

Waste for waste: a new management approach in deteriorated soil remediation

Orsolya Klebercz¹, Viktoria Feigl¹, Emese Vaszita¹, **Katalin Gruiz¹**, Judit Kovács², Viktor Kőfalusi²

¹Department of Applied Biotechnology and Food Science, Budapest University of Technology and Economics

².A.S.A. Magyarország Ltd.

Abstract

We can link the requirements of sustainable soil quality with the goal of waste utilisation by integrating modern environmental scientific knowledge and technological innovation into a risk based management system. As examples, two case studies are introduced in this article.

The area of the former lead and zinc sulphide ore mine in Gyöngyösoroszi, Hungary is heavily polluted with toxic metals. The combination of phytostabilisation and chemical stabilisation using wastes as amendment was applied to various metal sulphides containing mine wastes.. The fly ashes were able to reduce the water soluble Cd and Zn content by more than 99% and the acetate extractable content by 49%. The high toxicity of the mine waste was reduced, and healthy vegetation developed on the previously barren surface.

The waste deposit at Gyál, Hungary is covered by low quality waste soils. Soil amendment experiments with wastes as soil additives demonstrated that addition of a mixture of wastes to the soil can result in a fertile cultivation medium.

Introduction

Extensive soil contamination and deterioration are worldwide problems resulting bad quality soils, problems in ecosystem and agricultural land uses. Mining activities, constructions, industries and transport may result widespread areas of low quality, toxic surface materials, unable to fulfil the requirements of topsoils, leading to reduced yields in agricultural areas, and deterioration of the natural ecosystem. These processes result in barren or semi-barren sites, exposed to further erosion and degradation, therefore bad quality soils represent a risk for the whole ecosystem.

Waste utilization for soil amelioration in a scientifically substantiated and conscious manner provide an environmentally friendly and affordable alternative for the remediation of former mining sites, revegetation of brownfields, or amelioration of agricultural lands. The research undertaken in this field by the authors represents a new, holistic approach in waste management and soil amelioration, where the needs of bad quality soils can meet the values of waste materials. This paper presents two case studies, demonstrating that from two waste materials we can produce a higher quality material: a fertile cultivation medium, as a basis of a new soil formation.

1. Management framework of waste utilization for soil quality improvements

Risk Based approach combined with a Value Based evaluation of wastes makes possible matching certain wastes with low quality soils to find waste utilization technologies fitting for soil remediation or amelioration aiming at sustainable and eco-efficient soil use and waste elimination. The short and long term risk resulted from handling and storing of wastes and from soil degradation can be quantified and compared with that of utilisation of waste on soil. The values and benefits can also be quantitatively evaluated by technological and socio-economic evaluation tools. The two quantitative indicators together could serve as a basis for a good decision making on the proper utilisation of wastes for soil-improvement (Gruiz, 2009).

Figure 1 shows the scheme of the risk management concept of waste utilisation for soil improvement. The process starts with information collection on the waste, its origin, composition, land and soil use at the place of the planned application. The next step is the creation of the risk scenario for risk calculation. The hazard should be assessed based on the known adverse effects of the waste. The result of the hazard assessment is the “no effect” value, which is the predicted concentration or level, which does not pose risk to the environment (PNEC= Predicted No Effect Concentration) and to humans (DNEL=Derived No Effect Level). The calculated value should be compared to the predicted concentration in the environment after waste utilisation (PEC=Predicted Environmental Concentration). The ratio of PEC and PNEC/DNEL is the Risk Quotient (RQ). Calculation of RQ is an iterative process within which, data precision and calculation can be increased if the conservative estimates do not fulfil the $RQ < 1$ criterion. The hazard associated with the waste differs from the land-use specific risk of waste utilisation on soil, and the value-based benefits may compensate for the hazard-based risks.

