

## WP2 - COMBINING AOPs WITH BIOLOGICAL LANDFILL LEACHATE TREATMENT

# Optimisation of landfill leachate treatment in a moving-bed biofilm system by means of reactor staging and controlled ozonation: the **BIOZO** concept

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# Publications

- Plósz, B.G., Langford, K.H., Heiaas, H.H., Macrae, K., Liltved, H., Lopez, A., Vogelsang, C. (2010) Occurrence of xenobiotic organic micro-pollutants in landfill leachate and PAHs removal from the liquid and sludge phases in a biofilm system combined with ozonation. WATER SCIENCE AND TECHNOLOGY, Accepted.
- Plósz, B.G., Vogelsang, C., Jantsch, T.G., Liltved, H., C., Lopez, A. (2010) Ozonation as a means to optimise biological nitrogen removal from landfill leachate. OZONE: SCI. ENG., Accepted.
- Plósz, B.G., Vogelsang, C., Bomo, A.M., Rossetti, S. (2009) Controlled stratification of microbial populations in biofilm systems by reactor stages: A novel optimisation method using reaction kinetics data. Submitted to BIOTECHNOLOGY AND BIOENGINEERING.







# Outline

- Characterisation of the partly stabilised landfill leachate
- Biological treatment and reactor design;
- Biological treatment coupled with controlled ozonation

# Composition of the stabilised leachate used in our experiments

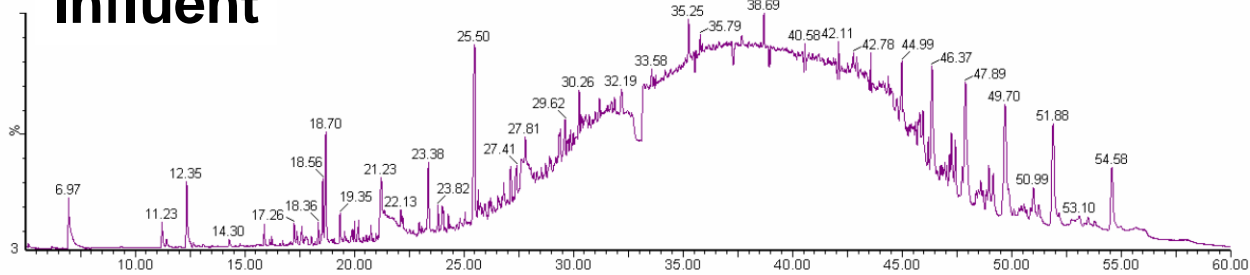
		Average
pH	-	7.48
NH <sub>4</sub> N	mg/L	197
NO <sub>3</sub> N	mg/L	4.69
NO <sub>2</sub> N	mg/L	0.75
COD <sub>Total</sub>	mg/L	3090
COD <sub>Solubl</sub> e	mg/L	1664

- *High concentrations of scavenger molecules!!*

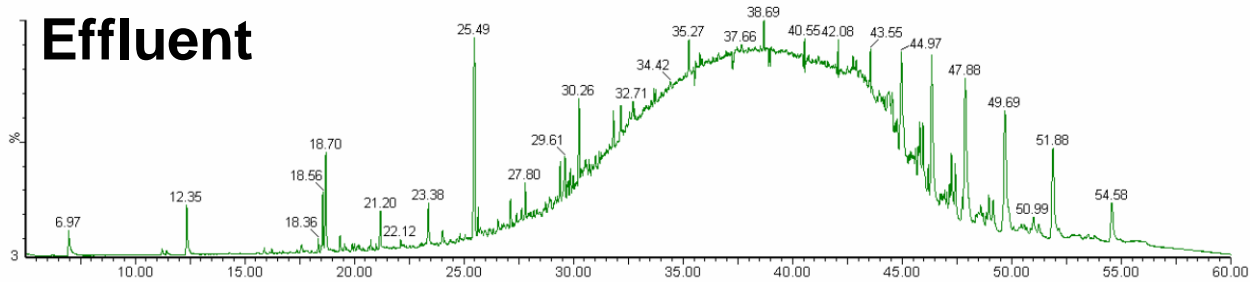


# Screening for priority pollutants in landfill leachate

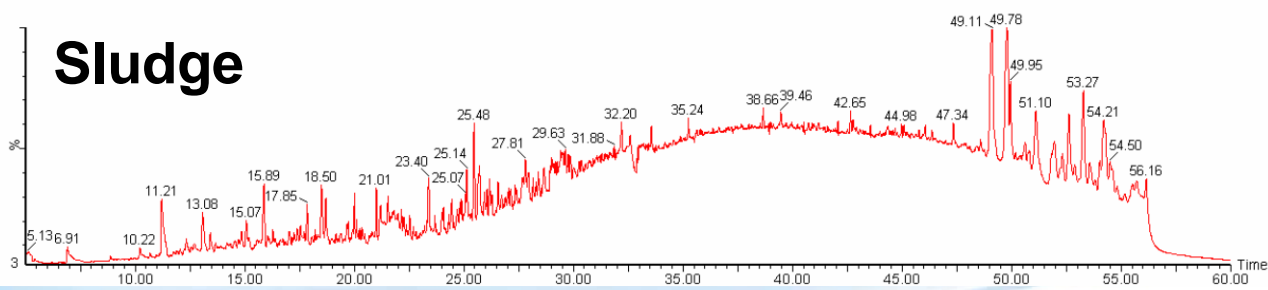
## Influent



## Effluent



## Sludge



- Samples analysed for organic micro-pollutants by GC-ToF-MS.
- Hybrid mass spectrometer ideal for applications in environmental chemistry. Coupling Gas chromatography (GC) with Time-of-Flight (TOF) and mass spectrometry (MS) technologies.
- All samples showed evidence of unresolved complex mixtures (UCM) as demonstrated by the humps in the chromatograms

## Prioritised XOCs in leachate

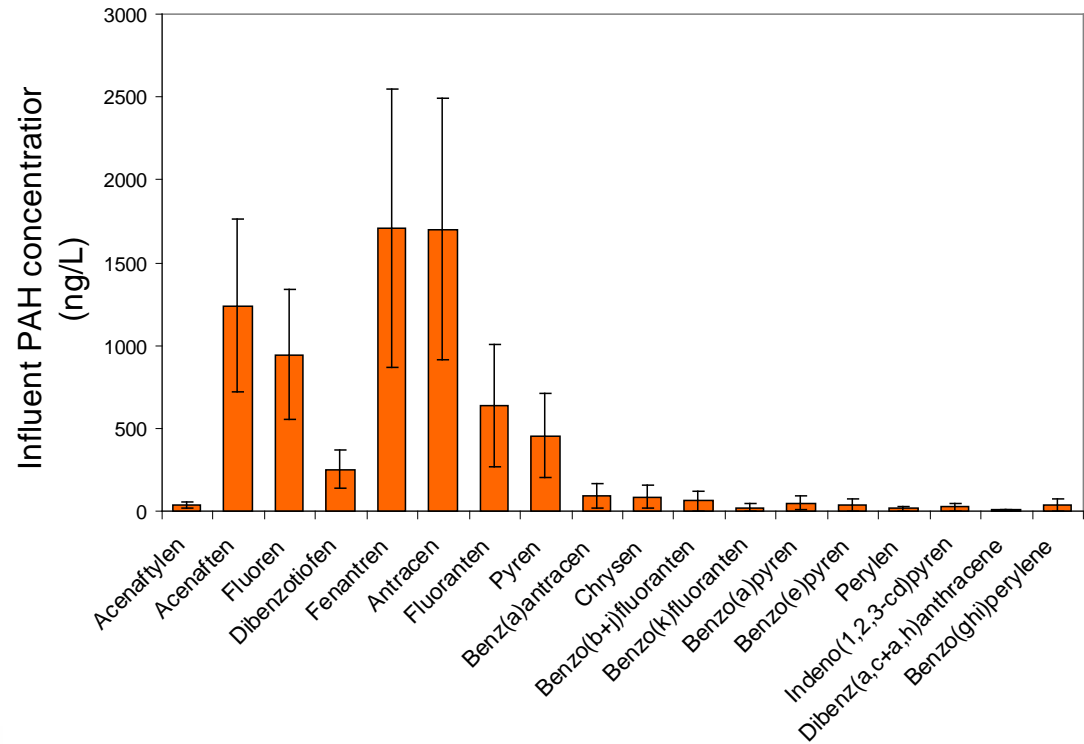
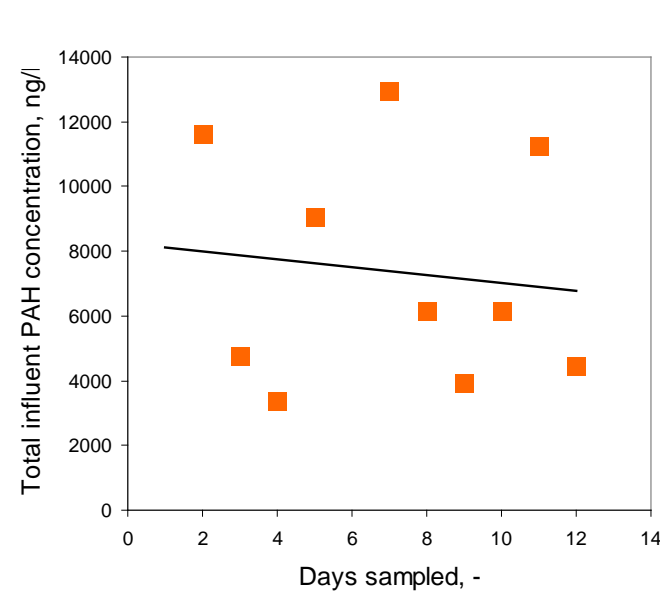
Compound	CAS no.	Charecteristics	Reference	Influent	Effluent	Sludge
Acenaphthene	83-32-9	Irritant	Faust, 1994	X	X	X
Anthracenedione	84-65-1	Irritant	ITII, 1988	X	X	
Anthracene	120-12-7	Photosensitising	ILO, 1983	X	X	X
Phenanthracene	85-01-8	Phototoxic, narcotic	Bellas et al., 2008	X	X	X
Benzophenone	119-61-9	Hepatotoxic, nephrotoxic	Chhabra, 2000	X	X	X
Bromoimidazole	2302-25-2	24h LD50 in rats: 1.7 - 3.4 millimoles/kg**	Verschoyle et al., 1987	X	X	X
Cyclopentaphenanthrene	80455-52-3	Mutagenic	Morrochi et al., 1996	X	X	X
Di isopropylnaphthalene	24157-81-1	Hepatotoxic	Hasegawa et al., 1982	X	X	X
Fluorene	86-73-7	Low toxicity	Bellas et al., 2008	X	X	X
Methyl octahydro anthracene	N/A	N/A	N/A	X	X	X
Methylthio benzothiazole	21564-17-0 *	7 day NOEC in crustacean C. dubia: 1.21mg/L**	Nawrocki et al., 2005	X	X	X
Phthalates	EDF-150	Estrogenic	Nakai et al. 1999	X	X	X
Phthalic acids	88-99-3	Estrogenic	Pavan et al. 2001., Ema et al., 1997	X	X	X
Tetramethyl Butylphenol	98-54-4	Narcotic	Russom et al., 1997	X	X	X

\*This is the code for TCMTB, the parent compound from which this one is a metabolite. The CAS number of MTBT is unknown for the authors.

