*Int. J. Environ. Res., 9(2):561-566, Spring 2015*
*ISSN: 1735-6865*

# Total Material Requirement assessment of Phosphorus sources from Phosphate ore and urban sinks: Sewage Sludge and MSW incineration fly ash

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Received 20 June 2014; Revised 28 Aug. 2014; Accepted 10 Sep. 2014

**ABSTRACT:** Diversification of phosphorus sources can bring substantial synergy effects within all the sustainable development domains: environmental, economic and social, and such an opportunity should not be overlooked. Urban sinks accumulate phosphorus and other elements and may serve as sources of secondary raw materials. This paper evaluates phosphorus sources based on their total material requirement (TMR). Resource requirements and emissions of the conventional phosphorus production from mining through the acid route processing have been quantified and have been used as a yardstick against which to measure the performance of two recycling options: spreading of sewage sludge and phosphorus recovery from municipal solid waste incineration fly ash (MSWA). The sludge spreading had the lowest TMR. Phosphorus extracted from the MSWA had four-fold higher TMR than the conventional production. However, method modifications were suggested and are currently being tested, which reduce the methods TMR well below the TMR of the phosphorus production from the ore. The entire impact of the Swedish mineral fertilizer demand can be avoided by recycling urban sinks of phosphorus. Sweden can become self-sufficient in mineral fertilizer because the phosphorus quantities imported in food and later found in the urban sinks such as food waste, sewage sludge and MSW incineration residues cover the entire necessary quantity.

**Key words:** Phosphorus,Urban sinks,Sewage Sludge,Fertilizer

## INTRODUCTION

Modern agriculture depends on the input of fertilizers for production of food, feed, fiber and biofuels. Nitrogen (N) and Phosphorus (P) are both essential to life and non-substitutable, however P is also a non-renewable resource derived from rock deposits. Due to the risk of aquatic pollution and eutrophication, phosphorus management has long been a concern. Recently, however, the focus has changed to the strategic importance of phosphorus in agricultural production, and food security in particular. Today, about 85% of the worldwide demand for rock phosphate relates to agricultural production (IFA, 2011). There have been warnings about probable future scarcity, as the global population continues to grow, and there is a reduction in under-nutrition, diets change, and the production of biofuels increases (Cordell *et al.*, 2009). The estimated remaining lifetime for the phosphate rock reserves varies between 400 and 1000 years, and the IFA (International Fertilizer Industry Association) recently refuted the “peak phosphate” theory (IFA, 2011). At the moment, the consensus is that there is no looming physical scarcity, whilst economic scarcity (i.e. due to the high price) is a reality (Scholz and Wellmer, 2013). However, it is a fact that phosphate rock is a non-renewable resource and its overexploitation should be avoided. Rock phosphorus demand can be mitigated by phosphorus recycling, i.e. by the secondary phosphorus sources. Such resources include agricultural waste such as manure and crop residues and human wastes such as sewage sludge and solid waste (Kalmykova *et al.*, 2012; Liu *et al.*, 2008). Crop residues are already recycled as animal fodder or left in fields. Manure is the largest secondary resource and as a rule also completely recycled. In contrast, sewage sludge and solid waste recycling is currently low. For example in the EU, less than 40% of the sewage sludge is returned to agriculture.

