Abatement of methane diffuse emissions by biotrickling filtration using polyurethane foam as carrier

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Main current challenges for air pollution

- Particulate matter
- Carbon monoxide and CO_2
- Volatile Inorganic Compounds (VICs)
  - Sulphur: SO_2, SO_3, H_2SO_4, H_2S, R-SH
  - Nitrogen: NO_x, NH_3, R-NH_2

- Volatile Organic compounds: VOCs
  - Ketones, aldehydes, acids, etc.
- Odours
  - H2S, mercaptanes, VFAs, etc.
- Non CO_2 – Greenhouse Gases (GHGs)
  - CH_4, N_2O

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Non-CO_2 Greenhouse Gases (GHGs)

- GHGs: Methane
  - CH_4 has 25 times more impact on global warming than CO_2
  - Wastewater treatment: 2.5% US emissions (2012)
  - Dumps, WWTPs and other wastes: up to 31% of CH_4 emissions (Spain, 2007)
- GHGs: Nitrous oxide
  - N_2O has 310 times more impact on global warming than CO_2
  - Wastewater treatment: 1.6% US emissions (2012)
  - Around 0.4% of the oxidized NH_3 during nitrification and 0.2% of reduced nitrate during denitrification is emitted as N_2O

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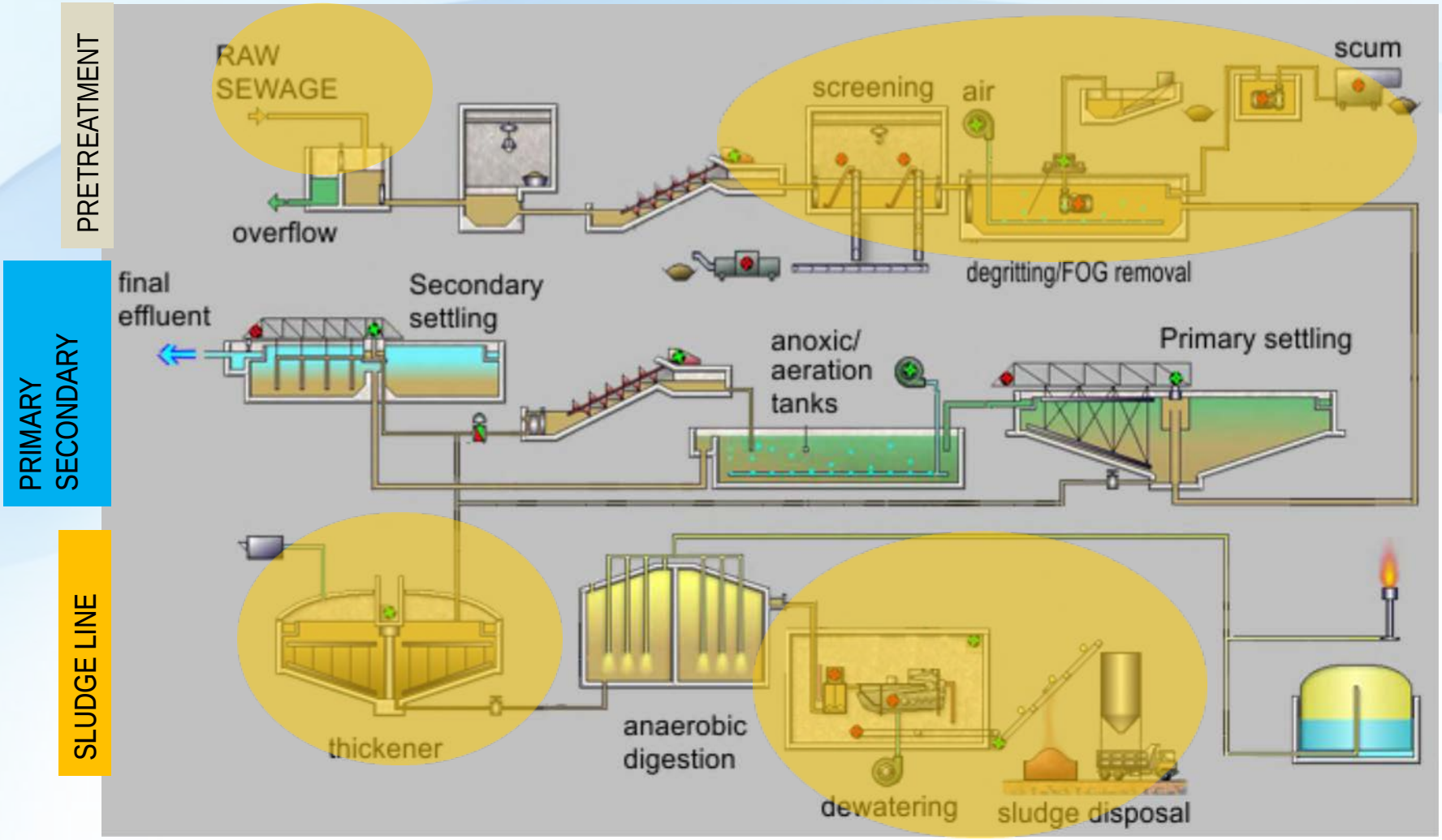
Occurrence in STPs

