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BIOTECHNOLOGICAL REMEDIATION STRATEGIES FOR MARINE SEDIMENTS HIGHLY CONTAMINATED WITH PAHs

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SEDIMENT, DIFFUSE POLLUTION AND SUSTAINABLE DEVELOPMENT GOALS

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Introduction

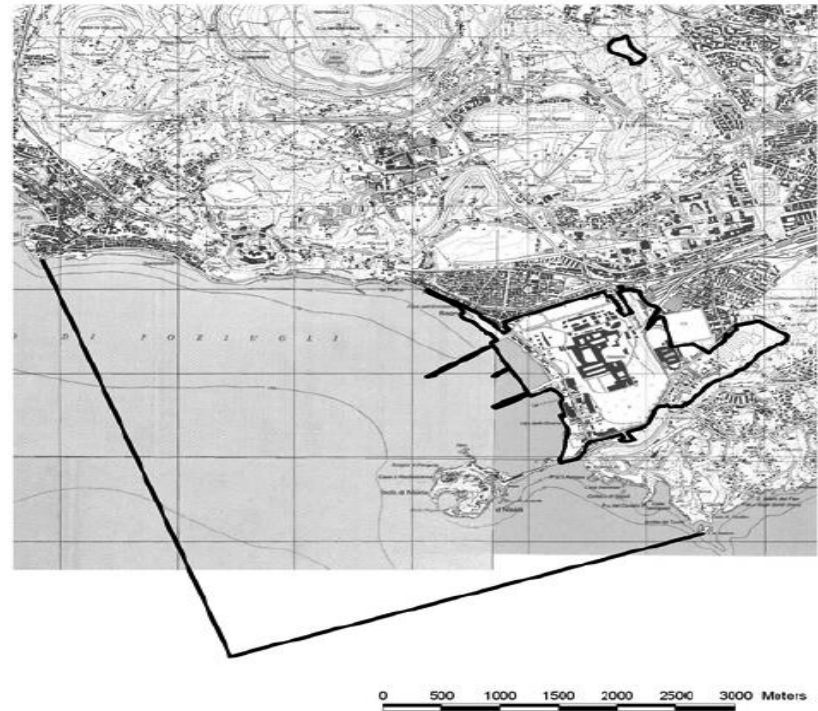


- Hydrocarbon pollution is widespread in marine environments as a result of spills from refining facilities and oil tankers, direct discharge from effluents and runoff from terrestrial sources

- Aquatic sediments represent a major sink of hydrocarbons
- High concentrations of hydrocarbons have been reported in several coastal urban areas worldwide



PAH contamination in the Bagnoli Site



The sediments of the Bagnoli site (Naples, Italy) in the Southern Tyrrhenian Sea is characterised by high concentrations of PAH and toxic metals (Romano et al. 2004).

PAH concentrations up to 2-3 orders of magnitude higher than those generally found on a global scale (Arienzo et al. 2017).

Conventional vs Biological Remediation

Conventional Remediation Strategies

Dredging

- Landfilling
- Thermal Treatment
- Open water Placement
- Confined Disposal

- In situ Capping
- Addition of Dispersants

Bioremediation Strategies

Biostimulation

- Nutrient Ammendment
- Biosurfactants
- Addition of Oxygen

Bioaugmentation

- Addition of Microbial degrading strains

Bioelectrochemical Systems

- Microbial fuel cell
- Oil spill Snorkel

Bioremediation approaches are more sustainable than the conventional remediation options which are characterized by high costs and greater risks of secondary pollution (Akcil et al. 2015).

Bioremediation and Metal Mobilisation

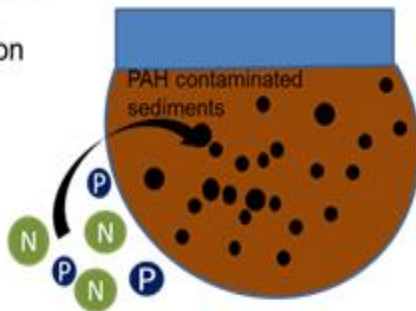


Bioremediation can influence metal mobility in sediments by altering the metal distribution in the various geochemical fractions

