PHOSPHOGYPSUM

Sustainable Management and Use

A Report for IFA Members

AE ''Johnny'' Johnston, General Editor

Paris, January 2016

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To obtain paper copies, contact IFA.

International Fertilizer Industry Association
28, rue Marbeuf
75008 Paris – France
Tel: +33 1 53 93 05 00
Fax: +33 1 53 93 05 45/ 47
publications@fertilizer.org
www.fertilizer.org
Twitter: fertilizernews

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Abbreviations and glossary

Abbreviations

ALARA	As Low As Reasonably Achievable
Bq	Becquerel
BSS	Basic Safety Standards
Ca	calcium
$CaCO_3$	calcium carbonate
CEC	Cation Exchange Capacity
CO_2	carbon dioxide
CSR	Corporate social responsibility
CX	Comprehensive extraction
DAP	diammonium phosphate
EC	electrical conductivity
ESIA	Environmental and Social Impact Assessment
EU	European Union
FA	fly ash
FCA	Full Cost Accounting
FEW	Food-Energy-Water Security
FIPR	Florida Industrial and Phosphate Research Institute
FSP	Fundamental Safety Principles
H_3PO_4	phosphoric acid
HAP	hazardous air pollutant
HF	hydrogen fluoride
HSE	health, safety and environment
IAEA	International Atomic Energy Agency
ICRP	International Commission on Radiological Protection
IFA	International Fertilizer Industry Association
IMO	International Maritime Organisation
K	potassium
LNT	Linear No-Threshold
MAP	monoammonium phosphate
Mg	magnesium
MSDS	Materials Safety Data Sheet
mt	million (metric) tonnes
NORM	Naturally Occurring Radioactive Materials
P	phosphorus
P_2O_5	phosphorus pentoxide (phosphoric acid)
PA	phosphoric acid
Pb	lead
PG	phosphogypsum ($CaSO_4 \cdot nH_2O$)
Po	polonium
PR	phosphate rock
OHS	occupational health and safety
Ra	radium
REE	rare earth elements
S	sulphur
SD	sustainable development
SO_2	sulphur dioxide
Sv	Sievert
TBL	Triple Bottom Line
Th	thorium
TSP	triple superphosphate
U	uranium
UNECE	United Nations Economic Commission for Europe
UNFC	United Nations Framework Classification
UNSCEAR	United Nations Scientific Committee on the Effects of Atomic Radiation
USEPA	United States Environmental Protection Agency

Glossary

Activity. The quantity A for an amount of radionuclide in a given energy state at a given time, defined as:

$A(t) = dN/dt$

where dN is the expectation value of the number of spontaneous nuclear transformations from the given energy state in the time interval dt.

The SI unit for activity is reciprocal second (/s), termed the becquerel (Bq). [170]

Activity Concentration. The *activity* per unit mass or volume typically measured as Bq/g or Bq/l. The term is used for any situation where the *activity* is in the form of *contamination* in or on a material). [170]

Arising. Materials forming the secondary or waste products of industrial operations.

Beneficial use. The desired outcome of the application of the Waste Hierarchy to materials under consideration for disposal is that beneficial use is found for them. This requires that the use should be technically feasible, environmentally neutral or benign and proportionate in regard to cost. It also requires consideration of use "as is" or "made useful" by further treatment or processing.

Characterisation. The mandatory first step in the decision-making process for determining the suitability of phosphogypsum (PG) for particular types of reuse or recycling is characterisation (Figure 7. PG Characterisation for use). Characterisation requires taking samples from a predefined series of locations whether at the filter or

the stacks and an analysis of the appropriate biological, chemical, physical and radiological properties to fully characterize the PG for the use for which it is intended.

Contamination. Radioactive substances on surfaces, or within solids, liquids or gases (including the human body), where their presence is unintended or undesirable, or the process giving rise to their presence in such places.

- Contamination does not include residual radioactive material remaining at a site after the completion of decommissioning. [8]

Co-product. The result of a chemical reaction from which two different products are formed, as for example phosphoric acid and phosphogypsum.

Decontamination. The complete or partial removal of contamination by a deliberate physical, chemical or biological process.

- This definition is intended to include a wide range of processes for removing contamination from people, equipment and buildings, but to exclude the removal of radionuclides from within the human body or the removal of radionuclides by natural weathering or migration processes, which are not considered to be decontamination.

Exemption. The determination by a regulatory body that a source or practice need not be subject to some or all aspects of regulatory control on the basis that the exposure due to the source or practice is too small to warrant the application of those aspects.[30] [8].

Exposure. The act or condition of being subject to irradiation. (IAEA Note: *Exposure* should not be used as a synonym for *dose*. *Dose* is a measure of the effects of *exposure*.)
Exposure can be divided into categories according to its nature and duration (see *exposure situations*) or according to the *source* of the *exposure*, the people exposed and/or the circumstances under which they are exposed.

Exposure situation.

- *Acute exposure.* Exposure received within a short period of time. Normally used to refer to exposure of sufficiently short duration that the resulting doses can be treated as instantaneous (e.g. less than an hour).
- *Chronic exposure.* Exposure persisting in time.
- *Existing.* Exposure already present before decision on control is made. (Retrospective)
- *Planned.* Applied as part of an intentional, planned, controlled situation or process. (Prospective)
- *Emergency.* Unexpected, uncontrolled or uncontrollable situation. (Reactive)

Green chemistry/Green engineering [23]. The invention, design and application of chemical products and processes to reduce or to eliminate the use and generation of hazardous substances.
www.acs.org/content/acs/en/greenchemistry/what-is-green-chemistry/principles/12-principles-of-green-chemistry.html
www.incaweb.org/research/green_chemistry/

Intervention. Any actions intended to reduce or avert exposure or the likelihood of exposure to sources which are not part of a controlled practice (or an exempt practice) or which are out of control as a consequence of an accident.

London Convention. The IMO "Convention on the Prevention of Marine Pollution by Dumping of Wastes and Other Matter 1972", the "London Convention" for short, is one of the first global conventions to protect the marine environment from human activities. It has been in force since 1975. www.imo.org/OurWork/Environment/LCLP/Pages/default.aspx

London Protocol. In 1996, the IMO "London Protocol" was agreed to further modernize the London Convention and, eventually, replace it. Under the Protocol all dumping of wastes to sea is prohibited, except for possibly acceptable wastes on the so-called "reverse list". www.imo.org/OurWork/Environment/LCLP/Pages/default.aspx

NORM (Naturally Occurring Radioactive Materials). *Radioactive material* containing no significant amounts of radionuclides other than *naturally occurring radionuclides*.

- The exact definition of 'significant amounts' would be a regulatory decision.
- Material in which the *activity concentrations* of the *naturally occurring radionuclides* have been changed by a *process* is included in *naturally occurring radioactive material*.
- *Naturally occurring radioactive materia*l or NORM should be used in the singular unless reference is explicitly being made to various materials.

Pathway (exposure). A route by which *radiation* or radionuclides can reach humans and cause exposure.

Phosphogypsum. Calcium sulphate. A co-product with phosphoric acid of the wet process production of phosphate fertilizers.

Practice. Any human activity that introduces additional sources of exposure or exposure pathways or extends exposure to additional people or modifies the network of exposure pathways from existing sources, so as to increase the exposure or likelihood of exposure of people or the number of people exposed.

Radioactivity. The emission of particulate or electromagnetic radiation as a result of decay of the nuclei of unstable elements, a property of all chemical elements of atomic number above 83. Scientifically, something is described as radioactive if it exhibits the phenomenon of radioactivity or if it contains any substance that exhibits radioactivity. Scientifically, therefore, virtually any material (including material that is considered to be waste) is radioactive. However, it is common regulatory practice to define terms such as *radioactive material* and *radioactive waste* in such a way as to include only that material or waste that is subject to regulation by virtue of the radiological hazard that it poses. [170]

Remediation. Any measures that may be carried out to reduce the radiation exposure due to existing contamination of land areas through actions applied to the contamination itself (the source) or to the exposure pathways to humans.

- Complete removal of the contamination is not implied. [17]

Resource. Naturally occurring materials for which there is a reasonable prospect of economic use (valorisation).

Soil fertility. The ability of a soil to produce the required or optimum level of yield and quality from a given crop, at a given time and under given growing conditions, assuming appropriate, measurable inputs.

Source. Anything that may cause *radiation exposure* — such as by emitting *ionizing radiation* or by releasing *radioactive substances* or *material* — and can be treated as a single entity for protection and safety purposes.

- *Natural source*. A naturally occurring *source of radiation*, such as the sun and stars (sources of *cosmic radiation*) and rocks and soil *(terrestrial sources of radiation*).

Sustainability. The preservation of the environment's ability to meet both present and future needs.

Sustainable development. Development that meets the needs of the present without compromising the ability of future generations to meet their own needs. It contains within it two key concepts:

- the concept of *needs*, in particular the essential needs of the world's poor, to which overriding priority should be given; and
- the idea of *limitations* imposed by the state of technology and social organization on the environment's ability to meet present and future needs. [15.]

Valorisation. The economic use of a resource across its whole life-cycle.

Waste (EU). "Any substance or object which the holder discards or intends to discard".

Waste (IAEA). "Material for which no further use is foreseen".

- *Exempt waste*. *Waste* that is released from *regulatory control* in accordance with *exemption* principles.
- *Mixed waste*. *Radioactive waste* that also contains non-radioactive toxic or hazardous substances.

NORM waste. N*aturally occurring radioactive material* for which no further use is foreseen.

Waste Hierarchy. The EU Waste Framework Directive [9] establishes a strategy for prioritising management of "waste" in a hierarchical form as follows (Figure 2):

1. Prevention
2. Re-use
3. Recycling
4. Processing or recovery
5. Disposal

Of these the least favoured option is disposal.

Foreword

Charlotte Hebebrand
IFA, Director General

I am pleased to present IFA members with this Report addressing rapidly changing scientific, regulatory and policy perspectives concerning phosphogypsum (PG) management and use worldwide. It has been authored and reviewed by many of the world's top experts on PG from academia, international agencies and representatives of IFA's own member companies.

There are three main reasons that propelled IFA to commission this work. Foremost, we felt it important to inform IFA members that the Phosphate Industry Safety Report published by the International Atomic Energy Agency (IAEA) in 2013 has classified PG as a co-product of phosphoric acid production, rather than as waste, and is hence safe to use. This conclusion was based on a landmark scientific risk assessment which concluded that it is the relative level rather than the mere presence of naturally occurring radioactivity in PG that needs to be considered when assessing its safety. The IAEA's principal finding is that there is no radiological reason for preventing PG use. This has already paved the way for rapid growth in beneficial applications of the material, notably in agriculture, mine restoration, construction and in reprocessing PG into valuable new products such as affordable fertilisers. Secondly, we wanted to document in some detail that although the regulation of PG varies greatly between countries, some major producing countries have already opted to remove obstacles to beneficial uses of PG leading to the consumption of millions of tonnes per year. Hence today, 25-30% of PG produced annually finds its way into diverse applications and use is growing strongly. Thirdly, these new horizons on PG use raise questions on the sustainability of long-term stacking or disposal at sea.

Changes in approach to PG are to be understood in the context of wider changes of attitude to disposal of any material considered a waste. The concept of a waste hierarchy, which sees disposal of any waste as a last and undesired resort, urges the global community to focus on recycling any materials before discarding them. This greatly strengthens the case for PG use rather than disposal. Against this background, it will be important to engage further with policymakers and stakeholder groups, who have an understandable caution in all matters relating to safe and beneficial uses of naturally occurring radioactive materials (NORM) which phosphates contain. This Report provides IFA members with some guidance on these matters and examples of successes.

At this time of change in the phosphate industry this document provides not an IFA position on PG but a factual review, for IFA members only, of key developments in hand. Our purpose is to enable members to better analyse and plan their own future strategies of PG management. Even when safety concerns are overcome, important questions remain with regard to the economics of PG use and the costs of storage. This dimension is given particular attention in the case studies in the Report.

In thanking all those who have contributed, allow me to single out Johnny Johnston of Rothamsted Research, who has played such a valuable role as General Editor and Chair of the Scientific Expert Panel. Thanks are due also to Brian Birky, Florida Industrial & Phosphate Research Institute, and Julian Hilton, Aleff Group, who with Johnny have led the editorial team. They have been ably assisted throughout by distinguished independent expert contributors and peer reviewers as well as by members of IFA's NORM Working Group who have provided valuable input into the PG Report and participated so actively in IFA discussions on this topic.

Last but not least, I want to acknowledge the efforts of Volker Andresen and Michel Prud'homme of the IFA Secretariat for so effectively moving this project forward.

Introduction

AE “Johnny” Johnston
Rothamsted Research,
General Editor

Phosphogypsum (PG) and phosphoric acid are the co-products from the dissolution of phosphate rock (PR) with sulphuric acid in the wet process that is used by most phosphoric acid producing IFA member companies. More than 90% of the PG is hydrated calcium sulphate (gypsum). This is precipitated in the latter stages of the reaction and is separated by filtration. The gypsum filtrate contains some phosphorus (P), hence coining the name phosphogypsum – often shortened to PG. Each tonne (t) of acid as P_2O_5 yields some 5 t PG as co-product. The central issue of this Report is what to do with very large amounts of PG produced each year in a financially viable and environmentally acceptable way.

It has long been known that PR contains Naturally Occurring Radioactive Material (NORM) notably uranium and radium. Radioactivity from any source is a major concern to many humans because it is very frequently linked to cancer without realising that it is the length of exposure and dose rate that are important. Both sedimentary and igneous PR contain heavy metals such as cadmium but all PR is very variable in composition. The concentration of each element in it varies widely between PR sources. In the wet process of treating PR with sulphuric acid, uranium and most of the heavy metals go to the phosphoric acid while other radioactive forms of elements, principally isotopes of radium, go with the PG.

Phosphoric acid has many and varied uses but the major applications are in crop production and animal feed supplements. Phosphatic fertilizers are essential to building up and maintaining the plant-available supply of P within the soil and likewise contribute vitally to animal growth and health. Both uses are at the heart of sustaining food production to feed the world’s rapidly growing population. Phosphogypsum has a number of well-established uses and an ever-growing array of new ones. Combined, these proven and potential uses could consume most if not all of the PG produced annually. But a long history of managing PG as a waste not a resource has left a legacy of concern in some countries about its safety which will take patience and careful engagement with stakeholders to resolve.

Current constraints on the widespread use of PG are related to its content of NORM. It is because of NORM issues that much of the PG produced globally in the past 25 years has not been used. Questions are raised about its safety given the link between exposure to radioactivity and the risk of cancer in humans. So the first and crucially important question this Report addresses is whether or not PG is safe to use. Is all the PG currently produced safe from a radiological perspective? Should there be limitations to its use? How should PG best be characterised such that any user can be assured about its safety both in respect of its intended application and in respect of its traceability to the source?

This Report provides IFA member companies with in-depth information on the safety aspects of PG (as for example in Section 1.3) including extensive references to peer-reviewed international literature, starting with two major publications of the International Atomic Energy Agency (IAEA). Founded in 1956, the IAEA is an international treaty organisation with 165 signatory Member States (as of September 2015). Under its Charter, Member States are expected to follow its standards in regard to radiation protection and radiation safety, which in most countries have the force of law. In 2013, IAEA published Safety Reports Series No. 78 on Radiation Protection and Management of NORM Residues in the Phosphate Industry. This IAEA Report concluded that provided that the ^{226}Ra activity concentration in PG was 1 or less than 1 Bq/g it was safe to use without restriction and even above this level uses are encouraged, but with certain restrictions. On this basis PG was classified as a co-product of phosphoric acid production. As most of the PG produced globally has less than 1 Bq/g, the IAEA advises that there is no obstacle to PG use on radiological grounds.

The publication in 2014 of the Revised Basic Safety Standards (BSS) of the IAEA, the standard international reference publication on all aspects of radiation protection and safety, has further strongly reinforced the findings of Safety Reports Series No. 78. The revised BSS

does not require the elimination of all radioactivity or all traces of radioactive substances before a material such as PG can be used. Instead it focuses on finding an appropriate balance between risk and benefits from radiation protection policies and practices. Numerical values are set for measures of dose or activity concentration that will ensure the desired level of radiation protection and safety for those managing or using these materials or products derived from them. But the chosen means for ensuring safety, including safe use, are expected to be reasonable, appropriate, proportionate, affordable and sustainable. A risk-based approach is required and there must be clear evidence that protective measures bring commensurate benefits. If such evidence cannot be adduced the measures should be adjusted or removed.

Against that background the primary conclusion of this Report is that PG is a safe resource for which there are many beneficial uses. Once both regulators and stakeholders understand the consequences of the recent reassessment of the safety aspects related to PG the task of revising regulatory positions to facilitate the use of PG should be much easier at the same time answering stakeholder concerns.

Long-established and large volume uses of PG are focused on agriculture and construction, including building materials. New applications are emerging such as in the marine environment, for example for coastal defences. PG is also a focus of attention for the recoverable resources it contains, such as sulphur and rare earths. Descriptions of these applications are given in some detail. Currently many technological improvements are being made or considered that will benefit the use of PG.

From an IFA member company perspective the PG producer does not have to be the user. Every opportunity should be taken to create business partnerships with other organisations that can use PG to the benefit of both organisations. Many of the examples given in this Report illustrate how this is already being done. A possible limitation to the rapid widespread use of PG will be the need to convince regulators to modify local regulations and for the many stakeholders to be assured that any relaxation of existing regulations is fully justified.

Because of the natural variability of phosphate rock, PG itself can be very variable in content. Once the decision is taken to use rather than discard it, it should be fully characterised, notably in regard to radionuclide and heavy metal content, which will greatly assist in finding appropriate and economically viable applications according to local needs and market conditions.

Already there are some answers to the issue of the safety of PG use in the long-term. One of the oldest is from the Rothamsted long-term, Classical Experiments. J. B. Lawes took out his patent for the manufacture of single superphosphate from various phosphatic materials, including phosphate rock (PR), in 1842. From 1843 until 1973 single superphosphate was applied each year at 410 kg/ha to supply 33 kg P/ha on phosphate-treated plots in a number of long-term experiments. Initially the superphosphate was made from bones but from the 1880s it was from PR. Thus all the calcium sulphate (PG), produced by the reaction of PR with sulphuric acid, and all the elements in the PR were added to the soil each year where single superphosphate was applied. There is no evidence that after 90 years of applying this large amount of single superphosphate each year there has been any adverse effect of the accumulation of these other elements on soil fertility or crop yields.

Although there has been some accumulation of cadmium in soil crop yields are not affected and there is little uptake of this cadmium into plants where soil acidity is maintained above pH 6.

The same experience is replicated worldwide: there does not seem to be any recorded evidence of the adverse effects from the global use of single superphosphate. Following the introduction of the wet process for producing phosphoric acid from PR and the separation of the PG from the acid it would be logical to assume that this PG would have no adverse effects as when applied as an integral part of single superphosphate. However, logic, based on evidence, does not appear to have prevailed everywhere!

In recent years global stakeholders have become more and more aware of the need to conserve global resources and protect the environment as the world's rapidly growing population puts increasing demands on the earth's resources. Their strongly and often correctly voiced views are requiring regulators to take appropriate action. Now that the safety issues surrounding PG are better understood and accepted its production and use can be seen within these wider objectives of resource conservation and environmental protection. To help achieve these wider objectives now is an appropriate time for the phosphate industry to strengthen its links with science and academia to seek new approaches to, for example improving the recovery of phosphorus from phosphate rock, removing radioactive elements from PG, recovering the rare earth elements, and recovering sulphur from PG. Looking even further ahead it would be worth a concerted research effort to identify an economically viable process that would make sufficient quantities of plant-available phosphorus from phosphate rock without the need for using strong acids.

Over the past decade the world has become more conscious of the adverse effects on the environment posed by disposing of waste, a trend which has significantly influenced the mining and processing industries, such as phosphate. Thus the topic of zero waste is becoming of major interest to stakeholders and regulators as they seek to protect the environment for the benefit of future generations. Many industrial processes were de-

signed initially to produce a specific product and anything else was considered a waste. In this context PG was seen as a waste to be disposed as cheaply as possible either by stacking on land or discharge to the sea. Both methods of disposal had little regard for any possible adverse environmental effects or resource conservation. As demand to limit waste to conserve resources and improve the environment has increased, the concept of a five step waste hierarchy for the prevention and management of waste (see Section 1.4) has been developed both in Europe and America. When applying the EU five-tier waste hierarchy, before the fifth and least desired step of seeking approval to dispose of any material as a waste, a producer has to consider and if possible adopt one or more of four steps namely, prevention, minimisation, reuse and recycling. Steps 3 and 4 are applicable to PG because it can be reused and recycled as a resource. This hierarchy has already been widely adopted by law- and policy-makers and is applied in many countries by regulators when permission to consider a material as a waste is being requested.

Now that PG can be considered as a co-product it is increasingly being seen as a resource of value within the context of sustainable development, and minimising adverse environmental effects. The world does not have limitless resources and safeguarding the resources we have is of paramount importance especially as the world population increases with increasing demand on those resources. Most of the examples in this Report that illustrate the use of PG also show its role as a resource, for example, in building materials, road construction, and for producing other compounds. Many of these uses of PG help conserve other virgin resources, for example using PG to replace mineral virgin rock in road construction.

In moving towards a strategy for use, perhaps the biggest challenge facing the IFA member company is how to engage with their national regulator, if one exists. The regulatory position worldwide is changing fast, but remains very diverse. Hence a company may be subject to one or more of five principal types of regulatory framework: 1. no framework; 2. encourages use by removing regulatory obstacles, as in Brazil and India; 3. requires a minimum level of use, as in China; 4. is changing fast, but at varying rates, as in the United Kingdom, Russia, Spain, and Sweden; 5. heavily restricts use, use is completely or almost completely prohibited as in the US. Where change is happening (category 4) it is quite radical. For example, in the UK for NORM residues where radioactivity is at or less than 1 Bq/g ^{226}Ra, PG is now deemed out of scope of regulation and can be used without restriction. Even calling PG radioactive is discouraged because it deters market uptake and labelling NORM residues of this kind as radioactive will be prohibited.

The trend at least is clear, that use is increasingly encouraged and with that encouragement use is growing. It is to be hoped in due course that a uniform, consistent and evidence-based approach will become the international standard, based on the IAEA reference publications. Each company can then seek beneficial uses and markets for the PG it produces with some key obstacles removed.

Using all the PG produced would be a major contribution to global resource conservation and to the benefit of the environment. But such uses must also be affordable, most of all for the producing company. The many safe and beneficial applications described in this Report will help IFA member companies to better analyse and plan their individual strategies for safe, affordable, even profitable uses of the PG they produce.

Executive Summary

Data supplied in response to an IFA survey demonstrates that phosphogypsum (PG) use worldwide has increased significantly since 2008. From a baseline of near zero use, by early 2015 IFA members were reporting that 35-40 million tonnes (mt) PG would be consumed during the year and more recent estimates suggest a usage level of some 25% of annual production by year end. There is every reason to expect that the growth trend will continue in the coming decade.

The large volume uses of PG are in mine restoration, agriculture and construction – including wallboard, cement, building materials and ceramics. But there are many more smaller volume uses, with an increasing number focused on value-added products and materials, such as the recovery of rare earth elements and sulphur, the production of premium high-strength alpha gypsum and the reprocessing of PG into other products such as ammonium sulphate and calcium carbonate. The change has come about at both international and national levels.

Phosphate production is classed as a NORM industry because phosphate rocks contain Naturally Occurring Radioactive Materials (NORM). For the past twenty-five years the presence of NORM in PG has been the single most significant obstacle to its use. In April 2013 the International Atomic Energy Agency (IAEA), the global reference authority for radiation safety, published a Safety Report on the phosphate industry (Safety Reports Series No. 78), detailing the findings of an evidence- and risk-based review and consultation started in 2006. The IAEA concluded that there were no well-founded scientific reasons for prohibiting or discouraging PG use on radiological grounds and accordingly re-classified PG as a co-product of phosphoric acid production and not a waste. It further concluded that using PG was preferable to storing it, not least on environmental grounds.

Some producers have a long history of treating PG as a co-product of phosphoric acid. More recently, major producer-consumer countries such as Brazil, China, India and Russia with active support and even leadership from their national regulators, have taken various steps to remove obstacles to creating a market for PG and PG-based products. For example, since 2008 India has declared agricultural applications free from all restrictions, while in China since January 2015 a minimum of 20% use is mandatory. One Chinese producer has already reached 100% use, including use for mine restoration, for which it has been recognised with a national Green Mine Award.

While some of this growth in PG use has resulted from direct engagement with modifying the perceived negative health, safety and environmental profile of PG to a more positive image as secondary resource, some of the growth has come about for other reasons such as denials by regulators or stakeholders of permits for extending stacks. In the EU, for example, the disposal of any material as a waste is the last and least desired outcome in a waste management strategy. So use of materials such as PG achieves the sustainable development goal of not imposing the burden of unwanted waste legacies on future generations. Before any material can be classified as waste all reasonable options for reuse or recycling must first be applied.

Where such options do not currently exist, or are too expensive, research and development is encouraged to seek alternatives. While IAEA has removed the radiological objections to PG use, the challenge remains of its volume. The chemistry of the wet process for making phosphoric acid by digesting phosphate rock in sulphuric acid, which is the industry-standard production method, brings with it a very large volume of PG – 5 t for every t of acid, as P_2O_5. Annual PG production now exceeds 200 mt. While investment is needed to develop new processing technologies to prevent or minimise PG production, in parallel wet process producers must continue their efforts to reuse and recycling PG as part of their wider commitment to following Sustainable Development Goals.

This commitment to use by IFA members is not new. Armand Davister started the process at an IFA Technical Committee meeting in 1998, arguing that PG is a resource not a waste. Paul Smith and Tibaut Theys made the case in 2000 for its profitable use. More recently the safe and sustainable uses of PG have been the focus of a series of successful meetings and workshops organised by the IFA NORM Working Group. The first was in Tashkent (2012) which was followed by Istanbul (2013), Amsterdam (2014) and Vancouver (2015). The success of these well-attended meetings and their importance to the phosphate industry encouraged the IFA leadership to support the publication of this Report - *Phosphogypsum: sustainable management and use*. It has been assembled, edited and peer-reviewed by an independent expert team but has also benefited from many active contributions of case

studies and wider experience from numerous experts from IFA member companies. This is the first time that IFA has been able to bring together in a single publication a snapshot of the very diverse aspects of PG worldwide. The aim is to enable IFA members to learn from the scientific community and from each other about options for PG use at a time of great and positive change in the policy and regulatory frameworks for NORM industries.

There is no cut and paste solution. No two IFA member companies are alike; there is no single message and no IFA position on PG. Each producer will find different opportunities for use, different market conditions and must satisfy specific local and regional economic, environmental and social conditions. Some producers may require years and incur significant costs in order to arrive at a clear action plan for transitioning from stacking to use. Consequently this Report includes business models and case studies that have been developed to facilitate such a transition. Producers can however be assured that such plans are being developed against an increasingly positive background. Worldwide, there is a clear trend towards increased secondary resource use, in many cases encouraged by regulators. In parallel, mining and extractive industries in general, faced with increasing public scepticism about waste-generating mining practices are progressively committing themselves to an operational policy of zero waste.

Leadership in such initiatives is coming from countries with a significant economic dependency on the mining sector: Australia, Canada, South Africa, and Saudi Arabia. Similar measures are being pursued in major mineral consuming economies such as China and India. The phosphate industry can contribute much to this goal.

The Report concludes that there is no known PG currently in production that cannot find a safe use, assuming four key conditions are met. First, a careful characterisation of physical and chemical properties with particular attention to radionuclide and heavy metal content will establish the basis on which the most appropriate uses can be selected. Secondly, national regulations may need to be aligned with IAEA recommendations to eliminate obstacles to markets and to use. Thirdly, any stakeholder concerns must be carefully and continuously addressed through science-based communications and education. Fourthly, leadership must come from within the industry itself to achieve a sustainable solution. This Report is part of that leadership initiative. The new policy and regulatory framework for NORM industries such as phosphates aims at a new equilibrium – to protect the environment and to promote economic opportunities and jobs. The need for such a new equilibrium was first articulated in economic theory by the Nobel Prize-winning mathematician and economist John Nash. Nash described certain types of economic processes in which parties in a transaction either both win or both lose. Applying this concept to PG shows that if PG is not valued and used as a co-product, both operator and society lose. This publication is designed to make it easier for operator and society to both win.

1 Phosphogypsum in the phosphorus life-cycle

KEY POINTS

- Phosphorus is critical for food supply and food security.
- Water soluble phosphate fertilizers, such as DAP, MAP, and TSP are essential for providing soils with sufficient phosphorus to assure crop yields.
- Commercial production worldwide uses the "wet process" for making phosphoric acid, which results in generating large tonnages of PG.
- There is a significant financial and environmental gain if PG can be used as a co-product and not discarded as a waste.
- Universal usage will require a paradigm shift in attitude and in some cases regulatory change based on better understanding of the range of safe, beneficial, tested options available. Such a shift will have distinct regional variations.
- The evidence is unequivocal: all known PG types can be used safely. Uses will vary according to characterisation and some may be restricted subject to their relative levels of radionuclides and heavy metal content.

1.1 Introduction to phosphates and phosphogypsum

Phosphorus (P) is essential to all life forms because it is a non-substitutable element in all plant and animal DNA. Plant roots take up P from the soil but it has to be in an available form. Since the mid nineteenth century phosphate fertilizers have been used to supplement the natural soil supply because most soils contain too little plant-available P. For the past sixty years the dominant method for making phosphate fertilizers has been from phosphoric acid. This acid, known industry-wide as phosphorus pentoxide, P_2O_5, is produced by the so-called "wet process" by which phosphate rock (PR) is digested in sulphuric acid.

It is generally well understood that managing the restricted global P resource efficiently, whether during fertilizer production by the operator or consumption by the farmer, is critical to the world's capacity to feed itself. It is not well understood that the "wet process" generates both phosphoric acid and a very plentiful co-product, phosphogypsum (PG) (Figure 1). PG consists primarily of calcium sulphate – gypsum – which, because it contains some residual P, is known as phosphogypsum.

In the context of a recent fundamental change in knowledge and understanding of the phosphorus life-cycle as a whole, PR is increasingly seen as a critical energetic mineral. Likewise, our understanding of the key role of P in maintaining soil fertility has advanced significantly such that phosphate use in agriculture can now be managed more efficiently [1]. The challenge these positive changes bring is how best to integrate PG into the management of the phosphorus life-cycle as a whole. This Report shows this need for integration is well accepted and is rapidly translating into new practices. Overall, our success as a global village in stewarding the whole life-cycle of our common phosphate resources will be a key indicator of how the wider goals of food, energy and water security are being met for a world population predicted to reach 9 billion by 2050.

FIGURE 1

Phosphoric acid and phosphogypsum

Five tonnes of PG are produced for every tonne of acid, as P_2O_5 making it by volume the phosphate industry's largest output. Assuming the wet process retains its position as the dominant production technology, the more phosphoric acid that is produced to manufacture fertilizers to feed the world - and we do need more - the greater will be the production of PG. Currently annual

production of PG is running at some 215 million tonnes (mt) per year[1], each mine and each production method generating subtly different types. What best to do with these large quantities of PG is now at issue, whether to use it as an essential resource in a number of tested and innovative applications or to continue the practice of the last 50 years in some producing countries and dispose of it indefinitely to land or to sea with the potential adverse environmental impacts associated with these methods of disposal.

This Report, prepared specifically for use by IFA Members, argues that PG has a number of very significant roles to play as a resource in today's world. As a soil amendment, PG can improve the yield potential of many soils, which will contribute significantly to food security and related efficiency of water uptake by crops. The many well-proven uses of PG in construction, construction materials and energy recovery help in significant ways to conserve virgin resources, such as aggregates and sulphur.

For a variety of reasons during the past 30 years PG has not been given the opportunity to fulfil these roles. Of these perhaps the most significant is the regulatory context, although the economics of use is clearly a major factor in some markets. Some jurisdictions effectively forbid the wide use of PG. Others now actively encourage it even to the point of setting minimum targets for use. In early 2015, China started requiring the use of 20% of production. Thus the situation is changing - in some countries very quickly - primarily in the direction of use. This process of change sets the stage for a radical reappraisal of PG as a resource to be conserved and used, and not as a waste to be discarded. The purpose of this Report therefore is to inform IFA members about the opportunities any changing regulatory context offers and to document the base of scientific knowledge and operational experience which has grown up over 40 years concerning beneficial use. Hence the great bulk of the content derives directly from the operators themselves supported by academic and scientific centres of excellence around the world.

