Electricity Driven Low Energy and Chemical Input Technology for Accelerated Bioremediation

Dear readers,

we are happy to present you the first issue of our ELECTRA newsletter.

ELECTRA is a flagship project of the EU-China Cooperation Initiative. It is a 4-year Research and Innovation Action. The consortium consists of 16 European partners receiving funding from the European Union H2020 programme, and 5 Chinese partners being funded by the National Natural Science Foundation of China (NSFC).

We work closely together on the development and implementation of bio-electrochemical systems for the bioremediation of contaminated wastewater, groundwater, sediments and soil. In this first newsletter, we are glad to present a first set of four technologies of the ELECTRA project as well as a brief summary of an impressive number of communication and dissemination activities performed within this first 18 months of the project.

The Coordination Team
Philippe Corvini
Korneel Rabaey
Shuang-Jiang Liu

EU – China Collaboration

Despite difficulties imposed by the COVID-19 pandemic, the European and Chinese partners of ELECTRA were able to set multiple scientific and technological collaborations, which have materialised in eleven joint publications and an oral communication. Most importantly, scientific missions of young Chinese researchers in the labs of European partners and reciprocally already took place. Three Chinese students visited European labs for research stays and six European students attended one highlight event in China, namely workshop for European and Chinese BSc., MSc., PhD and post-docs working in the ELECTRA project.

Philippe Corvini
Shuang-Jiang Liu

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Sino-European Environmental Biotechnology Cooperation Research Project Holds Academic Exchange for Young Scientists

Article source: Liu Shuang Jiang's group Published time: 2019.12.02

On November 28, 2019, the academic exchange activities of young scientific and technological investigators in the Sino-European Environmental Biotechnology Cooperation Project funded by the National Natural Science Foundation of China (NSFC) and the European Commission (EC) took place at our institute. European project leader Professor Philippe Corvini, Professor Korneel Rabaey, and Chinese project leader Professor Liu Shuang Jiang jointly organized and participated in the event.

Researcher Liu Shuang Jiang introduced the main research directions and contents of the Center for Environmental Microbiology and Biotechnology, and exchanged information on the implementation and progress of the ELECTRA project. Later, Professor Ou Fang and young scientific and technical researchers visited the Microbial Resources and Big Data Center. Director Ma Juncai introduced the big data information platform resource acquisition process, a resource sharing process and a microbiological information query process. In addition, the exchange group visited the strain collection center of the Institute of Microbiology, Chinese Academy of Sciences, led by Mr. Cai Man. Mr. Cai Man patiently answered all the questions posed by the European young scientists and technicians about the preservation process of the Institute strains. Finally, the exchange group visited the Herbarium, and Wang Ke introduced the history and current state of fungi research in China.

Young science and technology investigators play a vital role and key role in scientific research projects. The academic exchange activities between China and Europe young science and technology workers are an important form of promoting China-EU cooperation. The successful holding of this exchange event will further strengthen and promote the implementation and development of China-EU cooperation projects.

Some highlights from the development of ELECTRA technologies

Groundwater denitrification by microbial electrochemical technology (MET)

Groundwater is an important drinking water source for the majority population in the world. In the last decades, increase of nitrate-contaminated groundwater has become a major issue along with intensive agriculture and livestock production. Compared to the mostly used nitrate removal technologies, such as microbial reduction using methanol, ion exchange, reverse osmosis and reverse electrodialysis, bioelectrochemical denitrification has become a competitive alternative with several advantages: 1. low energy cost; 2. easy to monitor and control for long term operation; 3. no chemical (*e.g.,* organic content) addition and no waste generation.

