Comparison of alternative management methods for phosphogypsum waste using life cycle analysis

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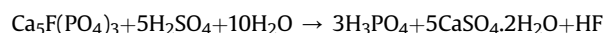
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ABSTRACT

The objective of this paper was to compare the following four alternative management methods of phosphogypsum (PG) waste using Life Cycle Analysis (LCA) and select the one with the lowest environmental footprint: (1) use of PG in brick production, (2) use of PG as soil amendment, (3) use of PG in road construction and (4) disposal of PG in a stack. The results showed that the use of PG waste as soil amendment has a lower environmental footprint than the respective use of conventional gypsum with commercial fertilizers. Similarly, PG waste has a lower environmental footprint when replacing conventional clay in road construction. In contrast, use of PG waste in brick production has a higher footprint than the respective use of clay and sand. Comparison of the four alternative management methods using LCA showed that the use of PG as a soil amendment had the lowest environmental footprint. Disposal of PG waste in stacks, which is currently the most common management method, was ranked as the least preferred one. As an example, using Ecoindicator 99 and the egalitarian perspective total scores were 12.807, −5.334, −0.064 and 15.484 Points (Pt) for brick production, soil amendment, road construction and stack disposal, respectively. Among indicative impact categories, carcinogen scores were 7.643, −0.228, 0.0001 and 0 Pt, respectively. Respiratory inorganics scores were −0.192, −4.540, −0.0383 and 0 Pt, respectively. The ranking based on cost is stack disposal<soil amendment≪road construction<brick production. For any alternative PG waste use, it is required that activity concentration of naturally occurring radionuclides is below the respective European Union standards. Using LCA is a useful approach for comparing PG waste valorization methods within the context of Circular Economy.

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1. Introduction

Phosphogypsum (PG) is a waste byproduct from phosphate rock processing by the wet acid method, for production of phosphoric acid used in fertilizer industry. The process is described by the reaction (Tayibi et al., 2009):

$$Ca_5F(PO_4)_3 + 5H_2SO_4 + 10H_2O \rightarrow 3H_3PO_4 + 5CaSO_4.2H_2O + HF$$

PG is the calcium sulfate dihydrate ($CaSO_4.2H_2O$), produced at a rate of 5 tonnes of PG per tonne of phosphoric acid. Impurities encountered in PG include phosphates (H_3PO_4, $Ca(H_2PO_4)_2.H_2O$, $CaHPO_4.2H_2O$ and $Ca_3(PO_4)_2$), residual acids, fluorides (NaF, CaF_2, Na_3AlF_6, Na_3FeF_6 and Na_2SiF_6), trace metals (Cd, Zn, Cu, Cr) and organic matter (e.g., amines and ketones adhered to gypsum

crystals) (Rutherford et al., 1996). PG also contains radionuclides. Approximately 80% of naturally occurring Ra in the phosphate rock ends up in PG, whereas 86% of U and 70% of Th end up in phosphoric acid (Tayibi et al., 2009). PG composition has been reported in several studies, e.g., Tayibi et al. (2009), Renteria-Villalobos et al. (2010), Macias et al. (2017a) and Zmemla et al. (2020). Hentati et al. (2015) and Park et al. (2016) reported on PG toxicity.

World phosphate rock production estimates range between 225 Mt in 2014 and 258 Mt in 2018 (Macias et al., 2017b). World annual PG waste production is estimated in the range of 100—280 Mt (Yang et al., 2009; Tayibi et al., 2009). Approximately 85% of PG waste produced is disposed of in stacks, without treatment, which occupy large land areas and cause significant environmental damage. According to Rutherford et al. (1994), storage of PG waste in stacks causes: (1) atmospheric pollution due to

emission of fluorides, toxic metals, radioactive dust and radon, (2) groundwater pollution due to emission of sulfate and phosphate anions, acidity, trace metals and radionuclides, (3) surface runoff, (4) workers exposure to γ-radiation and (5) problems related to geotechnical stack stability. The radioactivity content is a significant barrier to increasing the amount of PG waste for alternative use. Nevertheless, PG waste valorization in building materials, soil amendments and fertilizers and manufacturing of Portland cement has been employed, but accounts only for the remainder 15% of annual world production (Tayibi et al., 2009).

Several potential PG applications have been investigated and/or proposed within the context of circular economy. For example, PG application in pavement and road construction (Silva et al., 2019; de Rezende et al., 2017; Folek et al., 2011; Qiao et al., 2010), brick manufacturing (Yang et al., 2009; Ajam et al., 2009; Kumar, 2002, 2003), cement production (Rosales et al., 2020; Mun et al., 2007), binder production (Nizeviciene et al., 2018; Kuryatnyk et al., 2008), soil stabilization (Hentati et al., 2015; James et al., 2014) and coastal applications (Deshpande, 2003) have been reported. In addition, PG was used as raw material for recovery of rare earth elements (Rychkov et al., 2018), production of sodium sulfate and calcium silicate (Zemni et al., 2018), synthesis of α-hemihydrate gypsum (Ma et al., 2018) and hydrated gypsum following extraction of natural radionuclides (Moreira et al., 2018). Furthermore, Contreras et al. (2015) confirmed that PG could be used for mineral CO_2 sequestration. Following a detailed environmental and geotechnical laboratory scale investigation, Gaidajis et al. (2017) concluded that Greek PG may be considered for secondary embankment construction and mine and quarry rehabilitation.

Canovas et al. (2018) presented a review of PG waste valorization methods and critically described their advantages and disadvantages, also recommending the use Life Cycle Analysis (LCA) to assess their secondary pollution. LCA is a powerful decision-making tool and a methodology which can be used to assess alternative management methods for PG waste. LCA has been used to assess environmental impact of a process or a material during its life cycle and to compute its environmental footprint. This may include climate change, depletion of stratospheric ozone, creation of tropospheric ozone, eutrophication, acid rain, toxic stress to humans and ecosystems, depletion of natural resources, water use, land use, noise etc. (Rebitzer et al., 2004). LCA should be conducted according to International Organization for Standardization (ISO 14040, ISO 14044).

Many studies along with review papers dealing with environmental impact and potential applications of PG waste have been published (Canovas et al., 2018). However, to the authors' knowledge, there are very few studies comparing PG valorization methods. For example, Kulczycka et al. (2016) used LCA to assess environmental impact from PG land disposal and recovery of rare earth elements by leaching with sulfuric acid. Mohammed et al. (2018) used LCA to evaluate PG conversion to paper and fertilizer and compare with conventional products. Belboom et al. (2015) applied LCA to compare phosphoric acid production using the wet di-hemihydrate process and the thermal process. Zhang et al. (2017) performed LCA assessment of diammonium- and monoammonium-phosphate fertilizer production in China and compared the environmental impacts caused by PG utilization in production of cement relievers, ammonium sulfate, NPK compound fertilizer and landfill disposal. They concluded that production of cement relievers had the lowest environmental burden. However, there are potential PG applications, which have not been addressed by LCA studies.