\*\*This is the lethal dose for 50% of test group or the no observable effect concentration for these chemicals and not the type of toxicity

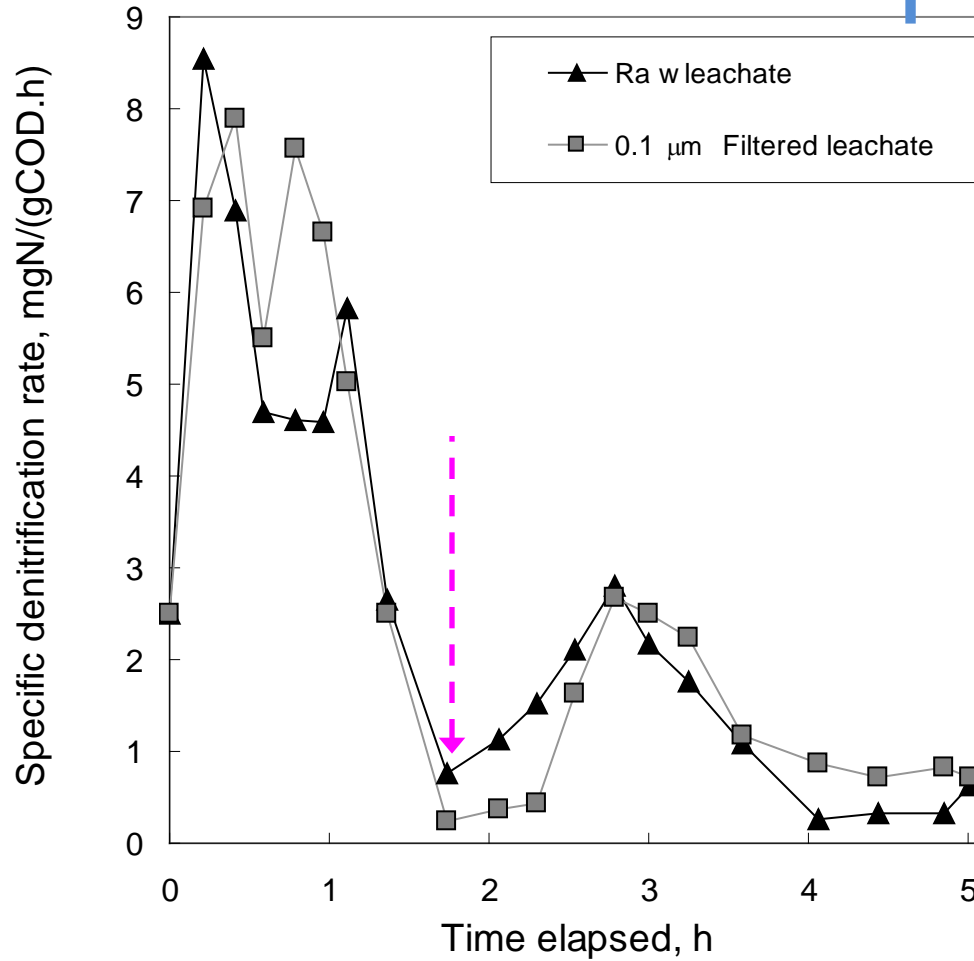


# PAH concentration in raw leachate



- Two main PAH groups (ave. conc.):
  - Group A: 254-1710 ng/L -
  - Group B: <93 ng/L

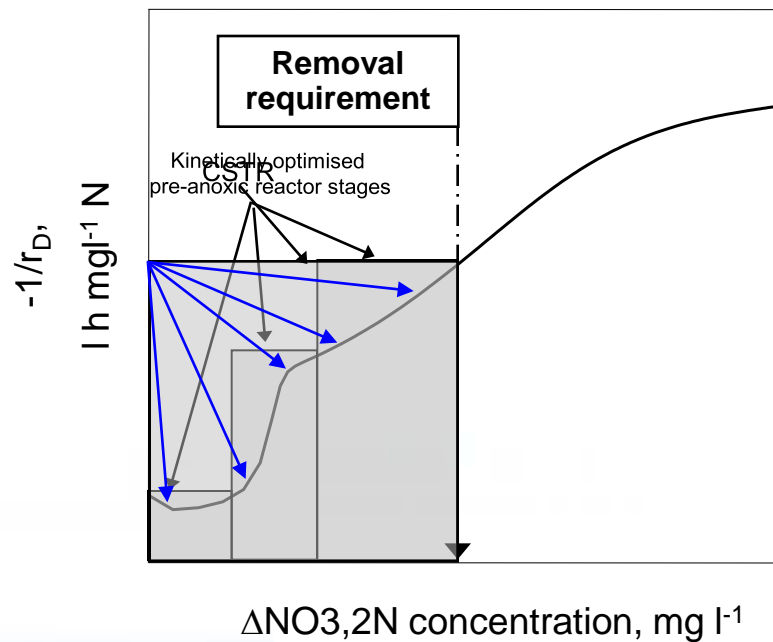
# Batch experiments



- Toxicity and inhibitory effects can effectively deteriorate denitrification rates
- Biodegradable substrates oxidised through two consecutive steps (no ASM);
- Biodegradation kinetics can limit denitrification capacity in CSTR anoxic reactors;

# Kinetic optimisation of anoxic bioreactor configuration

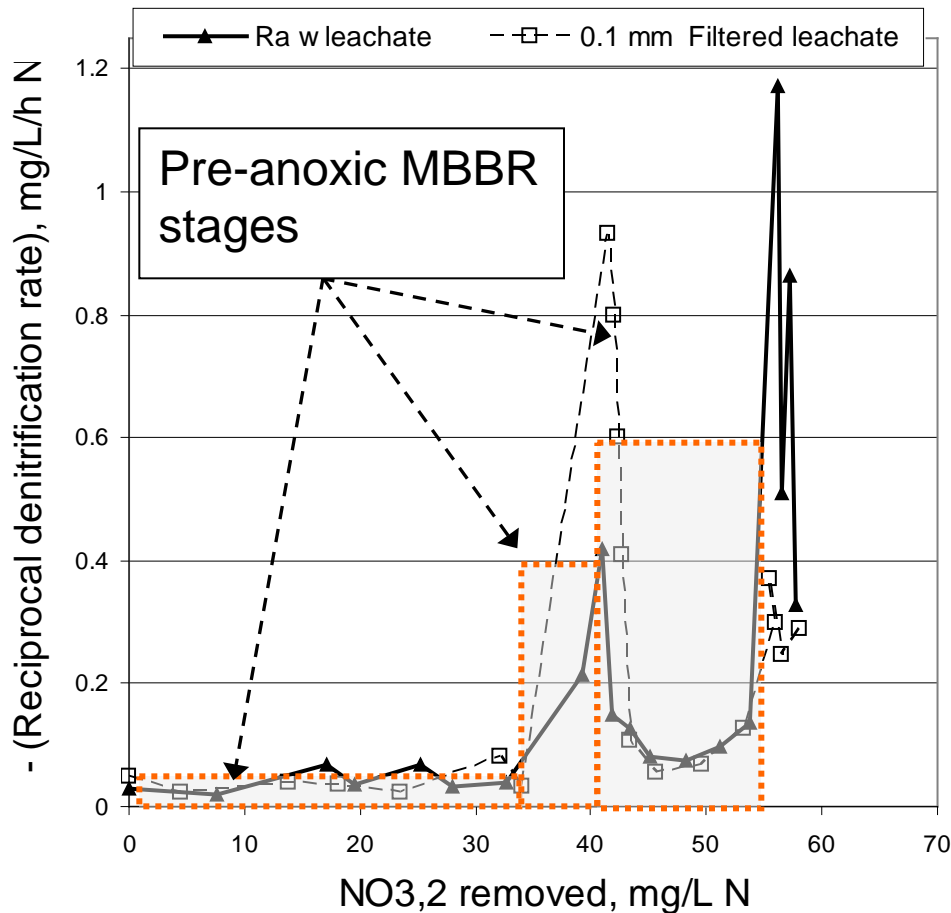
"Determining the best system configuration for a given conversion."



$$HRT_{Ax} = \frac{V_{Ax}}{F} = (\Delta NO_{3,2}) \times \left( -\frac{1}{r_N} \right)$$

