



Hazard of NORM from phosphorite of Pakistan

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ABSTRACT

In order to investigate the radiological hazard of naturally occurring radioactive material (NORM) in phosphorite deposits of Pakistan, 26 samples of phosphorite were collected from the phosphorite mines near Abbottabad, and 20 samples of single superphosphate (SSP) fertilizer were obtained from the warehouses in Pakistan. Activity concentration in all the samples was assayed using HPGe detection system. Specific activity values of ^{238}U , ^{40}K , ^{226}Ra and ^{232}Th in the samples of phosphorite were 550 ± 156 (329–845), 206 ± 72 (93–362), 511 ± 189 (316–830) and 52 ± 17 (23–81) Bq kg^{-1} , respectively; and those in SSP fertilizer due to these radionuclides were 637 ± 44 (596–687), 164 ± 24 (113–215), 589 ± 44 (521–671) and 29 ± 6 (16–45) Bq kg^{-1} , respectively. The results were compared with that of worldwide soil. Outdoor external dose rate due to gamma rays from phosphorite was calculated to be 276 ± 94 (177–441) nGy h^{-1} and external dose rate in a room made of phosphorite containing material was estimated to be 706 ± 243 (455–1129) nGy h^{-1} . The concentration of radon was measured in phosphorite mines and in the warehouses for SSP fertilizer by an active method. Protective measures have been proposed to control the pollution in the phosphorite mining and processing, and fertilizer storage areas.

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1. Introduction

NORM stands for naturally occurring radioactive materials, which may contain any of the primordial radionuclides or radioactive elements as they occur in nature, such as uranium, thorium, their radioactive decay products, and potassium that are disturbed in the earth's crust in variable amounts [1]. Radiation levels presented by NORM are generally referred to as a component of "natural background radiation". Natural uranium can substitute calcium in the phosphate rock (PR) structure due to similarity in ionic sizes between U^{4+} and Ca^{2+} , PR contains additional NORM due to uranium and its progeny [2].

Phosphorus (P) does not occur free in nature but its compounds are widely distributed, mainly as phosphates, and are found in many rocks and minerals. There are over 150 species of phosphate minerals of which the most common are apatite, phosphorite, wavellite and vivianite [3]. The phosphate ores that are in the form of calcium phosphates [$\text{Ca}_3(\text{PO}_4)_2$] are called phosphorites or apatites depending on origin of the ore. Phosphorites are very old marine sedimentary deposits associated with fossils while apatites are of igneous origin formed in situ [4]. Phosphorites are widely used as a raw material for the production of phosphate fertilizer (PF). In Pakistan, single super phosphate (SSP) fertilizer is manufactured from phosphorite. Hazara phosphorites are the only known

PR deposits in Pakistan. High-grade ore reserves of this area are being exploited for the manufacturing of SSP at Haripur and Jaranwala Fertilizer Plants in Pakistan [5]. Low-grade phosphate ore is used for manufacturing of feed for poultry and life stock. The impurities and contaminants in phosphorite are transferred to the fertilizer and feed made of it.

Phosphate make chemical complexes with uranium, therefore PR contains radioactivity of uranium and its progeny [6]. The high concentration of NORM is a source of radiological hazard that may cause significant exposure to radiation of people working in various phosphate industrial operations and of general public living in the premises of PR deposits. Radon gas emanates from the NORM in PR; and accumulates in caves, mines, houses made of/on PR, and in PF warehouses. Radon progeny can be a cause of lung cancer for the population exposed to high concentration of radon.

Human beings are exposed directly or indirectly to radiation of NORM in PR and PF. NORM can reach humans via several pathways, including the food chain, inhalation or ingestion of airborne radioactive dust and the inhalation of radon isotopes and their progeny, which reach the atmosphere as a result of the exhalation of radon isotopes from the ground surface or from the surface of building materials [7]. NORM is the source of background terrestrial radiation that varies from place to place depending on where one lives. Ingestion and inhalation of NORM can be a source of deleterious effects of ionizing radiation.