The overall objective of this work was to compare the environmental footprint of alternative management methods for PG waste using LCA and select the one with the lowest environmental footprint. The following four alternative management methods

were investigated: (1) Use of PG in brick production, (2) use of PG as soil amendment, (3) use of PG in road construction and (4) disposal of PG in a stack. Apparently, this work covers a knowledge gap regarding PG management methods, which were not addressed by previous LCA studies and this is the novelty of the work.

This is a research paper mainly addressed to the scientific community, but the conclusions could be presented to PG waste stakeholders, such as the fertilizer industry, the local government and decision makers and the general public, affected by PG stacks and seeking alternative management methods.

The analysis was conducted mainly from the egalitarian perspective (citizen group point of view) and was complemented by the hierarchist perspective (government point of view) and individualist perspective (market point of view).

2. Methodology

The ISO 14044 standards (ISO, 2006a,b) and the ILCD handbook guidelines (EC, 2010, 2011) were followed.

2.1. Goal definition

The goal of this work was to assess the environmental impact of four alternative management methods for PG waste, using LCA, and select the one with the lowest environmental footprint. The following four alternative management methods were investigated: (1) Use of PG in brick production, (2) use of PG as soil amendment, (3) use of PG in road construction and (4) disposal of PG in a stack.

The study has the following limitations: Except for radionuclide content, site-specific data from Greek PG production sites were not available by the producers, when this study was conducted. Therefore, assumptions were made and results of previously published studies were used as a basis to compute emission inventories for the alternative management methods described in this study. In addition, the main impact assessment method used (Ecoindicator 99) does not provide damage factors for emissions of some relevant contaminants, such as sulfate and phosphate anions, to soil and water. To address the problem of eutrophication and acidification, an alternative method (CML 2 Baseline 2000) was used (with other deficiencies, however) for conducting the impact assessment (Frischknecht et al., 2007).

2.2. Scope definition

2.2.1. Functional unit

The functional unit of LCA will be the management of 1 tonne of PG, taken from a stack. It was assumed that the moisture of the stack did not exceed 10%. This is a unitary functional unit, defined by its waste composition (Table 1). The majority of LCA studies on solid waste management used a functional unit of 1 tonne of waste (Laurent et al., 2014; Kulczycka et al., 2016). From the context point of view, the study is classified as a C1 case, according to the ILCD guidelines (EC, 2010). As such, attributional LCA with average data will be used (Finnveden et al., 2009; Laurent et al., 2014).

2.2.2. System boundaries

The system boundaries separate the system being studied from the ecosphere and the rest of technosphere. Therefore, energy and material flows in and out of these boundaries must be included in life cycle inventory (EC, 2010). In defining system boundaries, three components were considered: (1) capital goods, (2) collection and transportation of PG and other materials and (3) transport and treatment of secondary products and final residuals (Laurent et al., 2014).

Table 1
Chemical composition of North African PG, according to Renteria-Villalobos et al. (2010), Sebbahi et al. (1997) and Potiriadis et al. (2011).

Major component	%, dw	Trace element	ppm, dw	Trace element	ppm, dw
SiO_2	0.86 ± 0.01	Ba	98 ± 23	Sc	4.7 ± 0.7
Al_2O_3	0.19 ± 0.001	Cd	6 ± 1	Se	<1.75
Fe_2O_3	0.21 ± 0.01	Cr	20 ± 2	Sr	709 ± 115
CaO	38.14 ± 1.70	Cu	21 ± 2	Th	4 ± 0.64
SO_3	48.12 ± 9.04	Ga	1 ± 0.2	Ud	8.3 ± 1.8
K_2O	0.01 ± 0.001	La	86 ± 19	V	4.75 ± 0.63
Na_2O	0.17 ± 0.01	Nb	1 ± 0.2	Y	144 ± 38
P_2O_5	0.69 ± 0.01	Ni	<1	Zn	8 ± 1
TiO_2	0.01 ± 0.0004	Pb	6.2 ± 1.7	Zr	6.2 ± 0.8
F^-	0.15[a]	Rb	2 ± 0.1		
LOI	22				

Radionuclide[b]					Bq/kg
^{238}U					35 ± 18
^{226}Ra					515 ± 150
^{40}K					<20
^{232}Th					<15

[a] According to Sebbahi et al. (1997).
[b] Potiriadis et al. (2011).

The boundaries of the analysis begin with PG withdrawal from the stack and application of the alternative management methods (section 2.1) in a period of 100 years. Production of PG during production of phosphoric acid was not considered.

The required data were provided or computed to ensure a valid comparative study. It was assumed that PG withdrawal did not occur during rain periods and the moisture of the stack did not exceed 10%. Moisture is needed to assess PG dry mass and emissions presented in relevant tables and Appendix A. The assumed value is within the range 8−18% reported by Taha and Seals (1992). In any case, moisture may vary significantly, depending on local weather conditions. Capital goods (e.g., buildings, materials) were excluded, because it was considered that they were the same for both, the conventional materials and PG waste. Materials transportation is discussed in section 2.2.3. Fig. 1 presents material flows and the system boundaries with the dashed lines. Input of raw materials, air and water emissions and PG residuals are shown for each PG waste management method.

2.2.3. Materials transportation

In alternative methods 1 (use of PG in brick production) and 3 (use of PG in road construction), it was assumed that the distance between the PG stack and the point of PG use was longer than the distance between the origin of conventional materials and the point of their use by 10 km. In method 2 (use of PG as a soil amendment), no such difference in distance was considered. It must be emphasized that these assumptions were arbitrary and, depending on the difference in distance, LCA results may be affected. Therefore, a sensitivity analysis was conducted to assess the effect of distance.

2.3. Life cycle inventory analysis

In the alternative management methods listed above, PG waste replaces a conventional material. Therefore, the emissions resulting from the conventional material use were subtracted from the emissions resulting from PG use and were considered in computing life cycle inventories. All emissions were calculated or estimated based on previously published papers, as explained in section 3 (Alternative Management Methods) and in Appendix A. To complete the inventory, the data base Ecoinvent 2.0 (Ecoinvent Centre, 2007) of the Swiss Centre for Life Cycle Inventories was also used. Thus, a comprehensive emission and resource consumption

inventory was built. A table with LCI data for each alternative PG waste management method is shown in section 3 (LCIs of Alternative Management Methods), while the detailed LCI calculations are presented in Appendix A.