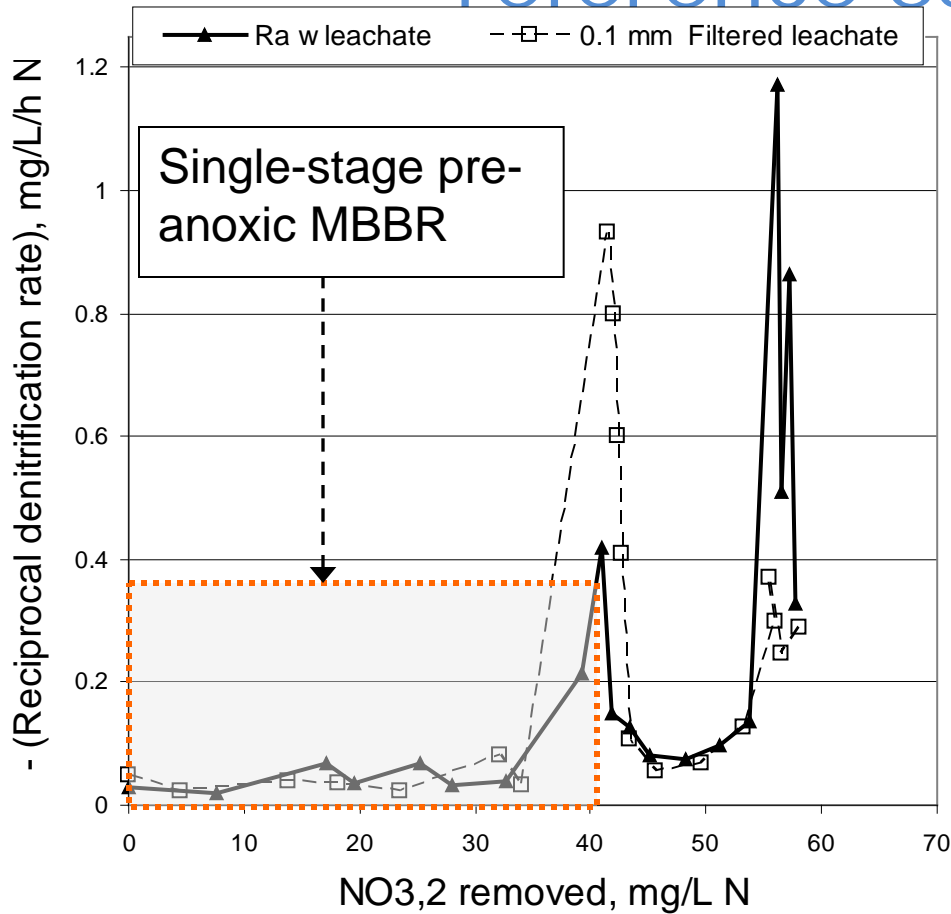
\* Plósz, B.Gy. (2007) Optimization of the activated sludge anoxic reactor configuration as a means to control nutrient removal kinetically. *Wat. Res.* 41(8), 1763–1773.

# 3-stage pre-anoxic MBBR design

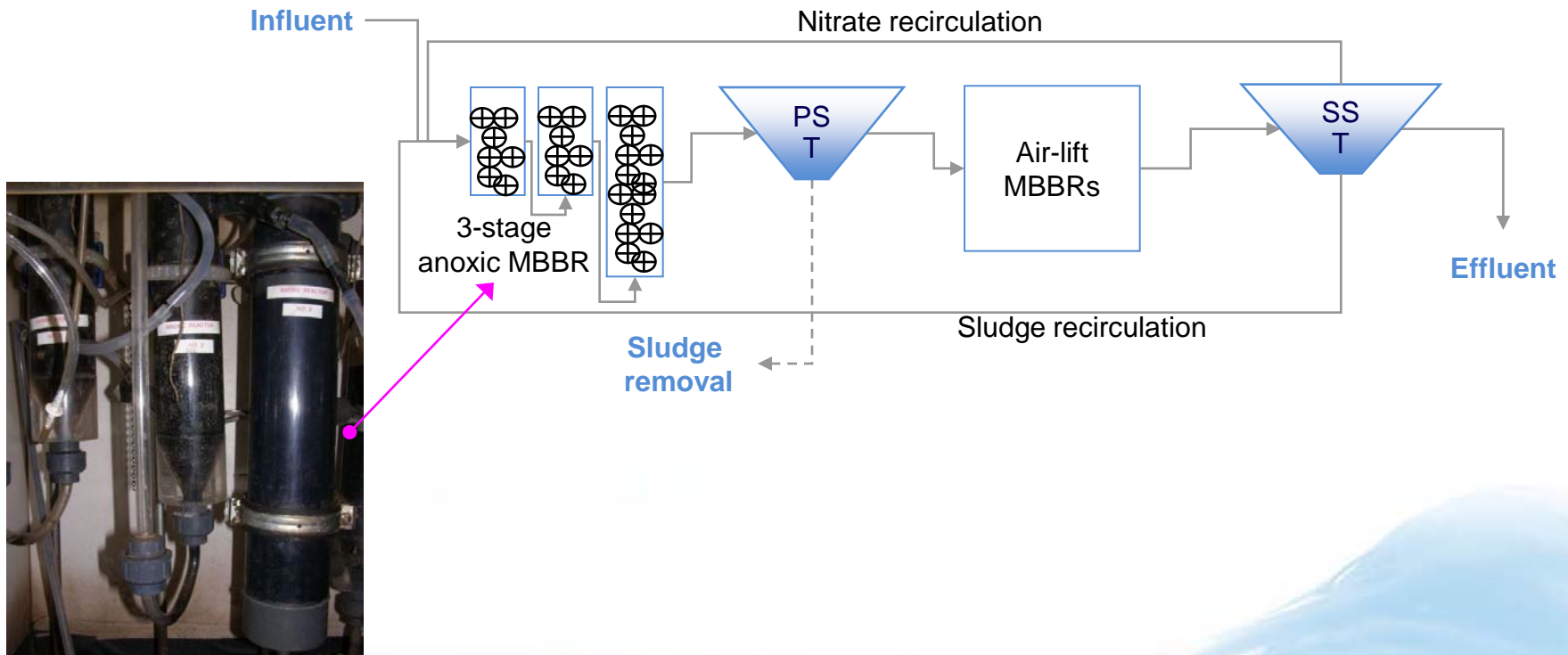




# Single-stage pre-anoxic MBBR – reference scenario

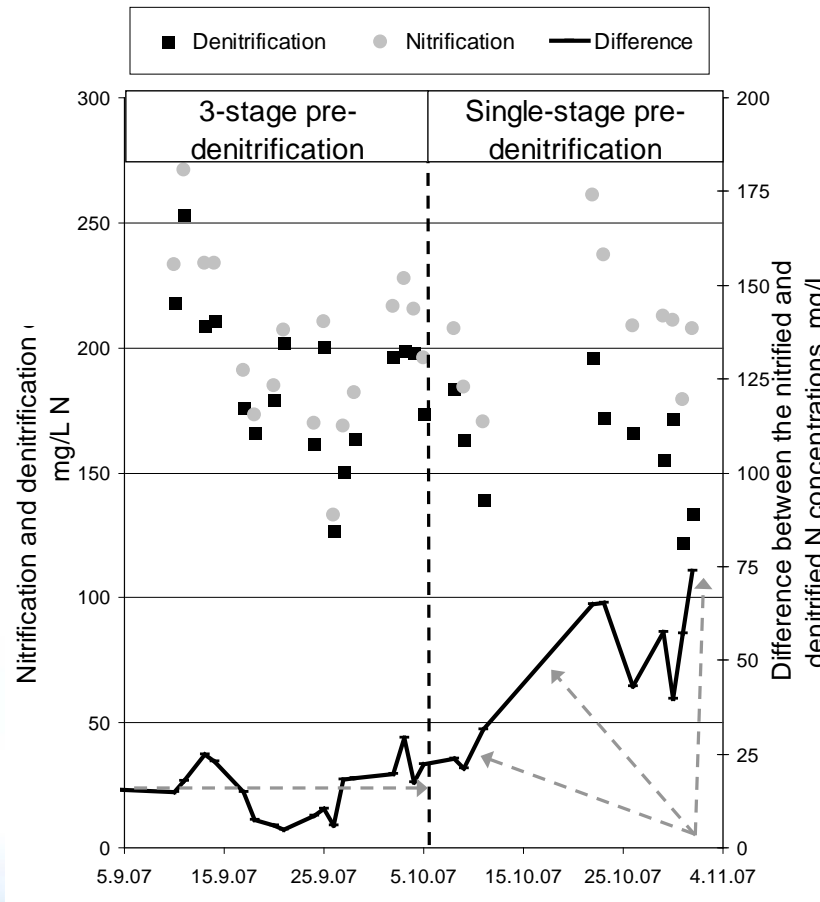


# Anoxic-aerobic, two-stage moving bed biofilm system



\* Plósz, B.Gy. (2007) Optimization of the activated sludge anoxic reactor configuration as a means to control nutrient removal kinetically. *Wat. Res.* 41(8), 1763–1773.

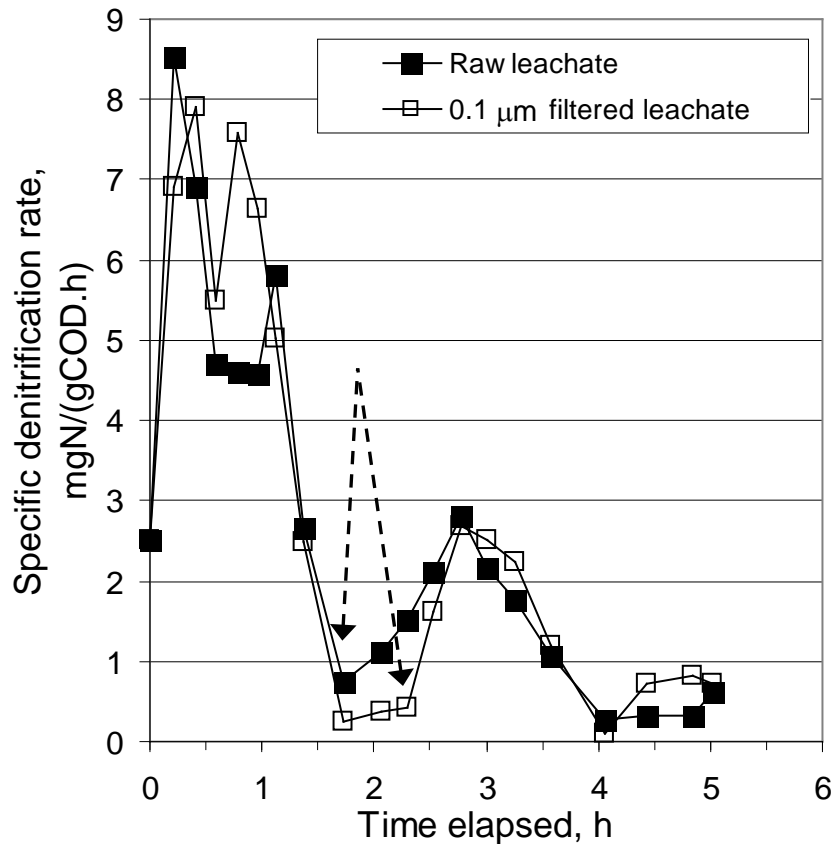
# Biological nitrogen removal



- The 3-stage reactor arrangement is shown to be effective to assure low effluent nitrate concentrations;
- For operating with a single-stage pre-anoxic unit using the same carriers, denitrification gradually deteriorated;

