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Title: Sewage sludge as organic amendment for quarry restoration: effects on soil and vegetation

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ABSTRACT

Quarry restoration in Mediterranean environments usually needs organic amendments to improve the substrates used for technosol construction. Digested sewage sludge from municipal wastewater treatment plants are rich in organic matter, N and P and constitute an available an economically interesting alternative for substrate amendment. However, their pollutant burden and labile organic matter content, involve an environmental risk that must be controlled. Moreover, ecological succession in restored areas can be influenced by the use of sludge and should be assessed. To minimize these risks, a new sewage sludge dose criteria according its labile organic matter and heavy metal content has been established. Sewage sludge doses currently ranges between 10 and 50 Mg ha^{-1}. In order to verify the suitability of this dose criterion, sixteen areas rehabilitated using sewage sludge located in limestone quarries on a Mediterranean climate in Catalonia (NE Spain) has been assessed. These evaluations focused in physicochemical properties of rehabilitated soils, land degradation processes and ecological succession. At short term, six months after sludge application, an increment of organic matter content in the restored soils was observed, without significant increases on electrical conductivity or heavy metals content, and with a dense plant cover that contributes to an effective soil erosion control. Two years after, ruderal plants were still present but native species colonized the restored zones in different degrees. These results suggest that sewage sludge, used as soil amendment according to the proposed methodology, improves technosol quality in safe conditions without constraints that compromise ecological succession.

KEYWORDS: soil rehabilitation, organic amendment, stability degree, Technosol, erosion control, quarry restoration

INTRODUCTION

Restoration ecologists have long recognized the role of soil, particularly its physical and chemical properties, in the successful revegetation of degraded sites (Jordan et al., 1987; Heneghan et al., 2008). Starting from this premise, ecological restoration principles applied to quarry restoration implies in many cases the use of own mine spoils (Tedesco et al., 1999; Ram et al., 2006; Jordán et al., 2008), mainly when topsoil stripping is not possible or not gives sufficient quantity of soil. However, these materials usually do not meet the minimum fertility requirements for their direct use as soil substitutes in land restoration and have to be improved using organic amendments. In this context, the use of sewage sludge as organic amendment could represent an economically and environmentally effective alternative to create a new fertile substrate, currently named technosol, for plant growth. Sewage sludge contains nutrients and trace elements essential for plant growth (Ortiz & Alcañiz, 2006), and organic matter which can act as a soil conditioner to improve the physical properties, such as soil aeration and water-holding capacity (Sort & Alcañiz, 1999; Singh & Agrawal, 2008). However, their pollutant burden, constituted by heavy metals and a variety of organic compounds, needs to be controlled. In this sense, there are some legislative regulations (European Union, 1986) and proposals (European Union, 2000) that establish maximum levels of heavy metals and organic pollutants in sewage sludge and receiving agricultural soils.

Organic amendments with high labile organic matter content are not suitable for land rehabilitation as this type of organic matter can be quickly mineralised, disappearing and releasing large amounts of nutrients, limiting its positive effects to a short time. Moreover, in studies carried out with different types of organic wastes, a negative correlation between the organic matter stability degree (the proportion of stable, not labile, organic matter) and toxicity to plants and/or soil fauna has been described (Fuentes et al., 2004; Domene et al., 2007; Ramírez et al., 2008). Furthermore, a strong correlation between the stability degree and total nitrogen, hydrolysable nitrogen and NH_4-N content has been found (Ramírez et al., 2008). At field level, high amounts of available nitrogen in rehabilitated soils promotes ruderal plant species predominance (Moreno-Peñaranda et al., 2004), making ecological succession difficult, and supposes a risk for groundwater contamination (Navarro-Pedreno et al., 2004). Regarding groundwater pollution by heavy metals, these don't suppose a real risk because heavy metals mobility in water is very low (Hornburg & Brummer, 1994).

In order to limit or avoid these unintended situations, a new dose calculation protocol for organic amendments to be used as soil amendments has been proposed (Alcañiz et al., 2009; Carabassa et al., 2010). Regarding to organic amendment characteristics, only stabilised amendments are allowed, recommending stable organic matter content greater than 30% (i.e. amendments containing a maximum amount of labile organic matter of 70%). In order to prevent heavy metal pollution, the protocol recommends to avoid sewage sludge and soils with metal concentrations above the limits proposed by the document of European Commission (2000), and do not reach these limits on the resulting technosoils. Moreover, in soils or substrates having more than 20 g kg^{-1} of organic matter, sewage sludge amendment is not recommended.

