Environ Earth Sci (2015) 73:7157–7164
DOI 10.1007/s12665-014-3895-0

ORIGINAL ARTICLE

Quarry rehabilitation employing treated residual sludge from dimension stone working plant

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Received: 31 July 2014 / Accepted: 15 November 2014 / Published online: 25 November 2014

Abstract Waste from stone and aggregate quarrying industry represents a serious environmental and economic problem in view of the difficulties related to its disposal, especially of the finest fraction. Although some attempts have been made to investigate possible reuse of these materials, little is known about their potential as components of a cultivation substrate. Their low physical and chemical fertility require the mixing with other materials to improve the general properties. The aim of this study was to evaluate if such a prospect product can be employed within the quarry for its environmental rehabilitation. Samples from gangue saw with abrasive shot (GSS), from diamond frame saw (DSS), and mixed sludge (MS)—from gangue and DSS—were collected and mixed with compost, green manure, and soil material. The resulting mixtures were further composted, distributed on parcels within the quarry area and sowed. The original materials and the mixtures were analyzed for metals and hydrocarbons (TPH) and for their phytotoxicity. The parcels were sampled and analyzed after 8 years. The results show that mixing with foreign materials can improve the overall quality and fertility of the sludge and that the mixture is not phytotoxic. Most metals concentrations decreased on mixing and further diminished after 8 years. TPH content was drastically reduced and fertility parameters tend toward equilibrium. This indicates that the residual sludge can be employed for quarrying rehabilitation after improvement of its fertility and of its environmental quality.

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Keywords Residual sludge · Stone industry · Quarry remediation · Cultivation substrate · Italy

Introduction

Quarrying is a very important industry in Italy especially for dimension stones and aggregates. The overall turnover of the production chain (quarries, working plants, laboratories, shops, etc) was nearly 40 billion € in 2011, almost 2 % of the Italian Gross Domestic Product (Ceruti 2013). In addition, Italian dimension stone exploitation during 2012 shows an export increment of 9.8 % from 2011 and an import decrement of −6 % (from 2011 to 2012). These values show the importance of quarrying industry which is, at present, characterized by an incertitude due to global crisis in the stone sector (Anonymous 2013).

Exploitation activities require an obligatory phase of environmental rehabilitation of both dumps and quarrying areas: the soil stripped during exploitation phases has to be separated from the waste rocks, and it should be saved for the subsequent vegetation re-establishment in the quarry area (Neri and Sanchez 2010). Topsoil from external sources is often employed for the purpose, however, due to the lengthy duration of the quarrying activities or whenever old abandoned quarries have to be remediated (Brodckom 2001; Milgrom 2008) and this results in an additional cost. On the other hand, quarries and working plants produce huge amounts of waste that has a potential for recovery (Luodes et al. 2012). The most difficult to recover is the residual sludge (EWC code 01 04 13—waste from stone cutting and sawing), a very fine material that can be used either as a filler for land rehabilitation or as a feeding material for different industrial uses, or dumped (Ministero dell'Ambiente 2006; Dino et al. 2013). The current cost connected to sludge landfilling may represent more than 3 % of the operating costs of dimensional stone working plants so there is an increasing interest into its recovery for environmental restoration.

The possibility of employing waste materials in quarry rehabilitation has been investigated by several authors. Bending et al. (1999) offered an overview of the possibility of using various materials for the rehabilitation of quarries. Sort and Alcañiz (1996) had studied the effects of sewage sludge additions to control erosion in limestone quarries. They reported a positive effect on soil physical properties in general and on soil loss in the plots treated with the sludge. More recently, the effects of applying urban wastes to quarries in arid environments were investigated by Castillejo and Castelló (2010). On the contrary, Bonoli and Dall'Ara (2012) observed negative effects in the environmental restoration of a former quarry by paper sludge with production of unwanted biogas.