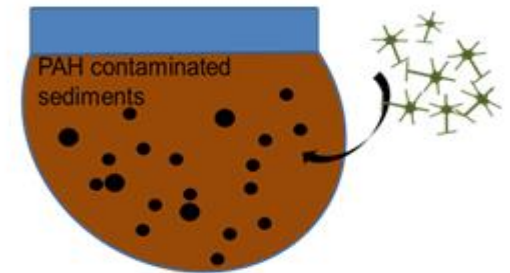
Objective

- Assessment of different bioremediation approaches on the sediments from the Bagnoli industrial area for enhanced PAH degradation.
- Evaluate the impact of bioremediation processes on metal mobility in sediments
- The bioremediation approaches investigated for the treatment of the contaminated sediments were:

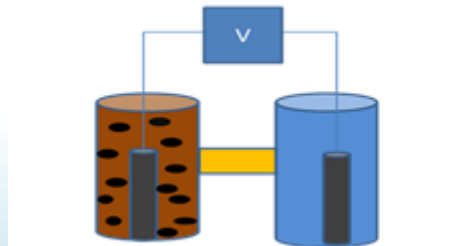
1. Biostimulation (N and P rich nutrient addition)



2. Bioaugmentation (Addition of *Aspergillus* strains solution)



3. Bioelectrochemical Stimulation



Bioremediation Experiments

Bagnoli sediment contamination

- Contaminant concentrations in the sediments were compared to the regulatory limits as prescribed by the Italian Decree number 173 of July 15, 2016.

Contaminant	Initial concentrations (mg/kg)	Italian Regulatory Limits (mg/kg)
As	210 ± 10	20
Hg	1.2 ± 0.2	0.8
Pb	820 ± 90	70
Mn	1900 ± 100	
V	120 ± 10	
Cd	8.8 ± 0.3	0,8
Cu	130 ± 40	52
∑ 16 priority PAH	130	4

Initial PAH and Metal Concentrations in Bagnoli Sediments

Materials and Methods: Biostimulation Experiment



- Addition of $(\text{NH}_4)_2\text{SO}_4$ and K_2HPO_4 (C: N: P ratio of 100:10:1).
- Operative conditions (O.C.): 28 days, 200 g/L of sediments.
- Control experiment was without nutrients

Materials and Methods: Bioaugmentation

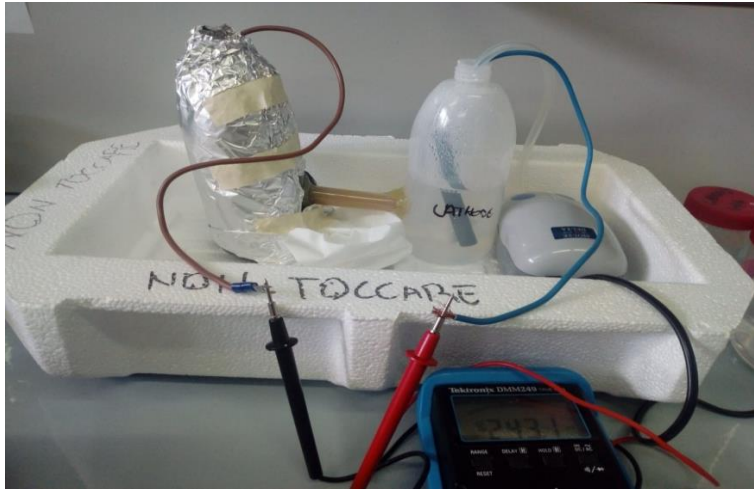


- Addition of *Aspergillus sp.*
- O.C.: 28 days, 200 g/L of sediments.

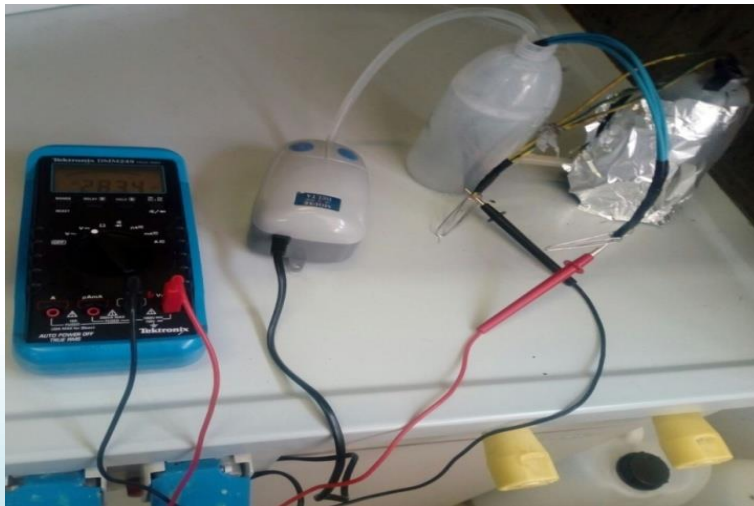


- Control experiment had a Glucose rich medium (GRM) to stimulate autochthonous microbial growth