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Solid waste is an underestimated source of phosphorus. Vast and ever increasing amounts of municipal solid waste (MSW) are produced worldwide. Several recent studies showed MSW to contain P quantities comparable to those found in sludge (Kalmykova *et al*., 2012; Ott and Rechberger, 2012). Food and food processing wastes is a major source of P in solid waste (4.0 g P/kg TS, total solid). Other sources of P in solid waste are wood, paper and textile (0.2 – 0.3 g P/kg TS). Only about 30% of the organic waste are currently collected separately and treated biologically in the EU (European Commission, 2008). Due to the challenge of the implementation of the comprehensive separate recycling of organic waste it is reasonable to expect that solid waste will contain considerable amount of P even in the foreseeable future. On the other hand, incineration of waste is increasingly common waste management option worldwide and the P, along with other elements, is expected to be stored in the incineration residues. A method for P recovery from the MSW incineration ash (MSWA) has recently been developed (Kalmykova and Fedje, 2013). In this paper we investigate how the newly developed method for recycling of P from the MSWA and the direct recycling of sewage sludge compare to conventional phosphorus production with respect to their environmental pressure. The life cycle assessment of the three options has been reported by Kalmykova *et al*. 2014a. The conventional production had slightly lower impact than the sewage sludge recycling, however, it has been stated that the actual impact of conventional production might be underestimated. In particular, the phosphate rock depletion as well as the growing impacts due to the lower grade of phosphates are not yet accounted for in the characterization methods. Therefore, the methods are compared in this paper from a different perspective, applying the total material requirement (TMR) indicator. In addition, the potential of urban phosphorus sinks for reduction of rock-derived phosphorus demand has been assessed.

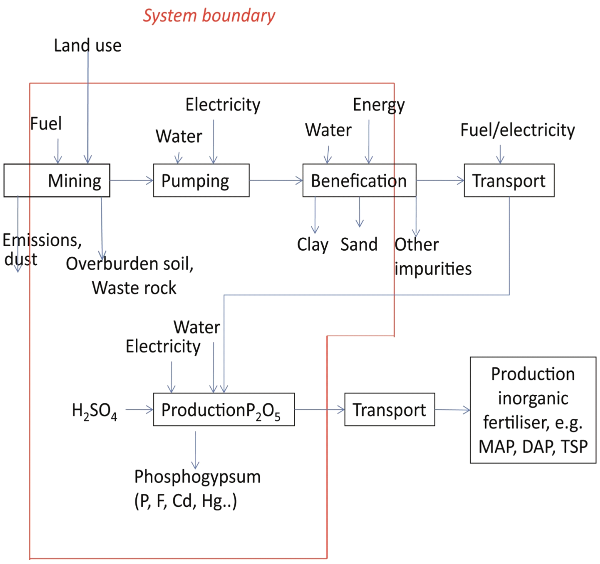

**Fig. 1. System boundary for the production from phosphate ore by the wet-chemical route**

## MATERIALS & METHODS

The Total Material Requirement (TMR) was used as the indicator of the environmental pressure. The TMR was calculated based on the used and unused (i.e. byproducts and waste) resources as well as the emissions of the P production process. For all the options, the system boundary was limited to the step where P is produced, the applied system boundaries can be found in Figs. 1-3. The functional unit was set to 1 tonne 100% phosphate ($P_2O_5$) produced or recycled. The potential of the recycling options to mitigate the demand for the rock-derived P was calculated using the available annual amounts of P in the secondary sources and the current recycling efficiencies. The possible different plant availability of the products from different processes was not considered. Sweden was chosen as a study area. Sweden has a national goal for phosphorus recycling from sewage sludge and for separate collection and treatment of the organic waste. However, despite two decades of separate organic waste collection, the collection has stagnated on the low level. In particular, only about 20% is collected from the households and even less from the wholesale, retail and the restaurant branches (Kalmykova, Harder 2012). On the other hand, in Sweden, more than 50% of the MSW is incinerated with energy recovery, therefore the P content of the organic waste become concentrated in the ash (Avfall Sverige, 2012). The two recycling options were assumed to be carried out in Gothenburg metropolitan area, Sweden. It is the second largest metropolitan area (0.95 million inhabitants) with one major sewage treatment plant and an incineration plant, whose incineration waste is used in the P recovery from MSWA method development.For the conventional production, the material requirement for the production of 1 tonne 100% phosphate ($P_2O_5$) by the acid route were compiled through a literature review.; see the calculations in Supplementary Table 1. In this paper we denote as phosphate rock the commercial grade rock, i.e. the raw material used by the chemical industry for phosphoric acid (phosphate fertilizers precursor) production after its concentration (beneficiation) from the crude phosphate ore. Phosphate ore is used to denote the crude ore, sometimes referred to as “the matrix”, which also contains minerals other than phosphate. The system boundary for sewage sludge spreading option is shown in Fig. 2. Processes and emissions of sewage treatment and sludge digestion to produce biogas were excluded. Only the additional process of sludge hyginisation, which is a requirement for its use as fertiliser, was considered. Similar to the other investigated options, energy for spreading of sludge was not accounted. According to estimates by (Tideström *et al*., 2009), the sludge is transported on average 15 km from the treatment plant to the nearest arable land. For the recycling from the MSWA option the material requirement is based on the laboratory procedure, which was scaled-up linearly (Fig. 3). In such a way, the material requirement is probably overestimated.