# Batch experiments

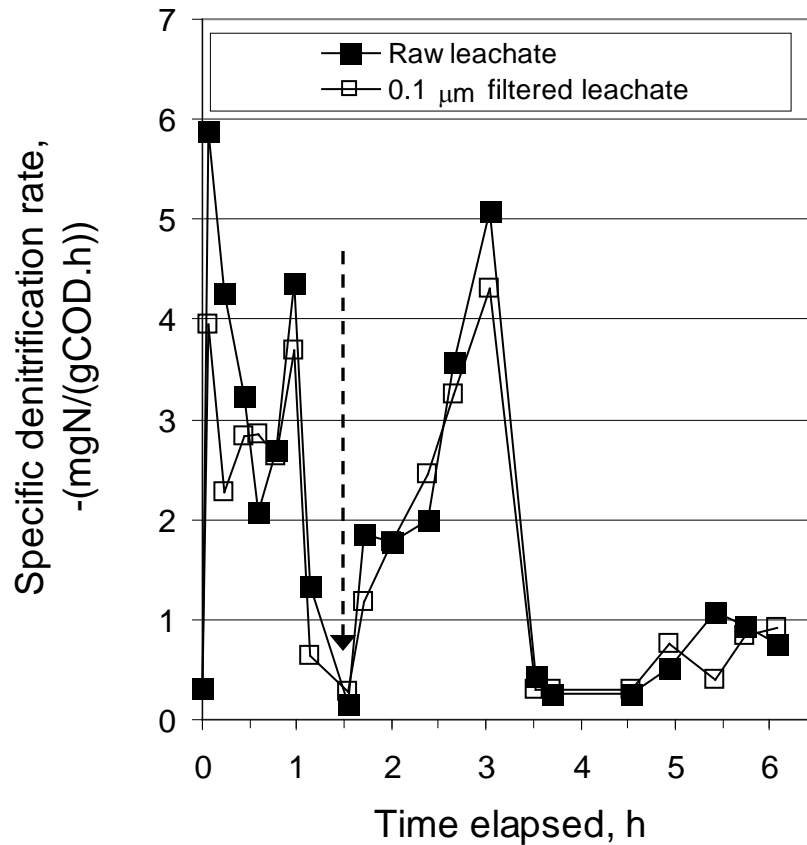
- Biofilm sample: stage 1





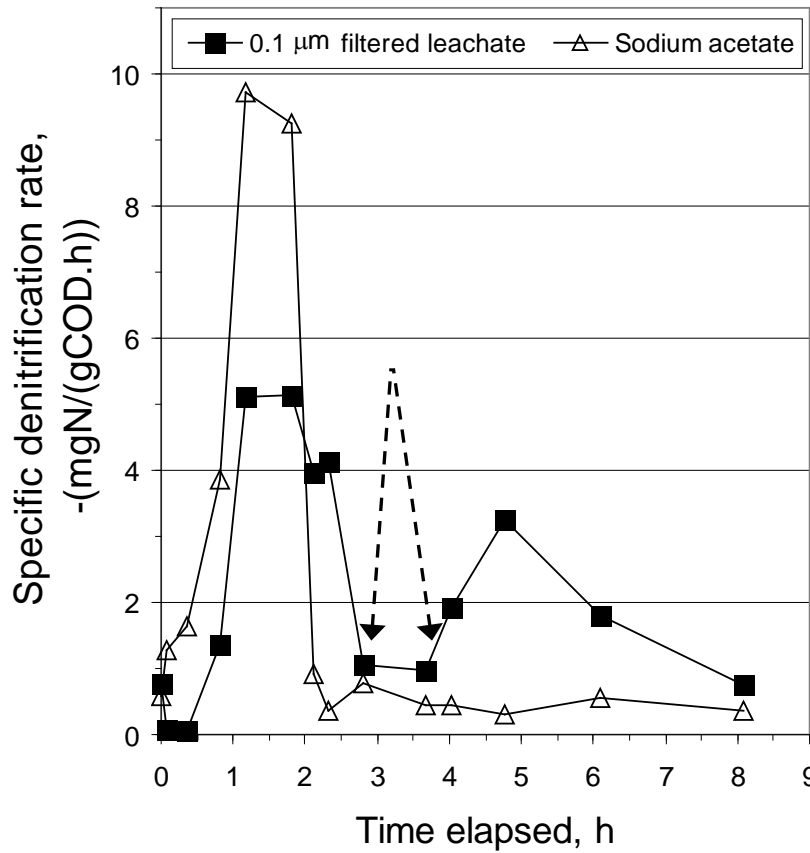
# Batch experiments

- Biofilm sample: stage 3

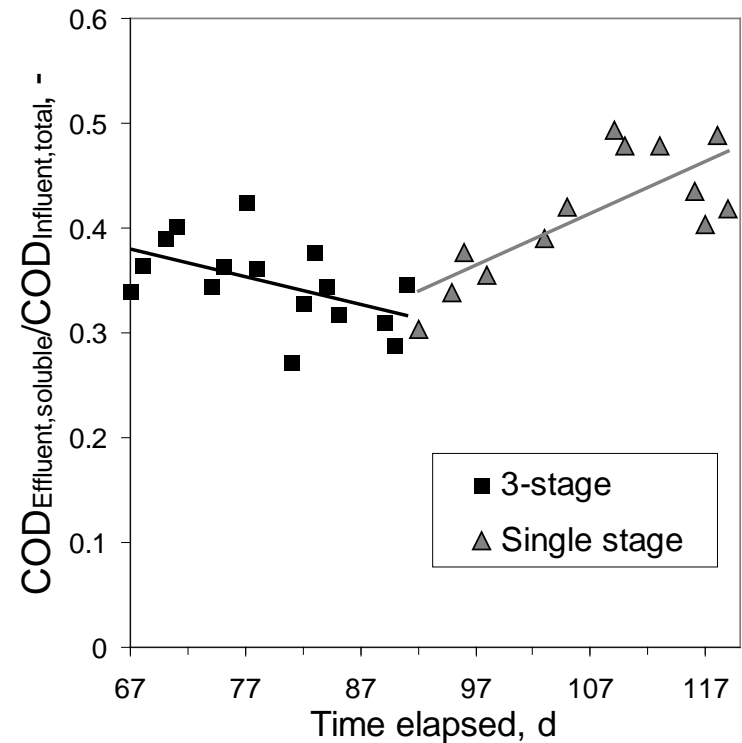
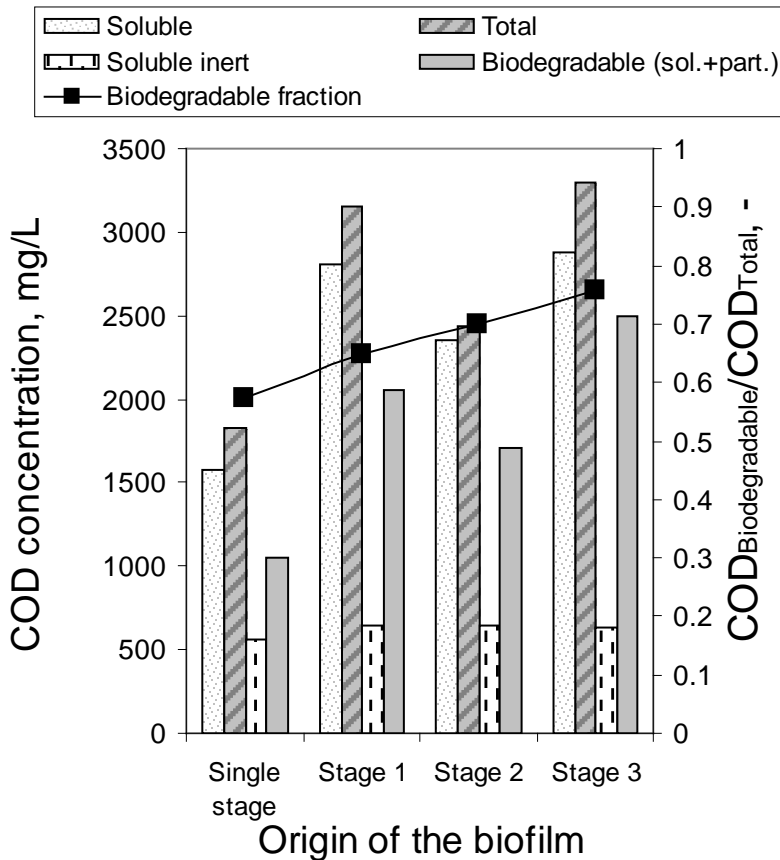


# Batch experiments

- Biofilm sample: single-stage with raw leachate and with effluent treated leachate spiked with Na-AcO