For the needs of "Hellenic Fertilizers" industry in Nea Karvali, Kavala, Greece, phosphate rock is imported from North Africa. Therefore, the chemical composition of PG waste, originating from raw materials from North Africa, was considered in this study (Table 1, Renteria-Villalobos et al., 2010). Radionuclide concentrations measured in PG from the Nea Karvali, Kavala, plant were: ^{238}U = 35 ± 18 and ^{226}Ra = 515 ± 150 Bq/kg (Potiriadis et al., 2011). Concentrations of ^{40}K < 20 and ^{232}Th < 15 Bq/kg were too low and were not considered in the analysis, whereas measurements of ^{210}Pb were not conducted (Potiriadis et al., 2011).

According to EU legislation (EU, 2014), waste for disposal, recycling or reuse may be released from regulatory control, provided that the activity concentration of naturally occurring radionuclides in solid materials does not exceed 1 kBq/kg for the ^{238}U series, 1 kBq/kg for the ^{232}Th series and 10 kBq/kg for the ^{4}K series. Activity concentrations measured in the Nea Karvali site are below the EU standards. Therefore, no regulatory control must be exercised for PG waste in this site.

2.4. Life cycle impact assessment

The software used for conducting the LCA according to the ILCD handbook guidelines in this paper was SimaPro 7.1, developed by Pre Consultants (Goedkoop et al., 2003). It can use different data bases and different assessment methods. According to Laurent et al. (2014), this was the most highly applied software for solid waste management in a total of 222 studies. It was also used by other PG LCA studies (e.g., Mohammed et al., 2018; Kulczycka et al., 2016).

The assessment method used was Ecoindicator 99, which applies a non-metric unit, called "Point" (Pt), to evaluate the impact of a process or a product. The Pt is the annual environmental load of an average European (Baayen, 2000; Acero et al., 2014). It was used in this study, because it is a highly applied impact assessment method (Laurent et al., 2014), incorporating many European data (Goedkoop and Spiensma, 2000). The method considers three damage categories, namely human health, ecosystem quality and resources, which were used for normalization and weighting. Ecoindicator 99 computes the following impact categories (Goedkoop et al., 2008): (1) Carcinogens, respiratory organics,
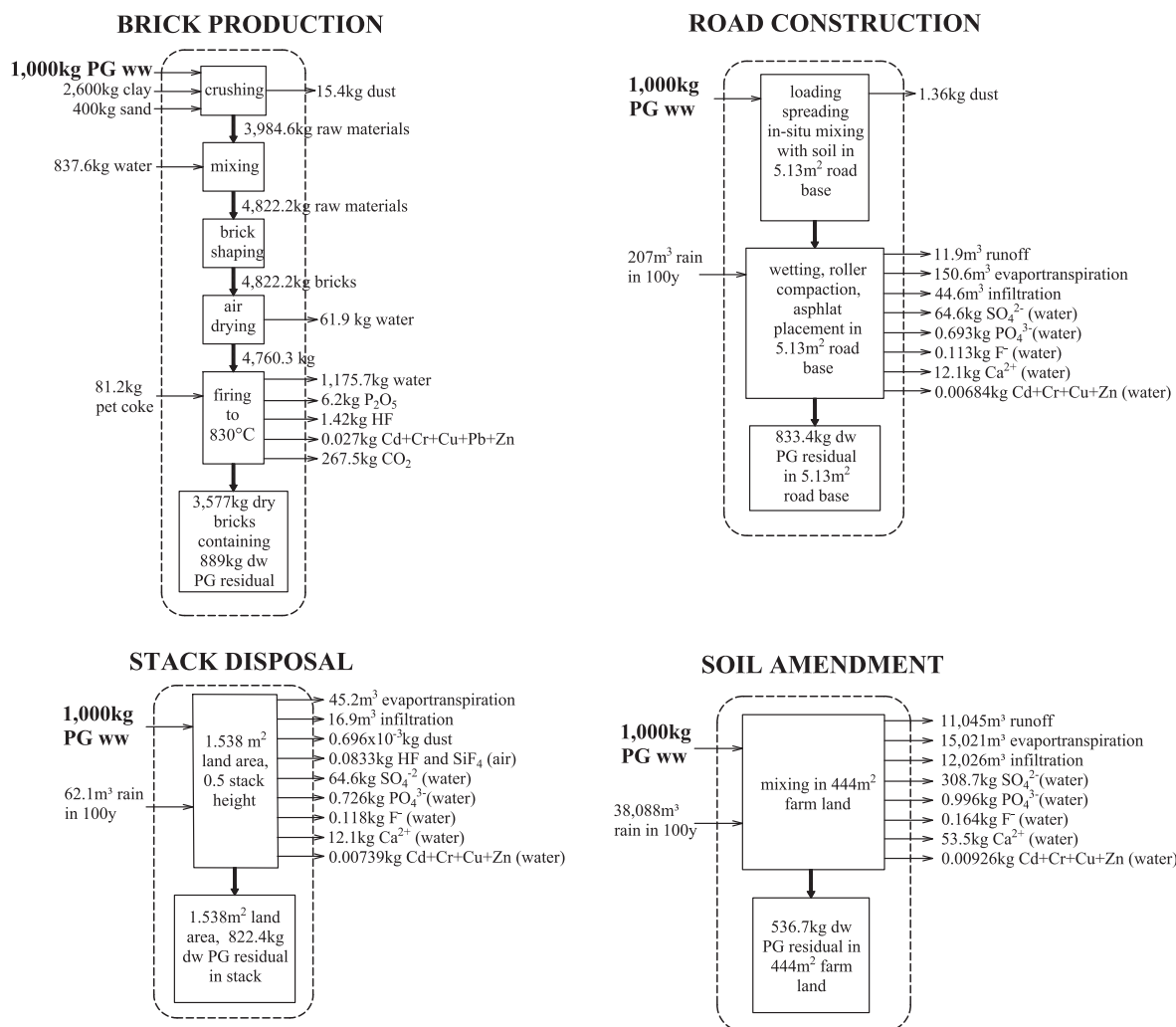
Fig. 1. Material flows and system boundaries.

respiratory inorganics, climate change, radiation and ozone layer for human health. (2) Ecotoxicity, acidification/eutrophication and land use for ecosystem quality. (3) Depletion of raw materials and energy measured by minerals and fossil fuels for resources. Damage categories were normalized on a European level (damage caused in one year by one European). Results can be computed from three different perspectives, namely the egalitarian, the hierarchist and the individualist perspective. Weighting from the egalitarian perspective (main perspective used) was 40% for human health, 40% for ecosystem quality and 20% for resources (Frischknecht et al., 2007).

