Towards Calibration of Phosphorus (P) Removal Plant-Wide Models

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INTRODUCTION

Over the past years, wastewater treatment plant (WWTP) mathematical models have been advancing towards their widespread application for sizing and operation of treatment plants to minimize energy consumption and cost while maximizing nutrient recovery and effluent quality. Effective utilisation of these models requires that they are well calibrated. However, difficulties (with important parameters not identified and uncertainties in intepretation of model output results) can be experienced in model calibration, especially due to (i) the intricate relationships of model output variables with model input factors (where parameters are inter-related to various model outputs), resulting in non-linearity, and (ii) the limitations (due to expensive and/or time consuming experimental methods) experienced in procuring and reconciling data required for determination of the model input factors (especially when the model has significantly large numbers of unknown parameters and model components).

The BIOMATH protocol, developed by Vanrolleghem *et al.* (2003), provides a systematic approach for calibration. The main objective of this paper is to apply the BIOMATH protocol in providing a guidance towards calibration of a plant-wide model that includes phosphorus. The three phase (aqueous-gas-solid) University of Cape Town plant wide (UCT–PW) model (Ikumi *et al.*, 2013) that was calibrated against the experimental layout described below is used as a case study for this calibration procedure.

EXPERIMENTAL SYSTEM LAYOUT

The experimental layout of Ikumi (2011) is used in this study. It replicates at laboratory scale three WWTP schemes, comprising (1) a Modified Ludzack – Ettinger (MLE) nitrification–denitrification (ND) activated sludge (AS) system treating raw sewage (MLE 1) with anaerobic digestion (AD) of its waste activated sludge (WAS) in AD system number 1 (i.e., AD1), (2) an identical MLE system (MLE 2) treating settled sewage with AD of its WAS in AD2 and (3) a membrane (MBR) University of Cape Town (UCT) ND enhanced biological P removal (NDEBPR) system treating settled sewage with (i) AD of its WAS in AD3 and (ii) anoxic/aerobic digestion (AAD) of its WAS in two intermittently aerated (3hour air on, 3hour air off) aerobic digesters, AAD1 fed with concentrated WAS (2x, 20gTSS/l) and AAD2 fed with dilute WAS (1/3, 3.3gTSS/l).

MODEL DESCRIPTION

The UCT three phase plant wide model was developed for simulating the biological processes to track and predict the output of materials (COD, carbon (C), hydrogen (H), oxygen (O), nitrogen (N), phosphorus (P), magnesium (Mg), potassium (K) and calcium (Ca)) along the unit processes of a WWTP. It comprises three sub-models, integrated for simulation of the entire WWTP under various configurations (e.g. NDBEPR AS system linked to an AD or an anoxic-aerobic digestion (AAD) for WAS stabilisation). These sub-models include:

1. The ionic speciation model (Brouckaert *et al.*, 2010). This model includes pairing of ionic components (the set of model ionic species is given in Table 3) and inter-phase transfers of component species. Table 4 gives an example of a set of equilibrium and mass balance equations used in the ionic speciation subroutine.
2. The ASM2-3P model: This is the Activated Sludge Model No. 2 (ASM2, Henze *et al.*, 1995), modified to include the ionic speciation model (Brouckaert *et al.*, 2010), the Inorganic Settleable Solids (ISS) model of Ekama and Wentzel (2004) and including multiple mineral precipitation according to Musvoto *et al.* (2000a,b).
3. The ADM3P Model: This is the University of Cape Town Anaerobic Digestion Model (UCTADM; Sötemann *et al.*, 2005), modified to include the hydrolysis of multiple organic sludge types (PS, ND WAS, NDBEPR WAS and PS-WAS blends), the Ekama and Wentzel (2004) ISS model, multiple mineral precipitation processes according to Musvoto *et al.* (2000a, b) and the Brouckaert *et al.* (2010) aqueous speciation model which facilitates ionic speciation (Ikumi *et al.*, 2011).

For their compatibility, the ASM2-3P and ADM3P models have the same comprehensive set of model components (supermodel approach, Volcke *et al.*, 2006; model components given in Table 1 and applied stoichiometric processes in Table 4), including parameterized stoichiometry for the bioprocesses and sharing the same ionic speciation subroutine model (1).

MODEL EVALUATION PROCESS

1. *Model Verification*: To initiate the evaluation of the UCT-PW model (Ikumi *et al.*, 2013), the systematic method proposed by Hauduc *et al.* (2010) was applied to verify that material (COD, C, H, O N, P, Mg K and Ca) balances were achieved in the determination of all stoichiometric processes.