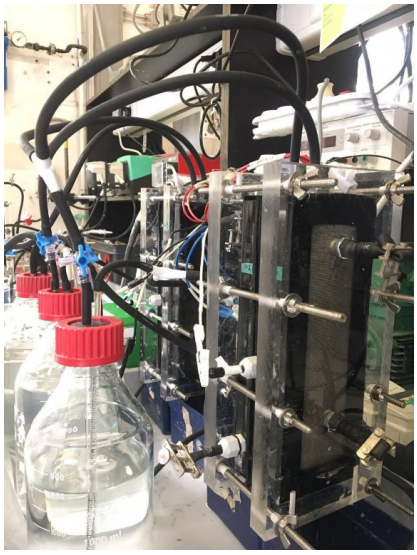

Figure 1: Picture of lab-scale MECs for groundwater denitrification

At Ghent University, two lab-scale bioelectrochemical systems (BESs) are successfully operated for nitrate removal in synthetic groundwater (Figure 1). As the anode potential is higher than the cathode potential, they are operated as so-called microbial electrolysis cells (MEC). Each MEC consisted of two identical Perspex frames and plates, and separated by a cation exchange membrane (CEM) with a working area of 100 cm^2. The anode was a dimensionally stable iridium mixed metal oxide (Ir MMO) coated titanium-electrode with a projected surface area of 100 cm^2. A stainless-steel wire mesh of the same exposed working area served as the current collector in the cathode. The cathode chamber was filled with granular graphite. The working volumes of the anode and cathode chambers were 0.2 L and 0.15 L, respectively. A reference electrode (Ag/AgCl (3 M KCl), + 0.210 V vs. SHE at 25 °C) was placed in the cathode chamber of each cell. A plastic spacer was used in each chamber to prevent the contact between the surface of the electrodes and the membrane. The cathode chambers of both reactors were fed under continuous-flow mode while the anolyte recirculated in a closed loop (Figure 2). The MECs were controlled by a potentiostat.

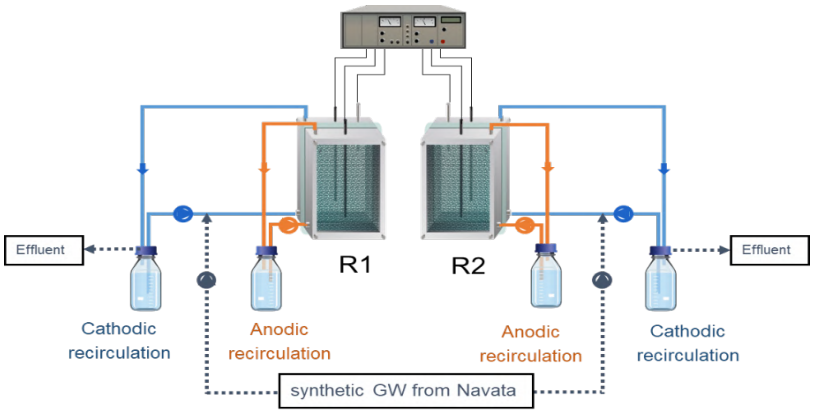

Figure2: Graphical representation of MECs

One MEC (R1) was operated under continuous polarization mode with cathode potential at -0.333 V vs. Ag/AgCl. The second MEC (R2) was operated under periodic polarisation mode with 30 s polarisation at the same cathode potential and 30 s open circuit (OCP). 1.1 L nitrate-contaminated (33 ppm nitrate-N) groundwater was fed to each cell per day and less than 5 ppm nitrate-N remained in the effluent. The denitrification rate was around 0.9 kg $NO_3/m^3_{cc}.d$ (Cathodic Compartment) under both polarisation modes.

In the next experiment, lab-scale fluidised-bed MECs (Figure 3) are constructed using the same material as described for the fixed-bed MECs. The fluidised particles will bring a more homogenous electron and substrate mixing and less scaling problems. A preliminary test showed that the denitrification rate achieved was higher than 1 kg $NO_3/m^3_{cc}.d$ under continuous polarization mode in fluidized-bed MECs. We aim to bring this technology to pilot scale.