1.1.1 Phosphate rock

Phosphate rock (PR) occurs in both igneous and sedimentary deposits. Sedimentary deposits are significantly more plentiful and are typically less costly to mine but they also contain the elements uranium and radium, classed as Naturally Occurring Radioactive Materials (NORM). Both uranium and radium are present in small concentrations broadly similar to the levels found in the world's soils. The rock also contains a very wide range of other elements, among them heavy metals such as cadmium and rare earth elements, most of potential commercial value. Initially, all these elements were deposited from the ocean into marine sediments.

When phosphate rock reacts with sulphuric acid, the calcium sulphate co-product incorporates any radium from the source rock and during processing substitutes for the calcium in it to make radium sulphate, the salt which naturally causes PG to be very slightly radioactive. When PG crystals are filtered from the phosphoric acid the radioactive elements and some other impurities, such as undigested rock, are also left in the solid PG.

For many years the combination of the presence of NORM and heavy metals such as cadmium has contributed to a perception of PG as a hazardous waste. Now a variety of factors, starting with an evidence-based reappraisal of the radiological risks posed by PG, has caused a fundamental rethink. The prevailing conclusion drawn from extensive current evidence is that the benefits of use far outweigh any real risks that PG may cause to people or the environment (Figure 6).

Finding a sustainable solution in operational terms to the most obvious challenge the phosphate industry faces is its sheer volume. But in some countries PG has such a negative image that the stiffer test is achieving public acceptance that its use is beneficial not harmful. Thus, producers will be required to change both the way they manage PG and the way they engage with stakeholders about its uses.

The fact that PG can be used does not mean it must be used: but when PG is used it must be in such a way that it aids not hinders the long-term operational and financial health of the producing company. At the same time its use must win the support and acceptance of all stakeholders in regards to its utility, its safety and its insignificant impact on the environment. but this places a strong emphasis on storage – whether short- or medium-term, or indefinite. Whatever the storage objective is, it must be managed and monitored in environmentally acceptable ways and according to technical and regulatory best practices.

1.2 A changing industry

In the first years after World War 2 the bulk of global phosphoric acid production was centred in Western Europe and the United States where resources were plentiful. Much of the PG produced was discharged to water, mostly to the seas and oceans but in some instances to rivers. Inland production sites tended to stack and some significant volumes of PG found use in both agriculture and construction, notably for roads. Saline soils in dry areas such as those in California showed very good crop response to applications of PG, and in the 1970s and 1980s demand by Californian farmers was sufficient for two stacks of PG to have been consumed fully by 1989. In a similar way PG was shown to be an excellent soil amendment for peanut

1 IFA survey and analysis, March 2015

farming and remains a product of choice in north Florida and Georgia.

In the 1980s as environmental pressures against discharging to sea increased, disposal to land (stacking) became either standard practice, as in Europe, or legally binding, as in the United States from 1989 [2]. By the late 1990s this had led progressively to the closure of most of the European phosphate industry where costs of stacking were prohibitively high or no suitable land was available. In the United States it led to the probably unforeseen creation of huge stacks of PG many now with footprints of several square kilometres.

Since 2000, the phosphate industry has experienced a period of intense change, initially a market-driven process, accompanied by an underlying Compound Annual Growth Rate (CAGR) in demand for phosphate fertilizers of 3-3.5%. In the European Union (EU), manufacturing sites in United Kingdom (UK), France, Greece, Italy, and Spain have all closed. More recently, driven by fears about risk of interruptions to the supply of PR to Europe, PR is now on the EU list of critical minerals. By contrast, the so-called BRICS economies (Brazil, Russia, India, China and South Africa) markedly increased their demand for fertilizers in general, and phosphates in particular, for food production [3].

Of these countries, China is undoubtedly the most significant in terms of phosphate production and consumption. In ten years China has risen from being a relatively minor player to become the largest producer/consumer in the world, displacing the United States from the top position. The country has changed from being a net importer to a net exporter as major new capacity has come on stream, to such an extent that it is now in structural over-capacity.

Brazil has increased phosphate fertilizer imports because its production capacity, although growing, is not keeping pace with domestic demand. India is almost completely reliant on imported phosphates, producing only 7% of its P_2O_5 needs from national resources, the balance of 93% relying on imports of rock and phosphoric acid. It does have some low grade PR deposits that may be exploited in the future because of their uranium content [4].

New plants are being planned or built in various parts of the world of which the most significant are the OCP Group Jorf Lasfar hub in Morocco and the Ma'aden Phosphate Company Umm Wu'al phosphate mine and processing complex and related Ras al Khair production facilities in Saudi Arabia.

1.3 The international framework for radiation protection and safety

Because phosphate rock contains NORM it typically falls under the framework for radiation protection and safety both at national and international levels. This means that from a regulatory point of view the lead international body is the International Atomic Energy Agency (IAEA). From a complementary scientific point of view, with particular emphasis on occupational, public and environmental health and safety, the lead body is the International Commission on Radiological Protection (ICRP). Typically, ICRP sets out the scientific principles and objectives for achieving an effective framework and IAEA sets out how to implement them in an operational setting.

1.3.1 The International Atomic Energy Agency

The International Atomic Energy Agency (IAEA)[2] is an autonomous member of the family of United Nations organisations, founded in 1956 under the rubric "Atoms for Peace". It has its own Statute [5], which came into force on 29 July 1957, and its own governance, the General Conference, which comprises representatives of all Member States. As of September 2015 it has 166 Member States[3]. Typically Member States have a permanent representative or Ambassador looking after day-to-day interests. The great majority of phosphate-producing countries are IAEA Member States, including all the leading producers.

A state joining IAEA as a full Member takes on a range of commitments and obligations as set out in a range of International Conventions and Legal Agreements of which IAEA is the depositary[4]. Additionally, the Agency is entrusted with responsibilities under treaties and agreements that States have adopted. The work of IAEA comprises three principal activities: i. nuclear technology and applications, ii. nuclear safety and security, and iii. safeguards and verification. These publications are commonly referenced in national laws and regulations either directly, or through regional counterparts based on the IAEA documents, such as Council Directive 96/29/Euratom in the European Union [6] "laying down basic safety standards for the protection of the health of workers and the general public against the dangers arising from ionising radiation".

The technical and scientific standards of IAEA are set out in a range of publications, some of which have the force of law, others of which are offered as guidance and good practice documents, while others provide technical and background information. Two of the most significant, legally binding documents, are the Fundamental Safety Principles (FSP) [7] and the Basic Safety Standards (BSS) [8]. These documents are kept under standing review and are periodically updated. The BSS was last updated in July 2014.

2 IAEA: www.iaea.org
3 List of IAEA Member States: www.iaea.org/about/memberstates
4 www.iaea.org/publications/documents/treaties

1.3.2 The International Commission on Radiation Protection

The scientific basis on which IAEA safety provisions such as the BSS rest are first developed by a complementary organization known as the International Commission on Radiation Protection (ICRP[5]). While IAEA is a treaty organization comprising Member States, ICRP, founded in 1928, "is an independent, international [professional] organisation with more than two hundred volunteer members from approximately thirty countries across six continents. These members represent the leading scientists and policy makers in the field of radiological protection". ICRP makes its work available to IAEA, national regulators, law makers and policy makers through a range of bi- and multi-lateral agreements, peer-reviewed publications, working groups and scientific meetings.

ICRP radiation protection measures are based on three principles which IAEA Member States universally follow. These are:

1. Justification;
2. Optimisation;
3. Dose limits.

These principles are of the highest significance in determining what measures for protection from risks from NORM may be justified and what may not.

1.3.3 The Basic Safety Standards 2014

The publication of the Revised Basic Safety Standards (BSS) 2014 [8] by the International Atomic Energy Agency (IAEA) marks a significant policy shift in the way radiation protection and safety measures are applied across the entire product life-cycle from mining to residue and waste management. This manifests itself in two main ways:

1. Numerical values such as measures of dose or activity concentration are set within the wider context of requiring Member States to put in place a strategic "framework for radiation protection and safety".
2. Desired outcomes form an essential part of the strategic framework – i.e. the aim when a particular regulation or safety measure is put in place has to be explained and justified, and means of verifying that the aim has been monitored and achieved have also to be shown.

The point is explained as follows in regard to remediation in the Revised Basic Safety Standards:

> "The government and the regulatory body or other relevant authority shall ensure that the protection strategy for the management of existing exposure situations, [...] is commensurate with the radiation risks associated with the existing exposure situation; and that remedial actions or protective actions are expected to yield sufficient benefits to outweigh any detriments associated with taking them, including detriments in the form of radiation risks.
>
> The implementation of remedial actions (remediation) does not imply the elimination of all radioactivity or all traces of radioactive substances. The optimization process may lead to extensive remediation but not necessarily to the restoration of previous conditions." [8]

Such changes in policy open significant new opportunities for the phosphate industry in that the demands placed on remediation projects where the eventual goal is the return of land used previously for PG disposal to productive economic uses. But this will also require regulators first to adopt these new policies and procedures and for industry to show compliance in following them. In particular any new policy framework must invoke the principles of proportionality and affordability such that remediation procedures should not be expected to remove all radionuclides or indeed more than is necessary to achieve a safe outcome.

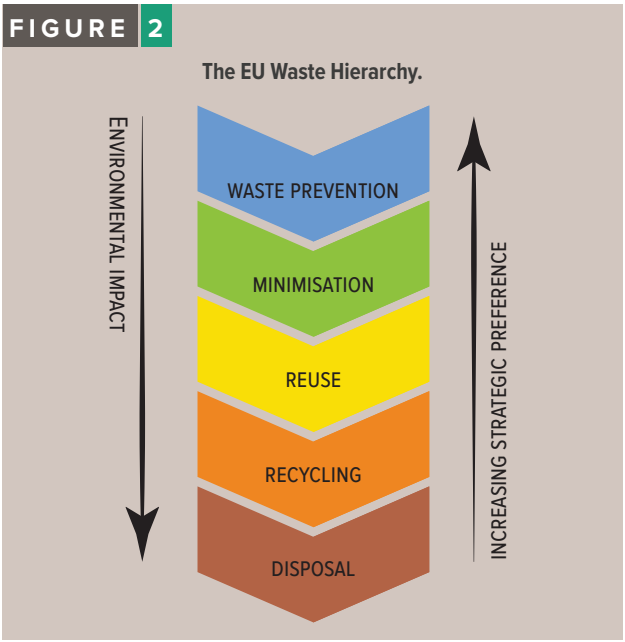
1.4 The waste hierarchy

The waste hierarchy concept is used increasingly worldwide in the prevention and management of waste, although there are variations as to what it means by region and country.

Figure 2 shows the European Union (EU) model which has been legally binding across the EU since 2008 [9]. To justify disposing of any material as a waste the four options – prevention, minimization, reuse and recycling (Figure 2) – must be reviewed first and shown to be technically impracticable or disproportionately expensive before disposal is authorised.

A fundamental requirement of the permitting process for the disposal of any material that is finally accepted for

FIGURE 2



5 ICRP: www.icrp.org/index.asp

designation as a "waste" starts with an Environmental Impact Assessment (EIA). It is part of the EIA to systematically evaluate beneficial use options which have to be rejected as unaffordable or inappropriate before resorting to disposal.

Currently, even the definition of waste is under review to align better with the objectives of the waste hierarchy. With the EU definition, the emphasis is on the human perspective - does the owner or user have any future intention for use of the material: if not, it is a waste. But this definition does not fit well with the waste hierarchy which rather emphasises the inherent properties of the waste itself. Hence the IAEA applies a more generalised criterion: is the material in question something for which a (beneficial) use may be "foreseen".

Individual intention is not taken into account whereas the inherent nature of the material – its characterisation and context – is. In this case, PG has very many foreseen uses and, therefore, is not *prima facie* a waste. Under the principles of sustainable development, the definition of waste is narrowed still further to exclude not just "foreseen", but also "foreseeable" uses, i.e. even those uses we are not yet aware of or which do not yet exist. This makes all PG *de facto* a resource.

In its meeting of April 2014 the United Nations Economic Commission for Europe (UNECE) Expert Working Group (EWG) on resource classification came into line with this trend to narrow considerably the definition of what should be classed as waste. Hence materials such as PG are termed secondary resources not wastes, and hence classed for resource reporting and evaluation purposes as such.

1.4.1 Changing policy and regulation

A number of major changes in policy both in regard to radiation protection and to wastes are reframing the context in which PG is both seen and used. These changes start with radiation protection itself, extend to the wider implications of the waste hierarchy, and end with the requirement to align all policies with those of sustainable development. All these factors underlie the IAEA phosphate industry Safety Report.

1.5 IAEA Phosphate Industry Safety Report

The crux of concern about PG use historically has been its radioactivity. Naturally occurring radioactive materials (NORM) in phosphate deposits have been studied since the early 1900s when it was realized that such deposits could have a potential value as a source of uranium if the concentration was sufficiently large [10, 11]. Although PG contains radioactive elements they are in approximately the same quantities, and in the same range, as are found naturally in many of the world's soils. Nevertheless, concern about radioactivity has been a recurring issue. Hence addressing the issue of radioactivity was one of the primary objectives of the 2013 publication of the International Atomic Energy Agency (IAEA) the Phosphate Industry Safety Report [12].

1.5.1 Phosphogypsum and Naturally Occurring Radioactive Materials

The phosphate industry is a NORM industry according to the IAEA [13], the EU [14] and many national regulators. The mining of PR, processing it into intermediate and end products and its residues, and the handling and use of all these materials can give rise to workers and members of the public being exposed to radioactivity depending how the radioactivity is distributed between the co-products.

Naturally occurring radioactive materials are quite variable in concentrations and chemical forms. Geologic deposits are formed in different ways and some radionuclides are associated with certain minerals while others are not. For example, uranium is typically associated with sedimentary phosphate rock while thorium is not, but uranium is not so strongly associated with igneous phosphate rock. There are also locations across the globe where radium in the ground decays to radon gas that percolates up through the ground in higher concentrations than is typical of most areas.

1.5.1.1 Radioactive decay chains and radon

There are three dominant radioactive "decay chains" in nature. These are headed by radioactive (unstable) forms of elements that "decay" into different radioactive forms of elements until a final, stable form is reached. The three chains are headed by uranium-238 (^{238}U), thorium-232 (^{232}Th), and uranium-235 (^{235}U). In sedimentary phosphate deposits, the ^{238}U chain (Figure 3), known as the "uranium series" predominates.

Radium-226 (^{226}Ra) (Figure 4) is one of the decay "daughters" of the uranium series decay chain. Radium in the soil, decays to radon gas and has a half-life of about 1,600 years. Radon-222 (^{222}Rn) is the progeny from radium, with a half-life of 3.8 days. It is found in the soil and the air. The dose to the world-wide population due to naturally occurring radioactive materials is predominantly due to radon's progeny.

Radon is an inert gas in that it cannot combine chemically with other elements. Radon cannot damage cells in the body; however, being a radioactive gas, when it decays, it emits energy that can cause damage. Also, after it decays, it becomes a different element that is also radioactive and not inert, which emits radiation and decays to other radioactive elements. These "daughters" emit radioactive particles and can damage the lungs.

The uranium series contains 19 primary members including the parent ^{238}U and the final stable member lead-206 (^{206}Pb). There are four other members that occur by

FIGURE 3

The uranium-238 decay chain through radon.

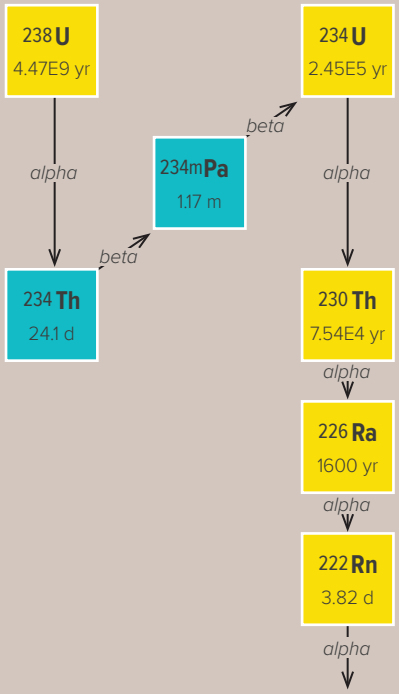
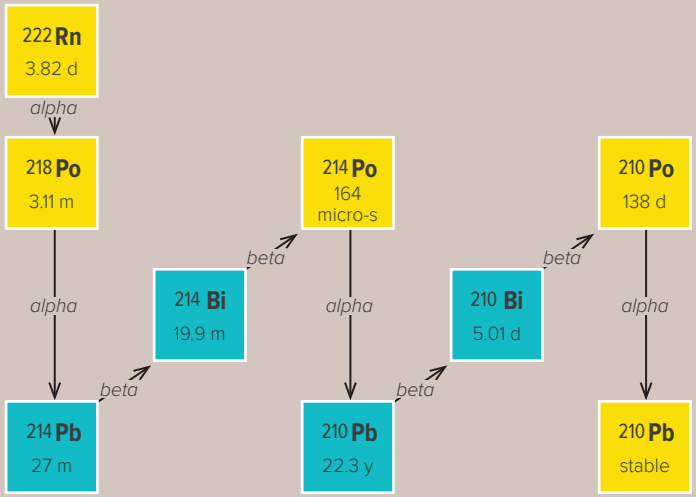


FIGURE 4

The decay chain after radon-222.



relatively infrequent alternative decays. Alternative decays occur because some modes of decay have different competing mechanisms that accomplish the same decrease in mass and energy in the effort to become more stable. The uranium decay series is shown in Figures 3 and 4 in simplified form, arbitrarily separated into the upper part through radon, and then radon and its daughters.

As the radioactive forms of elements, called radionuclides, decay they emit different types and energies of radiation. If the radiation impacts a person, either externally or internally, the energy of the radiation is absorbed in tissue and the person has received some amount of radiation dose. Radiation regulations are based on the principle that every amount of radiation dose, no matter how small, increases the recipient's risk of developing some form of cancer. If the dose is small, the risk will also be small and risks typical of the ones we accept in everyday life, e.g. the small risks of fatality from falls or fires in the home, are considered to be "safe." These risk levels are used in reference publications primarily the Basic Safety Standards [8], to calculate corresponding radiation doses and the concentrations of radioactive materials in consumer goods that could result in those doses and risks.

1.5.1.2 Other potential health risks from phosphoric acid

Products derived from phosphoric acid can give rise to risks to human health and the environment from non-radioactive constituents, notably fluorides and heavy metals such as arsenic, cadmium, lead, and mercury. These risks can make radiological considerations of secondary importance. In view of the very large amounts of PG, which may be stored in above-ground engineered containments, often for indefinite periods, or may be disposed of in surface water bodies such as estuaries and the sea, such concerns may apply equally to PG and to PG-based products.

1.5.2 Findings of the IAEA Safety Report

The IAEA Safety Report [12] signaled the removal of the primary obstacle to the definition and use of PG as a co-product, namely concern about its radioactivity and as a consequence the definition by some regulators that PG was a hazardous waste. The IAEA concludes that PG as a co-product presents no radiological grounds for preventing its beneficial use.

> "A particular example in this regard is phosphogypsum, a co-product of phosphoric acid production that, because of the very large amounts involved, is stored in aboveground engineered containments known as 'stacks', often for indefinite periods, or is sometimes disposed of in surface water bodies such as estuaries and the sea. Best practices established for the management of non-radiological risks to humans and to the environment have generally proven

to be effective in minimizing any risks arising from the residual radioactivity content. Nevertheless, the radioactivity aspect continues to arouse public concern in some countries." [12]

More widely, the indefinite disposal of PG in stacks is called into question:

"The storage of phosphogypsum in stacks, irrespective of its radioactivity content, creates potential environmental and physical safety hazards and therefore needs to be controlled by the relevant authorities in a consistent and harmonized manner. Similar controls are necessary when a stack is undergoing closure. Evidence shows that, with such controls in place, there is no necessity for additional regulation for purely radiological purposes. Future liabilities associated with the continued presence of large phosphogypsum stacks place a considerable burden on future generations. This, together with the increasing rate of phosphogypsum production, provides a very compelling reason for creating a regulatory environment that is conducive to identifying and promoting further ways for safely using phosphogypsum as a product rather than having to manage it as a waste.

For the foreseeable future the discharge of phosphogypsum to water bodies is largely being phased out, but is likely to continue in some countries. Experience has shown that, to ensure acceptable levels of risk to humans and the environment, such discharges would need to be regulated as part of an authorized practice on the basis of a situation-specific risk assessment.

All evidence suggests that the [radiation] doses received as a result of the use of phosphogypsum in agriculture, road construction, in the marine environment, and in landfill facilities are sufficiently low that no restrictions on such uses are necessary. The use of phosphogypsum in structural panels for the construction of a house could, in extreme circumstances, result in the occupant receiving an annual effective dose exceeding 1 mSv. Therefore, it would be prudent for the relevant authority to ensure that an appropriate situation-specific risk assessment is carried out[6] in order to determine whether any restrictions on this particular use of phosphogypsum are needed. For all other uses of phosphogypsum in home construction, including its use in cement, bricks, plasterboard and tiles, the annual effective dose received by the occupant is unlikely to exceed 1 mSv and restrictions on such use would appear to be unnecessary." [12]

As indicated in the introductory comments to this Report, regulators are gradually taking into account these conclusions in rewriting their regulatory requirements for PG, but the process will take time to work its way fully into everyday operations.

1.5.3 Evidence-based approach

Recognising that PG varies quite considerably in character from producer to producer based on the rock being processed, the IAEA Safety Report sets out four overall findings in regard to the material in use which may be summarised as follows:

1. Phosphoric acid (PA) and phosphogypsum (PG) are co-products of wet process phosphoric acid (PA) production
2. There is no radiological objection to use of PG; some uses may merit some restrictions depending on the precise characterisation of the PG under consideration
3. Use is environmentally preferable to stacking or disposal to sea
4. Regulators should be encouraged to promote beneficial uses as alternatives to disposal, in line with the waste hierarchy.

In classing PG as a co-product and not a waste IAEA took an evidence-based approach to the material based on published literature and case studies from across the world. It encouraged PG use as a better option environmentally than disposal to land or to sea, and importantly it also reflected a change of attitude on the part of policy-makers and the public that nothing should be classed as a waste before all potential beneficial uses of that material have first been considered. Such has been the impact of the IAEA Safety Report that some regulators are now introducing minimum use targets for PG rather than the previous default practice of mandatory disposal in stacks. The leading example is from China which as from 2015 requires 20% usage of PG and from 2025 the figure rises to 30%. In practice, some Chinese producers are already ahead of this target, Kailin having reached 100%.

1.5.4 Sustainability and the Triple Bottom Line

In the aftermath of the publication in 1987 of *Our Common Future*, also known as the Brundtland Report [15] there was an increasing recognition that the success of commercial enterprises could not be judged solely by their financial return. Taking these factors together John Elkington coined the term “Triple Bottom Line” [16] to reflect a responsible approach from corporations and business leaders to meeting a wide range of needs of the stakeholders in their affairs. In terms of the Triple Bottom Line (TBL) a company’s achievement is judged by a balanced combination of its financial, social and environmental performance.

6 A dose of 1 mSv corresponds to a risk of stochastic effects, both cancer and heritable, of 5.7 in 100,000 for the population as a whole and 4.2 in 100,000 for adults only (ICRP 103) [158].

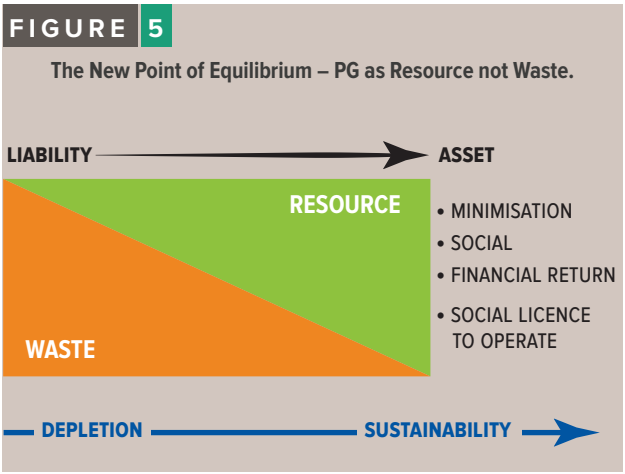
In commercial terms, every year the phosphate industry mines vast quantities of PR, makes more than 40 mt of phosphoric acid and generates more than 200 mt of PG co-product. In social terms, the industry is a major global employer, and in some markets one of, if not, the main economic pillars of the economy. It is an essential service provider to farmers and to the wider global population through its role in supplying phosphatic fertilizers for food production. Environmentally the disposal of PG to the sea or to the land is not consistent with the objectives of sustainability and resource conservation. Hence in this publication the beneficial use of PG is shown to be an essential and integral part of the wider strategy of meeting food, energy and water security goals, and of minimising waste and maximising recycling.

Phosphate fertilizers produced by the phosphate industry are at the centre of meeting the fundamental need for food by the present and increasing global population while the many and varied uses of co-product PG will play an increasing role in agriculture, construction and construction materials. These together will contribute to food security, and energy and water use efficiency, and provide employment. The more economic growth and stability worldwide is dependent on meeting the fundamental needs of food, energy and water security, together with the imperative of providing employment for an increasing number of people by 2050, the more deeply the global fertilizer industry and the phosphate-producing community in particular will necessarily be engaged in this undertaking. A measure of the extent to which the industry is willing to rise to these challenges may be taken from the welcome its leadership accorded the agreement in September 2015 by the United Nations to the Sustainable Development Goals[7] [17].

As many in the phosphate industry have already realized, it will have, in part, to reinvent itself starting with its range of products. In the past decade there has been a significant effort to find what the economic Nobel Prize winner John Nash called a "new point of equilibrium" [18,19], namely one that retains profitability while being significantly more inclusive of stakeholder interests. This has led to a significant growth in measures to demonstrate corporate social responsibility, a commitment to sustainability and the social licence to operate.

1.5.5 The new NORM equilibrium

The trend to use PG, whether for commercial or regulatory reasons, is perhaps the most obvious sign that a new point of equilibrium is being established in line with the IAEA co-product classification of PG. For the new equilibrium to be sustainable PG must be accepted as a safe, beneficial resource, by all three major stakeholders, the producers themselves, their customers and the regulators. Achieving such acceptance holds out major prospective financial and environmental benefits, turning waste into resource and hence liability into asset (Figure 5).



1. What has hitherto been seen in many markets as a waste can be reclassified as a resource; hence on the balance sheet it moves from the liability to the asset column.
2. In some markets PG can now be sold at whatever price the market will accept; in others finding acceptable solutions for use may not be market-driven.
3. The major negative externality of indefinitely stacking PG can be removed and value can be recovered from the sunk cost whether as product or recovered land, or both. Using Full Cost Accounting principles (below)) [20]. In many markets life-time disposal cost is an estimated US$ 25-35/t sunk cost. Such costs are influenced by factors such as regulatory requirements for managing the acidic pore water contained in the stacks. These will vary from market to market. Overall, when it is considered that some 4 billion tonnes of PG are currently stored in stacks worldwide, the capital drain on an already capital-intensive industry risks being too large to be sustained.
4. A key outcome from use of PG is that large areas of land that have been consumed by stacking PG, typically of potentially very high value, can be restored to productive use. (9.3, 9.5, 9.6).
5. Managing PG as a co-product with marketable uses reinforces the case for ceasing marine discharge on economic as well as environmental grounds.

7 United Nations Sustainable Development Goals: www.un.org/sustainabledevelopment/sustainable-development-goals/

1.5.6 Full Cost Accounting

Any sustainable solution to PG usage must take financial considerations into account. This Report is global in nature and hence is not intended to provide a detailed techno-economic feasibility analysis of specific PG types in specific markets. That task should be done by individual operators or by individual policy-makers and regulatory bodies prior to deciding which uses to prioritise. It is however, clear that the choice of the financial accounting model on which costs analysis should be conducted is a critically important one. Of the options considered, Full Cost Accounting (FCA) [21] is significantly the best suited for identifying and evaluating cost factors and drivers. It is perhaps doubly appropriate because one of its chief advocates is the United States Environmental Protection Agency (USEPA) which based its FCA Manual on the principles of the waste hierarchy.

One of the advantages of the FCA approach is that the emphasis is on preventing legacy sites that require post-closure remediation, with all the associated social, environmental and economic costs. Hence the cost structures identified FCA also include potential revenue streams under "any sale of by-products". It is increasingly clear that social factors – the so-called social licence to operate (SLO) [165] – are increasingly critical in the success or failure of any mining and processing project, especially in regard to wastes and it is striking that FCA pays full attention to this aspect, though not explicitly as an SLO issue.

1.5.7 Core principles of NORM waste management

Against the background of a general shift worldwide in regulatory approaches to NORM industries, many countries have developed a new NORM strategy within the context of a changing framework for radiation protection as set by the revised Basic Safety Standards (2014). Against that background, the fundamental principles according to which the new NORM strategy [22] have been developed by countries such as the UK and regions such as the EU is that any management regime applied to NORM must be:

- reasonable,
- appropriate,

TABLE 1

Full cost accounting, cost categories and pathways.

Cost category	Sub-category	Sub-category	Sub-category	Sub-category
Up-front	Public education and outreach (stakeholder engagement)	Land acquisition	Permitting	Building construction and modification
Operating	Normal (business as usual): · Operation and maintenance (O&M) · Capital (CAPEX) · Debt service	Unexpected	Revenue generation from recycling/reuse Sale of by-products	
Back-end (End of Life EoL)	Site closure (planned)	Building/equipment decommissioning	Post-closure care and maintenance	Social costs: · employee benefits eg healthcare and retirement
Remediation – brownfield or abandoned sites	Investigation, containment and clean-up of wastes and effluents	Post remediation care and maintenance	Fines or regulatory interventions	
Contingent	Remediation of legacy sites (past, undocumented sites or future sites)	Legacy liabilities: · personal injury or illness · property damage – negative impact on land and property values · damage to/contamination of primary or secondary resources		
Environmental	Environmental degradation	Avoidable need to use (waste) primary resources	Reputational damage to operator	
Social	Broken social licence to operate · negative impact on quality of life · heightened risk of illness or stress	Reduced property and land values	Planning/development blight	Aesthetic degradation

- proportionate,
- affordable, and
- sustainable [22].

In this list of five attributes, it is assumed that to be sustainable a practice must also be safe, but safety aspects are dealt with separately in a mandatory Safety Case, which derives from an equally mandatory Environmental and Social Impact Assessment (ESIA). These principles define, for example, the 2014 UK NORM Waste Management Strategy [22]. The UK position reflects a growing consensus both in the EU and beyond as to how NORM industries should in future be regulated. A key role is assigned in the strategy to finding commercially sustainable, market-based solutions for turning NORM "wastes" into commercially useful resources.

The UK NORM Strategy, in line with the EU Waste Hierarchy, also does not use the term waste as a first resort. Rather it uses the term "arisings" to encompass all materials not immediately classed by the operator as a primary material. IAEA uses a similar strategy by referring variously to co-products, by-products and residues as terms to be applied to materials other than the primary mineral target in preference to referring to a material as a waste [22].

1.6 Phosphogypsum management under the waste hierarchy

Historically since 1989 the management of PG, as reflected also in the majority of stack design and operations worldwide, has been with the end intention of permanent disposal. There have however always been exceptions. For example, Prayon in Belgium has seen itself primarily as a PG producer with phosphoric acid as a by-product and its PG has gone straight to wallboard production, eliminating the need for long-term stacking, while Omnia, South Africa is operating a new facility (2015) where the PG goes straight to the cement producer. Some PG produced in India likewise goes straight from the filter to the cement works.

As patterns of use in recent years have accelerated, stacking has become increasingly temporary in nature, often only to ensure the continuity of feedstock required while marine discharge has been curtailed based on recognition of potential ecological issues. In Brazil fresh PG is taken directly from the stack for use; in the Philippines PG is first limed to neutralise the acidic water in the PG and is then taken for cement production. Wengfu Group has redesigned its phosphoric acid facilities to take PG directly into the production process for construction materials. In general the industry faces a strategic management challenge for PG, especially in countries where the volumes produced far exceed national demand, to design and operate stacks in a manner that both protects the environment and promotes use, whether short- or long-term. Meeting this challenge is only just beginning.

When applied to PG, the primary focus of the waste hierarchy is on level 3 (Figure 2), reuse (as is, with no additional treatment) and level 4, recycling:

1. REUSE: Use as is in:
   - agriculture,
   - building and road construction materials,
   - embankments and sea defences,
   - filler for fertilizer that supplies Ca and S,
   - mine restoration.
2. RECYCLING as a resource in:
   - ammonium sulphate production,
   - sulphur recovery,
   - alpha gypsum (high strength),
   - calcium carbonate.

For producers where there is insufficient immediate market demand for PG, a sustainable approach might be to reverse engineer a pathway to a solution by starting from the desired outcomes which may be combinations of social, economic, environmental issues, and work back to the problem.