On the other hand, enclosed to this dose calculation protocol, a site-aptitude evaluation methodology is designed with the objective of avoiding applications on unsuitable areas such as sites vulnerable to pollution, protected groundwater recharge areas, etc. The items (topics) included on this evaluation procedure are the proximity of the zone to be restored to wells or watercourses, the location of quarry on a zone of aquifers vulnerable to nitrate contamination, the water table depth, the accessibility and the space for storage and mix sludge with soil, site visitation frequency, farming utilization and the proximity to inhabited sites. Additionally, if the evaluation procedure determines that a site is suitable for sewage sludge use, a maximum area of 2 ha per year is allowed to be restored using this amendment. This protocol is being used nowadays by the Waste Agency of Catalonia (NE Spain) and by waste management companies to calculate sludge doses and sludge application conditions for their use in quarry restoration works (DTS, 2015).

The main goal of this paper is to introduce the dose criteria proposed and to evaluate the effects of sewage sludge application as a technosol amendment under an ecological restoration point of view. To do this, soil quality parameters, degradation processes and plant composition, as indicators of ecological succession, have been evaluated.

METHODS

Sludge-dose criteria

According to the protocol described in Alcañiz et al. (2009) and Carabassa et al. (2010), sewage sludge is dosed regarding the stability degree of its organic matter, determined by weight loss-on-ignition (LOI) at 550°C after acid hydrolysis. The protocol proposes a 5 g kg^{-1} as a maximum amount of labile organic matter added by the sludge on the amended soil. Other parameters as the organic matter content of the sludge, the thickness of the technosol layer to be applied (0.4 m maximum), the bulk density and the percent of <2 mm size fraction are considered in the dose-calculation formula:

$$D_{DM} = T \cdot BD_E \cdot FE \cdot \left(5 + \frac{5 \cdot S}{1 - S}\right) \cdot \frac{1}{OM_S} \cdot 10$$

Where D_{DM} is the dose of sludge (Mg ha^{-1}, dry weight); T is the desired thickness of the substrate layer to be applied (m); BD_E is the bulk density of the mineral substrate (Mg m^{-3}); FE is the proportion of <2 mm size particles of the mineral substrate (g kg^{-1}); S is the stability degree of the sludge (g kg^{-1}) and OM_S is the proportion of organic matter in the sludge (g kg^{-1}).

Moreover, as a prevention criteria, this protocol fixes a maximum dose of sludge (50 Mg ha^{-1}, dry weight), based on earlier experience obtained from ecotoxicological assays using plants and soil fauna (Domene et al., 2007, Tarrasón et al., 2008).

Restored zones selected

A set of 16 areas located in 11 quarries that were restored using respective mine spoils and amended with sewage sludge according to the current protocol has been selected (table 1). These quarries are located in Mediterranean environments encompassing a climatic gradient from semiarid to sub-humid. Mining activities exploiting diverse calcareous materials that could influence the restoration processes have been included. Mine spoils were the main substrate used to create a technosol for topsoil rehabilitation. All the substrates were calcareous (30% to70% of carbonate content) and stony, but with more than 30% of <2 mm particle-size fraction and a loamy texture.

The average sewage sludge dose was 40 ± 13 Mg ha^{-1} (mean ± SD, dry weight basis). All sludge applied came from municipal wastewater treatment plants close to the mining areas, being subjected to an anaerobic digestion process and partially dehydrated (table 2). The organic matter content of these sludges and their stability degree was relatively high. They had high concentration of nitrogen and phosphorous whereby are currently applied to agricultural soils. The heavy metals content was low and always met the requirements of the European Union (2000).

Steeped slopes (60-75%) are the predominant restored surfaces in the mine sites selected, with someone that exceed 100%. On these slopes, a layer of about 20 cm of amended substrates (mainly mine spoils mixed with sewage sludge) were spread on. The average area of restored slopes is 4,500 m^2.

