Life Cycle Assessment of Alternative Road Base Materials: the Case of Phosphogypsum

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Abstract: Phosphogypsum (PG) is the most abundant by-product generated by the phosphate fertilizer industry. Formed during the production of phosphoric acid from natural phosphate rock, PG is mostly disposed in stacks or released into coastal regions' waters. Due to the expected increase in the world PG production – currently estimated to be around 250 Mt per year – and the environmental impacts of actual waste management scenarios, there is a call for a paradigm shift, by considering PG not as a waste but as a resource. About 15% of the total PG production is nowadays used as fertilizer, retarder, road base material or building material. The load of radionuclides and heavy metals contained in PG however questions the sustainability of such valorization routes. This paper aims to compare the environmental impacts of different PG valorization scenarios through life cycle assessment. Based on Moroccan conditions for phosphoric acid production, it discusses the key parameters influencing the assessment as well as assumptions regarding the allocation of emissions and resource use over PG and phosphoric acid.

1. Introduction: Phosphogypsum: waste or resource?
Phosphogypsum (PG) is the most abundant by-product generated by the phosphate fertilizer industry. It is formed during the wet production process of phosphoric acid, after digestion of natural phosphate rock with sulphuric acid (eq. 1). On average, the production of 1 ton of phosphoric acid (as P₂O₅) generates 5 tons of dry PG [13].

\[
\text{Ca}_5\text{F(PO}_4\text{)}_3 + 5\text{H}_2\text{SO}_4 + 10\text{H}_2\text{O} \rightarrow 3\text{H}_3\text{PO}_4 + 5\text{CaSO}_4 \cdot 2\text{H}_2\text{O} + \text{HF} \quad (\text{eq. 1})
\]

PG is mainly composed of gypsum but also contains impurities such heavy metals (Pb, Cd, As...) and radionuclides (Uranium-238, Thorium-232 and their daughter elements such as Ra-226), naturally occurring in the phosphate ore [3],[7]. Because of its low radioactivity, PG has been considered as a waste and is mostly stored indefinitely in open stacks (Florida, China) or discharged into the sea close to coastal regions (Morocco, Tunisia, South Africa...). Both PG disposal routes have environmental impacts and costs [20]; [4];[10];[17].

In 2013, the International Atomic Energy Agency concluded that “[a]ll evidence suggests that the doses received as a result of the use of PG in agriculture, road construction, marine application and in
landfill are sufficiently low that no restrictions on such uses are necessary.” [11]. As a consequence, there is a call for a paradigm shift, by considering PG not as a waste but as a resource [12],[5].

Due to the increasing demand in phosphate fertilizers at global scale, PG production – currently estimated to be around 250 Mt per year [19] – is expected to increase, as well as the environmental impacts of actual waste management scenarios. About 15% of the total PG production is nowadays used as fertilizer, retarder, road base or building material [6]. The sustainability and environmental suitability of such valorization routes are however to be investigated.

The objective of this paper is to assess the environmental impacts of PG valorization as road base material throughout its life cycle. It aims at 1) comparing environmental burdens of “conventional” versus alternative road base materials, respectively granulate and PG mixtures; 2) assessing the potential displacement of environmental impacts from a life cycle stage to another; and 3) discussing the influence of allocation approaches on the assessment. It presents preliminary results of a study conducted at the Ecole des Ponts ParisTech in collaboration with the OCP group. Based on Moroccan conditions for phosphoric acid production, it discusses the key parameters influencing the assessment, and assumptions regarding the allocation of emissions and resource use among PG and phosphoric acid.

2. Goal and scope definition

2.1 Goal

The main objective of this study is to compare the potential environmental impacts of formulations used as base layer containing different proportions of PG.

2.2. Functional unit (FU)

The functional unit (FU) considered in this case study is an experimental road pavement structure of 200 m length, with a width of 7 m. The PAP is 25 years. The average annual daily traffic (AADT) is considered to be equal to 15 heavy duty vehicles (HDV)/day. The FU is defined as a “unit of pavement that can safely and efficiently carry the same traffic over the same PAP” [15].