## RESULTS & DISCUSSION

About 90% of the phosphate rock worldwide is processed by the so called wet-chemical or acid route to produce phosphate fertilizers (Fig. 1 and Table 1). The majority of the phosphate mining is done by surface mining, i.e. open-cast or strip mining (Prud'homme, 2010). This uses large areas of land (Central Florida – 570 billion ha), may require altering of groundwater aquifers, destroys ecosystems, and impacts biodiversity and land productivity (UNEP, 2001). The overburden soil to ore ratio is 3.5:1 (Notholt *et al*., 1989). Average mining extraction efficiency is 80% and lower-content ore is left in place (Prud'homme, 2010). The tailings and overburden soil leach phosphorus and may cause eutrophication (Kuo and Munoz-Carpena, 2009). Phosphate ore is beneficiated through separation from sand and clay to produce phosphate rock concentrate of 28-40% $P_2O_5$ (IFA, 2011). Each ton of rock is accompanied by two tonnes of sand and clay (BCS Inc., 2002). The processes used are wet or dry screening, washing, flotation and drying. They consume water, chemicals and energy, and result in sand and clay tailings but also in contaminated effluents (Fig.4). After transportation to the chemical plant (often in a different country and/or continent), the phosphate concentrate is reacted with sulfuric acid to produce phosphoric acid. Globally, more than 60 percent of the industrial sulphuric acid is used to produce fertilizers (Scholz and Wellmer, 2013). The reaction between phosphate and sulfuric acid produces waste calcium sulfate (phosphogypsum, PG) and gaseous emissions of hydrofluoric acid (HF) and SiF4. Fluorides have been shown to have adverse impacts on grazing cattle (Rutherford *et al*., 1994). Five tonnes of PG is produced for each ton of phosphoric acid (EFMA, 2000). Billions of tonnes of PG have been produced to date and 160-200 million tonnes are added to the stacked PG piles each year. Although this waste product could be used