Evaluation parameters

The parameters evaluated on the restored zones concern soil quality, degradation processes and vegetation. Soil samples were taken 4-6 months after sludge application but degradation processes and vegetation data were assessed 24 months after. Soil sampling was done taking a composite sample of cores (n =12-20, d=0-20 cm) for each restored zone. The analysed parameters in soil samples were: particle size determined by sedimentation-Robinson pipette (Gee & Or, 2002), equivalent $CaCO_3$ by CO_2 volume released after HCl addition -Bernard calcimeter method (Nelson, 1982), electrical conductivity of 1:5 water extract (Rhoades, 1982), organic carbon content by acid dichromate oxidation (Nelson & Sommers, 1982), total nitrogen using the Kjeldahl method (Bremner & Mulvaney, 1982), available phosphorous-Olsen phosphorous (Olsen & Sommers, 1982) and potassium (Knudsen et al., 1982), and heavy metals by ICP-MS analysis (Thomas, 2004). Geotechnical and soil degradation processes were estimated through direct measures and observations at field, and erosion losses were estimated by measuring the rill volume. Vegetation measures were taken establishing transects and sampling plots (10m transects, 100m^2 plots, 3 per area).

RESULTS AND DISCUSION

Soil quality indicators of technosols were evaluated after one growing season (spring or autumn) (table 3). The electrical conductivity of amended soils remained low, with the only exception of two cases that were greater than 2 dS m^{-1}. Organic C contents were almost always upper than 10 g kg^{-1}, with an average increment of 11.2 ± 7.8 g kg^{-1} (mean ± SD) caused by the addition of the sludge to the mineral substrate that constitutes the mineral fraction of the technosol. Phosphorous contents tends to be high and correlated with the amount of sewage sludge added. Total nitrogen concentrations were balanced to the organic matter content having a C/N ratio close to 10, and according to the sludge dose applied. Available potassium content tends to be low, mainly in very rich calcium soils. Heavy metals content were low on the amended soils. Minor increases in the concentrations of these elements were observed after the addition of sewage sludge, all below the European upper limits for agricultural soils (table 4).

Soil quality parameters indicate that sewage sludge application causes a substantial improvement of organic matter and nutrients content on the amended technosols as reported in other works (Albiach et al., 2001; Heras et al., 2005). Despite the noticeable increase in the soil nutrients due to sludge mineralisation, electrical conductivity did not raised greatly. Available phosphorous levels increased by sludge addition but were in a high-medium range regarding agricultural soils of the region. However, partial immobilization could take place due to the alkaline pH of these highly calcareous soils. According to sludge composition, mineral nitrogen levels may rise just after sludge application, mainly in ammonium form (not measured in soil samples). This ammonium nitrogen may turn fast into nitrates in these calcareous soils (Kleber et al., 2000), that could leach and contaminate aquifers. However, nitrogen leaching, if occurs, should take place mainly in the first four months after sludge application due the high mineralization rates of the organic-N from sludge. After this, leaching risk decreases fast due to the nitrate absorption by roots of the growing plants, reducing the risk of aquifer eutrophication (Tarrassón et al., 2008; Jordán et al., 2017). Moreover, it has to be taken into account that this risk is relatively low due to the small surface restored (<2 ha in a quarry per year) and because sludge is applied only one time, compared to agricultural applications where sludge is applied recurrently at the same plot. On the other hand, a fraction of organic matter from sewage sludge persists on soil due to its recalcitrant composition (e.g. some lipids and aromatic hydrocarbons), and a labile organic matter fraction could also remain in soil protected into aggregates or by sorption on mineral surfaces (Ojeda et al., 2015).

The total amounts of heavy metals in the amended technosols never exceeded the concentrations fixed by the European Union proposal (2000), which is stricter than the current European Directive that regulates the agricultural use of sewage sludge. This is due to the relatively low concentration of these elements in the sewage sludge but also to the moderate doses applied. Moreover, the metal bioavailability in the studied technosols is expected to be low, according its pH and lime content (Ortiz & Alcañiz, 2006).

No noticeable water erosion or other soil degradation processes were observed 24 months after sludge application. Only in two cases stability issues (landslides and fallen rocks) were reported due to the excessive slope (greater than 100% in some sites). Rill erosion has been detected in seven restored zones although the estimated erosion rates were always below 6 Mg ha^{-1} y^{-1}, which can be considered an acceptable rate in restored areas, with the only exception of Las Cuevas quarry, where Z1 zone is severely affected (table 5).