VOCs and Odours



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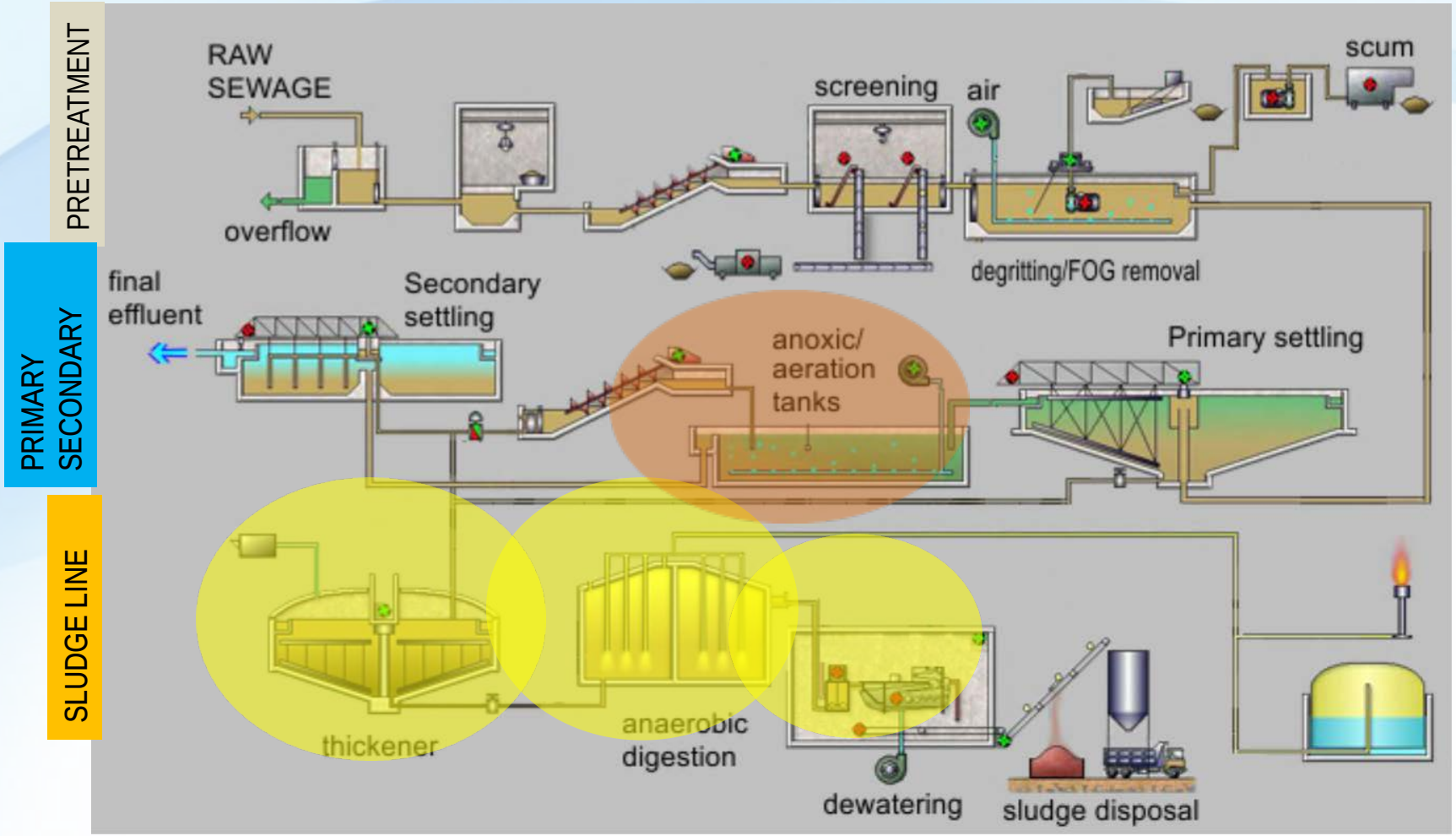
GHGs (CH_4 and N_2O)



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Introduction

Regarding Greenhouse gases emissions

CH_4 and **N_2O** (**Greenhouse gases, GHG)**, are 20-25 and 310 times more detrimental to the environment than CO_2, respectively

CH_4 concentration increased 143 % over the last 250 years and its contribution on the overall greenhouse effect is around 4-9 %

The **typical ranges** measured for methane are **500-5000** ppm, and for nitrous oxide are **0-500** ppm

Introduction

Regarding methane biofiltration

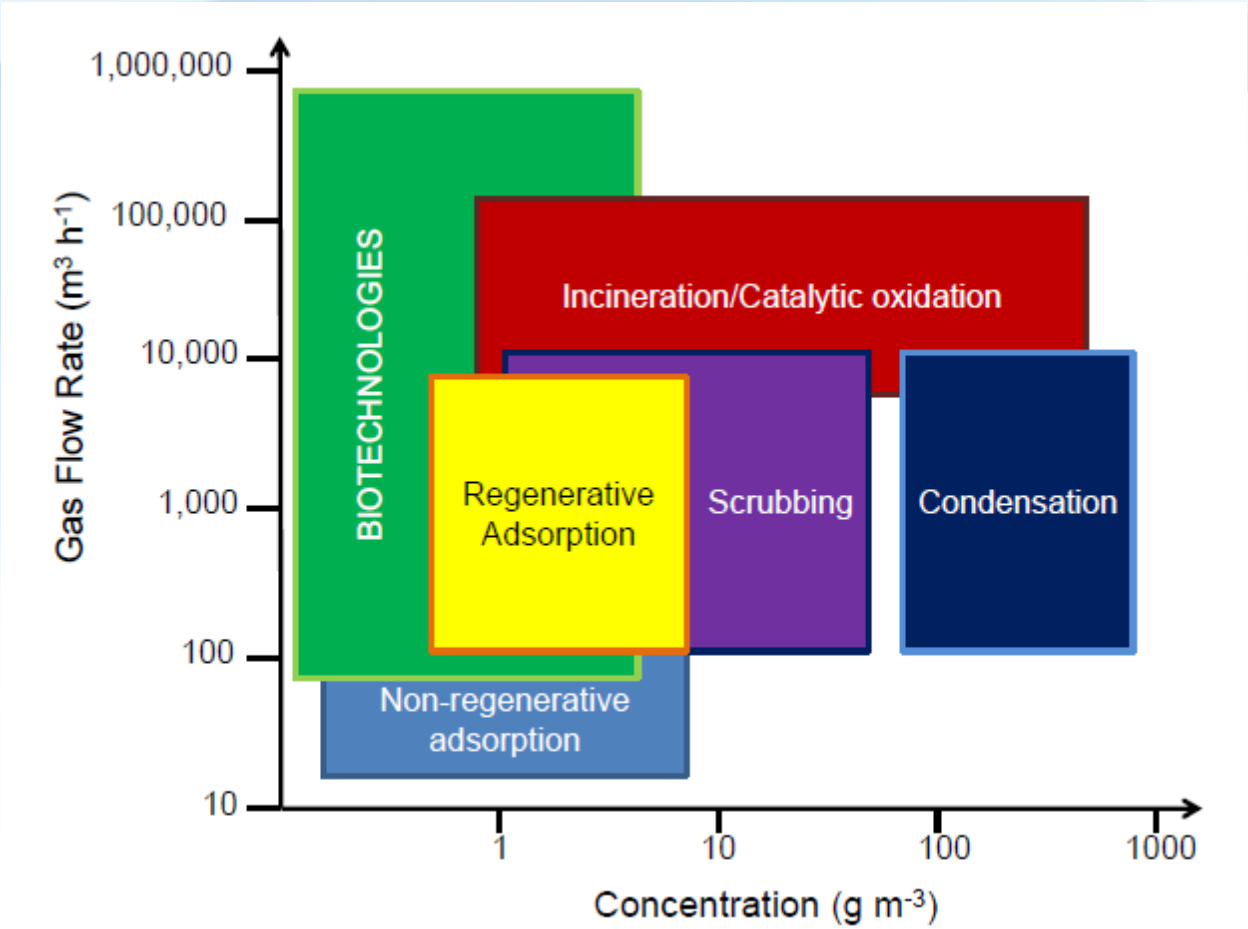
- The global reaction for **methane oxidation** in a biofilter can be expressed as:

$$CH_4 + \alpha O_2 + \text{Nutrients} \xrightarrow{\text{Methanotrophs}} \varepsilon CO_2 + \eta H_2O + Q + \text{new biomass}$$

