CONCEPTUAL MODEL ELABORATION FOR THE SAFETY ASSESSMENT OF PHOSPHOGYPSIUM USE IN SANITARY LANDFILLS

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ABSTRACT

Phosphogypsum is a by-product of the phosphatic fertilizer production from the beneficiation of phosphate minerals (apatites). Produced in large quantities throughout the world and stored temporarily in stacks, the final destination of this product is nowadays a subject of investigation. Due to the presence of radionuclides ($^{226}$Ra, $^{232}$Th and $^{40}$K, mainly), possible applications for the phosphogypsum must be verified for radiological safety. The goal of this paper was to elaborate a representative water flow conceptual model of a sanitary landfill for the safety assessment of the impact of using phosphogypsum as a cover material. For this, the ground water flow in variably saturated conditions and solute transport model HYDRUS-2D has been used for simulating the impact in the saturated zone of potential radionuclides leaching. The conceptual model was developed by collecting and analyzing the data from environmental license documentation of municipal sanitary landfills located on the State of Minas Gerais, Brazil. In order to fulfill the requirements of HYDRUS-2D model in terms of the necessary parameters, the physical characteristics and typical configuration of the landfills, as well as the hydrogeological parameters of soils and aquifers related to the local of placement of the landfills, were taken in account for the formulation of the conceptual model.

1. INTRODUCTION

Earth surface contains naturally occurring radioactive materials (NORM), such as the radionuclides from $^{235}$U, $^{238}$U and $^{232}$Th decay chains. However, some industrial processes may cause the concentration of NORM materials within the final products or wastes, creating the so-called TENORM (Technologically-Enhanced Naturally Occurring Radioactive Materials). These processes include mining and processing of minerals and sand, fertilizer production and its utilization, fossil fuel burning, petroleum extraction (tube incrustations and oily wastes) and refining of metals. In particular, it can point the mining and processing of uranium minerals and the production of phosphate fertilizers [1].

The most used process for phosphate fertilizer production is the phosphatic rock attack with concentrated sulfuric acid and water. In this case, the main products from chemical reactions are phosphoric acid, simple superphosphate (SSP), and triple superphosphate (TSP). As wastes from processing, phosphogypsum (dihydrate calcium sulfate) and hydrofluoric acid are produced [2].

At present, this material is stored in piles located near the fertilizers factories. Even though phosphogypsum is composed mainly from dihydrate calcium sulfate, it can present elevated levels of impurity from the matrix rock. These rocks transfer insoluble or acid soluble
impurities to phosphogypsum, such as heavy metals (Cd and Zn), metalloids (As and Se), fluorides, and natural series radionuclides [3]. As a consequence, phosphogypsum disposal in piles may result in environmental impact as leachate and superficial drainage of toxic elements, fluoride, heavy metals, sulfates, and radium ($^{226}\text{Ra}$ and $^{228}\text{Ra}$ in soluble form) can result in contamination of water bodies, as well as direct gamma irradiation from piles (harmful to workers), release of aerosol caused by eolic erosion of the pile, and $^{222}\text{Rn}$ gas inhalation (also harmful to workers) [4,5].

The rate of phosphogypsum generation is of approximately 4.8 tons for each ton of produced phosphoric acid. Worldwide annual production can be estimated in 150 million tons. From this total, around 12 million tons come from Brazil [6], and most of it from Minas Gerais State, Uberaba region. Thus phosphogypsum reutilization would contribute to preserve natural gypsum reserves, guaranteeing preservation of natural resources; a fundamental principle of sustainable development.

Most of radionuclides contained in NORM materials belong to decay chains that start with $^{235}\text{U}$, $^{238}\text{U}$, and $^{232}\text{Th}$. However, during the past years, many industries increased the proportion of NORM materials in final products or wastes, generating TENORMs (Technologically Enhanced Naturally Occurring Radioactive Materials). Such activities include general and sand mining and milling operations, fossil fuel burning, incrustation and sludge from petroleum extraction, metal refinement, uranium mining and milling, as well as manufacturing of phosphate fertilizers [1].

Several studies have been carried out all over the world in order to find applications for phosphogypsum, such as the usage as fertilizer and soil conditioner, as base layer on the road pavement, cover layer in sanitary landfills, production of gypsy plaques for ceiling cover, and as cement additive [4]. In Brazil, this material has been used for decades as a calcium and sulphur source, soil conditioner and for the correction of soils saturated with sodium and potassium. However, the phosphogypsum production rate is still much larger than the amount necessary for agricultural uses.

