



Volume :2, Issue :4, 574-579  
 April 2015  
 www.allsubjectjournal.com  
 e-ISSN: 2349-4182  
 p-ISSN: 2349-5979  
 Impact Factor: 3.762

**Lee Fergusson**  
 Principal Researcher, Prana  
 World Consulting Pty Ltd,  
 Oxenford, Queensland,  
 Australia

## Soil washing, ozofractionation and metal sequestration: removing organic and inorganic species from contaminated soil and water

**Lee Fergusson**

### Abstract

Most research of soil washing conducted around the world has involved the removal of inorganic species from contaminated soil matrices. Few have considered its potential for removing organic species, although it has been used to remove volatile organic compounds and diesel. Similarly, a significant amount of research has considered the roles of advanced oxidation and ozonation in destroying organic species in water, but few have examined the potential of ozofractionation to perform this role.

The purpose of this study was to assess the capacity of soil washing to remove both organic and inorganic species from a complex contaminated industrial soil, and once contaminants had been liberated from the solid phase, examine whether ozofractionation destroyed organic and inorganic species and whether a chemical reagent sequestered inorganic species in the flushing solution.

Findings suggest that soil washing had a salutary effect on liberating organic and inorganic species from contaminated soil, reducing total petroleum hydrocarbons, for example, from 1,290 mg/kg to 320 mg/kg. The study also found that ozofractionation destroyed organic and inorganic species, for example reducing polycyclic aromatic hydrocarbons from 13.2 mg/L to <0.5 mg/L and cyanide from 5.9 mg/L to 0.02 mg/L, and reagents sequestered heavy metals in the flushing solution.

**Keywords:** soil washing, surfactant, flushing solution, ozofractionation, sequestration

### 1. Introduction

Washing contaminated soil with freshwater in agitated mixers to liberate inorganic species, such as lead and zinc, has been widely documented in the last 20 years (Abumaizar & Smith, 1999, Dermont *et al.*, 2008; Mann, 1999); this practice is particularly common in mine site remediation (e.g., Moutsatsou *et al.*, 2006). Soil washing is an *ex-situ* remediation process which involves washing contaminated soil in an agitator then settling treated solids from the contaminated rinse water (the so-called “flushing solution”), and has become more widespread in commercial remediation applications in the last ten years, particularly in Europe where the greatest expertise resides.

Soil washing is based on the principle that a contaminant will bind to the soil’s fine particles, which in turn bind to coarser particles, and thus by separating the coarse fraction from the fine fraction using agitated fresh water, contaminants along with the finest soil fraction can be separated from the soil (a process called “soil partitioning”), thereby liberating contaminants from the coarse fraction. This process then allows the separated contaminated flushing solution to be treated and discharged, leaving behind a remediated coarse soil fraction which can be reused as backfill or for other purposes. Some soil washing techniques also use a surfactant or other chemical additive to aid in the liberation of contaminants from soil (Chu & Chan, 2003; Torres *et al.*, 2012), and some deploy novel agitation mechanisms, including jet reactions and ultrasonic washing (e.g., Feng *et al.*, 2001).

However, the vast majority of soil washing applications are designed to liberate inorganic contaminants from soil, usually heavy metals (Gusiatin & Klimiuk, 2012; Yang *et al.*, 2012); few are used to liberate both organic and inorganic contaminants, and when such techniques are applied to remove organic contaminants the process is designed specifically to target one or two contaminant types, such as diesel fuel or volatile organic compounds (e.g., Dermont *et al.*, 2008; Semer & Reddy, 1996), rather than simultaneously targeting different contaminant types.

In wastewater treatment, a wide variety of physical, chemical and biological technologies have been developed and deployed to destroy or remove organic and inorganic contaminants (European Commission, 2012). Under the general category of “oxidation” for example, some systems use advanced oxidation processes (AOP) and ozonation (e.g., Gogate & Pandit, 2004;

### Correspondence:

**Lee Fergusson**  
 Principal Researcher, Prana  
 World Consulting Pty Ltd,  
 Oxenford, Queensland  
 Australia

Panizza & Cerisola, 2001); more recently, some have combined soil washing with various forms of oxidation and biological treatment of the flushing solution (Haapea & Tuhkanen, 2006; Villa *et al.*, 2010).