Burragato et al. (1999) investigated the possibility of employing a mud from siliceous sand treatment for environmental rehabilitation. They observed that the size of mud particles and the lack of pores were conducive to progressive compaction and rendered the sludge virtually impermeable. This may represent a threat for the soil surface erosion and its permeability, and ultimately may increase the risk of landslides. Therefore, the physical and chemical structure of the sludge matrix should be properly adjusted. To this aim, Barrientos et al. (2010) advocate the use of granite sawdust as a soil surface sealant in view of its physical properties. Similarly, a recent paper by Sivrikaya et al. (2014) illustrated the effectiveness of stone waste in improving the physical quality of clayey soils. A direct agricultural application of such materials is seemingly hindered by their low chemical and physical fertility but they could be treated and recovered to obtain a cultivation substrate. If used in quarry rehabilitation the *cradle-to-cradle* principle would be respected as all the exploited material would be sold as products and the waste could be employed for the environmental rehabilitation of the *cradle* quarry site. Little is known, however, about the possible success of the field application of these materials.

The paper outlines the results obtained from a remediation treatment of residual sludge coming from dimension stone working plants to obtain an artificial cultivation substrate. The tested bioremediation treatment consists in the composting of a mixture of waste materials (mineral and organic), added with specific activators (Dino et al. 2006, 2012). The aim of the research is to evaluate if this prospect product, the artificial substrate, can be employed in quarry rehabilitation.

Materials and methods

Geographic and geological setting

The materials employed come from local plants which work *Luserna Stone* blocks, exploited in the area between Luserna San Giovanni and Bagnolo Piemonte (SW Piedmont region, Italy, Fig. 1). *Luserna Stone* is the commercial name of a gray-greenish gneiss, which locally changes the color to light blue. Petrographically, it is a leucogranitic orthogneiss characterized by a micro-"Augen" texture (Sandrone et al. 2000). It pertains geologically to the Dora-Maira Massif (Sandrone et al. 1993) and it outcrops in quite a wide area (approximately 50 km^2) in the Cottian Alps (Fig. 2); it shows a sub-horizontal attitude, with a marked foliation that is mostly associated to a visible lineation. The layers rich in mica give the flat schistous aspect to the rock; the average distance between the layers is 7–8 cm (range 2–40 cm). The principal components of the rock are quartz, mica, and feldspar (Table 1) and its chemical composition is shown in Table 2.

Sludge from the working plant

The products which reach the stone market are mostly slabs, squared blocks, often sold to other companies, *mosaico* (split face pieces for "crazy" paving), belgian blocks, and kerbstones. The transformation from block to slab is carried out by means of gangue saw with abrasive shot (GSS), or diamond frame saw or by manual wedging. Samples from GSS, sludge from diamond frame saw (DSS), and mixed sludge (MS)—from gangue and diamond frame saw in the case of a single productive line—were collected in accordance with the method ISO/DIS 10381-1-2. Samples were air-dried, gently crushed and sieved to <2 mm with plastic sieves to reduce metal contamination. Grain-size analysis was performed by sieving according to ASTM standards (D421-85; D422-63). A portion of the sample was further ground to <0.15 mm for *aqua regia* (HCl/HNO_3, 3:1 solution) digestion. Metal content (As, Cd, Co, Cr, Fe, Hg, Mn, Ni, Pb, and Zn) was determined by ICP–OES (TJA IRIS Advantage/1000 radial plasma spectrometer) *in the aqua regia* extracts. The analyses were performed by following the standard methods of the Italian Ministry of Agriculture (Mipaf 1999). Triplicates were made for all samples and results accepted when the coefficient of variation was within 5 %. A blank and soil CRM BCR 141 R reference material (Joint Research Centre—Institute for Reference Materials and Measurements, Geel, Belgium) were included in each batch of analyses for quality control. Results were considered satisfactory when within a range of ±10 % from the certified value. Furthermore, total petroleum hydrocarbons (TPH) were determined by ISO 16703:2004 method.