Hence sustainability may be the key determinant of how to approach the current situation, assuming:

1. Full characterisation of materials to confirm predicted low-level of radionuclide activity and confirm the consistency of the PG in the stack;
2. Use of a graded approach to mapping and sampling by stack for potential uses (Figure 8);
3. Environmental and social impact assessment;
4. Risk and exposure pathway assessment and resultant Safety Case (including As Low as Reasonably Achievable (ALARA)/justification/optimisation/dose limits);
5. Beneficial uses options (including proportionality) based on waste hierarchy principles;
6. Long-term storage options (future proofing);
7. Ongoing monitoring and assessment (intensity and strategy to be determined following mapping and characterisation).

1.6.1 Sustainability and the application of Green Chemistry and Green Engineering principles

Sustainable development focuses strongly on the concept of meeting needs both present and future, a process which entails careful stewardship of resource and the natural environment as a whole. In consequence, to the extent possible it is now regarded as good practice to substitute where possible secondary for primary resources and to regard as waste only those things for which no further use is foreseen (or even foreseeable).

The role of PG, as a co-product rather than as a waste is being reinvigorated within wider sustainability

objectives. These include those associated with the principles of green chemistry and green engineering [23], the progressive reduction of greenhouse gas emissions, the pursuit of low or zero carbon energy resources and the conservation of economically critical mineral resources. For some years PG has been an item of commerce in many countries, with a well-established market value in a variety of applications, especially in agriculture and construction, both of which have the capacity to consume large amounts of PG [20].

1.6.2 Implications of a 100% solution

There are 2 ways to get to 100% elimination of the risk of legacy wastes from PG – 1. Use and 2. Prevention. Use is the focus of this Report because prevention implies the end to the wet process of production which in the current market situation seems highly unlikely. But it is possible to envisage a hybrid where some producers change their flowsheets, e.g. to return to single-super phosphate (SSP) or nitro-phosphate fertilizers (Yara process) and others adopt new or updated technologies such as the "Improved Hard process" (IHP).

In this sense, the use pathway is one of "optimisation" meaning that the end point optimises a solution that uses 100% of the volume and returns as much value as possible to the producer and to stakeholders resulting in no legacy waste. So for example the stakeholder will benefit from the complete elimination of the needs for indefinite stacking and any land used in the past for storage can be progressively returned to other uses. This outcome has been achieved by various means in Sfax, Tunisia (remediation), the Western United States (use), North Carolina (return to the mine), Canada (remediation), China (return to mine and use combined), and Brazil (use). The only known producer to be currently at 100% use i.e. in equilibrium between production and consumption is Kailin, with a mix of mine restoration (40%), agriculture (30%) and construction (30%) as the means to that end.

The obvious drawback of putting PG back in a mine is that the valuable sulphur content is lost, together with small quantities of phosphorus. Thus sulphur recovery would ideally be best done before any remaining material is returned to the mine.

Where PG is rich in P_2O_5 the use of that material is sometimes problematic in construction materials because a P_2O_5 content higher than 0.5% may cause difficulties for the manufacturing process of products such as cement and building blocks. For road construction a slightly elevated P_2O_5 value may not matter, but the loss of the phosphate, which serves no useful purpose for the road, makes the practice questionable because the P has much higher value-add when applied to the soil.

The alternate pathway in technological terms would be termed "disruptive" i.e. a new dominant production technology displaces the current one. Given the long life of a given industry site and the overall capital intensity of the industry any disruption is highly unlikely to change the industry over night. Nevertheless, over time there will be technical innovation both of the optimisation kind but also completely new in nature, which will be accelerated if there are regulatory and fiscal incentives to promote them. Either way a 100% solution can be envisaged for PG use.

2 Phosphogypsum characterisation and availability

KEY POINTS

- All PG is suited for safe, beneficial uses.
- Characterisation will determine the most suitable, safest and affordable uses. Local and regional factors will determine selection criteria.
- Accurate characterisation, as shown in a Materials Safety Data Sheet (MSDS), will provide the customer or end user with reassurance about the traceability of all resource content.

2.1 Characterisation

Fluorapatite is the predominant phosphate mineral in PR worldwide, and when it is digested, most commonly with sulphuric acid, to make phosphoric acid the primary chemical reaction can be expressed as:

$$2Ca_5F(PO_4)_3 + 10H_2SO_4 + 10nH_2O \rightarrow 10CaSO_4 \cdot nH_2O + 6H_3PO_4 + 2HF$$

Depending on the value of n, the process is defined as the dihydrate (n=2) (DH) process, the hemihydrate (n=½) (HD) process, or anhydrate (n=0) process. The term $CaSO_4 \cdot nH_2O$ in the equation is generally referred to as PG. The DH process is used most widely while the HD process is becoming popular, especially for new plants. Currently the anhydrate process is not used on a commercial scale. Some hybrid processes combine some steps of the DH and HD processes.

Phosphate rock is processed by being fed into a reactor containing a circulating slurry of phosphoric acid and PG. Then a mixture of recycled phosphoric acid and sulphuric acid is added. This produces both phosphoric acid and insoluble calcium sulphate - PG. The phosphoric acid, which contains almost all the uranium from the source rock, is typically separated from the PG by filtration, and producing one tonne of P_2O_5 (as acid) produces approximately five tonnes of PG.

The reaction between phosphate rock and sulphuric acid is self-limiting. In the reactor, or attack tank, PG forms on the surface of particles of rock, inhibiting the reaction. This problem is kept to a minimum first by controlling the particle coarseness, and subsequently keeping the rock in contact with recirculated phosphoric acid to convert it as far as possible to soluble monocalcium phosphate. The PG is precipitated by the addition of sulphuric acid. In greater detail, the chemistry is as follows:

$$Ca_3(PO_4)_2 + 4H_3PO_4 \leftrightarrows 3Ca(H_2PO_4)_2$$

$$3Ca(H_2PO_4)_2 + 3H_2SO_4 \leftrightarrows 3CaSO_4 + 6H_3PO_4$$

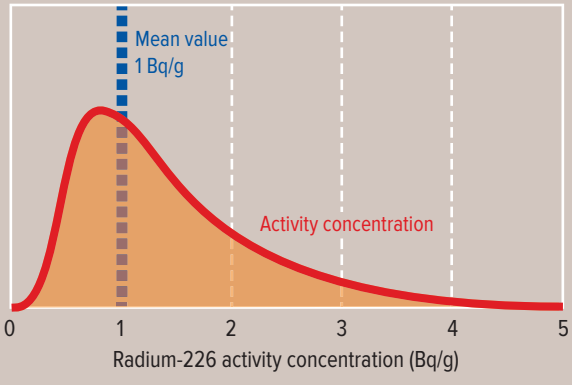
2.1.1 Radioactivity in phosphogypsum

Phosphate rock contains Naturally Occurring Radioactive Materials (NORM) notably radium (Ra) and uranium (U) in the pervasive forms ^{226}Ra and ^{238}U respectively.

When phosphate rock is digested in sulphuric acid during the wet process, the majority of the U remains with the acid while the Ra goes to the PG as radium sulphate and the levels of radioactivity are not increased. Almost all the ^{226}Ra and a small amount of U (5-10 ppm) in the U^{4+} ionic form migrates to the PG, with mean activity concentrations of at or below 1 Bq/g (Figure 6). This data is based on the average radioactivity in the global phosphate reserves, hence the actual data from which the curve in Figure 6 derives does not apply to any particular

FIGURE 6

Estimated worldwide distribution of ^{226}Ra activity concentrations in phosphogypsum.



deposit or site. Consequently, while values of 5 to 10 ppm U are typical they may be an order of magnitude less for some sites. For example, in central Florida, which produces PG at ~ 1 Bq/g ^{226}Ra, the U in the feed rock is some 140 ppm while the U content in the PG may be as low as 0.5 ppm (Table 2).

In reality by tonnage almost all commercially produced PG falls short of the 1 Bq/g threshold, much of it significantly so. 1 Bq/g is used as a threshold value because it is the upper limit of naturally occuring radioactivity in soils – hence regulatory attention may be required above this level. The low levels of radioactivity typically found in PG are key factors in the policy shift towards designating PG as "out of scope" for regulatory purposes.

Trace quantities of U may be present for five main reasons:

1. Unreacted rock particles containing U^{4+} may become coated with PG;
2. Some substitution by U^{4+} occurs in the crystal lattice of PG;
3. Residual amounts of phosphoric acid, containing uranium phosphate, remain with the PG after filtration;
4. U^{4+} may adsorb on the surface of PG as UO_2HPO_4.

The residual phosphoric acid remaining with the PG may also contain a small amount of U as uranyl ions (UO_2^{2+}), while the substitution of Ca^{2+} ions by UO_2^{2+} may occur on the surface of the PG crystal lattice [24].

In the context of discussing Figure 6 it must be remembered that in nature due to the decay chain of ^{238}U (Figure 3) Ra is produced by U. When the phosphate rock digestion process works efficiently the activity concentrations of ^{238}U are mostly below 0.1 Bq/g, although occasional higher values may indicate a lower recovery of phosphate in the attack tank. The activity concentrations of ^{226}Ra and its progeny are generally in the range 0.2-3 Bq/g for material derived from sedimentary phosphate ore, but are much lower for PG derived from igneous ore, ranging from less than 0.01-0.7 Bq/g.

The worldwide distribution of ^{226}Ra activity concentrations, estimated by fitting a lognormal curve to the average radioactivity in the global phosphate reserves, is shown in Figure 6. The arithmetic mean of the distribution is 1 Bq/g, which is also the maximum value found in any of the world's soils although, in reality, 99% of the world's soils have much lower activity concentration levels, typically 30-40 mBq/g. The 1Bq/g value for PG is used both by the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR)[8] [25] and other bodies which follow UNSCEAR such as IAEA and ICRP for determining the level above which it is reasonable for a regulator to intervene because this is the upper limit of what is found in nature. Thus when the PG ^{226}Ra level is equivalent to or lower than that in the soil it is deemed by the regulator to be inherently safe because its use does not appreciably enhance the radioactivity of the soil nor is there any evidence that such values harm people or the environment. Equally, it is deemed legally absurd to try to regulate a naturally occurring material to require it to be present in levels lower than those which are found in their natural state. It is also notoriously difficult to calculate what the risk might be to workers or the public simply from these activity concentration figures. Other key factors such as risk pathways, time and source-receptor geometries are also involved. For this reason IAEA has an extended discussion on how such risk figures are eventually arrived at in the Basic Safety Standards, notably in those sections dealing with the so-called "graded approach" [8].

According to the graded approach to NORM regulation up to 1 Bq/g ^{226}Ra is an amount of activity in a material that is expected to be safe for many applications. Once the level exceeds 1 Bq/g the materials of interest should be evaluated. In some cases it may even be advisable to take lower values into consideration – the number is a guide and not a limit set in stone. Many factors go into the decision to apply or not apply regulatory controls and, if so, to what extent. A person who is exposed to radioactive material will incur a radiation dose and there will be some associated risk of fatal cancer. Limits are derived to manage risk to an acceptable level typical of risks faced in commonly accepted activities. The dose corresponding to that risk level is generally accepted as 1 millisievert (mSv) per year. Such a dose yields a conservatively estimated mortality risk of 5.7 in 100,000 for the general population. In practice, the dose depends on the exposure pathway. For example, if PG is used in construction materials, there can be direct irradiation from those materials, or emanation of radon gas and its decay products into the air breathed in a building, which are inhaled and incorporated into the body. If PG is used in agriculture, there could be a soil ➔ grass ➔ cow ➔ milk ingestion pathway. There are many possible exposure pathways. IAEA has considered these and back-calculated to an activity concentration of 1 Bq/g ^{226}Ra that will be safe in most cases.

Activity concentrations vary between stacks and even between different levels in stacks, depending on factors such as age and the source of the PR. Some of the radionuclides in PG could be removed while still in slurry form, while attempts have been made to remove them using a cyclone because there are indications that a higher proportion of the radioactivity is found in the fine fraction of PG. The sulphates of radium and calcium differ in ionic radius, crystalline structure and solubility (calcium sulphate is slightly soluble and radium sulphate is highly insoluble) [26] and separation might be possible during crystallization. Any large-scale separation of radium radionuclides from PG is dependent on economic factors, notably the cost.

8 UNSCEAR mandate: www.unscear.org/unscear/en/about_us/mandate.html

2.2 Amounts of phosphogypsum produced

2.2.1 Amounts stacked

It has been estimated (Table 3) that, by 2006, a total of 2.6-3.7 billion t PG had been accumulated in stacks worldwide. This represents 44-62% of the total amount of PG produced by then, and of this total about 1.7 billion t was stacked in the USA (mostly in Florida).

Table 3 also gives an overview of what has happened to all the PG produced up to 2006, which includes two estimates for those countries where there is uncertainty about the amounts of PG discharged, used or abandoned relative to the amount retained in stacks.

Phosphogypsum is currently being added to stacks at an annual rate of about 40 mt in the USA and 120 mt elsewhere. The balance is discharged to sea. Although the trend is to find beneficial uses for PG, with volumes now approaching 35 mt/year (Table 2), at the current net rate of stacking to land the total amount stored in stacks could nearly double to 7-8 billion t by 2040. Further reductions in the discharge of PG to water bodies, as a result of acceptance and compliance with international conventions and treaties, corresponding to a better understanding of marine impacts, will add significantly to the volume being stored on land.

The discharge of PG to surface water bodies has significantly reduced as a percentage of production as a result of progressive changes in environmental regulation, notably within the European Union in the 1990s and under the London Protocol of the International Maritime Organisation since 1996 [27]. This has resulted in more PG being stored on land, and PG stacks becoming long-term disposal facilities rather than short-term holding piles [28]. However, the volume of PG discharged to water bodies still remains at some 50 mt/year because of the increase in overall production.

TABLE 2

Global PG production and use (2015) – IFA member companies.

	WWPPA production (2013) (t)	Potentiel PG co-product (t)	Ratio
Global IFA PIT production statistics (59 companies/ associations, 33 countries) *PG generation is estimated, not collected by PIT service*	42,500,000	216,750,000	5.1
Global IFA Tech & SHE PG Data Collection (38 companies/ associations, 21 countries)*	31,586,124	161,344,849	5.1
Including China	17,000,000	88,400,000	5.2
Including India	1,400,000	7,250,000	5.1
Including USA	7,039,576	37,786,195	5.4

PG regulated number of countries (A)	PG disposed or stacked (B,C) (t)	Agriculture usage (D) (t)	Construction usage (E) (t)	Other usage (F) (t)
19 yes / 2 no	109,098,567	2,193,993	11,603,631	15,228,502
		Total *recorded* usage (2013): **29 026 126 t** (G)		

*Background information:
· Significance of IFA PG data vs production statistics: 74% (Table 1)
· Some companies/associations did not provide B-F details (Table 2)
· 21 member companies and 12 countries did not participate

(A) PG has been regulated to various degrees arould the world
(B) At least 3 out of 38 companies are disposing PG
(C) At least 27 out of 38 companies are stacking PG
(D) At least 11 out of 38 companies are agricultural PG users
(E) At east 12 out of 38 companies are construction PG users
(F) At least 3 out of 38 companies are other PG users
(G) *Anecdotal feedback*, however, suggests that total usage of PG is likely to be at least 35,000,000 t/year worldwide

Source: Global IFA PG data collection (February 2015).

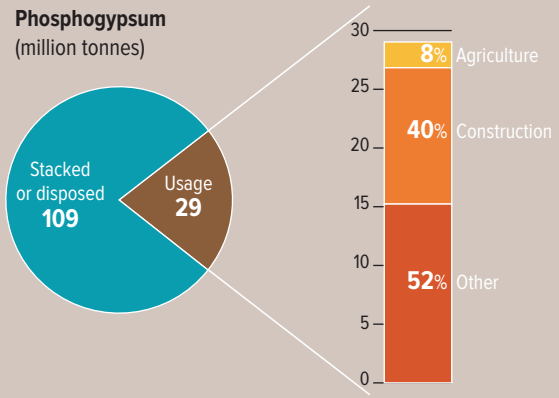


TABLE 3

Cumulative amounts of phosphogypsum up to 2006[9].

	'Minimum stockpile' estimate		'Maximum stockpile' estimate	
	Amount (billion t)	Proportion of total	Amount (billion t)	Proportion of total
Stockpiled in stacks				
USA	1.7	29%	1.7	29%
Other countries	0.9	16%	2.0	34%
Total	2.6	44%	3.7	62%
Discharged to water bodies				
USA	0.5	8%	0.5	8%
Other countries	2.5	42%	1.3	21%
Total	3.0	50%	1.8	29%
Used or abandoned	0.3	5%	0.5	8%
Total amount produced	6.0	100%	6.0	100%

2.2.2 Phosphogypsum production and use

As shown in Table 2 the relative balance between disposal and use is now shifting in favour of use, with an emphasis on mine restoration, agriculture and construction.

2.2.3 Phosphogypsum discharge to sea

Of the PG currently produced worldwide, some 20-30% is still discharged to sea as slurry consisting of PG suspended in salt water. Salt water is taken directly to the filter from the sea and this separates the PG crystals from the acid. The slurry of PG crystals suspended in seawater is then returned to the sea. The practice has been declining progressively and is now limited to a small number of producers, but the volumes discharged to water bodies remain significant.

2.2.4 Range of uses for phosphogypsum

In 2013 the IAEA Safety Report focused on a range of uses of PG, the largest number being in agriculture and construction. Once accurately characterized, in particular according to its content of radionuclides and heavy metals, as shown in Figure 7, all types of PG can be assigned to one of three basic categories of use. The degree of regulation and concomitant restrictions on the types of use deemed safe, increases progressively in line with the so-called "graded" approach to risk management as required by the Basic Safety Standards [8]. In parallel, any such use will depend on public opinion being assured that there is no risk from the radionuclides and heavy metals.

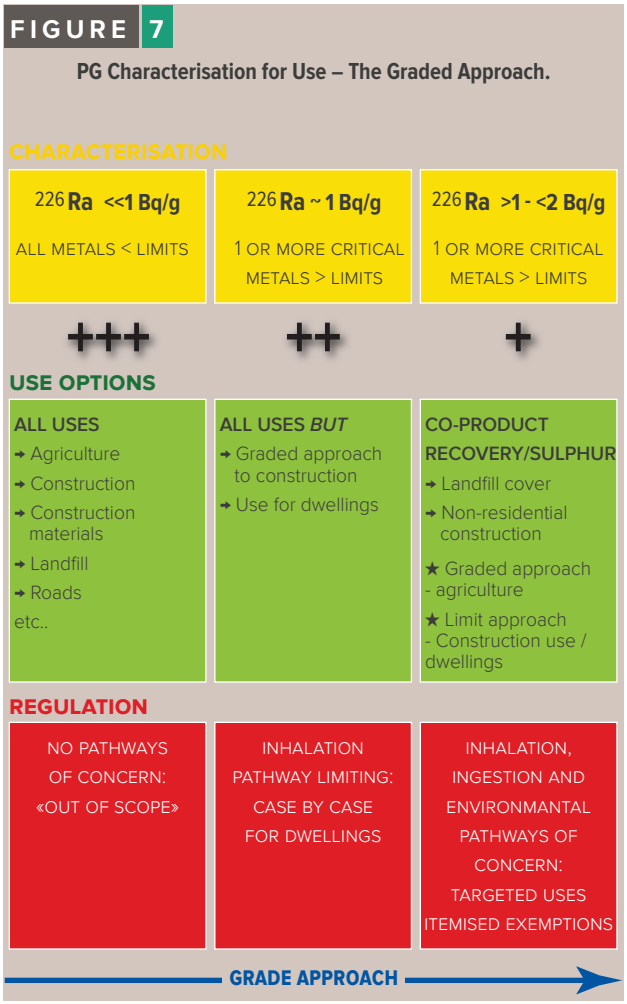
Following the graded approach as set out in Figure 7, it is possible to align the nature of the PG to hand to the best use for that PG given local social, environmental and economic requirements. This supposes that any particular PG used would a) have a full Materials Safety Data Sheet (MSDS) containing its specific characterisation including any regulatory requirements associated with use (the EU licence for PG which is based on a specific characterisation[10]) and b) its traceability showing where all the source materials, in particular where the PR and came from.

2.2.5 Mine restoration as use

The PCS Aurora phosphate facility in North Carolina, USA has long practised a USEPA-approved method of back-filling PG into its mine [29]. This type of procedure

FIGURE 7

PG Characterisation for Use – The Graded Approach.



9 The date 2006 is chosen because this was the first year in which such figures were estimated as part of the "Stack Free by '53: Beneficial Uses of Phosphogypsum" project. The figures were published in [20], and in turn fed directly into the IAEA Safety Report. The figures are referenced in the Safety Report as shown.

10 EU calcium sulphate licence

is increasingly seen not as disposal but as a high volume use, in this case for mine restoration[11].

In the PCS process, PG is mixed with the phosphatic clay suspension arising from the upstream beneficiation process. The mixture comprises approximately 3 parts PG to 1 part clay. The mixture dewaters and consolidates over approximately 1 year after which time it is possible to establish grass, trees and other vegetation. However, after PG and clay are mixed it would be difficult and very expensive to recover any of the useful components in the PG for future use and recycling.

USEPA identifies two critical factors in being able to use this method:

1. Proximity to the mine (to minimise transportation costs);
2. Calcium carbonate content must be at a level sufficient to neutralise the acid content of the PG (typically pH 2 in production).

Two benefits identified by USEPA in line with FCA are i) aesthetic, in that the "lunar landcape" effect from mining is fully remediated such that the mine site looks restored to its original state and ii) environmental in that fugitive dust and erosion problems are obviated.

2.3 Stepwise progression towards value-added use – example India

While there is no single pathway operators can follow towards PG use, there are common features across those countries showing a marked trend towards use. These features are well illustrated in India. Significantly, the first move is typically made by the regulator by removing regulatory obstacles to use. That in turn stimulates industry to develop products and markets.

2.3.1 Regulatory context

Following the exclusion of PG from the list of hazardous wastes, the Atomic Energy Regulatory Board (AERB) of India in a letter dated 20 March, 2009 clarified that it sees PG as a by-product not a waste. It confirms:

1. there is no restriction for use of PG in agriculture applications;
2. that AERB approval would not be required for companies wishing to sell PG for processing as building and construction materials, e.g. for panels and blocks if the activity concentration of ^{226}Ra is at 1 Bq/g or lower, which is the value set by IAEA;
3. If the ^{226}Ra in the PG exceeds this level it can be blended with other PG to reach the permitted level. For building blocks themselves the permitted level is set at 40 kBq/m^2.

Subsequent to the 2009 letter of authorisation the Central Pollution Control Board (CPCB) of India issued a Guideline for Management, Handling, Utilisation and Disposal of PhosphoGypsum generated in Phos. Acid plants (16 August, 2014) [30]. This document includes guidelines for PG use in seven different applications:

1. installing captive manufacturing unit for plaster/gypsum board;
2. use in plaster, blocks or gypsum board manufacturing industry;
3. cement manufacturing unit;
4. manufacture of ammonium sulphate;
5. recovery/manufacture sulphur/sulphuric acid;
6. reclamation of alkali soils and saline-alkali soils and use as fertilizer in agriculture
7. road construction.

2.3.2 Industry overview

In India there are 11 phosphoric acid plants with an annual capacity of around 1.4 mt/year P_2O_5 generating an estimated 7.1 mt/year PG (2014 figures)[12]. Of the total quantity of PG produced in 2013-14 (Table 4), some 55% was at one site with two large capacity phosphoric acid plants and no local cement producer to take the PG.

The situation is however easing as compared with the previous decades. In 2008, the Ministry of Environment, Forests & Climate Change, Government of India, categorized PG as a high-volume low-effect waste and excluded it from the list of hazardous waste under the Hazardous Wastes (Management, Handling & Transboundary Movement) Rules. Use in agriculture in particular is unrestricted. These Rules stipulate that separate guidelines for the management of PG should be prepared by the Central Pollution Control Board (CPCB). These Guidelines were published in August 2014.

TABLE 4

Phosphogypsum use in India 2011-2014.

Year	PG generation (mt)	Utilization of PG (mt)			
		Cement	Agriculture	Other	Total
2011-12	6.92	3.91	0.73	0.27	4.91
2012-13	6.07	3.58	0.67	0.19	4.44
2013-14	6.38	3.60	0.62	0.29	4.51

11 A forthcoming IAEA publication, the Comprehensive Extraction Policy Manual, recognizes this type of practice as a use secondary resources not as waste disposal, in line with zero waste objectives. In that context it advises careful characterisation of the materials use for eventual future recovery. The Manual is due for publication in 2016.

12 Data from the Fertiliser Association of India (FAI) as kindly supplied by IFFCO, September 2015.

2.3.3 Developing patterns of use, sector wide

The developing pattern and range of uses in India that has been stimulated by the change of regulatory framework is illustrated by recent company activity[13]:

1. **Paradeep**:
   PG Production – 2.724 mt PG (2012-13); 2.764 mt (2013-14)
   Annual sales - 0.33 mt for cement manufacture; 0.22 mt to agriculture
2. **PPL**:
   PG Production – 1.040 mt (2012-13); 1.136 mt (2013-14)
   Annual sales – 0.442 mt to cement/plaster of Paris manufacture; 0.580 and 0.580 mt, to fly-ash brick manufacturing units for export to Bangladesh and Nepal
3. **Coromandel**:
   PG Production - 0.553 mt (2012-13); 0.609 mt (2013-14)
4. **Gujarat State Fertiliser & Chemicals Ltd.**, Vadodara:
   PG Production – 0.37 mt (2012-13); 0.34 mt (2013-4)
   Annual use – 0.34 mt (2012-13), 0.32 mt (2013-14) for soil conditioning
5. **RCF:**
   PG Production – 0.14 mt (2012-13); 0.11 mt (2013-14)
   Annual sales – 0.18 mt (2012-13) and 0.12 mt (2013-14) for cement/Rapid Wall Panels/and putty
6. **TATA Chemicals Ltd.**, Haldia, WB:
   PG Production – 0.062 mt (2012-13)
   Annual sales – 0.12 mt (2012) for cement.

2.3.4 Commercial innovation

In addition to the general uses shown in 2.4.1, some specific new enterprises have also been created:

1. RCF has adopted Rapidwall technology for converting PG into load-bearing wall panels a low-cost, prefabricated walling product with broad construction applications. These are manufactured at a plant in Trombay using technology from M/s Rapidwall Building System, Pvt. Ltd, Australia. The plant annually produces 1.4 million m^2 of wall panels, 40,000 t of wall plaster and 6,000 t of wall putty, all from PG.
2. A similar project has been established in Kochi by the FACT-RCF Building Products Ltd (FRBL), a joint venture of FACT with RCF to recycle PG. The two FACT plants are in Cochin (~ 1800 TPD PG used) and Udyogamandal (~300 TPD PG used). The product has also received in-principle approval from the Building Material Technology Promotion Council (BMTPC) under the Ministry of Housing and Urban Poverty Alleviation.
3. PPL is manufacturing a PG based by-product, Zypmite, a NPK granulated fertiliser with added sulphur, zinc, boron, calcium and magnesium which helps improve soil fertility, and increases the uptake of plant nutrients and improves crop quality and yield. The plant has a 240 TPD capacity.
4. Coromandel is developing a Green Belt on an abandoned Gypsum Pond by using "Bio Remediation Technology" of TERI. This is an 18 acre plantation of about 18,000 trees (section 9.6. See also section 9.5 where a similar remediation project is under way in Canada led by Agrium.

2.4 Co-product strategies and comprehensive extraction

In the light of the IAEA Safety Report there is now increased attention to PG as a co-product and related interest in a comprehensive extraction approach to all materials of interest in phosphate ores. For some producers, such as Prayon, this is a belated recognition of an existing business model, for others it is causing a complete overhaul of the business model.

2.4.1 Prayon – phosphoric acid as co-product of phosphogypsum

Some producers have always worked to the principle of treating PA and PG as co-products. Prayon even goes further seeing PA as a by-product of its PG business [31].

Prayon has been producing phosphoric acid continuously at its main site of Engis, Belgium, since 1943 using a dihydrate process with a capacity of 35 mt per day P_2O_5. The PG produced was originally discharged into the nearby river where it settled and, twice a year it was removed (dredged) and stacked on land. In the mid-1960s, the capacity was increased to 150 mt per day P_2O_5, and with the increasing costs of dredging and transport, Prayon developed a new phosphoric acid production process which produced PG that could be sold because it is essentially dry and free of impurities. A global view of the plant and the site is shown in Figure 8.

2.4.1.1 Value-add co-products – construction materials, Belgium

Since the 1970s, Prayon has been collaborating with the German company Knauf to use PG in the plaster production, and in 1974, Knauf erected a stucco plaster plant adjacent to the PG curing area (Figure 8). In 2013, Prayon

13 Data from FAI India, courtesy IFFCO.

FIGURE 8

Global view of the Engis site.

FIGURE 9

Phosphogypsum being loaded on barges.

produced about 135,000 mt P_2O_5 and about 658,000 mt PG. The PG is sold mainly to produce stucco plaster, cement (as setting retarder) and fertilizers.

The merchant grade PG passes through a simple roller crusher to break lumps before screening and conveying to the plaster plant. Another conveyor feeds the screened PG to a loading station from where trucks or barges take it to cement factories and other users (Figure 9).

Depending on the year, between 80% and 90% of the total PG produced is sold. In 2013, 496,000 t were sold for construction, cement and plaster, and 14,000 t to the fertilizer industry. Unavoidably some of the gypsum produced is not sold because of:

1. Manufacturing process fluctuations (start-up, shut down, sulphate control...), the quality of the calcium sulphate does not always comply with customers specifications and around 10% of total gypsum is rejected (44,000 mt in 2013);
2. Limited demand in the gypsum market which cannot absorb the whole quantity of Prayon merchant-grade gypsum.

Unsold gypsum must be stacked locally on land or sent to an appropriate landfill (Section 10.)

2.4.2 Wengfu Group and Kailin – towards comprehensive extraction

The practice of comprehensive extraction starts from the premise that all materials are resources and hence can be turned into products. Figure 10, the product display room at Wengfu Group headquarters, shows some of the many co-products, including PG, that can be produced when PR is treated in the wet-acid process. These include U, REEs, Th, F, Cd and I.

As shown in Table 5 [12] (taken from IAEA Safety Report 78, Table 55) the typical concentrations of most trace elements in PG are significantly lower than those in the source rock. Small amounts of radioactive elements such as radium and uranium and of non-radioactive heavy metals such as arsenic, barium, cadmium, chromium, lead, mercury, selenium and silver, as well as phytotoxic fluoride and aluminium, are present in PG and its pore water [12]. Their concentrations vary widely, depending on the composition of the PR used as the feedstock and to a lesser extent on other factors such as differences in operation of the processing plant [32].

With some exceptions, the concentrations of cadmium and other heavy metals in PG derived from igneous PR tend to be lower than those in PG derived from sedimentary PR, while the concentrations of lanthanides (rare earth elements) tend to be higher. Heavy metals tend to concentrate in PG particles smaller than 20 µm [33, 34]. As shown in Table 5 in respect of cadmium, there is a degree of control the operator exercises over the

FIGURE 10

The Wengfu Group product display including PG construction products.

TABLE 5

Heavy metals and rare earth elements in phosphogypsum [12].

Element	Concentration (ppm)		
	Minimum	Maximum[a]	
Ti	26	470	
V	2	40	(190)
Cr	1,6	75	(594)
Mn	3,5	20	
Co	0,05	2,3	
Ni	1,7	250	
Cu	2	195	(508)
Zn	4	315	(351)
As	1,0	42	
Se	0,5	75	(249)
Sr	10	1118	(1606)
Y	2	156	
Zr	10	110[b]	(398)
Mo	1	16	
Ag	0,4	5	(73)
Cd	0.8[c]	40	
Ba	20	236	(810)
Hg	0,005	10	
Pb	0,5	16	(73)
Th	0,4	4	(75)
U	0,5	13,8	(19)
La	42	90	(419)
Ce	21	143	(425)
Nd	30	67	(352)
Sm	5	13	(60)
Eu	1,1	3	(15)
Yb	2,1	3,2	(21)
Lu	0,3	0,4	(2)

[a] Values in parentheses are maximum values reported for the fine fraction (<20 microns). [138,139]

[b] A value of 700 ppm was reported for PG of igneous origin. [48]

[c] It is reported that, by adapting the parameters of the sulphuric acid digestion process, the cadmium concentration in the PG can be reduced to less than 0.5 ppm, although this is offset by a higher cadmium concentration in the phosphoric acid. [85]

Source: IAEA Safety Report 78, Table 55.

extent to which the metal reports to the PG or stays with the acid, the range found in the PG being 0.8-40 ppm. As shown in Figure 8, given the considerable variability of PG, for each individual stack or production unit a separate characterisation study is required to make sure that heavy metals as well as radionuclides are accurately measured and their presence and relative quantity is factored into the choice of use, and the degree to which monitoring and surveillance of that use is warranted.