Oxidizing agents include ozone ( $O_3$ ), potassium permanganate ( $KMnO_4$ ), sodium persulfate ( $Na_2S_2O_8$ ), ascorbic acid ( $C_6H_8O_6$ ), Fenton reagents such hydrogen peroxide ( $H_2O_2$ ), or other sources of chemical energy. Irrespective of whether oxygen species are mixed as  $O_2$  in oxidation or AOP systems or injected as ozone, they are designed to destroy organic compounds, including such measures as chemical oxygen demand (COD), via chemical degradation or reduction reactions.

Most oxidation processes utilize aeration with atmospheric oxygen or oxygen injection (such as those related to biological aeration in bioreactors), but some use chemical reactions produced by hydroxyl radicals, such as those generated by potassium permanganate and hydrogen peroxide; these reactions are generally short-lived and sometimes violent in nature. For this reason, reaction vessels must be designed to accommodate short bursts of high temperature and safety of operators and out-gassing are important concerns. While ozonation has mostly been used to breakdown organic contaminant species in wastewater (e.g., Peña *et al.*, 2003), it has in the past also been applied in conjunction with other chemical additives to treat heavy metals in acid mine drainage (Department of Applied Science, 1970).

A less understood oxidising process in wastewater treatment is ozofractionation, most commonly associated with purifying water in aquariums. Like ozonation, in industrial applications ozofractionation injects ozone into the liquid waste stream but with one key process difference: where standard ozonation “bubbles” ozone into the liquid waste stream, ozofractionation injects it into a foam fractionator.

Where simple oxidation and biological aeration inject atmospheric oxygen into a waste stream in order to create an air/water interface which causes contaminants to oxidise and ozonation bubbles ozone through the liquid stream in order to expose contaminants to oxygen atoms with a larger oxygen/water interface to generate greater oxidising reactions, ozofractionation pumps ozone into a fractionator containing wastewater where millions of tiny bubbles of oxygen are generated (the foam-like bubbles have a diameter  $<150 \mu m$ ). These bubbles attract and oxidise contaminants in a way which is many times more aggressive than simple oxidation or ozonation because of the far greater agitated oxygen/water surface area generated by the fractionator.

The process is further enhanced by the fact that ozone is more soluble in water than either atmospheric air (which is only 20% oxygen) or pure oxygen. While it is not within the scope of this paper to describe all known aeration, AOPs, ozonation and ozofractionation reactions and processes (indeed many chemical reactions in both ozonation and ozofractionation are largely unknown to the scientific community due to difficulties associated with measuring oxidizing reactions which occur within nanoseconds inside closed-system reaction chambers), suffice it here to say that ozofractionation is a worthy candidate for examination in the context of soil washing, particularly as it has been used effectively to destroy complex organic and inorganic compounds, such as pesticides.

For example, this author has demonstrated that ozofractionation can reduce chlorpyrifos ( $C_9H_{11}Cl_3NO_3PS$ ) from  $7.2 \mu g/L$  to  $<0.5 \mu g/L$ , dichlorodiphenyltrichloroethane

(better known as DDT,  $C_{14}H_9Cl_5$ ) from  $108 \mu g/L$  to  $<2.0 \mu g/L$ , and DDT breakdown products dichlorodiphenyldichloroethylene (DDE,  $C_{14}H_8Cl_4$ ) from  $9.5 \mu g/L$  to  $<0.5 \mu g/L$  and dichlorodiphenyldichloroethane (DDD,  $C_{14}H_{10}Cl_4$ ) from  $15.2 \mu g/L$  to  $<0.5 \mu g/L$  in industrial wastewater at a pesticide and fertilizer plant (Fergusson, in press); an Australian researcher has also been granted a U.S. patent to use ozofractionation in combination with chemical additives to treat acid mine drainage and other industrial wastewater (Dickson, 2014).