Because of certain limitations of Ecoindicator 99 in the impact category acidification/eutrophication, impact assessment was also conducted with the CML 2 Baseline 2000 method (Frischknecht et al., 2007) and the results were compared.

2.5. Interpretation

The LCA results were interpreted based on the goal and scope of the study, according to the ILCD Handbook guidelines (EC, 2010). Sensitivity analysis was conducted with respect to some parameters, which were uncertain. These included the dose of PG when used as a soil amendment, the height of stack and the PG transport distance. In addition to Ecoindicator 99, the four PG waste management methods were compared using several other LCA methods, including CML 2 Baseline 2000.

3. LCIs of alternative management methods

3.1. Brick production

The first alternative management method concerns the use of 1 tonne PG to replace a portion of clay and sand in conventional brick production. The bricks would, then, be used for an external house wall construction in a populated area. The standard formulation of conventional bricks is 70% clay and 30% sand by weight. Ajam et al. (2009) studied PG valorization in fired bricks and proposed an alternative formulation for industrial brick production, with 65% clay, 25% PG and 10% sand, considering physical, chemical, mechanical and environmental tests. This formulation was adopted in this study.

Besides the economic benefit, PG addition reduces the required firing temperature, the plasticity and the drying shrinkage of the bricks, while brick shaping becomes easier and paste porosity is increased. Furthermore, PG helps curing and gives a skeleton to the raw and final paste. Valorization of 1 tonne PG results in 4 tonnes brick production. Raw materials savings account for 0.2 tonnes of clay and 0.8 tonnes of sand, but an additional 0.204 tonnes of water

is required, compared to the conventional brick production. Firing temperature is reduced from 1000 to 830 °C, resulting in fuel savings equal to 17,05 kg pet-coke/tonne of PG added. Air emissions (e.g., fluorides, P_2O_5 and metals) during brick firing and radon emission from the bricks over the life cycle of the constructed wall were estimated. Liquid emissions from the constructed wall were considered zero, because the wall will be covered with plaster and does not encounter water. Table 2 shows the LCI data for brick production, while respective calculation details are presented in Appendix A.

3.2. Use of PG as soil amendment

The second alternative management method concerns the use of PG as soil amendment to correct the calcium levels in agricultural land and improve the physical-chemical characteristics of the soil. Canovas et al. (2018) reviewed the studies of PG application in agriculture for different purposes, such as improving soil structure and crop yield, reducing soil erosion, treating acidic or metal-rich soils and increase Ca, P and S levels. Characterization of PG impurities and their mobility and bioavailability in soil is required for safe long-term application. The treatment involves PG application at a rate of 22.5 tonnes/ha every two years and mixing in the 0−40 cm soil horizon (Abril et al., 2009). It is considered that PG application replaces equal amounts of conventional gypsum and saves 6.2 kg of P_2O_5, which would have been added in agricultural land with commercial fertilizers. Emission of radioactive dust in the atmosphere was estimated, during PG application to the soil. Liquid emissions were, also, estimated as follows: (1) The liquid to solid ratio (L/S) for the top 0−40 cm soil horizon, based on infiltration of rain and irrigation water, was computed, using HELP model. (2) Leaching of fluoride, copper, cadmium, zinc and chromium was estimated, based on the work of Rutherford et al. (1995). (3) Radium leaching was estimated, based on the work of Haridasan et al. (2002). (4) Leaching of sulfate anions was estimated, based on the work of Kijjanapanich et al. (2013). Table 3 shows the LCI data for PG use as soil amendment, while respective calculation details are presented in Appendix A.

3.3. Use of PG in road construction

The third alternative management method concerns the use of PG to replace clay in secondary road construction (Fig. 2). According to IAEA (2013), use of PG in road base application could consume up to 25,000 tonnes/lane km, thus satisfying the high use requirement for a waste produced in enormous amounts. PG waste is discharged at the construction site and spread evenly by a bulldozer to the required thickness. A pulvimixer is then used to thoroughly mix the PG with the subgrade material. Then, the cross-section profile of the road is shaped and appropriate slope is provided, following the design specifications. After the necessary compaction, the finished base surface is sealed with asphalt (Chang et al., 1989). The conventional road construction would require equal amount of clay to be mixed with the subgrade material and, thus, stabilize on-site soil. Therefore, PG waste realizes savings of 1 tonne of clay. Emission of particulates and radioactive dust due to PG spreading and road shaping was estimated. No significant change in radon emission was found, compared to the use of conventional materials (Chang et al., 1989). Liquid emissions were, also, estimated, based on the computed L/S = 15 ratio. Leaching of fluoride, heavy metals, ^{226}Ra, sulfate and phosphate was estimated, based on the work of Rutherford et al. (1995). Table 4 shows the LCI data for PG use in road construction, while respective calculation details are presented in Appendix A.

3.4. Disposal of PG in a stack

The fourth alternative concerns the most common management method, which is the disposal of PG in a stack. The stack height chosen was 0.5 m, occupying a land area equal to 1.538 m^2. SimaPro 7.1, the software used for conducting LCA in this work, considers the occupied land area multiplied by the occupation time by PG. Thus, for a 100 year occupation time by the stack, the software considers (1.538 m^2)(100 a) = 153.8 m^{2}a. Estimated air emissions from the stack included hydrogen fluoride (HF), silicon tetrafluoride (SiF$_4$), radioactive dust and radon (^{222}Rn). Liquid emissions were based on the L/S ratio for the stack geometry (in our case L/S = 17) and included fluoride, trace metals, sulfate and phosphate anions and radon. Table 5 shows the LCI data for PG disposal in a stack, while respective calculation details are presented in Appendix A.

4. Results and discussion

4.1. Life cycle analysis of PG alternative management methods

The impact assessment method used by the LCA software was the Ecoindicator 99. The results of the analysis are presented in the form of bar diagrams, in which the positive sign indicates

Table 2
LCI data for 1 tonne PG valorization in brick production.

