FIFTEEN YEARS ELECTRO-RECLAMATION IN THE NETHERLANDS

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Prologue

The suggestion that electrochemical techniques and more in general the use of electricity could compete with traditional methods of land clean up seemed somewhat ludicrous during the nineteen-eighties. Especially in the Netherlands, where at that time soil remediation was synonymous with digging, dumping or incinerating contaminated soil and pumping and treating hundreds of thousands of cubic meters of mostly just lightly polluted groundwater. Those were the years of ex-situ ground processing and as a consequence of the Dutch clean-up policy of 'multifunctionality', (the multifunctional use of a site after remediation, meaning in practice unrealistic and extremely low clean-up values); there never really was a market for in-situ remediation.

At the end of the 1990's, however, the situation changed dramatically and in-situ remediation was much sought after and so were in-situ technologies. One of those is electroreclamation, but publications and symposia prolong the suggestion that it is still an emerging technology, or a development of recent years. Nothing is farther from the truth.

Historic background

Examination of the history of electrokinetics in soil will reveal it has its beginnings over 70 years ago. Some highlights are the work carried out by Casagrande[i] in stabilising clays by electroosmosis and his attempts to remove water from lime sludge in the 1930's and 1940's. In the 1950's Collopy[ii] patented the use of electromigration for reclaiming saline soils. Russian workers[iii] used electromigration in prospecting for metals in the 1960's. Co-workers at Battelle[iv] used electromigration of chloride ions into surface mounted membranes as a method of reducing corrosion of the reinforcing steel in salt contaminated concrete in the 1970's. The beginning of the 1980's marked the interest both in Europe[v] and in the USA[vi] of using the technology for the removal of toxic ions from the soil. Most of the early work and in particular the field experiments, however, were inconclusive due to the failure to manage the electrochemical changes in the soil around the electrodes, a neglect of ion exchange capacity of real soils compared to the kaolinite model systems used (and still being used!!) in the laboratory and the focus on electroosmosis[vii] as a possible means of removing pollutants from contaminated soil.

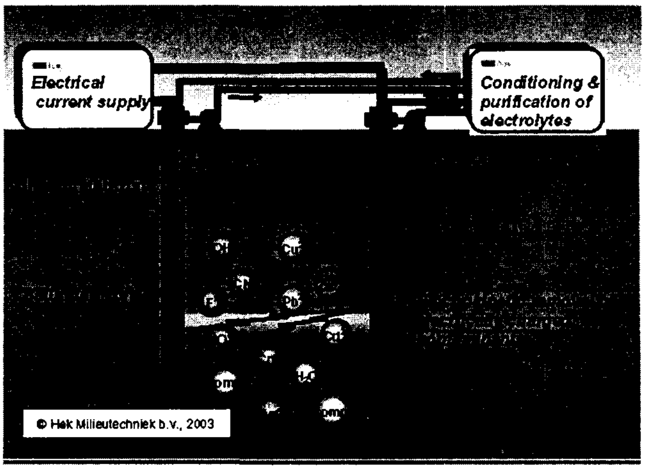

Figure 1. Schematic view of electroreclamation of ionic and/or polar contaminants using electrolyte management

The breakthrough came in 1987 with Lageman, Pool and Seffinga[viii] who focused on electromigration and patented the use of circulating electrolytes and the use of ion permeable wells to manage and control the anolyte and catholyte (figure 1).

Table 1. Results in-situ electroreclamation of inorganics at Loppersum

Soil type	Heavy clay	Average concentration at end	10 mg/kg
Depth of pollution	2 m	Arsenic removed	52 kg
Maximum concentration at beginning	500 mg/kg	Energy consumption	150 kWh/ton
Average concentration at beginning	115 mg/kg	Remediation period	80 days of 18 hrs.
Maximum concentration at end	29 mg/kg		

The first commercial and successful in-situ electroremediation project ever was carried out in 1988 by their company Geokinetics at the site of a former wood impregnation plant, which was heavily polluted with arsenic (table 1).

