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Environmental impacts, management and potential recovery of residual sludge from the stone industry: The piedmont case

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ABSTRACT

The management of residual sludge from dimension stone processing activities (EWC 010,413) represents a large concern for stone industry. Such sludges are characterised by a fine size distribution, high heavy metal contents and, potentially, TPH (Total Petroleum Hydrocarbon) contents. The objects of the present study are represented by gang-saw with abrasive shot (GSS), diamond frame-saw (DSS) and mixed (MS) sludges. The first one is high in Cr, Ni, Cu and TPH, the second presents high contents in Co, Cr and TPH, and the third represents a mix of the first two ones. Present legislation indicates that they can be disposed of in landfills or can be recovered (R5-R10) as filling material for environmental rehabilitation or as feeding materials for cement production.

The sampled materials were obtained from the Verbano Cusio Ossola (VCO) and Luserna Stone quarry basins (Piedmont, Northern Italy). These materials were physically and chemically characterised, and magnetic separation was performed to rapidly evaluate the heavy metal contents in the sludge. Afterwards, three different experiments were carried out for environmental and civil applications. The first one, for substrate production, was carried out using GSS, DSS and MS from VCO area. The second one, for CLSM and plaster production, was performed on materials from VCO and Luserna stone quarry basins (CLSM using GSS from VCO and plaster using DSS from Luserna stone quarry basin). The results here reported are promising for the three applications.

1. Introduction

1.1. State of the art

The management of residual sludge (EWC 010,413) produced during dimension stone processing still represents a concern in terms of the environmental and economic impacts. The main challenges to deal with are as follows (Careddu and Dino, 2016):

- the huge volume produced in single quarrying areas;
- the very fine particle distribution, with a consequent axphictic attitude;
- the presence of high heavy metals and total petroleum hydrocarbons (TPH), sometimes exceeding the threshold values and representing a potential source of pollution if not properly managed.

On the other side, in several EU and non-EU countries, the residual sludge can be recycled. The amount of material that can be recycled as a by-product to be used for other processes is high, representing a considerable economic and environmental advantage. Several studies have investigated the issues connected to sludge management and to the production of new potentially marketable products. In this paper, the most promising applications are reported.

Several studies have investigated the **use of residual sludge as secondary raw material (SRM) for the ceramic and glass industries.** In particular, because of flotation processes on siliceous residual sludge, quartz and feldspar have been enriched to meet the requirements of these industries (Curreli et al., 1992; Sassone and Danasino, 1995). A more recent study by Silva et al. (2018) discusses the appropriate treatments of siliceous (granite) sludge on the basis of the products required by the ceramic industry.

Since the 1990s, different studies have been investigating the use of residual sludge, as such or mixed with clay or bentonite, as **waterproofing material for municipal landfills and clay soil** (Bertolini and

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Celsi, 1989; Dino et al., 2013, 2015b; Frisa Morandini and Verga, 1990; Sivrikaya et al., 2014). In a recent study, Sivrikaya et al. (2014) obtained stabilised artificial clay samples by adding 5, 10, 20, 30 and 50% of waste calcitic marble, dolomitic marble and granite sludge to three clays with different plastic properties. The obtained results evidenced that, among all waste, natural stone powders suitable as stabilisers, dolomitic marble powder was the most effective one due to its chemical composition.

In addition, the use of sludge as **substrate for land reclamation** has been investigated since the late 1990s (Barrientos et al., 2010; Burragato et al., 1999; Dino et al., 2006, 2013, 2015a). Recent studies by Dino et al., in 2006 (and in 2015a with further tests) evidenced the option to recover residual sludge to produce artificial substrate for land reclamation in quarry contexts. The investigated sludge was obtained from the Luserna Stone quarry basin. Different mixes were produced, using different percentages of sludge (Gangue-saw sludge, Diamond frame-saw sludge and Mixed sludge, GSS, DSS and MS respectively) added to shredded green material, compost and natural topsoil. The obtained mixes showed a decrease in the TPH content compared to the value present in the sludge as such, and decreased metal concentrations below the threshold limits. The substrate had a good quality and was suitable as artificial soil (*technosoils*) in quarry remediation.

Several studies have investigated the use of residual sludge as **SRM for the construction industry.** In particular, two different applications, objects of the present study together with the application for substrate production, are reported here.

a **Artificial aggregate for concrete, cement and mortar (**Fang-Chih Chang et al., 2010; Al-Hamaiedeh and Khushefati, 2013; Allam et al., 2014; Al-Zboon and Al-Zou'by, 2015)

In 2010, Fang-Chih Chang et al. recycled waste stone sludge and waste silt for construction materials. Using fine waste powder compacted via vibratory compaction, obtained a material with a high compressive strength value and a lower water absorption compared to general cement concrete.

In 2013, Husam, Al-Hamaiedeh and Khushefati studied the effect of adding different amounts of granite sludge powder to cement, mortar and concrete. They conducted two sets of investigation activities: in the first one, they partially replaced cement with the sludge at different mass ratios, while in the second one, they added granite sludge to cement at different mass percentages. According to the authors, the replacement of concrete cement by granite sludge did not affect the mechanical properties of the concrete, while the addition of sludge to cement increased the compressive strength of concrete.

In 2014, Allam et al. investigated the re-use of granite sludge for the production of green concrete. They tested six mixes, three replacing cement with sludge in different quantities and three replacing sand with sludge. They concluded that granite sludge provided a good cohesiveness due to the very fine particle size distribution of the sludge. In addition, while the replacement of sand with sludge increased the mechanical properties of the concrete, the replacement of cement with sludge resulted in a decrease of the mechanical properties.

In 2015, Al Zboon and Al-Zou'by carried out a study on recycling stone cutting sludge in concrete mixes, substituting potable water with sawing sludge in the slurry state. Such replacement resulted in an increase of the mechanical properties of the concrete, but also in a reduction of the slump value and, consequently, of concrete workability.

b **Brick products** (El-Mahllawy, 2008; Ribeiro and Holanda, 2014)

In 2006, Medhat and co-authors investigated the characteristics of acid-resisting bricks made from granite-basalt fine quarry residues and waste steel slag. The obtained bricks showed a higher resistance to chemical reactions, in particular to sewage waters. The authors also studied the re-use of granite cutting sludge in soil cement bricks and found that a replacement of soil with granite sawing sludge, in the range of up to 30% wt., produced a soil-cement brick with good physical and mechanical properties.

The re-use of residual sludge as SRM or as new product in the cited applications, together with the monitoring and decrease of polluting sources because of sludge treatment, is in line with the circular economy strategy and with the definitions and measures based on the concept of the best available practices for the management of the waste of extractive industries, also for the circular economy approach (Directive, 2006/21/EC and Directive 96/61/EC).

1.2. Objectives and expected results

The present research investigates several aspects connected to residual sludge from the stone industry: from its chemical and physical characterisation (fundamental to evaluate environmental impacts and potential recovery) to the treatments, at laboratory scale, to separate heavy metals and/or to decrement the TPH content up to the potential re-uses of treated sludge. It focuses on the methodology adopted and the results obtained from the characterisation and treatment of residual sludge from the Verbano Cusio Ossola (VCO) and Luserna Stone (LS) quarry basins (two main quarry basins in the Piemonte region), derived from activities connected to slab production (starting from rock blocks).