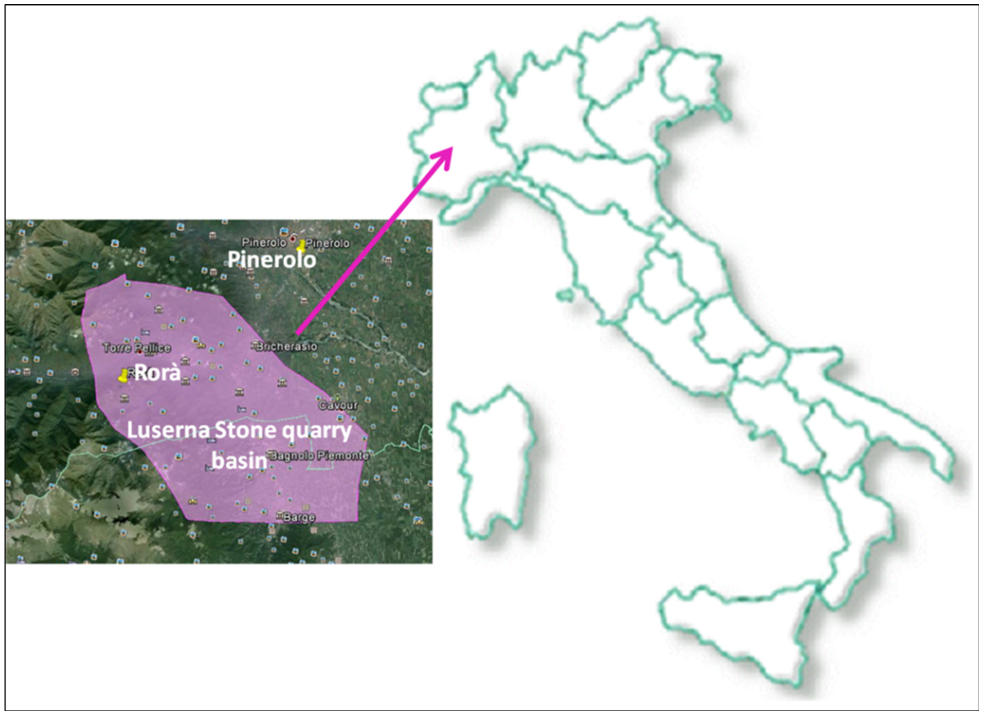

Fig. 1 Geographical setting: in the enlargement on the left the quarry in Rorà (TO): Cava del Tiglio–Pra del Torno (44°47′36″N 7°11′07″E) is indicated

Mixing materials

The materials employed for the experimentation were compost, shredded pruning material (green manure), and a soil material taken from a grassland near the quarry area. The compost from municipal organic waste and the green manure (clippings from gardening operations) were obtained from the ACEA Pinerolese company which operates in Pinerolo (Turin, Italy). These materials were characterized by following the same methodology employed for sludge characterization. Organic C and total N were measured by dry combustion (Methods ISO 10694; 13878). Approximately 35 m^3 (60 t) of each sludge type (humidity around 30 %) and 60 m^3 (40 t) of mixing material were used to obtain the mixtures. Previous experiments had demonstrated that these proportions generated the optimal conditions for the physical and biological functioning of the mixture. In particular, another research (Dino et al. 2003) on the recovery of diamond frame saw sludge (gneiss material from Canton Ticino, Switzerland) was conducted; the products obtained from sludge bioremediation treatment were employed for environmental rehabilitation.

Composting process

The composting of the four mixtures (Fig. 3) was started in the spring and was carried on for 12 weeks. The mixtures were arranged in triangular-shaped heaps (70 m^3: $2.5 \times 1.6 \times 35$ m) placed under a shed. Completion of the composting process was ensured by regular suction filtration and air regulation, wetting, and mechanical mixing and homogenization of the material. These operations were programmed on the basis of the results obtained by periodical physical and biological parameter tests. Process operating conditions (temperatures, colony forming units-CFU/g, pH, and moisture) were monitored and occasionally reassessed during the fermentation (4 weeks), as well as during the maturing phase (8 weeks) (Dino et al. 2006). The mixtures were chemically and physically characterized following the same methodology employed for their starting materials.

Phytotoxicity of the composted mixtures

Phytotoxicity was tested in greenhouse bioassays, in order to evaluate their effects on the germination and growth of crops. Corn, winter wheat,and lentils were planted (10 seeds/pot) in each of the mixture in a random block scheme (five replicates). After 4 and 17 weeks, total number of plants, plants height (in cm), and dry matter production were determined. The compost and the soil used for the mixtures were used as controls.