The results were presented at the NATO-CCMS conference in Copenhagen in May 1989 and at the Forum on Innovative Hazardous Waste Treatment Technologies in Atlanta in the same year. Worldwide interest in the technology has been rising ever since. In the US some commercial vendors have been trying to circumvent the patent of Geokinetics or just copied it. During the first half of the 1990's the Lasagna group[ix] from Monsanto, General Electric and Dupont pursued the use of electroosmosis as the driving and collecting mechanism for removing organic materials from contaminated land, but it has not developed into a commercially viable technique. In Germany 'Geooxidation', was promoted during the middle 1990's as an in-situ technique to 'mineralise' organic contaminants[x]. It was based on the idea that as a result of the induced electrical field, individual soil particles are acting as electrochemical electrodes, giving rise to anodic oxidation and cathodic reduction processes at each individual soil particle. The claims of success, however, could never be sustained and it was one of the reasons that in Germany electroreclamation has not been taken seriously for a long time. In Japan interest in electrokinetic techniques started in the middle 1990's. Many patents have been filed, most of them adaptations to existing American and European patents. Until recently, however, practical deployment of electroreclamation for the removal of ionic species has not been successful in that country.

But most of all, electroremediation became a popular subject for Research Institutes and Universities all over the world. Numerous scientists have been busy with laboratory experiments, sometimes it seemed as if trying to invent the wheel again. Looking back at 12 years of electrokinetic research and publications, we cannot but conclude that all this research has been of little practical value. Theory and practice of electroreclamation are not easily matched. Only in the field can electroreclamation be developed into a practical and economically viable remediation technology.

For the American market a joint venture company Geokinetics International Inc. was established in 1994 in Berkeley, California. In the Netherlands Geokinetics was acquired by Hak Milieutechniek BV. at the end of the 1990s. At the beginning of 2003 Hak Milieutechniek started an in situ test electro-reclamation in Japan with the aim to confirm the technology for a group of five Japanese companies, which are planning to deploy electro-reclamation in the near future.

Adapting to changing conditions

With the exception of 'pump and treat', in-situ remediation projects in the Netherlands were not very common at the beginning of the 1990's. Nonetheless, we continued applied R & D in this field. Until 1992 research was primarily focussed on increasing the efficiency of electroreclamation of heavy metals. After 1992 we shifted our main attention and research from electroreclamation of inorganics to electroreclamation of organics. There were three reasons for this change in company policy. First, at the beginning of the 1990's, the environmental regulators considered heavy metals in tight soils to be less of a problem, which resulted in a serious decrease in the number of remediation projects. The second reason was that groundwater remediation of organic pollutants by pump and treat as executed by other remediation companies, turned out to be not very successful. Initial optimistic estimates of remediation periods of several years had to be changed to several decades or even longer. In general the biggest problem when dealing with organic contaminants like e.g. aromatics, PAH, kerosene, petroleum diesel, heavy fuel oil, creosote and volatile chlorinated solvents (VOCs) are the so called smear zone around the phreatic surface, high concentration areas (hotspots) with free, undissolved product and tight soils like peat and clay. The composition of the contamination, together with the adsorption of compounds within the soil, the presence of colloidal suspensions and oil in water and water in oil phases means that conventional groundwater decontamination methods, such as vacuum extraction and pump and treat methods do not achieve the desired results. Pump and treat which in fact is similar to 'flushing' the saturated zone will almost never be enough, even when extremely long extraction or flushing periods are involved and even when the aquifer is characterised as "good". Contaminants that are present as droplets in the pores of the sediment and as thin film layers around soil particles will dissolve slowly and in small quantities only. Desorption of contaminants in the saturated and or unsaturated zone will generally not be enough in order to finalise remediation within an acceptable period of time (less than 10 to 20 years. In short, there was still an important and big market to be conquered. And lastly we had noticed that during our electroreclamation projects of inorganic contaminants temperature increase of the soil (Joule heating) resulted in higher permeability of clay soils and seemed to have no adverse effect on micro-organisms.

Applied R & D continued

During the period 1990 - 1996 we developed and tested many concepts for further enhancement of remediation techniques based on electrical current, some of which are discussed in more detail:

Dielectrophoresis: This research project was meant to investigate the possibilities of dielectrophoresis as a means to remove polar and a-polar toxic organic contaminants from the soil. It turned out that aromatics, PAH, pesticides, phenols, PCB etc. could be removed quite efficiently, but it also showed that this complex technique was not suited for in-situ remediation.

Electrokinetic fences: Based on a real situation of a groundwater plume contaminated with zinc up to a depth of 80 m b.g, we investigated the possibilities of electrokinetic fences using computer simulations (figure 2) and a laboratory model with soil and groundwater from the site. It turned out that the computer simulations and laboratory tests matched very well. In the laboratory model > 99 % of the zinc was captured by the fence. The investigation also showed that important parameters like electrode spacing and electrical power could be transformed into realistic practical values.