One of the main problems related to the management and future recovery of sawing sludge is the presence of heavy metals due to the tools used during the different cutting steps. The quantity of heavy metals in the sludge depends on rock hardness and on the cutting technology used. Even if the anisotropy and the workability of the stones affect the wear of the tools (Zichella et al., 2018), also sawing by abrasive shot, by diamond wire or by diamond blade result in different concentrations of metals in the sludge (Zichella, 2019). It is possible to assert that sludge derived from cutting with the gang-saw has a greater magnetic percentage compared to sludge derived from the diamond wire technology, but the percentage varies depending on the type of the cut stone.

Against this background, the objective of the study is to obtain by-products that can be reused in other production processes, exploiting the intrinsic characteristics of the sawing sludge (particle size distribution, metal content); three main potential applications were investigated:

- Environmental application: substrate production
- Civil applications:
  - o control low-strength material (CLSM) production
  - o plaster production

In this context, an overview of the three applications is provided. The obtained results were positive for all three applications. This work focused on the reuse of residual sludge as substrate. The other two applications in the civil field, such as plaster and CLSM production, are here presented as a feasibility study, but will be detailed in dedicated articles.

We report, for the first time, specific indications in the recovery of siliceous sludge depending on the metal content. Knowing the contents of the metals, it will be possible to use it as SRM in civil applications, as plaster (low metal content) or CLSM (high metal content), or in agricultural applications (low metal content), without performing any preliminary treatment.

2. Materials and methods

2.1. Investigated materials

The investigated materials were obtained from working plants which mainly treat local granite (lake granite from VCO) and gneiss (different kinds of *Serizzo* and *Beola* from VCO and LS), in two important Italian quarry basins: Verbano Cusio Ossola and Luserna Stone quarry basins (Piemonte Region, Northern Italy). Such materials are often characterised by different compositions due to the features of the original stones (*Serizzo*, *Beola*, Luserna Stone, Montorfano and Baveno granites). Table 1 shows the chemical and mineralogical parameters of the original rocks (XRD-QPA and ICP-MS).

Three typologies of residual sludge can be distinguished:

- GSS: sludge from gang-saw using abrasive shots;
- DSS: sludge from multi diamond-saw block cutter;
- MS: mixed sludge from gang-saw and block cutter.

Several materials from both quarry basins were sampled, characterised and used for the experiments. From the VCO, we took five samples for GSS, seven samples for DSS and three samples for MS. It should be highlighted that for the LS quarry basin, periodic sampling campaigns were carried out on three plants producing GSS, DSS and MS. A representative final single sample was obtained for each technology because of the more homogeneous characteristic of the LS material compared with that from the VCO.

In 2016, the total volumes of produced sludge in the VCO and LS quarry basins were, respectively, 17.700 t and 4.700 t (Zichella, 2019), although these volumes decreased during the last 10 years (70,000 and 16,000 t/y from VCO and LS, respectively; Dino et al., 2006), residual sludge management still represents an unsolved problem for the stone industry.

2.2. Material characterisation

The Italian legislation (Decree n. 152/2006 Annex 5 Part IV) defines the threshold limits for recovering waste or allocating it to a specific landfill category. The analyses that best identifies the quantity and type of metals present in the sludge are chemical analysis, leaching tests and magnetic separation; in particular, the most rapid analysis identifying the quantity of metals present in a sludge is magnetic separation. Chemical characterisation through chemical analysis and leaching tests can also be used, but is not as rapid as magnetic separation. The purpose of this work is to find different uses of the sawing sludge, according to the metal concentration.

The flowchart described in Fig. 1 outlines the different procedures.

Magnetic separation can be considered both as a material characterisation (1), complementary to the chemical analyses, and as a pre-treatment (2) after the cutting of ornamental stones.

(1) If indicated as material characterisation technique, magnetic separation discriminates sludge with a high metal content from those with a low content. In this way, it is possible to predict different recoveries such as plaster or substrates for sludge with a metal concentration <2% and CLSM (control low-strength material) for sludge with a metal concentration > 2%. In the case of CLSM, the presence of metals increases the thermal conductivity of the finished product. In this way, CLSM are not good insulators, but are characterised by a high thermal conductivity due to their high density. In road surface application, where electric cables and sub-services are present, a high density and a low porosity are required due to the heat dissipation produced by the cables. An increase of humidity and density induces an increment of thermal conductivity, and the presence of high percentages of metals in the sludge induces an increased density of the final products, satisfying the thermal conductivity properties of CLSM.
(2) As a pre-treatment technique, magnetic separation can be carried out to obtain two by-products: the amagnetic (not-magnetic) fraction, which can be recovered as substrate or plaster, and the magnetic fraction, which can be sent to specialised landfill, thus reducing the quantity and costs related to disposal, or recovered for specific industrial sectors (after further treatment of the metallic fraction).

Chemical characterisation was performed on samples dried at 40°C. Samples were analysed for their pseudo-total metal contents using the EPA 3051A:2007 microwave digestion method (U.S. EPA, 2007). Extractable (pseudo-total) contents of Sb, As, Be, Cd, Co, Cr tot, Cr VI, Hg, Ni, Pb, Cu, Se, Sn, TI, V and Zn were determined in bulk samples using ICP-OES. Cyanide levels were determined according to the EPA 9013A method (U.S. EPA, 2014); fluorides were determined by UV-VIS spectrometry (Colombo and Miano, 2015). The TPH concentration was determined according to the method ISO 16,703:2004 (ISO, 2004). The analyses were performed in the CSL-VCO Laboratory.

The same samples were subjected to a standard leaching test (UNI EN 12,457–2:2004), which consisted of parallel extractions of the material at a liquid-to-solid ratio of 10 (L/kg) for 48 h at a series of pre-set pH values (UNI, 2004). On the eluates, pH was determined potentiometrically, nitrate, sulphates, chlorides and fluorides were determined using UV-VIS spectrometry (Colombo and Miano, 2015), while the other contaminants were determined with the same methods used for solid materials.

Together with the chemical characterisation, fast tests concerning magnetic separation were performed, using a Kolm-type high-gradient magnetic separator (Eriez Magnetics), operating in a discontinuous mode. Magnetised coils produce a strong magnetic field, and a canister containing ferromagnetic grids of different mesh sizes was placed between the magnetised coils, producing a strong magnetic field, inside which ferromagnetic grids of different mesh sizes were inserted. The sawing sludge in a fluid slurry was fed from the top of the apparatus device through a funnel, passing through the canister and the grids among the magnetised coils. (Fig. 2).

When the apparatus is operative, the magnetic particles are trapped, while the amagnetic fraction ends up in the container. In a second step, trapped magnetic particles are easily washed out when the applied field is reduced to zero. To better separate the two fractions, it is necessary to perform three steps for both fractions (magnetic and amagnetic). The set parameters of the chosen separator device were as follows: magnetic field 260 mT, 10A and 140V.

In this manuscript, the content of the magnetic fraction, characterising the sludge from VCO and LS areas (using both gang-saw or diamond frame saw), was determined.

The results of the physical characterisation, including grain size

Table 1
Mineralogical analysis of the most important stones exploited in the VCO and LS quarry basins (data from Dino, 2004 and Zichella, 2019).