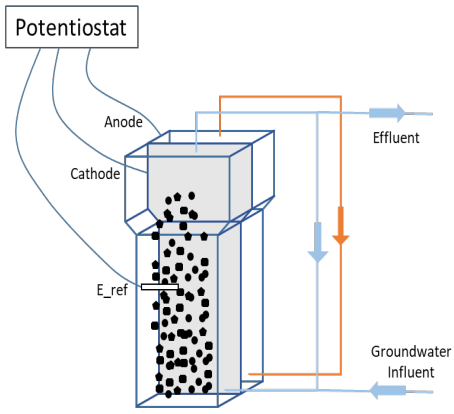

Figure 3: Schematic of a fluidized-bed MEC

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All-in-one: removal of oxidisable and reducible contaminants from contaminated groundwater in a single bio-electrochemical reactor

In recent years, microbial electrochemical technologies (MET) have attracted considerable attention for soil, sediment, and groundwater remediation. MET are anaerobic systems in which microorganisms catalyse oxidation or reduction reactions using solid-state electrodes which, upon deployment within the contaminated matrix, act as virtually inexhaustible metabolic electron acceptors or donors, respectively. A key advantage of MET, compared to state-of-the-art (bio)remediation technologies, is that the removal of contaminants by naturally occurring microbial communities can be enhanced with no need for adding expensive chemicals into the subsurface, with only modest energy consumption, and can be fine-tuned and continuously monitored using relatively simple electrochemical means [1-2]. To date, MET have shown great promise, at least at the laboratory scale, for the treatment of a broad spectrum of subsurface contaminants, such as aliphatic and aromatic petroleum hydrocarbons (*e.g.,* alkanes, BTEX), chlorinated solvents (*e.g.,* TCE, 1,2-DCA), nitrate and sulfate [3]. In spite of their potential, however, field scale applications are still limited, primarily due to the lack of scalable and economically viable system configurations. Furthermore, MET application potential for the simultaneous treatment of complex mixtures of pollutants (*e.g.,* containing both oxidizable and reducible substances) often co-existing in the environment, has been so far only marginally verified.

Recently, IRSA-CNR has developed a novel MET configuration, which addresses most of the above-mentioned limitations. This novel electrobioremediation technology, named 'the bioelectric well', can be easily integrated into existing groundwater well designs and bioremediation schemes [4]. Importantly, the "bioelectric well" has specific design and functional characteristics which make it highly attractive for field application (Figure 4). As an example, the system has a concentric geometry (*i.e.,* an inner stainless-steel cathode positioned in very close proximity of an outer graphite anode) which allows minimising voltage and energy losses. The anode and the cathode are not separated by a membrane or other expensive separators. Instead, they are hydraulically connected and

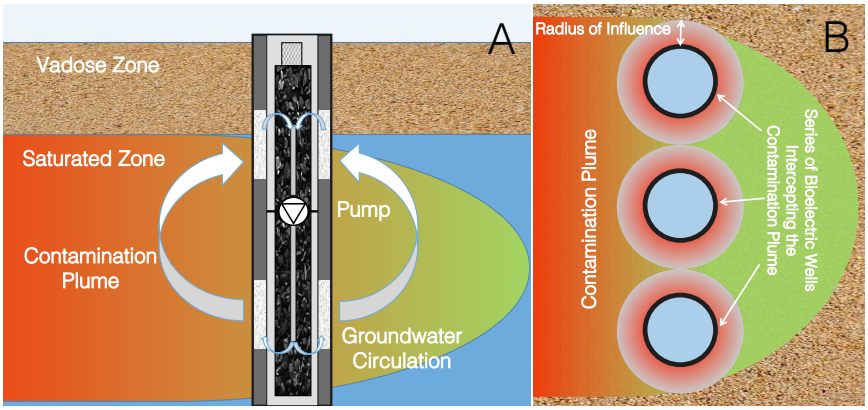

Figure 4: Cross-sectional (A) and plan (B) view of an in-situ groundwater bioremediation system based on the bioelectric well concept.

simultaneously exposed to the same reaction environment (*i.e.,* the contaminated groundwater), which makes it possible to drive concurrently oxidation (at the anode) and reduction (at the cathode) reactions, hence permitting the treatment of complex mixtures of pollutants. So far, a laboratory-scale prototype of such a system has been extensively and successfully employed to treat (synthetic) groundwater containing phenol or toluene.

IRSA-CNR is now facing the challenging task of employing the "bioelectric well" for the treatment of real groundwater originating from the Assemini site (Italy), which contains multiple harmful contaminants, namely BTEX, PAH, chlorinated solvents (primarily 1,2-DCA), and sulphate. A schematic drawing of the lab-scale "bioelectric well" prototype and of the possible oxidative and reductive biodegradation reactions it can drives is displayed in Figure 5.