Materials and Methods: Bioelectrochemical System

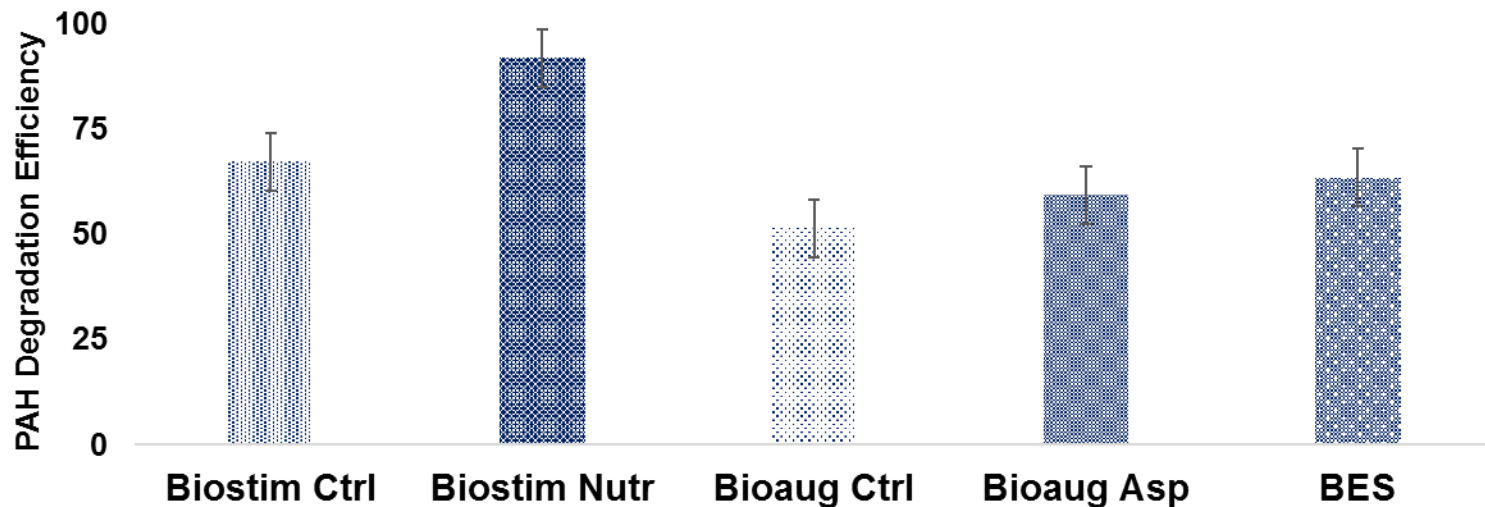


- Insertion of graphite rod to stimulate PAH oxidation.
- O.C.: 45 days, 2500 g/L of sediments.



Results : PAH Degradation

Biostimulation with the inorganic nutrient addition was the most efficient approach with over 85% PAH degradation rates obtained against around 60% with the other biotech approaches tested.



PAH Degradation Efficiency obtained in the various Bioremediation Experiments

BES and bioaugmentation efficiencies could be further enhanced by using different configurations and/or electrode materials and by the use of autochthonous strains

Results: Predictive Model

- A predictive model was developed to study the PAH degradation rate with respect to microbial growth rate and the variation in the control and nutrient amended experiments.

$$\frac{dN}{dt} = kN(1 - \beta N) + k_0 \int_0^t N_{(t)} dt \quad (1)$$

$$\frac{dC}{dt} = \frac{1}{Y} \frac{dN}{dt} \quad (2)$$

With N the bacteria number (10⁹ cells/g dry sediment)

C the PAH concentration (mg/kg)