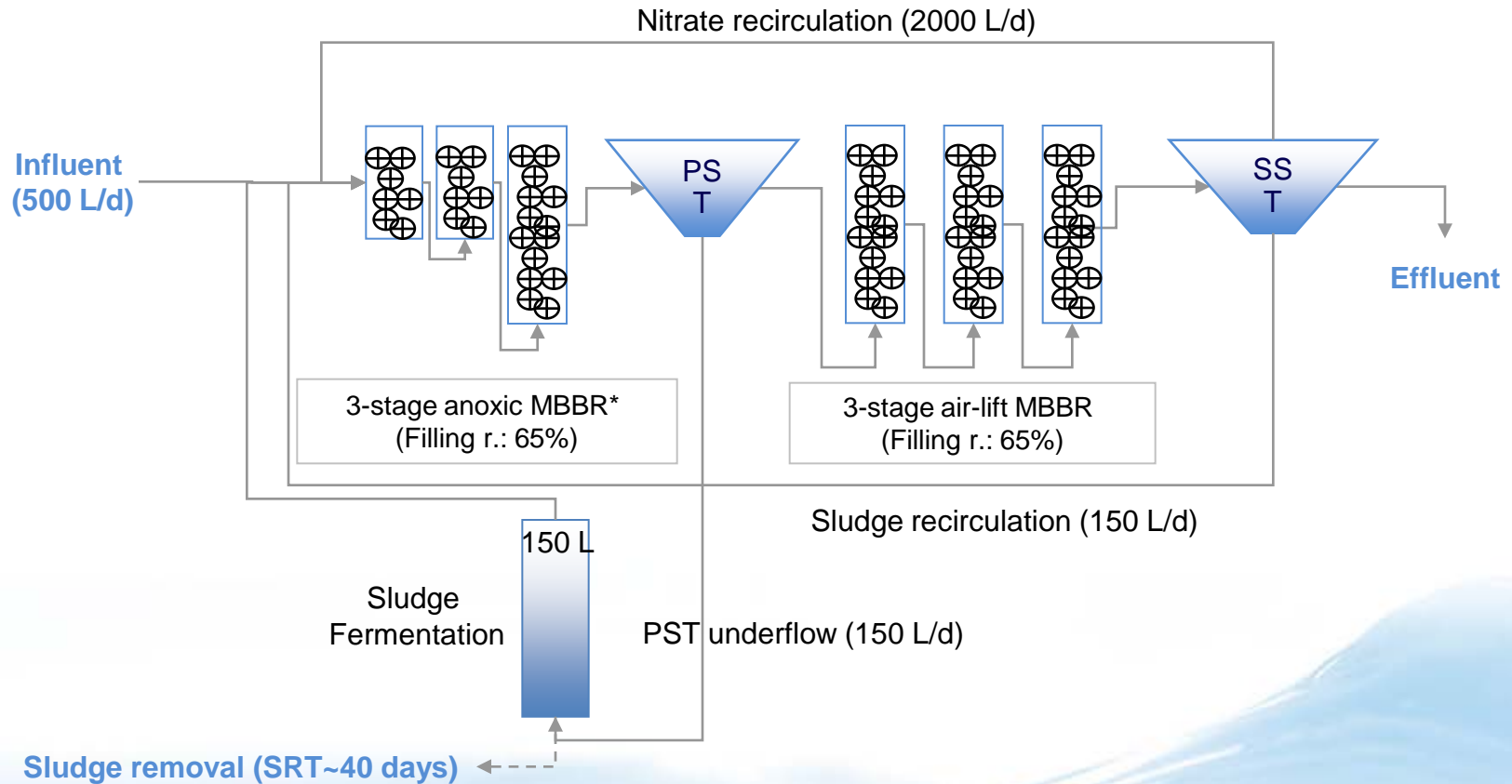


# Biodegradable COD



# BIOZO system – Biological treatment

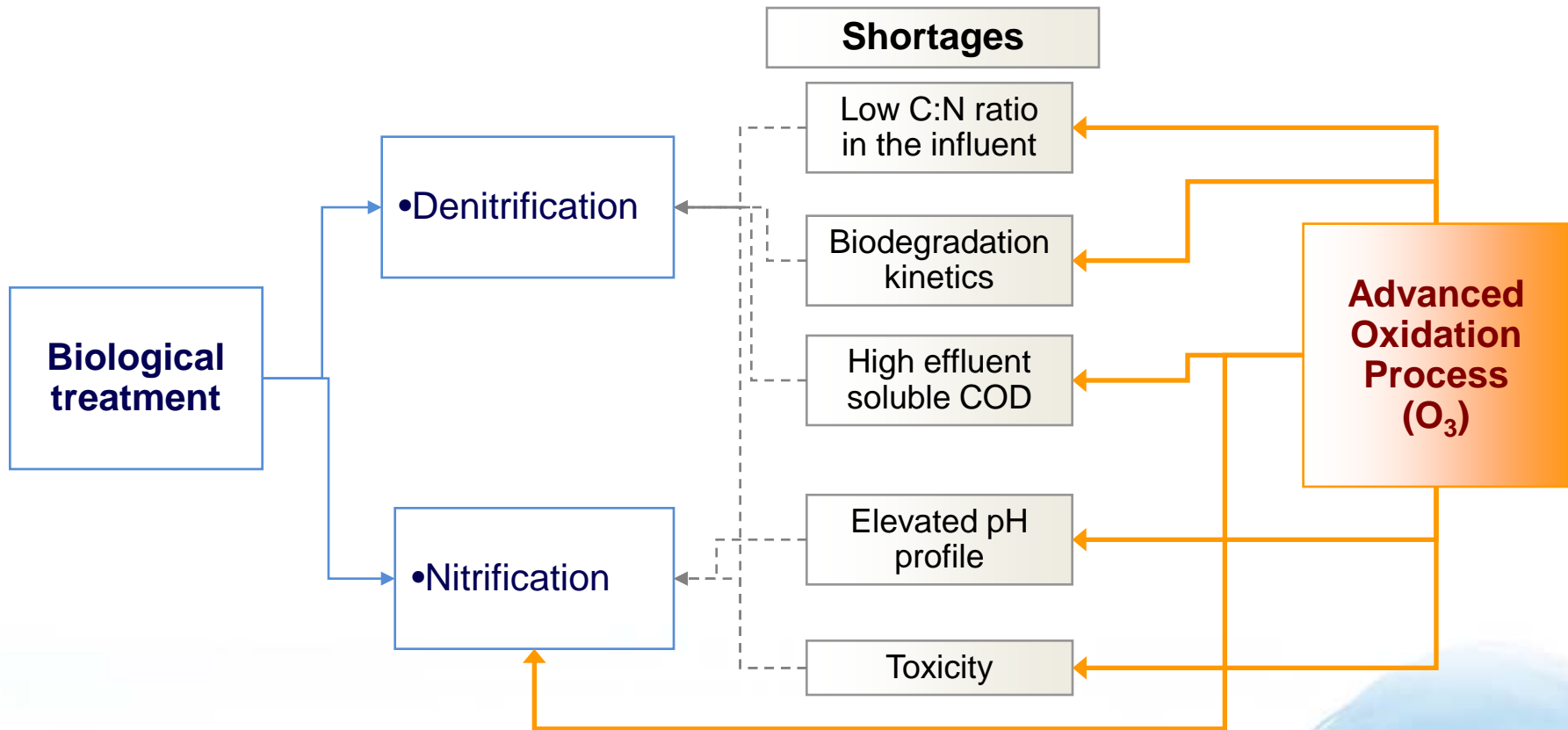
- Anoxic-aerobic moving bed biofilm system



\* Plósz, B.Gy. (2007) Optimization of the activated sludge anoxic reactor configuration as a means to control nutrient removal kinetically. *WATER RESEARCH* 41(8), 1763–1773.



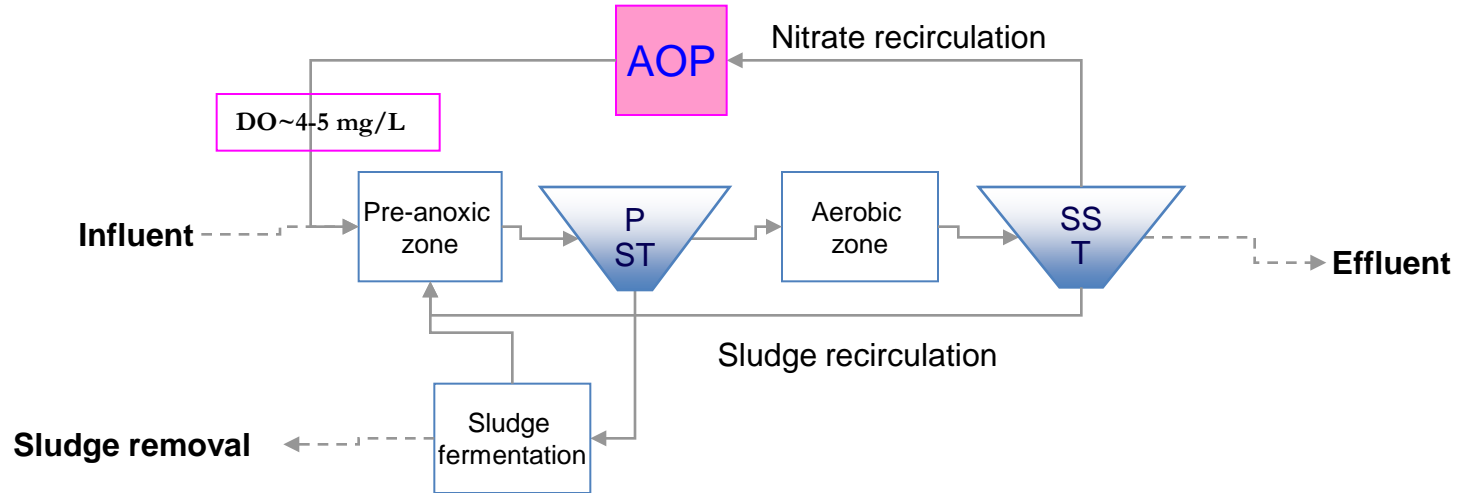
# Landfill leachate treatment



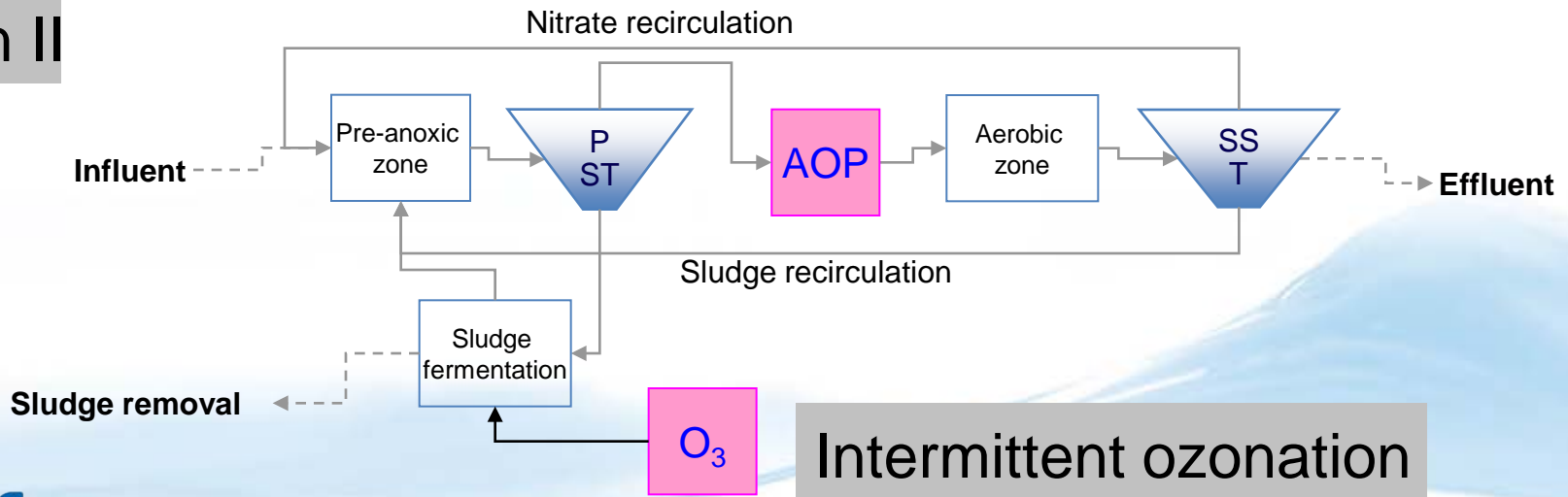
- What is the best reactor configuration to couple biological treatment with AOP?

