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Ammonia volatilisation in waste stabilization ponds: A cascade of misinterpretations?

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Abstract

Ammonia volatilisation has generally been reported as, or assumed to be, the main nitrogen removal mechanism in waste stabilization ponds (WSP). Nitrogen removal via ammonia volatilisation is based on two observations: (a) in-pond pH values can reach high values (>9, even >10), so increasing the proportion of the total ammonia present as the un-ionized form or free ammonia (NH₃); and (b) in-pond temperatures can also be high, so improving the mass transfer rate of free ammonia to the atmosphere. Consequently, one of the most widely accepted models for ammonia removal in WSP is that reported by Pano and Middlebrooks in 1982, which was developed to reflect the occurrence of these two observations. This work reports how simple mathematical models for ammonia volatilisation in WSP, in spite of the possibility of their giving good predictions, may not accurately describe the main pathways and mechanisms involved in ammonia removal in WSP.

Keywords

Ammonia volatilisation; biological nitrogen uptake; maturation ponds; nitrogen removal

INTRODUCTION

Ammonia volatilisation has been reported as the main nitrogen removal mechanism in waste stabilization ponds (WSP) during periods when temperature and pH (>10) are favourable (Maynard *et al.*, 1999). Ammonia removal in WSP has been estimated by using the model developed by Pano and Middlebrooks (1982), which is one of the most widely accepted to describe ammonia volatilisation processes in facultative and maturation ponds. This model is based on first-order kinetics in a completely mixed reactor and is dependent on variables such as pH, temperature and hydraulic loading rate. Similar conclusions with regard to the importance of ammonia volatilisation on nitrogen removal in WSP have been also reported by using mass-transfer models which specifically consider the aqueous ammonia-water equilibrium system (Rockne and Brezonik, 2006). Slight variations from the original Pano and Middlebrooks model have been reported in order to introduce the effect of local weather and operational characteristics, and their authors agreed that ammonia volatilisation could be the predominant pathway for nitrogen and ammonia removal in WSP (Silva *et al.*, 1995; Soares *et al.*, 1996; Bastos *et al.*, 2007). Indeed it is also assumed to be the predominant pathway in current models developed for plug-flow and complete-mix conditions and used for predicting total nitrogen removal in WSP (Crites *et al.*, 2006). However, none of these models has been calibrated or validated by means of direct measurements of ex-pond ammonia volatilisation rates. Recent work reported by Zimmo *et al.* (2003), Zhou *et al.* (2006) and Camargo Valero and Mara (2007a) have pointed out that ammonia volatilisation makes only a small contribution to the overall performance of nitrogen removal in WSP, despite the fact that the Pano and Middlebrooks model and its variations still make reasonable predictions of ammonia removal within facultative and maturation ponds. In this paper we review the evidence for and against the usefulness of the Pano and Middlebrooks model for ammonium removal in facultative and maturation ponds.

AMMONIA VOLATILISATION FROM ALKALINE WATERS

Krefx *et al.* (1958) published a short note on the possibility of removing ammonia from wastewater effluents by raising the pH. They were working on wastewater works effluent and found that ammonium disappeared from solutions of the order of 20–50 mg N/L within hours when these solutions were buffered to a pH <8.4, whereas at pH values of 4.5–6.5, there was no appreciable disappearance after a week (Harrison *et al.*, 1960). Indeed, the chemical equilibrium between ammonium (NH_4^+ , ionic form) and free ammonia (NH_3 , un-ionized gaseous form) makes ammonia volatilisation a feasible mechanism for ammonium removal in alkaline waters. Ammonia in an aqueous solution acts as a weak base in a dynamic equilibrium between ammonium and ammonia; this equilibrium (equation 1) depends mainly on pH but also on the dissociation constant (K_b) which is temperature-dependent (equations 2 and 3; Emerson *et al.*, 1975). The mass transfer process of ammonia from an aqueous solution into the atmosphere depends on the concentration of free ammonia in the liquid phase and its partial pressure in the atmosphere above the water surface (Henry's law), as well as on other factors such as the water surface turbulence, air velocity above the water surface, pH, temperature and mixing properties of the liquid (Stratton, 1969). Therefore at appropriate temperatures and pH values ammonia can be released from water by volatilisation to the atmosphere. This principle has been in use for the design of desorption towers in industrial wastewater treatment, which require a high air-to-water ratio as the ammonia mass transfer is a gas phase-controlled process (Patoczka and Wilson, 1984).