One of the most promising usage alternatives in terms of phosphogypsum production and utilization balance is its application in sanitary landfills. In this case, the sulfate-enriched phosphogypsum can be used in the anaerobic (oxygen depleted) environment, such as landfills, to enhance microbiological processes to decompose municipal solid waste, and thus, extend the lifetime of landfills [7].

There is good evidence to suggest that the addition of phosphogypsum could enhance biodegradation of municipal solid waste (MSW) in the landfill. During the early stages of waste decomposition in a landfill, the degradation process is essentially aerobic i.e., with available oxygen), and carbon dioxide is the principal gas produced. As oxygen in the landfill is depleted, the decomposition process becomes anaerobic, and other gases, principally methane, are generated in significant quantities. In an anaerobic environment, bacteria depend on bound oxygen to serve as electron acceptors in oxidation-reduction reactions. Since the phosphogypsum is enriched with sulfate, it is reasonable to assume that a sulfate using bacterial colony present in landfills will use phosphogypsum as an energy source after oxygen is depleted. It is therefore anticipated that the use of phosphogypsum as landfill cover will enhance biological decomposition of MSW and at the same time reduce the
accumulation of phosphogypsum and the volume of cover material remaining in a MSW landfill [7].

However, the presence of heavy metals and radionuclides in the phosphogypsum requires that any new usage proposal must be preceded by the elaboration of safety and risk analysis to evaluate its environmental and human health impact associated to that particular practice. In the case of waste disposal in landfills, this study is performed using hydrogeological models that describe the temporal and spatial behavior of the contaminated plume, subjected to the solute transport phenomena (advection, dispersion, natural attenuation, etc).

The methodology for the development of a mathematical model for simulating hydrogeological processes includes several steps, as follows [1]: (1) elaboration of a representative conceptual model, including hydrogeological, hydrological, and geological characteristics, among others; (2) selection of the computational code; (3) model design that aims to set the conceptual model in a format suitable for being modeled; (4) model calibration to certify that the model predictive capability; (5) model verification to increase the confidence degree of the model; (6) application of the model to predict the behavior of the system to future events or scenarios. The conceptual model is the model specification and the aim is to choose the model that will give the best overall performance for the project.

This paper aims to present and discuss the initial steps of the development of a mathematical model for the simulation of the hydrogeological processes related to the application of phosphogypsum as cover material in a sanitary landfill. For this, it is described the criteria for the elaboration of a representative conceptual flow model for a sanitary landfill, as well as for the selection of the computational code for the analyses.

For modeling the landfill, flow and transport simulations will be considered as decoupled, meaning that flow simulation will be made prior and separated from transport simulation. The safety and risk analysis will take place based on scenario simulations for certain exposition pathways (water ingestion, intrusion, etc).

2. GENERAL CHARACTERISTICS OF A SANITARY LANDFILL

Landfills must be designed to protect the environmental from leachate contaminants. An ideal landfill project should avoid the location on environmentally sensitive areas and to foresee a preventive monitoring system.

A Brazilian law [8] enforces that municipalities must have a suitable disposal system for urban solid wastes. The article 2 requires that all municipalities, independently of the size, must accomplish the following requirements:

- To dispose the waste in soil or rock with low permeability, declivity lower than 30%, good accessibility, minimum distance of 300-meters from surface water bodies and 500-meters from population bodies, away from roads, erosions and permanent preservation areas;
- To implement a pluvial drainage system within the whole landfill area, to minimize rain infiltration in the waste area;
- To compact and cover waste with soil or debris;
To isolate the area with a fence and bushes or trees to make the access difficult for people and animals;

To forbid people from remaining at the waste deposition area for waste collection. The municipality is responsible for providing suitable alternatives in technical, sanitary and environmentally-friendly perspectives to allow recycling activities.

Still according to this regulation, waste must be covered with soil with the following frequency:

- Municipalities with urban population lower than 5,000 inhabitants – at least, once a week;
- Municipalities with urban population between 5,000 and 10,000 inhabitants – at least, twice a week;
- Municipalities with urban population between 10,000 and 30,000 inhabitants – at least, three times a week;
- Municipalities with urban population greater than 30,000 inhabitants – daily.

