \quad (1)$$

$\text{NH}_3$  is relatively volatile and can be removed from solution to the atmosphere via diffusion through water to the surface and through mass transfer from the water surface to the atmosphere. The ammonia volatilisation rate was found to depend on pH, water temperature

[19–21] and mixing conditions [6]. Ammonia volatilisation can be described mathematically by the use of the two-film theory of mass transfer in which the water phase is assumed to be well mixed except near the interface. The mass transfer equation Eq. (2) with the assumption that the concentration of the ammonia gas in the atmosphere is zero was used to estimate the average ammonia volatilisation rates from each pond of the two systems:

$$N_{\text{NH}_3} = -K_1[\text{NH}_3], \quad (2)$$

where,  $N_{\text{NH}_3}$  is the mass transfer rate of ammonia ( $\text{mg l}^{-1} \text{d}^{-1}$ ),  $K_1$  is the convection mass transfer coefficient in the liquid phase ( $\text{d}^{-1}$ ),  $\text{NH}_3$  is the concentration of ammonia in the liquid phase ( $\text{mg l}^{-1}$ ). This first-order equation for ammonia mass transfer rate has been supported by Stratton [21] who obtained the following expression for the mass transfer coefficient:

$$K_1 = \frac{0.0566}{d} \exp[0.13(T - 20)], \quad (3)$$

where,  $d$  is the depth of water column in the pond (m) and  $T$  is the water temperature ( $^{\circ}\text{C}$ ).

## 3. Results

### 3.1. Ammonia concentrations and environmental conditions

Average influent and effluent  $\text{NH}_4^+$ -N and corresponding  $\text{NH}_4^+$ -N removal rates from the ABPs and DBPs during the experimental period are shown in Table 1. Average concentrations decreased throughout the ponds of the two systems and the decrease varied with the season and the wastewater nitrogen content (Table 1). Comparing the pooled volatilisation rates for all duckweed ponds ( $n = 114$ ) with the pooled data for all algae ponds ( $n = 114$ ) showed significant (oneway ANOVA,  $P < 0.05$ ) higher  $\text{NH}_4^+$ -N removal rates in

Table 1

Average influent and effluent  $\text{NH}_4^+$ -N concentrations (standard deviations) and  $\text{NH}_4^+$ -N removal rates from the ABPs and DBPs during the experimental period

Sample source		Summer 5/6/99–22/9/99 <i>n</i> = 17	Autumn 11/10/99–18/11/99 <i>n</i> = 4	Winter 7/12/99–22/3/00 <i>n</i> = 16	Spring 29/3/99–14/7/00 <i>n</i> = 12	Period during 1/8/00–1/2/01 <sup>a</sup> <i>n</i> = 17
$\text{NH}_4^+$ -N concentration in effl. Of DBPs ( $\text{mg l}^{-1}$ )	Influent	60.2 (5.6)	60.4 (4.4)	59.8 (6.4)	60.0 (3.5)	110.0 (9.5)
	D1 effl.	50.5 (5.2)	50.4 (2.2)	52.8 (4.1)	49.3 (2.8)	91.1 (6.4)
	D2 effl.	41.0 (4.1)	41.0 (0.7)	46.1 (4.2)	39.2 (2.1)	73.3 (10.1)
	D3 effl.	32.0 (4.1)	32.1 (1.5)	39.3 (4.1)	29.3 (1.7)	54.4 (9.1)
	D4 effl.	23.7 (4.3)	23.2 (1.6)	32.8 (3.2)	19.8 (1.7)	36.0 (7.1)
$\text{NH}_4^+$ -N removal rates ( $\text{mg-N m}^{-2} \text{d}^{-1}$ )	Influent-D4 effl.	1174	1197	869	1293	2380
$\text{NH}_4^+$ -N concentration in effl. Of ABPs ( $\text{mg l}^{-1}$ )	A1 effl.	31.6 (4.0)	34.8 (7.3)	45.2 (5.4)	32.9 (4.5)	85.0 (8.6)
	A2 effl.	20.1 (3.2)	23.4 (2.2)	33.4 (5.4)	23.6 (3.7)	65.7 (9.1)
	A3 effl.	12.5 (1.9)	15.4 (1.9)	23.3 (3.5)	16.9 (3.4)	40.3 (17.5)
	A4 effl.	8.7 (1.9)	9.1 (1.2)	14.6 (2.2)	10.1 (2.8)	23.5 (12.8)
$\text{NH}_4^+$ -N removal rates ( $\text{mg-N m}^{-2} \text{d}^{-1}$ )	Influent-A4 effl.	1657	1650	1454	1605	2782

Note: *n* is the number of samples analysed for each pond during the stated period.