Environmental risk management of waste utilisation on soil

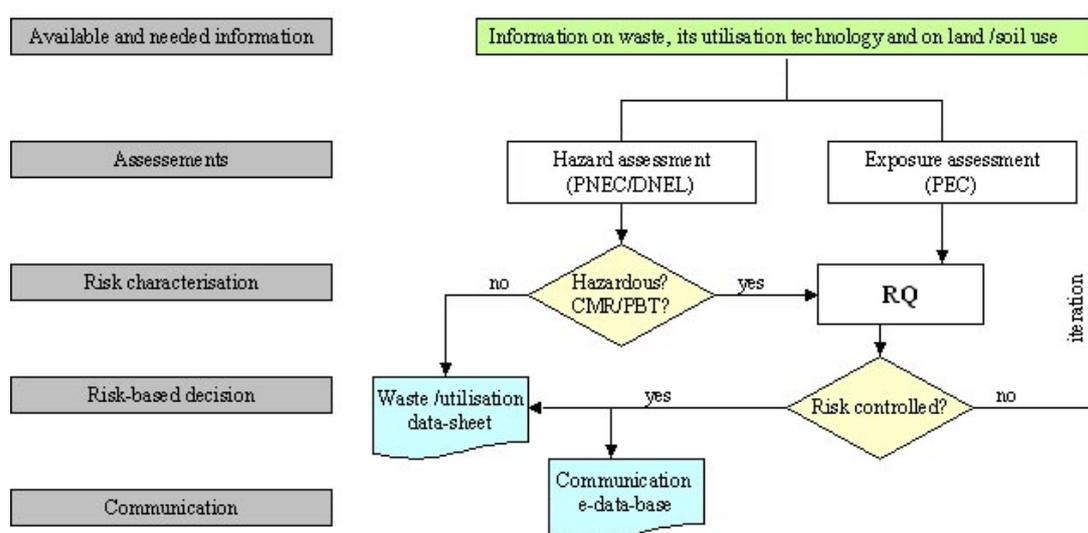


Figure 1 Environmental risk management scheme of waste reuse and recycling

The essential management tools are based on comparative evaluation and verification of waste utilisation, soil amelioration and remediation technologies. Verification is performed based on demonstration projects employing a well fitting, targeted, integrated technology and environment monitoring during the waste utilisation on soil. In this paper we chose to solve two soil problems: by remediation of metal contaminated sites, and amendment of nutrient depleted soils of non-adequate texture for plant growth.

2. Examples of soil amelioration using wastes

2.1. Waste utilization for the remediation of metal contaminated sites

Problem of toxic metal contamination and solutions using waste

In situ chemical stabilization of toxic metals in abandoned mining sites using waste products as amendment is one of the most urgent problems studied by several researchers. In Europe Vangronsveld et al. (1995) were the first who applied successfully fly ash and organic wastes to reduce the risk of a diffusely metal-polluted site. Since then there are many successful applications by Ruttens et al. (2006), Dermatas et al. (2003), and by our group (BUTE) as described hereinafter.

Many other waste-origin materials are proven to be able to reduce the mobility of toxic metals. Adding communal sewage sludge in proper ratio to the soil can both reduce the mobility of heavy metals in the soil (Theodoratos et al 2000) and have a positive effect on the produced vegetation. Several researches show the effectiveness of red mud (Lombi et al 2001; Gray et al 2006; Friesl et al 2005). Lime-containing waste materials like sugar beet lime (Madejón et al 2005) gypsum-containing wastes (Garrido et al 2004), phosphogypsum (Abril et al 2008), bone meal (Sneddon et al 2008) are also used as stabilizing soil amendment materials. The effect of these materials can be improved by combining materials with different metal stabilizing mechanism, like Ruttens (2006) does with compost (bondage to organic material), cyclonic ashes (bondage to puzzolanic material) and iron forges (coprecipitation with Fe). These combined wastes can not only have synergic effect to each other, but thanks to the different mechanisms, the stabilizing effect can be more durable and more indifferent to changes in environmental surroundings (e.g. soil pH).