- CH_4 Henry constant value is 27.2 g aq^{-1} → **low solubility** → **mass transfer limitations**

- The **inoculum** employed is a key factor → enriched methanotrophs culture might reduce startup of the bioreactor; i.e. type I (MG705, MG84), type II (MA450)

Gaseous effluents treatment technologies



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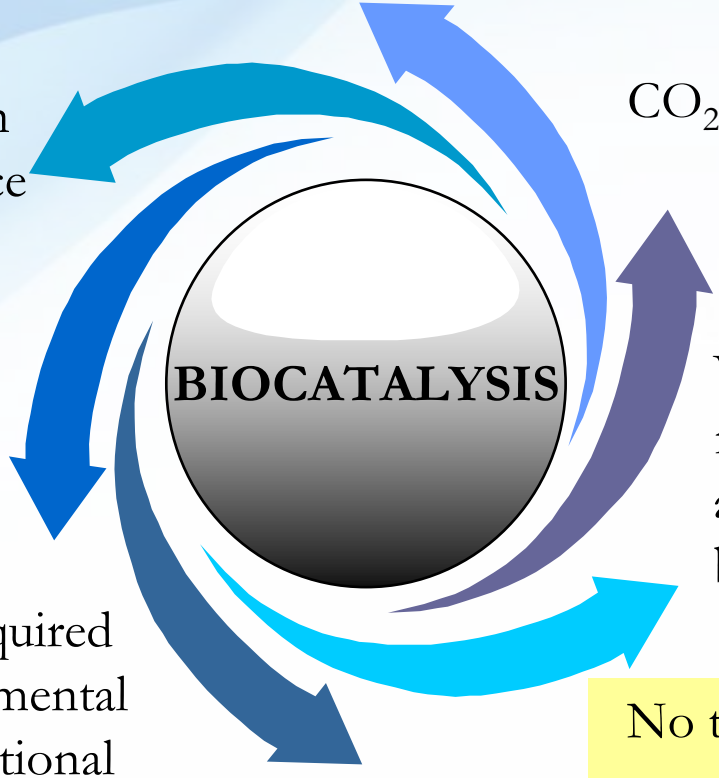
Biological treatment

$$COV + O_2 + \text{Nutrients (N, P, S, trace elements)} \xrightarrow{\text{Microorganisms}} CO_2 + H_2O + \text{Microorganisms} + \text{Energy}$$



VOCs as carbon and energy source

Ambient T and P conditions

Less energy is required (thus less environmental impact and operational costs)

CO_2 footprint is much lower than incineration
No fuel is required

VOCs are transferred from gas phase to aqueous phase prior to biodegradation

No toxic byproducts are used/generated

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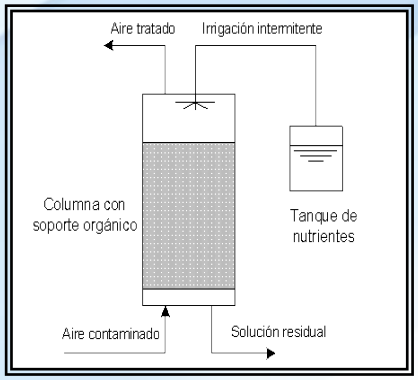
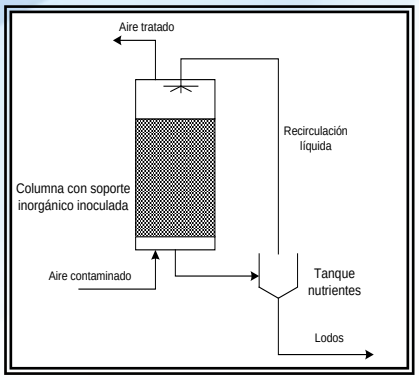
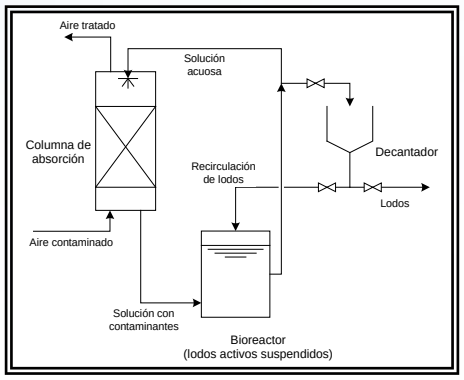
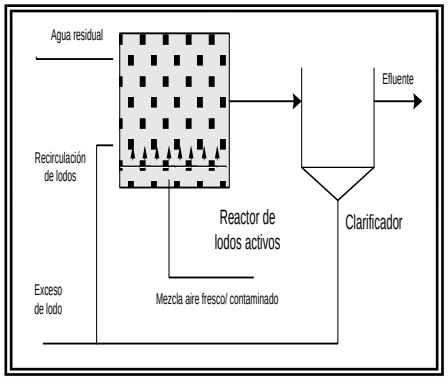
Biotechnologies

Biofilter (BF)	Biotrickling filter (BTF)	BioScrubber (BS)	Activated Sludge Diffusion (ASD)









Stationary aqueous phase	Mobile aqueous phase	Mobile aqueous phase	Stationary aqueous phase