Mineralogical analysis (%)	Serizzo	Beola	Luserna Stone	Montorfano Granite	Baveno Granite
Quartz	30	30	50	40	35
Plagioclase	25	15	15	20	29
Ortoclase	26	34	20	35	29
Biotite	10	5	–	5	4
Muscovite	5	14	5	–	–
Chlorite	–	–	5	–	2
Epidote	3	–	5	–	–
Accessory minerals	1	2	–	–	1
Chemical Analysis (weight, %)	**Serizzo**	**Beola**	**Luserna Stone**	**Montorfano Granite**	**Baveno Granite**
SiO_2	64.5–68.1	72.8	71.6–75.	63.7	76.5
TiO_2	0.4–1.1	0.4	0.1–0	0.4	–
Al_2O_3	18.3–19.0	16.1	13.1–15	18.8	15.4
Fe_2O_3	2.4–4.1	1.4	0.8–1.	2.3	1.6
FeO_2	–	0.1	–	2.1	0.9
MgO	0.8–1.3	0.4	0.4–1	5.4	0.4
CaO	2.7–3.8	1.4	0.8–1	0.4	1.0
Na_2O	2.8–3.8	4.7	2.9–4	4.7	4.2
K_2O	2.6–3.2	3.5	3.7–5	2.2	–

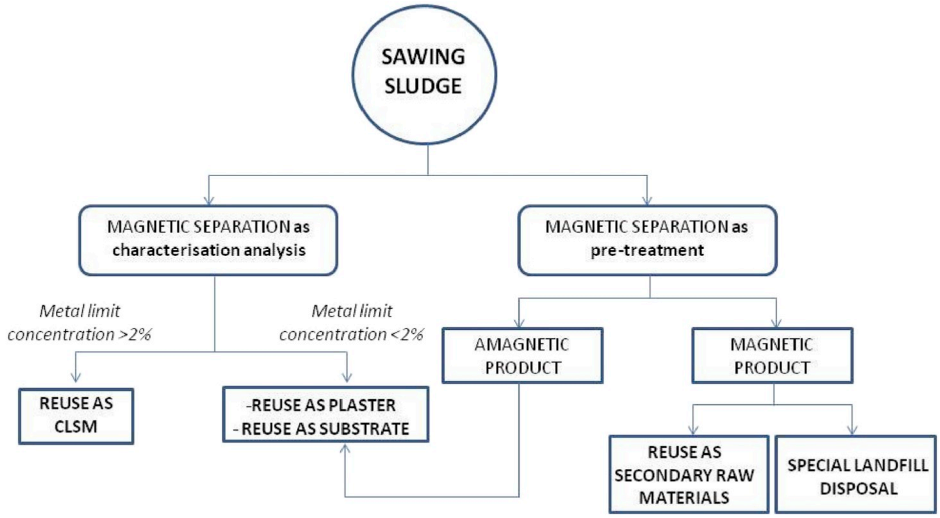


Fig. 1. Flowchart of the potential applications for sawing sludge.

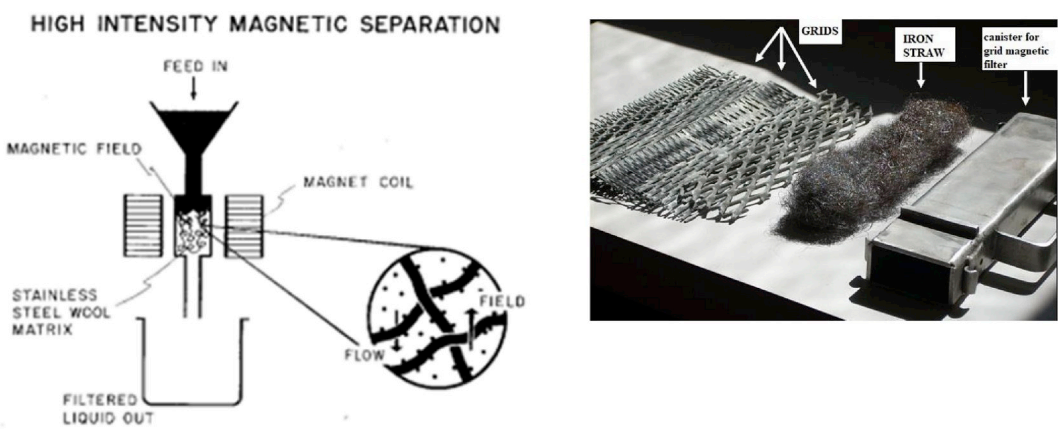


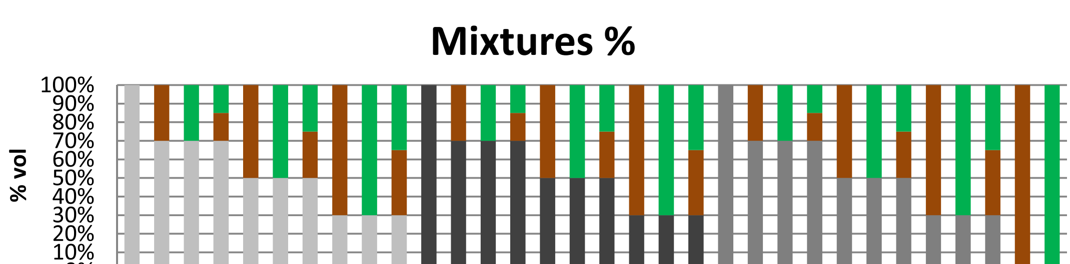
Fig. 2. Scheme, grids and canister of the Eriez Separator for magnetic separation (Zichella, 2019).

analysis and water content, are not reported in the present paper, as they have already been published elsewhere (Careddu and Dino, 2016; Zichella et al., 2018).

2.3. *Potential applications*

2.3.1. *Environmental application: substrate production*

The experiments tested, at laboratory, greenhouse and field scales, the effectiveness of the materials as artificial substrate, using different percentages of DSS, GSS, MS, compost (Verbania Cooperative Social Resources) and a control substrate (topsoil) of sand (Holcim VA.GA. s.r.



	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	21	22	23	24	25	26	27	28	29	30	31	32
Compost			30	15		50	25		70	35			30	15		50	25		70	35			30	15		50	25		70	35		10
Top soil		30		15	50		25	70		35		30		15	50		25	70		35		30		15	50		25	70		35	10	
MS																					10	70	70	70	50	50	50	30	30	30		
GSS											10	70	70	70	50	50	50	30	30	30												
DSS	10	70	70	70	50	50	50	30	30	30																						

Fig. 3. Different substrate compositions for seed germination and plant growth experiments.

l.) and peat (Vitaflor, Blumenverde) 1:1/V: V.

Seed germination tests and plant growth experiments were carried out on 32 mixtures prepared as in Fig. 3. Two of them were used as control (100% compost or topsoil) and three were composed solely by one type of sludge.

Seed germination tests were conducted using the methodology reported in Nappi and Consiglio (1986), using *Lepidium sativum* (Wundram et al., 1997; Blok et al., 2008). Seeds were placed in Petri dishes on filter paper moistened with 5 ml of either double-distilled water (control) or test solution. The test solution was prepared using 200 g of sample brought to a moisture content of 85% and agitated for 2 h. The aqueous extract was filtrated (test solution) and diluted with distilled water to obtain concentrations of 30, 50 and 70%; the experiments were run in five replicates. Dishes were incubated in a thermostatic chamber for 24 h at 27°C. After this, germinated seeds were counted, and the root elongation was measured with a ruler.

Plant growth experiments were performed in triplicate. The different mixtures were placed in 300-ml pots at 27°C and watered daily for 21 days. After this, the plant tops were cut and oven-dried at 40°C, and biomass was determined by weighing.