2. *Parameter Values*: The initial values for suitable kinetic and stoichiometric parameters as obtained experimentally or from literature were entered, and given the typical value range, determined according to the methods proposed by Brun *et al.* (2002).

3. *Senitivity Analysis*: The parameters were subsequently evaluated using two different methods of global sensitivity analysis: (1) Standardised Regression Coefficients (SRC) and (2) Morris Screening. The results obtained using these methods are used to identify important parameters (prioritisation of those with greatest effects), non-influential parameters (those that can be ‘fixed’ at any value within their range without effecting outputs) and interacting parameters (Neumann, 2012).

4. *Model Calibration and Validation*: Non-influential parameters were set at their default values and random samples were drawn from the remaining subset of parameters. For the

sampled parameter sets simulations were conducted and predicted model outputs were compared with observed outputs. During this calibration phase, a consistent set of parameter values was used to simulate all experimental systems/periods, and detailed explanations of observed discrepancies (if any) were reported by Ikumi (2011). The sensitivity analysis together with intuitive observation on a steady state stoichiometric model was a significant role in selecting the 'best' set of parameters. It was noted that influent sewage and sludge characterisation, and determination of hydrolysis kinetic rates were important requirements prior to any simulation. In AD, the hydrolysis process is the slowest one and requires the best possible calibration. The hydrolysis kinetic constants were fit to match experimental data by Ikumi *et al*. (2013), using the non-linear regression. The biological reactions following hydrolysis are limited by the hydrolysis rate, hence their kinetic parameters are not identifiable from these experimental data, and so were adopted without adjustment from literature. Most of the parameterised influent and sludge characteristics could be obtained or calculated from directly measured results (Ikumi, 2011). This is because the unbiodegradable fractions of influent organics and the characteristics of the biodegradable feed components have a significant effect on the quality of model predictions, but are usually specific to the feed source.

5. *Model Performance Results*: Below are observations in experimental behaviour of P removal systems, replicated by the UCT-PW model as required to promote confidence in its application:
    i. Applying the ASM2-3P model to an MLE system with ND does not stimulate EBPR (i.e., there is no PAO growth, hence no polyphosphate (PP) storage) and its effluent P comprises mainly the OP not utilized by the biomass (mainly OHOs) for growth.
    ii. For MLE systems with little or no nitrification taking place, high quantities of P and acetate in the un-aerated ('anoxic') zone will result in the growth of phosphorus accumulating organisms (PAOs) rather than ordinary heterotrophic organisms (OHOs) only as expected in fully aerobic or nitrogen (N) removal systems. The concentration of acetate available for this PAO growth (and associated EBPR) depends on the rate of fermentation of biodegradable soluble organics (BSO) that occurs and the concentration of nitrate that gets recycled to the anaerobic reactor in these systems. Moreover, as noted in 3 and 5-stage Bardenpho systems – in winter, when denitrification is lower, the nitrates recycled to the anaerobic reactor can be sufficiently high to suppress EBPR. The ASM2-3P model predicts this behaviour qualitatively well.
    iii. Applying ASM2-3P for sludge treatment with anoxic-aerobic digestion (AAD), the absence of VFA and an anaerobic period renders the PAOs unable to compete with the OHOs. Consequently, the PAOs do not grow and undergo endogenous respiration and die, releasing their stored PP as magnesium (Mg), calcium (Ca), potassium (K) and orthophosphate (OP). Struvite ($MgNH_4PO_4$) precipitation occurs when the concentration of Mg, ammonia and OP is high enough (i.e., the struvite is supersaturated) in the mixed liquor. If the ammonia is low (< 1mg/N/*l*), due to nitrification, K-struvite ($MgKPO_4$) forms.
    iv. In AD, organically bound N is released with the hydrolysis of biodegradable organics in the non-ionic NH_3 form, which are non-reference species (reference species of a weak acid system being one that, when added to pure water, creates a solution state, relative to which the alkalinity of the weak acid system is measured) for the ammonia weak acid/base system. Therefore, the aqueous alkalinity increases by the concentration of NH_3 transferred from the organics (the NH_3 being an intrinsic alkalinity content of the organics) to the aqueous phase. This is the main aqueous $H_2CO_3^*$ alkalinity generation

process in an AD treating PS or WAS that is not P-rich. For P-rich systems with PP, the aqueous H_2CO_3* alkalinity increase also depends on PP and cell bound P release because PP is released as H_2PO_4 and biomass P is released as H_3PO_4, which interact with the other weak acid/base systems and influence pH. The ADM3P model predicts the pH for both these systems (P- rich or not) really well.