Knowledge of the concentration of the other components in PG can be important for defining technical solutions for the production of marketable product to be sold (Figure 9) and for the management and environmental policies when PG is stacked.

For the case studies that follow, please note that regulatory policies and limits vary significantly between jurisdictions. Consequently, an acceptable practice described in a case study from one region may be unacceptable in another. Carefully consider concentrations of radionuclides and metals in a specific PG source in comparison to policies and limits in the application region when evaluating options for PG use.

3 Phosphogypsum in agriculture

KEY POINTS

- Crop yields are being adversely affected by sulphur deficiency because of declining atmospheric inputs.
- The use of irrigation water with high salt levels has caused an accumulation of sodium and magnesium in many soils significantly reducing crop yield.
- PG offers very considerable advantages in dealing with this situation. It will be used at two different rates per unit surface area:
  1. At low rates to supply calcium and sulphur, especially to remedy any sulphur deficiency;
  2. At very large rates for soil remediation where soils are affected by excessive quantities of sodium or magnesium.

3.1 Background

Phosphogypsum is used in agriculture throughout the world [20] in a wide variety of climates and on a wide variety of soils and crops. It is applied to soils, for example, in Australia, Bangladesh, Brazil [48], Egypt, India, Kazakhstan [35] (Figure 15), Pakistan, Spain [36], the Syrian Arab Republic and the USA [37]. In other countries such as Tunisia and Jordan where regulatory constraints prevent large-scale use it has been intensively studied for use in agriculture. Its use contributes to soil fertility and crop production in various ways including:

1. Remediation of saline and sodic soils – Section 3.3;
2. Soil conditioning to prevent crusting leading to enhanced water infiltration and prevention of run-off;
3. Fertilization of soil for growing crops and pasture
4. Reduced sodium or aluminium toxicity in soil;
5. Increased supply of calcium and sulphur dissolved from the PG;
6. Increased ammonia retention by the soil;
7. Greater water use efficiency;
8. Reclamation of estuarine marsh to productive agricultural soil;
9. Affordability – Section 3.4.1.3 .

These uses, which are described in more detail in Sections 3.2.1 and 3.2.2., require very different amounts of PG, up to 1 t/ha to remedy sulphur (S) and calcium (Ca) deficiency and as much as 30-40 t/ha for soil remediation. Phosphogypsum was used extensively in US agriculture until the promulgation of the USEPA PG Rule in 1989 led to an almost total ban. Since that time use of PG and the many crops that benefitted by its application has been reviewed extensively [38], as for example in the increasingly significant application of PG to ameliorate subsoil acidity [39]. In particular in irrigated soils, acidity limits crop yield because roots cannot grow into acid soil to find water and nutrients [40]. Such is the extent of this problem that as of September 2015 it is listed as Goal 15 of the 17 UN Sustainable Development Goals [41].

There is significant potential to use PG with too high a concentration of P for use in construction (i.e. in excess of 0.5% P_2O_5 content) as an affordable fertilizer, for example by a deliberate increase the amount of P it contains to 2.5% or higher. Further value addition could be achieved by augmenting the product with nitrogen (N), potassium (K) and micronutrients, to make a general purpose NPK fertilizer. Where there is additional acid water in the PG, finely ground phosphate rock may be used as the additional P source. Trends in this direction in the industry are already discernible. Coromandel (India) for example markets a product it calls “phosphogypsum and sulphur pastilles”[14].

In Brazil PG is taken fresh from the production facility (Figure 19), in other markets it may be allowed to dewater and weather before use. For example, PG taken for agricultural use from the inactive portion of a stack in northern Florida, USA is first exposed to rainwater for about a year to displace the acidic water and the pH of the PG to increase above pH 5.

14 Coromandel PG and sulphur pastilles: www.murugappa.com/investors/coromandel/coromandel.htm

For some types of PG it may be necessary to remove other impurities such as cadmium if the levels it contains could be damaging to the soil or the crop grown. If the costs of removal become disproportionate, these PG types are likely to be better used in construction. In general, removing impurities can make the use of PG uneconomic compared with the use of natural gypsum.

3.1.1 Risk and environmental impact assessment

Table 5 shows the heavy metals and radioactive elements in PG. Concerns have been expressed about the risks that may be posed to human health when these materials are added to soil, an issue which has been studied extensively.

Risks are associated especially with the following pathways:

1. The uptake of radioactivity and heavy metals (such as cadmium) from the amended soil by edible crops;
2. The uptake of radioactivity and heavy metals (such as cadmium) into herbage for animals used for human consumption;
3. The inhalation of radionuclides in airborne dust during the application of PG;
4. External exposure rates from the amended soil
5. Groundwater contamination;
6. Radon emission from the amended soil.

Various controlled experiments (Figure 11) and computer-based modelling scenarios have evaluated the application of PG as a soil amendment and fertilizer. In one computer-based study the conservative baseline scenario assumed a PG with a ^{226}Ra activity concentration of about 1 Bq/g (the highest value found in commercially produced PG) and a PG application to the soil every second year for 100 years following an initial application of twice the biennial application rate. Six comparative scenarios were then considered, involving rates of PG application ranging from 1.66-10 t/ha and tillage depths of 22-46 cm. At the end of the projected 100-year period, ^{226}Ra concentrations in the soil were estimated to be only 0.03-0.12 Bq/g [42] presumably due to dilution within the large volume of soil to 46 cm depth.

FIGURE 11

Glasshouse studies on various crops assessing the risks of using phosphogypsum at Sevilla, Spain [43].
(Courtesy: University of Seville, Spain)

The levels and behaviour of radionuclides have also been studied when PG was applied as a source of calcium for cotton [43]. Phosphogypsum with a ^{226}Ra activity concentration of 0.51 Bq/g was applied at rates of 13 and 26 t/ha in conjunction with 30 t/ha organic manure. The concentrations of ^{226}Ra in the water draining from the PG- treated areas were similar to those reported for non-treated areas (2.6-7.2 mBq/l).[15] The activity concentrations of ^{226}Ra in the crops were not affected by the addition of PG and there was no accumulation of radioactivity in the soil. Similar studies to investigate the build-up of radioactivity in soil or uptake by crops show no significant uptake in most cases [44, 45, 46, 47].

As a general rule, concentrations of radioactive materials are frequently reported in terms of the activity of a particular radionuclide per mass or volume of some item of interest, such as water to be consumed, or air in the home. In this instance, the scientists were concerned about the activity in water draining from the treated site. This would usually be reported in becquerels (Bq) (the unit of activity) per litre of water, i.e. Bq/l. In this case, the ^{226}Ra activity is so small that it is reported in thousandths of a becquerel, mBq/l, which is similar to untreated sites.

When considering the impact of repeat application of PG to soils, and the consequent potential for build-up of radionuclides, it must be remembered that the volume of PG added to soil is very much less than the volume of soil that is impacted by the PG. Consequently, when the ^{226}Ra concentration in the PG is very low, it is very difficult to detect any increase in radioactivity in the soil even with very sensitive instruments. Such additional radioactivity will remain in the soil until removed by weathering or crop uptake, but in concentrations that are hard, or effectively impossible, to distinguish from existing background levels.

15 While the ^{226}Ra concentrations in the drainage water were not increased, the concentration of ^{238}U was found to be 200 mBq/l, an order of magnitude higher than normal. However, the ^{234}U to ^{238}U isotopic ratio in the uranium-enriched drainage water was 1.16, as opposed to a ratio of 1 in the PG and other phosphate fertilizers used, and it was concluded that most of the additional uranium in the drainage water did not originate from the PG, but most likely had been desorbed from the uranium naturally present in the soil (at a ^{238}U activity concentration of 0.025 Bq/g).

Transfer of radioactivity from soil to plants is complex. It is dependent upon many soil and crop factors such as plant physiology, soil characteristics and the radioactive element of interest. Where there is concern, the uptake by the plant of any particular element or radionuclide of interest can be measured under actual, or closely-simulated, farming conditions.

To that end, possible effects of radionuclides in PG were investigated in a field experiment in the Cerrado region of Brazil on two typical local soils, one clayey and the other sandy. The mineralogy, organic matter content and concentrations of P, K, Ca, Mg and Al were determined. The organic matter content of the soil was low and the potential acidity high. The concentrations of radionuclides and metals in the PG were measured. The mean ^{226}Ra activity concentration in the PG was 0.252 Bq/g, which was less than the background concentration in the clayey oxisol soils of Sete Lagoas, Minas Gerais, Brazil. It was concluded that using PG as a soil amendment in agriculture would not have a significant impact on the environment [48].

A three-year field study in Florida, USA, in which PG was applied at relatively low rates (up to 4 t/ha), showed no statistically significant increases in radionuclide concentrations in soils and groundwater or in the levels of airborne radon and gamma radiation measured 1 m above the soil [49]. A subsequent study has become a benchmark because of its comprehensive methodology and scope [50]. It assessed the possible environmental impact over time of applying PG at rates up to 20 t/ha on an established Bahia grass pasture, in terms both of the radionuclides and the heavy metal impurities. The results were as follows:

1. Exposure from the inhalation of radon progeny was determined from measurements of the ^{226}Ra activity concentration in the soil, the radon flux from the soil and the radon concentrations in the air. For an application of PG at 0.4 t/ha over a 100 year period, the incremental radon flux from the amended soil was projected to be about 40% of the mean value for undisturbed land (with no phosphate fertilisation) in the region. For a house constructed on the PG-amended soil, it was estimated that the indoor radon concentration would increase by about 1-10 Bq/m^3 (representing, for a typical house, an increase of about 2-20%). For a cumulative application of up to 40 t/ha, the PG was predicted to contribute less than 3.7 Bq/m^3 to the radon concentration over the field.
2. Exposure to external gamma radiation was determined from dose rate measurements made following a single application of PG up to 40 t/ha. After the first year, no gamma exposure attributable to the PG could be detected. It was concluded that the radionuclides from the PG had penetrated the soil or had been removed by weathering or harvesting. The incremental annual effective dose received by an individual remaining permanently on the treated land was projected to be 0.028 mSv after 100 years[16].
3. Exposure from the ingestion of radionuclides in water and food was determined from measurements of the activity concentrations of ^{226}Ra, ^{210}Pb and ^{210}Po in samples of soil, water and forage. The results suggested that the radionuclides contained in the PG had limited mobility in surface water and groundwater during the first two years after application to the soil. However, it is possible that they may have been gradually mobilized, appearing in the groundwater at a later date. The activity concentrations of ^{226}Ra in shallow groundwater after 100 years of PG use were projected to be about 0.1 Bq/l. Levels of ^{210}Pb were projected to be similar to the baseline levels in runoff and shallow groundwater (<0.04 Bq/l). Doses to humans from the ingestion of animal products that had been contaminated with radionuclides taken up by the forage appeared to be within the range of variation in a normal diet.

3.2 Role of phosphogypsum in soil fertility and crop production

3.2.1 Basic soil properties related to the use of phosphogypsum in agriculture

Food security above all requires fertile soils. Safeguarding soil fertility, and thus crop yields, requires that the world's soils have the best possible biological, chemical and physical properties. Foremost amongst these is soil structure.

Soil develops as rocks weather, decomposing with rainfall, freezing and thawing, biological activity into particles of varying size. Particles in decreasing size order are called coarse sand, fine sand, silt and clay. Most of the soil's determining properties in relation to crop production are concentrated in the smallest particles, notably those less than 2 mm in size. Soil structure develops as the different sized particles, especially the silt and clay, are bound together into aggregates (crumbs). Calcium is an important binding agent.

Soils typically have a good structure when they contain adequate calcium. Between the crumbs are spaces (pores/voids), which must hold both water, essential for the plant to grow, and oxygen, because plant roots respire and do not grow into soil where there is no oxygen.

16 Such a dose corresponds to a risk of 1.6 in one million.

A good soil structure is one with a range of different sized crumbs, on which nutrients are held, and which creates a network of pores of different sizes, small diameter pores retaining water for the crop, large ones allowing excess water to drain away quickly and allow air to enter the soil mass. In such a soil roots can grow to find water and nutrients so that crop growth is not limited. Smaller soil crumbs are especially important in the top 5cm soil into which seeds will be planted to ensure good germination and early growth of crops.

Soil structure is adversely affected when salts, added in irrigation water, flooding with sea water, or from weathering of soil mineral particles, accumulate because there is not sufficient water to drain through the soil and remove excess salt. This process of salinisation is the oldest soil pollution problem. The collapse of the Babylonian Empire is considered to be partly the result of the failure of irrigated crops due to the accumulation of salts. Today about 60% of the world's irrigated land is still subject to degradation and loss of production despite our understanding of the problem and its management. It is estimated that about 60% of the US$ 27 billion global annual cost of salt-induced land degradation in irrigated areas is due to lost crop production alone [40]. Well known examples of salt-affected soils in irrigated areas include the Aral Sea Basin (Amu-Darya and Syr-Darya River Basins) in Central Asia, the Indo-Gangetic Basin in India, the Indus Basin in Pakistan, the Yellow River Basin in China, the Euphrates Basin in Syria and Iraq, the Murray-Darling Basin in Australia, and the Cauca River Valley in Colombia.

The accumulation of sodium (Na) in saline/sodic soils and magnesium (Mg) in Mg-affected soils create two distinct problems. Excess Na causes soil aggregates to breakdown and the silt and clay particles disperse filling spaces between the larger-sized sand particles so that there is no pore structure into which water can enter or roots can grow. As the soil dries cracks develop as shown in Figure 12. By contrast, excess Mg binds the finer silt and clay sized particles together tightly to form large clods (Figure 13). Such clods are difficult to breakdown into smaller sized aggregates required for a seedbed into which seeds can be drilled. The effect of the elevated levels of Na and Mg in sodic and Mg-affected soils, respectively, is the degradation of soil physical properties leading to low water infiltration rate and low hydraulic conductivity that impedes water flow resulting in poor water distribution and plant growth.

3.2.2 Soil degradation due to excess sodium and magnesium

Soil degradation due to excess Na and Mg occurs when these elements occupy a larger than normal proportion of the cation exchange sites in the soil. These sites develop when, during the weathering of the rock from which the soil was formed, there is a loss of positively charged ions from the molecules that were part of the original crystal structure of the rock minerals. Consequently, there are sites on the clay and silt particles with negative charges. To maintain soil electrical neutrality these negative charges are balanced by positive charges on cations like Ca^{2+}, Mg^{2+}, K^{+} and Na^{+}, of which Ca, Mg and K are important plant nutrients. The total number of negative charges which can be neutralised by positive charges is called the soil Cation Exchange Capacity (CEC). Usually, total CEC need only be determined once but the base cations, Ca^{2+}, Mg^{2+} and Na^{+} are determined periodically to follow the effects of treatments on the concentrations and ratios of these elements on the CEC. The unit of measurement is $cmol_c$ /kg (centimole charge per kilogram soil). The CEC is a very important characteristic of soil fertility. For many

FIGURE 12

Poor soil condition soil before PG application.

FIGURE 13

Magnesium-affected soils form massive clods upon drying after irrigation events, thereby resulting in poor water distribution, Kazakhstan [35].

agricultural soils with pH in the range of 5.8 and greater, Ca^{2+} and to a lesser extent Mg^{2+} usually occupy most of the sites on the CEC. In acid soils (pH 5.5 or lower) large amounts of aluminium and iron, in ionic form, are held on the CEC. There is no specific or ideal ratio of these ions on the CEC in fertile soils. However, many soils, especially those with a pH 6.0-7.5, contain predominantly Ca^{2+} and Mg^{2+}, K^{+} is often about 5% and Na^{+} much less than 1%. The ratio of Ca^{2+}:Mg^{2+} varies between 5:1 and 1:2 the latter in Mg-affected soils.

Soil properties involving the CEC, which are frequently determined in relation to saline/sodic and Mg-affected soils, are: the electrical conductivity (EC_e), the exchangeable sodium and magnesium percentage (ESP and EMP), and the sodium adsorption ratio (SAR):

1. Electrical conductivity (EC_e) measures the total salinity of the soil solution. The unit of measurement is dS/m (decisiemens per metre), and the values increase with increasing salt content measured in a saturated water extract of soil. The simplest classification of salt-affected soils is: saline soils, EC_e > 4 dS/m, non-saline soils EC_e < dS/m. The electrical conductivity of irrigation water (EC_w) is also measured to indicate the amount of salts being added.
2. Exchangeable sodium percentage (ESP) and exchangeable magnesium percentage (EMP) is the amount of exchangeable Na and Mg, respectively, expressed as a percentage of the CEC. The simplest classification of soil sodicity is: sodic soils ESP > 15%, non-sodic soils ESP <15%. The adverse effect of Mg occurs when EMP is 25-40%.
3. Sodium adsorption ratio (SAR) rates the tendency of irrigation water to increase ESP. SAR is the ratio of the concentration of Na to the square root of the sum of the concentrations of Ca and Mg: $[Na^{+}]/([Ca^{2+}] + [Mg^{2+}])^{1/2}$, where [Na, Ca, Mg] is the concentration of the three ions in mmol/l.

3.2.3 Analysis of soils and phosphogypsum

There is ample evidence, some of which is presented in the following sections, that there are important benefits from ameliorating sodic and Mg-affected soils by using PG. However, its use has been limited due to the general perception that it contains radioactivity, and in some countries this aspect continues to be a public concern. Phosphogypsum also contains non-radioactive constituents, notably fluorides and heavy metals, such as arsenic, cadmium, lead and mercury, which in excess may create problems for soil fertility and crop quality. Both short- and long-term studies have shown that these impurities do not increase significantly in soil to which PG has been applied. However, to convince all stakeholders that there is no problem with radioactivity and other constituents in PG it will be essential to fully characterize PG which is to be used in agriculture.

In addition, analytical services should be available to the farmer to monitor changes in the soils to which PG has been applied. This could be a suite of analyses including CEC, EC_e, ESP, EMP, SAR some of which can be done once (CEC), some which should be done periodically, ESP, EMP plant-available P, K, Mg to monitor changes due to treatment. Analysis of soil to which PG has been added for other constituents, like heavy metals, need only be determined if the analysis of the PG has shown elevated concentrations. Such monitoring programs for PG and soil will inform all stakeholders and help to establish the evidence-based protocols for regulating the use of PG in different agro-environments and soil conditions.

3.2.4 Economic considerations

There is a cost to using PG in agriculture but when considering this cost it must be remembered that besides larger yields there are additional benefits. Growers are more productive per unit of land, labour and water used. If there is no immediate market for the increased produce, then a smaller area of land can be used and the other land used to grow other crops. Ameliorating sodic and Mg-affected soils and then maintaining these soils by periodic applications of PG as required, makes the farming of such soils more sustainable and the livelihoods of those farming them more secure.

A major issue with the use of PG in agriculture is the cost of transport to the fields where it is needed. In the case study from Kazakhstan (Section 3.4.1), PG was transported about 300 km from the production facility to the experimental fields. Of the total cost of applying the PG, 80% was for transport and 14% for purchase and application. Where possible bulk transport by freight train or by river by barge might decrease transport costs. Moving large amounts of PG for use in agriculture may co-benefit a range of stakeholders not involved in the phosphate industry and agriculture such as:

1. large stacks of PG near the factory site are removed at little or no cost to the producer, rather there could be some economic gain;
2. environmental benefits to the society at large as PG stacks are considered environmentally hazardous;
3. opportunities for the transport sector in moving PG from the production site by the most appropriate method;
4. opportunities for infrastructure development such as roads and warehouses as a result of PG and farm produce transport and storage.

3.3 Different amounts of phosphogypsum are needed in agriculture

3.3.1 Use of small amounts of phosphogypsum to improve crop production

Phosphogypsum applications between 100 and 600 kg/ha have increased yields of a wide range of crops shown in Table 6 under specific soil and climatic conditions [20]. Where it is seen, this beneficial effect is probably related to the supply of sulphur and/or calcium to soils where the plant-available supply in the soil is limiting. Most crops contain as much S as they do P and in many soils worldwide the supply of S is declining and crops do not then achieve their yield potential. Sulphur was added to soil together with P in single superphosphate, which is now rarely available. By contrast, the widely-used triple superphosphate (TSP) and mono- and di-ammonium phosphate (MAP and DAP) fertilizers contain no S because it is removed as PG in the production of phosphoric acid used to produce TSP, MAP and DAP.

During the period of heavy industrialisation using coal as a fuel source, the other major addition of S to soil came through aerial deposition in industrial areas from factory emissions. This supply (known as acid rain) is rapidly declining as a result of the requirement for industry to minimize all forms of air pollution, notably those containing sulphur. There is no mechanism to retain sulphate ions in most soils. Sulphate is stripped when water from rain or irrigation drains through the soil rather than being held in it. Consequently, PG should be used in small amounts on a regular basis to replenish plant-available S in the soil. Although PG does contain some readily plant-available P, perhaps up to 4 g/kg, applying PG at the small rates used for the roles described here will not add sufficient P to meet the P requirement of most crops.

3.3.2 Use of large amounts of phosphogypsum for soil remediation

Ameliorating saline and sodic soils requires Ca to replace the excess Na and Mg on the CEC on the mineral and organic components of the soil, and for the displaced Na and Mg to then be leached from the root zone which occurs when excess water drains through the soil. The common source of Ca for the amelioration of Na- and Mg-affected soils is gypsum ($CaSO_4$), which is the major component of PG. The amount of Ca required is based on the amount of exchangeable Na or Mg to be replaced and this is usually determined in the laboratory. The water used for leaching must have a low EC_w. Large amounts of Ca are needed and improving soil structure and crop yield potential takes time because not only has Ca to replace Na on the CEC but the sodium sulphate has to be leached from the depth of soil explored by roots in their search for water and nutrients.

Typically PG rates of 3+ t/ha are applied to remediate degraded lands and such rates may be used every 3-5 years depending on the severity of the problem [35]. Varying levels have been tested in lysimeter studies. For example, in a lysimeter study in Jordan [51], on a saline-sodic soil (EC_e = 4.8 dS/m, and ESP = 58) PG was applied at the equivalent of 5, 10, 15, 20, 25, 35, 40 t/ha. The soil ESP was significantly decreased with increasing amounts of PG up to 35 t/ha PG. Due to the higher cost and different uses of other Ca-supplying soil amendments it was recommended in this case to use PG because of the large amount of Ca required.

Phosphogypsum is a more effective Ca source than chalk/limestone ($CaCO_3$) for ameliorating sodic and Mg-affected soils because chalk/limestone is only very slightly

TABLE 6

Crops that have shown response to phosphogypsum under specific soil and climatic conditions.

Alfalfa	Carrots	Lemons	Peaches	Sugar beet
Apples	Citrus	Lentil	Peanuts	Sugar cane
Avocado	Coffee	Limes	Pepper	Sweet sorghum
Bahia grass	Corn	Lucerne	Pineapple	Tea
Barley	Cotton	Maize	Rapeseed	Tobacco
Beans	Cover crops	Mustard	Rice	Tomato
Beets	Forage	Niger	Rye grass	Turnip
Bermuda grass	Frasier fir	Onion	Sorghum	Upland rice
Black gram	Groundnuts	Oranges	Soya beans	Vegetables
Cabbage	Guinea grass	Pasture grass	Squash	Wheat

soluble in the soil solution at pH > 7, which is the usual pH range of these soils in irrigated areas. However, the solubility of both sources of $CaCO_3$ increases tremendously in soils with low pH and it is widely used to ameliorate soil acidity. Using PG to supply Ca offers additional value to farmers because when used in the large amounts needed for soil remediation PG also supplies P and the cost of PG can be lower than that of other sources of gypsum.

3.4 Case studies demonstrating the beneficial effects of phosphogypsum use in agriculture

3.4.1 Kazakhstan

In this region cotton is the important cash crop and in 2006-07 some of the first scientifically-based field trials were started to measure the effect of PG both on the physical and nutrient condition of some of the badly degraded soils on which yields were poor [35, 52].

3.4.1.1 Ameliorating degraded soils

In a 4-year study involving farmers in southern Kazakhstan [35] PG was used to ameliorate Mg-affected soils with electrical conductivity, EC_e = 1.4-1.6 dS/m and exchangeable Mg percentage, EMP = 28-42. Three treatments were replicated four times: (1) control (no PG); (2) PG at 4.5 t/ha; and (3) PG at 8.0 t/ha.

Phosphogypsum was applied once to the respective treatments at the beginning of the 4-year period. The total amount of rainfall in March and April and irrigation prior to sowing the crop was sufficient to begin the dissolution of PG into the soil solution with increased levels of plant-available Ca and replacement of Mg by Ca on the CEC. The amount of water draining through the soil was sufficient to leach out some Mg as indicated by the decrease in EMP by 18% and 25%, compared to the pre-PG application levels, where PG was applied at 4.5 and 8.0 t/ha, respectively. Similar results were reported in another 2-year study on a similar type of Mg-affected soil

When PG was applied to Mg-affected soils at 3.3 and 8.0 t/ha [35] before and after snowfall, in addition to decreasing EMP, there was an improvement in total and available soil moisture as well as irrigation efficiency. Significantly more moisture was retained in the soil treated with PG compared to the control treatment (Figure 14). The increase in soil moisture was due to the improvement in soil structure and hydraulic properties of the soil (infiltration rate and hydraulic conductivity) and enhancing water movement into and within the soil profile. The high water-holding capacity of PG also contributed to water storage in the soil, the water stored in the root zone contributing to the increase in crop growth and yield. Irrigation efficiency was significantly higher in the PG treatments compared to the control; the average irrigation efficiency of the control was 52%, compared to 59-60% with the PG treatments (Figure 14). Percentage irrigation efficiency in was calculated as:

$$\text{Irrigation Efficiency} = (\text{NIR} / \text{GIR})\ 100$$

where NIR referred to net irrigation rate (m^3/ha) and GIR represented gross irrigation rate (m^3/ha).

Farmers with sodic or Mg-affected soils are eager to use calcium-supplying amendments to improve the productivity of their soils. For example, there are more than 140,000 ha in southern Kazakhstan with Mg-affected soils. Local farmers in informal interviews largely indicated their willingness to use PG as a soil amendment, but needed help in terms of:

1. bulk transportation of PG from the production site, which is some 300 km from the area where PG is needed
2. quantifying the amount of PG to apply per unit area based on the amount of Mg to be replaced for effective soil remediation
3. guidelines on the on-farm methods of application and irrigation management practices
4. recommendations on when to apply PG again to maintain good productivity of their ameliorated land.

3.4.1.2 Increasing crop production

Results from the studies made by participating local farmers on using PG to ameliorate Mg-affected soils in southern Kazakhstan show the beneficial effects of this amendment [35]. There was improvement in soil structure, which led to improvements in germination of cotton (Figure 15) and the formation of buds and bolls all of which contributed to the increase in crop yield.

FIGURE 14

Average values of total and available soil moisture in the 0.9 m soil profile and irrigation efficiency as affected by phosphogypsum treatments (Based on the data from [35]).

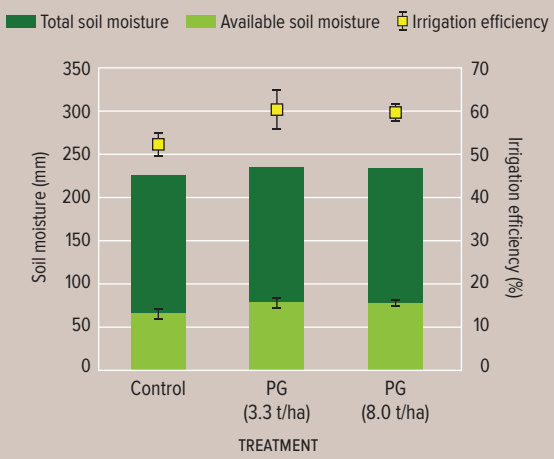


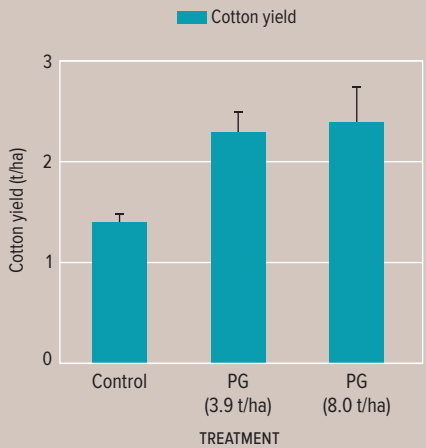
FIGURE 15

Patchy germination and growth of cotton on a magnesium-affected soil (left) **and improved germination and growth of cotton on the same soil** (right) **after phosphogypsum application in southern Kazakhstan [35].**

The average yields of cotton (lint plus seed) from the different experiments were 2.4 and 2.3 t/ha where PG was applied at 8 and 3.9 t/ha, respectively, compared to the much smaller yield of 1.4 t/ha where no PG was applied (Figure 16) [35]. The largest cotton yields (2.7-3.0 t/ha) were obtained during the first year of the PG treatment compared to the fields without PG where yields were only 1.3-1.4 t/ha (Figure 16). In subsequent years, the average cotton yield without PG remained about 1.3-1.4 t/ha but on the PG treatments yields gradually declined. In part this was because no more PG was applied in these experiments and Mg and other salts were probably applied in the irrigation water, highlighting the need to ensure that irrigation water has a low EC_w. This decline in yield where PG was applied indicates that there will be a need to apply PG periodically, perhaps every four to five years, to maintain optimum soil conditions, including soil structure to maintain yields.

FIGURE 16

Multi-year average yields of cotton harvested from phosphogypsum treated soil compared to the control without phosphogypsum (Based on multi-location data from [35]).



The results of these field experiments show that it is possible to restore the yield potential of such degraded soils by applying PG (Figure 16). However, the results also show that the benefit from using PG is not permanent and further experiments are need to show how frequently it is necessary to repeat the application of PG to maintain acceptable yields.

Currently, there are some 10 mt of PG available at two phosphate fertilizer factories in Taraz and Shymkent. Applying PG at an initial application of 5 t/ha, the amelioration of 140,000 ha of Mg-affected soils in the area would need 7.0 mt PG. The remaining PG could be applied in subsequent years to maintain the yield potential of the improved soils or it could be used elsewhere on similar soils in the region, for example: Bayaut district, Uzbekistan; Akaltyn area in Gulistan district, Uzbekistan; Chimbay district, Karakalpakstan, Uzbekistan; Makhtaaral district, Kazakhstan; and Dashauz district, Turkmenistan.

3.4.1.3 Economic benefits

Besides measuring the effect of PG addition on soil properties and yields of cotton, an assessment of the economic benefits and improvements in livelihoods was also made. The economic benefit was based on the following cost components:

1. purchase, transportation and field application costs of PG when applied in the first year as no amendment was applied subsequently
2. farm operations consisting of ploughing, furrowing, harrowing, chiselling, purchasing cotton seed, sowing, weeding, harvesting and transportation of the harvest material
3. fertilizer purchase and application
4. irrigation water provision.

The gross income was calculated from the cotton yield and market price of cotton in the region in the year the cotton was grown [35]. The net income from PG application at 4.5 t/ha (US$ 522/ha) and 8.0 t/ha (US$ 554/ha) was double that of the control (US$ 241/ha). For the

PG treatments, the additional economic gain (US$ 32/ha) when PG was applied at 8.0 t/ha was marginal; suggesting that the lower rate of PG was optimal but this could be tested using a larger number of rates of application to more accurately indicate the best economic rate to use.

3.4.1.4 Improvement in livelihoods

Based on the economic analysis of the data collected from the farmers using PG in southern Kazakhstan, the farmers have a potential opportunity to become more independent of the local cotton-pricing system, which is largely influenced by private companies trading in cotton. Most growers and companies establish contracts based on the 'future price' of cotton, which is usually predetermined by both parties. Farmers take loans from the companies to meet the costs of farm inputs and operations with an agreement to pay back the loan at harvest time. Such agreements benefit the companies because the cotton's 'future price' is always kept lower than the probable actual market price at harvest and a farmer is bound to sell his cotton to the company he has contracted with. Where farmers continue to grow cotton on Mg-affected land, low yields often mean that the income they receive does little more than pay off the loan taken out at the beginning of the season, with little more money to meet their basic needs. Through PG application there is the opportunity to produce more cotton, and hence income and farmers become more independent by selling cotton in the open markets at more competitive prices.

3.4.2 Canada

Hitherto there has been little use of and research on PG in agriculture in Canada. But as the new, world-wide approach to using PG gathers momentum the farming community itself is showing interest in using PG, in some cases without being prompted by the producer.