Case study: remediation of the former mining area of Gyöngyösoroszi

Phytostabilisation combined with chemical stabilisation is the chosen best technology for the diffusely contaminated large area of the abandoned lead-zinc sulphide ore mine situated in the Mátra Mountains, Northern Hungary (Gruiz et al 2009). Mine waste, barren dead rocks and flotation tailing material, as well as secondary sources like river-bed and pond sediments are the sources of high As, Cd, Cu, Pb and Zn contamination in the catchment.

Phytostabilisation combined with chemical stabilisation is a proven technology, as indicated by successful studies performed in both Europe and USA (Schwitzguébel et al 2002, Vangronsveld et al 2009). To enhance plant growth and prevent transport of toxic metals by runoff into surface waters and by infiltrating waters into

subsurface waters as well as by plants into food chains *in situ* chemical immobilization (stabilisation) of the metals is necessary. The chemical stabilisers lower the mobility and bioavailability of toxic metals in the soil, therefore reduce erosion and leaching and enable settling of plants, without letting them to take toxic metals up into the above-ground parts. The complex stabilisation system lowers the environmental risk of diffuse pollution and of plants grown on it.

Experimental design

Mine wastes (weathered acidic rock) and mine waste contaminated soils from the phytostabilisation experimental field in Gyöngyösoroszi were treated in laboratory microcosms with different amendments of waste origin:

- five different types of fly ashes, two from the power plant in Oroszlány, Hungary (fly ash “OA” pH=12.6 and “OB” pH=9.7) in 1 w%, 2 w% and 5 w%, two types from Tata, Hungary (fly ash “TA” pH=7.2 and “TB” pH=6.8), and one from Visonta, Hungary (fly ash “V” pH=6.4), added to the soil in 2 and 5 w%,
- two types of Fe-Mn-hydroxide precipitate from drinking water treatment from Budapest (precipitate “R” and “C”, in 2 and 5 w%), and
- red mud (2 and 5 w%) from bauxite processing, stored in Almásfüzitő.

Mine waste rock and metal contaminated soil treated with additives and the untreated control were placed in 2 kg pots each. They were incubated at 25 °C, were mixed and moistened regularly. The duration of the experiments varied from 2 to 3 years.

As a result of the addition of stabilizers to the mine waste, complex processes take place in the microcosms. Metal mobility, solubility and bioavailability are changing continuously until reaching a new equilibrium. The process should be monitored on the long term, to demonstrate the efficiency of the additive during a longer period. The stabilisation and as a consequence the decreased mobility, solubility and bioavailability of toxic metals in the treated mine waste was monitored by an integrated methodology, which combined physico-chemical analysis with biological and ecotoxicity testing (Feigl et al 2009a). The integrated methodology included the analysis of toxic metal content (ICP-AES) with different separation methods (Aqua Regia, Ammonium-acetate buffer solution and distilled water), *Sinapis alba* rapid metal bioaccumulation test, *Vibrio fischeri*, *Azotobacter agile* and *Sinapis alba* direct contact laboratory tests.

Metal stabilization of mine wastes and mine waste contaminated soils using waste products as additives

The mine wastes and soils of Gyöngyösoroszi are mainly contaminated with As, Cd, Cu, Pb and Zn. The most mobile metals are Cd: its concentration at the site is 4.2–23.3 mg/kg and Zn: 926–4420 mg/kg. According to the applied extractants of gradually growing acidity 26–34% of Cd and 23–24% of Zn are present in a highly mobile form (acetate extract % of Aqua Regia extract) while 7–13% of Cd and 6–11% of Zn is water-soluble (in % of Aqua Regia extract).

Table 1 shows the efficiency of the stabilizing agents resulting in the decrease of the mobile Cd and Zn fractions compared to the untreated (0%). Amongst the tested fly ashes, type “OA” was the most efficient in reducing the mobile metal content of the mine waste contaminated soil: it decreased the water soluble Cd and Zn content by more than 99%. Its immobilising effect was observed already 21 days after addition and remained unchanged after 2 years. The larger the quantity of the added fly ash (1, 2, 5 w%), a larger decrease in the mobility was measured. The non-alkaline fly ashes (“TA”, “TB” and “V”) were less effective, but still resulted in 84–96% decrease in water soluble Cd and Zn contents. Their efficacy can be further improved by the addition of lime (Feigl et al 2009a).