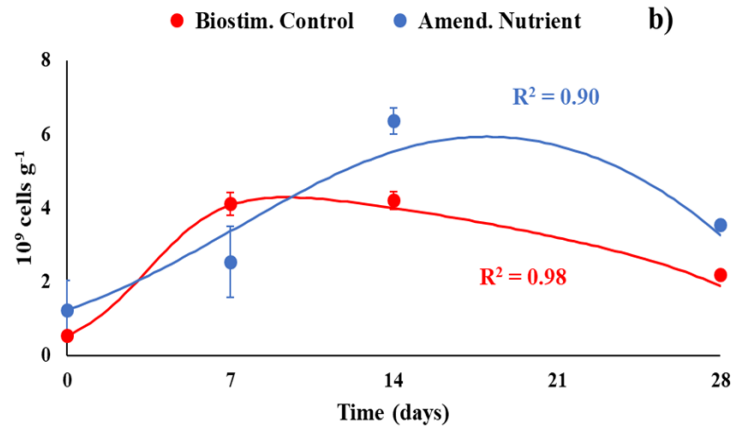
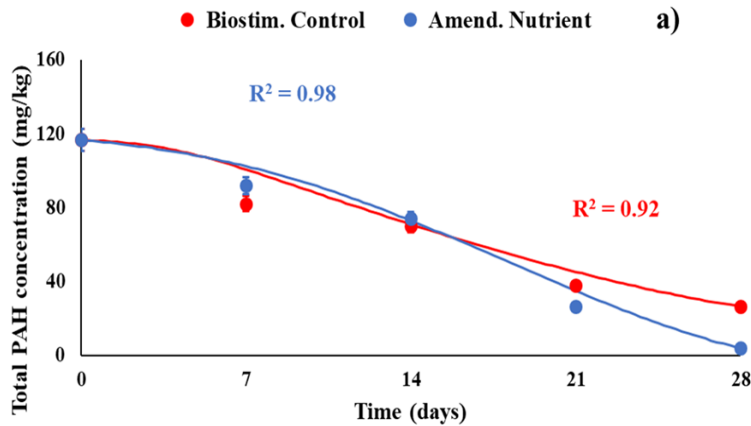
k and β bacteria growth and inhibition factor resp.

k₀, death constant

Y degradation efficiency.

- The experimental data fitted well in Equations 1 and 2 of the model

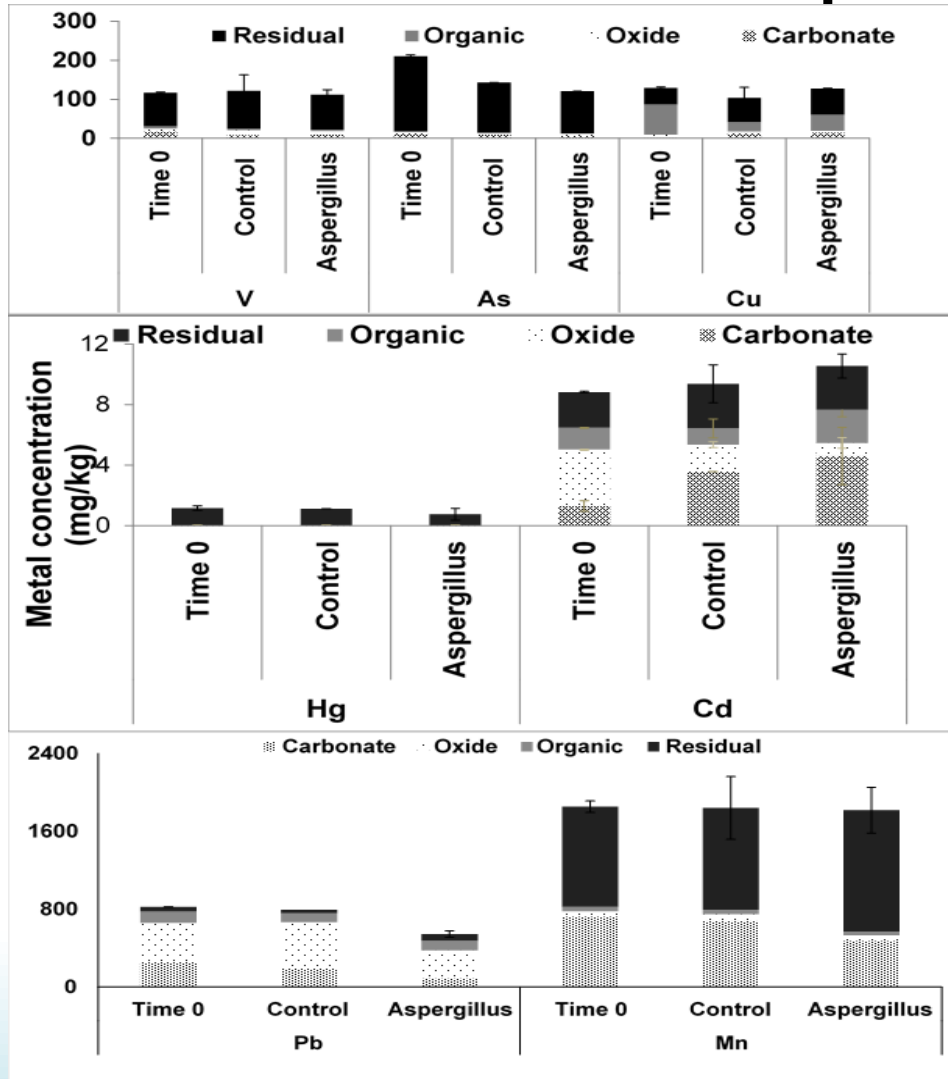
Results: Predictive Model



Temporal changes of the total PAH concentration (a) and the prokaryotic abundances (b) during bioremediation experiments.