3.4.2.1. Experimental use of phosphogypsum in dairy operations

In 2006, Agrium[17] was approached by a local dairy farmer requesting PG for use as an amendment for composting dairy manure. Previous research supported by Agrium had shown that addition of the small amount of PG to composting cattle manure can offer significant benefits by increasing the Ca, SO_4 and N concentrations in the composted manure [53]. Phosphogypsum is added at a 10% rate, using approximately 1000 t PG a year. The piles of amended manure are turned every two to three weeks until the composting process is complete, taking typically some four months. The finished compost is then spread on agricultural fields to improve soil properties and to reduce the cost of mineral fertilizers.

The farmer also uses PG, at about 1 t PG/cow/year, as part of the bedding in the dairy barn (Figures 17, 18). The mix comprises 30% PG and/or ground, recycled drywall with 70% wood shavings. This yields a dry and very dense bedding, significantly more absorbent than traditional bedding material and lasting three to four weeks compared to only one week for the traditional bedding of wood shavings alone. The farmer reports that with the gypsum-amended bedding the cows are noticeably dryer and cleaner with reduced levels of mastitis compared to using conventional bedding materials. Overall, significant financial savings are achieved in respect of bedding costs.

3.4.3 Brazil

The Brazilian savanna (Cerrado) – a region of some 2,036,448 km^2 – is now one of the most productive agricultural areas in the world growing about 60% of Brazil's

17 Content kindly provided by Dr. Connie Nichol, Agrium.

FIGURE 17/18

Dairy bedding composed of gypsum and wood shavings, Canada.

grain[18]. Until the 1960s these vast flat areas of acidic soils, largely devoid of plant nutrients, were used very inefficiently by beef cattle grazing the very poor quality herbage. The dramatic changes in cropping and productivity have occurred through longstanding investments in research in soil management and crop adaptation.

Initially acidity in the surface soil was corrected with liming materials, such as calcitic ($CaCO_3$) or dolomitic ($CaMgCO_3$) limestone, which added Ca and Mg or both. Later it was shown that where PG was used there was an added benefit. Both the Ca and the SO_4 in the PG moved down through the soil profile and the Ca corrected the acidity at depth. This increased the volume of soil for root growth and gave roots access to water deeper in the soil profile [54]. This process has clearly demonstrated that such poor soils can be improved by systematic use of large amounts of PG, a lesson which can be applied elsewhere to similar soils. More recently the area of the original savanna in the Cerrado available for grain production has been decreased by more than 50% so that there is now need for further intensification of crop production on the existing agricultural land to prevent further loss of the original vegetation.

Annual production of PG in Brazil in São Paulo and Minas Gerais states is 4.5 mt of which 1.7 mt is used as a soil amendment, 0.7 mt in the cement industry, and the remaining 2.1 mt is stacked in the open. In addition, there is about 150 mt of PG in existing stacks. For tropical soils, one of the best options for the use of PG is its application to soils where there is too little plant-available Ca and too much available aluminium (Al). Applying PG to such soils, improves Ca availability to plants and increases crop productivity and resilience to drought. Although adding PG does not always increase yield immediately, increases in exchangeable Ca and SO_4 in subsoil have been observed which can benefit crops grown in the future. Appropriate management of PG is paramount; but its cost-effectiveness depends on the distance from the source to the farming areas where it is needed.

In Brazil, recommendations on application rates of PG take into account both clay content and the level of Al and Ca in the soil at various depths. For annual crops the 0-20 and 20-40 cm depths of soil must be considered and for perennial crops the 40-60 cm depth is included also. Generally PG must be applied if Al saturation is > 20% (depending on the crop/source) and/or exchangeable Ca < 5 $mmol_c/l$ Where these guidelines for the use of PG are followed crop yields have increased in many cases together with plant resistance to dry periods. Success from using PG is explained when it is realized that

18 This section is based on a case study specially prepared for this Report by Marchi, G., Martins, E.S., Spehar, C.R., Guilherme, L..R.G., Phosphogypsum research perspectives for the Brazilian Cerrado. The authors' contribution is gratefully acknowledged.

TABLE 7

Recommended application of PG based on the average sulphate-S in soil samples from 0-20 and 20-40 cm divided by 2, extracted by aqueous 0.01 M $Ca(H_2PO_4)_2$ (soil: solution = 1:2.5), and the clay content of the soil.

Amount PG kg/ha	Sulphate-S mg/l
Soil with ≥ 20% clay	
10 × clay (%)	≤ 4
5 × clay (%)	5 to 9
-	≥ 10
Soil with ≤ 20% clay	
200	≤ 4
100	5 to 9

[1]Adapted from Sousa *et al.* (2011). [55]

86% of Cerrado soils have < 4 $mmol_c/l$ plant-available Ca. The recommended amount of PG to apply is based on the formula proposed by Sousa & Rein (2011) [55]:

PG (kg/ha) = 50 × Soil clay content at the depth of 30 to 50 or 40 to 60 cm

This empirical formula can be adjusted for different soil types and needs.

Phosphogypsum may also be applied to pastures as a source of S where this nutrient is deficient. The amount to apply is based on the sum of the available sulphate-S in the 0-20 and 20-40 cm depths divided by 2 and the clay content of the soil as shown in Table 7.

On average, applying 1 t/ha PG (~ 15% water) produced in Brazil adds about 200 kg Ca, 160 kg S, and 8 kg P_2O_5 to the soil. This amount of Ca is able to increase exchangeable Ca in the top 20 cm soil by 5 $mmol_c/l$. In general it is recommended to allow a period of five years before considering applying PG again and then the decision to apply should be based on whether soil analysis shows a need for a further application.

Applying PG can have benefits other than the supply of plant-available Ca. Although adding PG does not change soil pH significantly, the increased concentration of Ca and SO_4 ions in the soil neutralises the repulsive forces on clay particles so that they tend to aggregate (flocculate) rather than disperse [56] (Sutherland *et al.*, 2014). This improves soil structure, increasing pore size and creating conditions for fine root development. Pronounced effects on soil structure were obtained with large applications of PG but recommended amounts based on soil analysis should be used and the effects on soil nutrient and physical properties monitored by regular soil analysis. Similar effects of using large amounts of PG are discussed in Section 3.4.1.

Applying 5 t/ha PG to sugarcane ratoon increased the yield of stalk and straw by 16.9% and 17.1%, respectively, over a 4-year period and also increased the amount of carbon stored in the soil [57] (Araújo, 2015). Where PG was applied carbon in the 0-100 cm soil depth increased by 5 t/ha, presumably as a result of increased root growth, and of this increase 80% was in the 40-100 cm soil depth. Increasing soil carbon as organic matter below 40 cm in soil contributes to crop resistance to dry spells.

A downside to the application of PG to Cerrado soils has been concern about losses of cations, principally Mg, from these sandy soils with a low CEC. As noted in Section 3.3.1 the sulphate ion (SO_4^{2-}) is not retained in most soils and when it is leached in water draining through the soil a positive cation like Mg^{2+} is removed also beyond the depth at which roots grow. Losses of Mg can be corrected by the application of dolomitic limestone but this is an additional cost. However, applying the recommended amount of PG and maintaining an appropriate ratio of plant-available Ca and Mg in soil, leaching in Cerrado soils has been small and does not adversely affect crop yields (Sousa *et al.*., 2001, 2007) [58] [59].

3.4.3.1 Phosphogypsum: environmental concerns in Brazil

In Brazil the main environmental issues with PG when it is stacked are pore water, low pH, hydrofluoric acid (HF), phosphate, radioactivity and heavy metals (Bilal *et al.*, 2010) [60], and radioactivity and heavy metals when PG is applied to soil. According to current Brazilian legislation, PG is classified as a class-II solid residue (i.e. not dangerous and not inert). This classification was made by the Brazilian Association for Technical Standards (ABNT)[19] [61]. A class-II solid residue means that after all (economically viable) available treatments and recovery are considered, it may be disposed of in environmentally acceptable ways. This procedure is used by Brazilian legislators to classify materials and ensure proper use, storage, and commercialization [62]. In Brazil more PG is produced than can be used so it has to be stacked at the production site.

19 ABNT: www.abnt.org.br

FIGURE 19

Trucks collection fresh PG from Uberaba, MG, for transportation to the Cerrado region, Brazil.

3.4.3.2 Specifications for agricultural use

The Brazilian Ministry of Agriculture (MAPA) has ruled that PG for use in agriculture must contain at least 16% Ca and 13% S, according to Normative 5 (IN 5) [63]. The heavy metal content, determined according to IN 28 (MAPA, 2007), must not exceed values specified by IN 27 [64]. Maximum concentrations, in mg/kg, are: arsenic, 10; cadmium, 20; lead, 100; chromium, 200; mercury, 0.2. Levels of these heavy metals in PG from the three main Brazilian producers (Copebras, Fosfértil and Ultrafértil) were below these limits. Different limits apply when PG is registered for use as soil acid neutralizer or as a soil conditioner. Saueia *et al.* [65] [66] have shown that the application of PG in agriculture is safe as far as contamination by metals and radionuclides is concerned.

3.4.4 California, South Carolina and North Florida

In California, USA, before the use of PG in the USA was restricted in 1989, there was a market in PG for agricultural purposes, PG selling at some US$ 25 per short ton excluding freight. During the 1970s and 1980s two complete stacks of PG originating from PR from phosphoric acid plants in Idaho and Wyoming, were completely used up for agricultural purposes. Still today, under an exemption in the 1989 Rule, a very small quantity of PG from the PCS White Springs facility, Florida, continues to be used as a soil amendment on farms growing peanuts in north Florida and South Carolina. This PG has a very low radium content and is hence accepted for use by US EPA; but in volume terms it represents only 0.03% of total Florida PG production. Peanuts (ground nuts) have a high Ca requirement during pod-filling (nut growth) stage and rates of application of calcium sulphate generally range between 500 and 1500 kg/ha depending on soil Ca levels and the type of peanut being grown.

3.4.5 European Union

In the European Union, PG is permitted for use as a soil amendment under the product category calcium sulphate [67, 68]. In Spain, freshly produced PG, which is unweathered and moist, has been used in this form for nearly 70 years. It is applied directly to the surface of the soil using conventional equipment (Figures 20, 21).

FIGURE 20

Top dressing of PG, Huelva, Spain.

FIGURE 21

Soils reclaimed from estuary, SW Spain.

3.4.6 India

India[20] is experiencing a time when food and nutrition security have become a major issue because of increasing population and shrinking natural resource base. Fertilizer use patterns are in a general state of imbalance, both in respect of the major nutrients, NPK, but also through neglect of secondary and micro nutrients. This has led to widespread nutrient deficiency, among which sulphur deficiency is commonly the most acute. Overall the current sulphur deficit in Indian agriculture is estimated at ~1.6 mt/y.

Sulphur is one of the main inputs for increasing the yield of cereals and oilseeds. Paradeep Phosphates Limited (PPL) has supported a number of trials and demonstrations on various crops across India. Conducted both by agricultural universities and by PPL's own agricultural experts the results have demonstrated that PG is the most affordable and best source of S in the Indian domestic market. The application of S to S-deficient soils using PG increases the yield of oil-producing seeds such as groundnut, soya bean, sunflower and mustard by 20% to 30%. For crops such as paddy, wheat, pigeon pea and green gram yields are increased by 10% to 20%. It is a paradox yet to be resolved, that in Indian there ison the one hand an immediate need for S to produce larger yields and on the other hand PG as an affordable source of S is stacked unused at different factories.

3.4.6.1 Field trials and general observations

From 2007-2012 field trials were conducted across India on a range of crops studying the effect of PG on crop productivity, crop quality and environmental impact. The specific goals were to:

1. Measure the effect of PG on the growth, yield and the yield characteristics of different crops;
2. Determine the residual effect of PG on succeeding crops;
3. Assess the effect of PG application on soil, groundwater and crop quality especially with respect to fluorine;
4. Undertake adsorption and leaching studies in the laboratory to assess the extent of adsorption and rate of leaching of fluorine;
5. Establish PG as safe fertilizer for agricultural use;
6. To compare the relative efficiency of PG as a source of sulphur against mineral gypsum and single super phosphate.

The general observations of Odisha University of Agriculture and Technology (OUAT) Orissa were:

1. PG helps to improve crop quality especially oil and protein content in oil seed and pulses;
2. Gypsum can be used to good effect in acid soils.
3. Doses of PG for growth enhancement vary from crop to crop;
4. PG does not significantly lower the soil pH;
5. Combined application of a lower dose of lime combined with PG proves better for acid soils;
6. PG is a good, affordable source of S. Its efficiency is as high as other sources of S;
7. PG contains radionuclides, notably ^{226}Ra but its use in agriculture is not restricted by AERB as these limits are well in line with naturally occurring background levels;
8. No significant impact of fluorine is found with the use of PG.

The findings of such studies clearly contributed to the general conclusion of the Indian regulatory authorities that PG may be used in Indian agriculture without restriction.

20 Section content kindly provided by Ranjit Misra, Paradeep Phosphate Limited.

4 Use of phosphogypsum in building materials

KEY POINTS

- The need for affordable housing is universal and PG offers a number of options in regard to both primary and finished construction materials, such as cement, building blocks and wallboard.
- The substitution of secondary for primary materials is encouraged worldwide and there are significant savings to be made from using PG. Some recycling procedures such as the Merseburg process yield a bonus in regard to carbon sequestration through the derivate product calcium carbonate.
- A number of independent studies have shown that radon gas is not emitted in significantly greater quantities in dwellings made from PG-based materials than from conventional materials.

All PG must be thoroughly characterized prior to any decision on how it is to be used in commerce (Figure 7). The radionuclides and heavy metals in PG will vary in identity and concentration according to the rock or blend of rock sources used in its production and according to site-specific production methods. Hence each PG has its unique signature. The choice of appropriate applications will follow from the characterisation profile but will also be determined by factors such as regulatory requirements and markets factors.

An accurate and independent characterisation procedure is of the greatest significance when considering use of PG in construction especially for homes. Because of well-publicized incidents, notably in the United States, of contaminated construction products made from natural gypsum entering commerce, PG use in construction in some markets may face reputational and perceptual obstacles resulting from lack of public confidence concerning safety. Manufacturers must take such aspects fully into account in their safety and quality assurance procedures.

FIGURE 22

Wengfu Group - residential block built from PG-based materials.

Phosphogypsum has been used as a raw material for cement [69, 70], for plasterboard, and for the production of bricks [71], blocks [72], tiles, artificial stone and even boats [73].

Countries currently using PG for building purposes, usually with certain restrictions in respect of dwellings (Figure 7) include China (Figure 22) Belgium, Brazil and India, while it is considered as a potential building material in South Africa for the construction of affordable housing[21]. Almost all the PG used for building products requires the removal of the residual acidic water by washing, and in some cases further processing to remove residual phosphate.

When used to manufacture building materials [74, 75, 76, 77] the regulatory authorities have taken a great interest in establishing the necessary conditions for its safe use [78, 79, 80, 81]. For example, exposure levels have been found to depend strongly on exactly how the PG is used – exposure from PG panels, for instance, depends on panel design, thickness and density. Although most existing studies on such uses show that exposure levels are within internationally recognized regulatory limits (IAEA, UNSCEAR, ICRP) and do not give

21 joburg.org.za/index.php?option=com_content&view=article&id=7407:a-new-way-to-build-houses&catid=123:housing&Itemid=204

rise to concern, two factors in particular seem to prevent or delay more extensive use:

1. unduly restrictive national regulatory criteria for the use of PG in building materials;
2. unduly narrow criteria for evaluating the benefits of use as compared with the long-term economic and environmental costs of indefinite on-land disposal of PG.

Ideally, any regulatory regime should be grounded in a risk- and evidence-based approach, taking account of prevailing economic and social factors, including stakeholder concerns. In some instances local criteria tend to be based on a more simplistic approach using theoretical modelling. The outcome is likely to be generic, conservative and limited to purely radiological considerations. Consequently, the risk assessments tend to be unnecessarily conservative [12].

When only the direct technical and economic merits of using PG as a substitute for other building materials, such as natural gypsum, are considered, the less direct benefits such as the opportunity for reducing the financial and environmental liabilities associated with the indefinite storage of PG may not always be taken into account. This can result in unnecessary barriers to use, an issue which EU policy reform is now addressing (Section 1.4.6). Table 15 shows that when judged by published research papers PG use in building materials has been the primary focus of research and development in the period 2010-15. This finding agrees with the amounts of PG being used in 2015 (Table 2) which shows a strong emphasis on use in building materials, for example in the cement industry.

4.1 Phosphogypsum in cement

Some 5% gypsum/anhydrite is added to cement clinker to make Portland cement. The gypsum acts as a retardant, slowing the setting of the cement when mixed with water. Provided that PG does not contain more than 1% P_2O_5 it can be used as a substitute for natural gypsum without prejudicing the quality of the cement and plaster [82]. Some cement manufacturers specify an even lower value of 0.5% P_2O_5 which leads some PG producers to add lime before despatch to cement works and others to wash it to remove P to the level desired. The use of PG is well established in the cement industries of China, India, Indonesia and the Philippines and very large volumes are used [83, 84, 85]. In China a new method for commercial-scale reduction of P_2O_5 levels in PG to below the desired 0.5% threshold has recently been introduced with a further market advantage that the process results in a white cement product which has a competitive advantage in the Chinese market over grey cements.

In Spain, radioactivity concentrations were measured on two types of cement containing PG identified as PG-1 and PG-2 [86]. Table 8 shows that the activity concentrations of ^{226}Ra, ^{210}Pb and ^{210}Po in the two cements were much lower than those in the PG. The activity concentrations of ^{40}K were higher, while the activity concentrations of ^{232}Th were similar. The significant dilution of activity content indicates the potential for using PG in building materials. Detailed information based on dose assessments can be used to determine the applications of PG as an additive to cement for building applications on a case-by-case basis.

TABLE 8

Activity concentrations in PG and in cement containing PG.

Material	Activity concentration (Bq/g)				
	^{226}Ra[a]	^{210}Pb	^{210}Po	^{232}Th	^{40}K
PG-1	0.205	0.161	0.214	0.019	-
PG-2	0.17	0.161	0.174	0.006	0.013
Cement 1	0.027	<0.056[b]	0.021	0.017	0.73
Cement 2	0.027	<0.036[b]	0.025	0.013	0.613

[a] The activity concentrations of ^{226}Ra and ^{232}Th were determined by equating them to the measured values of ^{214}Bi and ^{228}Ac, respectively.
[b] Detection limit.

4.2 Phosphogypsum wallboard and panels

From the 1960s, in Europe, for example in Belgium and France (when PG was still produced), the opportunities for significant commercial benefit from the use of PG in value-added products such as cement and plaster led producers such as Prayon to focus on these higher-value markets rather than on other applications such as road construction [87]. In China reprocessing PG into “alpha” high-strength gypsum is now being investigated as another value-added option.

The attraction of affordability has seen the development of PG as a low-cost resource for housing construction, for example in Australia, Brazil, China, India and South Africa. The Australian company Rapidwall can process both natural gypsum and PG into high-strength wallboard[22]. More widely industry associations see such resource substitutions as key to their contributions to sustainable development goals [88]. In a safety study in Brazil [89, 90, 91] exposure levels were measured inside an experimental house (Figure 23) constructed using relatively thin, high strength PG panels. The walls were composed of two panels, each 1.5 cm thick with a 15 cm gap between the two panels. For ceiling panels the thickness

22 Rapidwall: www.rapidwall.com.au

FIGURE 23

An experimental house in Brazil constructed using PG panels.

was 1 cm. The mean radionuclide activity concentration in the PG panel material from three production locations was 0.02-0.39 Bq/g for ^{226}Ra, 0.03-0.85 Bq/g for ^{210}Pb, 0.03-0.25 Bq/g for ^{232}Th and <0.08 Bq/g for ^{40}K.

The data taken from the safety study conducted in Brazil present activity concentration ranges and not means. There is a very conservative screening formula used to determine the safety of building materials based on ^{226}Ra, ^{232}Th, and ^{40}K activity concentrations.

External and internal hazard indices are calculated using the following equation (1) and (2), respectively:

where

$$\frac{C_{Ra}}{370} + \frac{C_{Th}}{259} + \frac{C_{K}}{4810} \leq 1 \text{ for external exposure} \quad (1)$$

$$\frac{C_{Ra}}{185} + \frac{C_{Th}}{259} + \frac{C_{K}}{4810} \leq 1 \text{ for internal exposure} \quad (2)$$

C_{Ra} is the activity concentrations of ^{226}Ra (Bq/kg),
C_{Th} is the activity concentrations of ^{232}Th (Bq/kg),
C_{K} is the activity concentrations of ^{40}K (Bq/kg).

If the result is less than or equal to unity for both of the indices, the building material is safe to use without further assessment [92]. When the upper end of the range for each radionuclide is substituted in the equations, the index for external exposure is 2, and the index for internal exposure is 3. Therefore, the panels must be evaluated further prior to use in a habitable structure. Since the screening method is conservative, there is no compensation for the density and thickness of the panel, which affects its ability to attenuate radiation and thereby reduce the radiation impacting humans. It also does not consider radon emanation and panel fabrication or post-construction modifications that affect emanation. In this way they can definitively evaluate the safety of the panels.

The findings from this procedure were presented in a research paper at NORM VII, April 2013, Beijing [93]. Given the very conservative assumptions on which the model was based the results of modelling external and internal hazard indices for PG panels were initially above the recommended levels. Therefore, further studies using more realistic models were used to determine the safety of PG as a building material. The ^{222}Rn exhalation rates from PG plates and bricks were subsequently found to be of the same order of magnitude as ordinary building materials, such as sand and concrete, therefore it was concluded that the use of PG would not put occupants of dwellings at any additional risk due to radon inhalation.

4.3 Comparison of phosphogypsum and conventional bricks for housing

As background, Tunisia has a long and distinguished history in research and development into PG uses. This is well summarised in the 2013 study by Bouchhima *et al.* [94].

In respect of brick-making a unique two-part study in respect of uses of PG in housing was conducted in Tunisia under the auspices of the Centre National des Sciences et Technologies Nucléaires (CNSTN) over a six month period [95]. The first part addressed the feasibility of incorporating PG into the manufacture of fired bricks, by substituting PG for part of the sand used in making conventional bricks. The second part addressed the health, safety and environmental considerations of using such bricks for domestic housing, in particular the potential impact of radon on the occupants.

4.3.1 Phosphogypsum characterisation

Table 9 shows that the ^{238}U and ^{232}Th concentration in Tunisian PG (0.47 Bq/g and 0.15 Bq/g respectively) is comparable to the average concentrations of these radionuclides found in Tunisian soils (0.17 Bq/g for U and 0.20 for Th respectively [96]) and thus using PG does not present any risk for the environment. Additionally, ^{226}Ra activity concentration in Tunisian PG (0.22 Bq/g on average) is lower than that found for the majority of the PG in the world [97], and furthermore, ^{226}Ra, which is the principal environmental concern, remains within the PG [97].

TABLE 9

PG radioactivity (Bq/g) in Tunisian PG. [96]

Origin of PG	Origin of phosphate	^{238}U	^{226}Ra	^{232}Th
Sfax, Tunisia	Tunisia	0.047 (0.035-0.066)	0.22 (0.209–0.223)	0.015 (0.008-0.020)

4.3.2 Composition of the bricks

In Tunisia, standard bricks consist of 70% clay and 30% sand. For physical and mechanical reason the optimum mixture for the experimental bricks was 65% clay, 10% sand, and 25% PG [95].

4.3.3 Room construction

Two identical-size rooms were built on the CNSTN campus, near Tunis, each 4 m x 4 m. The first was constructed using standard bricks and the second using bricks containing 25% PG. To minimize any interference in the measurement procedure, they were built 2 m apart (Figure 24) replicating likely housing density in Tunisia.

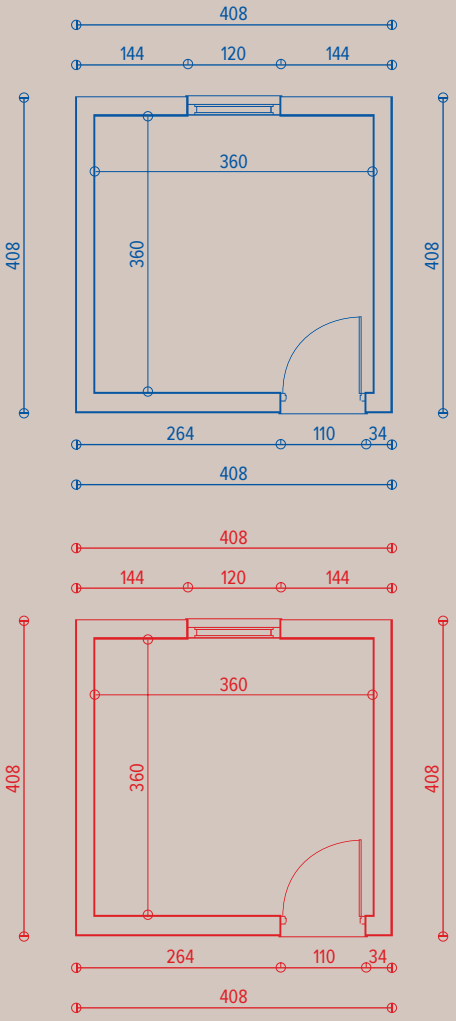
4.3.4 Radioactivity measurements

All radionuclides likely to be detected in the standard- and PG-bricks were measured and the data analysed to address and allay known stakeholder concerns regarding radioactivity in PG. Table 10 shows in detail the concentration of the radionuclides in the various construction materials that were used to make the rooms.

All the activity values in these materials satisfy the universal standards limiting the radioactivity clearance levels within the safe limits of 1000, 1000 and 10000 Bq/kg for ^{238}U, ^{232}Th and ^{40}K, respectively[23] [8]. Radionuclide activities for the conventional bricks are comparable to those made with 25% PG except for uranium which is slightly lower.

FIGURE 24

Layout of the CNSTN experimental rooms (top) – **PG and conventional materials compared** (bottom).

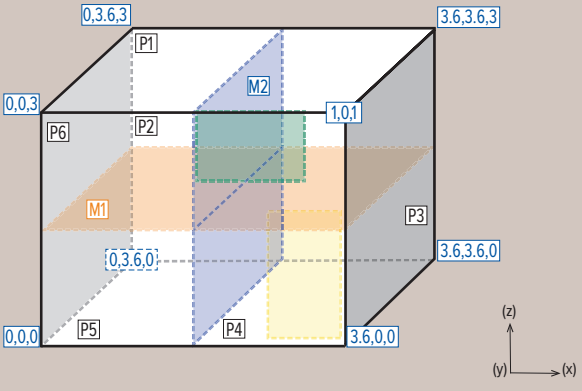


4.3.5 Radon measurements

As the principal public health concern to be addressed was radon, some 36 radon sensitive detectors were placed at various positions inside the rooms (Figure 25).

FIGURE 25

Radon dosimeters positioned in each room.



4.3.5.1 Measurements during construction

To determine the radon emanation from the bricks alone, radon sensitive films were put in both rooms before they were finished, and the rooms were sealed (Figure 26).

As there is no regulation in Tunisia concerning radon, instead the CNSTN study referenced the European Commission 1990 recommendation 90/143/Euratom on the protection of the public against indoor exposure to radon [6]. This recommendation defined 400 Bq/m^3 as the level for considering remedial action in existing dwellings and 200 Bq/m^3 as the reference level for new dwellings.

Radon concentrations in the room built with standard bricks in the horizontal mid-plane (h = 1.5m) varied from 17 ± 6 Bq/m^3 (middle of the room) to 53 ± 11 Bq/m^3 (near walls and corners). Radon concentrations in the room built with PG bricks in the horizontal mid-plane (h = 1.5 m) varied from 20 ± 7 Bq/m^3 (middle of the room) to 63 ± 12 Bq/m^3 (near walls and corners). Radon concentrations in both rooms are comparable and they are significantly below regulatory limits (200 Bq/m^3) [6].

23 BSS Schedule I.

TABLE 10

Radionuclide activities for different materials used in the evaluation of Tunisian bricks [95].

Sample	^{238}U	^{214}Pb	^{214}Bi	^{226}Ra	^{40}K	^{232}Th	^{228}Ac	^{212}Pb
Sand yellow	19.75	7.01	6.18	6.6	65.95	6.82	7.85	5.78
Sand red	14.18	5.41	4.94	5.17	64.14	5.53	6.51	4.55
Soil 0-30 cm	34.63	22.16	21.04	21.60	399.6	27.21	29.96	24.46
Soil 30-60 cm	32.32	24.95	27.44	26.2	479.83	30.13	33.40	26.86
Sol 60-90 cm	28.98	27.77	25.39	26.58	471.42	29.86	32.60	27.12
Brick 12H Ag	30.48	28.42	29.76	29.09	444.26	31.31	34.93	27.70
Brick 6H	26.67	39.00	36.01	37.50	505.21	32.37	35.21	29.53
Brick 12H PG	66.54	38.60	36.59	37.59	440.09	31.16	34.33	27.99
Cement 32.5	25.57	11.54	10.84	11.19	265.09	11.75	13.42	10.09
Gravel 4/15	35.99	5.12	4.68	4.90	48.83	4.11	4.74	3.48
Cayes 25/40	33.18	6.81	4.02	5.42	68.19	4.96	6.06	3.87

In comparing the values to the limit of 200 Bq/m^3, the observed values of 17 and 20 are only 8.5% and 10%, respectively. of the limit. and they differ by only 1.5% (relative to the limit). Similarly 63 and 53 are 31.5% and 26.5%, respectively, below the limit and they differ by 5% (relative to the limit). This was deemed acceptable by CNSTN. CNSTN also took into account uncertainty values for these measurements which were between 7 and 12% for all measurements. Taking these uncertainties into consideration, it can be stated that 17 and 20 and 53 and 63 are broadly comparable. The observed increase was hence statistically too small to be significant.

The rooms were then sealed for a period of ten weeks to maximise radon detection (Figure 27).

Radon levels tend to be quite variable even within a single structure. Ranges with lows and highs within 20% of each other are reasonably comparable, i.e. not orders of magnitude apart. For this reason the USEPA uses an action level of 148 Bq/m^3. The levels recorded in the CNSTN study are well below that action level.

4.3.5.2 Measurements post-construction

Radon concentrations were measured again in both rooms 1.5 m above the floor (Figure 28) during a 10-week period once they were finished.

In the finished rooms (Figures 28, 29), radon concentrations in the horizontal mid-plane (h = 1.5 m) in the room built with conventional bricks varied from 24 ± 8 Bq/m^3 (middle of the room) to 48 ± 11 Bq/m^3 (near walls and corners), while those in the room built using PG-containing bricks varied from 21 ± 7 Bq/m^3 (middle of the room) to 69 ± 12 Bq/m^3 (near walls and corners). Radon concentrations in both rooms were comparable and significantly lower than the standard limit of (200 Bq/m^3).

FIGURE 26

Dosimeters installed in the sealed room under construction.

FIGURE 27

Rooms under construction sealed for radon measurements.

FIGURE 28

Dosimeters inside the finished room.

FIGURE 29

Finished rooms.

4.3.6 Benefits from using phosphogypsum in fired bricks for housing

There are both tangible and intangible benefits from using PG as an alternate material for brick making:

1. *Primary resource conservation:* substituting secondary PG for sand achieved a 25% reduction in sand use.
2. *Energy saving (manufacture):* substituting PG for sand in manufacturing fired bricks results in important savings in energy and money with no reported health risk to the user[24].
3. *Environmental gains:* increased use of PG in construction will lead to the removal of huge stacks of PG with both visual and physical benefits to the environment, including freeing up of land for productive uses.
4. *Sustainability and social benefit:* by using PG as a resource an avoidable waste is prevented, as envisaged under the waste hierarchy, while using it for reducing the cost of housing offers significant socio-economic benefits.

It is known from other studies, such as those conducted in China and India that there are significant energy savings achievable from some mixes using PG in various construction materials including bricks and wallboard.

24 The energy saving aspect is subject to a pending patent application (September 2015) details of which cannot yet be disclosed.

5 Use of phosphogypsum in road construction

KEY POINTS

- PG has been used in road beds all over the world in a variety of climates, notably with wet/dry and freeze/thaw cycles.
- Secondary resources may be combined (e.g. PG and fly ash (FA)) and lead to conservation of primary (virgin) resources of up to 65% and consequent savings on mining materials and energy inputs.
- When mixed with the appropriate quantities of cement (typically 5-7%) PG has shown in a number of independent studies to harden over time as a road base, meaning its life-cycle performance, durability and resistance to rutting, is high and its overall life cycle cost is low, notably when compared with other secondary resources used in roads.
- Environmental impact studies have been conducted, some for 25 years continuously, showing no adverse impact to the environment from either radionuclides or heavy metals

The use of PG in road building has been widely studied and tested, both as a resource in its own right, and as a means of substituting secondary for primary materials in stabilised base and sub-base construction [98, 99]. As PG use in road construction is, by comparison for example with fly ash or steel slag, still in its relative infancy, PG has frequently been tested in experimental or pilot schemes when mixed with a variety of other secondary resources, and in varying combinations. These include sand, cement, sand plus cement, fly ash, fly ash plus cement, recycled crushed stone as aggregate plus a stabiliser including PG, and PG-based concrete. The PG is stabilised using either lime and pozzolanic fly ash, or Portland cement, or self-cementing fly ash. It is produced, placed, and compacted in essentially the same manner as other lime-fly ash or cement-stabilised base materials.