Biomass in biofilm	Suspended biomass

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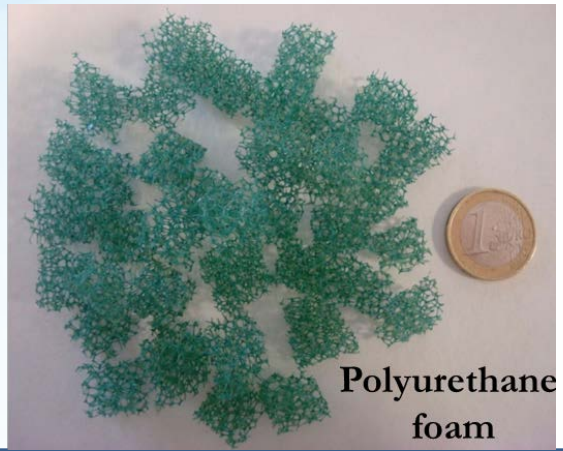
Characteristics of some common packing materials used in BTFs

	Size mm	Specific surface area $m^2 g^{-1}$	Void fraction -	Density $kg m^{-3}$	Bulk cost $€ m^{-3}$
GAC	2 - 5	800 - 1200	0.30 - 0.60	400 - 500	300 - 500
Porous ceramic rings	10 - 15	300 - 400	0.55	600 - 700	
Lava rock	20	0,60 - 0,65	0.50	750 - 850	40 - 80
Pall rings - polypropylene	50	100 - 300	0.90 - 0.95	50 - 80	400 - 500
Perlite (expanded)	4 - 6	2 - 10	0.40 - 0.60	50 - 150	60 - 100
Polyurethane foam	25	500 - 600	0.90 - 0.97	20 - 50	30 - 40

Kennes & Veiga, 2013



BIOTRICKLING FILTER

Advantages

- Easy control of the operation parameters (pH, moisture, nutrients)
- Microorganisms retention
- Low EBRT

Disadvantages

- Low transfer area
- Low efficiency for hydrophobic compounds
- Excessive biomass growth

HIGH EFFICIENCIES AT LOW EBRT: 1-10 s

H_2S 90-100 % COVs < 40 %

Henry Law < 1 ($H = C_g/C_l$)

Investment 5-20 € /(m^3h^{-1})

Operation 2-8 € /(m^3h^{-1})

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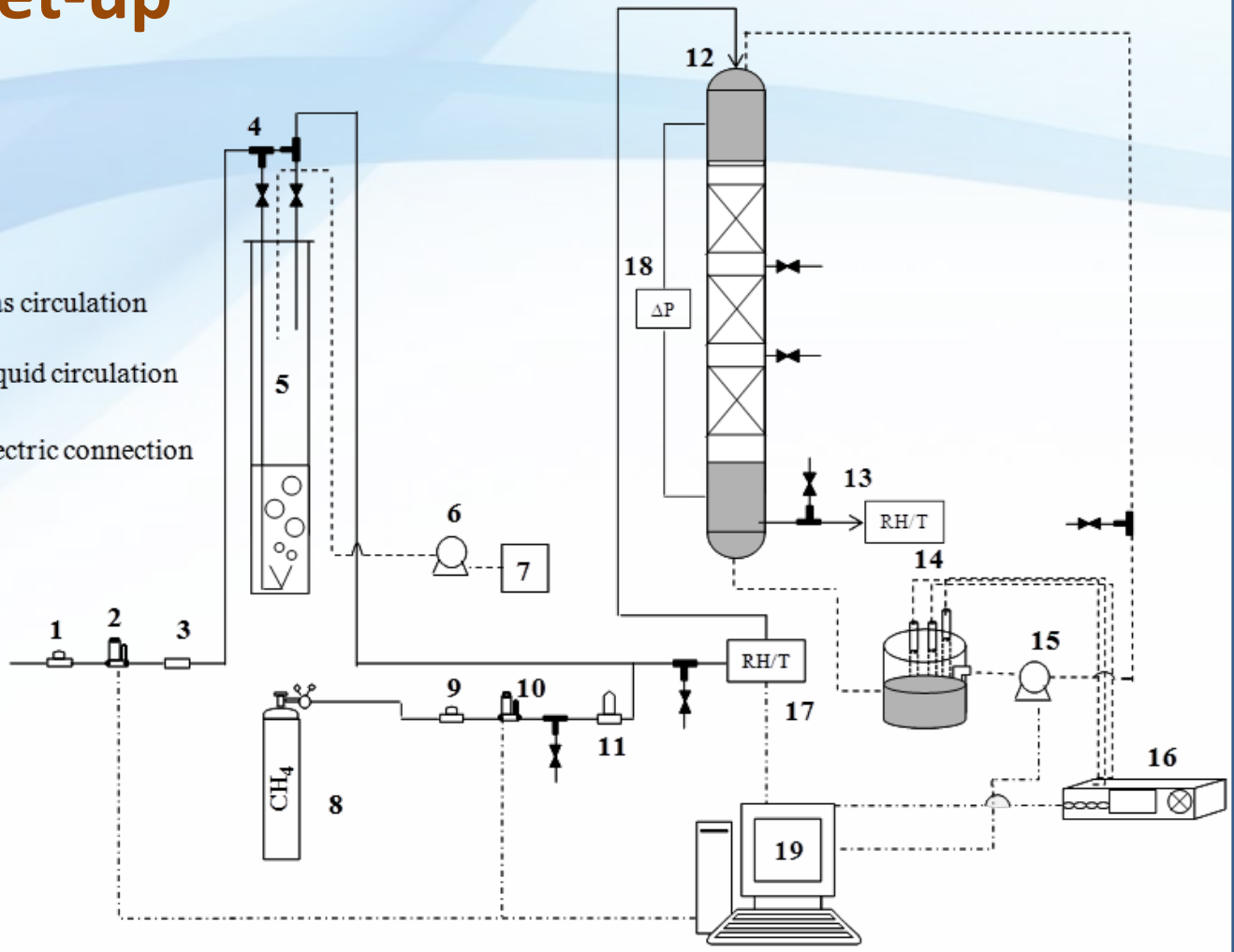
Methane biological abatement: Experimental set-up

- **Biofilter volume:**
  - Total: 13,4 L
  - Packing: 5,9 L
- **Packing:**
  - Polyurethane foam
- **Feeding:**
  - Mixture air + 0,2 - 1% CH_4