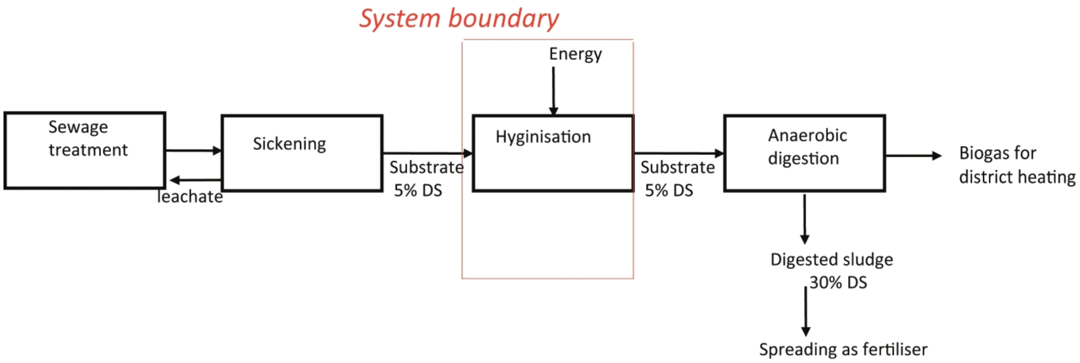

**Fig. 2. System boundary for the recycling from sewage sludge**

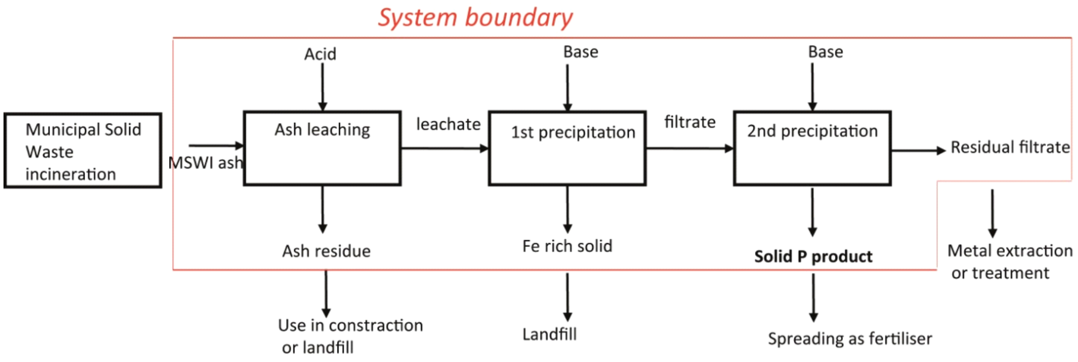

**Fig. 3. System boundary for the recycling from MSW incineration fly ash**

**Table 1. Material requirement for phosphorus production by the acid route. See Supplementary Table 1 for the calculations**

| Material, Tonnes | Flow | Per 1 tonne $P_2O_5$ (100 %) produced | | | | Per 10 Tonnes P in mineral fertilizer |
|---|---|---|---|---|---|---|
| | | Mining | Beneficiation | $H_3PO_4$ production | SUM | |
| Crude phosphate ore | Input | 15.6 | | | 15.6 | 68 |
| Water | Input | | 15.6 | 147 | 163 | 711 |
| Energy, toe | Input | 0.02 | | 0.01 | 0.03 | 0.1 |
| Benefication chemicals | Input | | 0.04 | | 0.04 | 0.2 |
| $H_2SO_4$ | Input | | | 2.7 | 2.7 | 12 |
| Overburden soil | Output | 54.6 | | | 54.6 | 238 |
| Sand and clay | Output | | 6.0 | | Accounted in crude ore | |
| Phosphogypsum | Output | | | 5.0 | 5.0 | 22 |
| **TOTAL** | | | | | **240-1=239** | **1047** |

in the manufacturing of sulfuric acid and as a road aggregate, this is rarely done and the USA EPA has banned all use of PG due to its radium content (above 10 pCi Ra/g, decomposition releases radon) and heavy metals. Dry PG piles may cause pollution through dust.Rock phosphate processing and use cause mobilization and dissipation of a range of contaminants at the mining and beneficiation sites, but also through fertilizer application. Sedimentary phosphates (83%, the rest are of igneous origin) contain low levels of more than a dozen elements—including As, Cd, Cr, Hg, Pb, U, F and V—which in excessive concentrations pose hazards to health. Cadmium is the most enriched element in phosphate rocks, occurring in concentrations almost 70 times higher than in average shale (van Kauwenbergh, 2010). The average global Cd level in phosphates is about 21 mg Cd/kg of rock, but some Moroccan rocks contain up to 40 mg, and phosphates from Togo and Tunisia contain up to 50–55 mg Cd/kg. Potential radiological impacts result from direct exposure, surface run-off and emissions to air from phosphogypsum piles. Guimond and Hardin (1989) reported that phosphate rocks generally have high concentrations of 226Ra. Uranium isotopes form highly soluble compounds with phosphate ions, while Ra isotopes 210Pb and 210Po concentrate in phosphogypsum. Saueia *et al.* (2005) found percentages (phosphogypsum to ore rocks) of 90% for 226Ra, 100% for 210Pb and 78% for 210Po.