Stabilised base mixtures containing PG have strength development and durability characteristics that are comparable to those of conventional stabilised base materials. Where construction difficulties have been encountered with PG the problems were related either to excessive moisture, over-stabilisation (accompanied by swelling), incomplete mixing, insufficient compaction and sealing, or incompatible stabilisers and prime coats [100].

Unpaved PG roads are commonplace at phosphoric acid production facilities, especially on and around PG stacks. Initial investigations of the use of PG for paved roads in Belgium [101, 102] and France [103, 104, 105, 106] in the 1970s and 1980s focused on its role as filler in embankments [107, 108] and for road stabilisation [109]. These investigations ceased when PG found a more valuable market when sold into the wall board industry. While some investigations of PG use in embankments continued elsewhere [110], the spotlight fell on its potential role in constructing road beds. A number of unrelated studies found that PG mixed with 5-7% cement and, in some cases, fly ash had the best properties both for construction purposes and for cost-effective life-cycle management [98]. The construction of “pilot” roads built in this manner (to assess the methodology) has been successful in climatic regions with wet-dry cycles as in Florida, USA [111] and South Africa [112] and with freeze-thaw cycles as in Finland [113].

Estimates suggest that an average of 25,000 t PG could be used per lane km, which would equate to an annual PG consumption of some 140 mt in the USA alone. Existing pilot studies suggest that using PG as a road bed material is no more expensive than the use of more traditional materials and may be considerably less expensive when the full life-cycle cost is considered [112]. In Florida, USA, in areas near readily-available large amounts of PG, the construction cost was found to be close to one third of the cost using conventional road-bed material. These estimates do not factor in the additional benefit of conservation of primary resources and the concomitant uses of secondary resources, with a potential virgin resource savings of up to two-thirds [113].

5.1 Constraints on use in roads

The major constraints on the use of PG in road construction are economic [114], cultural and regulatory:

1. *Economic constraints due to transport costs:* It has been estimated that to be viable the PG source would have to be within 150–200 km of the construction site [115].
2. *Cultural constraints:* There would have to be a cultural shift in the attitudes of road-builders to using PG [112], perhaps triggered initially by some financial incentive. This would not be the first time the road construction industry would be encouraged to change its mind about as secondary resource as both iron slag [116] and fly ash have been through similar attitudinal shifts [98] that these materials are resources of value not wastes.
3. *Regulatory constraints:* Even where the environmental impact is considered acceptable once the road is built, other regulatory constraints may still exist. In the USA, for instance, the use of PG for road construction is, in effect, prohibited because authorization cannot be obtained for the transport of the material to the construction site [117].

A further consideration is that given the value of PG as a soil amendment and a source of P, producers may be inclined to focus on the higher added-value applications in agriculture.

5.2 Characterisation of phosphogypsum for road construction

5.2.1 Physical properties

Appropriate selection and management of PG according to its physical properties is critical for ensuring its successful use in road building.[25] Phosphogypsum is a damp, powdery, silt or silty-sand material (Figure 1), with moisture content in the range 8-20%. The maximum granular size range is between approximately 0.5 mm (No. 40 sieve) and 1.0 mm (No. 20 sieve), but the majority of the particles are finer than 0.075 mm (No. 200 sieve). The silty size range classifies it as an A-4 soil in the AASHTO soil classification system [118]. The specific gravity ranges from 2.3 to 2.6 and dry weight density from 1470 to 1670 kg/m^3. The addition of fly ash or Portland cement to PG yields slightly higher maximum dry density and optimum moisture content values for stabilised PG mixtures, in comparison with unstabilised PG blends.

Historically, most of the experience in road building has been with dihydrate PG ($CaSO_4 \cdot 2H_2O$); there have been fewer tests with hemihydrate PG ($CaSO_4 \cdot ½H_2O$), the former being generally more finely graded.

25 Some material from this section is taken from www.recycledmaterials.org/tools/uguidelines/sw1.asp, to which reference is made.

5.2.2 Mechanical properties

The shear strength of unconsolidated, undrained and unstabilised PG has average internal friction angles of 32° and a cohesion value of 125 kN/m^2 (18 lb/in^2). Cement-stabilised PGs have internal friction angle values ranging from 28° to 47°, and cohesion values from 76-179 kN/m^2 (11 to 26 lb/in^2) [119]. Coefficient of permeability values for unstabilised PGs range from 1.3 x 10^{-4} cm/sec down to 2.1 x 10^{-5} cm/sec.

5.2.3 Engineering properties

Some of the engineering properties of PG that are of particular interest in stabilised base and sub-base applications include gradation, moisture content, specific gravity, moisture-density relationship, and unconfined compressive strength and typical values for these properties are given in 5.2 above.

5.3 Evaluating secondary resources for road building – AASHTO methodology

In the USA, the evaluation of the environmental and engineering performance of recycled materials in the highway environment [120] should follow the AASHTO Standard Recommended Practice (SRP). This is consistent with the Federal Highway Administration Report suggested framework guidelines in Publication No. FHWA-RD-00-140, Framework for Evaluating Use of Recycled Materials in the Highway Environment (2008).

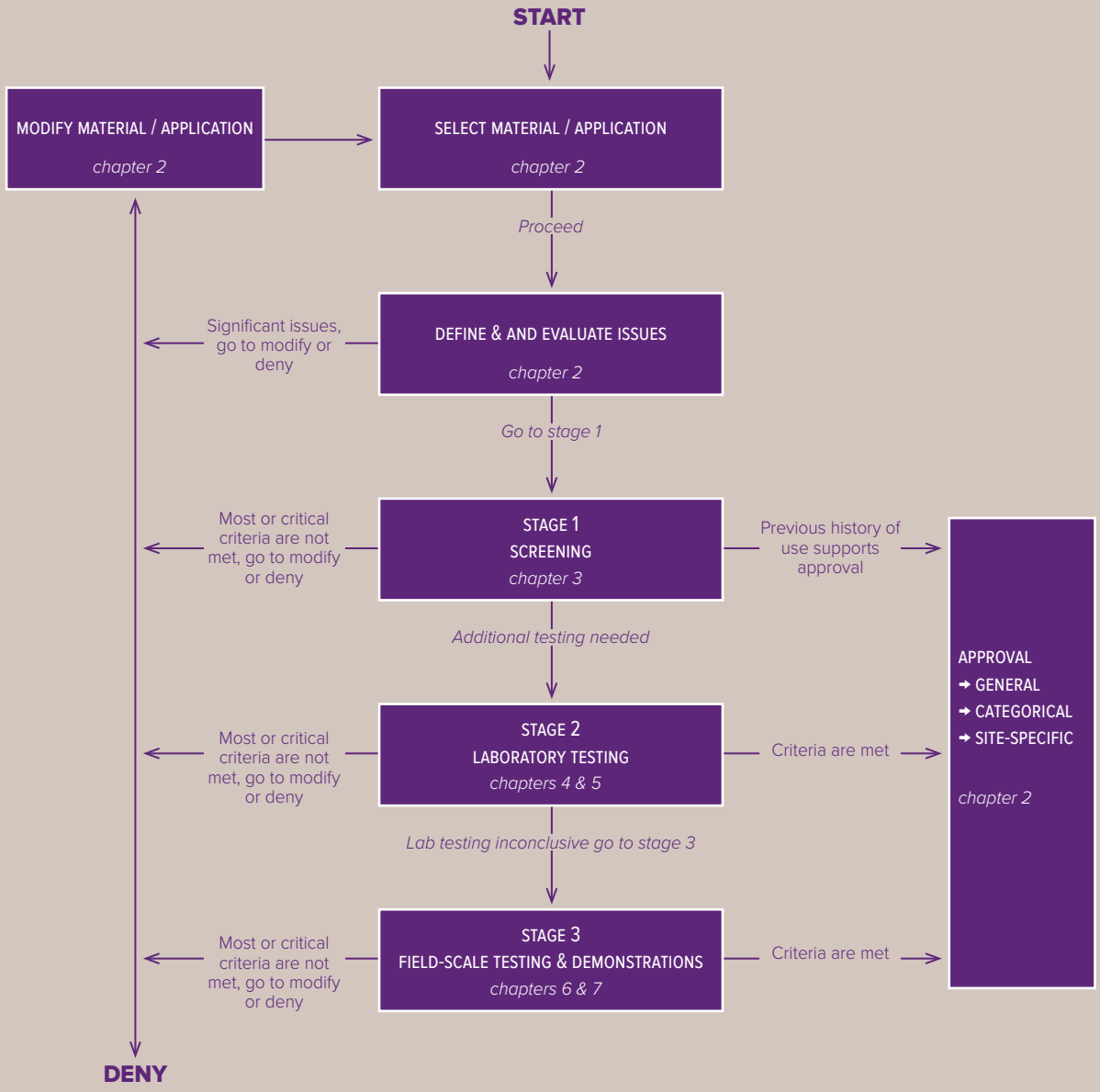
Until the mid-2000s methods for evaluating the engineering and environmental suitability of many materials historically used in the highway environment, such as recycled concrete material, recycled asphalt pavement, blast furnace slag, and coal fly ash had not been formally developed. Some countries adopted regulatory or procedural frameworks for examining the potential for using recycled materials but the absence until recently of definitive methods of evaluation and specific criteria for determining their suitability have in most instances limited the utility of these procedures. To assist with resolving this constraint a five-step evaluation framework (Figure 30) for evaluating the feasibility of using recycled materials in the highway environment, was initially developed in the United States but of use in other settings is suggested. It is now of further value in that it aligns with September 2015 SDGs, notably Goals 9 and 12.

Step 1 – Select material and application

Select a material and an application (e.g., use PG in embankment construction) and submit the application to the evaluator or decision maker.

FIGURE 30

Road bed materials evaluation and selection flowsheet [120].



Step 2 – Define and evaluate issues

Collect all relevant information for input into the decision-making process, including all related historical data, engineering and material property data, environmental, health and safety data, implementation constraints, recycling issues, and economic issues.

Step 3 – Stage 1 Screening evaluation

A Stage 1 screen should include an assessment of all the data collected in Step 2. The purpose of a Stage 1 screen is to determine whether the data collected in Step 2 are sufficient to approve (or reject) the proposed application without additional study. A Stage 1 approval means that the evaluator has a high degree of certainty that the applicant has provided sufficient information to justify acceptance of the proposed material and application. The applicant will typically be required to demonstrate that the proposed material is sufficiently similar to reference materials, which have been used successfully, to warrant approval.

Step 4 – Stage 2 Laboratory evaluation

A Stage 2 laboratory evaluation is recommended if a Stage 1 review determines that existing information collected in Step 2 is insufficient to either accept or reject the application. For a Stage 2 laboratory evaluation) a test

plan should be prepared that delineates the samples to be tested and the tests to be used. Acceptable specifications or performance criteria are identified so that the results can be statistically evaluated to determine if specifications are met or if performance is similar to appropriate reference materials, especially in relation to engineering and environmental parameters.

Step 5 – Stage 3 Field-scale testing and demonstration

Field testing may be warranted at Stage 3 if the available data are still inconclusive after both Stage 1 and Stage 2 evaluations. This Stage is intended to provide, if required, field-scale data on both (1) engineering and material properties, and (2) environmental, health, and safety properties of the material to be recycled so that comparisons can be made with established performance criteria or with reference materials (e.g. a control section).

Engineering monitoring identifies construction and performance aspects that may be affected by the use of a new material. Environmental monitoring identifies impacts to nearby air, soil, and water resources, as well as to the health and safety of those working with the material.

Both short-term and long-term monitoring may be required. Short-term monitoring evaluates how the new material might affect the application during the end-product production process, such as asphalt or Portland cement concrete production, and during and/or immediately after construction. Long-term monitoring evaluates how the proposed application performs during the post-construction period and can involve a time period ranging from several years up to the design life of the application.

A Stage 3 evaluation requires: (1) preparation of a test plan that delineates the field monitoring requirements, (2) listing acceptable specifications or performance criteria against which the results of the field demonstration can be evaluated, and (3) statistically evaluation of the data to determine if the specifications are met or if performance is similar to that of appropriate reference materials.

Field monitoring activities will differ, depending on the type of application being proposed.

5.4 Mixing fly ash and phosphogypsum

There is considerable interest in, and a growing body of experience from, using a mix of fly ash (FA) and PG.

Coal fly ash, a by-product of the combustion of bituminous coal is a pozzolan, and is frequently used in stabilised base mixtures often referred to as pozzolanic stabilised base (PSB) mixtures. Pozzolans contain finely divided amorphous siliceous or siliceous and aluminous material that will, in the presence of water, react with an activator, which contains sufficient calcium and magnesium compounds, to produce a material that has cementious properties. Descriptions of various kinds of pozzolans and their specifications are provided in American Society for Testing Materials (ASTM) C618 [121].

Usually PSB compositions consist of fly ash with lime or Portland cement, or kiln dust, plus water, to form the matrix that cements the aggregate particles together. When used with a chemical reagent[26] typically lime, Portland cement, or kiln dust this type of PSB normally comprises between 10 and 20% by weight of a stabilised base or subbase mix. When used with lighter weight aggregates (such as coal bottom ash), the PSB component may be 30% or more.

5.5 Phosphogypsum use in road construction in different climates and countries

5.5.1 Global experience

World-wide, experience with PG as a material for road-building, notably as a key component of road bed, has been documented in a wide range of countries, settings and climatic conditions, as shown in Table 11. For all stakeholders involved in road construction, however, a paradigm shift is required to give added momentum to PG use, especially as the cost of using PG places it within the range of costs for road building [112, 113].

5.5.2 Case studies

The following four examples of the use of PG in road construction show how it can be used to conserve virgin road construction materials, have negligible environmental impact, and serve specific local needs.

5.5.2.1 Finland

Use of PG as a road bed material in Finland is of particular interest from both its use in construction and as a means of resource conservation. The EU funded a major study of PG as a road- bed material in a rural area in Maaninka, Finland [122], in a climate and conditions of freeze/thaw. Phosphogypsum was used together with FA to construct two roads leaving one year between their construction to assimilate lessons learned in building the first road and apply them when building the second. The project included the opportunity to assess the potential offered by PG to conserve local virgin aggregates [123], to use PG from a large local stack, and assess the likely level of PG use in further road building. Conventional equipment was used to construct the roads, but it became clear that a critical factor was the quality of the mixing of PG and FA, and a new type of mixer, a "counterstroke mixer",

26 Typical chemical reagents for PSBs: www.fhwa.dot.gov/publications/research/infrastructure/structures/97148/cfa55.cfm

TABLE 11

Road building with phosphogypsum – case studies.

A.	Public Road: La Porte, Texas, USA, 1983
B.	On-site Roads: at Phosphoric Acid Production Facilities, White Springs Florida, Experimental, 1985 and on-going
C.	Public Road: Parrish Road, Polk County, Florida, USA, 1986-2009
D.	Public Road: White Springs Road, Columbia County, Florida, USA, 1987
E.	Parking Lot: Phosphogypsum in roller compacted concrete (RCC) – Bartow, Florida , USA, 1988
F.	Public Road: Edenvale and Tembisa, Lethabong Metropolitan Local Council, Republic of South Africa, 1999
G.	Public Roads: Maaninka (Käänninniementie, PT 16207, and Pulkonrannantie, PT 16177) Finland, 1999-2001
H.	Embankments and Shoulders: Northern France (1970-1989)
I.	Proposed Construction: Thornhill Road, Polk County, Florida (1989)
J.	Feasibility and Materials Testing: Sfax Phosphogypsum, Tunisia, 1991-2008
K.	Materials Testing: Central Florida Phosphogypsum, Gainesville, 2007-08
L.	On-site Road: Aqaba, Jordan – as of 2010
M.	On-site Roads: Gela, Sicily and Uberaba, Brazil – as of 2009
N.	Feasibility Study, Onsite Road: Morocco, 2007-2009
O.	Public Road: Former Soviet Union

would be required to reach the necessary quality of mixing if PG was to be used more widely.

The Manager for the Maaninka project, P. Lahtinen, sets the project in direct alignment with Triple Bottom Line (TBL) thinking [16], although he does not use Elkington's terminology:

1. The total amount of industrial residues that is available for National Recycling Centre (NRC)-technology is estimated (by the author) to be 1.9 mt each year. Presently, all of these residues are disposed of in landfills or used as secondary fillers. The disposal of this amount of industrial residues requires 30 landfill hectares each year (supposing the average height of the course is six meters and the average bulk density about 1 t/m^3). The construction costs of a landfill can be relatively high, largely depending on the prevailing requirements for the construction of landfill liners and covers. For a common municipal waste landfill, the costs of a bottom lining system is about 250 FIM/m^2, and the costs of a surface cover structure about 150 FIM/m^2 (based on calculations of different projects by SCC Viatek Oy). Thus, the construction costs of 30 landfill hectares will be approximately 120 million FIM. In addition to construction costs there will also be costs for transport, waste handling and waste taxes. Additional factors to be considered are the loss of land value in and around landfills, and environmental damage. It can be estimated that the total waste costs for disposal of industrial residues will be between 100 and 250 FIM/t. Thus, total waste costs to industry for disposal of the 1.9 mt would be between 190 and 475 million FIM annually (without waste taxes). [113]

While the economics of PG use in Finland, as in South Africa (Section 5.5.2.4), are broadly competitive with other materials, when the wider TBL context is considered the benefits of using PG are very compelling.

5.5.2.2 Phosphogypsum and fly-ash mix

The first demonstration site was at Käänninniementie in Maaninka where the rural road suffered badly from frost damage every spring. The test road had two sections each about 1.7 km long (Figure 33), and these were constructed on 7-18 June 1999.

Before construction began the site was inspected with georadar and reference samples collected to obtain background information. In addition, some 1.6 kilometres of the road was repaired using traditional methods and materials for comparison with the trial sections.

For both trial sections existing material was pushed into banks on either side of the road to give support during compaction (Figure 33). One trial section had a new structure course of PG+FA+aggregate; the other was stabilised with a binder mixture of PG+FA alone with an activator. The PG and FA were delivered by trucks and stored in piles at the mixing station (Figure 31).

The activator was kept in a silo to keep it dry and to make it easier to measure for mixing. The materials were mixed twice with a compost windrow turner in batches of 50 t and moved immediately after mixing to the construction site. A stack mixer was used for mixing PG and FA in batches of up to 200 t (Figure 32).

For the section with a new structure course the road surface was levelled with a planing machine (Figure 33) before the mixture of PG+FA+aggregate was spread and levelled to a 20 cm depth. A first compaction was by the trucks driving over the material. Crushed stone was spread on the structure course and compacted with a vibrating roller.

For the section using stabilised materials, the road surface was levelled after the big stones had been harrowed out. Then the binder mixture was spread to a 5 cm depth over the road surface and mixed with a 20 cm layer

FIGURE 31

Phosphogypsum and fly ash waiting to be mixed as binder.

FIGURE 32

Mixing PG and fly ash with a stack mixer.

of crushed stone scavenged from the old road structure. Mixing was done using a milling cutter and, when necessary, water was added during this work. Compaction was with a vibrating roller. A week later a further layer of crushed stone was spread and the surface was completed later in August. The four principal stages of construction are shown in Figure 33.

During the construction of this pilot project, quality control measurements were made according to a quality assurance plan, which included *in situ* laboratory work. The main control parameters were:

1. water content;
2. thickness of the structure courses;
3. success of the compaction.

During and after construction several test pieces were made of the materials used.

The trial sections demonstrated that this is a sustainable construction process simply by considering the nature and quantity of materials used, and that significant resource conservation was achieved. Some 3,200 t of PG and FA were consumed compared to more than 8,000 t of gravel and crushed stone that would have been required for conventional construction methods.

5.5.2.3 Conclusions from study in Finland

The principal conclusions from the Finnish pilot study were:

1. Correct mixtures of PG with FA and activators are excellent binder admixtures for the stabilisation and improvement of low-quality gravel road courses. Structural road courses based on PG-FA mixtures function quite well in comparison with conventional structures, and have been resistant to freezing and thawing cycles and to the frost heave of soil courses below these structures. Subsequent measurements have also shown that the strength of the stabilised courses continued to increase for more than two years.
2. With the help of effective activators and efficient mixing, the PG-FA mixtures have potential to function as hydraulic barriers having $k < 10^{-8}$ water permeability values. Best results can be obtained with the addition of a small quantity of bentonite and by using an impact mixing method.
3. Several alternative activators can be used with the mixes of PG with FA, and one of the most effective is blast furnace slag plus cement. The activator accelerates the strength development process and improves the long-term durability of both materials and structures.
4. The use of PG, an industrial co-product is environmentally safe and sound, as first shown in laboratory tests but more significantly in the follow-up at the Pilot 1 site.
5. The new types of thin structure for gravel roads seem to function very well where there are low traffic volumes. Thin material structures are low-cost and sustainable and they make it possible to conserve natural, non-renewable resources. Conventional road renovation methods can of course also be used to obtain durable and resistant structures; but this only works where the crushed stone or gravel are of excellent virgin quality and the depth of the courses is not less than 50 mm.
6. The combination of equipment and methods used in both pilot roads proved that effective PG-FA construction processes are possible. However, efficient mixing of materials is essential. Both the stack mixer and the new batch mixer for mixing PG and FA proved to be adequately effective. The impact mixer prototype that was used at the second pilot site in

FIGURE 33

Road construction in four principal stages, Finland.

2000 proved very promising but still needs further development.

7. The success of PG and FA mixes depends on two critical factors:
   i) the geotechnical properties and
   ii) the environmental acceptability of the recycled materials and their mixes.

Typically, and unless Finnish national legislation prescribes otherwise, the use of industrial by-products or, what has been officially considered a "waste" in road construction requires an environmental permit to be granted by the appropriate environmental authority. The material tests before and during the project as well as the follow-up tests at the pilot sites have proved that materials based on PG and FA mixes meet the criteria for geotechnical properties and environmental acceptability.

5.5.2.4 South Africa

The use of PG in pilot road projects in South Africa demonstrates both conservation of natural resource materials and confronting a paradigm shift in road building culture. Paige-Green and Gerber (1999) [112] describe a pilot PG road project in South Africa against a well-established background in South Africa of research and development into alternative, sustainable materials for road building. The case for use is primarily social and economic, but is anchored in sustainable development policy as noted by the authors:

1. Much of the development that will take place in the next decade will be associated with the expansion of existing urban areas and their adjacent peri-urban areas. As the urban areas develop the inter-urban communication routes will also require upgrading. It is in these developing areas and corridors that natural construction materials are being depleted and stockpiles of waste and by-product materials are increasing. A potentially valuable resource is waiting to be utilised. Large stockpiles of industrial and mine discard occur in South Africa, many of them well within economic haul distance for road construction in urban areas. These stockpiles sterilise large areas of property that is urgently needed to assist with the provision of housing and have significant environmental implications, frequently resulting in air, water and soil pollution. Materials such as mine dump rock and slimes, steel

slags, power station and industrial ash, fly ash and old rubber tyres have already proved to be useful in road construction in South Africa while materials such as broken glass, sulphur produced from the oil and petrochemical industry, crushed concrete rubble, shredded plastics and recycled asphalt paving (RAP) have been used for road construction in other countries.

For the project described here stockpiles of 2 mt of dihydrate PG were readily available at the inactive Chloorkop plant of AECI together with some 5 mt at the Potchefstroom plant where some 0.24 mt were being produced each year. These sources were within economic haul distance [150-200 km] for road projects in Gauteng Province as well as other areas in North West Province and Mpumalanga. Other deposits in South Africa could be economically viable for use in growth centres in KwaZulu Natal and in the Western Cape Province.

5.5.2.5 Conclusions: South Africa

Although the results of these pilot studies showed that PG could be used successfully in road construction, the engineers in charge of the project concluded: "A paradigm shift is necessary to fully exploit the potential of PG as a road material" [112], meaning cultural resistance from road builders to adopting new materials such as PG are the primary obstacle to use. South Africa has also commonly experienced media stories[27] citing the US EPA Rule which describes PG as "radioactive waste":

1. PG has properties significantly different from conventional natural materials, but laboratory testing has shown that stabilised PG material has comparable strength and durability with any C4 [high strength] type material and can be used provided it can be obtained cost-effectively.
2. Construction experience has shown that the material can be worked effectively using conventional equipment used in road construction.
3. Monitoring over an 18 month period indicated that the materials were performing well with an unexpected strengthening (86-121%) of the stabilised PG sub-base which was significantly greater than in the control (30-50%).
4. Local research has supported results from other countries showing that PG is a potentially useful road construction material, particularly when stabilised with cement. A number of trial roads have been constructed with adequate confidence.

5.5.2.6 Phosphogypsum use in Florida and Texas

Phosphogypsum has been used successfully as a binder for base-course mixtures and it has a number of advantages compared to clay-based mixtures. The compacted mixture does not absorb significant amounts of water, minimizing construction delays due to rain; shrinkage cracks and swelling are greatly reduced. Compacted PG mixtures are more stable than clay mixtures and if the amount of PG used is correctly optimized, there is a progressive strengthening of the road bed over time [112, 113].

5.5.2.7 Florida

In Florida, USA, two experimental roads built in 1986-1987 used both dihydrate and hemihydrate PG [124, 125]. The results [126, 127], showed that PG, when well mixed and compacted, was effective as a binder to stabilize on-site soil. Mixtures of PG and sand, stabilized with a small amount of cement, possessed a load bearing capacity greater than that of locally mined limestone. Additionally, incorporating PG into a cement- based mixture for making 'roller compacted concrete' (a form of concrete widely used for road construction) led to improved compaction and strength [128]. Figure 34 shows Parrish Road, Polk County Florida after 21 years, indicating very clearly

FIGURE 34

Comparative sections of Parrish Road, Polk County, Florida – PG road bed (upper) **and conventional lime rock** (lower).

27 Radiation fear over plan to build homes, March 20, 2012: www.iol.co.za/mercury/radiation-fear-over-plan-to-build-homes-1.1260349?ot=inmsa.ArticlePrintPageLayout.ot

(ipper picture) how the PG road bed had not failed in that time while the conventional road bed (lower picture) had.

5.5.2.8 Texas

In Texas, USA, road bases for city streets, shopping centres, truck terminals, parking lots and loading platforms have been successfully constructed using cement and FA-stabilized PG and fluorogypsum (a residue from hydrofluoric acid production) [129, 130]. Several experimental sections of road were constructed to establish criteria for the selection of materials and suitable mixes and construction procedures to test using stabilized PG road bases [131]; first in 1991 using a road base consisting of a 7% cement-stabilized PG mixture with an unconfined seven day compression strength of 3.1 MPa; then in 1992 using a road base containing equal proportions of bottom ash (comprising three parts boiler slag to one part cinder ash) and PG. Two additional road sections built in 1993 used PG blended with fly ash and a cement–fly ash, respectively [98]. After two years, these sections were performing well.

5.5.2.9 Environmental impact

Any potential environmental impact from the dissolution and/or leaching of the road bed mix, especially the PG, and any volume change due to changes in water content and/or chemical reactions were evaluated in laboratory tests. These showed that there was little leaching from cement-stabilized PG. Even in the TCLP (Toxicity Characteristic Leaching Procedure) test, designed to simulate the leaching of toxic elements such as mercury and cadmium from material disposed to landfill, the leaching of heavy metals from the PG was less than that from the cement component of the mix. Compaction and cementation effectively reduced the leaching of salts. Compaction also reduced radon exhalation.

The conclusion was that the concentrations of leached chemicals were below levels of environmental concern and in many cases negligible [129], even where the road base was exposed to free water. The water environment under a road does not change very quickly because the road bed has a low permeability and the road surface is designed to be impermeable.

In central Florida, USA, ground-water monitoring adjacent to one of the two experimental roads constructed in 1986-1987 (Figure 36) was undertaken four months prior to construction and thereafter for a further 27 months [132]. Follow-up monitoring continued until 2008. The concentrations of leached chemicals all remained below levels of environmental concern [133]. Similar monitoring of the second experimental road, constructed in 1986-1987, showed that there was no measurable impact on the quality of the groundwater.

5.5.2.10 Radiological impact

The two experimental roads constructed in Florida, USA were subjected to a radiological assessment by monitoring the air, soil and groundwater before and after construction [132] (Figure 35).

Gamma exposure measurements showed that the absorbed dose rates 1 m above the paved road surface was 0.015-0.035 µGy/h higher than pre-construction values and did not change appreciably over time. These incremental dose rates are within the normal range of variability in background levels.

Gamma radiation monitoring was also carried out above an experimental road surface made from roller compacted concrete containing PG [128]. The absorbed dose rate 1 m above the road surface was 0.009 µGy/h, about 50% lower than that above a nearby asphalt road (0.020 µGy/h) and about 25% lower than that above a nearby non-paved surface (0.012 µGy/h).

Outdoor radon and ^{226}Ra in samples of groundwater and soil after construction were not significantly different over a one-year period from those measured before construction [134]. It was concluded that the inclusion of PG in the construction of the roads had had no appreciable effect on radon and ^{226}Ra levels in the surrounding environment.

An investigation of the radiological implications for various age groups and types of receptor activities, of using PG in roads in an urban or suburban setting in 2000 [135] provided conservative estimates and concluded that of those affected road construction workers receive the highest risk (1.5×10^{-4}), but no age group and no type of receptor gave any grounds for concern.

FIGURE 35

Monitoring of gamma radiation above an experimental road in Florida, USA (courtesy FIPR).

FIGURE 36

Aerial view of an experimental road in central Florida, USA, showing the positions of the monitoring wells (courtesy FIPR).

5.5.2.11 Phosphogypsum use in Jordan

This example discusses a specific local need using the on-site PG road at the Aqaba facility of the Jordan Phosphate Mines Company (JPMC). The road was built to give access to the on-site PG stack located immediately behind the chemical processing facility. Dry stacking techniques are used and after 20-30 years of use the stack is now very high. The heat and friction of the tyres of the heavy vehicles moving over the surface of the PG compacts it and it exhibits pozzolanic characteristics without the addition of cement or binder and a hard crust is formed at the surface (Figure 37).

The Aqaba site road has developed over time and this has probably contributed to its structural integrity, consequently, the sides of the road are clearly defined and the integrity of the off-the-road geographical boundaries is not significantly disturbed and remains intact so that traffic can pass easily on either side and there is minimum rutting. There are no requirements for drainage, such as ditches or culverts, because there is very little regular rainfall.

5.5.2.12 Conclusions from Jordan

The JPMC Aqaba PG site road (Figure 37) illustrates beneficial properties and use of PG in an appropriate environment, namely:

1. there was no need for additional materials such as fly ash or cement, or special construction techniques. A truck both moves the material and scrapes and blades the piles, and compacts the material into a road by driving on it
2. the use of materials requiring little or no transportation or additional manipulation is highly desirable and demonstrably effective, it conserves other resources and is economic
3. in the light of the provenance of the road and its compelling reason to exist, no paradigm shift is required on the part of those who select PG as a building material as it is self-evidently the best suited material to use and familiarity on a day by day basis with the performance of the road enables the users also to be the maintainers of its functional value
4. in the context of benchmarking and best practice, the mix of sand and PG which appears to have occurred spontaneously, or by a process of trial and error, has self-evidently resulted in a highly sustainable, low cost solution
5. the structural integrity and security of the road is assured because it guarantees access to the site both for storage and possible use when the PG becomes an asset with economic value.
6. the boundaries between construction and maintenance are removed, in that the extension of the road, even its formation, are linked both physically and causally to the need for the road (Figure 38).

Health, Safety and Environmental oversight for the road is assigned to the on-site manager with responsibility for

FIGURE 37

Compacted PG site road and dry-stacking conveyor-belt system.

FIGURE 38

A PG site-road – a continuous self-justifying build.

the site as a whole. This achieves an effective integration of responsibilities, the road being in practice treated as one more item of on-site equipment. The external agencies responsible for such measures have no difficulty therefore, identifying the chain of command and the Responsible Person.

5.5.2.13 Russia

The Balakovsky branch of JSC "Apatite" (a part of the PhosAgro Holding) has been a leader in using PG in road construction in Russia, first in Moscow and the Rostov region, then in Lithuania, Ukraine and Central Asia[28]. The PG is produced using the local PR and the level of radioactivity is such that there is no local restriction on its use for road construction.