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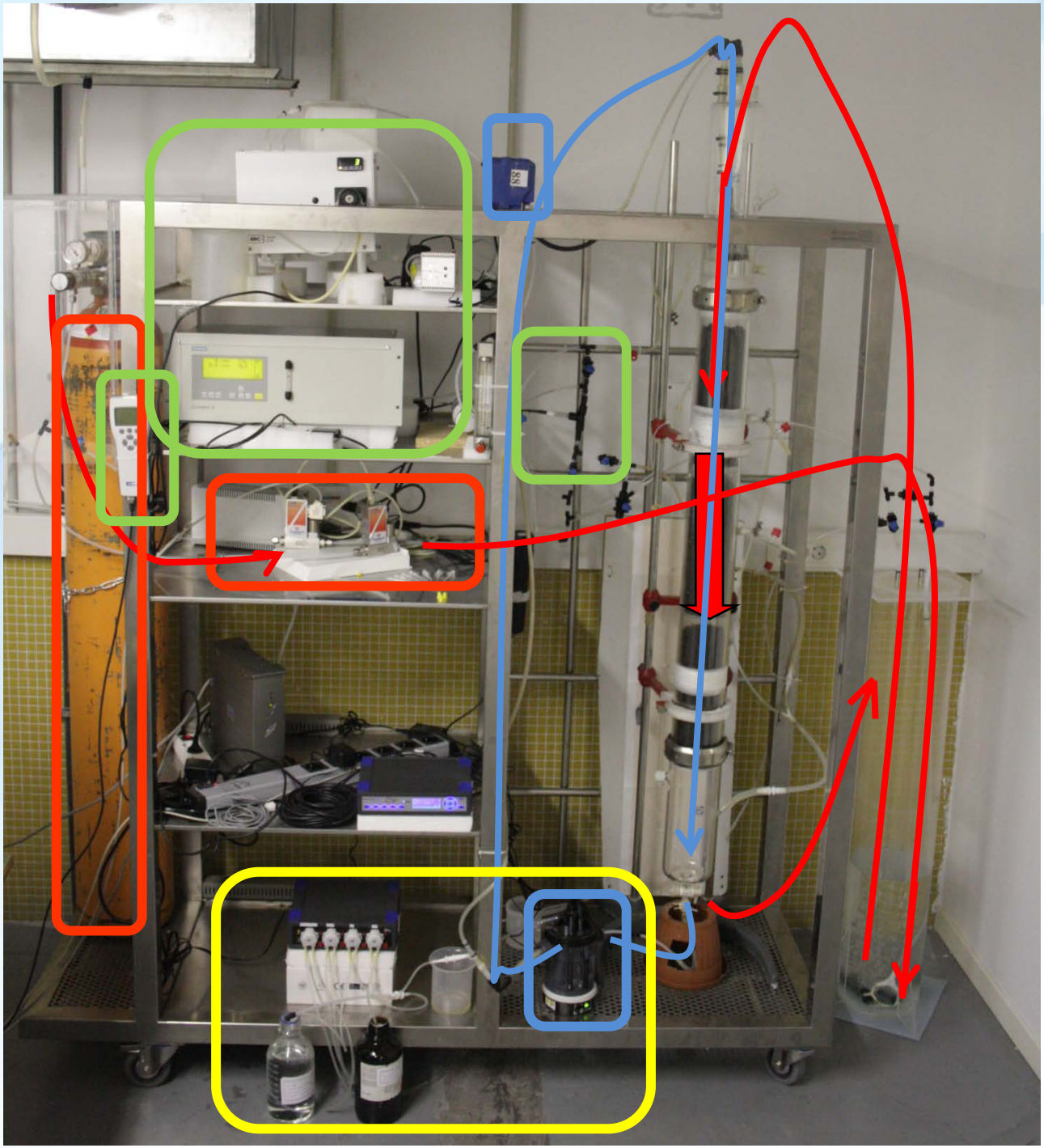