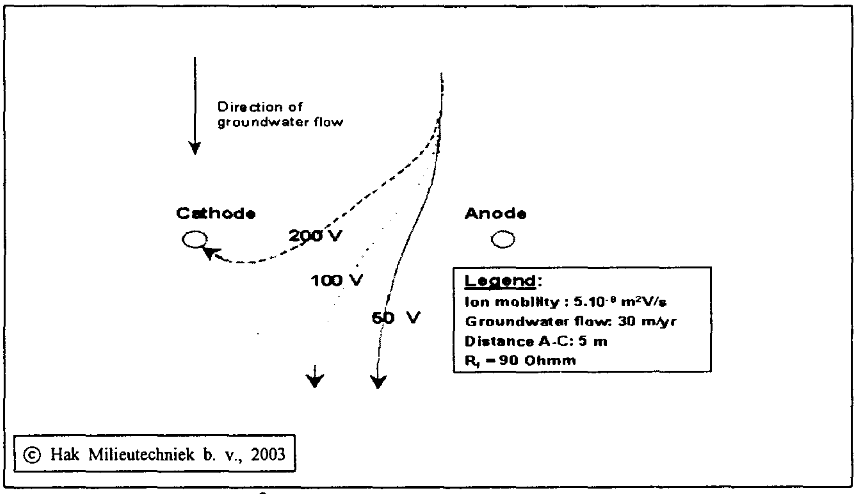

Figure 2. Calculated flow paths of Zn^{2+} ion entering the fence area upstream of the anode region using different potentials. At 200 V between anode (A) and cathode (C), all zinc ions are captured by the fence and the efficiency is almost 100 %. At 100 V between anode and cathode the efficiency of the fence is around 50 %.

Enhancement of biodegradation using electrokinetics: This investigation showed that the bio 'friendly' nature of electrochemical energy supply and electrokinetic nutrient supply can be used to dramatically accelerate adaptive growth of micro-organisms. In this case nutrients were added through the electrode wells and the electrokinetic processes were used to disperse these materials to the bacteria already present in the soil. pH was kept neutral by mixing cathode and anode solutions

Electrokinetic remediation of river and harbour sludge: A semi-technical set-up showed that by applying elongated ceramic elements for sludge and electrode compartments both inorganic and organic contaminants could be removed efficiently and economically. Lack of long term guaranteed contracts for large-scale sludge treatment was the main reason that no further development has taken place so far.

Electroreclamation of former gasworks sites: This field trial with some 150 tons of soil contaminated with cyanides and PAH, both in concentrations many times above the Dutch intervention values (I-value), was the first step in investigating the possibilities of remediating former gasworks sites with electroreclamation. At the end of the pilot project of 12 weeks, there was no trace of free cyanide left, total cyanide had been reduced to just underneath ½ * I-value and PAH concentrations were just above the target value.

Electroreclamation of TNT contaminated soil: In another project, 50 tons of soil contaminated with trinitro toluene (TNT explosive) from an old World War I munitions factory in Germany (which exploded in 1918) was treated by electrokinetically enhanced bioremediation in a period of about 3 months. During the process nutrients were added to the electrolytes and the soil was heated to 25-30 °C as a result of Joule heating. In addition As, Pb and other ionic contaminants were migrated out of the soil and recovered from the electrolytes. After three months of treatment the following results were obtained:

Table 2. Results of batch remediation of TNT by electrokinetically enhanced bioremediation.

Applied energy kWh/m^3	Conc. TNT mg/kg	Conc. DNT mg/kg	Conc. DNB mg/kg	Conc. PAH mg/kg	Conc. organic As mg/kg
0	49	188	553	40	11
31	70	10	2.7	nd	n.m
49	10.1	3.3	6.8	nd	0.11

TNT= trinitrotoluene. DNT= dinitrotoluene. DNB= dinitrobenzene. PAH= polycyclic aromatic hydrocarbons.
n.d= not detected, n.m= not measured

Note the removal of arsenic during the process into the anolyte wells and then recovery as an arsenate solution. Like the trials with soil from former gasworks sites, this is a good example of treating mixed wastes with electrokinetic technologies.

