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Summary

Gentle remediation options (GRO) include various and in general plant-based options to remediate trace element contaminated soils (TECS) at low cost and without significant negative effects for the environment. Although GRO comprise very innovative and efficient technologies, they are still not widely used as practical site solution due to several hindrance reasons. Greenland has been launched on January 1 2011 to address several issues:

- Sustainable management adapted to TECS and deployment of GRO at field scale;
- Valorisation of plant biomass produced on trace element (TE) -contaminated sites;
- Harmonization of methods to assess the bioavailability of TE and development of a tool set to monitor the sustainability of GRO;
- Improving GRO through plant selection and modifications in soil TE bioavailability;
- Appraisal of current GRO practice, and development of implementation guidance and decision support frameworks.

Various current gaps / shortfalls in GRO application and development, i.e. lack of rigorous large-scale applications of GRO, variability in methods used for analysis and appraisal, potential for increasing the efficiency of GRO through biotechnology, technologies for biomass valorization and their uncertainty, best-practice guidances for the application of GRO at field scale, including appraisal of the various options available and their uncertainty, methods for monitoring, development and evaluation of a decision support tool (DST) focused on GRO, which can be integrated into existing, well-established and utilized DSTs / decision-frameworks, etc. are addressed. The overall aim of GREENLAND is to make GRO fit for purpose, which will substantially contribute to improvement of soil quality and ecosystem services on the local level (land owners, communities), but also at the European level. In addition to that, the use of biomass produced on GRO-treated contaminated land will contribute to socio-economic development at the local level and help to fulfil the increasing demand for biomass use in raw material and energy production all over Europe while avoiding the competition for food production and land change use.

During the first 3 years of Greenland, major achievements have been reached in all workpackages. The field experiments have been successfully maintained, in spite of partly challenging conditions, e.g. severe drought periods. Several biomass valorisation options were tested. Based on test results, literature overview and ongoing interviews with biomass processing enterprises, a list of feasible options is prepared. Suitable indicators of gentle remediation progress and success were identified in a comprehensive ring test on soil samples from field trials. The results obtained so far were summarized and submitted for publication. A second ring test will confirm the preliminary results. To enhance the efficiency of GRO, several plant-associated bacterial strains and combinations of soil amendments were evaluated. For bringing GRO into practical application, a decision support system has been published, along with valuable information on stakeholder engagement and empowerment. A technical guideline, which will be available at the end of Greenland, is now prepared.

Introduction

Contamination of soils with trace elements (TE) is a worldwide and thus also a pan-European problem. The concern refers both to soil contamination in itself and to the dangers for other compartments, mainly water and food production, which affect ecosystems and human life. In EU countries, the number of potentially contaminated sites adds up to nearly 3 millions (EEA, 2007). Around 80,700 sites have been cleaned up over the last 30 years in the countries with available data on remediation. According to the European Environment Agency (EEA), at least 250,000 polluted sites in the member states require an urgent clean up. Current investigation trends will increase by 50% the number of sites needing remediation by 2025. Mineral oil (38%), TE (37%) and PAH (13%) are the most frequent soil contaminants at investigated sites.

Many industrial sites severely polluted with TE emitted from local sources have been identified in all European countries. Even wider areas are slightly or moderately contaminated with TE from diffuse sources such as traffic, waste management and application, mining and industrial activities. The low and moderate levels of pollution, which are clearly above European standards, have led to limitations in land use, especially to protect safety of plant-derived products and consumers. TE loading in the soil decreases the chances of the establishment and survival of non-metal tolerant plant species and may strongly decrease the biodiversity of local ecosystems; the loss of biodiversity is also reflected in the animal community of these ecosystems. In addition, microbial biodiversity is affected by TE load. Microbial activity plays a pivotal role in maintaining soil fertility, controlling carbon and nutrient cycling in the ecosystems and promoting plant growth.

Over the last two decades, various gentle remediation options (GRO), which are less invasive, more cost-effective and preservative or even restorative to soil structure and functions than civil engineering techniques (e.g. encapsulation, vitrification, soil washing) and more sustainable than ‘dig and dump’ strategy, have been developed at bench and greenhouse scales. These new techniques are broadly based on phytoremediation, the use of plants and associated microorganisms to clean up the contaminated soils, partly assisted by the use of various amendments and soil management practices. As well as remediation / soil restoration, these options may be valuable in the production of usable biomass from formerly unproductive land.