Gas phase circulation

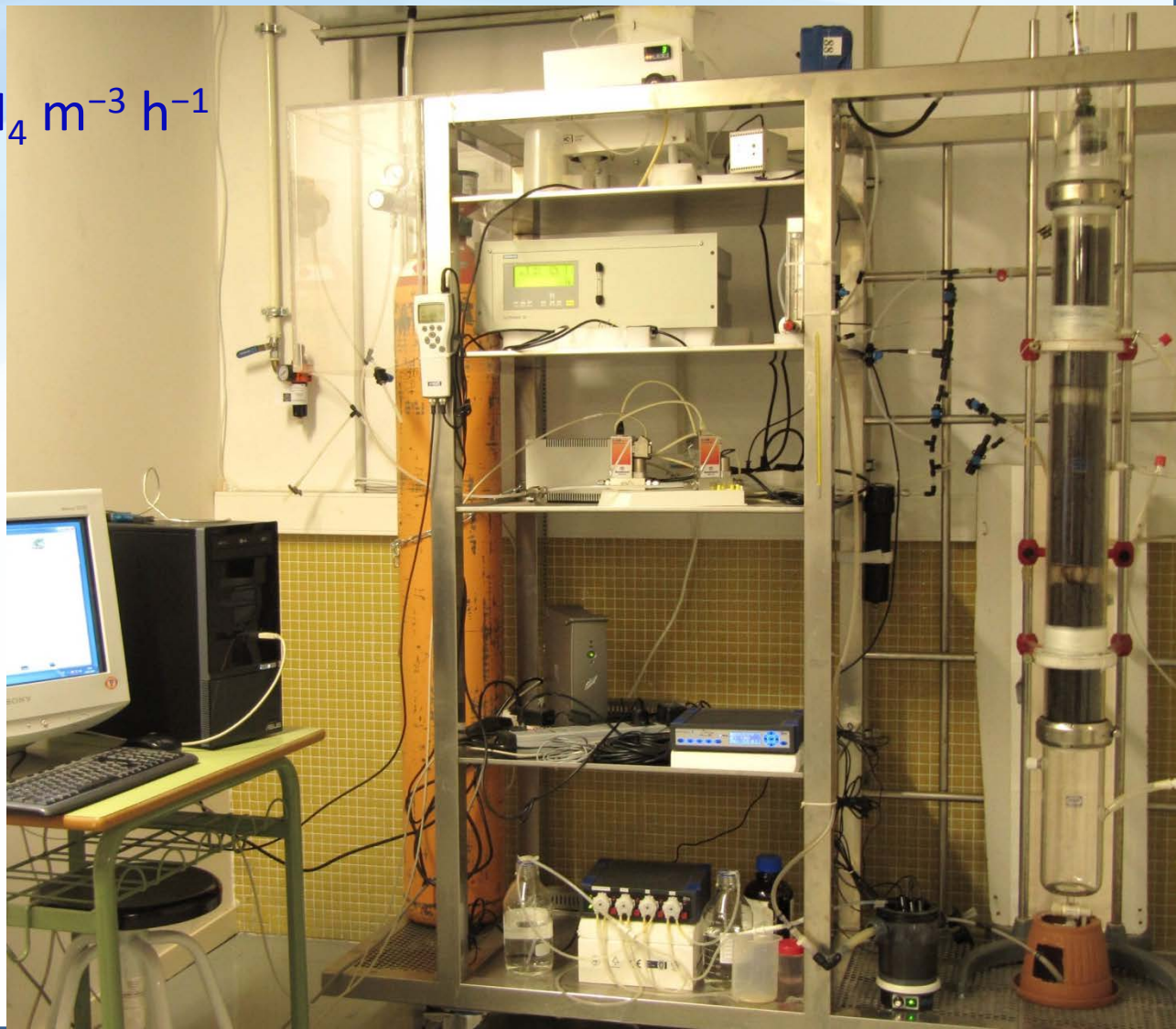
Liquid phase circulation

Liquid phase control and monitorization (pH, conductivity, temperature, etc…)

Gas phase measurements (CH4, CO2, HR and temperature)

Biofilter conditions of operation

- **$CH_{4,inlet}$=** 0.2 - 1 %
- **IL_{CH4} =** 24 – 74 g CH_4 m^{-3} h^{-1}
- **EBRT** = 4 - 8 min
- **pH** = 6.5-7.0
- 6 months



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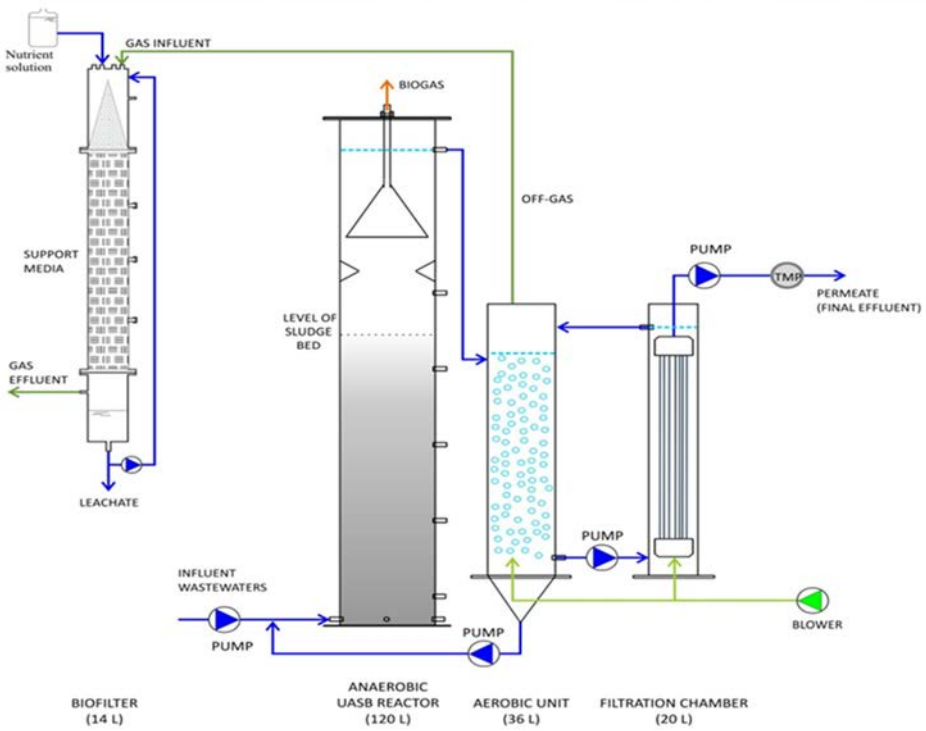
Analytical methods

- Gas phase
  - Methane (on-line, off-line)
  - Carbon dioxide (on-line)
  - Temperature
  - Relative humidity
  - Pressure drop
- Liquid phase
  - N-species (NH_4^+, NO_2^-, NO_3^-)
  - pH
  - Conductivity
  - Temperature
  - SSV
  - Exopolymeric substances

Start-up

- Packing material
  - Volume: 4.5 L
  - Open pore polyurethane foams cubes
  - 75 – 90 % porosity
  - 2.8 cm^3 cube size
  - 3 independent sections
- Inoculation
  - 4.5 L of sludge
  - Aerobic chamber of a three stage Anaerobic Hybrid Membrane Biological Reactor



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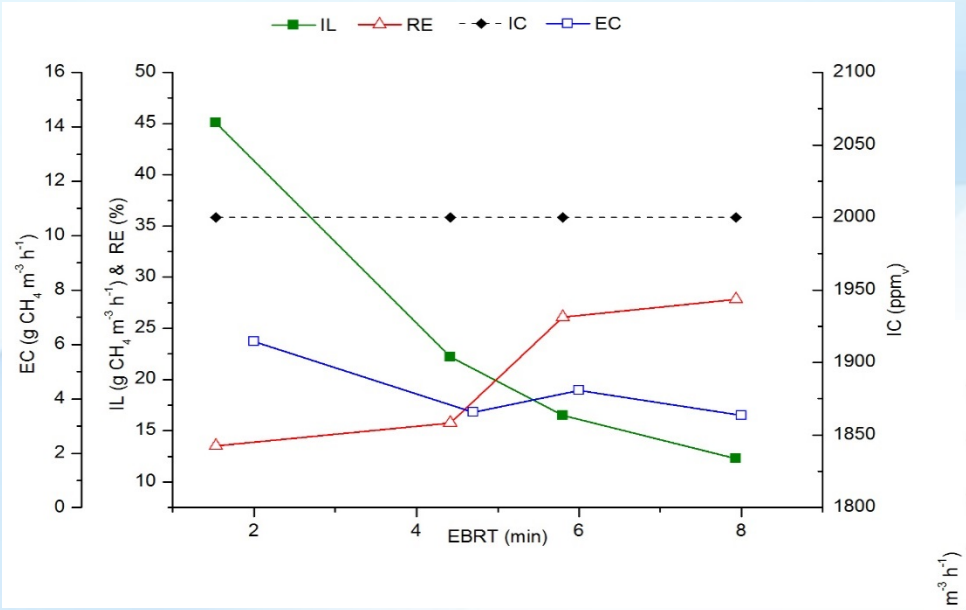
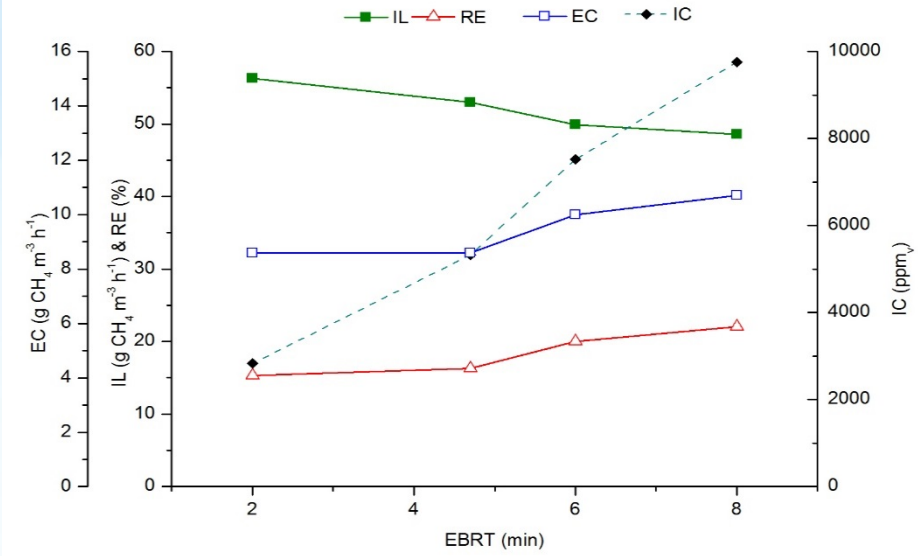
Periods of operation