Expanding the technology

Based on the foregoing R & D results and market situation, from 1994 onwards, the field applications of electroreclamation for in-situ removal of organic contaminants have been actively pursued and developed into a practical technology[xi]. The "electro" part pertains to low (< 100 °C) temperature heating of the soil, which results in accelerated desorption and mobility of the organic contaminants. The difference between the specific mass of the soil compartment being heated and the surrounding colder soil complex results in an upward pressure. In combination with soil vapour and groundwater extraction via relatively closely spaced extraction wells, this upward pressure is even more enhanced. In order to maintain the heat economy of the subsurface a low pumping rate is used for groundwater extraction.

Apart from these physical/chemical effects of electrical heating, temperature increase has a pronounced and positive effect on biodegradation. Earlier expectations, predicting adverse effects when increasing temperature above 30 to 40 °C, have not been observed. On the contrary, investigations during gradual heating of the soil up to 80 °C over a period of 6-8 weeks showed abundant biological activity, even at these relatively high temperatures. At temperatures of 70 to 80 °C thermophylic bacteria populations are observed at significantly higher levels than are active at temperatures of 20 to 30 ° C. After closing down the electrical system these populations can be observed adapting again to the changing (cooling) temperature conditions. Biodegradation is once more accelerated by periodic injection of nutrients (N, P), and, depending on the type of contamination, carbon sources, oxygen and/or electron donors or acceptors. Results of finished and ongoing electro(bio)reclamation projects at sites contaminated with VOCS, have shown that first order biodegradation constants which were initially used for estimation of total remediation time increased one to two orders of magnitude (table 3).

$$PCE \xrightarrow{k1} TCE \xrightarrow{k2} Cis\text{-}DCE \xrightarrow{k3} VC \xrightarrow{k4} Ethene$$

'normal' conditions[1]	Enhanced conditions[2]
k1 = 0.0023	k1 = 0.221 - 0.361
k2 = 0.0033	k2 = 0.193 - 0.317
k3= 0.0038	k3 = 0.051 - 0.09
kK4 = 0.0015	k4 = 0.0062 - 0.051

Table 3. First order biodegradation constants without[1] and with[2] electro(bio)reclamation

It seemed obvious to call this technology **electro(bio)reclamation** (figure 3)

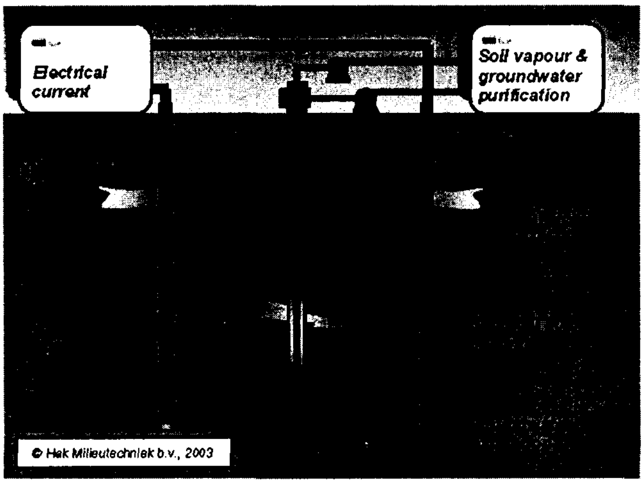

Figure 3. Schematic view of in-situ Electro(bio)reclamation of organic contaminants

In 1996 electro(bio)reclamation could be seamlessly incorporated into the newly adopted remediation policy of the Netherlands where multifunctional soil remediation was to a large extent changed into functional soil remediation which can be related tot the concept of 'Brownfields' and 'Greenfields'. The rigid principle that every contaminated site had to be cleaned up to some fictitious so-called background concentration level (former A-levels) was replaced by a much more differentiated and pragmatic policy, i.e. what is the future use of a polluted site, which remediation targets are realistic and what are the risks if some rest contamination remains ? And if that is the case, can we reach a so-called 'stable end situation', i.e. a situation where no further increase in size and/or concentration levels of the groundwater plume is observed? For Electro(bio)reclamation of organic species this new policy led to the following phased remediation approach:

1. Intensive phase — AC-heating of soil and groundwater in areas with high concentrations (free product zones and hotspots). Direct heating of the saturated zone and Indirect Heating of the unsaturated zone (capillary fringe and smear zone) by rising heat from the saturated zone. Heating is combined with groundwater and soil vapour extraction and periodical injection of nutrients, carbon sources, oxygen and/or electron donors/acceptors, depending on the nature of the pollutants.
2. Attenuated phase — During this phase attention is focused on biodegradation in the remaining source areas and adjoining groundwater plumes. Electrical heating is stopped. Groundwater and soil vapour extraction continue as well as periodical injection of nutrients oxygen and/or electron donors/acceptors and carbon sources.
3. Monitoring phase — Monitoring and control on the basis of periodic sampling and analyses, if clean-up criteria have not yet been met completely or there is risk of possible recontamination.