An ERANET-SNOWMAN funded European report (SUMATECS: http://www.snowman-era.net/downloads/SUMATECS_FINAL_REPORT.pdf) has provided a detailed overview on the state of the art of GRO, including their potential environmental and socio-economic impacts as well as their integration into decision tool systems. However, SUMATECS has further identified the major reasons of hindrance for applying GRO as practical solution and derived the respective research needs in order to make GRO more attractive for decision makers and stakeholders to use these methods as practical site solution:

- Performance on large field scale (up-scaling) remains to be demonstrated (Mench et al. 2009, 2010)
- Treatment / Valorisation of TE-contaminated plant biomass needs to be tested
- Stakeholders and Decision makers are little informed about GRO
- Decision support tools do not sufficiently consider GRO (Onwubuya et al, 2009)
- The success of GRO is mostly reflected by changes of TE bioavailability, which is currently not sufficiently considered by legal frameworks; changes in other pollutant linkages should be also assessed.
- On contaminated land under agricultural use the switch towards GRO is under strong (financial) competition from regular farming activities producing food and feeding stuffs depending on the risk level of TE transfer into products.

The work carried out in GREENLAND (www.greenland-project.eu, Jan. 2011 - Dec. 2014) aims at bringing GRO into application and to deploy them on sites tandem with ecological and financial returns from biomass valorisation. In order to address remaining questions regarding the practical application of GRO, a set of field experiments (case studies) was set up or continued if already established and shared as a common set of research platforms in GREENLAND. This approach has allowed to

- to summarize and exchange information on all aspects related with the application of GRO as practical site solutions
- to compare the performance of different GRO under different conditions (contaminants, climate, soils, history of land use, etc.), paying attention to various issues (food safety after soil clean-up, quality of (micro)habitat, biodiversity, sustainability, biomass quality regarding its processing, compliance with future needs of communities, ecological consequences, by-products, wastes and side-effects, relation with water system, etc.)
- to compare the different methods of initial and residual risk assessments, which will be finally the basis for harmonisation of methods and to define the most suitable methods (and as far as possible

standardized) for the assessment of the success of remediation including assessment of environmental performances of GRO

- to compare the quantity and quality of obtained plant biomasses, to evaluate their potential uses through various processes resulting in financial returns, including an energy balance and a C balance aiming at evaluating the C credits for GRO sustainability, (ecological returns) and
- to further enhance the GRO efficiency by biotechnological means (plant breeding, clone selection, isolation and characterisation of rhizosphere microbes, etc.)
- to evaluate environmental and socio-economic side effects.
- to publish a best practice guide and a decision support tool (DST) enabling companies to apply and stakeholders to decide for GRO

Materials and Methods

1. Appraisal of GRO options: The efficiency and effectiveness of different GRO technologies, inluding phytoextraction, (aided) phytostabilisation and phytoexclusion, are compared based on 17 case studies established on 13 long-term European case studies. The experiments cover different conditions regarding the pollution level, climatic conditions as well as the remediation time (3-19 years).