Activity	Inventory	Name in Ecoinvent
Materials savings		
Clay	−0.2 (tonne)	Clay, at mine/CH U
Sand	−0.8 (tonne)	Sand, at mine/CH U
Pet-coke	−17.05 (kg)	Petroleum coke, at refinery/RER U
Additional materials		
Water	0.204 (tonne)	Water, well, in ground
Transportation		
Lorry	10 (tkm)	Transport, lorry 3.5−16t, fleet average/RER U
3.3−16 tonne		
Air emissions		
HF	1.417 (kg)	Hydrogen fluoride (high pop.)
P_2O_5	6.2 (kg)	Phosphorus pentoxide (high pop.)
Cd	2.92×10^{-3} (kg)	Cadmium (high pop.)
Cr	3.6×10^{-3} (kg)	Chromium (high pop.)
Cu	10.8×10^{-3} (kg)	Copper (high pop.)
Zn	6.26×10^{-3} (kg)	Zinc (high pop.)
Pb	2.13×10^{-3} (kg)	Lead (high pop.)
^{222}Rn	371 (Bq)	Radon-222 (indoor)
^{238}U	143 (Bq)	Uranium-238 (high pop.)
^{226}Ra	2106 (Bq)	Radium-226 (high pop.)

Table 3
LCI data for 1 tonne PG use as soil amendment.

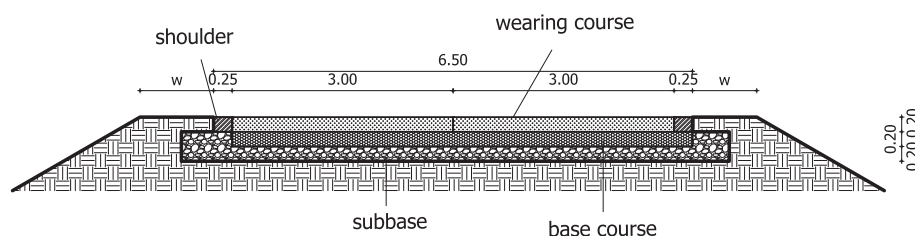| Activity	Inventory	Name in Ecoinvent
Materials savings		
Gypsum	−1 (tonne)	Gypsum, mineral, at mine/CH U
P_2O_5	−6.2 (kg)	Phosphoric acid, fertilizer grade, 70% in water, at plant/GLO U
Radioactivity		
^{226}Ra	0.525 (Bq)	Radium-226 (low pop.)
Liquid emissions		
F^-	0.164 (kg)	Fluoride (groundwater)
Cr	8.62×10^{-4} (kg)	Chromium (groundwater)
Zn	5.71×10^{-3} (kg)	Zinc (groundwater)
Cd	1.89×10^{-3} (kg)	Cadmium (groundwater)
Cu	7.94×10^{-4} (kg)	Copper (groundwater)
^{226}Ra	41.7 (kBq)	Radium-226 (groundwater)

Fig. 2. Typical e2 secondary road cross-section, according to Road Design Studies Guidelines (OMOE, 2001) of the Greek Ministry of Transport, Infrastructure and Networks.

Table 4
LCI data for 1 tn PG use in road construction.

Activity	Inventory	Name in Ecoinvent
Materials savings		
Clay	−1 (tn)	Clay, at mine/CH U
Transportation		
Lorry 3.3−16tn	10 (tkm)	Transport, lorry 3.5−16t, fleet average/RER U
Radioactivity		
^{238}U	24.75 (Bq)	Uranium-238 (high pop.)
^{226}Ra	364.1 (Bq)	Radium-226 (high pop.)
Liquid emissions		
F^-	112.86 (g)	Fluoride (groundwater)
Cr	8.62×10^{-4} (kg)	Chromium (groundwater)
Zn	3.56×10^{-3} (kg)	Zinc (groundwater)
Cd	1.62×10^{-3} (kg)	Cadmium (groundwater)
Cu	7.94×10^{-4} (kg)	Copper (groundwater)
PO_4^{3-}	0.693 (kg)	Phosphate (groundwater)
^{226}Ra	18.54 (kBq)	Radium-226 (groundwater)
SO_4^{2-}	64.59 (kg)	Sulfate (groundwater)
Ca^{2+}	12.08 (kg)	Calcium, ion (groundwater)

environmental burden and the negative sign indicates environmental benefit, with respect to energy and materials involved in the study.

4.2. Brick production

Fig. 3 compares the scores for each impact category for the brick production scenario. The second, third, fourth and fifth bars represent impact due to material transport, clay production, sand production and pet-coke production, respectively. The first bar represents all the other emissions and the materials of the brick production scenario. Thus, on the environmental burden side, air emissions/resources (first bar) cause the largest impact, accounted for by 15.815 Pt, which was itemized to ecotoxicity (8.164 Pt) and carcinogens (7.651 Pt). Material transport (second bar) contributes to environmental burden by 0.251 Pt, with contributors fossil fuel consumption (0.133 Pt), respiratory inorganics (0.072), acidification/eutrophication (0.016), climate change (0.013), ecotoxicity

(0.007), land use (0.007), minerals (0.002) and carcinogens (0.001 Pt). On the environmental benefit side, savings of clay, sand and pet-coke contribute by −0.064, −0.137 and −3.059 Pt, respectively. The two most important impact categories affecting clay and sand savings are fossil fuel savings (−0.023, −0.071 Pt, respectively) and reduction in emission of respiratory inorganics (−0.022, −0.056, respectively). Pet-coke savings are mostly influenced by fossil fuel savings (−2.716) and reduction in respiratory inorganics (−0.186 Pt). The final score of this analysis was 12.807 Pt, which indicates that PG valorization in brick production to replace clay and sand has a higher environmental footprint than conventional brick production and, therefore, is not recommended.

4.3. Use of PG as soil amendment

Fig. 4 compares impact categories for the soil amendment scenario. The second and third bars represent gypsum and phosphoric acid production, respectively, whereas the first bar represents all

Table 5
LCI data for 1 tonne PG disposal in stack.