For many practical purposes the percentage of un-ionized ammonia can be expressed as a function of pH and K_b values at a specific water temperature (equation 4).



$$K_b = \frac{[\text{NH}_4^+] \times [\text{OH}^-]}{[\text{NH}_3]} \quad (2)$$

$$pK_b = 0.09018 + \frac{2729.92}{273.2 + T} \quad (3)$$

$$\% \text{NH}_3 = \frac{100}{1 + 10^{pK_w - pK_b - \text{pH}}} \quad (4)$$

where $[\text{NH}_4^+]$, $[\text{NH}_3]$, $[\text{H}^+]$ and $[\text{OH}^-]$ are molar concentrations; $pK_b = -\log_{10}K_b$; $pK_w = -\log_{10}K_w$; K_w is the ionic product of water – i.e., $[\text{H}^+] \times [\text{OH}^-]$; and T is the water temperature ($^{\circ}\text{C}$).

Stratton (1968) determined ammonia losses in an open channel ($0.15 \times 6.00 \times 0.03$ m) in which 48 litres of synthetic river water (distilled water enriched with ammonium sulphate (20 mg NH_4^+ -N/L and potassium phosphate buffer) was circulated along a closed loop system. A set of experiments to calculate ammonia losses was carried out under different pH values (7.5, 8.0, 8.5, 9.0 and 9.5) and water temperatures (15, 20, 25 and 30°C), and the results showed that on average 50 percent of the total ammonium was removed in 14 hours at pH = 8.5 and $T = 20^{\circ}\text{C}$. Subsequently, and assuming that ammonia losses in alkaline waters follow an exponential decay pattern (equation 5), Stratton reported the relationship between the rate constant for ammonia losses with water temperature (at pH 8.5) and pH (at 20°C) (equations 6 and 7, respectively). Equation 5 is a first-order equation which describes the decrease of ammonia nitrogen concentration as a function of time (t), depth of flow (d) and a rate constant (k'). The k values in equations 6 and 7 correspond to the net disappearance rate of ammonia from clean water which is an ideal aqueous system with no biological activity; thus k values are not mass transfer coefficients for either the aqueous ammonia-clean water system or the aqueous ammonia-pond water system.

$$C = C_o \times \exp\left(\frac{-k' t}{d}\right) \quad (5)$$

$$k = 0.048 \times \exp\left[0.13 \times (T - 20)\right] \quad (6)$$

$$k = 0.080 \times \exp\left[1.57 \times (\text{pH} - 8.5)\right] \quad (7)$$

In equation 5 C_o is the ammonia-nitrogen concentration at time zero (mg/L) and C is the ammonia-nitrogen concentration at any time t (mg/L).

Based on his initial results, Stratton concluded that, considering that algae can increase the pH in surface water bodies up to ~10, large ammonia losses to the atmosphere could be expected in shallow streams in which algae and water plants were growing. In order to confirm this hypothesis, Stratton (1969) carried out outdoor experiments subjected to natural conditions favourable for ammonia volatilisation (i.e., summer conditions). Firstly, a set of four 6-litre battery jars containing synthetic lake water [i.e., deionised water enriched with ammonium nitrogen (20 mg NH_4^+ -N/L) and 1M phosphate buffer to pH 7.5, 8.0, 8.5, 9.0 and 9.5, separately] were placed outdoors. Ammonia nitrogen determinations were made every 48 hours for about 21 days. The results were in agreement with previous work with synthetic river water in an open channel (Stratton, 1968) and high ammonia volatilisation rates were reported (e.g., ~2,153 g N/ha d at pH=8.5, T= 19.6°C and an initial total ammonium concentration of 10 mg NH_4^+ -N/L). It is important to note that Stratton reported no evidence of biological activity during both experiments with synthetic alkaline waters.

Secondly, Stratton (1969) made direct measurements of gaseous ammonia nitrogen liberated through the surface of two small reservoirs (~2 ha each) with clear evidence of biological activity. Ex-pond ammonia gases were collected by using a floating cell and acid trap similar in principle, but different in design, to the one developed by Camargo Valero and Mara (2007a). Site 1 was Elfin Forest Lake (Escondido, CA) and site 2 was a pond receiving the effluent from a wastewater treatment plant (Santee, CA). Tests were conducted for a period of one hour and sampling points were selected based on the presence of high algal activity to guarantee high pH values. The measured rates of ammonia losses in site 1 and 2 were considerably below the predicted volatilisation rates from the outdoor experiments with synthetic lake water (approximately 14% and 23% of the predicted value, respectively). For instance, the average ammonia volatilisation rate for site 1 was 513 g N/ha d (9.5 < pH < 9.8; 28 < T < 29°C); however, this figure did not consider diurnal pH variations and hence the average daily ammonia volatilisation rate would have been much lower. Even so, it was estimated that ammonia volatilisation would contribute to remove only ~5.8 percent of the total daily influent ammonia entering Elfin Forest Lake.