Upon successful verification of the lab-scale performance of the system, an up-scaled prototype will be constructed, tested and optimised in the field under highly representative environmental conditions (*i.e.*, TRL 6).

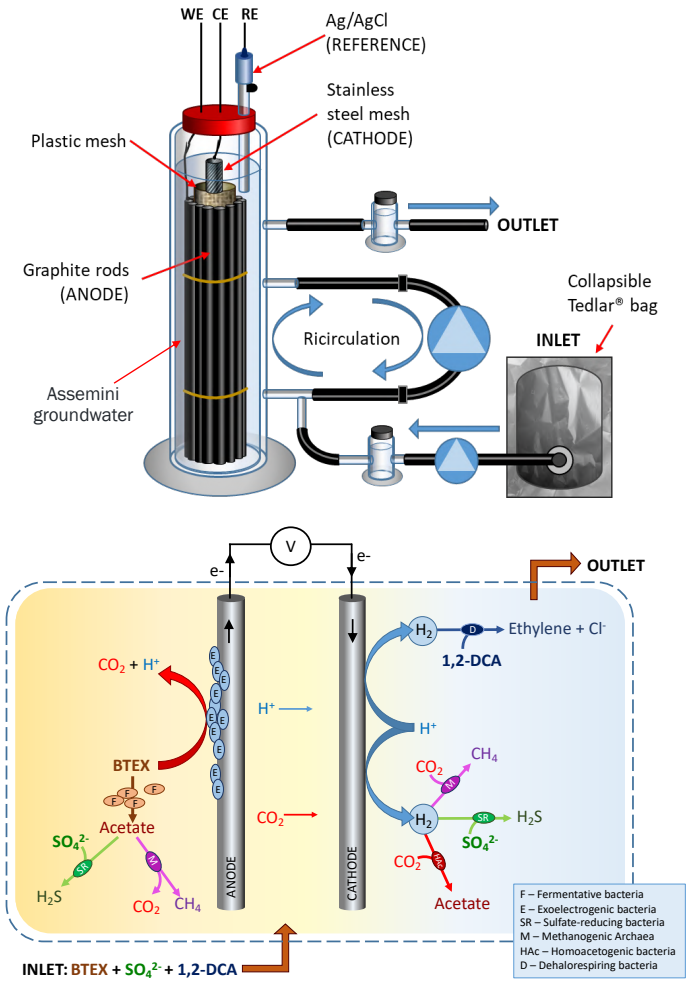

Figure 5: Schematic drawing of the lab-scale "bioelectric well" prototype (top) and of the involved oxidative and reductive biodegradation reactions (bottom). WE: working electrode; CE: counter electrode

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Bio-reactive barrier for in situ remediation using conductive nanomaterials

The microbial degradation of pollutants in the environment is often limited by the availability of electron acceptors. This can be improved by the direct interaction of microorganisms with a solid-state electron acceptor in the form of an electrode (anode). UDE developed an *in-situ* bioremediation process using conductive nanomaterials for the bioelectrochemical removal of contaminants, *e.g.,* polycyclic aromatic hydrocarbons (PAHs) by enhancing the anaerobic degradation.

