Journal of Hazardous Materials 261 (2013) 584–592

Contents lists available at ScienceDirect

Journal of Hazardous Materials

journal homepage: www.elsevier.com/locate/jhazmat

A new approach to determine the phosphogypsum spread from the deposition site into the environment

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HIGHLIGHTS

- Stationary diffusion model was used to describe phosphogypsum particle transfer.
- Active electronic dosimeters ALARA were used to measure dose rates.
- The mass concentration of particles was determined fiducially and realistically.
- The model enabled to reduce the expenses and time of the measurements.

ARTICLE INFO

Article history:
Received 14 May 2013
Received in revised form 18 July 2013
Accepted 4 August 2013
Available online 12 August 2013

Keywords:
Phosphogypsum
Model
Stationary diffusion
Migration
Radioactivity

ABSTRACT

Phosphogypsum (PG), a waste product in phosphate fertilizer production, is characterized by technically enhanced natural radioactivity. The presented investigation was performed on a PG deposition site in Croatia. A new approach in the determination of the spread of PG particles from the deposition site into the environment based on the PG particle radioactivity measurements is suggested and explained. The stationary diffusion model was assumed and employed to describe long term PG particle transfer into the surroundings. The advantage of this method is that it requires a minimal number of measurement locations and offers a realistic and reliable distribution of PG particles. The mass concentration of PG particles decreased to the distance of about 3 m, at most up to 10 m from the deposition site edge. The results indicate that a unique mechanism of particle transport exists and the migration of PG particles by surface water is the dominant way of their spread. The particle current deduced from the measurements was very low, approximately 4.05×10^{-2} kg/h, and the migration coefficient was approximately 1.69×10^{-5} m^2/h. The obtained results confirmed the initial hypothesis of the stationarity of the migration process, reached within about 6 years.

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1. Introduction

As a waste product in phosphate fertilizer production, phosphogypsum (PG) is characterized by technically enhanced natural radioactivity originating from phosphate rock, a raw material used for phosphate fertilizer production [1–5].

One of the major concerns resulting from the impact of the phosphate industry on the environment arises from a large number of PG deposition sites worldwide. Due to the possible spread of PG, concern exists not only for the environment, but also for occupationally exposed workers and local inhabitants [6–9].

Up to now, investigations of the impact of PG deposition sites were based on measurements of activity concentrations of different natural radionuclides in soil, water and biota samples in the vicinity of deposition sites [1,9–12]. The majority of these investigations do not report significant increase in activity concentrations in soil, water or biota samples surrounding the deposition site. All of these investigations focused on measurements of activity concentrations in PG but the literature lacks data on mass concentration of PG particles around the deposition sites. Namely, natural radioactivity is not the only concern arising from PG impact on the environment. There are also compounds like fluorides, sulphates and trace elements as well as the acidity of PG [1].

The mass concentration of PG particles is hard to measure accurately and correctly since the area of interest includes a large surface area around the deposition site. Porta [13] reviews different methods for the identification and determination of gypsum ($CaSO_4 \cdot 2H_2O$) in soils which include: wet chemical methods (dissolution in acid and water), electroconductometric methods, ion

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http://dx.doi.org/10.1016/j.jhazmat.2013.08.012

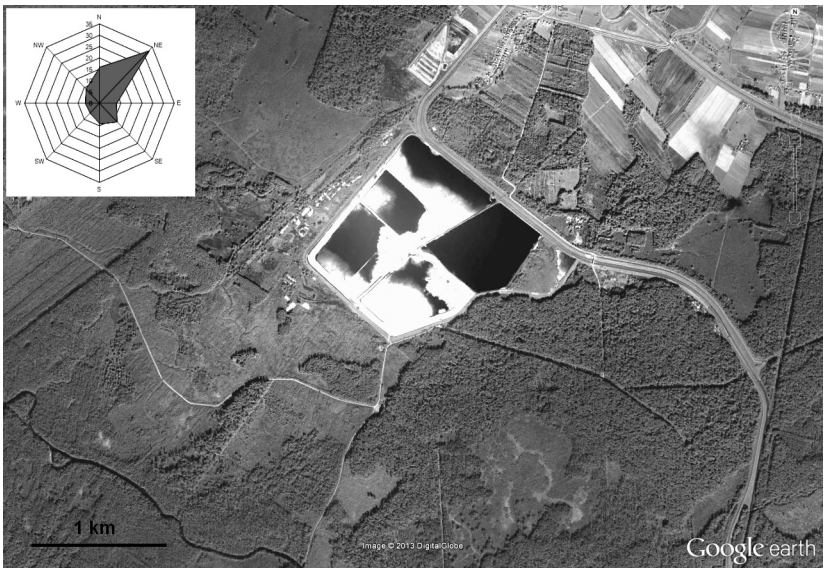

Fig. 1. Phosphogypsum deposition site located ~4 km southward from the fertilizer plant Petrokemija, Kutina, Croatia (Google Earth, 2013).