Table 1: Long-term field experiments in the Greenland project

No.	Experimental coordinator	Strategy and gentle remediation technology	Plant species	Contaminants	Site type	Duration (2014)
1	UHASSELT	phytoextraction using SRC and crops	poplars, willows, maize, rapeseed	Cd, Zn	agricultural soils	8 yrs
2	SLU	phytoextraction using SRC	willows	Cd, Zn (Cu, Ni, Cr, Pb)	Commercial sludge-amended fields	19 yrs / 8 yrs
3	INERIS	phytoextraction using HA and high biomass crop	*Arabidopsis halleri* rapeseed	Cd, Zn, Pb	Marginal lands in surrounding industrial facility	6 yrs
4	CSIC	phytoextraction using HA and SRC	*Noccaea caerulescens,* willows	Cd, Zn	tailings	4 yr
5	INRA	aided phytoextraction using high biomass crops from fast-track breeding	sunflower, tobacco sorghum	Cu and Cu/PAHs	industrial soils	6 yrs
6	LfULG	phytoextraction using SRC	poplars, willows	Cd, As, Pb	agricultural soils	8 yrs
7	PT-F	phytoextraction using high biomass crop from fast-track breeding	sunflower, tobacco	Cd, Zn (Cu, Ni, Cr, Pb)	sludged and agricultural soils on landfill	8 yrs
8	INRA	phytostabilisation and rhizodegradation (SRC and grassy cover)	Poplars, willows, grasses, vetiver	Cu and Cu/PAHs	industrial soils	8 yrs
9	CSIC	phytostabilisation	Tobacco, willows	Cu	tailings	4 yr
10	INERIS	aided phytostabilisation	Miscanthus, spontaneous grasses, shrubs, trees	Cd, Zn, Pb, As, Cu	dredged sediments	3 yr
11	AIT	in situ stabilization/phytoexclusion	barley, maize	Cd, Pb, Zn, (As, Cu)	agricultural soils	11 yrs
12	IUNG	in situ stabilization (lime, sludges)/phytoexclusion	grassland	Cd, Zn, Pb	Post-industrial soils	18 yrs
13	LfULG	in situ stabilization/phytoexclusion	crops, grassland	Cd, As, Pb	agricultural soils	8 yrs

2. Biomass valorisation options: There are three main tasks focussing on this topic: a) literature, report and data collection on existing and on-going valorization processes regarding biomass and, in international and national regulation state of the art for valorization processes; b) Interviews of companies using non-food biomass (e.g. to produce bioenergy, biofuel, fiber timber, technosoil and compost) are planned to identify the potential limitations (technical, economical, and regulatory) related to the admittance of plant biomass cultivated on contaminated lands in current (and forthcoming) plants and facilities; c) several options of

biomass valorization were tested using biomass obtained from different case studies listed in Table 1: combustion, anaerobic digestion, solvolysis, and microwave thermal treatment.

3. Harmonisation of methods used for GRO evaluation: Different methods were compared and evaluated for their reliability to quantify the potentially toxic trace element (TE) fractions among selected case studies and for their dependability as indicators of GRO success. Two test batteries were pre-selected, a chemical one for quantifying TE exposure in untreated soils and soils managed using GROs (i.e. phytoextraction, aided phytostabilisation, and in situ stabilization/phytoexclusion) and a biological one for characterizing soil functionality and ecotoxicity.

4. Increasing the GRO efficiency: Plants and microbes as well as soil amendment materials are tested in various experiments for their potential to increase the GRO efficiency. In addition, agronomic measures are evaluated for their potential to increase the GRO efficiency.

5. Stakeholder engagement and empowerment and development of a decision support tool (DST): Current sector practice in stakeholder engagement and its importance when implementing GRO and other remediation options were reviewed. From this, knowledge gaps were identified, and strategies to promote more effective stakeholder engagement during GRO application are outlined development, using data from selected case studies. In addition, a practical and simple decision support tool (DST) focused on regulators, consultants, site managers and planners was developed.

Results

1. Appraisal of GRO technologies: The case studies listed in Table 1 cover a range of different GRO options under different conditions and for different time periods. Here, the main results of selected case studies are presented:

1a GRO option in situ immobilisation/phytoexclusion - case study Arnoldstein (Austria): Lime powder and a mix of gravel sludge and iron oxide were applied to an arable field moderately polluted by Zn, Cd and Pb. Due to the treatment, the accumulation of these elements in maize was reduced to levels below the Austrian threshold value for fodder. Further reduction in Cd accumulation was achieved by changing to a metal-excluding cultivar of maize (Friesl-Hanl et al., unpublished).

1b GRO option phytoextraction – case study Bettwiesen (Switzerland): Non-GMO mutants of tobacco and sunflower were repeatedly planted and harvested on a Zn-contaminated site. Over the 5 year phytoextraction period a 45 - 70% reduction of the initial labile zinc top soil concentration was found. Without the phytoextraction the soluble top-soil concentration remains almost constant over time (Herzig et al., 2014)

1c GRO option Aided phytostabilisation – case study Piekary: On a site, where metal smelter slags were deposited, 300 t/ha biosolids and 30 t/ha lime has been applied in 1995. Previous results were published by Stuczynski et al. (2007). Continuous monitoring of soil health parameters and metal extractability has shown that water-extractable Zn has decreased from ~350 mg kg^{-1} to < 10 mg kg^{-1} and water-extractable Cd from ~10 mg kg^{-1} to < 0.1 mg kg^{-1}.