# Position of AOP

## Position I



## Position II



# Combined system

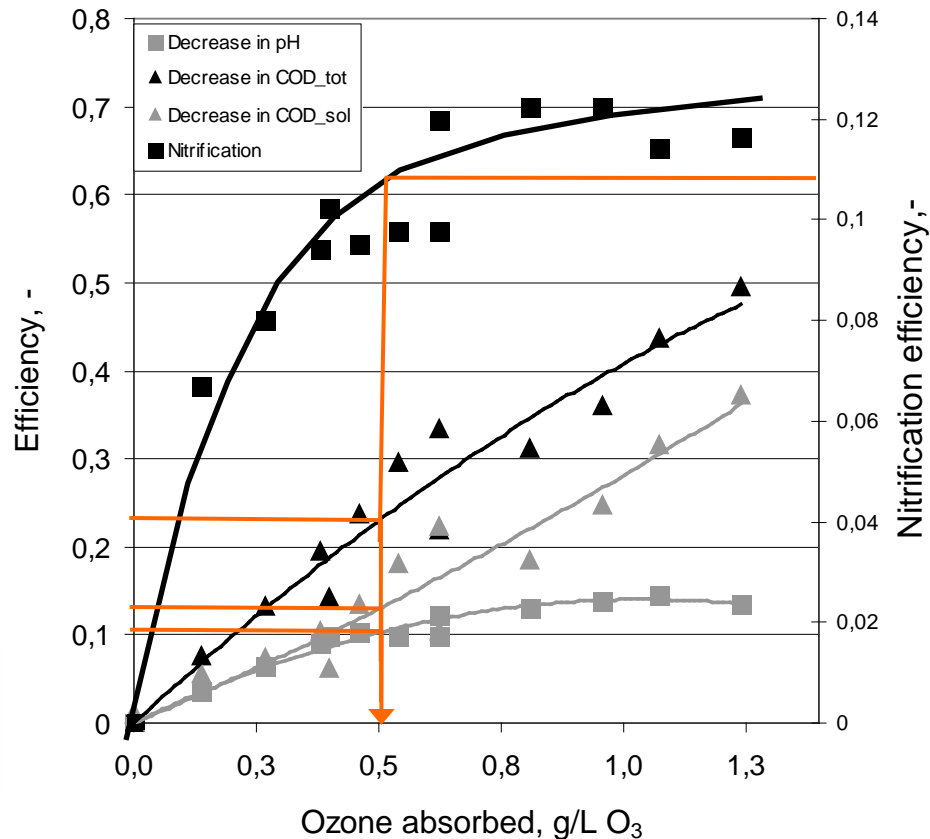
## Position 1

- (i) the relatively low alkalinity and ammonium/ammonia concentration, i.e. lower **scavenging effect**;
- (ii) the need to increase the biodegradation substrate concentration and thus the **C:N** ratio in the anoxic zone.
- *Pitfall*: the elevated dissolved oxygen (**DO**) concentration in the influent of the anoxic zone (on average 4-5 mg/L).

## Position 2

- (i) the more effective oxidation – the AOP unit situated in the **main line** and not in the recirculation stream;
- (ii) its potential to **reduce toxicity** in the influent of the aerobic zone, and thus the inhibitory impacts on the autotrophic nitrifiers;
- (iii) the potential of **nitrifying bacteria** to degrade degradation products derived from XOM oxidation in the AOP;
- (iv) the need to decrease the **pH** and alkalinity in the aerobic zone;
- (v) the high **DO** concentration in the effluent of the AOP.

# Ozonation efficiency



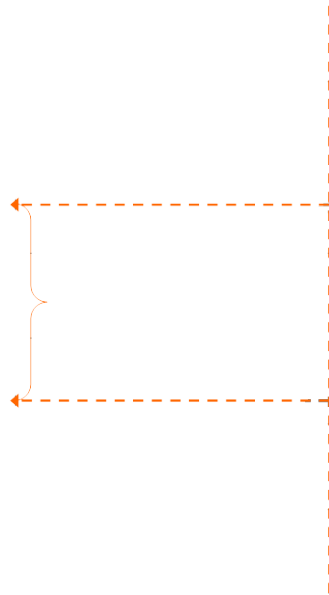
Using the effluent of the biological treatment system, an optimum ozone dosage of approx. 0.5 g/L O<sub>3</sub> can efficiently :

- Increase nitrification capacity (ca. 10 %)
- Decrease total COD (ca. 20%)
- Decrease soluble COD (ca. 10%)
- Decrease pH (ca. 10%)

Initial concentration values in the leachate solution

NH <sub>4</sub> N (mg/L)	COD <sub>tot</sub> (mg/L)	COD <sub>sol</sub> (mg/L)	pH (-)
64,2	856	670	8,04

# Cost assessment

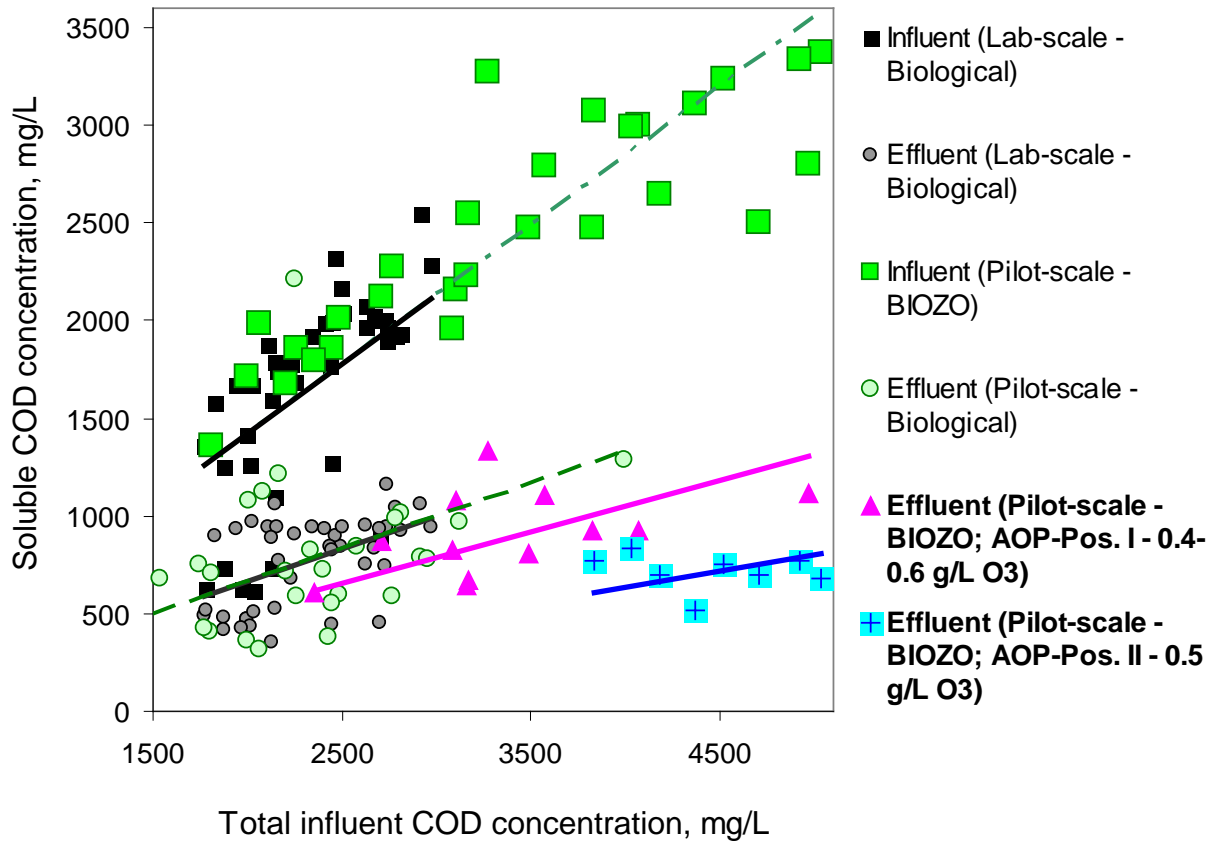


- Operation costs:  
0.34 – 1.16 euros per m<sup>3</sup> leachate treated
- Average cost values with 1 and 2 euros per kg O<sub>3</sub>:  
0.45 and 0.91 €/m<sup>3</sup>, respectively using an average soluble effluent COD value of **911** mg/L.