Not unlike oxidation processes, there are a great many filtration systems (e.g., ion exchange and granulated activated carbon), flotation systems (e.g., dissolved air flotation and induced gas flotation), and direct chemical additions (e.g., magnesium oxide ( $MgO$ ), calcium hydroxide ( $Ca(OH)_2$ ) and sodium hydroxide ( $NaOH$ )) used throughout industry to reduce or remove inorganic species from wastewater. These have been well documented over many years (Andriamirado *et al.*, 2007; Spellman, 2008). Some of these methods immobilize inorganic contaminants, usually metals, in the settled (or floated in the case of froth flotation) solid stream, usually as a semi-dry filtrate or centrate, which is subsequently disposed to landfill. However, immobilized or de-mineralized metals can readily release back into the environment under acidifying and reducing conditions, such as those that exist in landfills; sludges generated from immobilizing agents are chemically unstable because the ionic bonds which initially adsorb metals onto the reactive surface of reagents become weaker over time (Fergusson, 2009). The chemicals typically used in immobilizing technologies are generally water soluble, leading to higher volumes of sludge due to higher moisture contents increasing cost of disposal.

Metals sequestration is also used to treat wastewater via direct chemical addition, but the process does not result in bioavailable metals or higher volumes of sludge; in fact, metals sequestered in these type of sludges and sediments become more tightly bound as time passes (to use the colloquial language of criminology, immobilization “arrests” the contaminant whereas sequestration “locks them up”) and sludges contain less moisture due to the insoluble nature of sequestering chemicals (Fergusson, 2009).

The reagents used in metals sequestration are derived from a modified form of alumina refinery residue and contain a complex cocktail of metals and minerals, including hematite ( $Fe_2O_3$ ), boehmite ( $\gamma-AlOOH$ ), gibbsite ( $Al(OH)_3$ ) and sodalite ( $Na_4Al_3Si_3O_{12}Cl$ ), anatase ( $TiO_2$ ), argonite ( $CaCO_3$ ), brucite ( $Mg(OH)_2$ ), diasporite ( $\beta-Al_2O_3 \cdot H_2O$ ), ferrihydrite ( $Fe_5O_7(OH) \cdot 4H_2O$ ), gypsum ( $CaSO_4 \cdot 2H_2O$ ), hydrocalumite ( $Ca_2Al(OH)_7 \cdot 3H_2O$ ), hydrotalcite ( $Mg_6Al_2CO_3(OH)_{16} \cdot 4H_2O$ ), and p-aluminohydrocalcite ( $CaAl_2[CO_3]_2(OH)_4 \cdot 3H_2O$ ).

Of significance in these formulae is the presence of abundant hydroxides and oxyhydroxide compounds, which contribute to the acid neutralizing capacity of reagents, as well as the positively charge iron-, aluminium-, magnesium- and titanium-based molecules which not only adsorb metals but also lead to precipitation and isomorphic substitution reactions; these reactions are largely responsible for the long-term sequestration phenomena of inorganic species described above. Examples of how metals sequestration and oxidation work synergistically in environmental remediation have been discussed in detail elsewhere (Fergusson, 2009; 2012).

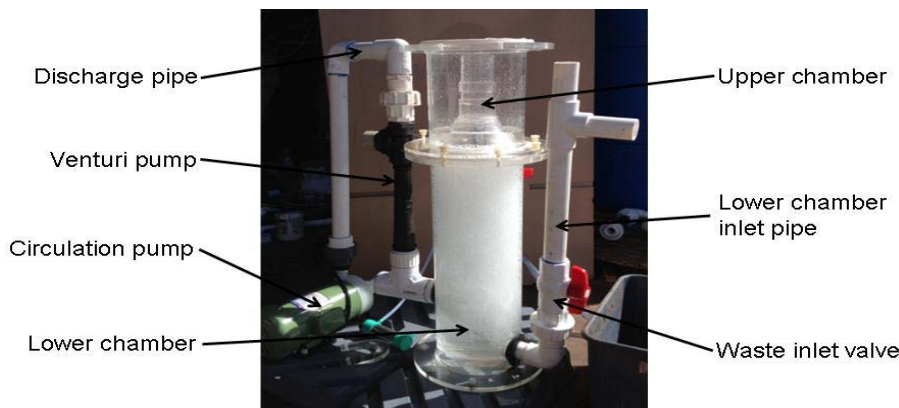
This overview of soil washing, ozofractionation and metals sequestration leads to the present study, which asks the following three research questions: 1) does a standard soil

washing technique using a surfactant remove a complex range of organic and inorganic species from contaminated soil; 2) does ozofractionation destroy organic species in the resultant flushing solution; and 3) does metal sequestration bind and reduce metals in a post-ozofractionation flushing solution?