<sup>a</sup> During this period, ABPs and DBPs were fed with raw sewage characterised by high nitrogen content.

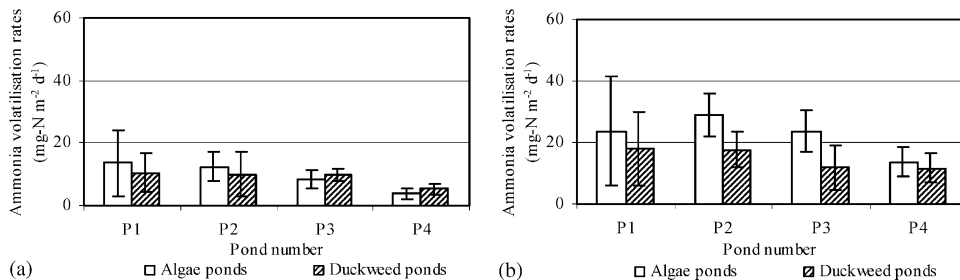


Fig. 3. Comparison of average ammonia volatilisation rates in ABPs and DBPs from June 1999 to July 2000 (a) and from August 2000 to February 2001 (b). Number of measurements for each individual pond was 27. The error bars indicate the standard deviations.

ABPs. Significantly higher removal rates were obtained when the pilot plant was fed with wastewater of higher ammonium content. In ABPs the pH values were generally above 8.0 whereas in the duckweed ponds, the duckweed mat prevented substantial algae photosynthesis and as a result lower pH values were observed [15]. During the daytime (only during the warm seasons) the shading provided by the duckweed cover in DBPs caused the temperature in DBPs to be 1°C lower than in ABPs.

### 3.2. Volatilisation rates and effect of environmental factors

Fig. 3 shows average ammonia volatilisation rates based on 24h measurements in the ABPs and DBPs,

with influent ammonium being 60 and 110  $\text{mg l}^{-1} \text{N}^{-1}$ , respectively. When influent ammonium was higher, higher ammonia volatilisation rates were measured in comparison with the previous period of operation. Partial disappearance of the duckweed cover from the first three ponds of DBPs was observed when the nitrogen concentrations in the system were increased, probably due to ammonia toxicity [18,22,23]. Despite the disappearance of duckweed cover, ammonia volatilisation in ABPs was higher than DBPs (Fig. 3b). This was possibly because in DBPs algae were not yet developed. In individual ponds similar volatilisation rates were obtained at similar ammonium concentrations and similar environmental conditions. Linear correlation factors  $R^2$  in the range 0.87–0.90 were obtained when unionised  $\text{NH}_3$  concentrations calculated

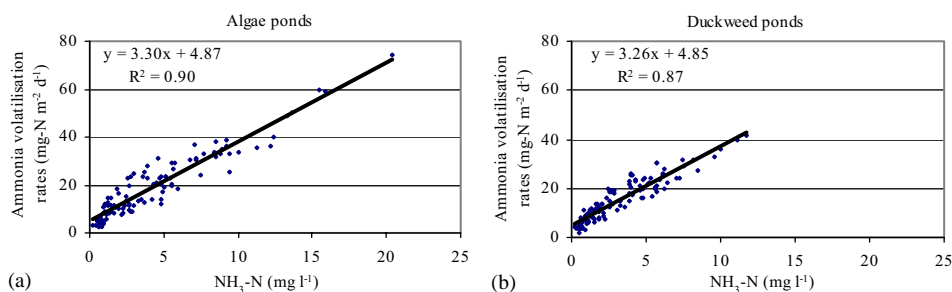


Fig. 4. Variation of ammonia volatilisation rate with pond water ammonia concentration in ABPs and DBPs from June 1999 to February 2001.

as a function of water temperature, pH and  $\text{NH}_4^+$  concentration Eq. (1) were plotted versus the measured ammonia volatilisation rates (Fig. 4).

#### 4. Discussion

This study describes the application of a simple method for direct assessment of ammonia volatilisation rates from stabilisation pond systems treating domestic wastewater. This method can also be used to assess the quantitative contribution of treatment systems to other gas emissions with potential negative environmental impact [24]. For verification of the method, ammonium in the second trap was measured several times during the experimental period. In addition, air from the vicinity of the pond was pumped through the traps. Ammonia concentrations in the traps in both cases were negligible. Placing a chamber on the surface may have influenced the gas emission, especially by affecting the temperature. However, it appeared that the air flow through the chamber was sufficient to minimise temperature increases. Therefore it can be assumed that the measured rates are equal to the actual rates of volatilisation occurring under natural quiescent conditions.