Further results of this part are presented by Michel Mench et al. [abstract no. 168]

2. Biomass valorisation:

The results of this part are presented by Valerie Bert et al. [abstract no. 98]

3. Harmonisation of tools:

The best correlations were obtained between NH_4NO_3-, followed by $NaNO_3$-, extractable TE and the ecotoxicological responses. Pseudo-total (*aqua regia*- extractable) concentrations correlated with few biological responses for Cd, but correlations were more frequent for Pb and Zn. Biometrical parameters and biomarkers of dwarf beans were the most responsive indicators for the soil treatments and changes in soil TE exposures. Plant growth was inhibited at the higher extractable TE concentrations, while plant stress enzyme activities augmented with increasing TE extractability. Based on these results, a minimum risk assessment battery to compare/biomonitor the sites phytomanaged by GROs might consist of the NH4NO3 extraction and the bean Plantox test including the stress enzyme activities. The results have been summarized in a paper submitted to *Science of the Total Environment* (Kumpiene et al., unpublished)

4. Improvement of GRO technologies:

a) Screening of mutant lines of tobacco and sunflowers in Bettwiesen (CH), Biogeco (FR), Lommel (BE), Touro (ES) and Pedrafita do Cebreiro (ES) (see Table 1): For tobacco, the highest biomass production was obtained in the Bettwiesen (CH) and Lommel (BE) sites, while the lowest biomass production was found in the mine tailings of the Piedrafita (ES) site. Zinc extraction potential was generally highest for Bettwiesen (ranging from 1243 – 3858 g/ha/yr), while Cd extraction was higher at the Lommel site (with NBCu 10-8 F2). In general, mutant tobacco lines showed a higher extraction potential than the motherline, and this was most pronounced at the Bettwiesen site. Regarding the sunflower mutant lines, biomass production was generally higher in mutant lines than the motherclone. Zn extraction potential was highest at both the Bettwiesen and Piedrafita sites, and Cd extraction was highest in Piedrafita.
b) Use of microbial inoculants for improving plant performance and/or phytoremediation potential: A description of bacterial collections obtained by different Greenland partners has been prepared. Here, the results of inoculation trials (principally in pot experiments) using different strains from these collections were presented. Microbial inoculants aim to improve plant growth and establishment, and/or modify soil metal bioavailability and plant uptake.
c) Use of amendments for reducing TE bioavailability: Ten amendments were evaluated on an individual basis and/or in various combinations. The effect of amendments on plant growth, trace metal availability and leaching, and soil bio- and physico-chemical properties was evaluated. Plant yield generally increased with all amendments compared to controls. Yields were also greater at the second harvest than the first harvest, and in those treatments which included organic amendments (e.g. drinking water residue or compost). When amendments were combined with an organic material this also led to a significant increase in biomass compared to that produced with amendments added individually. Metal (Cd, Pb and Zn) availability generally decreased with time and was reduced by all the tested amendments compared to untreated soils. Reductions in metal bioavailability were more pronounced after the incorporation of combinations of amendment.
d) Evaluation of agronomic measures to improve GRO efficiency: various agronomic measures (including soil and crop management practices, fertilization, weed control, planting methods, harvesting management, irrigation, etc.) were evaluated and compiled in a manuscript, which will be submitted to *International Journal of Phytoremediation* (Kidd et al., unpublished)

5. Stakeholder engagement and empowerment and development of a decision support tool (DST):

Effective stakeholder involvement has been identified as a key requirement for the optimal application of sustainable remediation strategies (CL:AIRE, 2011). Stakeholder engagement when remediating land for soft end-use (where the soil remains unsealed, e.g. a community parkland), is perhaps more wide ranging and more complex than in many other remediation fields. Consequently, stakeholder engagement is likely to be critical to the acceptance of GROs, particularly for larger projects, because GROs are most likely to be used for sites where a soft end use is envisaged, and the biological component of the remediation (e.g. plant cover) is likely to be an enduring part of the overall regeneration of the land. The aims of stakeholder engagement are building up and maintaining an open and constructive relationship with stakeholders and thereby facilitating a project's management of its operations, including its environmental and social effects and risks (World Bank, 2012). Effective stakeholder engagement is also seen as key in reducing remediation project risks.
Stakeholder engagement associated with GRO application largely involves liaison with the site owner and the relevant regulatory authority, and educational and research/proof-of-concept activities. At these sites, stakeholder activities are dominated by communication and agreement rather than collaboration and empowerment (Fig. 1).