**Method**

To answer these research questions, 0.5 m<sup>3</sup> (about 750 kg) of contaminated soil was excavated from a derelict industrial site in Adelaide, South Australia. The sample was assessed

for the following analytes prior to intervention: pH; alkanic hydrocarbon fractions C6-C9, C10-C14, C15-C28, C29-C36 and total petroleum hydrocarbons (TPH); cyanide (CN); aromatic hydrocarbons acenaphthene; benzo(a)pyrene (B(a)P); benzo(g,h,i)perylene; fluoranthene, naphthalene, pyrene, and total polycyclic aromatic hydrocarbons (PAH); total copper, lead, nickel and zinc; and leachable copper, lead, nickel, and zinc. The sample was divided into 30 x 25 kg sub-samples. Each sub-sample was processed using the following three-stage intervention: 1) soil washing; 2) ozofractionation; and 3) metals sequestration.



**Fig 1:** Description of the process units used in ozofractionation.

Stage 1 followed these steps: place a sub-sample in a concrete mixer; add 50 L of fresh water; add 50 mL of sodium lauryl sulfate (CH<sub>3</sub>(CH<sub>2</sub>)<sub>11</sub>OSO<sub>3</sub>Na) as an anionic surfactant; mix for 20 minutes; decant flushing solution and store; repeat for each sub-sample; retain and sample soil; homogenize sub-samples and analyze. Stage 2 followed these steps: pump flushing solution into an ozofractionator with a retention time of approximately one hour (Figure 1 shows the basic configuration of the ozofractionator, which had a 20 L holding capacity; this image does not show the ozone generator, a small electrical unit generating ozone by coroner discharge); release liquid and retain flushing solution; sample and analyze.

Stage 3 followed these steps: in a mixing tank, add 3.0 g/L of sequestering reagent<sup>1</sup> to flushing solution; mix for 10 minutes; allow solids to settle for one hour; decant flushing solution; sample and analyze. Representative samples of soil

were collected after stage 1, and samples of water were collected after stages 2 and 3; these samples were sent to a certified laboratory within 24 hours of collection and tested for the above analytes using either gas chromatography mass spectrometry, inductively coupled plasma mass spectrometry, or APHA 4500 CN-L, as appropriate.

**Results**

Analytical results from stage 1 soil washing are shown in Table 1. This data indicate that pH remained unchanged as a result of soil washing, and while there were no alkanic hydrocarbons present in the C6-C9 and C10-C14 fractions, hydrocarbons in the C15-C28 and C29-C36 fractions totalled 1,290 mg/kg, which were reduced by 80% to 260 mg/kg as a result of soil washing. Similarly, cyanide was reduced by 87% from 0.03 mg/kg to 0.004 mg/kg.

**Table 1:** Result of washing organic and inorganic species from contaminated soil

Parameter	Soil Before Soil Washing	Soil After Soil Washing	Percent Reduction (%)	Detection Limit
<i>pH</i>	8.0	8.3	0	—
<i>C6-C9 fraction (mg/kg)</i>	<10	<10	0	10
<i>C10-C14 fraction (mg/kg)</i>	<50	<50	0	50
<i>C15-C28 fraction (mg/kg)</i>	750	150	80	100
<i>C29-C36 fraction (mg/kg)</i>	540	110	80	100
<i>TPH (mg/kg)</i>	1,290	260	80	50
<i>Cyanide (mg/kg)</i>	0.03	0.004	87	0.004
<i>Acenaphthene (mg/kg)</i>	1.9	0.8	58	1.0
<i>Benzo(a)pyrene (mg/kg)</i>	3.6	2.0	45	0.5
<i>Benzo(g,h,i)perylene (mg/kg)</i>	1.6	0.8	50	1.0
<i>Fluoranthene (mg/kg)</i>	6.4	6.3	2	1.0
<i>Naphthalene (mg/kg)</i>	0.8	0.05	94	0.5
<i>Pyrene (mg/kg)</i>	7.7	6.2	20	1.0
<i>Total PAH (mg/kg)</i>	41	32	22	1.0
<i>Total Copper (mg/kg)</i>	774	24	97	5.0
<i>Total Lead (mg/kg)</i>	470	—	—	5.0
<i>Total Nickel (mg/kg)</i>	71	8.0	89	2.0
<i>Total Zinc (mg/kg)</i>	933	242	74	5.0