The total (150 m^3) of the four mixes was employed for quarry rehabilitation in *Luserna Stone* Basin: it was distributed in plots of 10 m in width and length (thickness 8–10 cm) on a hill side in “Cava del Tiglio” Loc. Pra del Torno (Fig. 4), in Rorà village (Turin) and sown with a

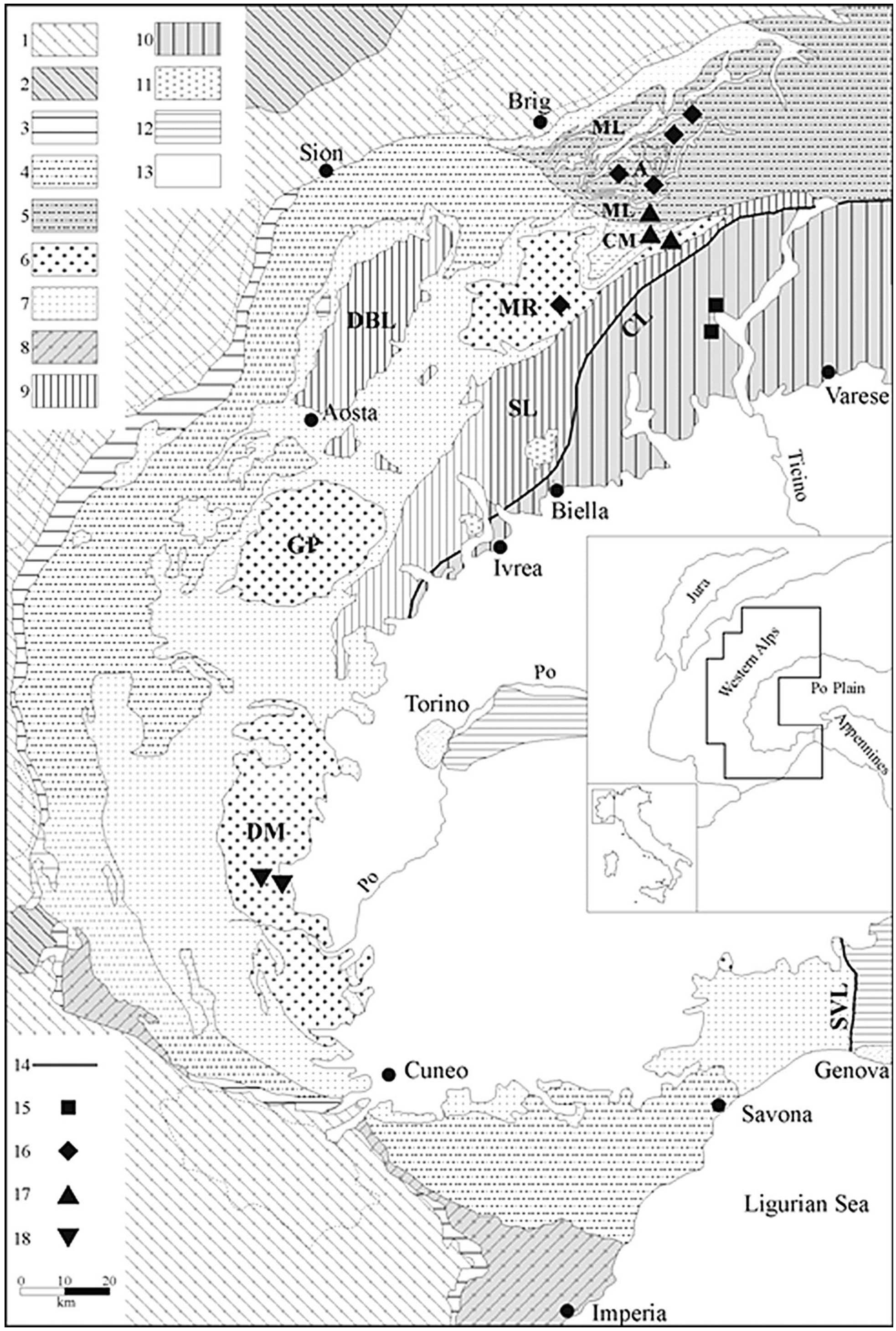

Fig. 2 Structural sketch map of the Western Alps (Dino et al. 2003). *1* Delphinois-Helvetic domain (*dashed line* external crystalline massifs). Penninic Domain—*2* Prealps, *3* Subbriançonnais Zone, *4* Briançonnais Zone, *5* Simplon-Tessin Nappes (*A* Antigorio, *ML* Monte Leone) and Camughera-Orselina-Moncucco Zone (CM), *6* Internal crystalline massifs (*MR* Monte Rosa, *GP* Gran Paradiso, *DM* Dora Maira), *7* Piedmontese Zone; North-Penninic Calcschists; Triassic-Neocomian succession of Versoyen, Montenotte and Sestri-Voltaggio Units, *8* Alpine Helminthoid Flysch of Ubaye-Embrunais and Liguria, *9* Sesia–Lanzo Zone (SL) and Dent Blanche Nappe (DBL), *10* Southern Alps, *11* Post-Alpine intrusives of Traversella and Valle del