- *Operating costs of ozonation should be contrasted against costs related to*
  - acid dosing;
  - external substrate dosing;
  - aeration for nitrification;
- *Additional benefits of ozonation: non-biodegradable COD removal*

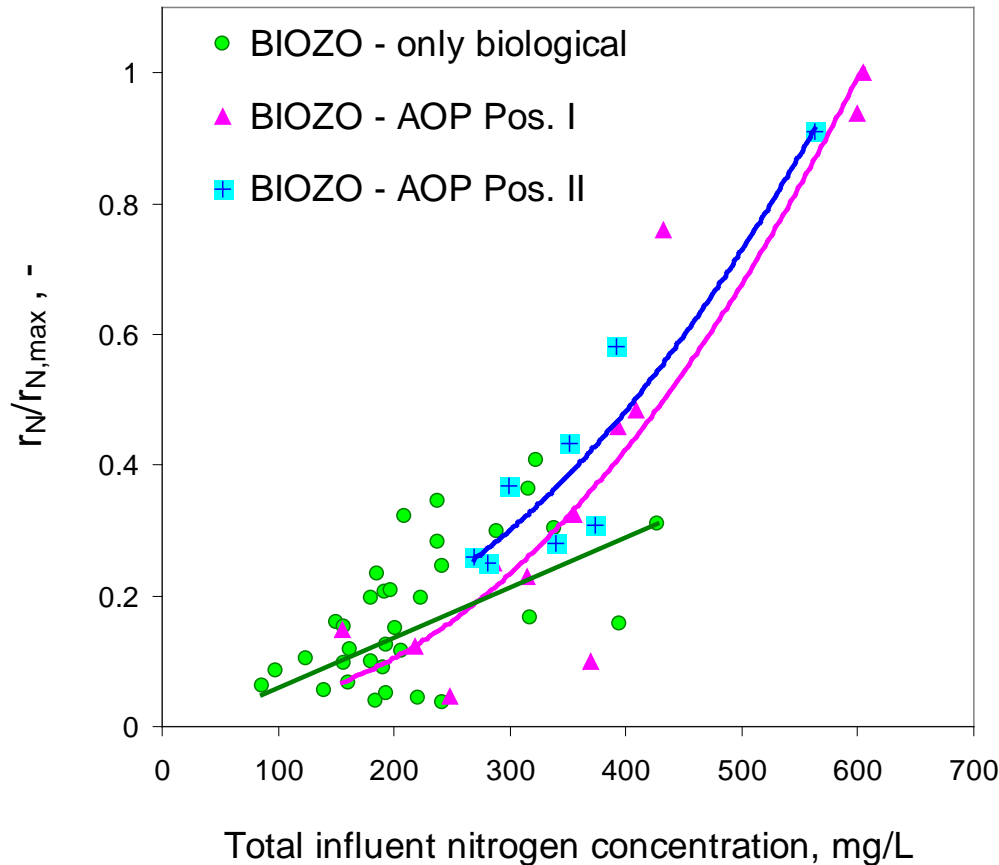


# COD removal



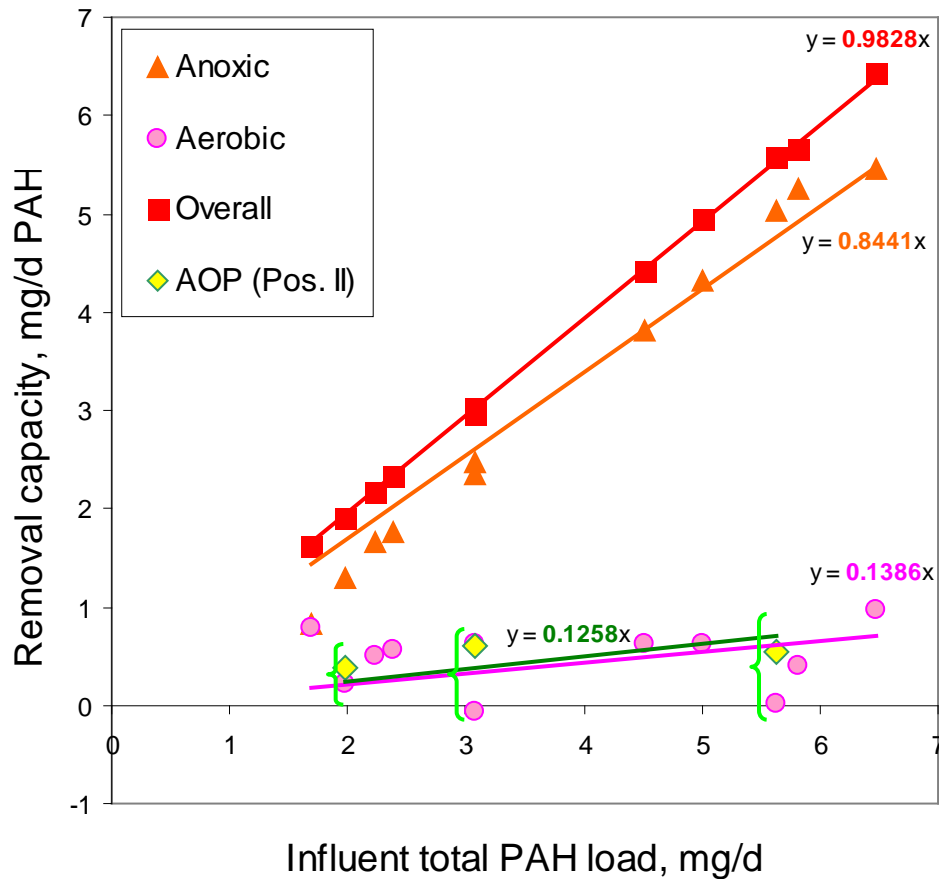
Combined operation with AOP installed upstream to the aerobic zone is an effective means to decrease  $COD_{Soluble, Eff}$ , thereby keeping it below 1000 mg/L COD.

# Nitrogen removal



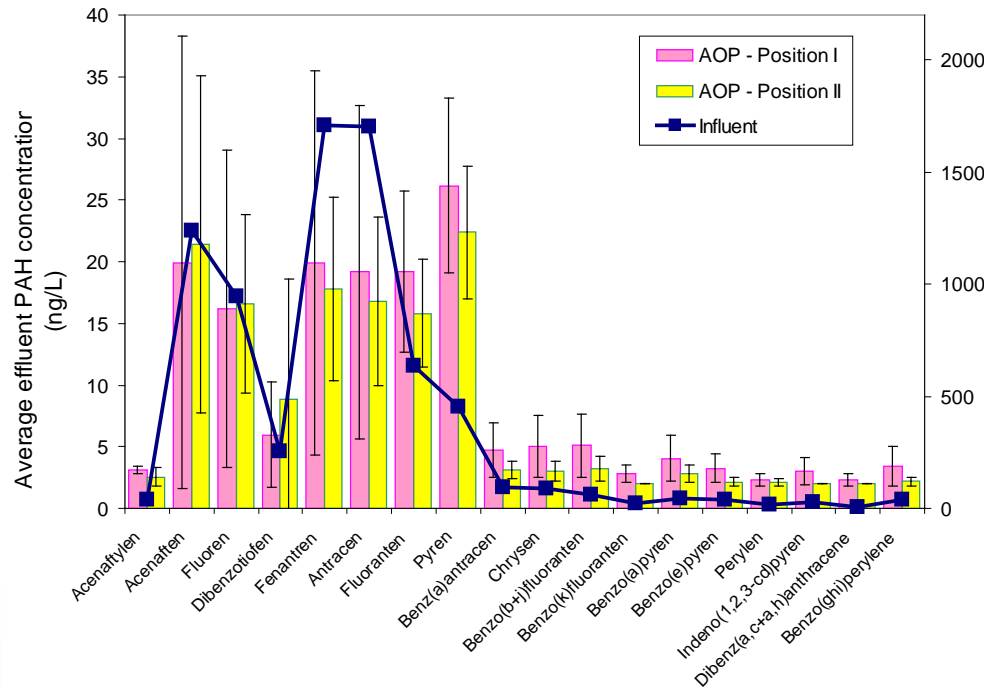
- Combined biological-AOP operation can significantly increase the overall nitrogen removal in the BIOZO system.
- Some but no significant difference between AOP installed in position II and position I.

# Total PAH removal



- High overall capacity to remove PAH;
- AOP installed in **Pos. II** can potentially decrease the PAH load on the aerobic zone, thereby mitigating
  - inhibitory impacts on nitrification and
  - decrease the effluent PAH.

# AOP positions and PAH removal

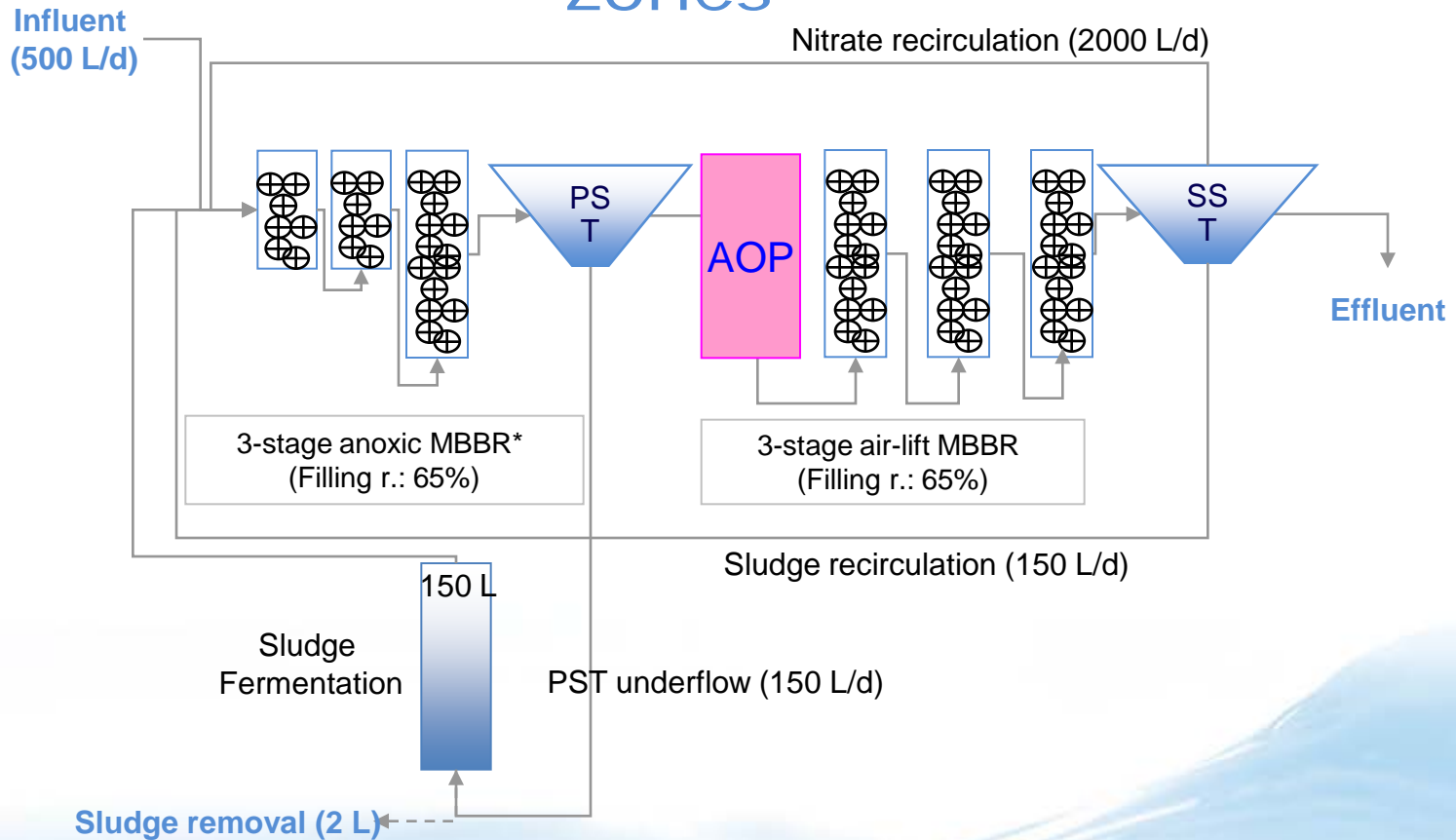


Influent PAH concentration (ng/L)