The conventional P production process is highly dissipative and its material requirement is 239 Tonnes per produced tonne of $P_2O_5$ (Table 1). The Total Material Requirement (TMR) was calculated based on the annual demand for mineral fertilizer phosphorus in Sweden, by adding the average amount of mineral fertilizer imported in 2009-2002 (SCB, 2013), 10 t P·y$^{1}$, to the material requirement for the production of 10 t P by the acid route. This means that the TMR for the phosphate fertilizer imported to Sweden annually (10 Tonnes) corresponds to 1047 Tonnes of consumed resources and produced waste. It should be noted that the most advanced phosphate producers, for example in Florida, USA, are recycling most of the process water and are obliged to use the overburden soil to restore the mining sites. However, these measures are not yet adopted worldwide. Sewage sludge is a known sink for anthropogenic phosphorus flows. Humans excrete 98% of the phosphorus consumed in food and this accumulates in sewage sludge. In the EU, 0.6 Kg P/cap per year is found in sludge (Dawson and Hilton, 2011).The simplest way to recycle phosphorus from sludge is to spread it directly onto arable land, where P and N can be recycled as nutrients, although its heavy metal, organic pollutant and pathogen contents may cause damage to both humans and ecosystems. REVAQ certification was introduced in Sweden to encourage the improvement of sludge quality through reduction in contaminants (REVAQ, 2013).

The disposal of sewage sludge to landfills, often via incineration, is common worldwide. The Netherlands incinerates 60% of its sewage sludge; Japan, Germany, the United Kingdom, and France incinerate at least 20% of their sludge (Dawson and Hilton, 2011; Matsubae-Yokoyama *et al.*, 2009). In Sweden, landfilling of sewage sludge was banned in 2005 and the Swedish government has set a goal of recycling 60% of the phosphorus from sewage sludge by 2015 (Swedish Government, 2005). Over 70% of the sludge in Sweden is certified as safe for recycling to agriculture, but only 25 % was recycled to agriculture in 2010, while the rest was used in construction (SCB, 2013). The low degree of sludge recycling is due to real or perceived contamination by heavy metals and organic pollutants. In Switzerland, The Netherlands and Germany, the spreading of sludge is banned and another recycling route is being developed through mono-incineration of sludge with subsequent phosphorus extraction from the ash (Adam *et al.*, 2009). The phosphorus content in the sewage sludge is 3% dry weight (DW) (7% as $P_2O_5$). This means that to recycle 1 tonne of $P_2O_5$, 14 tonnes of sludge DW has to be spread

**Table 2. Material requirement for phosphorus production from MSWA**

| Material | Flow | Quantity | Unit |
|---|---|---|---|
| MSW ash | Input | 84 | Tonnes |
| HCl* | Input | 31 | Tonnes |
| CaO | Input | 11.0 | Tonnes |
| Energy | Input | $0.5 \cdot 10^{-3}$ | Toe |
| Ash residue | Output | 63 | Tonnes |
| Filtrate* | Output | 830 | Tonnes |
| Fe- precipitate* | Output | 17 | Tonnes |
| **TOTAL** | | **1036** | |