The other examined wastes showed surprisingly high efficiency: From the two Fe–Mn precipitates of drinking water treatment the type “C” was more effective than the type “R”, especially in reducing the mobility of Zn, and both types were more effective for Cd than for Zn. Red mud reduced the mobility of all metals in the waste contaminated soil.

Table 1: Decrease in acetate extractable and water soluble metal concentrations in mine wastes and mine waste contaminated soils treated with stabilizing amendments: selected best long-term results (25–36 months) for each amendment

	Fly ash “OA”	Fly ash “OB”	Fly ash “TA”	Fly ash “TB”	Fly ash “V”	Prec. “R”	Prec. “C”	Red mud
pH – untreated	5.4	5.4	6.3	5.6	5.6	6.5	6.5	6.9
pH – treated	7.2	6.7	6.7	6.5	6.6	6.7	7.1	7.1
Extractable metal contents of untreated (mg/kg)								
Acetate extractable Cd	2.25	2.25	1.24	1.85	1.85	1.24	1.24	3.28
Acetate extractable Zn	302	302	187	356	356	187	187	354
Water soluble Cd	1.08	1.08	0.020	0.890	0.890	0.020	0.020	0.042
Water soluble Zn	152	152	2.27	213	213	2.27	2.27	2.38
Decrease due to treatment (%) compared to untreated (0%)								
Acetate extractable Cd	45%	30%	2%	55%	57%	51%	46%	40%
Acetate extractable Zn	49%	34%	18%	66%	63%	17%	38%	62%
Water soluble Cd	>99%	94%	48%	84%	90%	>79%	>79%	85%
Water soluble Zn	>99%	98%	35%	93%	96%	36%	60%	85%

The toxicity test results validated the results of chemical analysis. The best results were obtained by the addition of fly ashes “OA”. The bacterial *Vibrio fisheri* and *Azotobacter agilis* test showed that the toxicity decreased with ~30% particularly after 2 and 5 w% of fly ash treatment and the *Sinapis alba* plant growth test showed 60–70% reduction in toxicity. The other amendments were less effective, but also reduced the toxicity of the original materials (Feigl et al 2009a).

The self-developed rapid bioaccumulation test with *Sinapis alba* showed that addition of 5 w% “A” fly ash to the soil diminished the Cd and Zn uptake of the test plant by 58–74% (**Table 2**). Two other fly ashes (“TB” and “V”) also caused significant decrease in the bioaccumulated metal content (74–86%), while the other amendments were less effective, but still causing max. 29% decrease in metal uptake.

Table 2: Decrease in bioaccumulated metal concentrations in *Sinapis alba* test plants grown on mine wastes and mine waste contaminated soils treated with stabilizing wastes: selected best long-term results (25–36 months) for each amendment

	Fly ash “OA”	Fly ash “OB”	Fly ash “TA”	Fly ash “TB”	Fly ash “V”	Prec. “R”	Prec. “C”	Red mud
Bioaccumulated metal contents in <i>S. alba</i> on untreated (mg/kg)								
Cd	2.99	2.99	0.900	1.70	1.70	0.478	0.478	0.900
Zn	743	743	170	711	711	99.3	99.3	170
Decrease due to treatment (%) compared to untreated (0%)								
Cd	71%	58%	6%	74%	75%	8%	18%	19%
Zn	71%	74%	21%	86%	86%	21%	20%	29%

According to the results of the microcosm experiments field lysimeters and small-scale field experiments were set up as part of the scaled-up technological experiments (Feigl et al 2010a), which also confirmed the stabilizing and risk reducing effect of fly ash (Feigl et al 2009b).