Cervo, *12* Appennines and Tertiary sediments of the Turin Hill, *13* Quaternary and Tertiary sediments of the Ligurian-Piedmontese Basin, *14* Main tectonic lines (*CL* Canavese Line, *SVL* Sestri-Voltaggio line), *15* Granite, *16* Serizzo, *17* Beola, *18* Luserna Stone

Table 1 Modal composition (% w/v) range of Luserna Stone

Quartz	K-feldspar	Plagioclase (% anorthite)	Biotite and/ or chlorite	Phengite	Others
30–45	10–25	15–25 (2–5)	4–8	10–20	≤3

Data from Vialon (1966) and Barisone et al. (1979)

mixture of *Lolium perenne* L., *Festuca rubra* L., and *Poa pratensis* L. (Dino et al. 2006). A geotextile was placed over the plots to protect them from erosion.

In October 2013, all the plots were sampled, and all the sampled materials were characterized following the same methodology employed for the starting materials.

Table 2 Chemical composition (% weight of oxides) of Luserna Stone (Sandrone et al. 1982)

SiO_2	TiO_2	Al_2O_3	Fe_2O_3	FeO	CaO	MgO	K_2O	Na_2O	P_2O_3
71.6–75.8	0.1–0.2	13.1–15.6	0.4–1.0	0.3–0.7	0.8–1.2	0.4–1.1	3.7–5.2	2.9–4.7	0.1–0.4