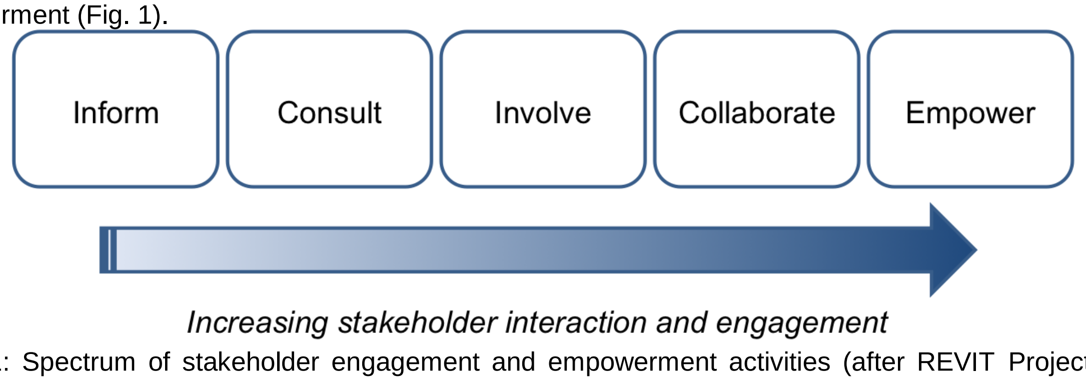

Figure 1: Spectrum of stakeholder engagement and empowerment activities (after REVIT Project, 2007). Note that the lower arrow shows direction of increasing depth of stakeholder interaction and engagement and is not intended to show a "flow" of activities, as stakeholder engagement activities are frequently iterative and non-linear in nature and may involve the full range of these involvement measures (reprinted from Cundy et al., 2013)

Stakeholder engagement in GRO application has been exemplified for two GREENLAND case studies. One of them, the BIOGECO site, is maintained by INRA. The purpose of the BIOGECO phytoremediation platform was to act as a demonstrator site highlighting to stakeholders the benefits and limitations of GRO at this and similar sites through application of phytostabilisation, aided phytostabilisation (combining in situ stabilisation and phytostabilisation), (aided) phytoextraction, and PAH phytodegradation. The need for a demonstrator site was partly driven by certain stakeholders, such as local and regional government, questioning the potential of GRO as practical site remediation tools. Initial stakeholder engagement was established with the local land owner. Later, regulatory and financing bodies (Direction Régionale de l'Industrie, de la Recherche et de l'Environnement (DRIRE, now renamed DREAL) in Aquitaine (regulation), Agence de l'Environnement et de la Maîtrise de l'Energie (ADEME), and the Aquitaine Region Council.
Finally, the following research needs were identified:

1. Integrating (sustained and long-term) stakeholder engagement strategies into decision support systems and tools for GRO (to raise the profile of the benefits of effective stakeholder engagement and participation, particularly with sector professionals)
2. Developing criteria for the identification of different stakeholders profiles/categories - their expectations, influence, characteristics, preferred approaches to engagement and levels of engagement.

The instructions for implementing GRO as a practical site solution will be compiled in a best practice guidance document. This book will include evaluation of implementation cost, remediation success / efficiency in terms of risk reduction, overall environmental and socio-economic benefits and impacts of the large-scale trials, recommendation of harmonized monitoring and analytical / risk assessment methods and analysis of the role of recent developments in biotechnology as well as options for biomass utilisation. The best practice guidance document is planned to have the following structure:

1. Definitions and context – what is GRO and how does it work?
2. Overview of current state of development and risk management capability
3. Case / success stories
4. Potential economic, environmental and social benefits
5. Operating windows for GRO
6. Further information sources

Appendices:
Appendix 1: Stakeholder engagement guidelines
Appendix 2: Cultivars and amendments
Appendix 3: Safe biomass usage
Appendix 4: Indicators of success and methods
Appendix 5: DST and cost-calculator

The delivered document is planned to be multi-lingual. The guidance will focus on practical rather than technical detail and will cross-reference the project DST and success stories / demonstrator sites (i.e. the Greenland sites, but also including sites where GRO has been applied as part of mixed site development e.g. reedbeds, urban parkland). The appendices will give more detail and refer the user on to more detailed scientific publications for support / validation.