Research and experimental work on using PG and phospho-hemihydrate in road construction was undertaken by the All-Union Institute for Designing Automobile Roads (Soyuzdor NII) which produced, "Guidelines for device bases of road clothes with fresh phospho-hemihydrate calcium sulfate" which is still of practical relevance.

Fresh PG in the hemihydrate form from the Balakovo branch "Apatite" is a warm, dry-bulk mass of light-grey colour with not less than 90% of calcium sulphate and with the optimum moisture level for use. The economic benefits of using PG in road construction are obvious, because 70% of the cost of road is for materials. Using this PG in road construction also conserves local virgin mineral resources (sand and gravel) and there is no cost associated with extracting, transporting and consolidating them, an important feature currently.

Phosphogypsum produced at the Balakovo site is preferable to the more traditional materials because it possesses high strength comparable with the strength of the concrete Mark 300. Consequently roads constructed with PG are suitable for cars and even for heavy vehicles because ruts do not form easily. As a sub-base on which to lay asphalt the optimum thickness of PG is about 30 cm. Moreover, the strength properties of a PG road increase with time, and such roads do not freeze so that seasonal soil swelling does not lead to road damage, and they do not become slushy and muddy after the rain.

During a period of five years specialists of the V.V. Dokuchaev Soil Science Institute monitored the possible impact on the environment of PG used in road construction. No adverse effects on the surrounding soil have been detected.

In August 2014 in the Saratov region a 600 m length of experimental road was built using PG. This formed a monolithic slab that was covered with a layer of asphalt aggregate. If the various observations on this experimental section currently in progress are positive a decision will be taken to use PG more widely in road construction. The use of co-product PG in road construction is most environmentally friendly allowing the use of a recycled resource that may become a profitable and promising business for Russian mineral fertilizer producers.

5.6 Overall conclusions on the use of phosphogypsum in road construction

Techniques for building roads with traditional materials are well established but in many countries a paradigm shift may be necessary to fully exploit the potential of PG in road construction. However, this may not be such an elusive a goal as it may currently seem because it is conceivable, even likely, that PG will follow a pathway to acceptance similar to that of FA, which was rejected as a valueless waste but has now become a material of choice in road construction.

5.6.1 General

1. Phosphogypsum is a suitable substitute for conventional material for road construction because it has a very low impact on operational requirements in terms of personnel or equipment.
2. Significant savings in respect of use of other virgin resources are possible. Construction costs with PG and other materials are broadly similar, with evidence that full life-cycle costs may be lower, in part, because of its property to continue strengthening over time. Where PG is readily available at reasonable costs for material and transport there will be a beneficial impact on materials costings.
3. Using PG in road construction will have a significant positive impact on reducing the continuing accumulation of PG with its associated costs.

28 Content kindly provided by Boris Levin, PhosAgro Holding.

4. When compacted, PG is a solid of high strength, and can be used effectively as a binder to stabilise on-site soil.
5. Phosphogypsum and sand mixtures, stabilised with a small amount of cement, typically 5-7%, possess a bearing strength greater than that of lime rock (currently used in Florida road base) and are suitable for use as base courses for roads.
6. Using the correct amount of PG in a cement-based mixture for Roller Compacted Concrete leads to superior compaction and improves pavement strength properties.

5.6.2 Environmental health and safety

There is no significant adverse impact on the chemical or radiological properties of adjacent ground water or air when PG is used in road construction:

1. In the Florida studies, Ca and ^{226}Ra correlated with the turbidity of the samples collected. At several wells immediately adjacent to the roadway, significant upward trends in sulphate (SO_4) were detected. However, the magnitude of the increases was not sufficient to cause the ambient levels of SO_4 to exceed drinking water standards.
2. Gamma radiation level measurements along the roadways indicate an average enhancement of about 2-4 uR/hour. after the construction of the roads. The maximum post-construction gamma radiation levels as measured are all within the normal range for background levels for soils in this region.
3. The radon levels, measured in the soil and air, as well as ^{226}Ra analysis in soil around the experimental roads, do not show any significant changes after the completion of PG roadways.

5.6.3 Life-cycle cost

Using PG in roads can have a major economic impact on road building where there is a readily-available supply and the cost of transportation to the construction site is acceptable.

At an operational level, the evidence overwhelmingly rebuts the practical barriers to use:

1. *Engineering:* PG is a highly suitable alternate road base material, if correctly selected, mixed and applied.
2. *Construction:* there are no major barriers to surmount in terms of design, operator training or equipment.
3. *Public and environmental health and safety:* There is no meaningful risk to people or the environment through exposure pathways in general, or through the accidental inhalation or ingestion of particulate matter (dust) [12]. Extensive tests on both the radioactive properties and the heavy metal content show that risks both to man and to the environment are typically at or below background levels and hence so low as to require no additional regulatory involvement. [136, 137]

If conservation and sustainability goals are to be met, there is an overriding obligation on road builders to follow best practices in the selection, transportation and use of all building materials, and PG is no exception.

At a time when public policy is focused on sustainability and conservation of resources, the ready availability of PG as a low-cost, narrow carbon footprint material, requiring little or no further processing for use, is highly attractive from a cost/benefit perspective, especially in markets which at typical road transportation costs can source within 150-200 km of a suitable (probably weathered) source. Outside that range, transportation costs may prove a greater constraint on use than any physical, environmental or engineering considerations.

6 Marine applications

KEY POINTS

- When correctly mixed with other suitable materials there are significant opportunities for large-scale uses adjacent to or in the coastal marine environment, as shown e.g. for strengthening sea defences and flood prevention.
- Extensive testing has shown that specific material combinations and designs are well suited to the marine environment and may be designed to withstand sustained immersion in seawater with no adverse consequences to the environment or marine life.
- Highly sensitive uses such as for making beds for oyster cultch have been successfully tested.

Many PG stacks are located near vulnerable, low-lying coastal areas and well within the economic transport range of 150-200 km from the stack making the use of PG in marine locations economically attractive. Uses that have already been developed include sea defences, stabilisation of vulnerable coasts, protection of coastal wetlands and other areas against the risk of flooding and re-establishing coastal industries such as shellfish production. Some countries also exempt PG from regulatory restriction when used in emergency situations, such as coastal flooding.

The use of PG in coastal and marine settings has been reviewed and researched. Researchers at the University of Louisiana, (LSU) USA demonstrated that there are a number of factors that favour the use of PG in such settings [138] [139]. These include:

1. the use of large quantities of PG for local construction purposes, notably levees or coastal defences;
2. the consequent savings achieved of primary resources, notably rip-rap from granite, by such use of secondary resources;
3. regulatory trends for PG stack management increasing the overall life-cycle cost of indefinite land-based disposal and the potential off-set in both direct costs (less disposal) and in direct costs (land use/less land required) favouring use
4. the annualized return on investment reclamation and remediation of inundated soils affected by sea water following such periodic events as hurricanes, e.g. annual impact of the Monsoon season in Bangladesh and after catastrophic storms such as Katrina in the United States or Yolanda in the Philippines;
5. in-water uses such as oyster cultch and artificial reefs.

6.1 Economic perspective

While the economics of PG briquette manufacture for coastal and marine applications was finely balanced in the first study conducted in 2001, the economics when reconsidered in 2010 were already significantly more favourable.

6.2 Leaching and bioaccumulation studies

Research has focused on finding a PG-FA-cement mixture that is stable in seawater, and measuring the effects of such stabilized PG mixtures on the surrounding environment, which includes leaching and diffusion of elements from them. Many bioaccumulation studies with different composites containing PG have shown very little evidence of leaching or bioaccumulation of toxic metals and ^{226}Ra.

6.3 Structural considerations

Typically, shoreline erosion dikes are 0.6 to 0.9m above the waterline and generally have a cross-section with a 1.2 m wide crown and a ratio of 2:1 or 3:1 for the back slope and a ratio of 3:1 or 4:1 for the front slope on the water side (National Gauging Database (NGD)). This requires significant quantities of material, generally riprap with high durability. Besides cost, the use of rip-rap causes excessive settlement of the dike due to its effect in consolidating the underlying soils. This problem can potentially be minimized by using lightweight materials, such as stabilised PG briquettes, as core material

in conjunction with geogrid (a form of synthetic soil reinforcement), with riprap used as a surface armouring. This configuration would dramatically decrease costs by reducing settlement and the amount of riprap needed.

The LSU field results indicated that a 62%, 35%, 3% mixture of PG-Class C FA-Portland Type II cement can survive in seawater for more than two years. Economic analysis indicated that the final cost of producing the briquettes was highly dependent on the cost of cement and Class C FA.

6.3.1 Briquette combinations

The LSU researchers selected the best four composite combinations for briquettes (Table 12) based on the physical integrity of the briquettes and their ability to withstand submersion in natural saltwater. Fabrication parameters are shown in Table 13.

The four composites showed no signs of degradation after being submerged in natural saltwater more than one year.

The range of effective Ca, SO_4 and ^{226}Ra diffusion coefficients was 1.36-8.04 x 10^{-13} m^2/s, 2.96-7.20 x 10^{-13} m^2/s[1], and 1.46 to 2.90 x 10^{-13} m^2/s, respectively. The predicted critical time (t_c) when leaching would cease for Ca, SO_4 and ^{226}Ra was 64-78, 122-137, and 150-470 days, as the leaching processes are balanced by precipitation reactions.

The ^{226}Ra concentrations in the TCLP leachate were well below the current USEPA regulation value for drinking water (5 pCi/l), and the metal concentrations were well below the USEPA toxicity characteristics limits. The effective diffusion coefficients of Cu, Cr, Zn, and Fe ranged from 10^{-12} to 10^{-16} (m^2/s).

The results of the engineering properties test indicated that the composite material could be classified as well-graded sand with little or no fines. The direct shear test determined the angle of internal friction as 49-50°. The USCS classification would also qualify the PG briquettes as a potential fill material in embankment construction projects because of the excellent workability characteristics.

The durability and saltwater survivability of stabilised PG briquettes under tidal conditions ("wet/dry" cycling) in the coastal area has not to date been adequately tested and would need to be investigated.

TABLE 12

Four best PG briquette combinations.

PG %	Class C fly ash %	Portland Type II cement %	Production cost/t US$ Florida (2003)
73	25	2	11.94
67	30	3	14.08
63	35	2	14.66
62	35	3	15.45

TABLE 13

Fabrication parameters used for the best four phosphogypsum combinations.

Briquette parameter	Final four combinations
Water content, %	4
Mean pressure, N/m^2	143.1 x 10^6
Mean solid density, g/cm^3	2.15
Mean mass, g	19.5
Mean volume, cm^3	9.0
Dimensions, (L x B x H) cm	42.5 x 23.5 x 14.5
Approx. surface area, cm^2	28.75

6.4 Conclusions

Research has focused on the stability of mixtures of PG, fly ash and cement in seawater environments, their effect on the surrounding marine environment, and the possibility for making oyster cultch or artificial reefs. It has also reviewed the economics. Mixtures containing various proportions of PG (55-73%), cement (2-10%) and fly ash (25-42%) suffered little degradation in seawater over periods of up to two years. Resistance to degradation in seawater and reduction in the diffusion of heavy metals and ^{226}Ra is the result of the formation of a calcite ($CaCO_3$) surface layer that acts as a physical barrier to seawater intrusion. Including fly ash in the stabilized PG mixture reduces the amount of cement to 2-3% and also promotes the formation of a calcite layer. The leaching of heavy metals and ^{226}Ra is comparable with that from pure cement and well below levels that might be of concern for health and environmental reasons. Stabilized PG mixtures are capable of supporting the growth of oysters and other shellfish without any significant accumulation of heavy metals.

7 Phosphogypsum as daily landfill cover

KEY POINTS

- Lack of available space for and social resistance to landfill, notably around the world's major cities, has led worldwide to a shortage of landfill sites.
- This space constraint has been compounded by the waste hierarchy which regards disposal of any material as the least desired outcome.
- Laboratory and commercial-scale pilot tests have shown the microbial decomposition of organic matter in landfill is quicker in the presence of PG used as a daily cover (in place of soil) in that the sulphur provides an energy source to the microbes, resulting in reduced volume up to 45% and eliminating the need for additional soil cover.
- Energy sources such as methane and H_2S can be recovered from the landfill and provide a source of revenue.

7.1 Biodegradation of landfill wastes

Experimental laboratory studies in Florida with FIPR funding [140] indicated that the addition of PG to municipal solid waste greatly enhances the rate of biodegradation, and consequently makes it possible to use a smaller landfill volume. Also in Florida, and maybe elsewhere, waste sent to landfill must be compacted and covered with soil every day to avoid trash being blown about and to reduce possible rodent infestations. This requires digging "borrow pits" to obtain the necessary soil, which leaves unsightly holes that provide an ideal breeding place for mosquitoes during the rainy season. Using PG for the daily cover material would eliminate the need for a soil cover.

Gas monitoring during microbial decomposition indicated that initially sulphate-reducing bacteria will use PG as an energy and oxygen source and carbon dioxide will be produced while the conditions are aerobic. Then as the oxygen in the landfill is depleted, the dominant digestion mode becomes anaerobic and methane and hydrogen sulphide will be produced.

The information from these bench and pilot scale studies, suggested that using PG in landfills offers some exceptional advantages:

1. up to about twice the volume of municipal solid waste could be disposed in any given landfill because of increased rate of biodegradation and compaction. Due to the more rapid decomposition of the waste, landfill gases would be generated in greater volumes earlier in the life of the landfill making their collection simpler and easier and energy could be recovered from them.
2. using PG as a cover material would make better use of land and achieve meaningful cost savings. There would be no need for borrow pits to provide soil as a cover material. Such pits are both unsightly and undesirable around the actual landfill site.
3. the leachate would contain less heavy metals when using PG if the microbial decomposition generates hydrogen sulphide leading to the formation of insoluble metal sulphides that are not subject to leaching.

These concepts have yet to be tested in long-term field conditions.

7.2 Field test, Minas Gerais State, Brazil

The technology of using PG in landfill sites has been used in Brazil following meetings of scientists, regulators, and producers from Brazil with colleagues in Florida [141]. The problems of landfill sites in Brazil are severe. For example, municipal waste is directly disposed on the land surface[7], as shown in Figure 39, in 73% of Minas Gerais cities.

While the use of HDPE liners in the USA is a reasonable requirement, it may not be as feasible in other regions, and natural local materials may be used instead. The study in Brazil used pathway modelling to determine the potential human exposure to radioactive materials and metals in a leachate plume. Mathematical modelling of contaminant transport in aquifers used experimental

FIGURE 39

Urban waste disposal to land, Brazil.

results from PG biodegradation with local landfill design specifications, including leachate confinement. The conclusion reached was that application of PG as a daily landfill cover does not result in significant risks to either human health or the environment under typical conditions in Brazil [142] and could offer major health and environmental improvements compared to current practices.

8 Secondary resource management

KEY POINTS

- In line with the waste hierarchy the use of any secondary material as a substitute for primary resources is encouraged. There are ~4 billion t of PG available for such use (2015).
- Secondary resources are now included in the UNFC resource classification system of the UN.
- Sulphur recovery for re-use in the wet process can assist those operators with no locally available source of sulphur making their process increasingly "circular" in nature.
- Reprocessing PG as a primary material for making ammonium sulphate and calcium carbonate shows how a previously linear life-cycle model for the wet process can be reengineered.

Led by the United Nations Economic Commission for Europe (UNECE)[29], the United Nations Framework Classification (UNFC) has been developing an update to the UNFC 2009 edition [143] which for the first time classifies both mineral and hydrocarbon primary resources and reserves according to a single three digit system. Since 2014, in alignment with the objectives of the waste hierarchy (Figure 2), this classification process has been further extended to include secondary resources. In respect of secondary resources, the inclusion has the explicit objective of encouraging the beneficial use of such resources rather than their disposal as wastes. Among the various secondary resources identified as suitable for inclusion under UNFC is PG[30].

As an example, Figure 38 illustrates the reprocessing of PG as a secondary resource to create both ammonium sulphate, for use in agriculture, and calcium carbonate for use in the cement industry, which has the further merit of sequestering significant tonnages of carbon dioxide (CO_2). It is likely that Ra in PG would migrate with Ca because they are chemically similar and both would be in the calcium carbonate, leaving the ammonium sulphate free of radium and the subsequent radon scrutiny created when it is used as a fertilizer. The production of ammonium sulphate from PG has long been practiced but the market is limited. The radiological aspects of biological recovery processes for S have not been studied, although the radionuclides would be expected to remain in the residue leaving the S essentially free of radioactivity [144].

8.1 Sulphur recovery

The recovery of S from PG has attracted global interest for a number of reasons, both economic and strategic. In the US, it would give a company control over its S supply and be attractive economically when the widely cyclical price of S remains high. In other regions native S may not be available or the accumulation of PG may not be tolerated. In China, for example, in addition to the economic benefit of not having to import S from Canada, there is also the immediate regulatory requirement to consume 20% of PG produced. Many process routes, both chemical [145, 146, 147, 148, 149, 150, 151, 152, 153] and biological [154, 155, 156, 157] have been explored to use PG as a sulphur source.

Traditional S recovery schemes producing SO_2 all have high energy input requirements, as well as the fact that PR processing plants have incorporated electricity cogeneration capabilities based on the heat generated in the production of sulphuric acid. Having the S enter the process as SO_2 eliminates a substantial portion of the heat source, because the heat of combustion of S would not be available. In addition, producing SO_2 from PG gives a concentration of only 6% SO_2 in the input stream to the sulphuric acid production unit which is half the 12% on which current plants are designed to operate. Increasing the concentration of SO_2 in the stream would require an additional, expensive step in the process. Conversely, using only 6% SO_2 in the stream would require a much

29 UNECE: www.unece.org

30 A presentation was made to the Plenary Session of the UNECE Expert Working Group by Hari Tulsidas, Roberto Villas Boas and Julian Hilton which included a case study of PG as a secondary resource (United Nations Office, Geneva, April 24, 2015).

larger plant, with a larger capex to handle the larger input volume required. Such a large plant would only produce the same output as a smaller plant operating at 12% SO_2. Even the FIPR circular grate process, which achieved an 8% stream through the addition of pyrites to the grate, would still fall far short of system requirements.

Recent FIPR efforts examining alternatives for S recovery have focused on PG conversion to hydrogen sulphide and then to elemental S though the traditional Claus process. Sulphur rather than SO_2 would then be available to the PR processing plants. Once reliably demonstrated, it is anticipated that there will be wide interest in implementation of the technology as regulatory pressures to consume PG to avoid environmental risks associated with stacking expand globally.

Other studies by FIPR compared the energy balances of recovering S by the SO_2 and hydrogen sulphide routes and the production of cement clinker from the calcium by-products. There was a substantial energy advantage using the hydrogen sulphide route together with a strategic advantage, because phosphate producers would be able to maintain control of their own S supply, and be less vulnerable to S price spikes and demands of S suppliers. Also, the substantial transportation costs associated with sulphur delivery would be eliminated.

8.2 Co-product strategies – ammonium sulphate and calcium carbonate

One commercially attractive use of PG as a secondary resource involves the well-established, Merseburg ammonio-carbonation process. This process (Figure 40), yields ammonium sulphate, an excellent fertilizer, and calcium carbonate, a useful product for neutralizing the acidic water produced in the wet process.

Almost all the radioactivity within the PG would be in the calcium carbonate, leaving the ammonium sulphate with a lower level of radioactivity than in most natural materials. A major ancillary benefit is that for the consumption of 1 mt of PG, some 170,000 t of CO_2 are sequestered. Although used on a commercial scale in China, India and Indonesia, the process has enjoyed only limited use because of abundant supplies of inexpensive ammonium sulphate from other industries.

FIGURE 40

Wengfu Group ammonium sulphate and calcium carbonate production.

9 Return of phosphogypsum disposal and storage land to productive use

KEY POINTS

- The stacking and disposal on land of PG takes up very considerable areas of land many of which are now experiencing significant levels of encroachment.
- Pressure on land use, rising land values, environmental and aesthetic concerns about stacking indefinitely, together with the waste hierarchy requirement that disposal of any material as a waste is the last resort has made the case for eliminating PG stacks increasingly strong.
- Full Cost Accounting pays considerable attention to the impact of waste disposal on land and property values and associated revenues.
- A number of reference examples now exist of the recovery of land from use for stacking to productive uses.

The disposal of PG whether to land or sea is not an ideal solution in an era when there is general agreement on policies of sustainability and the pursuit of zero waste and zero emissions. But changing established practices of disposal can be very expensive and, as has been seen in Europe, can come at the cost of closure of some parts of the industry. In any country challenged by difficult socio-economic conditions putting essential industries such as the phosphate industry at risk is unthinkable. How then to plan and execute policies focused on stacking for use not stacking for disposal that are economically, socially and environmentally acceptable is the key objective.

9.1 Stack life-cycles

All PG stacks have life-cycles, which currently are of three main types. Historically, the predominant type has been stacking for indefinite disposal. By contrast driven by sustainability goals there is now considerable interest in stacking for eventual complete use with the end goal of leaving no legacy stack. A third variant is already being achieved in some production facilities. For example, in south India, where a given land area is used for interim stacking, typically with a capacity equating to annual production. Then, at key times, such as after the monsoon rains, all the stored material is sold for use in soil remediation. In Belgium, Prayon uses all the PG it produces for supply into construction material processes on a continuous basis. In both cases there is no legacy stack.

Given the very large volumes of PG produced in the wet process even the largest countries have difficulty achieving equilibrium between production and use. By volume China is leading in use. Kailin as producer has achieved 100% use, including backfill to the mine. Overall consumption is growing but in a range of 40-60% in regard to total production capacity. Brazil is the first country to achieve the objective of fully consuming what it produces, some 5 mt/year. But this achievement is to some extent misleading in that only 40% of the phosphoric acid Brazil consumes is produced there. This means 60% of the acid, and hence 60% of the PG is produced elsewhere. So achieving full equilibrium between production and consumption for Brazil will involve solutions for the companies supplying Brazil as well as for Brazil itself.

9.2 Encroachment

In the immediate aftermath of World War 2 the phosphate industry rapidly expanded, driving global growth of the wet process. Plants were typically sited at or near ports making it easier to ship and receive PR, the PG slurry easier to discharge to sea and the product easier to ship to markets around the world.

Production facilities were located some distance from the cities with which they were associated and it was not anticipated that the local population would come into direct contact with them. In addition the global view of marine PG discharge has changed as greater appreciation of potential negative environmental impacts has evolved. But as the global population and in particular the world's cities have grown, so encroachment of people has affected an increasing number of production facilities in an increasing number of countries. Some port cities such as Athens and Cork no longer have their phosphoric acid plants, but they do have small PG stacks fully sur-

rounded now by settled populations. In Athens the stacks have been remediated for use as parks. Many producers in port locations where there is urban encroachment, such as in China, Jordan, Lebanon, Tunisia and Turkey are being put under considerable pressure to resolve the issue of stacking. In some instances this means continued discharge to sea, in some cases the solution is seen to be full use of the PG they produce, in other cases costly remote site storage is demanded, greatly increasing stacking and disposal costs.

In cases such as the city of Huelva, Spain (Figure 41) and Thessaloniki, Greece, the value of land itself is the key driver as land is required for housing and industry.

FIGURE 41

Huelva, Spain, city, industrial complex and PG stacks in close proximity.

9.3 Phosphogypsum stack site remediation – Taparura, Sfax, Tunisia

9.3.1 Background

The Groupe Chimique Tunisien (GCT) SIAPE B plant was constructed in 1964 in the then northern outskirts of Sfax, Tunisia, near the city's commercial port. For environmental reasons it was closed in 1991 when the plant and stack area had become completely encircled by the city of Sfax.There were releases of polluted water and air to the environment and constant leakage of PG slurry to the beach and the sea from the stack which was sited between the plant and the sea, as shown in an aerial photo, Figure 42, taken at the time of its closure.

After the plant itself had been decommissioned by GCT, the French monitoring agency ALGADE in collaboration with the Tunisian national regulatory centre for radiological protection CNRP (Centre National de Radio Protection) conducted a radiological survey of the beach and surrounding area. This study concluded that there was no radioactive (NORM) contamination of concern.

9.3.2 Taparura project - remediation scope and objectives

The name Taparura comes from the ancient name for Sfax.

FIGURE 42

Taparura, Tunisia, site prior to remediation.

Begun in 2006 the scope of the remediation project was to transform a 420 ha area previously owned and operated by GCT and return as much of the land as possible to productive use, including about 150 ha for residential, commercial and tertiary development. Additionally about 260 ha, which had eroded from the beach during the period of GCT operations, was to be reclaimed from the Mediterranean for an urban park and a 3 km stretch of restored beach.

A specialist company, Société d'Etudes et d'Aménagement de la Côte Nord de la Ville de Sfax (SEACNVS) was set up for the remediation project and for the development of the site. First the environmental impact assessment initially by ALGADE was confirmed. Then a chemical and radiological monitoring system was established.

In line with the waste hierarchy objectives of waste prevention and minimization two actions were taken to accompany the remediation work being done on- and off-shore. First, a circular HDPE lined impoundment system was set up (Figures 43 and 44) to contain the small volume of materials deemed too polluted to use for beach reclamation.

Once finished this area was to be capped, covered with top soil and turned into a small park (Figures 45, 46, 47, 48).

The remainder of the beach was reclaimed (Figure 47) and the park was opened (Figure 48).

Some small scale studies were also conducted for selecting locally grown plants for the Park area, and these included tomatoes (Figure 49) and figs (Figure 50).

FIGURE 43

HDPE barrier installation for PG impoundment.

FIGURE 44

PG impoundment for legacy waste and surrounding restored beach and building land.

FIGURE 45

Aerial photo of the remediated site.

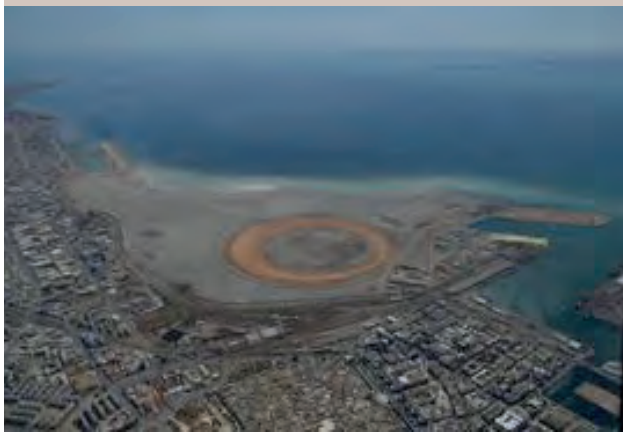


FIGURE 46

Aerial photo showing clear coastal water.

FIGURE 47

Restored beach.

FIGURE 48

New park.

FIGURE 49

Remediated stack growing tomatoes.

FIGURE 50

Growing figs.

The project overall has been so successful that feasibility studies are being conducted for remediating an area ten times as large (~4,000 ha) further along the coast.

9.4 Huelva, Spain and Gela, Sicily

Relative to the quantities of PG to be managed in other settings those at Sfax were relatively small, not least because a lot of the PG had washed away into the sea. But Sfax illustrates the extreme end of the encroachment process. Similar objectives of returning land to use under the waste hierarchy premise of minimisation are in place at other Mediterranean sites such as Huelva, Spain and Gela, Sicily where encroachment is happening but is not yet fully complete.

In the case of Huelva, as can be seen in Figure 41 and 51 the stacks are no more than 100m from the edge of the city. The contents of one closed stack have been used in agriculture and the site itself is now a popular park providing additional open space and amenities for the local urban population. Before the park was opened and in response to public concern about radioactivity, Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas (CIEMAT)[31] and the Universities of Huelva and Seville, in accordance with the specifications of Consejo de Seguridad Nucléar (CSN)[32], the Spanish regulator, monitored the site and the city (Figure 51). Naturally occurring background radiation in the city centre was higher than on the PG stack. At the same time, property developers are keenly aware of the value of the land surrounding the stack and have plans for development for commercial and domestic use.

In Sicily, the Gela phosphoric acid production and PG disposal site was located on a large complex together with other industries such as petro-chemicals. This co-location made the process of characterisation particularly challenging as no detailed records existed concerning what had been additionally disposed of in the PG stack. The closure and remediation process thus involved six different ministries and agencies of the Italian government.

FIGURE 51

Radiological monitoring, Huelva PG stack, close to the city.

All the surrounding land has been or will be developed for housing and commercial use, a process that has already started. But the stack site itself has been re-designated for use for renewable energy generation[33] (Figure 52) and is now producing energy from solar power, the solar units being sited on the stack surface.

31 CIEMAT: www.ciemat.es/portal.do;jsessionid=-7B8155E6744A5ADA96F476FC27006D90?IDM=6&NM=1

32 CSN: www.csn.es/home

33 Report of the solar energy facility at Gela, December 8, 2014, located on the former PG stack: http://espresso.repubblica.it/attualita/2014/12/05/news/gela-il-maxi-progetto-fotovoltaico-si-arena-il-sogno-da-300-milioni-e-diventato-un-incubo-1.190867?refresh_ce

FIGURE 52

The Eni Power Syndial Solar energy facility, sited on the closed Gela PG stack.

9.5 Phosphogypsum as fill for pond reclamation – Canada

The Agrium Fort Saskatchewan Nitrogen Operations site contains PG stacks and cooling ponds inherited from a previous operator. When it was suspected that one of the largest ponds, with an area of about 10,000 m^2, was a source of soluble material moving to groundwater it was decided to reclaim the pond by pumping out the water, filling it and establishing a cover of vegetation (Figure 53)[34].

The only clean fill that was both economically and readily accessible was the nearby PG stack and permission to use it was requested from the provincial government regulators.

Initially reluctant to give approval, the regulator eventually provided a letter of 'non-objection' after about a year of discussion, and reclamation work began in September, 2013. Over 64,000 m^3 of PG from the top of the adjacent stack was placed in the bottom of the pond (Fig-

34 Related Agrium website report: www.agrium.com/en/what-we-do/our-stories/2014-09-23/world-class-research-phosphogypsum

FIGURE 53

Historical phosphate holding pond (before remediation).

FIGURE 54

Filling the pond with PG from the adjacent stack.

FIGURE 55

Covering the compacted PG with soil.

FIGURE 56

Historical Phosphate Holding Pond (summer after filling with PG).

FIGURE 57

Stack remediation – before and after colonisation with saplings, Coromandel, India.

ure 54), and then compacted with heavy equipment and covered with about 15 cm of top soil (Figure 55).

To assess how well vegetation would grow on the heavily compacted gypsum a replicated randomized block experiment was started. The treatments include (1) shallow incorporation of soil into compacted PG with a heavy duty disc, (2) deep incorporation of soil into PG using a ripper blade on a caterpillar tractor, (3) deep incorporation with a ripper blade plus injection of manure pellets into the rooting zone of plants and (4) control – soil which is not incorporated into the underlying PG, in addition two different seed mixes will be tested.

Variables measured include species germination and establishment (Figure 56), biomass production, trace element uptake, water infiltration and changes in groundwater quality.

The purpose was to determine if PG is a suitable, environmentally protective fill material and if deep tillage is necessary for reclamation success on compacted PG. The project is in progress and is expected to be a research project for a university graduate student in 2015-2016.

9.6 Reclamation and restoration – India

Coromandel, India has successfully approached a challenge similar to that addressed by Agrium, how best to place a sustainable green cover over an abandoned PG pond[35]. It imposed on the challenge three constraints:

1. implement without use of soil;
2. optimise water consumption;
3. reduce the dust emissions.

The triggers for the project for Coromandel were a composite of "loss of land, dust emissions, poor aesthetics, nearby community discontent and regulatory pressures". The goal was to "to find sustainable solutions".

Initially PG storage from wet stacking was carried out at a site far removed from any settlement. But over time the company reports that small hamlets grew up around the site and significant numbers of people started living in the vicinity. The wind blew fugitive dust around and the PG storage areas became more and more of an eye sore.

Current production has changed from wet to dry stacking to facilitate the sale of PG into commerce. Two main channels are open, first into the cement industry and secondly into agriculture. But this positive change merely highlighted the need to find a solution to the problem of degraded lands caused by the legacy of wet stacking (Figure 57), a problem the company as well as local stakeholders were keen to resolve.

Initial company-internal attempts to remediate the stacks were not successful. So Coromandel sought partnership with the non-profit Energy Resources Institute, New Delhi (TERI)[36]. Together the partners identified the potential for a two-stage bio-remediation process allow-

35 The Coromandel case study has been kindly contributed by N S Subrahmanyam, Head, SHE, and Amir Alvi, -Executive Vice-President and Head Manufacturing (Fertilisers), Coromandel.