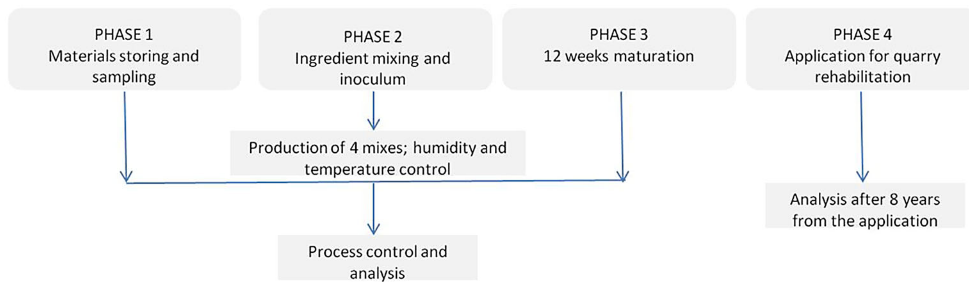

Fig. 3 Treatment flow sheet

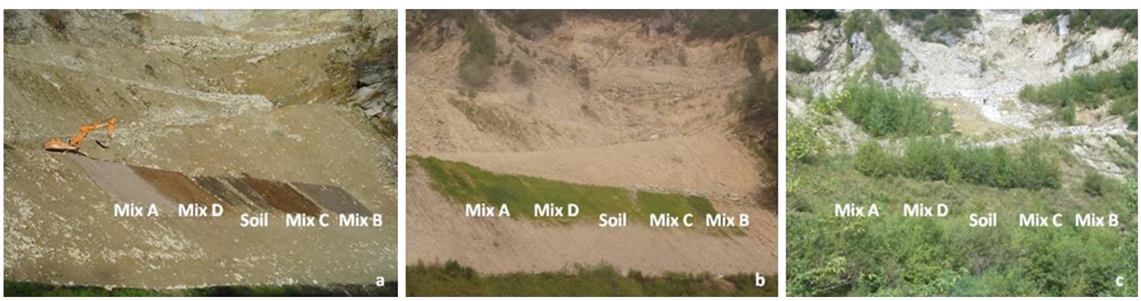

Fig. 4 Scheme of plots present on the hill side of "Cava del Tiglio" Loc. Pra del Torno—Rorà, Turin (44°47′36″N 7°11′07″E). **a** 1st step of quarry rehabilitation; **b** plant growth 3 months after seeding; **c** quarry rehabilitation after 8 years

Results and discussion

The characteristics of the residual sludge depend on the nature of the mother rock and on the working activities they undergo. In all cases, the sludge material was characterized by an alkaline pH, due to the use of lime as an antioxidant in the sawing operation. The average pH was 10.7 for GSS, 9.2 for DSS, and 11.2 for MS. The size distribution is fine (more than 40 % is <25 µm average diameter) all samples being clayey silt and weakly sandy. The solid fraction of the sludge constitutes, for its physical characteristics, an impermeable massive material which would not be usable, as such, for plant growing. The sludge contains variable amounts of metals and hydrocarbons (TPH) coming from working activities (Table 3). GSS contains notable amounts of metals coming from abrasive shot, in particular Cr and Cu, and Zn; DSS contains metals from the binder material of diamond cutting tools—Co, Cu—and has also the highest Zn concentration. The MS samples are heavily enriched in Cr, Cu, and Ni. Compared to the Italian legislative thresholds for soils (Ministero dell'Ambiente 2006), Cd, Co, Cr, Cu, and Ni and TPH in all materials result to be above the limits for residential areas reinforcing the notion that sludge cannot be used as such (Dino 2004). Magnetic separation of the metals from the GSS sludge was tested in a specific project (PIC Interreg III A "Residual sludge exploitation"). The metal concentration was decreased, but the technical feasibility of the method is still debated (Dino et al. 2003).

The sludge was mixed with organic materials and a soil in view of improving its fertility and environmental quality. The organic materials (Table 4) have a high pH and, while the compost has an equilibrated C/N, in the green manure carbon material prevails; the soil has a circum-neutral pH and an average content of C and N. The contents of contaminants (Table 4) are relatively high for some metals in the compost: Cr, Cu, and Zn are above the Italian legislative limits. This was taken into account in the composition of the mixtures (Table 5). The mixtures have properties that reflect their composition (Table 6). While the pH remains above neutrality, the C/N ratio increases in consideration of the inclusion of green manure. The general biological quality, as illustrated by the CFU count (Fig. 5), shows a definite improvement at the end of the process, fostering the use of the mixes for cultivation. The mixing and composting of the materials had some effects on the contaminant contents. While TPH decreased sharply, the metals showed somewhat erratic results as a consequence of their relatively high concentration in the compost. Co, Cr, and Cu are still above the legislative limits for soils of green and residential areas in some of the mixes. The performance of the mixtures was tested in terms of germination (Fig. 6) and yield (Fig. 7) of lentil (*Lens culinaris*, Medik.), corn (*Zea mays*, L.), and winter wheat (*Tricticum aestivum*, L.). Although in general the results were below the performance of a standard compost used as a benchmark, neither the mixes, nor the sludge itself appeared to be harmful to the plants tested. It should be noted, in addition, that the mixes in some cases produced a better results than those of the soil.

Eight years after the implementation, the substrate was again sampled and analyzed (Table 7). The plots had never been irrigated nor the plants were removed. The results show that while the pH has remained above 7 in all cases, the content of organic C and N tends toward equilibrium with a decrease of the amount of C and a consequent decrease of the C/N ratio. The metal content, as well as the TPH concentration all are reduced after 8 year with the notable exception of Pb. No sources of Pb contamination are present in the area. It is then possible that the inorganic forms of Pb have become more labile through the years and the *aqua regia* extraction—which does not extract part of the residual, more stable forms (Rauret et al. 1999)—was more complete at the end of the experiment. The increase in mobility of metals has been observed, e.g., in experiments of repeated soil reduction–oxidation cycles (Balint