The strong diurnal pH fluctuations as occurring in ABPs due to algal photosynthetic activity was not observed in DBPs [15]. Therefore the average ammonia concentrations in ABPs were higher than in DBPs, since at higher pH the equilibrium between ammonia and ammonium is displaced towards the free form. The higher ammonia concentrations in ABPs were expected to enhance ammonia volatilisation and indeed ammonia volatilisation rates measured in algae-based ponds were significantly higher than those measured simultaneously in duckweed based ponds. The variation of volatilisation rates over a 24 h period closely followed the variation in pH (data not shown), which, under constant concentrations of total ammonium nitrogen, was expected.

Ammonia concentrations not only differed between ABPs and DBPs, but also decreased along the series of

ponds. Accordingly the rates of volatilisation decreased along the series of ponds. Similarly ammonia concentrations were lower in winter due to lower pH and temperature, which resulted in lower volatilisation rates during that season. Good correlation coefficients were obtained to describe the relationships between ammonia volatilisation rates and ammonia concentrations (calculated as a function of ammonium concentration, pH and water temperature Eq. (1)).

Whenever ammonia concentrations were the same for any of the algae or duckweed ponds, also ammonia volatilisation rates were similar. Apparently, the duckweed layer did not provide a physical barrier for volatilisation of unionised ammonia, although the duckweed cover reduces the water surface that is directly exposed to the atmosphere. An explanation could be that gas emission from the water phase takes place through the stoma of the plant. This phenomenon has been reported for other gases and plants. For instance, methane emission from rice fields is mainly through the plants [25].

The simulated wind velocity in the Plexiglas box ( $1.1 \text{ m h}^{-1}$ ) corresponds to a situation when the air above the pond is nearly stagnant. It is expected that the ammonia volatilisation rates will increase at higher wind velocity as a result of a steeper ammonia gradient caused by the removal of ammonia accumulated in the gas film at the interface. If the measured ammonia volatilisation rates would be corrected according to data obtained by Wachs et al. [26], for the average wind velocity at the pilot plant site the volatilisation rates would be two times higher than rates indicated in Fig. 3. Our measurements of ammonia volatilisation rates are much lower than values reported for slurry and piggery wastes with higher  $\text{NH}_4^+$  and  $\text{NH}_3$  concentrations [2–5]. Our findings are in the same range as reported by Schroeder [21], Gross et al. [27] and Stratton [28]. They are also in agreement with others that found that ammonia volatilisation in waste stabilisation ponds accounts for only a small fraction of total N-removal from domestic sewage [7,12–14].

#### 4.1. Mechanisms and modelling of ammonia volatilisation

Pano and Middlebrooks [6] developed an ammonia nitrogen removal model for facultative ponds based on the theoretical ammonia stripping model. The authors assumed that nitrification does not account for a significant part of the nitrogen removal because of the low nitrite and nitrate concentrations observed. This assumption is not correct, because low nitrite and nitrate may be due to effective denitrification in the sediment layer [29]. Nitrification/denitrification may represent an important nitrogen flux [30]. Still, Pano and Middlebrooks could model  $\text{NH}_4^+$  removal accurate with only  $T$  and pH as predicting parameters. This may be due to the strong correlation between pH and algae growth. Algae growth is believed to be essential for nitrogen removal, either via uptake, decay and sedimentation or by providing attachment area and oxygen for nitrifiers. A model that includes algae decay and sedimentation, but not nitrification was proposed by Ferrara and Avci [7]. Their calculation for ammonia volatilisation was based on the assumption that ammonia mass transfer is first order with respect to ammonia nitrogen concentration in the fluid. By using the expression for mass transfer coefficient by Stratton [21] they concluded that ammonia volatilisation accounts for only a small fraction of total N-removal. Table 2 compares the average rates of ammonia volatilisation from the algae and duckweed-based ponds obtained by direct measurement in the present study with the average rates predicted using the models by Stratton and Pano and Middlebrooks. The predicted rates using these models were calculated for the average pond water pH and temperature measured over a 24h duration at 2h interval once every month

during the study period. Predicted values by the Stratton model were one to two times higher than the measured values, but values become similar when corrected for wind effects. Ammonium removal rates predicted by the Pano and Middlebrooks model were much higher than the measured values in our experiments. A possible explanation for that was discussed above. Our results suggest that stabilisation ponds are probably more environmentally friendly treatment systems than one might expect from results of Pano and Middlebrooks who suggested extreme high values for ammonia release and subsequent negative impact on the environment. Based on our measurements for ammonia volatilisation from ABPs and DBPs, the following equation is proposed to calculate the amount of ammonia volatilisation from stabilisation ponds:

$$Y = 3.30[\text{NH}_3] + 4.90,$$

where,  $Y$  is the ammonia volatilisation in  $\text{mg-N m}^{-2} \text{d}^{-1}$ ,  $[\text{NH}_3]$  is the calculated  $\text{NH}_3$  ( $\text{mg-N l}^{-1}$ ) as a function of pH, water temperature and ammonium concentration in pond water Eq. (1).