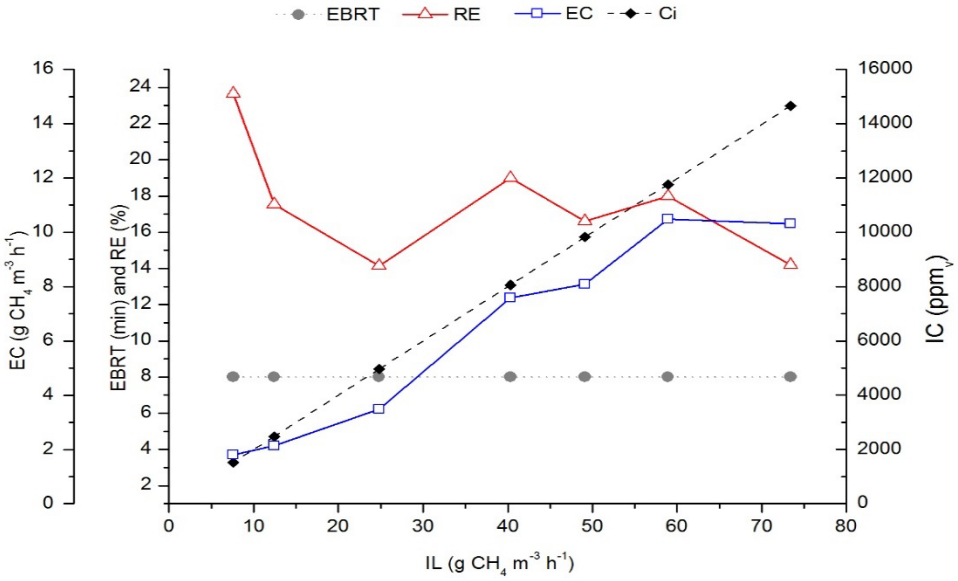
- Strategies for start-up and biomass colonisation (Period 1)
  - biomass development in BTF systems is a crucial stage
  - start-up of 15 days (Period P1a),
  - IL doubled from 23.5 to 50 g CH_4 m^{-3} h^{-1} (IC up to 0.99 % vv^{-1})
  - Correlation between N in the nutrient solution and REs and ECs
- Short-term optimisation tests (Period 2)
  - Water addition
  - Influence of the EBRT
  - Effect of the inlet load
- Final steady-state operation (P3)
  - IC of 0.5% and finally 0.2%
  - IL of 50 g CH_4 m^{-3} h^{-1} led to the achievement of maximum REs of 25.6%, corresponding to ECs of 14.7 g CH_4 m^{-3} h^{-1}.

Operation Days	*15*	*8*	*8*	*12*	*28*	*28*	*20*	*16*	*59*	*10*
Pressure drop (mm)	6	6	4	4	4	4	4	4	4	4
EBRT (min)	4	8	8	8	8	8	2.0 - 8.0	8	4	4
T inlet flow (ºC)	22.4	21.7	21.7	21.7	21.7	23.35	23.35	23.35	23.4	23.4
T outlet flow (ºC)	22	21.3	21.3	21.3	21.3	23.23	23.23	23.23	23.2	23.2
Gas Flow (L h^{-1})	74	34	34	34	34	34	33 - 135	34	67.5	67.5
Inlet Conc. (ppm)	2140	9900	9785	9800	9550	9763	2000 - 10000	1500 - 14600	5000	2000
Inlet Conc. (%)	0.2	1	1	1	1	1.5	0.2 - 1	0.15 - 1.46	0.5	0.2
Inlet Load (g m-3 h-1)	23.5	49.95	49.37	49.44	48.18	49.26	23.5 - 50	7.57 - 73.66	50.08	20.03
RE range (%)	0 - 16.5%	10 - 27 %	0 - 25 %	18 - 25 %	20 - 23 %	19 - 25 %	13 - 28	14 - 24	12- 25.6	7.1 - 29
RE av. (%)	9±4.0	19,3 ± 3,1	18.0± 8	21± 4.0	23± 3.0	20.0± 3.0	18.7± 5.5	18± 3.0	16.2± 3.8%	22.2± 8
EC range (g m-3 h-1)	0 - 3.91	5.0 - 14.0	3.0 - 12	9.0-11	10.0-11.0	8.9 - 12.61	3.4 - 10.72	1.8 – 10.49	7.4 - 14.7	1.0 - 7.0
EC av. (g m-3 h-1)	2.0± 1.3	9.23 ± 3.1	8.39± 2.99	9.9± 1.6	10.89± 3.9	9.47± 1.7		8.05± 3.7	8.88± 2.07	4.69± 2.4