<i>Leachable Copper (mg/L)</i>	30	6.6	78	0.001
<i>Leachable Lead (mg/L)</i>	0.7	0.1	86	0.001
<i>Leachable Nickel (mg/L)</i>	0.2	0.05	75	0.001
<i>Leachable Zinc (mg/L)</i>	10.6	2.7	75	0.005
<i>Total average organic species change</i>	—	—	53	—
<i>Total average inorganic species change</i>	—	—	83	—
<i>Total average change</i>	—	—	66	—

The presence of a range of aromatic hydrocarbons indicated the soil was complex, however, soil washing reduced acenaphthene, benzo(a)pyrene, benzo(g,h,i)perylene, naphthalene, pyrene, and total PAH by an average of 42%, although fluoranthene was unaffected. Total metals were reduced by 87%, with total Cu reduced by 97% and total nickel by 89%; leachable metals after soil washing were reduced by an average of 79%. The total average organic species change was 53% and the total average inorganic species change was 83%, for a total average change of 66%. From these data it was concluded that research question 1 can tentatively be answered in the affirmative, although reductions of PAH were unconvincing. It is also unclear what role, if any, sodium lauryl sulfate played in these outcomes. While the reduction in naphthalene was significant at 94%, that of B(a)P was only 45%; given that B(a)P in particular is

a contaminant of concern (in addition to being found in industrial coal tar wastes, cooked meats, and tobacco, B(a)P is also a known carcinogen, Jiang *et al.*, 2007), this result was unimpressive.

Results of stage 2 ozofractionation are shown in Table 2. Consistent with the results for stage 1, there were no alkanic hydrocarbon C6-C9 and C10-C14 fractions present in the flushing solution, but the C15-C28 and C29-C36 fractions were reduced by 100% and 73% respectively as a result of ozofractionation. Similarly, cyanide was reduced by 100% from 5.9 mg/L to 0.02 mg/L, and all aromatic hydrocarbons were completely destroyed by ozofractionation. The total average change in all organic and inorganic species before and after ozofractionation was 97%. From these data it can be concluded that research question 2 was answered in the affirmative.

**Table 2:** Result of ozofractionating organic and inorganic species in flushing solution

Parameter	Flushing Solution Before Ozofractionation	Flushing Solution After Ozofractionation	Percent Reduction (%)	Detection Limit
<i>C6-C9 fraction (mg/L)</i>	<10	<10	0	10
<i>C10-C14 fraction (mg/L)</i>	<50	<50	0	50
<i>C15-C28 fraction (mg/L)</i>	420	<100	100	100
<i>C29-C36 fraction (mg/L)</i>	670	160	73	100
<i>TPH (mg/L)</i>	1,090	160	86	50
<i>Cyanide (mg/L)</i>	5.9	0.02	100	0.004
<i>Acenaphthene (mg/L)</i>	<1.0	<1.0	0	1.0
<i>Benzo(a)pyrene (mg/L)</i>	2.1	<0.5	100	0.5
<i>Benzo(g,h,i)perylene (mg/L)</i>	1.3	<0.5	100	0.5
<i>Benz(a)anthracene (mg/L)</i>	1.3	<0.5	100	0.5
<i>Benzo(b)fluoranthene (mg/L)</i>	1.2	<0.5	100	0.5
<i>Chrysene (mg/L)</i>	1.1	<0.5	100	0.5
<i>Fluoranthene (mg/L)</i>	2.0	<0.5	100	0.5
<i>Indeno(1.2.3.cd)pyrene (mg/L)</i>	1.0	<0.5	100	0.5
<i>Naphthalene (mg/L)</i>	<1.0	<1.0	0	1.0
<i>Pyrene (mg/L)</i>	2.9	<0.5	100	0.5
<i>Total PAH (mg/L)</i>	13.2	<0.5	100	<0.5
<i>Total average change</i>	—	—	97	—