Germination and growth rates were calculated in terms of germination and plant growth indices as follows:

$$I_g = \frac{Gt \cdot Lt}{Gc \cdot Lc} \cdot 100 \quad (1)$$

$$G_m = \frac{Bt}{Bc} \cdot 100 \quad (2)$$

where.

I_g = germination index (in %);
Gc = mean value of germinated seeds using test solution from samples;
Gt = mean value of germinated seeds using control solution (double distilled water);
Lc = mean value of length of roots of seeds using test solution from samples;
Lt = mean value of length of roots of seeds using control solution;
G_m = plant growth index (in %);
Bt = weight of biomass of plants obtained using extractive waste in sand and compost mixture;
Bc = weight of biomass of plants obtained using only sand and compost mixture.

The 12 blends composed at least for 50% of sludge that yielded better results in seed germination and plant growth experiments were characterised at the end of the experiment.

In addition to the above-explained chemical analyses, materials were agronomically characterised using the official Italian methods (Colombo and Miano, 2015). All samples were analysed for pH (1:2.5, soil: water), total carbon (TC), nitrogen (TN) (CE Instruments, NA2100 Elemental Analyzer) and carbonates (volumetric method, ISO 10,693). Cation exchange capacity (CEC) was determined using barium chloride and triethanolamine (ISO 13,536); plant-available P was determined through the Olsen method (Colombo and Miano, 2015).

Two mixes were tested in a greenhouse experiment as growing substrate for green coverings against a control soil composed of sand, topsoil and compost (50:25:25%/V:).

For the greenhouse experiment, six vessels were filled with 18 l of each mixture and six vessels with the control soil and placed in a greenhouse for 30 days.

On these mixture, two seed mixes were sown:

- Mix 1 - grasses with a predominant function of the formation of a stable rind, which retains the soil avoiding erosion: *Lolium perenne* L. 30%, *Festuca rubra* L. 25%, *Poa pratensis* L. 40% (S.i.S. Società italiana s.p.a);
- Mix 2 - grasses and legumes with the purpose of improving the soil in terms of N addition: *Trifolium pratense* L. 3%, *Lolium perenne* L. 26%, *Trifolium hybridum* L. 6%, *Trifolium repens* L. 7%, *Dactylis glomerata* L. 15%, *Festuca arundinacea* L. 18%, *Lolium italicum* L. 15%, *Phleum climax* L. 10% (Continental Semences s.p.a.).

After 4 days, seed germination was verified with respect to the control soil. After 30 days, plant biomass production was evaluated by cutting all the plants to the collar. During the revegetation test, a photographic survey of the phenological development and of possible occurrences of plant diseases was carried out. After the cutting, the pots were left in the greenhouse under the same environmental conditions for 45 days. The samples were subjected to the same analytical set of initial samples. In addition, microbial biomass was determined via the fumigation method (MiPAF, 2004).

After the greenhouse experiment, one sludge mixture and one seed mixture were selected for the field test, conducted in the VCO area. We selected a site with cold winters, hot summers and abundant rainfall, at 200 m above sea level. On this site, four trenches with dimensions of 1.5 × 4 m were excavated to a depth of 65 cm. Two of them were filled with the control substrate, composed of 50% sand and 50% commercial soil, while the other two were filled with a mixture consisting of 50% MS, 25% sand and 25% compost.

The mixtures were placed on the top of a 15-cm layer of gravel and a 10-cm sand layer. The trenches were compacted and left to rest for 2 days before sowing. Sowing was performed using selected seeds. After 8 weeks, the mixtures and the reference (control substrate) were sampled and characterised.

2.3.2. Civil application: CLSM and plaster production

The CLSM are self-levelling cementitious materials used as a filling and must satisfy technical requirements according to ACI229 R-99. The CLSM applications considered in this study are road foundations, pipe laying beds and thermal insulation with air-based cement mortars. The most important requirement for CLSM is the effect of the thermal conductivity of the filling material on the transmission line performance. The sub-services, high-tension electric cables, present in the road sub-floors of the tunnels, generate heat. The surrounding material must therefore be able to dissipate heat to keep the cables safe and in good working order. For this purpose, the sludge sample VCO GSS was used, which contains a high percentage of heavy metals due to the metal grit used during cutting. The presence of heavy metals and the quartz content of the sawed silicate rocks increase thermal conductivity, which is required for CLSM. Two mix-design experiments were performed with different cement dosages: 50 and 100 kg/m^3. Based on the flowability test, the proper mix (cement, quartzite aggregate, sludge and water) was determined, and two cylindrical specimens per sample were prepared. The dimension of each specimen was 200 mm by height and 100 mm by diameter. Table 2 shows the data for the mix design used for the CLSM specimen preparation.

The analysis performed on CLSM mixes, and here presented, were thermal conductivity at 18 curing times and in dry condition, chemical analysis and leaching test. Leaching test and chemical analysis are

Table 2
Mix design of the CLSM samples tested (referred to 1 m^3 of sample for all the components).

Component (kg/m^3)	M50	M100
Cement	50	100
Quartzite 0-8	1236.3	1212.8
Quartzite 8-14	191.5	187.9
VCO GSS sludge	313.4	187.9
Additive	0.25	0.50
Water	308.9	305.6

carried out to verify the release of metals, comparing it with the results of leaching and chemical analysis of the original sawing sludge sample. With regard to this application, other complementary analyses will be reported in a future paper.

Plaster, in contrast to CLSM, must meet the following requirements: good adhesion with underlying support, impermeability, thermal and acoustic insulation, mechanical resistance and ability to allow transpiration processes through the wall perimeters. Plaster is used as a compact coating to cover the masonry, with a thickness of 1.5–2 cm; in some cases, thickness can reach 10 cm. It is composed of a binding mortar that incorporates sand with a particle size distribution that does not exceed 2 mm. The addition of silica generally increases the mechanical characteristics of the plaster and improves the resistance to salt crystallisation. For this purpose, the LS-DSS sludge was used both because of its fine particle size distribution and extremely low metal content. Table 3 shows the characteristics of the plaster mix design chosen for the presented tests.

The tests carried out on plaster were the same as those for CLSM. Thermal conductivity was determined to verify the thermal insulation, which represents a fundamental requirement for plaster application. Chemical analyses and leaching tests were performed to verify the metal release in relation to the original sludge sample. In our future article, regarding this application, more specific analyses will be reported.

3. Results

3.1. Material characterisation

The results of the chemical analyses on the contaminants carried out on the three sludge types are shown in Table 4, together with the Italian legislative thresholds for the use in residential (Column A) or commercial areas (Column B) (M.A.T.T., 2006).

The chemical composition of the wastes (Table 4) suggests an abundance of various metals and TPH, sometimes exceeding the threshold values for their use in residential (A) or in industrial (B) areas. This requires a standardised residual sludge management to prevent water and soil pollution.

Among the metals, Co was the most concentrated one in DSS samples, with concentrations higher than the residential area thresholds, while Cr VI presented problematic values in both categories, although they were higher in GSS samples than in DSS ones. Likewise, the TPH concentrations exceeded the limits for residential areas in almost all samples, with one value exceeding the threshold for commercial areas. These compounds were derived from lubricants used for both gang-saw and diamond frame-saw technologies.

The results of the leaching tests (not reported) presented concentrations below the threshold values established by the current legislation on wastes (M.A.T.T., 2006) for most of the samples and most of the elements, indicating that these materials were, generally, not harmful in regard to water pollution. Only one sample of DSS slightly exceeded the maximum permitted concentrations of Ni and Cr.