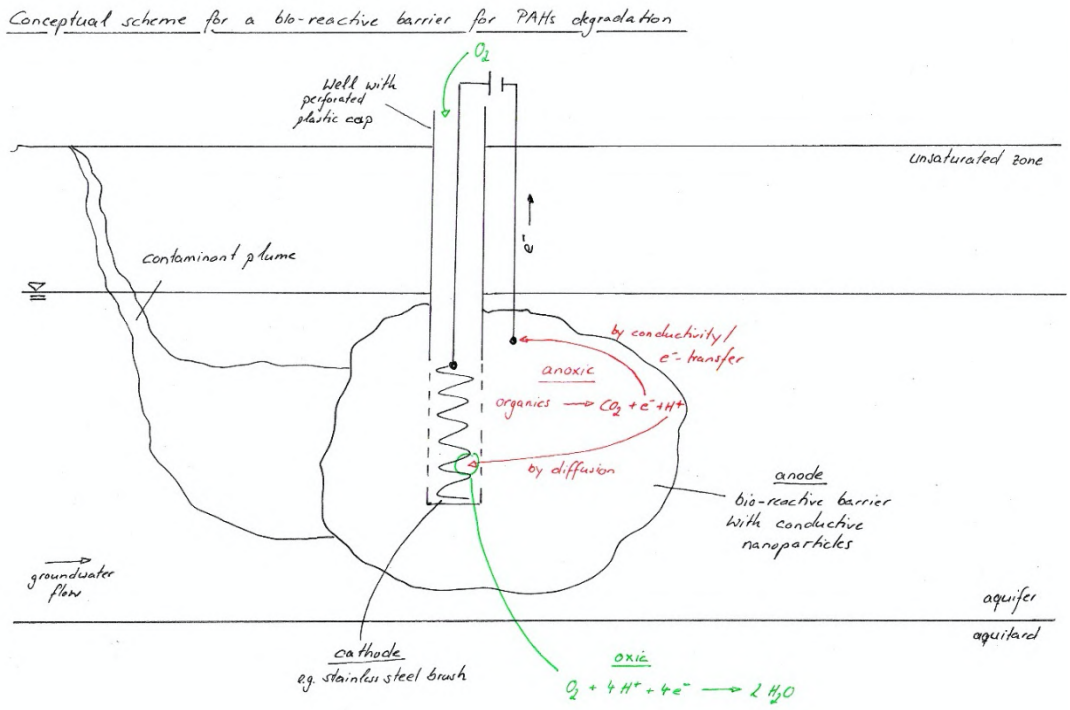

Figure 6: Conceptual scheme for a bio-reactive barrier for PAHs degradation.

To this end, the conductive nanoparticles are injected into sediments where they cover the matrix and form an electro-conductive network, which can work as an anode. UDE thus intends producing a bio-reactive barrier in the aquifer (see Figure 6). This increases the active electrode surface and the availability of electron acceptors. The technology should work in a minimal invasive manner and with minimal use of chemicals and energy.

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Field site study on groundwater remediation

In the context of the ELECTRA Project partners IEC and Sapienza University of Rome combined a Groundwater Circulation Well (IEG-GCW®) system and a reductive/oxidative bioelectrochemical sequential process for CAHs removal developed by University of Rome, La Sapienza, in a field test.

A former chemical facility located in the Rho district (Milano, Italy), was identified as the origin of a large chlorinated solvent plume. The highly contaminated source area was encapsulated by bulkheads in the 80s but it seems that there is still a leakage of pollutants from the isolated area to the deeper aquifer, caused by inhomogeneities in the underground. The vertical encapsulation system was set in a thin horizontal clay/sandy silt layer (about 1 – 2 m thick) which separates an upper shallow aquifer (5 to 10 m below ground) from a deeper one (12 to 40 m below ground). The groundwater samples from the shallow aquifer show high concentrations of trichloroethene (TCE) and *cis*-1,2-dichloroethene (*c*DCE) and some traces of vinyl chloride, in the deeper one the concentration levels decrease. It is planned to test a bioelectrochemical system of the University of Roma (Sapienza). For this purpose, IEG will install an IEG-GCW® system in the shallow aquifer (Figure 7). The GCW system consists of a lower screen section followed by a hydraulically inactive zone (packer) and an upper screen section. A submerged pump extracts the groundwater from the lower screen section and infiltrates in the upper screen section. The resulting hydraulic gradient generates a groundwater circulation from the top of the aquifer to its base. Pollutants in the circulation zone will be mobilised and flow to the well for treatment. In industrial remediation projects, the IEG-GCW® is typically combined with treatment units like vacuum stripping and/or adsorption, *etc.* In other cases, amendments will be infiltrated and distributed for reductive dehalogenation of solvents. At the Rho site, a certain amount of the circulating water will be