Results for stage 3 metals sequestration are presented in Table 3. It should be noted that some inorganic species may have been removed rather than destroyed by stage 2 ozofractionation. These results indicate that pH and total arsenic concentration in the flushing solution did not change as a result of chemical addition (lead has not been averaged due to it being at detection level prior to treatment). However, cadmium was reduced by 100% from 0.007 mg/L to below the detection limit, copper was reduced by 72% from 0.07 mg/L to 0.02 mg/L, molybdenum was reduced by

57% from 0.23 mg/L to 0.01 mg/L, nickel was reduced by 30% from 0.01 mg/L to 0.007 mg/L, and zinc was reduced by 100% from 0.08 mg/L to below the detection limit; the total average inorganic species reduction was 60%. It should also be pointed out that concentrations in the flushing solution before metals sequestration were extremely low and would not pose a significant risk in a real-world scenario. From data presented in Table 3 it can be concluded that research question 3 can only be partially answered in the affirmative.

**Table 3:** Result of sequestering inorganic species in flushing solution

Parameter	Flushing Solution Before Metals Sequestration	Flushing Solution After Metals Sequestration	Percent Reduction (%)	Detection Limit
<i>pH</i>	8.0	7.8	0	—
<i>Arsenic (mg/L)</i>	0.003	0.003	0	0.001
<i>Cadmium (mg/L)</i>	0.007	<0.001	100	0.001
<i>Copper (mg/L)</i>	0.07	0.02	72	0.001

<i>Lead (mg/L)</i>	<0.001	<0.001	—	0.001
<i>Molybdenum (mg/L)</i>	0.23	0.01	57	0.001
<i>Nickel (mg/L)</i>	0.01	0.007	30	0.001
<i>Zinc (mg/L)</i>	0.08	<0.005	100	0.005
<i>Total average change</i>	—	—	60	—

## Conclusion

This preliminary study showed that both organic and inorganic contaminants in a complex industrial soil matrix can be reduced by soil washing, and that ozofractionation can destroy organic and inorganic species and metals sequestration can reduce inorganic species in a flushing solution generated by the soil washing process. However, in some cases the results are far from significant, and the viability of these outcomes in a real-world setting will depend on a variety of factors in each jurisdiction, including starting contaminant concentrations, treatment targets, and regulatory limits for contaminated solids and treated wastewater for discharge to the environment.

Impacts associated with and better controls on the role, if any, of anionic surfactant addition rates and type of surfactant in liberating contaminants during soil washing, optimization of soil washing agitation times, optimization of soil:water ratio in the mixer, potential changes to addition rates of chemical reagent or varying sequestering reagents, and other changes to processes and inputs need to be the subject of future research on soil washing, ozofractionation and metals sequestration. These processes may need to be isolated from each other in a more controlled and repeatable research design in order to determine the primary contributors to observed outcomes.

Nevertheless, the present study does provide provisional evidence that these process steps can remove, reduce or destroy organic and inorganic contaminants in soil and water.

## Acknowledgement

The author has received assistance in the preparation of this study from Virotec Pty Ltd in Australia, with whom the author is affiliated; ViroBind reagent is a common law trade mark of Virotec.