The results of the compost and soil analyses were, for all samples, below the threshold limits established by the Italian legislation for the use as fertilisers (Mi.P.A.F., 2010), making them suitable as artificial substrates.

Table 3
Mix design of the plaster samples tested.

Component	Unit	M.U.
Portland Cement 42.5R	kg/m^3	313.0
Lime NHL 3.5	kg/m^3	38.0
Luserna Flaming sand 0/3 mm	kg/m^3	813.0
Filler MCD (*)	kg/m^3	187.0
Micro fibre (**)	kg/m^3	0.3
Natural Foam 72–75 g/m^3	m^3/m^3	0.37–0.38

Table 5 Percentages of the magnetic fraction for each sample. The samples from GSS had a greater magnetic percentage compared to those from DSS. Comparing the two sludges derived from gang-saw cutting, VCO-GSS and LS-GSS LS had a higher percentage of the magnetic fraction.

The typology of processed stone and its workability affects the wear of the tools and, therefore, the concentration of metals in the sludge (Zichella et al., 2018). High-field magnetic separation attracts not only magnetic metals, but also part of those minerals that are paramagnetic, such as biotites and amphiboles containing iron. Furthermore, it should be noted that not all metals are magnetic. Therefore, in the final magnetic product, it is also possible to find minerals, while in the amagnetic product, we can find metals such as Zn and Cu, which are diamagnetic Fig. 4 shows the comparison between the results of MVG sample before and after magnetic separation. It is possible to observe in the magnetic fraction also the presence of minerals as quartz and feldspar; in fact during the magnetic separation, iron-based minerals such as mica, feldspars and pyrite, have been also captured together with metals. For this reason, the efficiency of the separation was evaluated taking into account the amount of the non-magnetic fraction collected together with the magnetic fraction (trapping phenomenon).

3.2. Potential applications

3.2.1. Substrate production

The use as substrate has previously been assessed by Dino et al. (2006, 2015a), using a limited number of samples from a single quarry area located in the LS quarry basin, while in this experiment, for the first time, samples from different quarries and areas were assessed to obtain a broader overview of the possible applications of these substrates.

To select a better combination between sludge and organic materials (compost and topsoil), phytotoxicity tests were conducted on the mixtures prepared with different percentages of each material; the results are reported in Fig. 5. With regard to the germination index (IG), a lower percentage of sludge corresponded to a higher IG for most of the combinations (Fig. 5a).

All mixtures reached values higher than threshold values established for the use as fertiliser (an IG of about 60%) (Mi.P.A.F., 2010), although *Lepidium sativum* showed a better performance in a mixture of 50% sludge and 50% topsoil or compost.

The plant growth index (GM) for all mixes is represented in Fig. 5b, where the biomass produced from each mixture is compared to the production of the topsoil made of sand and peat (1:1, v: v). In this case, some of the mixtures, in particular those with DSS and MS, showed higher values than topsoil, with the better results achieved from the same mixture as in the IG tests. Conversely, GSS mixtures produced lower biomass quantities in all combinations.

Based on the results obtained from the phytotoxicity test, 12 mixtures were chemically and agronomically characterised (Table 6). All three sludge types had some common characteristics that could be problematic (although not prohibiting) for plant development, such as:

- very fine texture;
- strongly alkaline pH;
- absence of N and plant-available P (P Olsen);
- extremely low levels of organic C;
- poor cation exchange capacity (CEC).

These characteristics should be taken into account before using residual sludge as feeding material, together with organic and inert compounds, for artificial substrate production.

Most of the samples did not contain inorganic contaminants, except for the MS sample, presenting a concentration of Ni exceeding the limit allowed for residential soils (M.A.T.T., 2006).

As all mixtures presented similar characteristics from an agronomic point of view, we selected the worst cases in terms of heavy metal

Table 4
Chemical parameters of the three sludge types. All values are expressed in mg/kg of dry sample. Data for Be, Cd, Se, Sn, Tl, and cyanide are not reported as they were below the detection limit (<0.5 mg/kg) for all samples. Fluoride levels were below the detection limit (<2.5 mg/kg) for most samples. Values in bold exceeded the threshold values for residential areas.

Sample	Sb	As	Co	Cr tot	Cr VI	Hg	Ni	Pb	Cu	V	Zn	TPH
GSS1-VCO	2.4	11.7	9.4	67	**22**	<0.5	33	<2.5	116	37	60	**57**
GSS2-VCO	3.1	10.3	7.8	42	**31**	0.5	32	3.7	96	28	49	**116**
GSS3-VCO	2.2	10.8	5.2	25	**45**	8.7	30	2.7	109	28	50	43
GSS4-VCO	3.6	13.2	10	130	**70**	<0.5	87	4.7	**186**	22	54	**112**
GSS5-VCO	4.2	11.4	9.0	63	**23**	<0.5	25	<2.5	74	28	55	**80**
GSS-LS	/	<0.1	5.4	**250**	< 5	<0.01	**133**	53	**181**	<0.1	26	/
DSS1-VCO	1.8	**37**	**34**	8.7	**3.8**	0.7	6.4	19	53	16	103	**113**
DSS2-VCO	3.1	4.9	**67**	5.2	1.2	0.7	17	<2.5	49	22	53	38
DSS3-VCO	2.3	7.2	**20**	7.6	**2.5**	0.7	12	11	57	13	53	**167**
DSS4-VCO	2.6	4.4	**99**	16	**2.2**	0.7	9.7	<2.5	117	31	65	**889**
DSS5-VCO	2.0	5.9	**41**	6.6	**4.0**	0.7	4.5	<2.5	35	36	57	**433**
DSS6-VCO	2.7	4.5	**47**	<5	1.0	0.5	2.6	4.0	50	28	60	48
DSS7-VCO	3.1	<2.5	**83**	19	**15**	0.6	2.7	<2.5	94	26	51	**161**
DSS-LS	/	<0.1	**26**	18	**< 5**	<0.01	<0.01	23	42	<1	25	/
MS1-VCO	2.5	14.1	4	210	**64**	<0.5	117	2.5	**225**	31	56	**67**
MS2-VCO	3.2	9.2	10	56	**42**	0.6	39	<2.5	87	30	56	**80**
MS3-VCO	3.0	7.9	7.2	19	**12**	0.8	13	3.0	36	31	70	**121**
MS-LS	/	<0.1	**32**	8.7	<1	<0.01	18	19	108	<1	109	/
Limits column A	10	20	20	150	2	1	120	100	120	90	150	50
Limits column B	30	50	250	800	15	5	500	1000	600	250	1500	750

Table 5
Results of magnetic separation for VCO and LS samples.

Sludge code	Magnetic fraction [Wt, %]	Amagnetic fraction [Wt, %]
VCO - GSS	2.96	97.04
VCO - MS	2.7	97.3
VCO - DSS	1.5	98.5
LS - GSS	6.3	93.7
LS - MS	2.46	97.54
LS - DSS	1.95	98.05

contamination, thus we used the samples containing the maximum quantity of MS sludge having acceptable IG and GM results.

The mixtures selected for the greenhouse test are highlighted in Table 6: MS + Compost (50/50%, v/v), hereafter MIX A, and MS + Topsoil (50/50%, v/v), hereafter MIX B, together with a control substrate composed of sand, T and C (50%/25%/25%).