A working GRO-focused DST has been developed, following the "tiered" (or layered) model proposed in the earlier ERA-NET SUMATECS project and in Onwubuya et al. (2009), initially within the framework of the UK Model Procedures for the Management of Land Contamination (CLR11). Initial research underpinning this DST development has included validation and testing of this outline model using 3 case studies (sites in east London, Lommel, and the Biogeco platform), the latter two drawn from the Greenland test sites. Following this testing and evaluation of the outline model produced the project DST an MS Excel-based model has been developed. The model is designed to build in complexity and time effort (and technical detail) through its 3 phases, and will embed the best practice guidance document and the stakeholder engagement principles and identification criteria. Phase 1 will include definitions, "success stories" of GRO, and a simple contaminant matrix designed to simply and effectively identify the potential feasibility of GRO at a particular site. Phase 2 will include modules on sustainability assessment (drawing on published SuRF-UK sustainability indicators, www.claire.co.uk), stakeholder engagement (including stakeholder engagement principles, and stakeholder identification criteria), and economic assessment (via a simplified cost calculator, to identify the economic value proposition of GRO) to further support the decisional process.
In the economic assessment module, the simplified cost calculator is planned to be a MS Excel-based programme where the user enters local data (e.g. land cost, transport costs, location (remote rural, urban) etc) to support a cost calculation, supported by data from the Greenland sites to "calibrate" the model and give economic value examples. Additionally, a patent landscape for phytoremediation is being developed,

and will be included in this module of the DST. This is important for strategic matters regarding the introduction of an innovative technology such as phytoremediation.
Phase 3 of the model will provide a technical assessment of GRO, and will incorporate the appendices from the best practice guidance, to help the user identify “operating windows” for GRO.
Importantly, the DST will also cross-link with and complement work on decision support being undertaken in the EU HOMBRE project (www.zerobrownfields.eu). A complementary role for each DST was identified, with the Greenland DST focused on options appraisal, while HOMBRE takes a wider perspective at an earlier decision stage in site design for soft end-use and assessment of wider site “services”.

Expected final results and their potential impacts

The main aim of GREENLAND is to optimize the applicability of gentle soil remediation options (GRO) in the field and to make them fit for practical application. This includes technical issues on the one hand (use of biomass, evaluation of GRO efficiency, biotechnological improvements), but also regulatory issues as well as stakeholder engagement and empowerment. At the end of GREENLAND, the efficiency of GRO under various conditions (different pollution profiles and level, climate, soil conditions) will have been demonstrated. With similar importance, also the beneficial socio-economic impacts (profit from biomass valorisation, improvement of land value) will be shown. Both technological and socio-economic benefits will be important prerequisites for practical application. A decision support tool developed by GREENLAND will allow stakeholders to take a decision for GRO and a practical handbook will facilitate the implementation as a practical site solution. Overall, there will be a substantial improvement of soil quality and socio-economic conditions at the local level (land owners, communities), but also on the European level, since soil contamination is still one of the major threats for European soils. Soil is a non-renewable natural resource, which is constantly declining worldwide, but also in Europe, mainly due to the threats listed in the proposal for a Soil Framework Directive (COM(2006) 232). GRO will contribute to improve the natural basis for agricultural production (food and non-food crops) and bring marginal land back into productivity. The transfer of pollutants into the human food chain will be reduced due to GRO application.
During the second reporting period of the GREENLAND project, the fundamental basis for achieving the overall technical aims has been successfully extended and already partly completed. All milestones have been achieved and deliverables have been finalised. In the last reporting period, the major output, including the decision support tool and the handbook, will be prepared, finalised and published.

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We dedicate this paper to our friend/colleague Dr Anne Loppinet-Serani in memoriam.

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