Table 3 Contaminants in the sludge

		GSS	DSS	MS	Italian legislative threshold values for soils according to their use	
					Green, residential	Commercial, industrial
As	μg/kg	9.8	14.2	12.9	20	50
Hg	μg/kg	0.9	0.1	0.4	1	5
Cd	mg/kg	1.8	0.6	*3.1*	2	15
Co	mg/kg	14.4	*89.4*	18.7	20	250
Cr	mg/kg	*166*	24.5	*189*	150	800
Cu	mg/kg	*188*	49.9	*219*	120	600
Ni	mg/kg	95.3	10.1	*132*	120	500
Pb	mg/kg	12.6	8.2	14.9	100	1,000
Zn	mg/kg	75.7	98.8	38.8	150	1,500
TPH	mg/kg	*746*	*411*	*165*	50	750

Values in italics exceed the threshold values of the Italian legislations

Table 4 Characteristics and contaminants of organic materials and the soil (values are on dry weight—d.w.)

		Compost	Green manure	Soil
pH		8.6	7.5	6.4
Total organic C	%	24	32	1.9
Total N	%	2.3	0.51	0.18
C/N ratio		10.3	63.3	10.6
As	μg/kg	3.9	2.0	12.9
Hg	μg/kg	0.5	0.1	0.1
Cd	mg/kg	<0.5	<0.5	0.6
Co	mg/kg	bdl*	4	19
Cr	mg/kg	*154*	76	94
Cu	mg/kg	*142*	34	45
Ni	mg/kg	98	47	84
Pb	mg/kg	82	19	37
Zn	mg/kg	*330*	104	80
TPH	mg/kg	bdl*	889	319

* Below detection limit

Italic values exceed the threshold values of the Italian legislations

Table 5 Composition of the four mixes (% d.w.)

	Mix A	Mix B	Mix C	Mix D
GSS	–	–	62	–
DSS	59	59	–	–
MS	–	–	–	61
Compost	18	14	8	8
Green manure	23	17	20	20
Soil	–	10	10	11
Total	100	100	100	100

Table 6 Properties and contaminants of Mix A, B, C, and D after the composting process

		Mix A	Mix B	Mix C	Mix D
pH		7.6	7.6	7.6	8.0
Total organic C	%	7.7	4.9	7.6	6.9
Total N	%	0.26	0.18	0.23	0.28
C/N ratio		29	27	33	25
As	μg/kg	11.2	13.5	10.8	9.7
Hg	μg/kg	<1	<1	<1	<1
Cd	mg/kg	0.5	0.5	0.5	0.5
Co	mg/kg	*49*	*62*	13	14
Cr	mg/kg	41	36	*190*	122
Cu	mg/kg	72	63	*207*	*168*
Ni	mg/kg	31	30	116	90
Pb	mg/kg	22	14	26	23
Zn	mg/kg	116	91	99	93
TPH	mg/kg	10	10	10	10

Italic values exceed the threshold values of the Italian legislations as reported in Table 3 (values are on dry weight—d.w.)

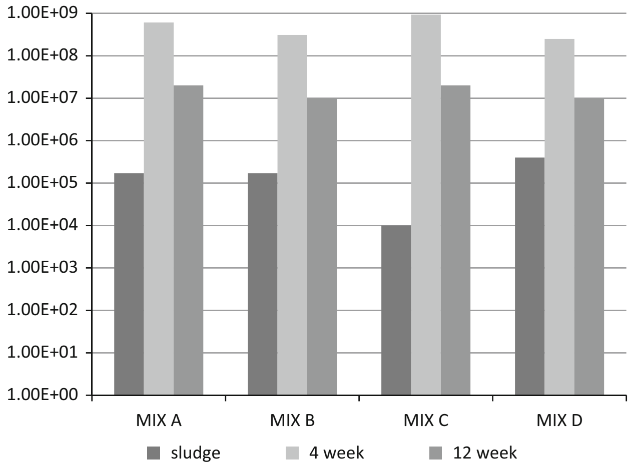

Fig. 5 Colony-Forming Unit (CFU) changes from t0 (start of composting) to t4 (4th week after start) and to t12 (12th week after composting start)