The two selected mixtures had different agronomic characteristics, with Mix B being sandier than Mix A, thus more easily colonisable by plants, although with a lower nutrient concentration.

Fig. 6 shows the results of this experiment; Mix A was the substrate with the higher biomass production for both seed mixes, most likely because of the considerably higher N and P concentrations.

From this starting point, we moved to the field trial, using Mix A as artificial substrate and the Seed Mix 2 to compare the effective use of such a substrate directly where the wastes are produced, using a circular and a zero waste approach. In this case, the substrates were studied in duplicate 6-m^2 trenches and were sampled 8 weeks after sowing. Table 7 shows the results related to the different substrates.

During the field trial, the chemical properties of the substrates did not change substantially; microbial biomass increased in both substrates, while Olsen P decreased. After 8 weeks, plants were harvested, and the produced biomass was weighed. The artificial substrate had a productivity similar to that of the control, although slightly lower.

3.2.2. CLSM and plaster production

Thermal conductivity analysis was performed by means of a KD2 Pro device. For this, a needle probe (RK1 model, 6 cm long, 3.9 mm diameter), introduced into the cylindrical specimen via a special hole, was used. The information provided by the device consisted of the value of thermal conductivity [W/m*K], specimen temperature [K] and the error of measurements. Measurements were performed at 18 curing days and under dry conditions (48 h in the oven at 60°C); Table 8 shows the results.

The cement content affects the results of the thermal conductivity test. At 18 curing days and under dry conditions, the trend was similar. To demonstrate the efficiency of CLSM, it is necessary to test it under dry conditions, i.e. in real conditions. One of the objectives of the use of CLSM is to maintain a high conductivity regardless of the humidity values. The material needs to be able to dissipate the heat generated by the cables under dry conditions. The thermal conductivity value for CLSM product with a standard aggregate was about 0.5–0.8 W/m*K. Replacing the standard aggregate with sawing sludge powder at 18 curing days, a higher thermal conductivity value was obtained due to the quartz and metal contents of the sludge. Also, under dry conditions, good values were obtained, similar to the standard CLSM product.

Thermal conductivity for plaster application showed a good behaviour of the plaster product already at 18 curing days and a good thermal stability of the mix design in comparison with the dry condition.

The results of the chemical analysis (Table 9) and of the leaching tests (Table 10) demonstrated the good chemical properties of the final products and showed that both products were not contaminated and did not release metals to the environment.

The only metal with values above the threshold values was Cr in the CLSM application, probably because of the release of this metal from moulds used to perform the specimens.

4. Discussion

Most of the sludge from sawing processes could be reused in industrial and commercial areas (limits below column B of Legislative Decree 152/2006). A preliminary analysis of the chemical composition is crucial to choose the correct future reuse of the sludge both in civil engineering and in environmental applications. For example, GSS sludge presented high Fe concentrations and might therefore be useful for CLSM production. In contrast, DSS and MS, with lower Fe contents, could be employed as feeding material for plaster and substrate production.

Magnetic separation proved to be useful to preliminarily characterise the sludge in terms of Fe content, with the aim to estimate the average of metals contents and, consequently, the potential application of the different sludge types. For sludge types with metal concentrations higher than the limits, pre-treatment could be provided through magnetic separation. Such an approach can be useful at the industrial level

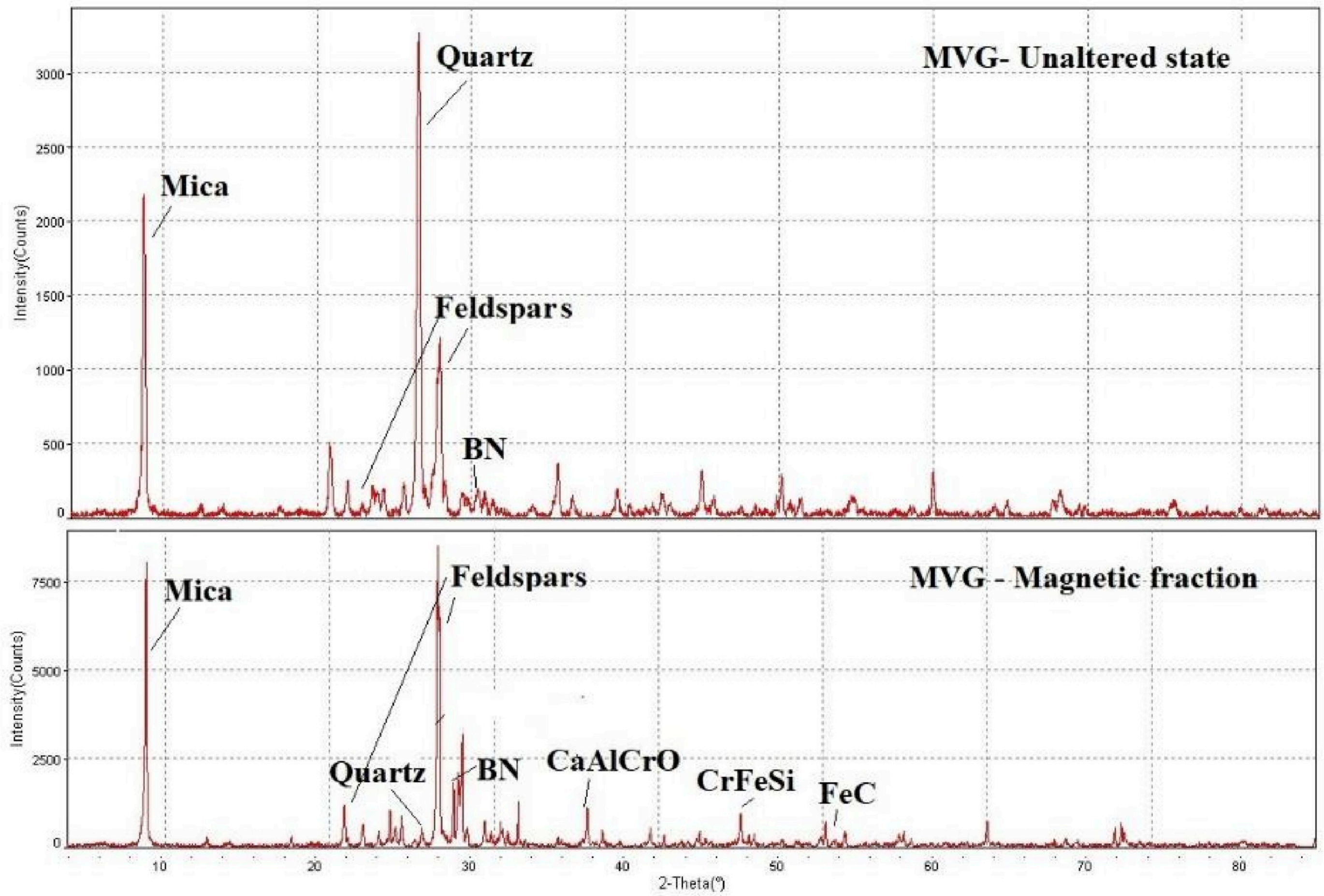


Fig. 4. VCO- GSS XRD measurement from Zichella (2019). In the upper graph, the XRD of the whole sludge sample is demonstrated, while the lower graph shows that of the magnetic fraction.