Dense plant cover has been observed on the evaluated zones two years after sludge application. Average plant cover is 70 ± 24 % and herbaceous plant height 0.45 ± 0.30 m. Concerning species richness, more than 20 plant species have been identified in evaluated zones. Regarding herbaceous vegetation (see figure 1), grasses were the most frequent functional group of plants ($P < 0.015$). However, some herbaceous reported species can be considered as ruderal plants that are growing in nutrient-rich and disturbed habitats, usually resulting from human activity. Leguminous are also well represented in sewage sludge revegetated zones at similar frequency as compositae or ruderals. No exotic or invasive species were observed in the evaluated zones, despite some individuals of *Arundo donax* were present in one zone (Falconera quarry) before sludge application.

Vegetation succession shows that native species start to colonize the amended zones two years after sludge amendment. Herbaceous species were the main colonizers, getting into half of the amended zones. Shrub species appeared in four restored zones, especially *Santolina chamaecyparissus* and *Rosmarinus officinalis.* Moreover, in approximately 60% of the rehabilitated zones where shrubs were planted, spontaneous reproduction of these species was observed.

Vegetation cover is a key parameter for soil stabilization and erosion control (Merlin et al., 1999; Bochet et al., 2010; Espigares et al., 2011), mainly in major civil works such as the construction of motorways, the rehabilitation of quarries or dumps, and even the creation of sky slopes. Several authors (Albiach et al., 2001; Pond et al., 2005) have demonstrated that soils amended with sewage sludge favor a fast vegetal recovery and plant development, especially for herbaceous vegetation, which is the best way to control soil erosion in steeped slopes. Moreover, sewage sludge promotes soil aggregation (Sort & Alcañiz, 1996; Sort & Alcañiz, 1999; Ojeda et al., 2008), reducing soil erodibility. These combined beneficial effects of sewage sludge on vegetation development and soil structure are probably the main reasons explaining the reduced erosion rates found on steeped slopes of the studied quarries that are especially vulnerable to soil erosion.

The relatively lower frequency of ruderal species found in this work (see figure 1) compared with those reported in a previous paper of the same team (Moreno-Peñaranda et al., 2004) that show a dominance of ruderal plants on sewage sludge amended zones must be discussed. This apparent discrepancy may be explained by the different doses of sludge applied, which are lower in the present work (calculated following the protocol reported in Carabassa et al., 2010) compared to other previous ones (Sopper, 1993; Alcañiz et al., 1996; Barnhisel et al., 2000; Jorba & Andrés, 2000; Morera et al., 2002; Moreno-Peñaranda et al., 2004). Therefore, the main difference falls on the quantity and quality of the organic matter added to the mineral substrate. As explained in the methods section, the new procedure calculates the dose of sludge according to their concentration of stable organic matter (stability degree), and fix the maximum dose at 50 Mg ha^{-1}. These criterions imply an addition of a limited proportion of labile organic matter and a relatively low addition of total organic matter associated to sludge application, which contributes to reduce the development of ruderal plants currently associated to over-fertilized soils. However, the presence of ruderal plants in restored areas may be common also when organic amendments are not used (Hobbs & Adkins, 1988; Alpert et al., 2000; Moreno-Peñaranda et al., 2004). Therefore, the use of sewage sludge in appropriate dose should not be considered as an unsuitable factor concerning plant community succession towards the natural surrounding vegetation. Furthermore, the noticeable recruitment of native shrub species may suggest a plant community convergence to adjacent undisturbed habitats at medium term. However, this process should be monitored at long term as is one of the main objectives of the ecological restoration (SER, 2004). Moreover, an increasing emphasis will be focused on proper ecological functionality of a restored site, and to a lesser extent on nudging a restored site back to previous conditions based on species composition (Harris et al., 2006).

CONCLUSIONS

On the range of climatic and soil conditions tested on this work, the use of sewage sludge for vegetation recovery purposes in restoration works is a good alternative that allows the valorization of sewage sludge and increases the quality and stability of restored areas, reducing the risk of soil erosion. One of the most important parameters to take into account for sewage sludge dosage is the amount of labile organic matter, in order to avoid compromising encroachment and reduce the risk of nitrate contamination. Moreover, an aptitude evaluation of sludge, mineral substrates and emplacement is mandatory before sludge application in order to prevent contamination risks and annoyances to inhabited zones.