Based on the results from experiments with synthetic water, Stratton's (1968, 1969) work clearly demonstrated that ammonia could be removed from alkaline waters without any biological activity; thus ammonia volatilisation could be considered the main mechanism for ammonium removal when waters are buffered to pH values higher than 8.5. However, and most importantly, he also showed that very low ammonia volatilisation rates were obtained in alkaline waters which had a high algal activity such as WSP waters.

THE PANO AND MIDDLEBROOKS MODEL

One of the most commonly accepted models for ammonium nitrogen removal in WSP was proposed by Pano and Middlebrooks in 1982. Data utilized to develop and validate that model were taken from three WSP systems located at Peterborough, New Hampshire (US EPA, 1977a); Eudora, Kansas (US EPA, 1977b); and Corinne, Utah (US EPA, 1977c). All three systems were exposed to similar climatic conditions and water temperature varies from 1 to 5°C in winter (ice cover is also

experienced in winter), whilst average water temperature in summer is 20°C but generally less than 25°C. These studies included a full year of data collection from pond influent and effluent of each system, including four separated 30-consecutive-day and 24-hour composite sampling periods, one each season (US EPA, 1983). The first model reported by Pano and Middlebrooks (1982) was based on Stratton's work with synthetic river and lake waters (Stratton 1968; 1969); therefore, ammonia volatilisation was initially adopted as the main nitrogen removal mechanism in WSP. Arguments made in support of this assumption considered that low temperatures, good mixing conditions and low biological activity would provide a perfect niche for the supremacy of ammonia stripping over other feasible ammonium removal processes (e.g., ammonia assimilation in algal biomass). Paradoxically, high biological – mainly algal – activity is the only available route to guarantee high pH values in WSP and consequently the presence of free ammonia.

The mass balance reported by Pano and Middlebrooks (1982) for ammonia stripping in a completely mixed reactor under steady-state and continuous flow conditions produced the mathematical model reported in equation 8 below. However, they considered that their model should contemplate other removal mechanisms such as ammonia removal through biological activity and ammonia releasing into pond water column from anaerobic activity at the bottom of the pond along with ammonia volatilisation. Therefore, rather than using the equation developed for ammonia stripping (equation 8), they decided to consider a more general expression for ammonia nitrogen removal which would follow first-order kinetics in a reactor with a flow pattern very close to completely mixed conditions (equation 9). The resulting models were calibrated and validated against monthly mean influent and effluent ammonium concentrations from the three WSP systems under study for temperatures up to 20°C and for those between 21 to 25°C (equations 10 and 11, respectively).

$$\frac{C_e}{C_i} = \frac{1}{1 + \frac{A}{Q} k_1 \left[\frac{1}{1 + 10^{pK_w - pK_b - pH}} \right]} \quad (8)$$

$$\frac{C_e}{C_i} = \frac{1}{1 + \frac{A}{Q} K \cdot f \cdot \phi H} \quad (9)$$

$$\frac{C_e}{C_i} = \frac{1}{\left\{ 1 + \frac{A}{Q} \left[0.0038 + 0.000134T \exp \{ 0.041 + 0.044T \} \phi H - 6.6 \right] \right\}} \quad T \leq 20^\circ\text{C} \quad (10)$$

$$\frac{C_e}{C_i} = \frac{1}{\left\{ 1 + \frac{A}{Q} \left[0.005035 \exp \{ 1.540 \phi H - 6.6 \} \right] \right\}} \quad 21 \leq T \leq 25^\circ\text{C} \quad (11)$$

where C_e and C_i are the total ammonium concentration ($\text{NH}_4^+ + \text{NH}_3$) in the pond effluent and influent, respectively (mg N/L); A is the surface area of the pond (m^2); Q is the inlet flow rate (m^3/d); k_1 is the ammonia mass transfer coefficient (m/d); and K is the rate coefficient for ammonia removal (m/d).