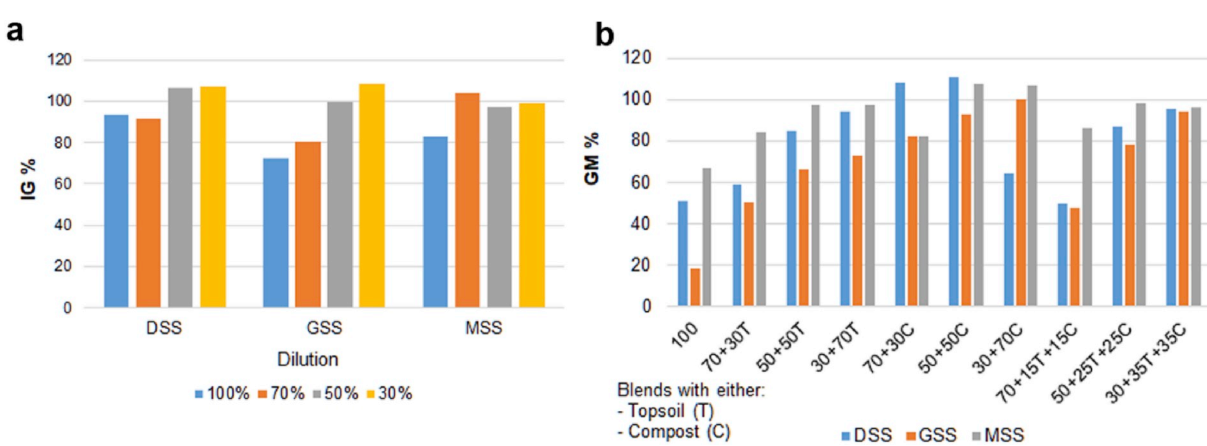


Fig. 5. Results of the phytotoxicity tests conducted on the mixtures composed of one of the three sludges and an organic material (either compost or topsoil). In graph b, the plant growth index (GM) is expressed as a percentage of the topsoil result.

as it does not change the state of the material. This method did not produce any other substances, simply splitting the sludge in two by-products (magnetic and amagnetic) with a high degree of purity. The magnetic product, rich in metals, can be reused in other productive sectors or disposed of in landfills (with an economic advantage due to the reduction of the quantities to be disposed). The amagnetic product, mostly lithoid material, could be reused in the building sector. The recovery of sludge as it is and/or after magnetic separation could turn a unidirectional system into a circular system.

For the use as substrate, we selected the worst-case scenario in terms of metal contamination from the mixes presenting good agronomic characteristics. The mixes containing the maximum quantity of MS sludge with acceptable IG and GM results were chosen as test materials for the greenhouse test. The MS sludge was less contaminated than sludge from DSS and GSS, respectively rich in Co, Cr and TPH (the first one) and Ni, Cr, Cu and TPH (the second one), thus less phytotoxic.

Table 6
Chemical-physical analyses of the starting materials and the selected mixtures with sludge. Bold values indicate values beyond the threshold value; n.d. = not determined. Selected materials for greenhouse tests are in red. GSS: Gangue-saw sludge, DSS: Diamond frame-saw sludge, MS: Mixed sludge. C: compost, T: Top soil.

		Blends		100			50 + 50 C			50 + 50 T			50 + 25 C + 25 T			70 + 30 C		
Parameter	**Unit**	**Compost**	**Topsoil**	**DSS**	**GSS**	**MSS**	**DSS**	**GSS**	**MSS**	**DSS**	**GSS**	**MSS**	**DSS**	**GSS**	**MSS**	**DSS**	**GSS**	**MSS**
	Sand %	n.d	n.d	n.d	n.d	n.d	53	62	69	62	72	77	60	76	75	41	57	67
Texture	Silt %	n.d	n.d	n.d	n.d	n.d	43	34	29	35	27	23	37	23	25	52	38	30
	Clay %	n.d	n.d	n.d	n.d	n.d	4	4	2	3	1	0	3	1	0	7	5	3
pH		8.2	7.1	10	10.1	10.2	8.9	9.7	9.7	8.4	9.7	9.7	8.6	9.8	9.8	9.2	9.9	9.8
P Olsen	mg/kg	223	0.26	n.d	n.d	n.d	71	52	61	n.d.	n.d.	n.d.	35	27	25	42	17	14
N tot	%	1.55	0.06	n.d	n.d	n.d	0.45	0.39	0.37	0.03	0.04	0.03	0.21	0.17	0.1	0.2	0.15	0.19
CEC	cmol/kg	64.1	3.4	2.5	2.1	4.7	20.5	23.4	17.4	4.8	5.1	5	11.2	10.5	9.9	9.9	10	7.7
C org	g/kg	26.0	3.2	0.1	0.6	0.5	5.9	5.5	5.1	1.7	2	1.3	3.5	3.3	1.9	2.7	2.6	3.2
Fe	g/kg	16	13	13	139	66	14	42	33	13	24	20	13	21	35	14	34	31
Cd	mg/kg	0.5	0.3	0.2	1	0.8	0.1	0.8	0.6	0.1	0.6	0.5	0.1	0.6	0.5	0.1	0.5	0.4
Co	mg/kg	10	9	**73**	14	18	**49**	13	15	**38**	12	14	**43**	12	17	**56**	13	14
Cr	mg/kg	77	84	8	88	119	29	89	107	59	101	112	42	97	114	27	84	116
Cu	mg/kg	125.8	10	82	81	117	68	98	104	41	58	69	59	80	96	62	97	85
Ni	mg/kg	11.8	34.9	18	96	**157**	42	95	**138**	68	106	**147**	58	105	**147**	40	90	**150**
Pb	mg/kg	37.6	11	17	15	14	29	26	22	14	14	13	21	18	15	21	26	18
Zn	mg/kg	136.4	28	63	70	62	104	103	90	46	53	50	70	75	65	77	96	62

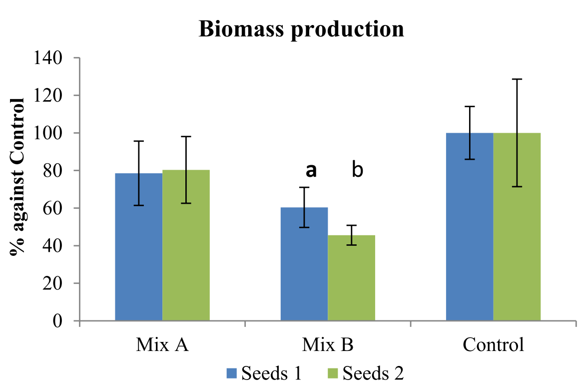


Fig. 6. Biomass production of each selected mixture and of a control substrate using two different seed mixes. Different letters indicate significant differences; bars represent error bars.

In the greenhouse experiment, Mix A (produced using MS mixed with compost) showed good performance in terms of biomass production, being not statistically different from the control substrate. This blend resulted in a lower microbial biomass than the control, probably because the microbial population came only from the compost and, because of the limited time, could not adapt to the sludge.

The results of the field trial were positive, providing outputs similar to the substrate used as reference. These results were probably associated with the chemical characteristics of this mixture, with high N and P

Table 7
Analysis of the substrates before sowing (T0), 1 week after sowing (T1) and at the end of the field trial (T8).