2.2. Revegetation and rehabilitation: creating a fertile topsoil layer

Problem of soil degradation and loss of fertility

Apart from soil contamination several other soil degradation processes caused by anthropogenic effects result in the damage of soil fertility, soil texture and ecosystem. Acidification, sodification and salinisation are the disturbances of chemical and structural equilibrium of soil composition. Erosion is the loss of soil due to degraded soil structure, often caused by the previously mentioned processes. Soil degradation is in close relation with the agricultural overuse of land, the exhaustion of organic material and nutrient supply and soil buffering capacity, and the regular disturbances of soil structure (tillage, irrigation etc). On the other hand, soil structure degradation results in the loss of nutrients, closing the vicious circle resulting in nutrient depleted soils with seriously damaged texture. Therefore, when planning soil amelioration we need to have a complex view over the composition of soil, the soil forming and degrading processes, including physical, chemical and biological effects.

Case study: development of fertile cultivation medium from wastes

Our demonstration site is located at the communal waste landfill of A.S.A. Hungary near Budapest. Communal waste deponies, raised gradually along with the incoming amount of waste, use large quantities of low quality soil and inert waste originating

from construction and demolition sites. This material is very heterogeneous, typically low in organic matter and nutrients and constitutes the surface layer of the deposit from the start of landfilling till the final recultivation in the next 10 or more years. Therefore a temporary vegetation is needed to protect the slopes from erosion and improve the esthetical view of the deposit close to the residential area. The goal was to develop a 40–50 cm deep layer of fertile cultivation media at affordable price, utilising the actual, yet barren covering material mixed with organic and inorganic wastes, which can sustain a continuous vegetation on the surface of the deposit during its continuous process of construction. We must find an amendment technology which can be used uniformly for the heterogeneous material of the deposit. Therefore we chose four sample sites on the slope of the deposit with different covering materials, and tested in microcosm experiments the effect on the covering material of fly ashes and pretreated sewage sludges as nutrient supply.

From all four types of slope material, there was an untreated control microcosm, and one treated with fertilizers as positive control. We applied three types of organic amendments (raw sewage sludge, digested sewage sludge, composted sewage sludge and the 1:1:1 mixture of these three) at 10% waste to soil ratio. As inorganic amendment, coal combustion fly ash and wood ash were used in 3% waste to soil ratio. The preliminary characterization of the planned waste additives showed that many of them have some toxic effects: fly ash, wood ash and digested sewage sludge showed toxicity for *Sinapis alba* (white mustard), and fly ash for *Vibrio fischeri* (bacteria). Nevertheless, at 10% mixing ratio into the soil they showed no toxicity, but a positive effect on nutrient content and soil habitat.

After one month of incubation, the microcosms were planted with a special grass mixture described by Feigl, 2010. The experimental matrix is shown in **Table 3**.

Table 3 Treatments of the ASA landfill slope material in microcosms

Treatment	site 1	site 2	site 3	site 4
Untreated control	x	x	x	x
Artificial fertilizer only	x	x	x	x
3% fly ash	x			
3% wood ash	x			
10% raw sewage sludge	x		x	x
10% digested sewage sludge	x		x	x
10% composted sewage sludge	x		x	x
10% sewage sludge mix	x	x	x	x
10% sewage sludge mix + 3% fly ash	x	x	x	x
10% sewage sludge mix + 3% wood ash	x	x	x	x

Table 4 shows the fertility change of the soil as a result of amendment addition. This was expressed by the measured biomass production increase of the grass planted in the microcosms.

Table 4 Grass biomass increase (%) after 4 month of growth.