The resulting equations reported by Pano and Middlebrooks (1982) for ammonia removal in WSP systems (equations 10 and 11) are actually simple first-order equations for a completely mixed reactor, which reflexes a statistically significant relationship with pH, water temperature and hydraulic loading rate. Pano and Middlebrooks' model does not prove that the most important mechanism in ammonia nitrogen removal in WSP is ammonia volatilisation. In fact, it does not provide any further information on the nature of the nitrogen removal mechanisms as most

biochemical reactions that involve nitrogen (e.g., biological nitrogen uptake, nitrification-denitrification) will respond as strongly to these three factors as ammonia volatilisation does.

Moreover, if ammonia volatilisation is the main mechanism for permanent ammonium removal in WSP, the Pano and Middlebrooks model does not explain why the annual average ammonium removal efficiencies in the WSP systems used to develop their model (52.8% in Peterborough, NH; 96.1% in Eudora, KS; and 99% in Corin, UT; Pano and Middlebrooks, 1982; Ferrara and Avci, 1982) are higher than the corresponding total nitrogen removal figures (43, 82 and 91%, respectively; Reed, 1985; Ferrara and Avci, 1982). This could infer that, although ammonium nitrogen is removed, it may be transformed into other nitrogen species (e.g., organic nitrogen), which leaves in the pond effluent; thus total nitrogen removal would be lower than ammonium removal. Therefore, any agreement of field results with values predicted by Pano and Middlebrooks' equations and their variations (Silva *et al.*, 1995; Soares *et al.*, 1996; Bastos *et al.*, 2007) does not confirm the validity of the mechanism(s) for ammonia removal in WSP assumed in their development.

AMMONIA REMOVAL IN WSP VIA VOLATILISATION PROCESS

Results from a set of experiments carried out by Camargo Valero and Mara (2007a) in a pilot-scale maturation pond in the UK confirm the poor contribution of ammonia volatilisation on nitrogen removal in WSP. They reported very low ex-pond ammonia volatilisation rates (from <1 to 27 g NH₃-N/ha) despite the highly efficient ammonium removal achieved (ammonium removal rate was 3,747g N/ha d; ≈90% removal efficiency) and the occurrence of favourable conditions for ammonia volatilisation process (i.e., summer conditions; 8.9 < pH < 10.2 and 15.2 < T < 18.2°C).

Ex-pond ammonia volatilisation rates reported by Camargo Valero and Mara (2007a) were much lower than initial results from lab-base experiments with alkaline water (2,517 g NH₃-N/ha; pH =10.1, T =17.1°C) and calculated values from theoretical models using mass transfer coefficients under field conditions (107–812 g N/ha d). Moreover, their results were even lower than (a) the annual value of 828 g N/ha d reported by Rockne and Brezonik (2006) who used an equilibrium-based model with wind speed adjustment for the WSP system at Harris, Minnesota (i.e., in a cold region with a long ice-cover season); (b) the values of 105–223 g N/ha d measured in situ by Zimmo *et al.* (2003) from an algal WSP located in Ginebra, southwest Colombia; and (c) the values of 198–811 g N/ha d calculated by Zimmo *et al.* (2003) using the Stratton's equation reported by Ferrara and Avci (1982).

Consequently, models based on mass transfer coefficients for the ideal water–ammonia–air system (e.g., Stratton's equation in Ferrara and Avci, 1982; Zimmo *et al.*, 2003; Rockne and Brezonik 2006) overestimate ammonia losses to the atmosphere. That is simply because they do not include the influence of strong interferences on free ammonia concentration (e.g., phytoplanktonic activity) and ignore the fact that the water in a WSP is a very complex matrix where ammonia may be involved in more than one chemical or biochemical transformation pathway simultaneously. Therefore, it is only to be expected that theoretical models for ammonia removal in WSP based exclusively on the ammonia volatilisation process would not be able to predict the real contribution of that mechanism on nitrogen removal in WSP systems. Moreover, high pH and water temperature values do not necessarily favour ammonia volatilisation over alternative mechanisms such as algal uptake.