Activity	Inventory	Name in Ecoinvent
Land use		
Land occupation	153.8 (m^{2}a)	Occupation, pasture and meadow (land)
Air emissions		
HF	38.4 (g)	Hydrogen fluoride (low pop.)
SiF$_4$	49.9 (g)	Silicon tetrafluoride (low pop.)
PM$_{30\mu m}$	0.696 (g)	Particulates, >10 μm (low pop.)
Radioactivity		
^{238}U	0.024 (Bq)	Uranium-238 (low pop.)
^{226}Ra	0.358 (Bq)	Radium-226 (low pop.)
^{222}Rn	392060 (kBq)	Radon-222 (low pop.)
Liquid emissions		
F$^-$	118.12 (g)	Fluoride (groundwater)
Cr	8.62×10^{-4} (kg)	Chromium (groundwater)
Zn	3.84×10^{-3} (kg)	Zinc (groundwater)
Cd	1.89×10^{-3} (kg)	Cadmium (groundwater)
Cu	7.94×10^{-4} (kg)	Copper (groundwater)
PO$_4^{3-}$	0.726 (kg)	Phosphate (groundwater)
^{226}Ra	9.27 (kBq)	Radium-226 (groundwater)
SO$_4^{2-}$	64.59 (kg)	Sulfate (groundwater)
Ca^{2+}	12.08 (kg)	Calcium, ion (groundwater)

other impacts, including radioactivity and liquid emissions. Negative impacts due to emissions account for 0.006 Pt, all attributed to ecotoxicity. Positive impact due to conventional gypsum savings account for −4.277 Pt, with dominant impact categories reduction in respiratory inorganics (−4.167 Pt), fossil fuel reduction (−0.074 Pt) and acidification/eutrophication (−0.033 Pt). Positive impact due to savings from phosphoric acid production account for −1.063 Pt, with dominant impact categories respiratory inorganics (−0.372 Pt), carcinogens (−0.211 Pt), fossil fuels (−0.195 Pt) and land use (−0.193 Pt). The final score of this analysis was −5.33 Pt, which indicates that the use of PG as soil amendment to correct calcium levels in agricultural land has a lower environmental footprint than conventional gypsum use. This happens because the positive impacts due to materials savings are larger than the negative PG impacts.

4.4. Use of PG in road construction

Fig. 5 compares impact categories for the road construction scenario. The second and third bars represent material transport

and clay production, respectively, whereas the first bar represents all other emissions, materials and energy of the scenario. Negative impacts due to materials transport and other emissions account for 0.2507 and 0.0058 Pt, respectively. The top three impact categories of materials transport are fossil fuels (0.1328 Pt), respiratory inorganics (0.0715 Pt) and acidification/eutrophication (0.0158 Pt). The impact categories affecting all other emissions are ecotoxicity (0.0058 Pt) and radiation (0.0001 Pt). Positive impacts due to clay savings account for −0.32 Pt, with the top three impacts fossil fuels (−0.1129 Pt), respiratory inorganics (−0.1099 Pt) and land use (−0.0613 Pt). The final score was −0.0635 Pt, indicating that the use of PG to replace clay in road construction works has a lower environmental footprint than the conventional scenario, which uses clay.

4.5. Disposal of PG in a stack

Fig. 6 presents the final score of 15.484 Pt for PG disposal in stacks 0.5 m high. This scenario does not include activities, such as savings of materials and energy, which were implemented in previous scenarios. The largest impact in the 100-year LCA was predicted for land use (15.295 Pt), followed by radiation emission (0.183 Pt) and ecotoxicity (0.006 Pt). A sensitivity analysis with respect to stack height was conducted to compute how its impact on land is affected. Therefore, alternative scenarios with heights ranging from 0.1 to 5 m were considered. Fig. 7 shows that stack height has a significant effect on land impact, with smaller heights being more important. This happens because the shorter stack occupies a larger area, therefore, the land coverage and air emissions from the top surface will be larger. In addition, the liquid to solid (L/S) ratio will be higher, thus, resulting in increased cumulative leaching. Other than its serious environmental impact, stacking has the disadvantage of considering a potential resource as a waste.

4.6. Comparison of alternative methods

Fig. 8 compares the four alternative management methods for PG waste, using the Ecoindicator 99 method and the egalitarian perspective. The use of PG as a soil amendment to replace conventional gypsum in agricultural land has the lowest environmental footprint, followed by use of PG as a subbase to replace conventional gypsum in road works. The PG valorization in brick
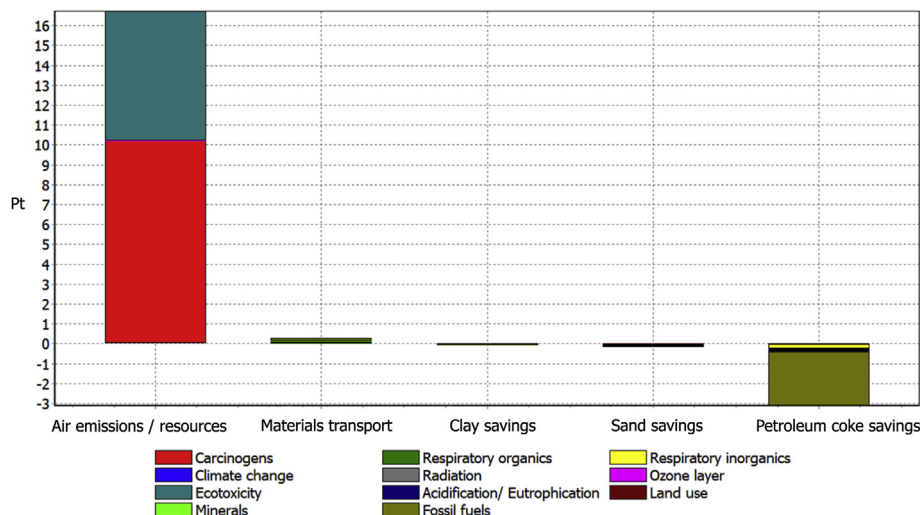

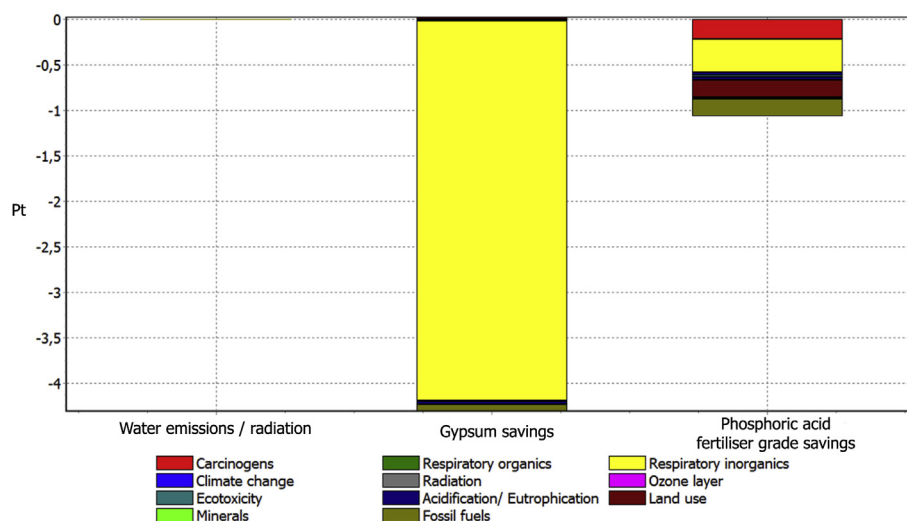**Fig. 3.** Final scores for the brick production scenario.