Treatment	Grass biomass increase compared to untreated control (%)				
	site 1	site 2	site 3	site 4	Average
Artificial fertilizer only	382%	100%	289%	132%	226%
3% fly ash	9%				9%
3% wood ash	64%				64%
10% raw sewage sludge	718%		442%	363%	508%
10% digested sewage sludge	482%		368%	253%	368%
10% composted sewage sludge	591%	163%	263%	89%	277%
10% sewage sludge mix	536%	175%	395%	263%	342%
10% sewage sludge mix + 3% fly ash	691%	171%	421%	232%	379%
10% sewage sludge mix+3% wood ash	609%	150%	395%	337%	373%

These results demonstrate that although a waste material may pose a certain risk for the environment, if used in the right concentration, its values can make up for their risks, and they may have good effect on soil.

3. Conclusion

Barren, or semi-barren areas with contaminated, degraded soil pose a constant risk to the ecosystem and lead to further contamination and deterioration. Removal, exchange and disposal of these soils is expansive and not sustainable. In-situ amendment or remediation with wastes has a double benefit: landscaping of these areas and utilization of agricultural and industrial wastes. The usable wastes can have also their own risks, but they can be decreased, and compensated by the benefits produced in changing soil properties. Although this approach has many positive points/aspects, demonstrated by several scientific experiments and demonstration projects, the industrial and legal practitioners are still not ready to accept it. To make it more accessible for decision-makers, a full-scale, scientific, management and legal background has to be developed.

Acknowledgements

Microcosm tests were incubated, and analytical measurements were conducted in the laboratory of Research Institute for Soil Science and Agricultural of the Hungarian Academy of Sciences (RISSAC).

This work is part of the SOILUTIL project, funded by the Hungarian National Technology Program's Liveable and Sustainable Environment sub program under the code number TECH 09 A4.

This work is connected to the scientific program of the " Development of quality-oriented and harmonized R+D+I strategy and functional model at BME" project. This project is supported by the New Hungary Development Plan (Project ID: TÁMOP-4.2.1/B-09/1/KMR-2010-0002).

References

- Abril, J.M., García-Tenorio, R., Enamorado, S.M., Hurtado, M.D., Andreu, L., Delgado, A (2008) “The cumulative effect of three decades of phosphogypsum amendments in reclaimed marsh soils from SW Spain: 226Ra, 238U and Cd contents in soils and tomato fruit” *Sci. of the Total Environ.*, 403, 80-88.
- Bihariné dr. Krekó I. (2002): “Komposztálás” *Környezetünk Magazin*, www.kornyezetunk.hu
- Delgado-Moreno, L., Sánchez-Moreno, L., Peña, A (2007) “Assessment of olive cake as soil amendment for the controlled release of triazine herbicides” *Sci. of the Total Environ.*, 378, 119–123.
- Dermatas, D., Meng, X. (2003) “Utilization of fly ash for stabilization/solidification of heavy metal contaminated soils” *Eng. Geol.*, 70(3-4), 377-394.
- Difpomine (2011) Information on www.difpolmine.org
- Feigl V., Uzinger N., Gruiz K. (2009a) “Chemical stabilisation of toxic metals in soil microcosms” *Land. Contam. Reclam.*, 17(3–4), 483–494.
- Feigl V., Uzinger N., Gruiz K., Anton A. (2009b) “Reduction of abiotic stress in a metal polluted agricultural area by combined chemical and phytostabilisation” *Cereal Res. Commun.* 37, 465–468.
- Feigl V., Anton A., Gruiz K. (2010a) “An innovative technology for metal polluted soil – combined chemical and phytostabilisation” Sarsby RW, Meggyes T (eds.) *Construction for a sustainable environment* Proceedings of the International Conference of Construction for a Sustainable Environment, Vilnius, Lithuania, 1–4 July, 2008, Taylor and Francis Group, London, 187–195
- Feigl, V., Gruiz, K., Anton, A. (2010) „Remediation of metal ore mine waste using combined chemical- and phytostabilisation” *Periodica Polytechnica*, 54 (2), 71–80.
- Friesl, W., Horak, O., Wenzel, W.W.(2004) “Immobilization of heavy metals in soils by the application of bauxite residues: pot experiments under field conditions” *J. Plant Nutr. Soil Sci.* 167, 54-59.
- Garrido, F., Illera, V., García-González, M. T. (2005) “Effect of the addition of gypsum- and lime-rich industrial by-products on Cd, Cu and Pb availability and leachability in metal-spiked acid soils” *Appl. Geochem.*, 20, 397–408.
- Horváth, B. and Gruiz, K.(1996) “Impact of metalliferous ore mining activity on the environment in Gyongyosoroszi, Hungary” *Sci. of the Total Environ.* 184, 215-227.
- Gruiz, K., Vaszita, E., Siki, Z., Feigl, V., Fekete, F. (2009) “Complex environmental risk management of a former mining site” *Land Contam. and Reclam.*, 17 (3–4), 355–367.
- Gruiz, K. (2009) “Scientific and engineering ‘improvement’ of environmental risk management” *Land Contam. & Reclam.*, 343–346. EPP Publications Limited, UK