v. In the dynamic model of the AD, initially, PP release and poly-hydroxy-alkanoate (PHA) storage by PAOs takes place with the uptake of acetate, as would happen in the anaerobic part of the parent NDEBPR system. This results in increased alkalinity because the PP is released as $H_2PO_4^-$. Because the PAOs also require alternating aerobic conditions for their growth, they cannot grow in the AD. Therefore, the PAOs are modelled to “die” in AD at a rate faster than their endogenous respiration; releasing their PHA and the remainder of their stored PP, adding more $H_2PO_4^-$ and alkalinity. Depending on the charge/proton balance requirements, some of the $H_2PO_4^-$ species become HPO_4^{2-} species by reacting with HCO_3^- to form HPO_4^{2-}, H_2O and CO_2. The increase in CO_2 gas increases the partial pressure of the gas phase, which influences the aqueous speciation. The split between the OP species co-dependent on the inorganic carbon (IC) system (and any other weak acid/base system that may be present), which together establish the AD pH.

vi. Because H_3PO_4 is the reference species for the OP weak acid/base system, the total alkalinity does not change with the slower release of organically bound P, but the species that represent it do.

vii. The rapid release of PP and associated Mg^{2+} and the slow release of biomass N and P generate high concentrations of P, NH_4^+ and Mg^{2+} species in the AD liquor, which promotes struvite precipitation. This struvite precipitation decreases the total alkalinity and so results in re-speciation of the IC system, which increases the CO_2 partial pressure and decreases AD pH.

COMPARISON OF EXPERIMENTAL AND PREDICTED RESULTS

Figures 1a to f show a comparison between the data measured and simulated by the three phase AD dynamic model for the AD1 fed with the NDBEPR WAS (i.e. the AD effluent from UCT NDBEPR linked to AD in a plant wide setting). Considering the general complexities in characterization of the AD influent (i.e. the NDBEPR WAS; Ikumi *et al*., 2013), the simulated results match quite well for COD removal (Fig 1a, which it should because the hydrolysis rates were calibrated on to the experimental results), and FSA (Fig1b) and OP (Fig 1c) release. Because all the AD products, including the H_2CO_3 and H_3PO_4 alkalinities and gas CO_2 partial pressure (p_{CO2}), are entirely dependent on the composition of the biodegradable organics (x,y,z,a,b,q,c,d,e in $C_xH_yO_zN_aP_b\ qMg_cK_dCa_ePO_3$), if the organics’ composition entered into the model is not “correct”, then the simulated and measured results will not match, even with 100% experimental material balances (which of course were not achieved on the UCT and AD systems). Improving the comparison between predicted and measured results is a complex exercise because multiple processes act on single compounds. For instance under-predicted FSA (Fig 1b) means the determined N content of the biodegradable part of the OHO and PAO biomass is too low, but this does not mean that the H_2CO_3 alkalinity also has to be under-predicted (through $NH_3 + H_2CO_3 \rightarrow NH_4^+ + HCO_3^-$, as it would for an AD fed with low P organics, Sötemann *et al.*, 2005a,b) because the release of PP also produces alkalinity (through $MePO_3 + H_2O \rightarrow Me^+ + H_2PO_4^{2-}$).