Camargo Valero and Mara (2007b) and Camargo Valero (2008) reported results from tracer experiments carried out in a pilot-scale maturation pond under summer and winter conditions (Figures 1 and 2, respectively). Figures 1 and 2 show the normalised concentrations (C/C_0) in the pond effluent, against normalised time (t/θ), of (a) ¹⁵N-ammonium tracer used to track ammonium transformations, (b) Rhodamine WT used to visualise the behaviour of a conservative tracer, and (c)

^{15}N -suspended organic nitrogen used to estimate the influence of biological uptake on ammonium removal [normalised concentration is the ratio between the actual effluent concentration (C) divided by the concentration expected if the mass of tracer were instantaneously completely mixed in the whole pond volume (C_0); and normalised time is equal to time (t) divided by hydraulic retention time (θ)]. The data presented in Figure 1 show how under summer conditions (i.e., during periods of high phytoplanktonic activity) ^{15}N -ammonium is rapidly incorporated in the pond biomass and then washed out of the system in the pond effluent as biomass; this explains why ammonium removals in full-scale WSP have reported higher values than corresponding total nitrogen removals figures. On the other hand, low water temperatures and short photoperiods (i.e., winter conditions) clearly affect algal activity; for these reasons ^{15}N -ammonium is mainly washed out in the pond effluent with only minimal incorporation into pond biomass (Figure 2).

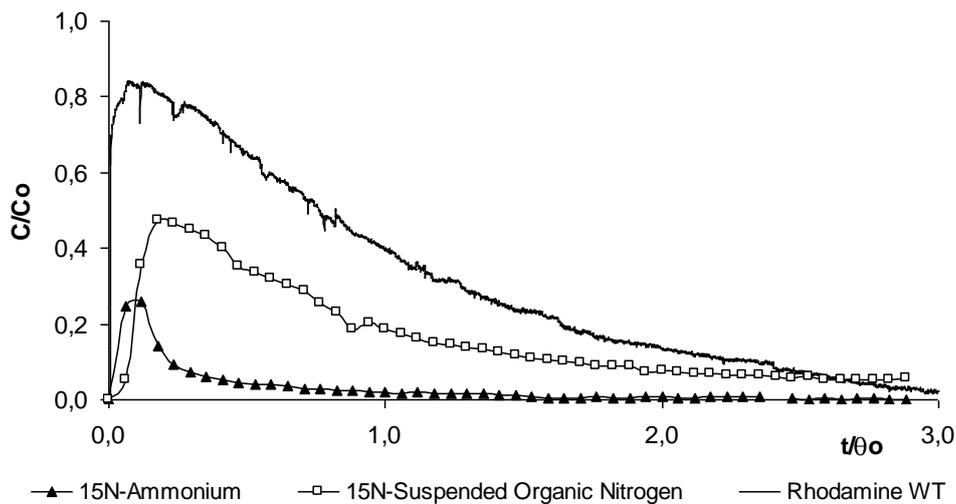


Figure 1. Tracer experiment results from a maturation pond in summer

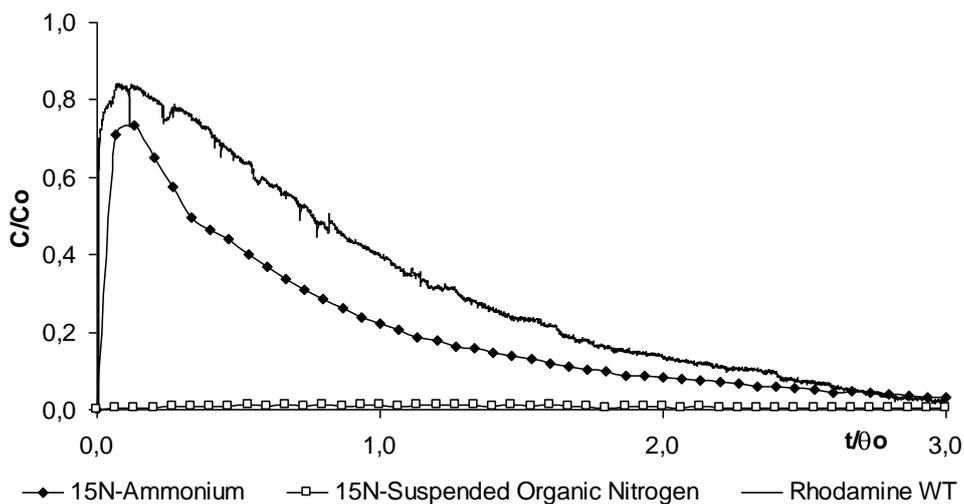


Figure 2. Tracer experiment results from a maturation pond in winter

These tracer experiments not only provide additional evidence to support the finding that ammonia volatilisation makes only a small contribution on the overall nitrogen removal in WSP, but also that biological nitrogen uptake strongly dominates ammonia removal under conditions favourable for algal growth. This is in agreement with the hypothesis formulated by Pearson *et al.* (1988) that increasing pond water temperature increases phytoplanktonic activity and consequently in-pond algal biomass takes up and removes ammonium at a faster rate than expected via ammonia stripping. The increment of pH in WSP is a consequence of algal activity and it makes a small

contribution to ammonia volatilisation as the ammonia concentration drops due to algal uptake. Moreover, recent findings by Camargo Valero *et al.* (2009a, 2009b) may explain high total nitrogen removals found in WSP in operation as a combination of sedimentation of dead biomass after biological nitrogen uptake and denitrification.