- Gruiz, K., Feigl, V., Vaszita, E., Klebercz, O., Ujaczky, É., Atkári, Á. (2010) „An integrated approach for the utilization of waste on soil – Innovative management and technologies” *ConSoil Conference, Proceedings CD*
- Gray, C.W., Dunham, S.J., Dennis, P.G., Zhao, F.J., McGrath, S.P. (2006) “Field evaluation of in situ remediation of a heavy metal contaminated soil using lime and red-mud” *Environ. Pollut.*, 142, 530-539.
- Klebercz, O., Gruiz, K., Feigl, V., Anton, A. (2010) „Introducing the SOILUTIL Project” *ConSoil Conference, Proceedings CD*
- Lombi, E., Zhao, F.J., Zhang, G., Sun, B., Fitz, W., Zhang, H., McGrath, S.P.(2002) „In situ fixation of metals in soils using bauxite residue: chemical assessment” *Environ. Pollut.*, 118, 435–443.
- Madejón, E., Pérez de Mora, A., Felipe, F., Burgos, P., Cabrera, F. (2005) “Soil amendments reduce trace element solubility in a contaminated soil and allow regrowth of natural vegetation” *Environ. Pollut.*, 139, 40-52.
- Ruttens, A., Mench, M., Colpaert, J.V., Boisson, J., Carleer, R., Vangronsveld, J. (2006) “Phytostabilization of a metal contaminated sandy soil. I: Influence of compost and/or inorganic metal immobilizing soil amendments on phytotoxicity and plant availability of metals” *Environ. Pollut.*, 144, 524-532
- Sneddon, I.R., Orueetxebarria, M., Hodson, M.E., Schofield, P.F., Valsami-Jones, E. (2006) “Use of bone meal amendments to immobilise Pb, Zn and Cd in soil: A leaching column study” *Environ. Pollut.*, 144, 816-825.
- Schwitzguébel, J., van der Leile, D., Baker, A., Glass, D., Vangronsveld, J. (2002) „Phytoremediation: European and American Trends” *JSS – J Soils and Sediments*, 2, 91-99.
- Theodoratos, P. et al. (2000) “The use of municipal sewage sludge for the stabilization of soil contaminated by mining activities” *J Hazard. Material* 77, 177–191.
- Vangronsveld, Streckx, Van Assche, Clijsters (1995) “Rehabilitation studies on an old non-ferrous waste dumping ground: effects of revegetation and metal immobilization by beringite” *J. Geochem.l Explor.*, 52, 221–229.
- Vangronsveld J., Herzig R., Weyens N., Boulet J., Adriaensen K., Ruttens A., Thewys T., Vassilev A., Meers E., Nehnevajova E., van der Lelie D., Mench M. (2009) “Phytoremediation of contaminated soils and groundwater: lessons from the field” *Environ Sci Pollut Res*, 16, 765–794.
- Williamson, J.C., Akinola, M., Nason, M.A., Tandy, S., Healey, J.R., Jones, D.L. (2009) “Contaminated land clean-up using composted wastes and impacts of VOCs on land” *Waste Management*, 29, 1772–1778