![Experimental road pavement cross section](image)

Figure 1 – Experimental road pavement cross section, modified from [14]

The pavement structure (Figure 1) is composed of 20 cm bank-run gravel type A (GNA), 20 cm bank-run gravel type B (GNB), and 35 cm phosphogypsum mixture (PG) as base layer, the latter being under scrutiny. Table 1 presents the formulations of the PG mixture used for the base layer, as described in [14]. The baseline scenario is a base layer composed of 100% granulate. The 4 phosphogypsum mixture scenarios have different proportions of PG and waste rock, and a constant 7% dry mass binder material, as cement 42.5. All materials are produced in Morocco, in less than 100 km radius from the road pavement construction site. As a first approach, it is assumed that the total dry weight of material required for the FU is equivalent for all scenarios (1200 t/UF). This assumption should be further discussed.

| Table 1 – Formulation of the phosphogypsum mixture used for the base layer (% dry mass equivalent) |
|---------------------------------------------------------------|
| **Baseline** | **Scenario 1** | **Scenario 2** | **Scenario 3** | **Scenario 4** |
| Granulate | 100 | 0 | 0 | 28 | 0 |
| Cement | 0 | 7 | 7 | 7 | 7 |
2.3 System boundaries

The system boundaries of this study (Figure 2) include the extraction and production of road base material such as granulate, cement and PG; the material transportation from the production site to the road pavement construction site; the on-site laying of the base layer; and its use over the PAP. The PG formulation is assumed to have no influence on the lifespan and maintenance of the road structure. Therefore, extraction and manufacturing of materials for upper layers GNA and GNB are not accounted for, as well as road maintenance processes and the end-of-life (see below). Waste rock generation during phosphate rock washing (see below) is also excluded from the system.

Figure 2 – Flowchart of major processes in the life cycle of a road pavement structure

In order to match Moroccan conditions, manufacturing of virgin materials (granulate and cement 42.5) are adapted from French processes, described respectively in [18] and [2]. Since no electricity mix exists for Morocco, Greek electricity mix from ecoinvent, “electricity, medium voltage | market for electricity, medium voltage – GR” is chosen for its proximity with the Moroccan mix (23% coal, 61% oil, 5% gas, 7% biomass, 2% renewable, 2% import, no nuclear energy, ref). It replaces the French electricity mix used in the processes where nuclear energy predominates.

PG production is adapted from the ecoinvent process for phosphoric acid production in Morocco, “phosphoric acid production, dihydrate process | phosphoric acid, fertiliser grade, without water, in 70% solution state, MA”, described in [1]. After extraction, phosphate rock is beneficiated through screening, washing and flotation. The beneficiation process does not involve adjunction of chemicals. Reaction between beneficiated phosphate rock and sulphuric acid produces phosphoric acid and PG. Substances reported in the original ecoinvent process and emitted during disposal of PG into the ocean are allocated among the use and end-of-life stages, where 100% of heavy metals and radionuclides are leached into groundwater. Emissions, raw material extraction and environmental impacts generated by
this multifunctional system are partitioned between the two co-products based on the “cut-off” approach, meaning that environmental burdens are attributed to phosphoric acid alone [16], as a first step. The influence of allocation methods is discussed below.

After attack of phosphate rock with sulphuric acid, the generated PG slurry (30% water content) is conveyed, stored in open tailings ponds and air-dried. Acidic pond water is partially recovered (about 15% of the total volume) and reinjected in the phosphoric acid production process, before adjunction of sulphuric acid [8]. Although this is common practice for Morroc an conditions, the recycling of weak phosphoric acid and the benefits associated with avoided sulphuric acid production are not included in the system. Air-dried PG (15% water content) is recovered for further use. PG recovery thus avoids waste management where PG slurry is diluted in the Atlantic ocean (see above).

Distances and vehicle types for road base materials transportation are taken from [14].

The on-site laying process is assumed to be the same for all formulations, i.e. 30 minutes mixing, except for the baseline scenario where no mixing is required [14].

During the use phase, only direct leaching to groundwater of water soluble substances contained in PG is taken into account. While all substances particularly trace elements and radionuclides contained in PG are directly emitted into sea water when disposed into the ocean (avoided waste management scenario), addition of cement to the PG mixture has an inerting effect [9]. Contaminants are trapped in a solidified matrix and only a part is leached with precipitation water. The crucial point is to determine the flows of pollutant potentially transferred into groundwater. Since no data are currently satisfactory to estimate transfer coefficients, assumptions about the inerting effect of cement are made. It is assumed that 100% of the impurities are emitted into groundwater (no inerting effect: worst case scenario), based on the transfer coefficients into sea water estimated in ecoinvent [1]. This assumption will not be discussed in this paper.

Exposure to radioactive substances resulting from airborne particle emissions are usually carefully considered [1]. In our case, phosphogypsum mixtures are used for road subgrade layer construction, the emission of airborne particles from abrasion during the use phase is considered inexistant.

For the end-of-life, it is assumed that the road pavement structure is rehabilitated after the end of the PAP, i.e. after 25 years. Rehabilitation of the road pavement implies scraping the surface course, excavating the base layers GNA, GNB and PG which are mixed together, compacting the road foundation, applying the mixture as base layer and renewing upper layers. As a consequence, the rehabilitated road pavement structure is raised 10-15 cm. The effective lifespan of the road is not known. Since it is assumed that the PG mixture does not influence the rehabilitation frequency and lifespan of the pavement structure, the only discussion is about the long term fate of contaminants and how to allocate pollutants release between the use and the end-of-life. Too many uncertainties are attached with the fate of such structure, the end-of-life is then excluded from the system.