The execution of the different phases depends on the results of the foregoing ones. Changing from one phase to the other also depends on the so-called efficiency profile(s) of the remediation, as pictured in figure 4. Electro(bio)reclamation is particularly suited to clean-up "hotspots" during the intensive remediation phase, creating optimal conditions for a subsequent second phase of extensive remediation or natural attenuation.

Some projects and achievements

During the past 8 years 18 electro(bio)reclamation projects have been finished successfully, while are still running. They are mostly projects, which are characterised as serious cases of soil contamination. It concerns contamination underneath buildings, in residential areas, to relatively great depths and in 'difficult' soils like clay and peat. It concerns pollutants like heavy metals and organic species like diesel, petrol, fuel oil, PAH, PCE, TCE and their degradation products. A few of these projects which will be discussed in more detail.

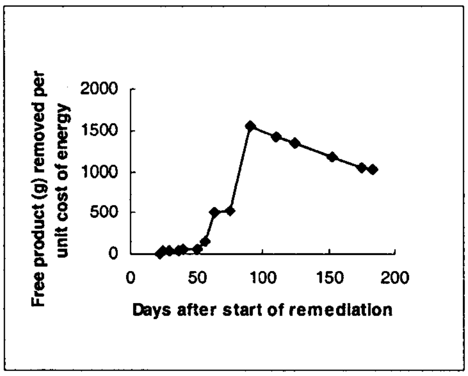

Figure 4. Efficiency curve during intensive phase of electro(bio)reclamation of PCE contamination. During first 50 days remediation efficiency is low. After 50 days efficiency increases dramatically as a result of desorption and mobilisation of PCE. After 100 days no free product is left and the amount of dissolved PCE removed against the costs of electricity result in a downward (negative) slope of the curve. By shifting to extensive methods, cost of energy will diminish and the curve will go up again.

Heavy metals

At the site of the military airbase at Woensdrecht, 2,600 m^3 of sludge, peat and clay contaminated with heavy metals have been remediated with electroreclamation during two years, in batches of 1,300 m^3 each (table 4). For energy about 200 kWh/m^3 was used, while some 330 kg of filtercake consisting of metal hydroxides were collected from the electrolytes. The target value was 50 mg/kg for Cd.

Table 4. Results of electroreclamation of heavy metals at Woensdrecht

	Cr	Ni	Cu	Zn	Pb	Cd
Concentration at beginning mg/kg)	7,300	860	770	2,600	730	660
Concentration at end (mg/kg)	755	80	98	289	108	47
Removal efficiency (%)	90	91	89	89	85	93

Chlorinated solvents

In the picturesque city of Nieuwpoort in-situ electro(bio)reclamation started in the middle of 1998 underneath a monumental building, and underneath and around some surrounding residential buildings up to a maximum depth of 12 m below grade. The site is part of a former silver factory and chlorinated solvents like PCE and TCE had been abundantly used. Of a total volume of 12,500 m^3 of contaminated clay and peat soil, 4,800 m^3 was remediated during and intensive phase of 2 years while 7,700 m^3 is still being remediated with attenuated remediation methods. Some 60 kg of free product has been removed to date. Investigation of the rate of biodegradation turned out that the initially used first order biodegradation constants had to be adjusted 1 to 2 orders of magnitude (see table 3). The project has been finished 2 years ahead of the originally calculated project termination.

Diesel and aromatics

In-situ electro(bio)reclamation up to a depth of 8 m below grade took place at the site of a former fuel storage facility and gas station in the city of Horst. During the heat-up phase to a maximum of 94 °C a free product layer of oil compounds, which was not present before, appeared on the groundwater level. At the end of the intensive phase, which lasted from January 1998 to July 1999 21,000 m^3 of free product had been removed. Attenuated remediation using enhanced biodegradation and periodic nutrient injection took care of the remaining contaminants during the following 2 ½ .

Table 5. Results of electro(bio)reclamation of mineral oil at Horst

Product removed	Amount (kg)
By excavation	1,000
By free product extraction	17,400
Via active carbon	2,000
Via air stripper	600
Total	21,000
Max. Temperature at 6 m depth	94 °C

Gasoline

On farmland near the city of Maasdam a subsurface fuel pipeline was damaged and under a pressure of some 17 bar an estimated 50,000 litres of gasoline was sprayed out over the land and flowed into the clayey soil, covering an area of some 3,000 m2. Most of the gasoline evaporated and free product leaked into the soil contaminating groundwater up to a maximum depth of 9 below ground level.