## References

- ViroBind reagent, which is composed mostly of modified alumina refinery residue, was supplied for this study by Virotec.
- Abumaizar, R.J. and Smith, E.H. Heavy metal contaminants removal by soil washing. *Journal of Hazardous Materials*, 1999, 70(1-2), 71-86.
- Andriamirado, L., Asensi, D., Ballard, T., Bele, P., Bernard, M., Bourdelot, J., Brunet, J., and Cachot, L. *Water treatment handbook*, (7th edition), Rueil-Malmaison, France, 2007.
- Chu, W., and Chan, K.H. The mechanism of the surfactant-aided soil washing system for hydrophobic and partial hydrophobic organics. *Science of The Total Environment*, 2003, 307(1-3), 83-92.
- Department of Applied Science. *Treatment of acid mine drainage by ozone oxidation*, Brookhaven National Laboratory, U.S. Atomic Energy Commission, for the U.S. Environmental Protection Agency, Water Pollution Control Research Series, 14010FMH12/70, New York. December 1970.
- Dermont, G., Bergeron, M., Mercier, G., and Richer-Lafleche, M. Soil washing for metal removal: A review of physical/chemical technologies and field applications. *Journal of Hazardous Materials*, 2008, 152(1), 1-31.
- Dickson, M.D. *Method for treating industrial waste*. U.S. patent no. US 2014/0190896 A1, publication date July 10, 2014.
- European Commission. *EU wide monitoring survey on waste water treatment plant effluents*, European Commission Joint Research Centre, Institute for Environment and Sustainability, Rome, Italy, 2012.
- Feng, D. Lorenzen, L. Aldrich, C. Maré, P.W. Ex-situ diesel contaminated soil washing with mechanical methods. *Minerals Engineering*, 2001, 14(9), 1093-1100.
- Fergusson, L. *Commercialisation of environmental technologies derived from alumina refinery residues*. Commonwealth Scientific and Industrial Research Organisation (CSIRO), Project ATF-06-3 "Management of Bauxite Residues", Department of Resources, Energy and Tourism (DRET), Commonwealth Government of Australia, Asia-Pacific Partnership on Clean Development and Climate, Canberra, November 2009.
- Fergusson, L. *ViroMine technology: A solution to the world's mining megawaste*, Prana World Publishing, Gold Coast, Australia, 2012.
- Fergusson, L. Pesticides and ozofractionation: An investigation to remove organic and inorganic contaminants from industrial wastewater, *Asian Journal of Chemistry*, in press.
- Gogate, P.R. and Pandit, A.B. A review of imperative technologies for wastewater treatment I: Oxidation technologies at ambient conditions. *Advances in Environmental Research*, 2004, 8(3-4), 501-551.
- Gusiatin, Z.M. and Klimiuk, E. Metal (Cu, Cd and Zn) removal and stabilization during multiple soil washing by saponin. *Chemosphere*, 2012, 86(4), 383-391.
- Haapea, P. and Tuhkanen, T. Integrated treatment of PAH contaminated soil by soil washing, ozonation and biological treatment. *Journal of Hazardous Materials*, 2006, 136(2), 244-250.
- Jiang, H. Gelhaus, S.L., Mangal, D., Harvey, R.G., Blair, I.A., and Penning, T.M. Metabolism of benzo(a)pyrene in human bronchoalveolar H358 cells using liquid chromatography-mass spectrometry. *Chemical Research in Toxicology*, 2007, 20(9), 1331-1341.
- Mann, M.J. Full-scale and pilot-scale soil washing. *Journal of Hazardous Materials*, 1999, 66(1-2), 119-136.
- Moutsatsou, A. Gregou, M. Matsas, D., and Protonotarios, V. Washing as a remediation technology applicable in soils heavily polluted by mining-metallurgical activities. *Chemosphere*, 2006, 63(10), 1632-1640.
- Panizza M, Cerisola G. Removal of organic pollutants from industrial wastewater by electrogenerated Fenton's reagent. *Water Research*, 2001, 35(16), 3987-3992.
- Peña, M., Coca, M., González, G., Rioja, R., and García, M.T. Chemical oxidation of wastewater from molasses fermentation with ozone. *Chemosphere*, 2003, 51, 893-900.
- Semer, R., and Reddy, K.R. Evaluation of soil washing process to remove mixed contaminants from a sandy

- loam. *Journal of Hazardous Materials*, 1996, 45(1), 45-57.
- 22 Spellman, F.R. *Handbook of water and wastewater treatment plant operations*, 2nd Edition, CRC Press, Boca Raton, FL, 2008.
- 23 Torres, L.G. Lopez, R.B., and Beltran, M. Removal of As, Cd, Cu, Ni, Pb, and Zn from a highly contaminated industrial soil using surfactant enhanced soil washing. *Physics and Chemistry of the Earth*, 2012, 37-39(A/B/C), 30-36.
- 24 Villa, R.D., Trovó, A.G., and Pupo Nogueira, R.F. Soil remediation using a coupled process: Soil washing with surfactant followed by photo-Fenton oxidation. *Journal of Hazardous Materials*, 2010, 174(1-3), 770-775.
- 25 Yang, Z., Zhang, S., Liao, Y., Li, Q., Wu, B., and Wu, R. Remediation of heavy metal contamination in calcareous soil by washing with reagents: A column washing. *Procedia Environmental Sciences*, 2012, 16, 778-785.