	Suggested treatment target for BIOZO (µg/l - µg/g DS)			
	Conservative		Minimum	
	Water	Sediment/sludge	Water	Sediment/sludge
Naphtalene	1.200	0.029	80	1
Acenaphthylene	0.130	0.003	3.3	0.085
Acenaphthene	0.380	0.016	5.8	0.358
Fluorene	0.250	0.026	5	0.511
Phenanthrene	1.300	0.500	5.1	1.168
Anthracene	0.110	0.030	0.36	0.101
Fluoranthene	0.120	0.013	0.9	0.097
Pyrene	0.023	0.140	0.023	1.4
Benzo(a)anthracene	0.001	0.001	0.018	0.009
Chrysene	0.007	0.003	0.07	0.028
Benzo(b)fluoranthene	0.030	0.024	0.1	0.049
Benzo(k)fluoranthene	0.027	0.174	0.1	0.387
Benzo(a)pyrene	0.050	0.250	0.1	0.5
Dibenzo(ah)anthracene	0.030	0.058	0.1	0.116
Benzo(ghi)perylene	0.002	0.002	0.1	0.0031
Indeno(123cd)pyrene	0.002	0.005	0.1	0.007

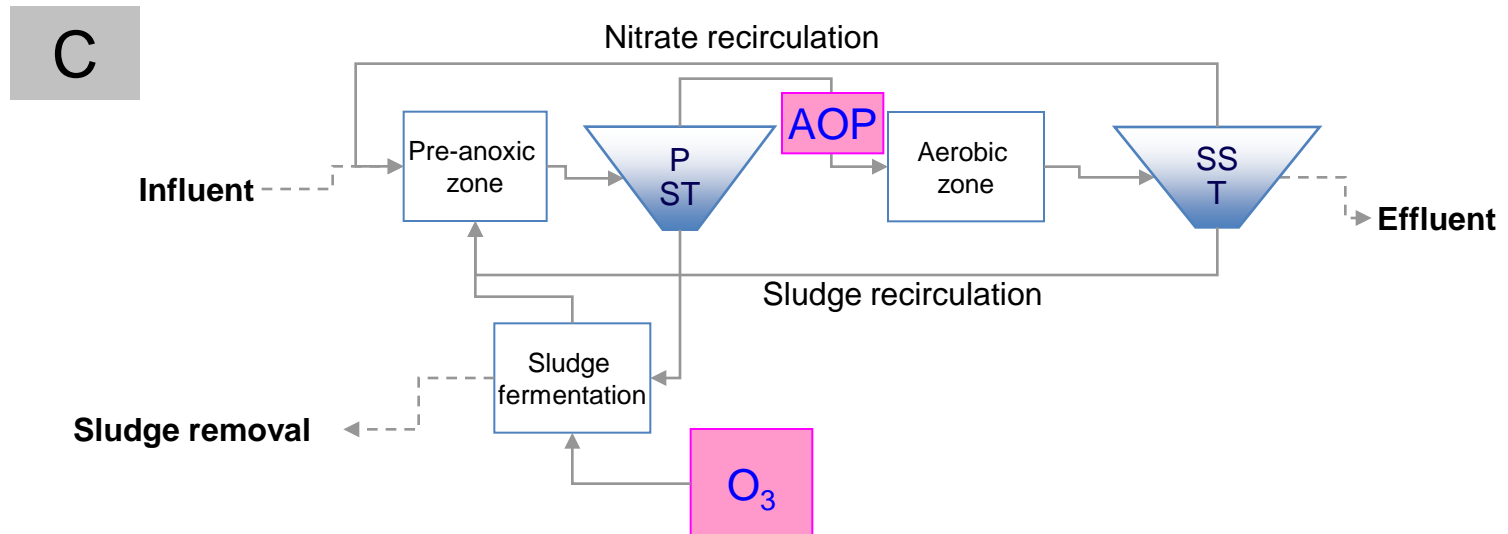
- BIOZO with AOP installed in Pos. II can meet the effluent criteria for all the contaminants.

## BIOZO system – Biological treatment combined with AOP between the pre-anoxic and aerobic zones



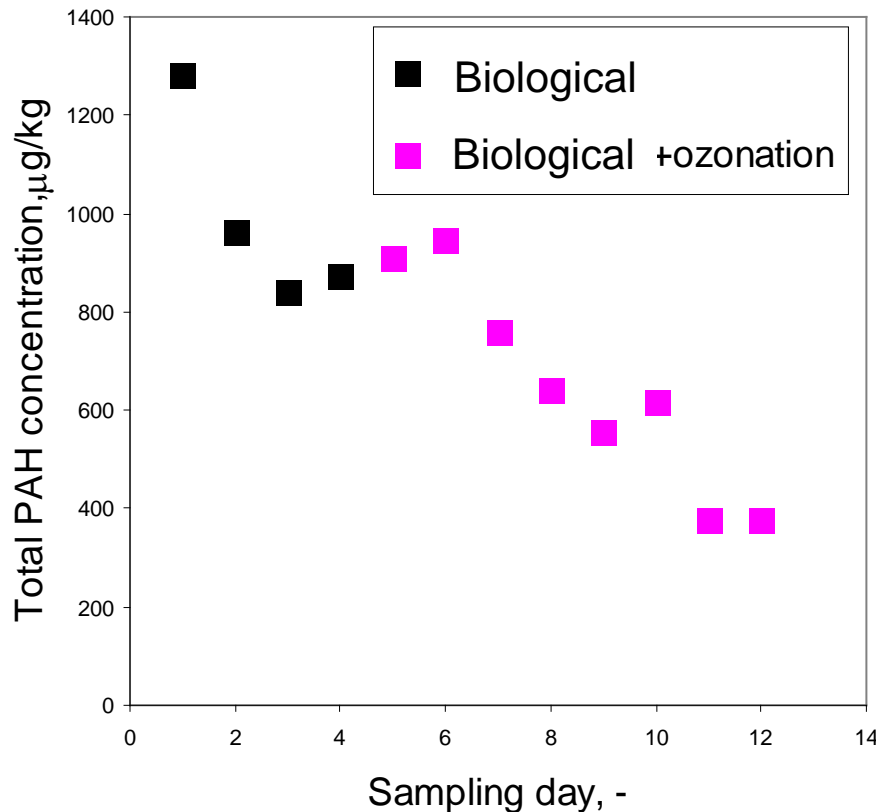


# Intermittent sludge ozonation



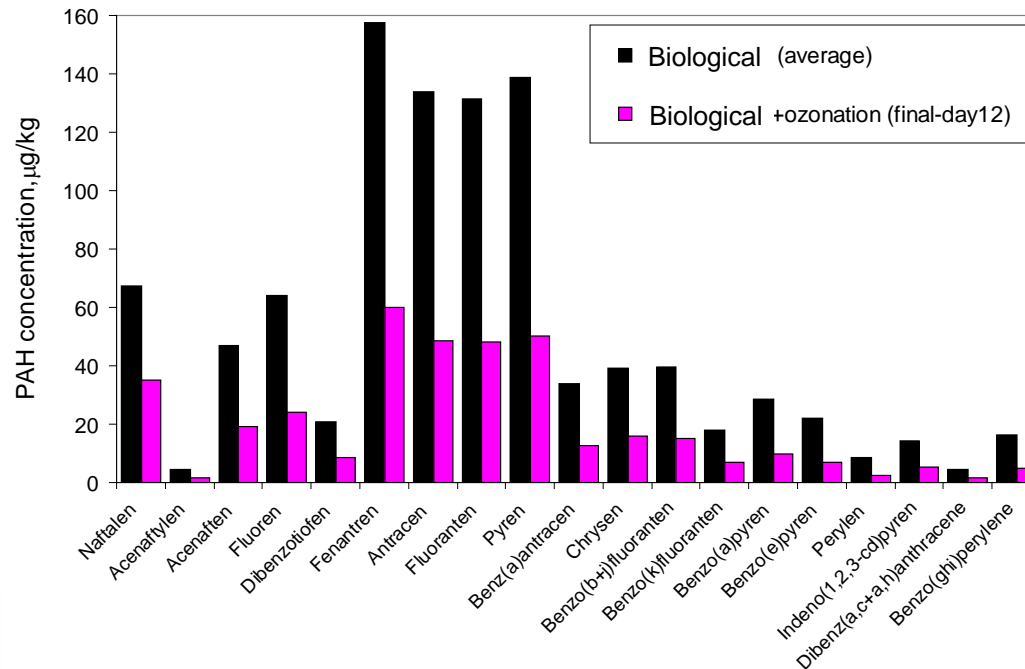
- **Assessment of the sequential ozonation of the sludge in the fermentation unit using 0.05 mgO<sub>3</sub> / mgTSS. the optimum secondary BIO-AOP layout**
- **To increase the biodegradable COD in the anoxic zone**
- **To improve sludge quality**
- **To decrease the sludge production**

# Sludge quality - PAH removal



- Sludge quality can be significantly improved by intermittent sludge ozonation.

# PAH solids concentration



# Conclusions

- **Testing of the BIOZO landfill leachate treatment systems is successfully completed in pilot-scale;**
- **In the combined BIOZO system, the AOP installed upstream to the aerobic zone is superior over the nitrate-recirculation line, and thus is selected as the final design;**
- **The BIOZO system can effectively remove COD, nitrogen and XOC from the leachate, thereby providing a robust engineering solution to complying with strict regulations for sewer discharge criteria;**
- **Intermittent sludge ozonation can significantly improve the solids quality and can improve nitrogen removal in the system;**

# Conclusions

- Biodegradation kinetics can limit denitrification capacity in CSTR anoxic reactors;
- Nitrogen and organic substrate removal can be increased by using the kinetically optimised biofilm reactor design because
  - of the higher denitrification rates in the system;
  - Bacteria can be more effectively acclimatised;



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# Acknowledgements

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