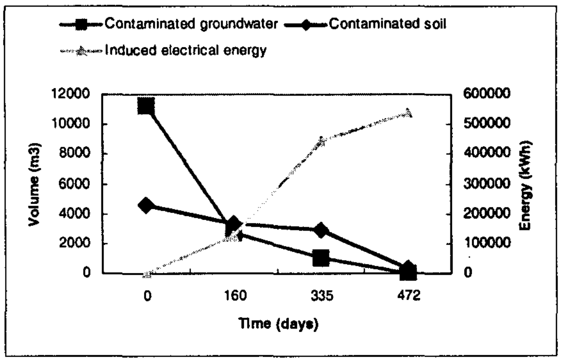

Figure 5. Decrease of contaminated volumes of ground and groundwater during electro(bio)reclamation at Maasdam

Within 80 weeks a volume of nearly 12,000 m^3 of contaminated groundwater and 5,000 m^3 ground has been remediated using electro(bio)reclamation. Total electricity consumption amounted to 540,000 kWh.

Fuel oil

In a commune for physically and mentally handicapped people at Woudenberg, an underground tank with fuel oil had been leaking and some 9,000 litres of oil had disappeared into the soil. During an intensive phase of electro(bio)reclamation of 13 months 7,800 litres of fuel oil have been recovered. During a following extensive phase of 19 months another 1,000 litres have been removed from the soil by enhanced biodegradation.

Cyanide and PAH

In the city of Oostburg a pilot project in-situ electroreclamation at the site of a former gasworks has recently been be finished. The project has been terminated one month ahead of the scheduled 3 months, because cyanide and PAH levels have dropped down to target levels much faster than estimated. This pilot study is of particular interest as during this study bioactivity has been stimulated in a zone between the electrodes to see its possible effect on degradation of cyanide complexes and PAH. In another zone injection of an alkaline solution was thought to enhance cyanide solubility and thus remediation efficiency. Final data, however, indicate that the best results are obtained in the zone where only DC has been applied, i.e. electrokinetic alkalisation seems to be the most efficient dissolution mechanism.

Electrokinetic biofence

In one of the northern provinces of the Netherlands at the city of Wildervank an electrokinetic biofence has been installed to a depth of 10 m b.g. in a groundwater plume contaminated with PCE, TCE, Cis-DCE and VC. This project is the outcome of renewed interest in our concept of electrokinetic fences along the borders of contaminated groundwater plumes, developed in the early 1990's. Such fences may function not only as capture zones of heavy metals, but also as elongated and relatively narrow zones wherein nutrients and/or electron donors are being dispersed electrokinetically, either from the electrode filters or from upstream infiltration wells. Within these zones, the groundwater contaminated with organic pollutants is being cleaned by electrokinetically stimulated biodegradation. Such an approach is characterised by avoiding the necessity to pump hundreds of thousands of cubic meters of groundwater and thus by not influencing the groundwater flow regime. The project has been scheduled for a duration of 4 years. For the electrical energy use is made of a combination of solar energy and electricity from the grid.

Nickel and Zinc

After almost 6 years of tranquillity on the heavy metals remediation front, there is also renewed interest in deploying electroreclamation at sites contaminated with (mobile) heavy metals. A project has started in 2003 at a site where galvanising activities have resulted in pollution of soil and groundwater with nickel and zinc up to a depth of 6 m b.g. The in-situ electroreclamation system has been installed inside the plant, allowing for almost undisturbed continuation of the production process. The cleanup operation has been scheduled for a period of approximately 2½ years.

Epilogue

At the start of the new millennium it can be established that electroreclamation and its derivative electro(bio)reclamation in combination with other in-situ techniques have been developed into a mature and versatile remediation technology, both for inorganic and organic contaminants. From the very beginning, the technology development has been driven by commercial pressures and practical clean-up operations. It means that theory and practice have been close partners, in fact they have been inseparable. Beyond that there has always been a strong believe in the necessity and functionality of in-situ remediation, independant of the prevailing opinion of environmental regulators. This has been the key that electroreclamation plays a very important part in current in-situ remediations in the Netherlands. It still is an innovative technology but certainly not an emerging one.

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