Short-term assays



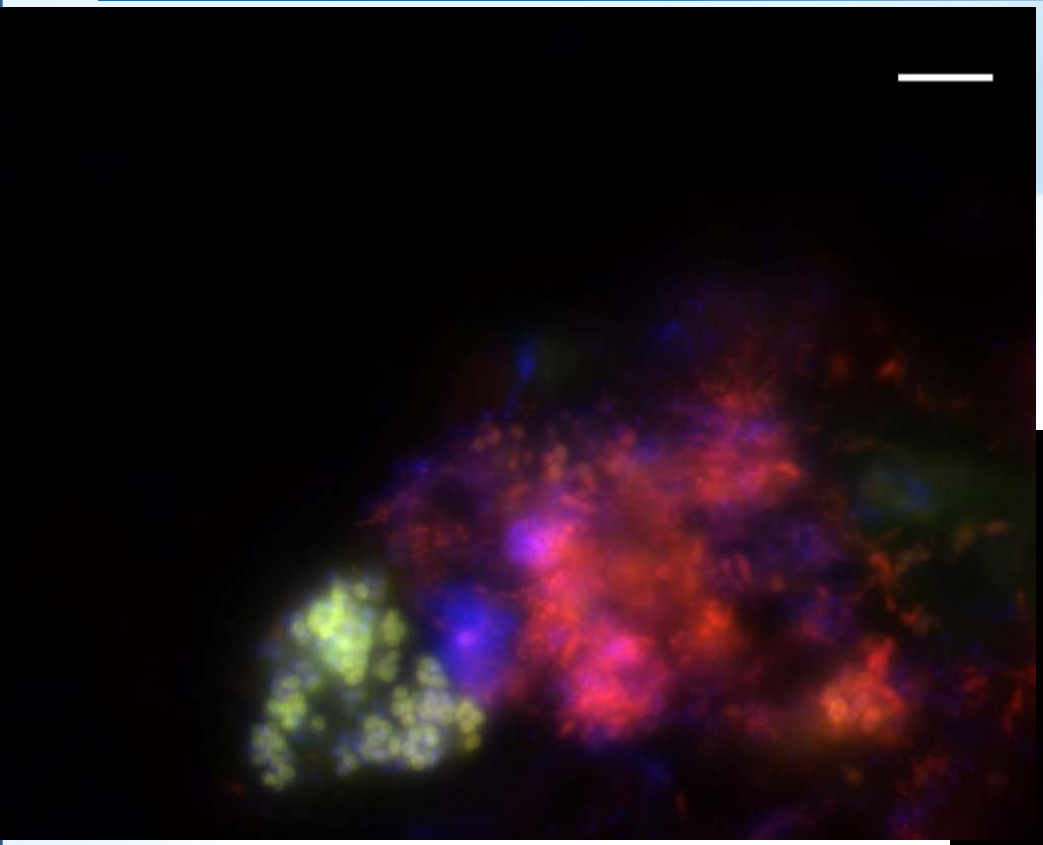


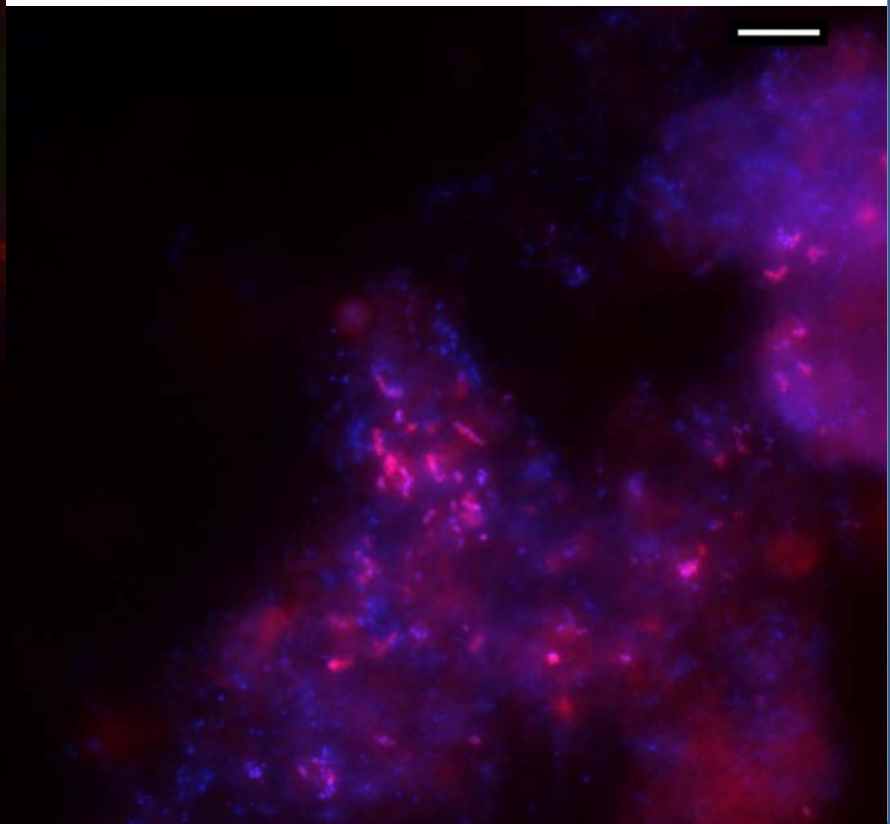


Microbiology

- Characterization of the inoculum
  - Illumina technology
  - high diversity of microorganisms since around 1200 genera and more than 800 species
  - Methanotrophs main families: *Protobacteria* phylum being *Gammaproteobacteria* (Methanotrophs Type I) and *Alphaprotobacteria* the dominant classes
  - 2.1 % of the microorganisms belong to the genera identified as methanotrophs
- Biomass developed in the BTF
  - Identify the presence of selected methanotrophs after 2 months
  - Identification of Type I methanotrophs and archaea, as well as the limited presence of Type II methanotrophs

Microbiology





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Conclusions

- A biotrickling filter packed with polyurethane foam was used to treat diffuse methane emissions (0.2-1% vv^{-1}) at long term operation (approximately 6 months) under stable conditions
- Removal efficiencies were always limited below 30% and maximum removal rates achieved values up to 14.7 g CH_4 m^{-3} h^{-1}
- Water addition necessary but should be optimised
- EBRT is the key factor to maximize methane abatement, with better results in the range of 6-8 minutes

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