Fig. 4. Final scores for the soil amendment scenario.
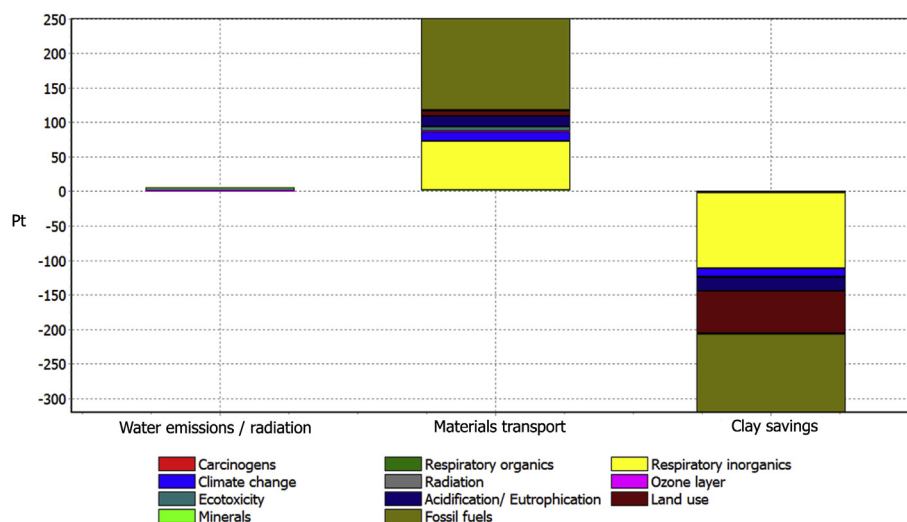

Fig. 5. Final scores for the road construction scenario.

production has the highest negative environmental impact, but this is offset by the significant positive effects of materials and energy savings. Therefore, this scenario was ranked in the third position, with PG disposal in 0.5 m high stacks having the least preference. Ranking the four PG waste management methods, from the point of view of the three perspectives, based on their total score, is presented in Table 6.

4.7. Sensitivity analysis

4.7.1. Sensitivity analysis with respect to number of PG applications as soil amendment

The results of Fig. 4 refer to two PG applications at a rate of 22.5 tonnes/ha every two years and mixing in the 0−40 cm soil horizon. This corresponds to 500 kg PG/222 m^2 per application. In an alternative scenario, the same total amount of PG was added in 50 applications of 20 kg each, every two years. In order to maintain the same application rate (22.5 tonnes/ha or 2.25 kg/m^2), the area was reduced to 8.8 m^2. The final score of the alternative scenario was −5.33 Pt, which is equal to the score of the two-application scenario. The results of the sensitivity analysis with respect to number of PG applications are compared in Table B1 (Appendix B).

4.7.2. Effect of stack height on alternative method ranking

The first three methods were compared with stack disposal, for a stack height of 5 m. This is the optimum stack height of those investigated (Fig. 7). The individualist perspective was used, because it results in the lowest grading (i.e., lowest footprint) for stack disposal. The results (Table B2) show that stack disposal ranking is slightly improved, as it moves from the 4th to the 3rd position. This indicates that increase of stack height does not significantly affect the results of the previous comparison (Fig. 8).

4.7.3. Neglecting materials transportation

In the previous comparison of alternative management methods, an arbitrary distance of 10 km was used (section 2.2.3 Materials transportation). To assess the effect of distance, a comparison was made neglecting materials transportation. The results
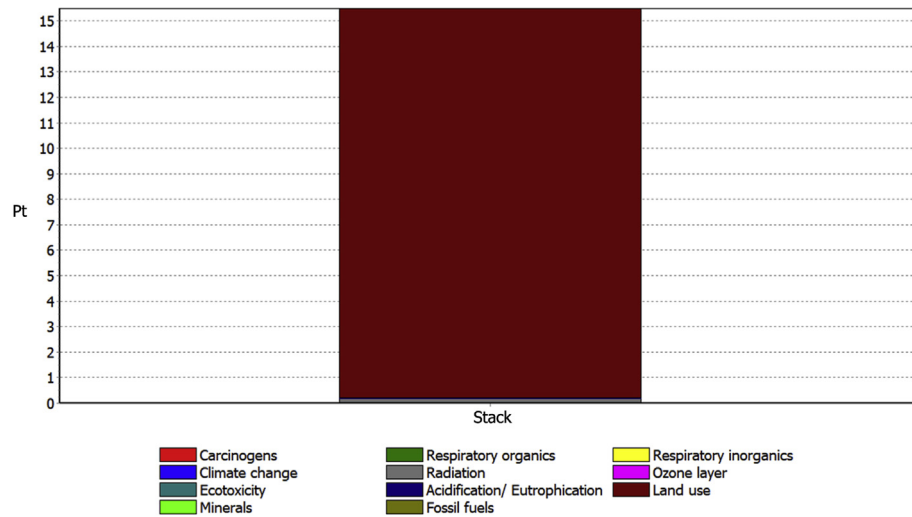
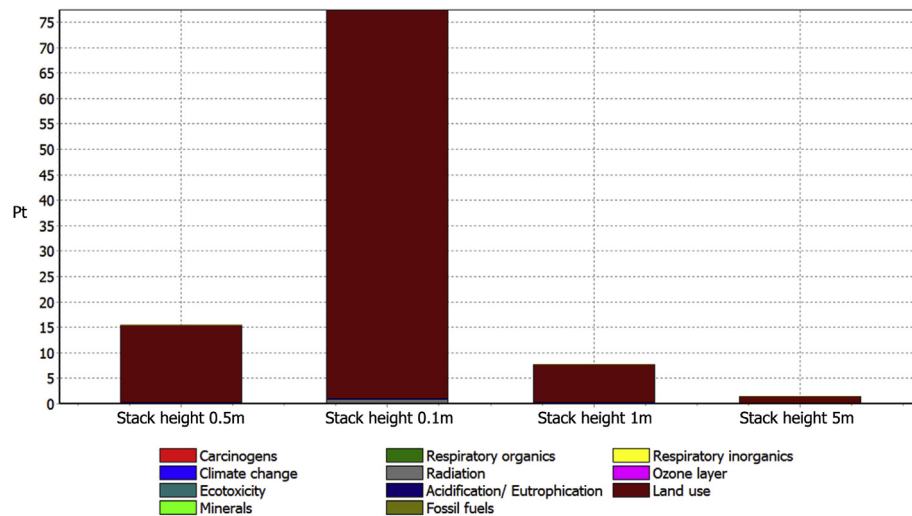**Fig. 6.** Final scores for the disposal in stack scenario.