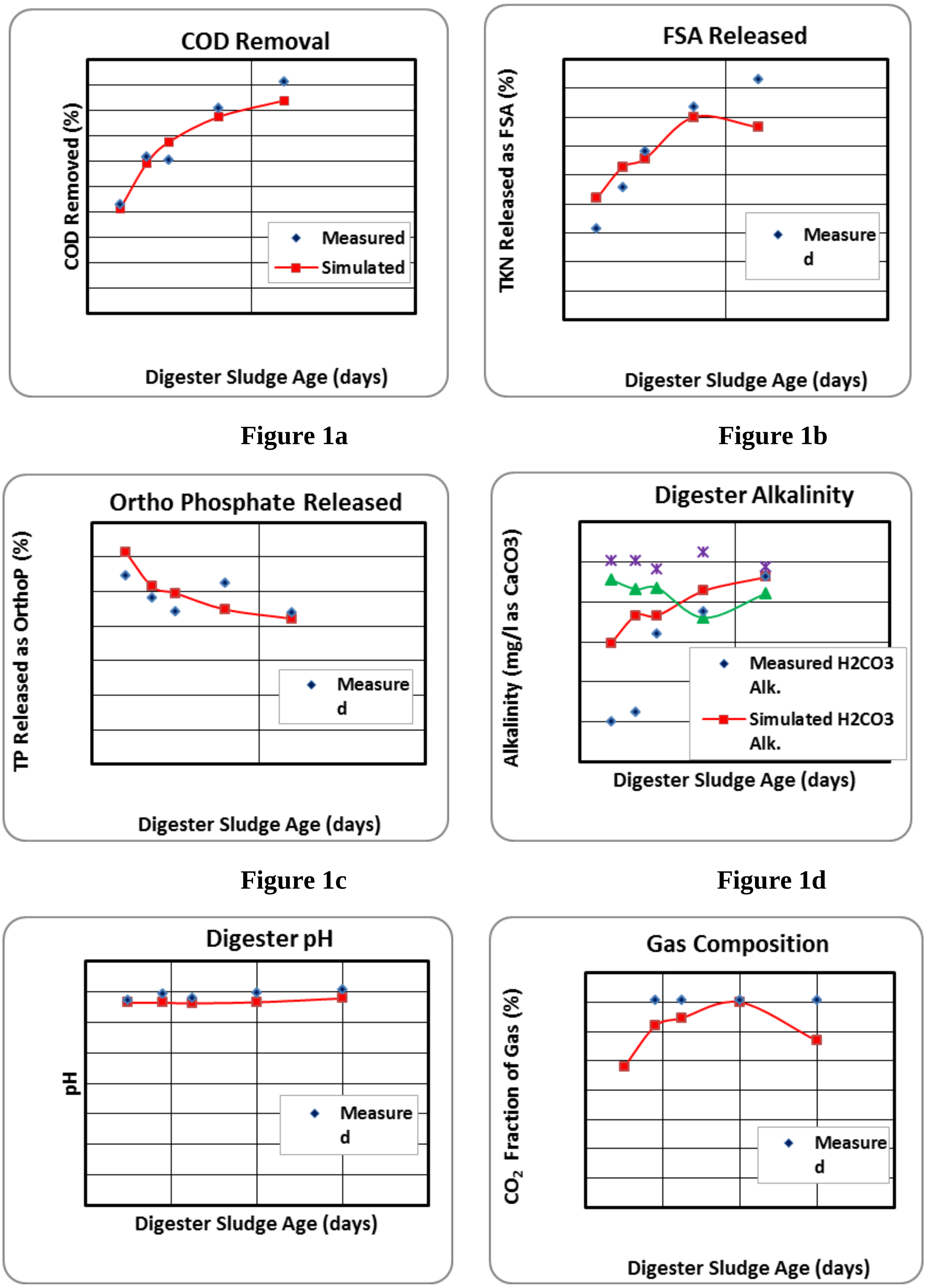

Figure 1a **Figure 1b**

Figure 1c **Figure 1d**

Figure 1e **Figure 1f**

***Figure 1**: Comparison between simulated and measured results for AD of WAS from the laboratory scale UCT NDBEPR system fed with settled WW and added acetate to increase BEPR.*

CLOSURE

The BIOMATH protocol was applied for the calibration of the UCT–PW model, for promotion of its widespread utilisation in a reproducible way. However, it is noted that the effective calibration of this model requires a further step - from modelling the laboratory scale systems (under controlled and completely mixed environments) to assessment of model predictions for full-scale wastewater treatment plant systems, interlinked to plant-wide configurations. This prospective work may be of particular interest to the IWA group on benchmarking of control strategies for WWTPs who are including P into an extended BSM model.

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Table 1: Universally selected model components for UCT three phase plant wide model (UCT-PW)