The nitrogen losses via ammonia volatilisation, after wind correction, in ABPs and DBPs were less than 2–3% of total  $\text{NH}_4^+$ -N removal. Therefore, ammonia volatilisation was not the major mechanism for ammonia loss in the pilot plant used in this study. This suggests that ammonia volatilisation is insignificant for nitrogen removal in natural treatment systems with wastewater characteristics and environmental conditions similar to the present experimental set-up. If the average pond water pH would rise to 9, the ammonia volatilisation is expected to account for 21% of the nitrogen mass

Table 2

Comparison of average ammonia volatilisation (AV) rates predicted using models by Stratton [21] and Pano and Middlebrooks [6] with rates obtained by direct measurement (this study)

Pond identification	$\text{NH}_4^+$ -N	$\text{NH}_4^+$ -N	$T$	pH	Fraction	$\text{NH}_3$ -N	Stratton	Pano and	Measured in
	$\text{mg l}^{-1}$	$\text{mg l}^{-1}$							
	In/out	Pond water	$^{\circ}\text{C}$	(–)	%	$\text{mg l}^{-1}$	AV rates in $\text{mg m}^{-2} \text{d}^{-1}$		
Inf.	70.1								
<i>ABPs</i>									
A1	46.0	59.1	16.7	8.1	3.8	2.2	81.1	3607	21.7
A2	32.4	43.7	17.0	8.1	3.6	1.6	60.5	2399	22.3
A3	19.4	27.3	17.3	8.0	3.0	0.8	37.7	1666	19.1
A4	11.4	18.3	17.1	8.0	2.8	0.5	19.8	1021	10.5
<i>DBPs</i>									
D1	58.4	67.4	16.5	8	3.0	2.0	63.0	3199	18.1
D2	47.7	52.6	17.3	7.8	2.0	1.1	41.8	2203	16.4
D3	37.0	43.4	16.9	7.8	1.7	0.8	28.3	1765	14.2
D4	26.8	31.7	16.5	7.7	1.5	0.5	17.7	1180	10.6

Note: Average values over the study period of influent and pond water ammonia concentrations,  $T$  and pH (daily average) are given.

balance. Although it is common to encounter pH values of 9–10 in the late afternoon in secondary facultative or maturation ponds, it is uncommon to have such values over a 24 h cycle.

## 5. Conclusions

This study clarifies the contradicting conclusions that others reached about the role of ammonia volatilisation in overall nitrogen removal from algae and duckweed based ponds. Ammonia volatilisation was highly correlated with  $\text{NH}_3$  concentration in pond water, which in turn was governed by the combined effect of pH and water temperature. Shading provided by the duckweed cover prevented strong diurnal changes in pond water pH such as commonly observed in conventional stabilisation ponds. The lower pH in DBPs in comparison with ABPs resulted in lower ammonia concentrations and hence in lower ammonia volatilisation. However, similar ammonia volatilisation from ABPs and DBPs at equal ammonia concentrations was observed. Apparently the physical barrier provided by the duckweed layer did not hinder ammonia volatilisation. Volatilisation rates decreased along the series of ponds as the unionised ammonia concentration decreased. Ranges of 37.4–7.2 and 31.5–6.4  $\text{mg-N m}^{-2} \text{d}^{-1}$  were observed in ABPs and DBPs, respectively. Nitrogen loss via ammonia volatilisation in the algae and duckweed ponds at calm conditions did not exceed 1.5% of total ammonium nitrogen removal in the treatment systems. This suggests that other nitrogen removal mechanisms such as nitrification/denitrification or sedimentation may be more important in overall nitrogen removal. The method for the measurement of ammonia volatilisation as developed and applied in this research could be adjusted for the quantification of wide range of different gas fluxes (such as  $\text{CH}_4$ ,  $\text{H}_2\text{S}$ ,  $\text{NO}_x$  and  $\text{CO}_2$ ) from stabilisation pond systems. The method therefore maybe useful to assess and compare environmental impacts from wastewater treatment systems due to (green house and acid rain) gas emissions.

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