36 Energy Resource Institute: www.teriin.org

FIGURE 58

Sapling planting propagation.

ing saplings to successfully colonise the PG. The first stage comprised a sequence of bio-remediation procedures recommended by TERI that created soil conditions suited to the propagation of saplings that would revegetate the stacking area see Figure 58. Key to this process was the application of mycorrhizal fungi a technique also recommended by the Food an Agriculture Organisation as an "inoculation technique currently suitable in plantation crops and trees"[37]. The second stage established the saplings on the PG stacking site.

When considered from a full cost accounting perspective [21] it is clear that the company sought a solution fully in line with FCA principles. It accordingly reports the benefits in similar FCA consistent terms:

Tangible benefits

1. Direct sustenance to phosphoric acid manufacturing by reducing life-cycle (EoL) costs and providing potential revenue from sapling growth and sale;
2. Resolving the environmental issue created space for future phosphoric acid manufacturing capacities
3. Reclaimed and secured 100 acres of land for business needs;
4. Provided a solution to similar issues faced by Coromandel JVs outside India;
5. Contributed to ensuring food security to the India through sustainability.

Intangible benefits

1. Effective scientific partnership with national independent centre of excellence (TERI) to solve a remediation problem;
2. Improved image among local stake holders;
3. Engaged local communities by providing employment opportunities during bioremediation;
4. Improved aesthetics of the site.

37 FAO advice on mycorrhizal fungi use in sapling development: www.fao.org/docrep/article/wfc/xii/0961-b1.htm

10 Regulatory context, background and trends

KEY POINTS

- Led by the IAEA Safety Report the global regulatory trend is to encourage and even require PG use not disposal to land or sea.
- In those jurisdictions where use of PG has been outlawed or restricted, primarily on radiological grounds, regulators in IAEA Member States have the opportunity to change their regulatory regimes. In some cases action has already been taken with regulators reclassifying NORM residues such as PG as "out of scope" for regulatory purposes.
- Some regulators are outlawing the use of radiological hazard labels on residues with activity concentrations of 1 Bq/g or less because the practice causes unnecessary concern among stakeholders and acts as an unwarranted deterrent to the market.
- This trend is reinforced by the universal progress of the waste hierarchy which treats disposal or any material as a last resort which should as far as possible be avoided
- Recent trends in agricultural science have indicated a significantly enhanced role for sulphur and calcium in soils, PG being rich in both
- There are therefore significant opportunities for "win/win" solutions where regulatory objectives and operational benefits converge.

10.1 Justification, optimisation and proportionality applied to NORM residues

The regulatory framework within which the phosphate industry is operating worldwide in many markets is in a period of rapid change. This change affects both the principles used for setting out the regulatory process and the way they are applied. Nowhere is the change more evident than in the emphasis on the requirement for countries to have a clear strategy and framework for radiation protection and safety, developed in consultation with stakeholders and based on clear outcomes, that the July 2014 revision of the BSS [8] requires. Hence the principles within which the BSS works aligns the classical ICRP principles of justification, optimisation and dose constraints [158], with the modern NORM regulatory policy principles of reasonableness, proportionality and affordability. This translates in practice into a new equilibrium in NORM residue management between benefit and risk. This affects all desired outcomes by requiring that they be "commensurate" with risk, including decontamination and remediation challenges:

1. The government and the regulatory body or other relevant authority shall ensure that the protection strategy for the management of existing exposure situations, (...) is commensurate with the radiation risks associated with the existing exposure situation; and that remedial actions or protective actions are expected to yield sufficient benefits to outweigh the detriments associated with taking them, including detriments in the form of radiation risks.
2. The implementation of remedial actions (remediation) does not imply the elimination of all radioactivity or all traces of radioactive substances. The optimization process may lead to extensive remediation but not necessarily to the restoration of previous conditions [8].

10.1.1 Regulatory implementation

In terms of the graded approach to regulation (Figure 7), the nature and extent of such measures will be commensurate with the type of practice and the levels of exposure, and entail the establishment of radiation protection

programmes, with suitable provisions for monitoring and dose assessment, within an agreed protection strategy.

10.1.2 Regulatory trends and policy drivers

While each regulatory jurisdiction and each market has its own specific policies, procedures and objectives, there are many similarities emerging between countries where there are fundamental changes in the regulatory framework for NORM industries, including phosphates. The range of countries is diverse including for example, Brazil, China, India, South Africa, United Kingdom and Sweden.

Among the more significant similarities are:

1. Commitment to change on the part of the regulator to rebalance environmental, social and economic objectives, based on agreed "commensurate" outcomes;
2. Complementary commitment on the part of company leaderships to effect operational change in the wider context of sustainability and resource conservation;
3. Engagement with stakeholders at each step to maximise acceptance and to win and keep the social licence to operate;
4. Recognition of the role of the market in achieving financial sustainability;
5. Removal of unnecessary or unjustified barriers to use, including the designation of all residues at or below 1 Bq/g activity concentration as "Out of Scope" even to the point of forbidding the use of the radiological hazard label on such materials so as not to deter the markets and to raise fear among stakeholders.

These changes are not just evident at national level but are also framing the policies and procedures of key international bodies such as IAEA and ICRP. ICRP for example is highly conscious of the risk to trade of inappropriate types of NORM regulation, as seen in the discussion in ICRP 104 [159] of commodities containing NORM, such as phosphate ores or fertilizers, and the need to avoid artificial barriers to trade. Section 7.5 acknowledges that some countries may be tempted to use this as a means of justifying trade restrictions [160] and in paragraph 177, notes that:

1. natural background exposure causes annual doses of at least a few mSv/y and, taking account of possible annual doses from authorized practices, this leaves an upper bound of the order of a few mSv/y for annual doses from all commodities to be exempted from intervention[38]. It is not likely that several types of commodities would be simultaneous sources of high exposure to any given individual [160].

38 Such a dose corresponds to a level risk below the USEPA's 3 in 10,000 value established for PG use.

10.1.3 Pathway to phosphogypsum use

From all these common factors it is possible to discern a reasonably predictable pathway to use of PG (Table 14) if the following steps are respected:

1. Follow the IAEA Safety Report and related framing documents such as BSS and ICRP;
2. Redefine PG as a co-product or by-product or re-categorise PG into a non-hazardous class;
3. Issue a positive guidance note from the NORM (or responsible) regulator;
4. Conduct field or demonstration trials under peer reviewed conditions, notably for agriculture and housing;
5. Engage stakeholders e.g. farmers, future residents;
6. Stimulate the market through measures such as deregulation and emphasis on affordability.

In India and Brazil these steps have been followed. Similar though less coordinated strategies have been used in South Africa. In Belgium, Poland and the UK need to responsibly preserve jobs in critical NORM industries has caused a major shift in policy away from rigid insistence on numerical values as applied to very low level wastes and residues towards science-based decision-making balancing economic and environmental considerations.

In sum, if the major parties cooperate based on common values and with common goals, socially accepted change is achievable, resulting in direct stimulus to substantial levels of PG use.

10.2 Health, safety, environmental (HSE) considerations

10.2.1 Health and safety

Specific radiological measures in the workplace such as control of the occupancy period or even shielding may sometimes be appropriate to minimize external exposure to NORM. Materials with relatively low activity concentrations give rise to modest gamma dose rates (typically no more than a few mSv per hour), even on contact. In such cases, discouraging access, for example by storing materials in mostly unoccupied areas, may be sufficient. In areas containing materials with relatively high activity concentrations, physical barriers and warning signs may be necessary.

Exposure to airborne dust is likely to be controlled already in many workplaces through general occupational, health and safety (OHS) regulations and good practices such as keeping the PG at a specific moisture level to negate fugitive dust formation Control of the air quality for the purpose of minimizing dust levels may also help to reduce radon and thoron concentrations. Therefore, the extent to which existing OHS control measures are effective

TABLE 14

Regulatory context.

Country/ regulatory body	Approach	PG use/disposal	Driver	Trend
Brazil (CNEN)	· Graded (follows IAEA) · Evidence- and risk-based	· Use in agriculture and construction	· Regulator position paper encouraging use (2008) · Subsequent change of regulations to allow/encourage use. · Agricultural field studies showing value-add and safety	· Equilibrium between domestic production and consumption (2014) · Remediation/use of legacy stacks for land recovery (City of Uberaba)
Belgium	· Graded (follows IAEA) · Evidence- and risk-based	· Long history of commercial use (80-90% of production) · Limited disposal - stored for potential future use	· Regulator respecting equilibrium between environmental and economic considerations, eg permit for unlined stack	· Science-based regulatory decision-making
Canada	· Graded (follows IAEA) · Evidence- and risk-based	· Small scale experimental uses (crops and livestock) · Innovative remediation	· Regulator issued "No Objection" notice	· Broader-based use
China	· Graded (aligned to IAEA) · Evidence- and risk-based	· Wide range of uses	· Environmental and social policy · Green Mine policy and awards	· Minimum use level – 20% from 2015 – 30% from 2025 · Zero waste goal
IAEA	· Graded · Evidence- and risk-based	· Use preferred to stacking, discharge to sea or disposal	· Basic Safety Standards · Safety Report 78	· Zero waste goal
India	· Graded (aligned to IAEA) · Evidence- and risk-based	· Agriculture – unrestricted · Construction – some restrictions	· Regulator authorisation (2008) · Agricultural field trials demonstrating value add and safety	· Value add uses, eg affordable sulphur rich fertilisers · Reclamation of legacy stacks
European Union	· Graded (follows IAEA) · Evidence- and risk-based	· Sustainable market participation = commercial solution	· Waste hierarchy · Removal of obstacles to market participation · Local variations	· UK to outlaw labelling NORM residues at 1 Bq/g or less as radioactive
Philippines	· Graded (follows IAEA)	· Use in cement and agriculture (~50% of production)	· Market factors	· Growth in agricultural use
Poland	· Graded (follows IAEA) · Evidence- and risk-based	· Early indications of market interest for agricultural use	· Regulator respecting equilibrium between environmental and economic considerations, eg permit for stack extension to safeguard production	· Science-based regulatory decision-making
Tunisia	· Graded (follows IAEA)	· Experimental uses in construction and agriculture · Discharge to sea being phased out	· Regulators (APNE) in process of changing the regulatory framework · Compliance with Barcelona · Convention and LBS discharge protocols	· Social acceptance remains to be renegotiated
Morocco	· Graded (follows IAEA)	· Developing "valorization" strategy for use	· Encouraging results · Experimental uses in agriculture and road construction	· New regulatory framework
South Africa	· Graded (follows IAEA)	· Experimental uses in agriculture, road building and affordable housing	· Regulations modified to encourage use	· Regulatory regime under review

in minimizing workers' radiation exposure is something that the regulatory body would first need to establish before deciding to impose additional control measures for purely radiological reasons. In some workplaces, existing OHS control measures alone may provide sufficient protection against internal exposure. In other workplaces, additional control measures specifically for radiation protection purposes may become necessary for achieving compliance with the Standards. Engineered controls are the favoured option, with working procedures and, finally, protective respiratory equipment being considered only where further engineering controls are unlikely to be effective or practicable.

Complete containment of material is often impractical, especially where large quantities of low activity concentration materials are involved and spills and the spread of materials outside the area are often of no radiological significance unless substantial and persistent airborne dust levels result. Prevention of re-suspension of dust is therefore likely to be the most effective approach. Specific measures to control surface contamination only become meaningful where materials with higher activity concentrations are present.

Worker awareness and training are particularly important for supporting the introduction of local rules and for creating an understanding of the precautions embodied in such rules. The work practices of individual employees may exacerbate dust generation and, in some cases, may completely negate the effect of any engineering controls installed. There may be deficiencies in the way in which equipment maintenance tasks are undertaken, implying the need for periodic review to determine if improvements are possible. The general standard of housekeeping and spillage control also needs to be kept under regular review. Even where the materials being handled have a low activity concentration, a reasonable standard of housekeeping may be necessary to ensure that dust re-suspension is adequately controlled.

10.2.2 Phosphoric acid and phosphogypsum as co-products – radiological and environmental considerations

The mining and beneficiation of PR involves mainly physical processes, and is therefore unlikely to affect the equilibrium between the components that generally exists in the ore. However, chemical or thermal processing of PR will mobilize the ^{238}U and ^{232}Th decay series radionuclides contained in the ore. This causes different radionuclides to migrate in different ways, thus destroying the equilibrium condition. Knowledge of the resulting radionuclide compositions of the various process materials and products is essential for determining the nature of any control measures that might be needed to ensure that the workers and members of the public are adequately protected.

As an illustration of radionuclide migration during the processing of PR into fertilizer, a mass balance and a radionuclide balance for a wet-process facility processing sedimentary rock are available in the IAEA Phosphate Industry Safety Report [12]. During the digestion of the rock with sulphuric acid, the uranium contained within it migrates primarily to the phosphoric acid, whereas most of the radium ends up in the PG. The remaining radionuclide composition of the phosphoric acid including the uranium is, in turn, carried through to the various fertilizer products derived from the acid.

10.2.3 USA-specific considerations

Phosphate fertilizer production has a long history in the United States that predates modern Health, Safety and Environment (HSE) concerns and practices. Its rapid development in the USA was focused on the wet process which came to dominate the world production for many decades. As radiological, environmental, and workplace regulation evolved in the USA, regulations were copied to various degrees in other countries. Consequently, the US regulatory framework for a long time was the *de facto* starting point for other regulatory regimes. As shown in this Report other national regulators have set revised or different limits and clearance levels according to their own risk analyses and national priorities, including a significant increase in the use of the "out of scope" classification for low level NORM co-products and by-products.

Under the USEPA Rule, PG is regulated according to its ^{226}Ra activity concentration, i.e. this single radium isotope and the fact that it is the precursor to ^{222}Rn, commonly known as radon gas. High concentrations of radon gas, together with its radioactive progeny, have been linked to lung cancer and it is assumed to have a proportional risk at low concentrations. However, the risk due to chronic low radiation doses from exposure to low concentrations of radon is not based on direct evidence.

10.2.3.1 The Linear No-Threshold Hypothesis

Under advice from UNSCEAR (2014) the international radiation protection community has advised against using the Linear No-Threshold (LNT) hypothesis for assessing the risk of cancer from human exposure to ionizing radiation. The dose-response model uses a straight line relationship between very high doses (such as people exposed to radiation from a nuclear fission reaction) and low doses (such as people receive from radon in their homes). This means that for every increment in dose, no matter how small, the model assumes an increased risk for some form of cancer. This simple, linear relationship ignores biological repair mechanisms that are very effective and efficient at low doses and low dose rates of ionizing radiations. Consequently, the effect of low doses and low rates may be substantially below what would be predicted by the linear model meaning that the

risk is much lower. Radiation is a very weak carcinogen, so the most probable outcome is that the health effect would be negligible or at least indistinguishable from the background incidence. Some regulatory regimes recognize this situation and the lack of any evidence of harm when they set levels for clearance of NORM.

10.2.3.2. The Clean Air Act

In the USA, the Clean Air Act requires the USEPA to regulate airborne emissions of hazardous air pollutants (HAPs) emitted from industrial "source categories" specified in their list. Radioactive isotopes of elements (radionuclides) are included in the HAPs. Each source category that emits significant quantities of radionuclides must meet control technology requirements and regulatory limits derived according to the risk represented by each radionuclide. These regulatory limits are the National Emission Standards for Hazardous Air Pollutants for Radionuclides (commonly known as the Rad NESHAPs).[39] Subpart R regulates emission of radon from PG stacks. Any approved use of PG must result in no greater risk to the public or environment than stacking (set at 3 in 10,000 risk of mortality). However, approval for use is difficult to receive. Also, Florida lawmakers passed a financial responsibility requirement whereby each stack operator must either set aside adequate funds or show adequate financial strength on their balance sheet for eventual stack closure.

Multiple HAPs are targeted, but the primary concern is hydrogen fluoride (HF) emissions from the stacks, which the stack size limitation is intended to contain. Consequently, new pressures have now been exerted for non-radiological reasons.

The USEPA allows only limited use of PG in agriculture if it averages less than 0.37 Bq/g ^{226}Ra. Other countries allow clearance of NORM at higher levels. For example, under the current UK regulations PG under 0.5 Bq/g has historically been considered non-radioactive, or outside the scope of the regulations[40], a level which under the 2014 NORM Strategic Framework is raised to 1 Bq/g [22] with exemptions possible up to 5 and even 10 Bq/g.

Disposal is not the same as an application, which would be reviewed for concentration potential and human exposure pathways. Products made with PG would not be "wastes" but rather "materials". Most products are mixtures of materials and would be very likely to fall below the same 0.5 Bq/g criterion at which they are also considered non-radioactive. The UK adopted an exemption level of 5 Bq/g for disposal of NORM waste and there is even a provision to extend this up to 10 Bq/g, but this requires a specific radiological assessment to demonstrate that the dose constraint targets are met. At 5 Bq/g, there is no published evidence that any PG ever produced in the world would not qualify as exempt. But note in detail, it is more complicated than this and there are other conditions imposed on exemption such as summing for the total activity to be disposed. In Scotland there are annual disposal limits on exempt waste by landfill site, e.g. 50 TBq/year. The Swedish radiation safety authority (SSM) has issued a new regulation on exception and clearance of NORM (SSMFS 2011:4) with a level of 10 Bq/g ^{226}Ra.

These levels were chosen because they correspond to some range of doses and risk. Nobody is "wrong". It depends on the basis for choosing the level. If the goal is to limit risk to < 1 in 3 in 10000 as is the case of the USEPA, then the corresponding activity concentrations will be low where the LNT model is applied. If the risk assessment relies more on physical evidence of harm, such as excess cancer mortality at low doses, such evidence will not be visible because the background incidence of cancer is enough to overwhelm and mask it. Consequently, regulators may establish higher risk levels that correspond to higher activity concentrations that, nevertheless, are still in the range of undisturbed NORM in the environment.

10.3 Environmental good practices for phosphogypsum use

New policies concerning resource conservation and critical materials and changing emphasis on regulatory control are becoming more important for environmental regulation in general and PG use in particular. These environmental policies require increased regulatory and stakeholder attention to:

1. Fertilizer use efficiency, maintaining nutrient inputs to crops and soils with just sufficient nutrient to deliver optimised yield and quality, with no run-off or avoidable leakage to the environment (the right fertilizer at the right time and in the right quantity);
2. Risks from point and area source pollution and eutrophication of surface fresh water when excessive amounts of phosphorus (P) are transferred from soil to water;
3. Observation of the waste hierarchy (Figure 2) that disposal of wastes to sea or land is the option of last resort and only to be accepted when all options for beneficial use have not met the criteria outlined above in Section 1.4.6;
4. Disposal permits can only be granted when all beneficial uses have first been explored and been found to be not viable for technical and/or economic reasons;
5. Environmental impact assessments (EIAs) which are mandatory for any such project as widespread PG use are now being extended in scope to include mandatory Safety Cases and the inclusion of social factors in EIAs, which are now termed ESIAs;

39 www.epa.gov/radiation/neshaps/
40 www.defra.gov.uk/publications/files/pb13624-rsl-guidance-110914.pdf

6. Recovery of phosphates from urban wastewaters and their substitution into the fertilizer production process instead of primary resources.

10.3.1 The principle of waste minimisation – the Prayon PG Stack, Belgium

As the Prayon case study demonstrates, despite best efforts to achieve a zero waste outcome, at least in regard to marketable residue, some of the PG produced does not meet market specifications and would be disproportionately expensive to remediate for sale. Hence the NORM policy principles consinue to accept that such materials can be stacked with no forseen plan for use. The design of a PG stack for materials of this type must ensure the prevention of pollution of the soil, groundwater or surface water and provide for efficient collection of leachate. This results in most cases in a regulatory requirement for the construction of a bottom liner while the stack is in operation and closure and capping with a second liner covered by top soil when the stack is no longer used. Liquid and gas drainage systems are also required. But in the case of the stack at the Prayon production facility in Belgium, the leaching tests at different pHs showed little release of metallic cations and other impurities into the environment and hence simple storage on the ground complied with acceptable limits established by the Belgian authorities. This eliminated the need for a bottom liner because of the very low level of impurities in the leachates; and it was further concluded from the risk assessment that it would be environmentally preferable not to use a liner in case at some future time the liner might be breached at a single point which could lead to a possible concentration of impurities in the leachate at the breach point [160].

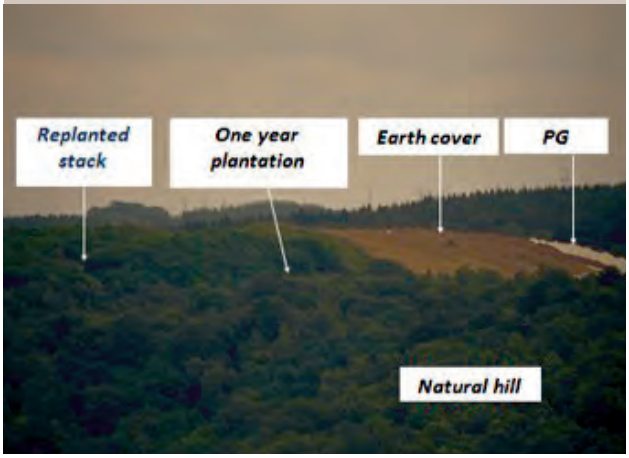
In respect of environmental monitoring, to ensure that the environmental impact remains low over time, several wells were dug all around the stack site. The water collected in the wells is analysed for elements or molecules that might leach from the stack. The results to date have shown that leachate from the stack is hardly detectable and hence well below Belgian national action levels.

Draining and runoff waters coming from the hill surrounding the landfill, whether from the stack or from pore water leaching out from the periphery of the stack, are collected into pipes. The water flows through these pipes naturally down hill into the Meuse river. To reduce the visual impact of the stack, the decision was taken not to cover it with an impervious cap but instead to mask it with a 1.5 m thick soil layer. To ensure the soil layer is fully compatible with the surrounding environment approximately one meter of soil is removed before the PG stack site is commissioned. This soil is then stored close to the stack and is finally to cover the stack once it has reached its final permitted height and is closed.

The soil is then planted with trees and shrubs transforming the stack in a “hill between the hills”. After a few years the results are impressive; the stack has blended in with its surroundings (Figure 59).

FIGURE 59

Revegetated closed stack integrated into the natural surroundings, Belgium.



10.4 Social factors

As the value of PG as a co-product becomes more widely recognized and it is used more widely, using PG will have an increasing social impact. For example, when used as a soil amendment in agriculture to improve the productivity of degraded and sodic soils, as trialled in Kazakhstan [35], food security and the livelihoods of those farming such soils are improved. When, as in China, using PG for making high-strength, thermally efficient construction materials demonstrably reduces the cost of building materials then a highly beneficial outcome is increased affordability of quality housing.

As highlighted in The IAEA Safety Report [12], concerns about the environmental impact of stacks have grown since the 1980s with problems such as slope failures, sink holes and acidic water spills causing considerable damage. The more people encroach on areas used for PG stacking the greater the potential for danger to their lives and property. There are clear benefits if the stacks are reduced in size and/or eliminated by use, as shown in Section 9.

10.5 Phosphorus life-cycle efficiency and comprehensive extraction

Policies related to sustainability and resource conservation have favoured the emergence of a new model for mining and processing, termed “comprehensive extraction” (CX), which takes as a voluntary that mining should only disturb the ground once [161] and that all resources found in the ore bodies mined should be recovered in a single process. This approach is attractive when winning, and

keeping, a social licence to open a new mine is becoming increasingly difficult. Comprehensive extraction seeks a strategic, long-term approach to resource extraction and processing rather than focusing on a single commodity. This has implications for the way both primary (phosphate rock) and secondary (PG) resources are assessed and extracted. One outcome from this approach is the emergence of concepts such as "energy basin management" [162] where the resources of a whole sedimentary basin, that might include coal, oil and gas, uranium, phosphates and rare earths, are managed as a single complex resource rather than as individual target minerals.

For the phosphate industry, the concept of comprehensive extraction includes the prospect of reintroducing uranium (U) extraction facilities at phosphate facilities, but also facilities for capturing other minerals such as rare earth elements (REE) or thorium. A further, highly significant attraction of comprehensive extraction [163] is the environmental benefit because the ground is only disturbed once and may not need to be disturbed elsewhere. For example, every tonne of U that can be extracted from PR offsets U that has to be extracted by conventional mining at another location. At a time when stakeholder acceptance of mining is declining worldwide, the environmental benefits of this approach can weigh heavily in deciding whether or not to allow a new phosphate mine. Projects such as Santa Quitéria in Brazil show that the comprehensive extraction model is starting to gain acceptance and open up phosphate deposits previously considered uneconomic for commercial use.

10.6 Research and development in phosphogypsum uses

Table 15[41], covering 2010-present, shows that research papers on PG uses continue to be published in large numbers as the trend to find beneficial uses and to address radiological impacts grows. The primary focus for use is construction. The primary concerns are radiological and environmental risks.

Although not categorized as such, other publications, including four of the six in Analytical Methods, are radiological in nature. PG Properties followed by Purification and Impurities are the next largest categories, respectively. They can be considered related because these are categories of interest to those attempting to use PG. Decomposition is tied with Purification and Impurities for similar reasons. Tied for fifth place are Environmental Impacts and Agriculture – Soil Amendment. Agriculture combined comprises the fourth largest category. Surprisingly, there were only 5 publications on roads perhaps because this use is well established. Some uses in agriculture and roads are seasonable and cannot be relied on to consume large volumes on a regular basis.

41 Data compiled by the FIPR librarian Karen Stewart

TABLE 15

Published phosphogypsum research references 2010 - 2015[41].

Category	Number of references
Analytical Methods	6
Binders	7
Coatings	1
Crystallization	1
Decomposition (eg for sulphur recovery)	16
Disposal	2
Environmental Impacts	12
Filtration	3
Heavy Metals	7
Hydration	2
Leaching & Leachate	4
Properties	29
Public Health Impacts	2
Purification & Impurities	16
Radiological Impacts	32
Recovery (secondary resource mining) – ammonium sulphate, calcium, calcium carbonate/ lime, calcium sulphide, phosphate, potassium sulphate, Rare Earths, sulphur, sulphuric acid, sulphur dioxide, trace metals	24
Refining	2
Remediation – Arsenic, Cadmium, Chromium, Copper, Lead, Zinc	9
Stacks	2
Stacks – Design	2
Uses – (Comprehensive Utilization)	2
Uses – Acid Mine Drainage	1
Uses – Agriculture – Soil Amendment	12
Uses – Agriculture – Soil Amendment – Sodic Soils	7
Uses – Agriculture – Soil Amendment – Soil Acidity	3
Uses – Biomaterial	1
Uses – Construction	55
Uses – Emission Control	1
Uses – Fertilizer Manufacture	3
Uses – Filler	5
Uses – Filtration Aid	2
Uses – Fluoride Sorbent	1
Uses – General	8
Uses – Liner Material	1
Uses – Marine Applications	2
Uses – Roads	5
Uses – Soil Amendment	1
Uses – Sulfate Reduction	2
Uses – Water Treatment	1
Total	300

The appeal to producers of construction materials is relatively simple to explain. Whether the construction application is in-house (such as offered by Wengfu Group[42]) or outsourced e.g. to a cement manufacturer, this use works on the basis of a predictable and regular off-take agreement consuming large volumes (more than 1mt/year) of PG.

10.7 Stakeholder engagement and communications strategy

Stakeholder engagement and communications are rapidly becoming a point of convergence between good practice, corporate social responsibility and regulatory obligation. From a corporate point of view, close engagement with stakeholders and shared values are fundamental to preventing reputational damage that can arise from the perception that the phosphate industry is a polluting industry. But this does not just affect industry. These are also now mandatory requirements for governments following the IAEA Basic Safety Standards (BSS) [8]. The BSS makes stakeholder engagement and social acceptance a key part of the development of the framework for radiation protection and safety.

A key part of the social acceptance process involves identifying at an early stage who the stakeholders are and how they are best approached. For example, social acceptance by the farming community in India for the use of PG as a soil amendment was achieved by educating farmers' groups from the outset in the advantages that PG had for them both in regard to its qualities and affordability. Farmers were directly involved in field trials with widely-grown crops to show the magnitude of the response that could be achieved. Similar experience was gained in Kazakhstan and Syria.

Such practices are part of a wider movement among mining and extractive industries to win and keep a "social license to operate" (SLO). This concept was first introduced by the industry in 2002 in the landmark publication "Breaking New Ground" [164]. Its adoption has been so widespread that not only has it become a fundamental good practice, but it is also seen by leading consulting firms such as Ernst and Young [165] and KPMG [166] as a core requirement for managing business risk in large-scale investments such as characterize the phosphate industry. Hence the SLO is a key way in which the industry is able to mitigate or eliminate the risk of reputational damage. A similar message is conveyed by the report: "Extracting with Purpose: Creating Shared Value in the Oil and Gas and Mining Sectors' Companies and Communities" [167]. As Michael Porter comments in his foreword:

1. Aligning the business interests of extractives companies with community needs and priorities is the only real solution for companies and communities alike. The root causes of community strife are lack of economic opportunity, poor health, lack of effective local or national governments, and environmental degradation. These issues are fundamental to business success due in part to the very long time horizons of oil and gas and mining operations and the deficits in the regions where these companies operate. Companies must tie community prosperity to the present long-term needs of the business in areas such as a qualified labour pool, capable suppliers, and well-functioning community infrastructure.

The findings apply as much to phosphates as to oil and gas.

42 Wengfu Group: www.wengfu.com/main.htm

11 Conclusions

The search for a sustainable solution for PG as a useful resource not a burdensome waste has deep roots in IFA, beginning with the presentation by Armand Davister at the 1998 Technical Conference in Marrakech [168]. In 2000 Paul Smith and Tibaut Theys showed the IFA community how PG could be used profitably [31]. In 2007, the FIPR "Stack Free" project, to which Armand Davister contributed and with encouragement from IFA, identified four conditions that would need to be satisfied for a new point of equilibrium to be reached for sustainable PG management and use as a co-product [169]. These were:

1. Technical feasibility;
2. Regulatory acceptance;
3. Commercial viability;
4. Policy desirability.

The "Stack Free" project further concluded that any solution would be "regional" in nature given the high degrees of variability in production, regulatory, market and policy conditions affecting operators around the world [170].

This Report has shown that all four conditions are being met, but in varying degrees, at varying speeds, and on a regional basis – as anticipated.

It is not the intention of the Report to argue that PG must be used, simply to show on the basis of a risk- and evidence-based review that it can be used, and used safely, assuming careful and accurate characterisation, appropriate selection of uses according to social, environmental and economic considerations and adherence to a "graded approach" which takes into full account the presence of trace quantities of naturally occurring radionuclides and of heavy metals.

Because the primary challenge posed by PG is its very large volume the emphasis on use is likely to remain in three broad areas, agriculture, construction and mine restoration. Of these, mine restoration may be the least favoured as it provides little or no added value, but has the considerable merit in the context of sustainable development goals of leaving no legacy wastes behind. One producer, Prayon, has achieved 80-90% uses over many years, focused on supplying PG into the construction industry; another Kailin, has reached 100% use with a combination of all three large volume strategies, of which 60% is in value-added activity in agriculture and construction; another PCS has long achieved 100% use through mine restoration. These high levels of use remain exceptions when viewed across the industry as a whole, but the global trend is now positively towards use, encouraged by some influential international and national policies and regulatory agencies. As of the year of first publication of this report, global reuse or recycling will reach some 20-25% of production and the upward trend towards increasing use is likely to continue.

A combination of powerful environmental, market and policy factors will stimulate further growth and with it potentially the development of new practices and products. Of these perhaps the most powerful is the UN led initiative to reverse the highly damaging trend, notably in irrigated agricultural areas, towards increasing salinity in soils. If this trend cannot be reversed the goal of feeding a global population of 9.5 billion by 2050 is seriously at risk. As this Report shows, PG has a potentially very major role to play in reversing this trend, with the added benefit of significantly enhanced efficiency of water use in crop production. Agriculture presents a second major opening for PG notably in addressing the problem of sulphur deficiency in soils. Some producers are already developing new specialised PG products, some with added micronutrients and in granulated form, to meet demand for affordable, agronomically efficient sulphur rich fertilisers.

While such initiatives demonstrate there is much that can be done following a path of optimisation of current knowledge and practices, the vigorous state of research and development concerning PG as evidenced by the many papers published on the topic and the numerous patents for specific applications indicates that "disruptive" technologies are also likely to emerge that either prevent PG production in the first place by modifying or replacing the "wet process" production of phosphoric acid, or which progressively embed PG in the circular economy, as for example through sulphur recovery and reuse as an energy source.

In seeking such innovations, there will many opportunities for collaboration between industry, government and centres of scientific and technical excellence. Out of such a collaborative approach it may also be possible to develop a coherent and consistent policy and practice of sustainable PG use worldwide.

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