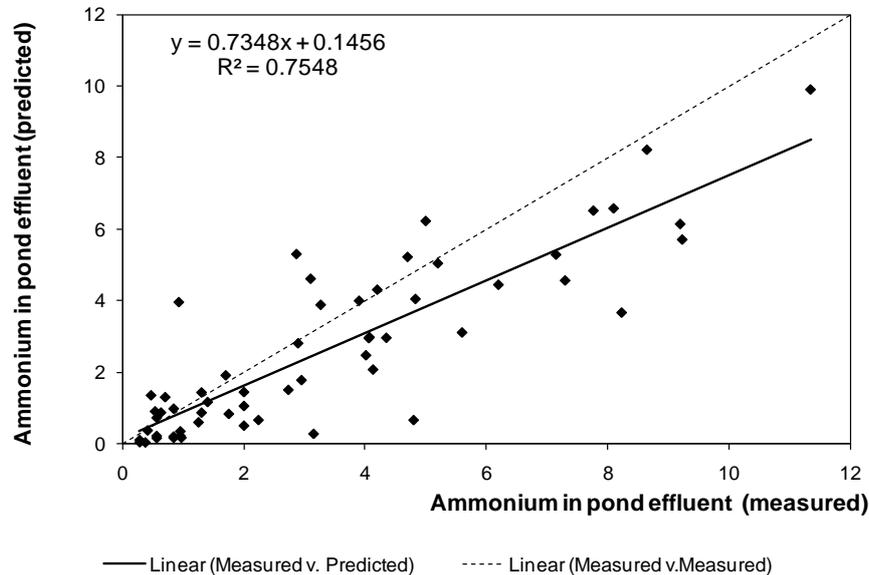


Figure 3. Prediction of ammonium concentration in the effluent of maturation ponds with the Pano and Middlebrooks model

Using the Pano and Middlebrooks model (equation 10), data collected in a three-year monitoring programme from two pilot-scale maturation ponds in series were used to predict the ammonium concentrations in each pond effluent and the ammonia removal efficiencies (Camargo Valero, 2008). Results reported in Figure 3 show that the linear regression coefficient ($R^2 = 0.7548$), the slope of the graph (0.7348) and the Coefficient of Determination (0.31) illustrate a poor linear correlation between actual ammonium values from pond effluents and those predicted by the Pano and Middlebrooks model. Therefore the accuracy to predict ammonium concentration in the pilot-scale maturation ponds was poor and the estimated error varied from -327 to 90 percent; nevertheless, average performance was reasonably well predicted (e.g., the average ammonium removal efficiency in summer was 90% and the predicted figure was 96%). However, better agreements have been reported by other researchers and in some cases upgraded versions of the Pano and Middlebrooks model have been developed (Silva *et al.*, 1995; Soares *et al.*, 1996; Bastos *et al.*, 2007). The Pano and Middlebrooks model, along with its improved versions, may predict ammonia removal in WSP quite well as algal nitrogen uptake can be simply modelled as a first-order reaction in a completely mixed reactor.

CONCLUSIONS

Based on the literature reviewed and the experiments undertaken, it is postulated that ammonia removal by volatilization makes little or no contribution to nitrogen removal by WSP either in summer or winter. High pH and water temperature values should not necessarily favour ammonia volatilisation over alternative mechanisms like algal uptake. An increasing pond water temperature increases phytoplanktonic activity and consequently, in-pond algal biomass would take up and remove ammonium to a faster rate than expected via ammonia stripping. The increment of pH in WSP is a consequence of algal activity and it makes a small contribution to ammonia volatilisation process as ammonia concentration drops due to algal uptake. Pano and Middlebrooks' model and its

variations could fairly predict ammonia removal in WSP, but they do not confirm ammonia volatilisation as the main mechanism for permanent nitrogen removal.

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