Fig. 7. Sensitivity analysis with respect to stack height.

presented in Table B3 indicate that the distance of materials transportation within the range 0–10 km does not affect the first two ranking positions of the previous comparison (Fig. 8), but places stack disposal in the third position.

4.7.4. Effect of method of analysis

The four alternative PG waste management methods were compared using all the available methods of analysis and the results are presented in Table B4. The number of substances not considered by each method of analysis is shown in the last column of Table B4. Therefore, the most appropriate methods are the ones considering the largest number of substances, i.e., have the smallest entry in the last column of Table B4. Most of the methods used maintain the ranking of Fig. 8.

4.7.5. Comparison with respect to acidification and eutrophication

Ecoindicator 99 has limitations with respect to impact categories acidification/eutrophication. Therefore, the impact assessment was also conducted with method CML 2 Baseline 2000, which

considers substances causing these impact categories. With respect to acidification, soil amendment has the lowest footprint, followed by PG use in brick production and use in road construction, with stack disposal having the highest footprint (Table B5). The ranking with increasing footprint with respect to eutrophication is: PG use as soil amendment, use in road construction, stack disposal and brick production (Table B5).

4.8. Economic analysis

Because of lack of literature data, an economic analysis was conducted to compare the four management methods of PG waste. The results are presented in Appendix C and summarized in Table 7. Cost estimates are normally based on full scale works, therefore this comparison, which is based on information from the current Greek market, should be considered indicative. Table 7 shows that cost estimates for PG works are lower than conventional works. The % cost difference with respect to PG works are 2.9 and 6.3% for brick production and road construction, respectively, but this difference
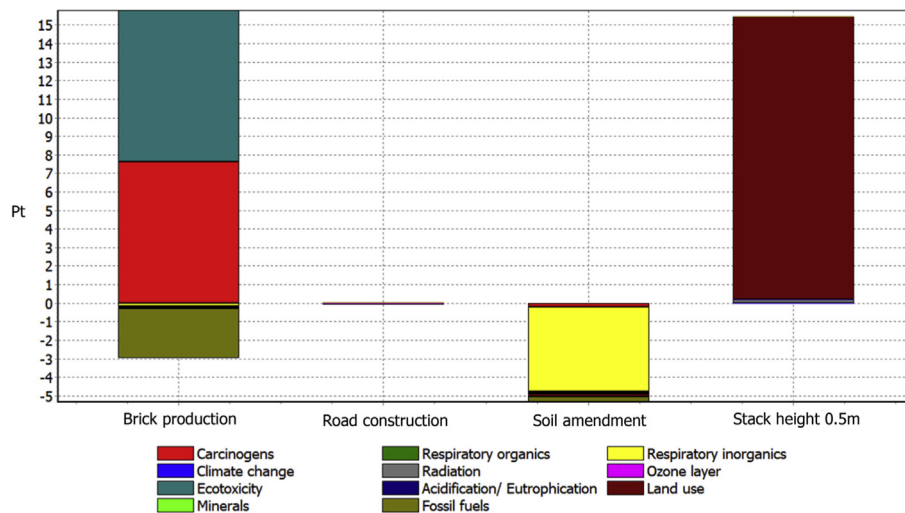
Fig. 8. Comparison of the four alternative management methods for PG waste, using Ecoindicator 99 and the egalitarian perspective.

Table 6
Ranking of the four PG waste management methods using the total score and the three perspectives.

Perspective	RANK			
	1st	2nd	3rd	4th
Egalitarian	Use as soil amendment −5.334	Use in road construction −0.064	Brick production 12.807	Stack disposal 15.484
Hierarchist	Use as soil amendment −6.925	Use in road construction −0.065	Stack disposal 12.486	Brick production 13.163
Individualist	Use as soil amendment −9.307	Use in road construction −0.074	Brick production 1.996	Stack disposal 14.314

for soil amendment is overwhelming (4450%). The ranking based on cost is soil amendment≪road construction<brick production. PG stack disposal is the least expensive, as indicated by the cost ratio, and partially explains its wide popularity compared to other management methods. However, stack disposal in most cases is uncontrolled landfilling with the highest environmental footprint.

5. Conclusions

The first objective of this work was to compute the environmental footprint of alternative PG waste management methods, using LCA. The data base Ecoinvent 2.0 with the assessment method Ecoindicator 99 was used. The results showed that PG waste valorization to replace sand and clay in brick production has a higher environmental footprint than the conventional brick production method and, therefore, it is not recommended. Use of PG waste as soil amendment to correct calcium levels in agricultural land has a lower environmental footprint than using conventional gypsum and it is recommended. Similarly, use of PG

waste as a subbase in road construction works has a lower environmental footprint than using conventional clay. The second objective was to rank the alternative PG waste management methods, using their environmental footprint. The results showed that the top ranked method is the use of PG waste as a soil amendment, with second the use of PG as a subbase in road works and third the use of PG waste in brick production. Disposal of PG waste in stacks, which is the most common management method, was ranked as the least preferred one. The ranking is based on the total scores −5.334, −0.064, 12.807 and 15.484 Pt, respectively, using Ecoindicator 99 and the egalitarian perspective. The ranking based on cost is: stack disposal<soil amendment≪road construction<brick production. Use in road construction and agriculture are the most promising valorization methods in terms of the amount of PG used, satisfying better the need for mass consumption of a waste material stacked in enormous amounts and with serious environmental impact. Limitation of this study is the lack of site-specific data from Greek PG production sites, except for radionuclide content. Therefore, future work could address this

Table 7
Economic comparison of the four management methods for 1 tonne PG.

Management method	Total cost, euro	% Difference with respect to PG method	Cost ratio to stack
Conventional brick production	245		76.6
PG brick production	238	2.9	74.4
Conventional soil amendment	273		85.3
PG soil amendment	6	4450	1.9
Conventional road construction	119		37.2
PG road construction	112	6.3	35.0
PG stack disposal	3.20		1.0

need, along with study of additional valorization methods, such as production of chemicals and cement. Using LCA is a useful approach for comparing PG waste valorization methods within the context of Circular Economy.

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Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

CRediT authorship contribution statement

Maria Tsioka: Methodology, Software, Validation, Formal analysis, Investigation, Writing - original draft, Visualization. **Evangelos A. Voudrias:** Conceptualization, Methodology, Investigation, Writing - original draft, Writing - review & editing, Visualization, Supervision.

Appendices A, B and C. Supplementary data

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