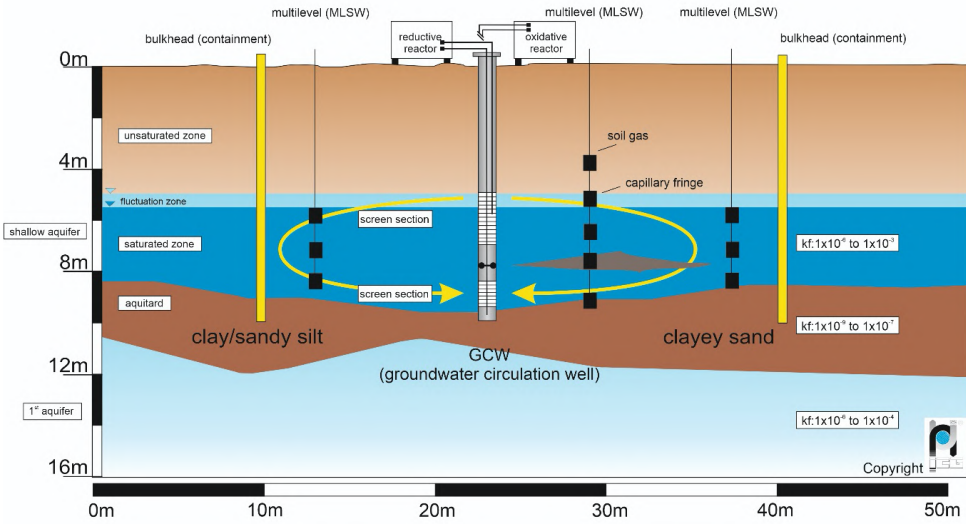

Figure 7: Schematic field installation layout

treated by an innovative reductive/oxidative bioelectrochemical sequential process under real remediation conditions.

The sequential bioelectrochemical process consisted of two separate tubular reactors which adopt a novel reactor configuration that avoids the use of an ion exchange membrane to separate the anodic and cathodic chamber (Figure 8). In the reductive reactor, a dechlorinating mixed inoculum received the reducing power to perform the reductive dechlorination of highly chlorinated aliphatic compounds by the cathode chamber while the less chlorinated daughter products (*c*DCE and VC) were removed in the oxidative reactor which supports an aerobic dechlorinating culture by *in-situ* electrochemical oxygen evolution performed by a mixed metal oxide anode. The separation of the reductive and the oxidative bioelectrochemical reaction allow the optimisation of the operating condition for each step such as the working potential and the hydraulic retention time.

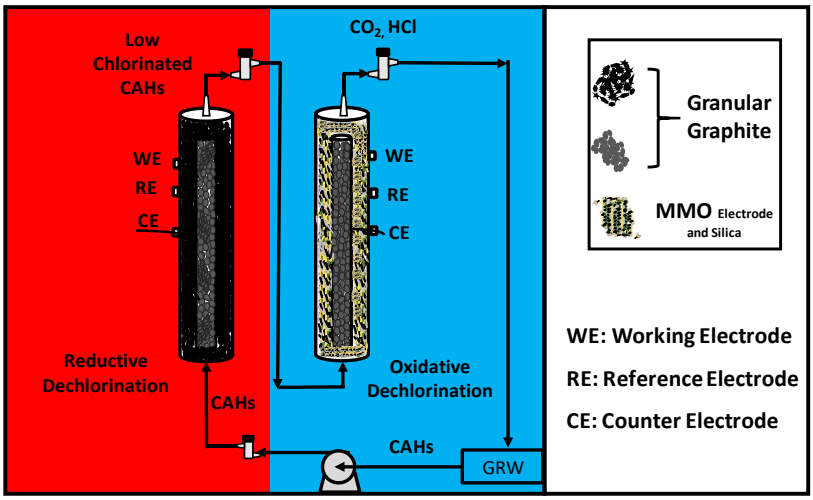

Figure 8: Schematic representation of the sequential reductive/oxidative bioelectrochemical process

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Scientific output

ELECTRA has disseminated its scientific achievements in a number of publications and conference presentations. For the complete list, refer to our website.