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Tables

Table 1. General description of the mine sites located in the NE Iberian Peninsula

Quarry	**Latitude (N)**	**Longitude (E)**	**Mean annual rainfall (mm)**	**Mean annual temperature (ºC)**	**Previous vegetation**	**Parent material**
Aiguamolls	41º 28´ 31´´	0º 49´ 39´´	450-500	14-15	Shrubland dominated by *Thymus vulgaris* and *Rosmarinus officinalis* as dominant shrubs	Marl
Calvari	41º 30´ 26´´	0º 57´58´´	350-400	14-15	Rainfed tall fruit trees	Marl
Ponderosa	41º 15´ 41´´	1º 09´ 27´´	550-600	15-16	*Pinus halepensis* forest	Limestone
Antonieta	42º 16´ 06´´	1º 22´ 11´´	750-800	11-12	*Pinus nigra* forest	Limestone
Lázaro	41º 12´ 07´´	1º 28´ 57´´	500-550	15-16	*Pinus halepensis* forest	Limestone
Orpí	41º 31´ 22´´	1º 36´ 15´´	600-650	13-14	*Pinus halepensis* forest	Limestone
Montlleó	41º 41´ 35´´	1º 49´ 39´´	600-650	13-14	*Thymus vulgaris* and *Rosmarinus officinalis* shrubs	Sandstone
Vallcarca	41º 15´ 13´´	1º 52´ 25´´	500-550	15-16	Mixed forest: *Quercus ilex* and *Pinus halepensis*	Marl and limestone
Falconera	41º 15´ 40´´	1º 53´ 12´´	500-550	15-16	*Pinus halepensis* forest	Limestone
Cubetas	41º 20´ 24´´	1º 53´ 33´´	600-650	13-14	*Pinus halepensis* forest	Dolostone
Cuevas	41º 16´ 16´´	1º 54´ 13´´	500-550	15-16	*Quercus coccifera* shrubs	Limestone

Table 2. Analytical characterization of sewage sludges applied on the selected mine sites.

Parameter	**Average**	**Max.**	**Min.**	**SD**
Dry matter (%)	24.5	26.8	22.5	12.6
Organic matter (%)	57.7	70.7	39.2	27.1
Stability degree (%)	48.1	60.1	31.8	17.2
Conductivity (dS m^{-1} 25ºC)	2.0	3.0	1.0	0.7
pH (water 1:10 w:v)	7.7	8.5	6.9	0.6
N-Kjeldhal (g kg^{-1})	36.8	63.4	11.5	19.8
N-ammonia (g kg^{-1})	10.7	21.5	1.8	7.3
P- total (g kg^{-1})	38.3	64.4	24.5	23.5
K (g kg^{-1})	2.8	5.7	1.0	1.5
Cu (mg kg^{-1})	322.3	580.0	102.0	170.6
Ni (mg kg^{-1})	23.0	43.3	15.5	13.9
Cr (mg kg^{-1})	56.6	85.6	12.5	35.7
Pb (mg kg^{-1})	54.3	64.2	29.5	28.1
Hg (mg kg^{-1})	2.0	4.2	0.2	1.3
Cd (mg kg^{-1})	3.7	10.0	0.9	3.6
Zn (mg kg^{-1})	1037.6	2199.0	375.0	512.6

Table 3. Summary of physical and chemical parameters of amended substrates (technosols). N=28. Data are referred to fine fraction (<2 mm), except coarse particle size fractions that are reported as percent of whole soil sample (ws).

Parameter	**Average**	**Max.**	**Min.**	**SD**
Fraction >5 cm (% ws)	3	10	0	3.5
Fraction 5-1 cm (% ws)	23	40	8	9.1
Fraction 1-0.2 cm (% ws)	21	42	8	8.1
Fine earth <0.2 cm (% ws)	53	87	31	15.7
Sand (%)	35	60	16	12.9
Silt (%)	41	63	25	11.3
Clay (%)	23	28	14	4.4
$CaCO_3$ eq. (%)	52	80	29	14.7
pH water (1:2.5 w:v)	8.1	8.5	7.9	0.2
Electrical conductivity (1:5 extract w:v, dS/m 25ºC)	1.02	2.94	0.3	0.8
Organic carbon (g kg^{-1})	11.3	23.4	2.1	4.1
N-Kjeldhal (g kg^{-1})	1.1	1.9	4.0	3.0
P-Olsen (mg kg^{-1})	66	103	13	25.4
K-available (mg kg^{-1})	116	244	76	41.0

Table 4. Heavy metals concentration on amended soils and average and maximum increments regarding to original mine spoils (nd: no detected; units: mg kg^{-1}). Z1, Z2 and Z3 refers to diverse slopes or zones in the same quarry. N=28.