3. Results and discussion

The OpenLCA software version 1.7.4 is used for the modelling of the scenarios and calculation of environmental impacts scores. The chosen impact assessment method is RECIPE (H), at midpoint level.

3.1 Impact assessment

The LCIA scores for the road pavement scenarios are calculated by applying the RECIPE midpoint (H) method. Environmental impact scores are calculated considering that first burdens related to phosphate rock processing is totally attributed to phosphoric production (cut-off approach: Schrivers et al (2016)), second no substitution related to the avoided waste management of PG (disposal into the ocean) is taken into account and third 100% of the heavy metals and radionucleids contained in PG are released into groundwater during the use stage of the road (worst case scenario).
The results presented in Figure 3 show that phosphogypsum mixtures scenarios are improving the environmental performance of the road pavement for impact categories such as ozone depletion, metal depletion and fossil depletion, compared with the baseline scenario (high impact related to transportation).

Figure 3 – Relative LCIA results for the road pavement scenarios, calculated with RECIPE midpoint (H), cut-off approach, 100% transfer to groundwater, without substitution

(a)

(b)
Figure 4 - Relative impacts for a) road pavement scenario 3 and b) road pavement scenario 4, life cycle stages, cut-off approach, 100% transfer to groundwater, without substitution, ReCiPe Midpoint (H).

Scenario 4 presents the highest impact scores for freshwater ecotoxicity, freshwater eutrophication, human toxicity, ionising radiation and marine ecotoxicity because of heavy metals and radionuclides transferred to groundwater (Figure 4). Scenario 3 shows the higher scores for impact categories such as terrestrial ecotoxicity, terrestrial acidification, photochemical oxidant formation and climate change. Effects on climate change are related to the production of cement as binder in the phosphogypsum mixtures, as production of cement induces decarbonation.

3.2 Sensitivity analysis: influence of allocation methods

PG is generated during the production of phosphoric acid. Previously, emissions, extractions and related impacts have been allocated between the two co-products based on the cut-off approach (Schrivers et al 2016). According to this approach, environmental impacts of phosphate rock extraction and processing are attributed to phosphoric acid alone. The choice of allocation methods however influences significantly the impact scores. Figure 5 compares the relative impacts of scenario 1 to 4 calculated based on the cut-off approach and two other partitioning methods (based on mass and market price of substituted material). With mass allocation, since 1 kg P2O5 in acid and 4.83 kg PG are produced during the processing of phosphate rock, 82.8% of the environmental burdens are attributed to PG. With economic partitioning, it is assumed that 1 t of PG substitutes 1 t of granulate (30 €/t); and 1 t P2O5 in acid worths 350 €/t. Therefore, about 8% of total impacts are attributed to PG.

Considering that 100% of the environmental burdens associated with the extraction and processing of phosphate rock are attributed to PG has a significant influence on the calculated impact scores. According to the mass allocation approach, the PG production stage generates the greater impact for almost all categories.
4. Conclusion

Preliminary results show that, with 100% transfer of heavy metals and radionuclides into groundwater, direct emissions during the use stage of the road pavement structure are responsible for major impacts on human health and ecosystem quality. This assumption however ignores the inerting effect of cement. It is then necessary to specify transfer coefficients from phosphogypsum mixture to groundwater and to take into account the temporality of such transfer. Allocating the effects of heavy metals and radionuclides leaching between the PAP and the long term (end-of-life) has to be discussed as well.

The impact of cement consumption in the PG mixtures predominates for climate change, particulate matter formation and photochemical oxidant formation. Decreasing the proportion of cement in the PG mixture should be investigated parallel to its inerting effect.

Preliminary results are based on the hypothesis of no abrasion during the use phase, because PG is used in the base layer mixture and not in upper layers. Scenarios taking into account abrasion processes and particulate dissemination in different environments (with low to high population density, for example rural and urban areas) are to be discussed for longer PAP.

The impact assessment shows the influence of several parameters. The choice of the electricity mix is important, especially regarding ionizing radiations. There is no ecoinvent electricity mix for Morocco and the available African electricity mix is not suitable. The allocation method has a great influence on the results. By allocating emissions and extractions based on mass, PG production is the life cycle stage with the higher impact score for most categories.
Finally, substitutions related to the replacement of virgin material (granulate) by phosphogypsum mixtures and the avoided waste management scenario (PG disposal into the ocean) are not taken into account. This should be further investigated.

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