3. METHODOLOGY

The proposed study was based on the development of three steps, as follows:

1) Data collection: Landfill project data and location-related information from licensing processes of the Minas Gerais State environmental authority (Fundação Estadual do Meio Ambiente – FEAM) were reviewed for selecting a specific landfill to be used as reference for the conceptual model elaboration. Sanitary landfills located at the Triângulo Mineiro area were selected since the larger Brazilian fertilizer industry is sited at Uberaba. Among them, taking in consideration the logistics required for the transportation of large amounts of phosphogypsum, necessary for the daily operation of waste covering in large municipal landfills (more than 30,000 inhabitants), Uberlândia’s municipal landfill was selected as the reference landfill for the safety analysis to be accomplished.

2) Flow conceptual model elaboration: the Uberlândia Sanitary Landfill’s main physical characteristics were evaluated for the definition of the basic dimensions and water flow across the landfill. This information was used for the development of the flow conceptual model, which has the following main characteristics: two-dimensional, cross-sectional model oriented towards the ground water discharge point (River); homogenized waste section, with hydrodynamic properties obtained by calibration; no flow barrier on the top of the waste layer (open landfill); one-meter clay layer barrier above the base of the landfill; drainage on the top of the clay barrier layer.

3) Selection of the mathematical model: The HYDRUS model [9] was selected for the flow and transport model development. This finite element model is able to describe
variably-saturated flow and multiple solute transports for several geometries, as well as temperature-driven processes. In a posterior step of this research, the developed flow conceptual model for the reference landfill will be implemented in HYDRUS 2D and calibrated. Later, the transport of radionuclides related to phosphogypsum leaching will be simulated, applying also HYDRUS 2D, and the plume concentrations will be used for evaluating the dose for different exposition pathways. Once the Brazilian regulations do not determine the maximum allowed concentrations for radionuclides in ground water, doses will be estimated using peak concentrations, according to the worst case criteria.

3.1. The Uberlândia Sanitary Landfill

All the information presented in this item were collected from FEAM licensing processes 353/1996/006/2003 and 353/1996/001/1996, and refers to the licensing of the Uberlândia Sanitary Landfill and of its expansion in 2004.

The landfill is located in an area of 22 hectares, 7 of them related to the waste disposal area and the remaining, to the permanent preservation area around the Uberabinha River. Originally, the landfill was designed to operate between the altitudes of 790 and 810 meters above sea level, with four platforms. After the implementation of an extension project, the maximum operational elevation was changed to 835 meters, with more five platforms.

The average temperature of the municipality of Uberlândia is around 20 and 22°C, with low average around 18°C. In the Triângulo Mineiro and Alto Paranáiba areas, the isohyets are between 1,300 and 1,700 mm, with 50% of the annual precipitation rate concentrated in December and January.

Regionally, Uberlândia is inserted in the Paraná geological basin. Locally, the landfill is directly sited over pre cambrian rocks of Araxá Group and lens of Botucatu Formation sandstones, with the Serra Geral Formation as substratum. Regional drainage is made towards the high curse of the Tijucoco River and the Uberabinha River, tributary of the Araguari River (Paraná River basin). The location map of the landfill is shown at Figure 1 below.

Specifically on the area of the landfill, three types of soils are found: dystrophic purple Latosoil to moderate, clayish texture; eutrophic Cambisoi to Chernozemic, clayish texture with rocky phase; and alic dark-red latosoi to moderate, with medium texture. After five meters deep, altered basalt is found.

Ground water was observed in two different situations. A phreatic aquifer, associated to the soil layer and the altered basalt, and a fissural aquifer, associated to the basalt fractures. Water flows following the land declivity and the contact between colluvial soil and the basalt. The ground water levels can vary from 2.57 m (rain season) and 5.12 m (dry season).

Trenches were excavated for permeability investigation, yielding that the area has three ranges of hydraulic conductivities: very slow (K=2.0 x 10^{-5} cm/s); slow (K=5.9 x 10^{-5} cm/s); moderate (K=7.8 x 10^{-5} cm/s).
Upon its arrival to the landfill, waste is spread and compacted to form 0.5 m thick layers. The waste is covered daily with 10-cm thick compacted soil. On the top of each platform, a uniform declivity of 1% is applied to allow drainage of pluvial waters. The platform finishing is obtained by setting a final covering of 60 cm of low permeability clay (K=1 x 10^{-6} cm/s), compacted with an energy equivalent to 10% of the standard Proctor compaction test. Finally, this layer will be covered with soil and a cover crop.