- The four parameters k , k_0 , β and Y were estimated through a nonlinear regression analysis
- Data from the model were similar to the experimental data
- Also a direct correlation was observed between bacteria growth and PAH degradation

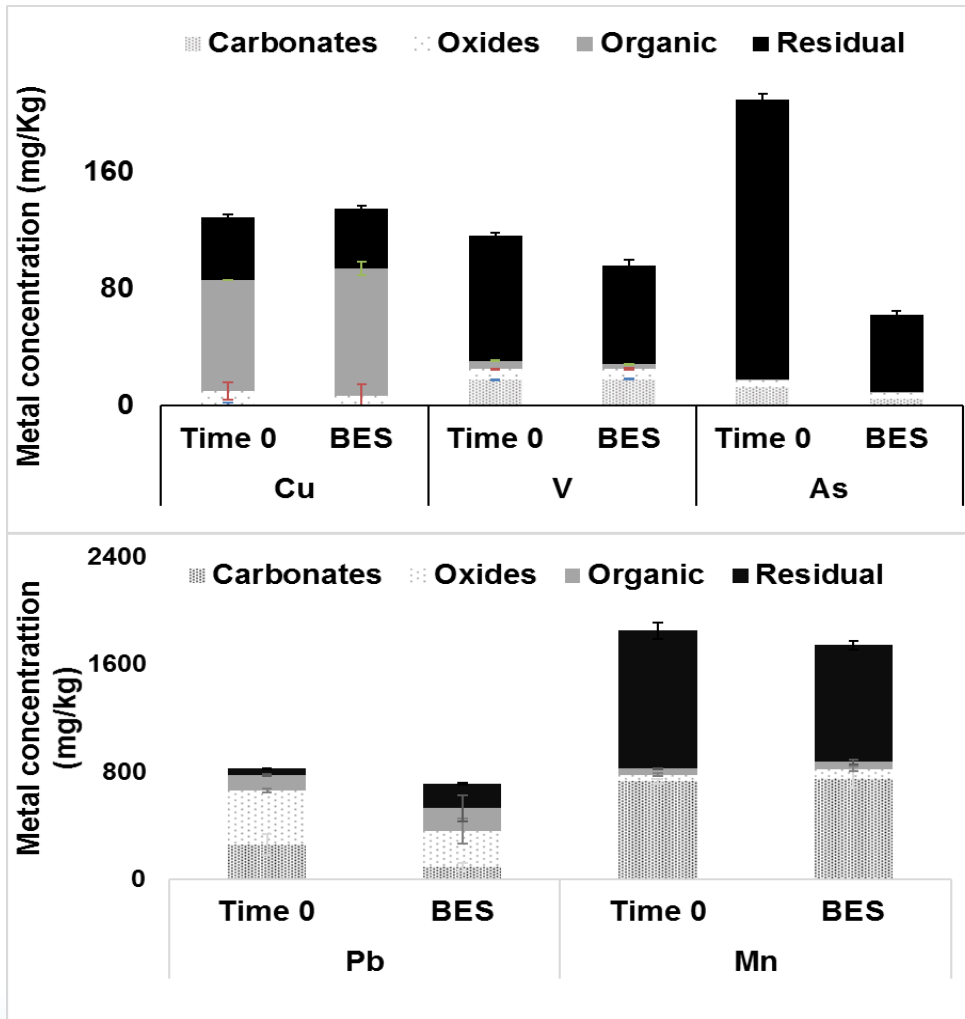
Results : Metal Mobilisation in Bioaugmentation Experiments



- Pb and As were mobilized by approximately 35% and 45%, respectively
- Cd showed a significant increase in the carbonate phase (2.5-fold), combined with a decrease in the oxides (around 80%).

Metal partitioning in the bioaugmentation experiments; Pb, Mn (a), Hg, Cd (b), V, As and Cu (c).