chromatography, thermogravimetric methods and X-ray diffraction techniques. All of these methods are either insufficiently accurate, lengthy, quite expensive or inapplicable to a large surface area. It is obvious that there are no easy and accurate quantitative methods to fiducially determine the mass of PG particles spread around a deposition site.

In this investigation, a new approach is introduced and tested. Based on the fact that there is almost unambiguous correspondence between particle density and the activity concentration on the soil surface, our concept was to use the natural radioactivity of PG as a marker – an indicator of particle presence in the environment. The measurements of dose rate were used as a method to determine the activity concentration of PG around the deposition site and to determine the mass concentration of PG particles. The data on the distribution of particle concentrations will enable us to determine the dominant way of particle migration and/or diffusion from the deposition site into the environment, and establish the extent of phosphogypsum spread.

For the determination of the way that PG particles spread from the deposition site to the environment, the *stationary diffusion model* was assumed to be valid. Contrary to usual types of measurements which require a large number of samples, large labor force and a lot of financial assets, the model is expected to simplify the procedure allowing for a fast, thorough and reliable assessment of PG particles spread to the deposition site surroundings. Measurement of ambient dose equivalent rate was performed using active electronic dosimeters (AED) ALARA with a time-resolution option.

The characterization of the deposition site, description of the stationary diffusion model, and measurement equipment with the locations of the measurements are described in detail in Section 2. Section 3 includes the presentation of experimental data, their analysis within the model and the discussion of the results.

2. Materials and methods

2.1. The deposition site

The investigation was performed on a PG deposition site in Croatia, located at the edge of Lonjsko Polje Nature Park, approximately 4 km southward from the "Petrokemija" fertilizer plant (N45°26′38.67″ and E16°44′40.76″). The pile consists of four ponds with a total surface of 1.6 km^2 and average depth of 4 m (Fig. 1). The capacity of the ponds is ~16 million tons, but only 4 million tons of phosphogypsum have been deposited until now [9].

Investigation of the soil composition underlying the deposition site showed that the clay forms a 2 m-thick upper layer (Fig. 2). Beneath the upper layer, there is a 4 m-thick aquifer and 1 m-thick insulating layer built from silt and clay. The second aquifer is located at a depth of 7–10 m [14]. In the area surrounding the deposition site, ground waterways move NW → SE. Fig. 2 also shows the earth embankment dam, built around the deposition site, which separates the PG pile from the surrounding area.

2.2. The stationary diffusion model

2.2.1. Migration and diffusion of phosphogypsum particles

PG particles can leave the deposition site in two possible ways. The first one is *migration*, a physical process in which PG particles are transferred as macroscopic particles by wind and/or are flooded undissolved by water (surface water and/or ground water). The second one is *diffusion*, a physico-chemical process in which dissolved PG particles are transferred through water distributed in the soil.

Since PG solubility in water is moderate (~2–2.5 g/L), no significant contribution of PG particle diffusion through soil was expected in the total number of particles deserting the deposition site.

2.2.2. Description of the deposition site within the model

The initial conditions of the model can be described as follows: the mass concentration of PG particles present on the deposition site itself is constant and maximal (constantly filled with new PG from the factory up to year 2009). In the environment, at least far enough from the deposition site, the mass concentration is equal to zero. Therefore, there is a constant difference in the concentration of particles which causes particle flow regardless of the way particles leave the deposition site (migration and/or diffusion). The migration and/or diffusion begin and the particle mass concentration increases at each location surrounding the deposition site. This process continues until a stationary state is reached, which means that, at each location surrounding the deposition site, the incoming particle current equals the outgoing particle current. Since the disposal of PG started in the early 1980s, we assumed that it was sufficient time for stationary conditions to be well established.

Two criteria must be fulfilled in order to consider a process as stationary. The first is that the mass concentration of PG particles

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