	Unit	T0	Mix B	T8	T0	Control	T8
			T1			T1	
pH		8.6	8.5	8.2	7.6	7.7	7.8
CEC	meq/100 g	2.7	2.1	4.5	17.7	12.5	12.7
P Olsen	mg/kg	17	10.5	13.4	3.8	2.7	2.5
N tot	g/kg	0.7	0.7	0.8	1.8	1.6	1.6
C org	g/kg	12.7	12.2	13.2	86.9	82.1	81
Microbial biomass	mg C/kg	511	473	589	1623	2338	2159
Biomass production				78%			100%

concentrations, necessary for biomass growth, similar to a typical agricultural soil. The MS sludge did not present high TPH concentrations, with metals being the major contaminants, but with this treatment, TPH contamination could also be reduced (Dino et al., 2015a).

These promising results, obtained by mixing sludge and compost, suggest the use of sludge as a substrate to be used for the recovery of industrial areas as a substitution for natural soils.

For civil applications, two recoveries were planned, based on the sludge metal content. The first recovery, as CLSM, provided the use of a filler that increased the capacity to dissipate thermal energy, due to the

Table 8
Thermal conductivity results on VCO GSS (gang-saw) sludge for two mix designs with cement contents of 50 and 100 kg/m^3, respectively, and LS DSS for plaster.

	Thermal conductivity at 18 curing days [W/m*K]	Thermal conductivity under dry conditions [W/m*K]
VCO GSS (50) for CLSM	0.97	0.51
VCO GSS (100) for CLSM	1.16	0.84
LS DSS for plaster	0.3	0.2

passage in the sub-road network of sub-services. The second recovery, as thermals-eco-mortar for plaster, provided the use of a filler that would allow thermal insulation, combined with good mechanical resistance. Concerning CLSM products, we suggest that they meet the requirements for this application. Some observations can be highlighted on the basis of the obtained results:

- an increase of cement content (50 to 100 kg/m^3) worsens the fluidity properties, while under dry conditions, resistance and thermal conductivity are improved;
- thermal conductivity values are better for CLSM with standard aggregates due to the sawing sludge quartz and metal content;
- the leaching test on CLSM samples shows that there is no release of metals, as the cement mortar well incorporates the sawing sludge metals.

Concerning thermal-eco-mortar for plaster application, DSS improved the rheological, thermal and physical performance, conferring a light macroporous cellular structure via the addition of organic foam. This characteristic facilitates plaster installation even at high thicknesses. The thermal conductivity obtained exceed the values of standard plaster, making this eco-friendly plaster an excellent product, ideal for energy saving in buildings.

5. Conclusions

The present research shows the results concerning the sustainable way to manage and recover residual sludge coming from the Verbano Cusio Ossola and Luserna Stone quarry basins. The characterisation phase is fundamental to decide upon the best applications to adopt, depending on the chemical and magnetic characteristics of the sludge as such. Furthermore, because of the introduced procedure for residual sludge characterisation (physical and chemical) and management (summarised in Fig. 1), costs and environmental impacts connected to their disposal can be reduced. It is fundamental to evidence that the products obtained from the treatment can be employed as integrative/substituting materials in addition to natural ones, reducing the pressure on natural resources.

Applying a fast characterisation by means of magnetic separation facilitates sawing sludge recovery without any preliminary treatment: on the one hand, products characterised by a low magnetic fraction content can be used for plaster or substrate production, on the other hand, products characterised by a high magnetic fraction content could be used for CLSM production. Thanks to this pre-screening, it is possible to save costs related to pre-treatment (the different materials can be directly employed as feeding materials for the above-mentioned products) and to reduce the impacts associated with landfilling.

Moreover, high-field magnetic separation can be the most appropriate pre-treatment, as it is simple, fast and effective in case the chemical characterisation shows a metal content above the established threshold limits. Consequently, as it does not change the initial state of the sample, but performs a simple separation of the components, high-field magnetic separation could be included in the Art. 5 of the Directive 2008/98/EC definition of "Normal industrial practice".

The use of residual sludge for substrate production seems to be highly promising: the substrate can be used for quarry rehabilitation and in the rehabilitation of industrial sites. This new product ("new soil") could be included in the list of the materials to use as addition to topsoil for civil works and infrastructures (e.g. railway embankments, roundabouts, etc …). It has to be highlighted that the presented methodology, first applied for sludge recovery in substrate production, has recently

Table 9
Chemical analysis of CLSM products and the plaster product (mg/kg). Last row: Standard threshold limit concentration according to Lgs. D. 152/2006, Annex 5, title IV and Art. 8 of M.D. February 05, 1998.

	Fe	Ba	Cu	Zn	Co	Ni	V	As	Cd	Cr	Pb	Be	Cr VI	Hg	Se
	mg/kg														
VCO GSS 50	5120	29	9	10	2	7	7	<1	<1	11	1	<1	<5	<5	<1
VCO GSS 100	6993	26	15	17	3	11	13	1	<1	17	2	<1	<5	<5	<1
LS-DSS plaster	7861	94	41	91	5	13	41	6	<1	15	10	1	<5	<5	<1
Limit column B	/	/	600	1500	250	500	/	50	15	800	1000	/	15	5	/

Table 10
Leaching test on CLSM products and the plaster product. Standard threshold limit concentration according to Lgs. D. 152/2006, Annex 5, title IV. In bold values exceeding the threshold.

	Fe	Ba	Cu	Zn	Co	Ni	V	As	Cd
	mg/L	mg/L	mg/L	mg/L	μg/L	μg/L	μg/L	μg/L	μg/L
VCO GSS 50	0.03	<0.01	<0.001	<0.001	<1	<1	2.28	<1	<1
VCO GSS 100rowhead	0,1	0.03	<0.001	<0.001	<1	<1	<1	<1	<1
LS-DSS plaster	0.40	0.02	0.019	<0.01	<0.005	<0.005	<0.005	<0.01	<0.001
Limit	/	1	0,05	3	250	10	250	50	5

	Cr	Pb	Be	Cr VI	Hg	Se	NO_3	F	SO_4	Cl
	μg/l	μg/l	mg/l	mg/l	mg/l	mg/l	mg/l	mg/l	mg/l	mg/l
VCO GSS 50	95	<1	<0.001	0.08	<0.0005	<0.025	1.5	0.02	49.5	3.1
VCO GSS 100	85	<1	<0.001	0.06	<0.0005	<0.025	1.1	0.02	13.3	3.6
LS-DSS plaster	0.08	<0.01	<0.001	0.07	<0.0005	<0.025	/	/	/	/
Limits	50	50	10	/	1	10	50	1,5	250	100

been adapted and tested for other kinds of waste materials, such as the fine fractions from construction and demolition waste, from rock and soils from excavation works, and from aggregate production. Tests at laboratory, greenhouse and field levels are in progress; the first results evidence the suitability of these new products in environmental applications. The use of "new soil" has to be intended as a sustainable alternative to contrast soil consumption: topsoil has to be used for agricultural purposes or for residential areas, where rehabilitation concerns industrial areas, areas to be remediated, quarries, etc.; these "artificial" substrates, produced and tested on the basis of validated procedures and characterisation, can be applied.

Concluding, the recovery of sludge as such or through pre-treatment would lead to the passage from the currently linear system (waste to landfill) to a circular one (waste to material): this change of perspective is slightly in line with the circular economy politics fixed by the EU commission in 2017. To boost the reuse of residual sludge, it is fundamental to think about it not as a waste, but **as a sustainable resource to exploit**.

The presented and tested applications seem to be the most promising for the real market: LCA and CBA have to be predicted in specific case studies in order to objectively evaluate the profitability and sustainability of the recovery from both environmental and economic points of view.

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