Results: Metal Mobilisation in BES



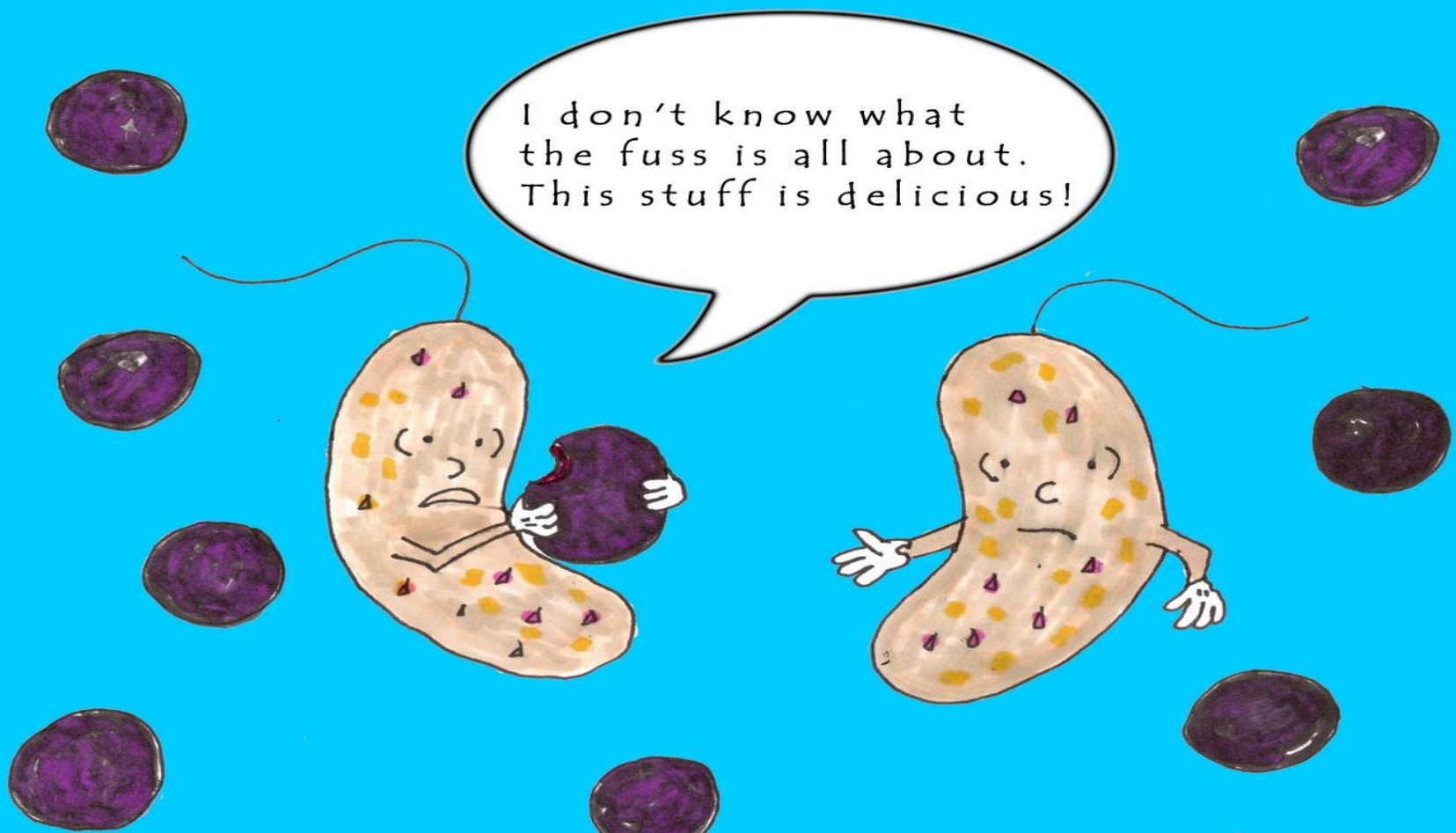
➤ Similarly, the BES system increases the mobility of Pb and As.

➤ As mobilised by 70% and Pb by 15%

(a) Lead and Manganese (b) Cu, As and V
Partitioning in the Sediments treated in MFC systems

Conclusions

- Biostimulation with inorganic nutrients was the most efficient method.
- A direct correlation was observed between PAH degradation and microbial growth rates
- A semi empirical model can help predict the time required to achieve a specific level of degradation during a bioremediation project
- Bioremediation can induce mobilisation of metals and this should be carefully taken into account especially if bio-treatments are applied in situ



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