Zone code	**Cd**	**Cu**	**Cr**	**Hg**	**Ni**	**Pb**	**Zn**
Average	0.100	20.8	25.1	0.062	19.1	27.2	77.5
Max.	0.500	50.0	40.0	0.097	29.0	55.0	190.0
Min.	0.100	6.0	7.0	0.041	11.0	11.0	18.0
SD	0.005	10.7	9.4	0.024	5.7	14.9	39.9
Limits proposed by EU (2000) for alkaline soils	1.5	100	100	1	70	100	200
Average increase	nd	0.3	1.8	nd	2.1	2.1	4.4
Maximum increase	nd	2.1	11.5	nd	12.0	11.5	14.5

Table 5. Surface of soil affected by water erosion. erosion rates and soil loss on the evaluated zones of quarries where soil erosion has been detected (Z1, Z2 and Z3 refer to diverse slopes or zones in the same quarry).

Zone code	**Affected area (m^2)**	**Erosion rate ($Mg\ ha^{-1}\ year^{-1}$)**	**Soil loss ($Mg\ ha^{-1}$)**
Aiguamolls Z1	1,225	2.3	5.3
Aiguamolls Z2	1,870	0.3	0.7
Aiguamolls Z3	1,950	2.2	5.0
Lázaro	1,800	0.4	0.9
Cubetas	8,040	1.9	4.5
Cuevas Z1	2,000	17.3	55.4
Cuevas Z2	4,400	3.2	10.0

Figures

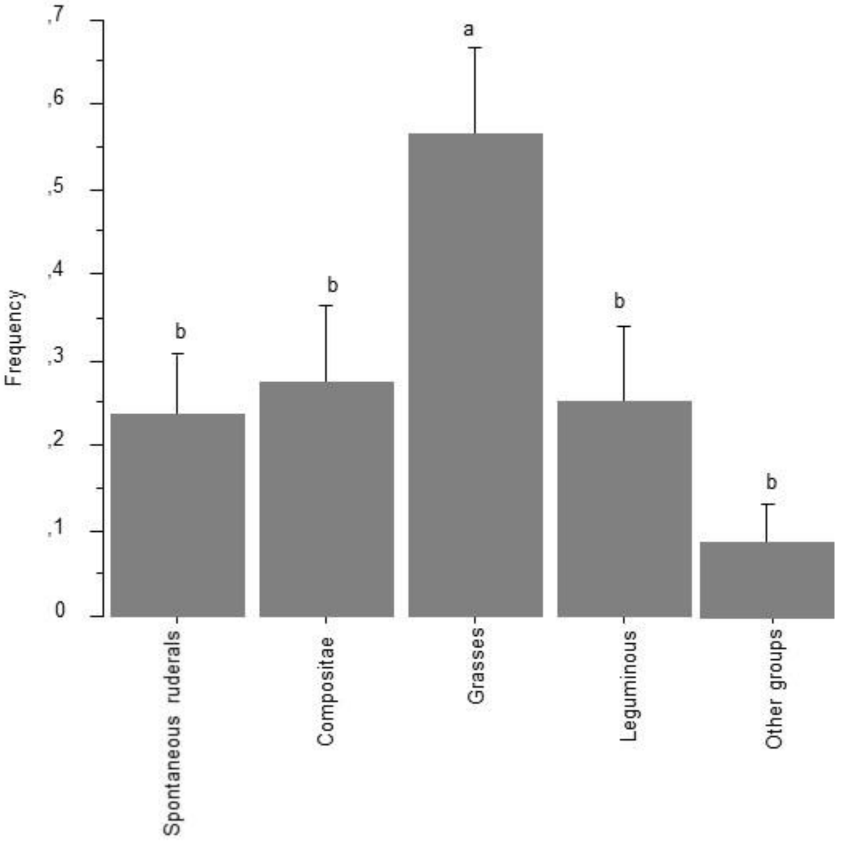

Figure 1. Frequency of different herbaceous plant functional groups in the evaluated zones of quarries. Error bars indicate standard error ($P < 0.02$).