Name	Empirical formula	Description	Units
H2O	H2O	Water	m^3/d
S_H	H^+	Hydrogen ion	gH/m^3
S_Na	Na^+	Sodium	gNa/m^3
S_K	K^+	Potassium	gK/m^3
S_Ca	Ca^{2+}	Calcium	gCa/m^3
S_Mg	Mg^{2+}	Magnesium	gMg/m^3
S_NH$_x$	NH_4^+	Ammonium	gNH$_4$/m^3
S_Cl	Cl^-	Chloride	gCl/m^3
S_VFA	CH_3COO^-	Acetate	gAc/m^3
S_Pr	$CH_3CH_2COO^-$	Propionate	gPr/m^3
S_CO$_3$	CO_3^{2-}	Carbonate	gCO$_3$/m^3
S_SO$_4$	SO_4^{2-}	Sulphate	gSO$_4$/m^3
S_PO$_4$	PO_4^{3-}	Phosphate	gPO$_4$/m^3
S_NO$_x$	NO_3^-	Nitrate	gNO$_3$/m^3
S_H2	H_2	Dissolved hydrogen	gH$_2$/m^3
S_O2	O_2	Dissolved oxygen	gO$_2$/m^3
S_U	$CH_{Yu}O_{Zu}N_{Au}P_{Bu}$	Unbiodegradable Soluble Organics	g/m^3
S_F	$CH_{Yf}O_{Zf}N_{Af}P_{Bf}$	Fermentable Biodegradable Soluble Organics	g/m^3
S_Glu	$C_6H_{12}O_6$	Glucose	g/m^3
X_U_inf	$CH_{Yup}O_{Zup}N_{Aup}P_{Bup}$	Unbiodegradable particulate organics	g/m^3
X_B_Org	$CH_{Ybp}O_{zbp}N_{Abp}P_{Bbp}$	Biodegradable particulate organics	g/m^3
X_B_Inf	$CH_{Ybps}O_{Zbps}N_{Abps}P_{Bbps}$	Influent biodegradable particulate organics	g/m^3
X_PAO_PP	$K_{kp}Mg_{mp}Ca_{cp}PO_3$	Polyphosphate	g/m^3
X_PAO_Stor	$C_4H_6O_2$	Poly-hydroxy-alkanoate	g/m^3
X_Str_NH4	$MgNH_4PO_4.6H_2O$	Struvite	g/m^3
X_ACP	$Ca_3(PO_4)_2$	Calcium Phosphate	g/m^3
X_Str_K	$MgKPO_4.6H_2O$	K-struvite	g/m^3
X_Cal	$CaCO_3$	Calcite	g/m^3
X_Mag	$MgCO_3$	Magnesite	g/m^3
X_Newb	$MgHPO_4$	Newberyite	g/m^3
X_ISS		Influent inorganic settleable solids	gISS/m^3
X_OHO	$CH_{Yo}O_{Zo}N_{Ao}P_{Bo}$	Ordinary heterotrophic organisms	g/m^3
X_PAO	$CH_{Yo}O_{Zo}N_{Ao}P_{Bo}$	Phosphate accumulating organisms	g/m^3
X_ANO	$CH_{Yo}O_{Zo}N_{Ao}P_{Bo}$	Autotrophic nitrifying organisms	g/m^3
X_ZAD	$CH_{Yo}O_{Zo}N_{Ao}P_{Bo}$	Acidogens	g/m^3
X_ZAC	$CH_{Yo}O_{Zo}N_{Ao}P_{Bo}$	Acetogens	g/m^3
X_ZAM	$CH_{Yo}O_{Zo}N_{Ao}P_{Bo}$	Acetoclastic Methanogens	g/m^3
X_ZHM	$CH_{Yo}O_{Zo}N_{Ao}P_{Bo}$	Hydrogenotrophic methanogens	g/m^3
X_U_Org	$CH_{ye}O_{ze}N_{ae}P_{be}$	Endogenous residue	g/m^3
G_CO$_2$	CO_2	Carbon dioxide	gCO$_2$/m^3
G_CH$_4$	CH_4	Methane	gCH$_4$/m^3

Table 2: Ionic species selected for the UCT three phase model (UCT-PW)