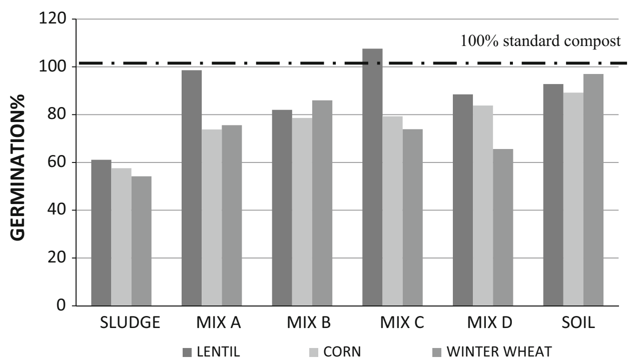

Fig. 6 Germination ratio versus standard compost (100 % = standard compost)

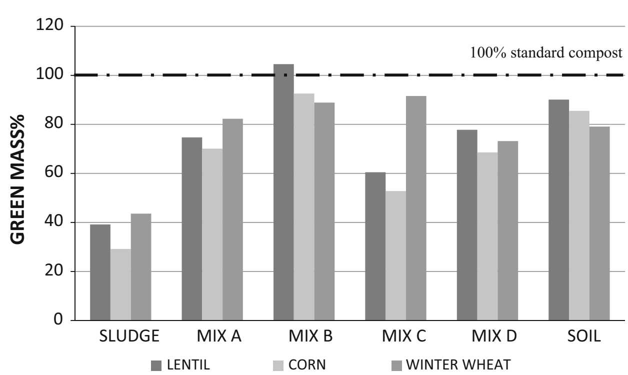

Fig. 7 Green mass development versus standard compost (100 % = standard compost)

et al. 2014) and the effect of the vegetation itself should be taken into account. In a comprehensive study of Pb in the area of Oslo (Norway) Reimann et al. (2008) demonstrated a distinct effect of the components of the biosphere on the cycling of Pb in the environment.

Table 7 Characterization and contaminant contents of the four mixes after 8 years

		Mix A	Mix B	Mix C	Mix D
pH		7.5	7.4	8.1	8.1
Total organic C	%	3.2	3.0	4.0	2.9
Total N	%	0.22	0.25	0.29	0.19
C/N ratio		14	12	14	15
As	μg/kg	11.2	12.1	10.8	9.7
Hg	μg/kg	<1	<1	<1	<1
Cd	mg/kg	0.5	0.5	0.8	0.7
Co	mg/kg	*32*	*54*	*24*	16
Cr	mg/kg	40	32	130	84
Cu	mg/kg	65	59	*174*	91
Ni	mg/kg	32	29	103	63
Pb	mg/kg	30	20	39	31
Zn	mg/kg	83	78	99	79
TPH	mg/kg	10	10	10	10

Italic values exceed the threshold values of the Italian legislations as reported in Table 3 (values are on dry weight—d.w.)

Conclusions

A pilot experiment was conducted for the remediation of a quarry site using dimension stone sludge, soil, and/or organic waste materials such as composted municipal waste and green manure. Despite the presence of numerous inorganic and organic contaminants, the mixtures gave positive results both in terms of abatement of contaminants and improvement of mid-term fertility. After eight years from the implementation, and without external intervention—irrigation, fertilization or cropping—the mixtures host a composite vegetation and show no sign of surface erosion. The environmental quality of the substrate, in terms of concentration of contaminants, could be improved. A more scrupulous consideration of the proportion of the components in terms of environmental quality would easily improve the response of the method. The employment of such waste materials appears to be promising in view of the general adoption of the *cradle-to-cradle* principle.

Acknowledgments The ACEA Pinerolese company, Pinerolo (TO) is gratefully acknowledged for providing the MSW compost and the facility for conducting the preparation of the mixes. The collaboration of the Reviplant nursery, CORIVER branch, Moncalieri-(TO), with the germination and yield tests is also acknowledged. We are grateful to Prof. M. Fornaro, Eng. E. Fornaro and Eng. E. Corio for their precious help throughout the study.

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