Below the waste section, a drainage system allows recovering the waste leachate for treatment. In the expansion area, a new drainage system, as a complement of the original one, was implemented. It is constituted of three draining layers of different sizes of gravel, with a thickness of 55 cm, in total. Within the gravel layers, tubes disposed as “fish spine” shape will collect and remove the leachate.

Finally, below the drainage system, a one-meter clay layer will act as water flow vertical barrier. This clay layer has permeability of 1x10^{-6} cm/s, compacted with an energy equivalent to 95% of the standard Proctor compaction test.

![Figure 1](image)

Figure 1 – Uberlândia sanitary landfill implantation map; A-A’ cut (red line) indicates the profile that will be used for the model development.
4. LANDFILL FLOW CONCEPTUAL MODEL

The flow conceptual model of the landfill was elaborated based on the localization, dimensions and physical characteristics of Uberlândia Sanitary Landfill, as described above. The conceptual model will be used to describe the flow in a cross-sectional area of the landfill, as shown in the Figure 2. As usual for a safety assessment in a screening level, the worst case criteria were always applied.

The selected cross-section was oriented to follow the largest length of the landfill towards the Uberabinha River, the main discharge point of the area (see indication at Figure 1). This cross-section describes the center line of a potential contamination plume, where the concentrations are expected to be the largest. As a two-dimensional, cross-sectional model are being built, all flow rates must be represented as volumes/time per length unit parallel to the cross-section. Conservative values of water and contaminant flow rates for the whole landfill can be obtained by multiplying these values by the landfill’s maximum width.

On the context of this conceptual model, a 45-meters height trapezoidal waste section of the landfill (actually formed by nine platforms) was considered to be homogeneous: a blended material resultant from the mixture of the waste layers and the cover material (phosphogypsum). The hydrodynamic properties (hydraulic conductivity, porosity and retention curve parameters) of this material are meant to be obtained by calibration or by literature review. The amount of water infiltrating (Q) the landfill was estimated by the Swiss Method [10] as approximately 190 m$^3$/year per unit of landfill width, using 2,000 mm of annual precipitation rate, 380 m$^2$ of area (base length of 380 m and a width of 1 m) and a K coefficient (fraction of the precipitation that is converted in percolate) of 0.25, assuming that the specific weight of the wastes is around 7 kN/m$^3$.

![Figure 2 - Cross-sectional area of the landfill.](image-url)
As the waste profile is considered to be in hydrostatic equilibrium, humidity vertical distribution will be given by the retention curves. No flow barrier on the top of the waste layer will be included, simulating the situation of an open landfill during its operational phase. The only considered barrier for the rain infiltration is the one-meter clay layer located beneath the waste section. With a hydraulic conductivity of $1.10^{-6}\text{cm/s}$, the clay barrier layer was considered to be fully saturated.

On the top of this barrier, a drainage system promotes the removal of the water that accumulates due to the decrease of permeability. Constituted by three layers of gravel, this 55-cm, high permeability layer is considered to be half-height saturated. In order to simulate this system, a gravel layer was included, where will be applied a specified flux boundary condition to simulate the water removal by the drain.

Below the clay layer, on the contact with the soil, seepage condition was established. A 35-meters layer of local soil (clayed sand) and altered basalt precedes the contact with the impervious rock. The water table was considered to be located at 2.6 meters below the clay layer. All local flow is drained by the Uberabinha River, with River stage located at 750 m.

## 5. CONCLUSIONS

Based on environmental license report of a typical municipal solid waste landfill in operation in Minas Gerais state, a representative conceptual model of the basic dimensions and of the water flow across the landfill was developed. The two-dimensional version of HYDRUS [9] was selected for modeling the water flow and contaminants across the landfill.

This study is a part of another more comprehensive research to evaluate the environmental and healthy impact associated to the use of phosphogypsum as a daily cover in order to extend the life of a landfill, which include the following steps: 1) to set up a set of experiments in order to investigate whether the use of phosphogypsum in municipal solid waste (MSW) landfills can speed bacterial decomposition and extend the landfills' life; 2) to measure the concentration of toxic elements (radionuclides, metals and metalloids) in the leachate and 3) to perform a safety and risk analysis to evaluate its environmental and human health impact associated to that particular practice.

The safety and risk analysis will involve not only the characterization of the landfill itself, in order to define situations leading to releases of radionuclides and other toxic substances, but also the characterization of the human behavior in order to define situations leading to intrusions, and of the interfaces between the two subsystems, in order to identify situations which could lead to a radiological risk for man.

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REFERENCES