	Formula	Description		Formula	Description
1	H^+	Hydrogen ion	23	$NH_4SO_4^-$	Ammonium sulphate
2	Na^+	Sodium	24	$MgPO_4^-$	Magnesium phosphate
3	K^+	Potassium	25	$CaCH_3COO^+$	Calcium acetate
4	Ca^{2+}	Calcium	26	$CaCH_3CH_2COO^+$	Calcium propionate
5	Mg^{2+}	Magnesium	27	$CaHCO_3^+$	Calcium bi-carbonate
6	NH_4^+	Ammonium	28	$NaSO_4^-$	Sodium sulphate
7	Cl^-	Chloride	29	$MgHPO_4$	Magnesium hydrogen phosphate
8	CH_3COO^-	Acetate	30	CH_3COONa	Sodium Acetate
9	$CH_3CH_2COO^-$	Propionate	31	H_2CO_3	Di-hydrogen carbonate
10	CO_3^{2-}	Carbonate	32	$MgSO_4$	Magnesium sulphate
11	SO_4^{2-}	Sulphate	33	HPO_4^{2-}	Hydrogen phosphate
12	PO_4^{3-}	Phosphate	34	NH_3	Ammonia
13	NO_3^-	Nitrate	35	$MgCO_3$	Magnesium carbonate
14	OH^-	Hydroxide ion	36	$ACPO_4^-$	Calcium Phosphate
15	CH_3COOH	Acetic acid	37	$MgHCO_3^+$	Magnesium hydrogen carbonate
16	CH_3CH_2COOH	Propionic acid	38	$CaHPO_4^-$	Calcium hydrogen phosphate
17	HCO_3^-	Bi-carbonate	39	$NaCO_3^-$	Sodium carbonate
18	$CaSO_4$	Calcium sulphate	40	$MgH_2PO_4^+$	Magnesium di-hydrogen phosphate
19	$H_2PO_4^-$	Di-hydrogen phosphate	41	$NaHCO_3$	Sodium hydrogen carbonate
20	$MgCH_3COO^+$	Magnesium acetate	42	$NaHPO_4^-$	Sodium hydrogen phosphate
21	$MgCH_3CH_2COO^+$	Magnesium propionate	43	$CaOH^+$	Calcium hydroxide
22	$CaCO_3$	Calcium carbonate	44	$MgOH^+$	Magnesium hydroxide

Table 3: Example for equilibrium and mass balance equations for ionic speciation

Weak Acid Sub-System	*Aqueous Phase Equilibrium Equations	Mass Balance Equation
Ammonia	$[NH_3] = \frac{K_{NH_4} \cdot [NH_4^+]}{(H^+)}$ $[NH_4SO_4^-] = \frac{[SO_4^{2-}][NH_4^+]}{K_{NH_4SO_4}}$	$NH_x = [NH_4^+] + [NH_3] + [NH_4SO_4^-]$

*Where (H^+) is the hydrogen ion activity, [X] the molar concentrations of species X and $K_{X'}$ is the thermodynamic equilibrium constant for species X, adjusted for Debye Hückel effects to account for the activity of ions in low salinity water (Stumm and Morgan, 1996).

Table 4: Processes used in the application of UCT three phase plant wide model

Name	Description
AerHydrol	Aerobic hydrolysis of biodegradable particulate organics (BPO)
AnHydrol	Anoxic hydrolysis of BPO
AnaerHydrol	Anaerobic hydrolysis of BPO
AerGrowthOnSf	Aerobic OHO growth on fermentable soluble organics (FBSO)
AerGrowthOnSa	Aerobic OHO growth on Acetate
AnGrowthOnSfDenitrif	Anoxic OHO growth on FBSO
AnGrowthOnSaDenitrif	Anoxic OHO growth on Acetate
Fermentation	Fermentation of FBSO
LysisOfAuto	Storage of poly-hydroxy-alkanoate (PHA) by PAOs
StorageOfXPP	Aerobic storage of PP with PHA uptake
AerGrowthOnXPHA	Aerobic growth of PAOs
LysisOfXPP	Release and hydrolysis of polyphosphate (PP)
LysisOfXPHA	Release and hydrolysis of PHA
GrowthOfAuto	Aerobic growth of ANOs with nitrification
OHO_Lysis	Lysis of OHOs in aerobic systems
LysisOfXPAO	Lysis of PAOs in aerobic systems
LysisOfAuto	Lysis of ANOs in AS system
Aeration	Oxygen supply to aerobic reactor
FSO_Hydrolysis	Hydrolysis of FBSO in AD system
BPO_Hydrolysis	Hydrolysis of BPO produced by dead biomass
BPO_PS_Hydrolysis	Hydrolysis of BPO from primary sludge (PS)
OHO_Lysis_AD	Lysis of OHOs in AD system
PAO_Lysis_AD	Lysis of PAOs in AD system
PP_Release	Release of PP with uptake of PHA in AD system
PP_Hydrolysis	Release and hydrolysis of PP in AD system
PHA_Hydrolysis	Release and hydrolysis of PHA in AD system
Acidogenesis_L	Low hydrogen partial pressure (p_{H2}) Acidogenesis
Acidogenesis_H	High p_{H2} Acidogenesis
AD_decay	Lysis of acidogens
Acetogenesis	Growth of acetogens in AD system
AC_decay	Lysis of acetogens
Acet_methanogenesis	Growth of acetoclastic methanogens in AD system
AM_decay	Lysis of acetoclastic methanogens
Hyd_methanogenesis	Growth of hydrogenotrophic methanogens in AD system
HM_decay	Lysis of hydrogenotrophic methanogens