*are avoided in the modified method

onto the land. Under hyginisation the substrate is heated to $70^0$C during one hour, which requires 333 kWh per tonne sludge DW (Pettersson, 2001). In such a way, the 14 tonnes of sludge DW require about 0.42 toe (tonnes oil equivalents) of energy. In fact, the biogas from the anaerobic digestion is used for sludge hyginisation. The alternative use of the biogas is the district heating. In Sweden, more than 50% of the MSW is incinerated with energy recovery (Avfall Sverige, 2012). Even though incineration offers several advantages over e.g. landfilling, including mass and volume minimization and destruction of microorganisms and organic compounds, the residues, i.e. the ashes, have to be landfilled or used as a construction material within landfills. However, the ashes are also a potential resource, as they are a major sink for the material flows of society, including phosphorus and metals. A lab-scale method for phosphorus recovery from MSWA has recently been developed (Kalmykova and Karlfeldt Fedje, 2013). The final product is a solid containing about 3 w% of P (7% as $P_2O_5$). The resource requirements are based on laboratory experiments (Table 2). Electricity is required for the slurry agitation in a reaction chamber and for the anaerobic digester in the waste water treatment. An electricity consumption of 0.008kWh/m$^3$ for a cylindrical tank with turbine mixer was assumed and calculated for 830 m$^3$ of filtrate. The method is material-intensive due to the large volume of the water and the acid inputs required for controlling the pH. In addition, the residual products, such as ash residue, Fe rich intermediate precipitant, and the filtrate were all considered waste in this assessment. On the other hand, several modifications are currently being tested, which decrease methods resource requirement to 146 tonnes and below the conventional P production TMR. In particular, clean water is completely replaced by the acidic process water, produced at incineration plants with wet flue gas cleaning (a regular technology). Today, this liquid waste is pre-treated by addition of a base at the plant and released to the municipal waste water treatment plant. Therefore, also use of chemicals like $CaCO_3$, NaOH and TMT used today to raise the pH of the acidic water is avoided, which accounts to 0.1 tonne chemicals per each tonne of the fly ash produced, i.e. 8.5 tonnes for the 84 tonnes of ash required to produce 1 tonne $P_2O_5$. Approximate calculations on an average incineration plant show the process water is sufficient in volume to treat the fly ash generated in the same plant. However the process water is generally not acidic enough to create the low pH necessary for effective P leaching and consequently, some HCl (one fifth of the original volume, i.e. 7.5 tonnes to produce 1 tonne $P_2O_5$) is required. Another method modification aims to enable metals to be recovered from the ash prior to the phosphorus recovery. This would produce additional resources and also allow phosphorus extraction to be carried out in one step, which avoids the intermediate Fe-rich precipitate. One of the drivers for the development of phosphorus recycling is mitigation of the environmental impacts associated with its conventional production from ore. The impacts relate to both resource consumption and emissions, as illustrated in Fig. 4. The entire impact of the Swedish P demand can be avoided by recycling urban sources of P such as food waste, sewage sludge and MSW incineration residues. No inflow of the mineral fertilizer to the country is required in order to satisfy the P demand, because the necessary P amount is imported to the country as the content of food (Linderholm *et al.*, 2012). Consequently, the P flow in sewage sludge has been estimated to 4095 ton$^{\bullet}$y$^{1}$ P and in food waste to 2870 ton$^{\bullet}$y$^{1}$ P (Linderholm *et al.*, 2012). The estimated amount of phosphorus in MSW ash and slag is 4400 ton$^{\bullet}$y$^{1}$ P (Kalmykova et al., 2012). Applying the 70% recovery of phosphorus obtained in the initial laboratory test, this would yield 3080 ton$^{\bullet}$y$^{1}$ P (Kalmykova and Fedje, 2013).

## CONCLUSIONS

From the point of view of the TMR, sewage sludge recycling is the best of the investigated options. Only hyginisation needs to be added to the current sludge treatment. Also recycling of P from MSWA could offer a lower than conventional method TMR, provided the described method modifications are implemented. The entire impact of the Swedish mineral fertilizer demand can be avoided by recycling urban sources of phosphorus such as food waste, sewage sludge and MSW incineration residues. This would contribute to resource security and avoid the impacts of the production from the non-renewable rock resources.

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