Year 2019

1. Yi-Xuan Wang, Wen-Qiang Li, Wen-Ming Zong, Tian-Yu Su, Yang Mu* (2019) Polyaniline-decorated honeycomb-like structured macroporous carbon composite as an anode modifier for enhanced bioelectricity generation. Science of The Total Environment. 696, DOI.
2. Álvaro Pun, Karina Boltes, Pedro Letón, Abraham Esteve-Núñez (2019), Detoxification of wastewater containing pharmaceuticals using horizontal flow bioelectrochemical filter, Bioresource Technology Reports, 7, DOI.
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23. Riccardo Perri, Boris A. Kolvenbach and Philippe F. X. Corvini (2020) Subsistence and complexity of antimicrobial resistance on a community-wide level. Environmental Microbiology 22(7), 2463–2468 DOI.
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Keynote and Plenary Conference Presentations

ELECTRA partners have been invited to and participated in several international conferences and gave a number of keynote and plenary lectures:

1. **KEYNOTE:** Puig, S. "Niches for Electro bioremediation of contaminated waters". International society for microbial electrochemistry and technology: International Meeting (ISMET 7). 7-10 October 2019. Okinawa, Japan.
2. **KEYNOTE:** Korneel Rabaey, "Scaled up, high rate production and extraction of volatile fatty acids from CO2 and electricity", International society for microbial electrochemistry and technology: International Meeting (ISMET 7). 7-10 October 2019. Okinawa, Japan
3. **KEYNOTE:** Abraham Esteve-Núñez. "METlands for wastewater treatment". Northwest China Water Environment Young Scholars Forum. Xi'an (China) 25 May 2019.
4. **KEYNOTE:** Philippe Corvini, "Bacteria feeding on antibiotics – eating the poisonous". 2nd International Meeting on New Strategies in Bioremediation Processes- BIOREMID 2019, Porto, Portugal (24-25 Nov 2019).
5. **KEYNOTE:** Federico Aulenta (2019) "Novel electrobioremediation strategies for cleaning up sediments contaminated by petroleum hydrocarbons". 2nd International Meeting on New Strategies in Bioremediation Processes- BIOREMID 2019, Porto, Portugal (24-25 Nov 2019).
6. **KEYNOTE:** Philippe Corvini, "Bacteria feeding on antibiotics – eating the poisonous". ISPTS-2019, Stevens Institute of Technology, Hoboken, NJ (Oct 25-28, 2019)
7. **PLENARY:** Jiandong Jiang. The IUPAC2019 GHENT (Crop Protection Chemistry), Advances in pesticides biodegradation and metabolism: Mechanisms, applications and regulatory issues. 19-24 May 2019, Ghent Belgium. Microbial catabolism of chemical pesticides: the mechanism and its potential application.
8. **KEYNOTE:** Puig, S. Niches for Electro bioremediation of contaminated waters. Conference 7th International Society of Microbial Electrochemistry and

Technology conference (ISMET7-2019). 7th-11th October 2019 in Okinawa (Japan).

9. **KEYNOTE:** Puig, S. Electro bioremediation of contaminated waters. Conference Technology Innovation for Sustainable Water. 3th-6th September 2019 in Daegu (Korea).
10. **PLENARY:** Nicolas Kalogerakis "Ecosystem and Engineering process applications of air nanobubbles", 5th International Conference on Chemical Engineering (ICCE-2020), 28-30 October 2020, Iasi (Romania).

News & Announcements

ELECTRA had arranged special sessions in AP-ISMET and EU-ISMET meetings, however, both were postponed from 2020 to 2021!

In addition, ISPTS-2021 organized by partners TU-Crete and Nanjing University in September 2021 will also host a special session on bioelectrochemical systems.

ELECTRA will also participate actively at the upcoming 11th International Conference on Environmental Engineering & Management (ICEEM-11) in Muttenz (Switzerland) organized by the coordinating partner FHNW also scheduled in September 2021.

Consortium

European partners funded by H2020

University of Applied Sciences and Arts Northwestern (Switzerland); Philippe Corvini (Coordinator)

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Technical University of Crete (Greece)

Universitat de Girona-LEQUIA (Spain)

Universiteit Gent - CMET (Belgium)

University of Rome La Sapienza (Italy)

National Research Council - Water Research Institute (Italy)

Helmholtz-Centre for Environmental Research (Germany)

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