The purpose of present study was to determine the level of NORM in the phosphorite of Pakistan and in the fertilizers derived from it; therefore, radioactivity was measured in the samples of PR

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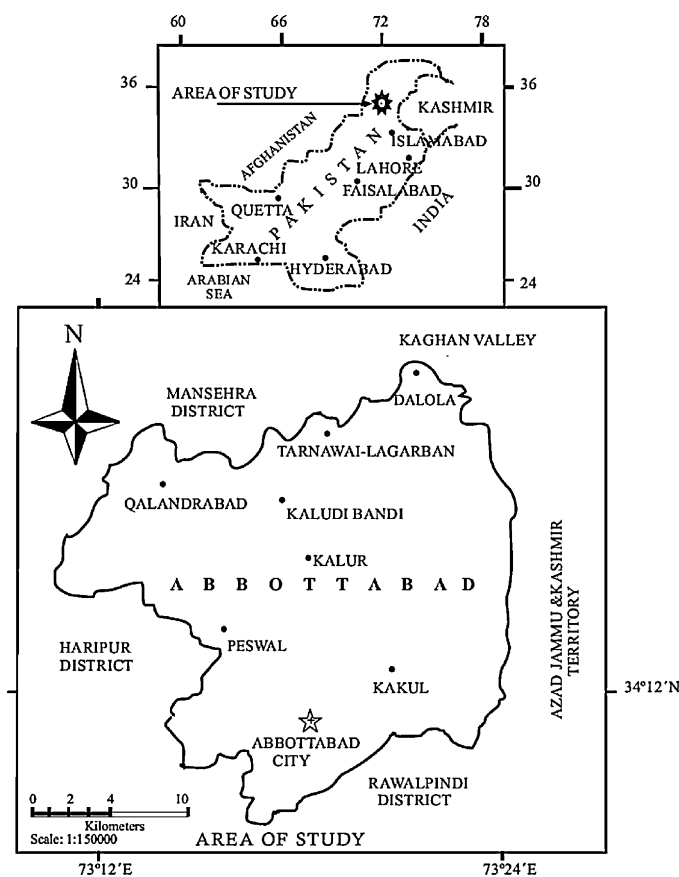


Fig. 1. Location of phosphorite sampling sites in the area under study.

collected from the mines in Hazara division and in the SSP fertilizer manufactured from this rock in Pakistan. The most widely used technique of gamma ray spectrometry based on HPGe (high purity germanium) detector was employed. The amount of radioactivity in the study area was compared with the data compiled from literature for PR and PF of some counties of the world. Radiological hazard was assessed in terms of radiation dose from the PR and radon emanating in the phosphorite mines in the study area.

2. Material and methods

2.1. Study area

The deposits of phosphate belong to the sedimentary rock formations that occur in Hazara division in the North West Frontier Province (NWFP) of Pakistan. These are the only economical deposits of phosphate in the country. Majority of phosphorite deposits of this region exist in an area of about 30 km² in linear dimension within latitudes 34°5' to 34°30'N and longitudes 73°15' to 73°20'E. The area includes Kakul, Kalur, Kakul West, Lambidogi, Tarnawai, Kaludi-Bandi, Guldanian, Dalola and many other localities in a belt near Abbottabad city in Hazara division of Pakistan. It is about 3819 feet above sea level. The study area is shown in Fig. 1. The range of P₂O₅ content in these sites is 25–30% [8]. Around 750,000 tons of PR of acceptable quality is available at Kakul mine alone [9].

2.2. Sample management

The sampling of PR was carried out from: (a) the open pit and underground mines of Kakul, (b) the open pit mine of Lambidogi, and (c) the rock crushing and milling plant of Sarhad Development

Authority (SDA) at Kakul near Abbottabad in the NWFP, Pakistan. Phosphorite samples were collected from various sections of each mine including hanging wall (formation immediate above the main phosphorite body), main phosphorite formation, and footwall (formation immediate below the main phosphorite body). A total of 26 samples were collected: 20 samples from various sections of different mines, and 6 from PR crushing plant near Kakul. Each sample was a representative of 5 specimens; one specimen was collected from the central specified site, and the others were obtained from the 4 peripheral locations around the central point [10]. The rock specimens were crushed to small pieces and mixed uniformly.

The phosphorite of Hazara deposit is mainly used for the manufacture of SSP fertilizers in Pakistan. The samples of SSP fertilizer were collected from (a) the fertilizer plants at Haripur and Jaranwala in Pakistan, and (b) the fertilizer warehouses in various cities of Pakistan. The samples from fertilizer factories were consisted of the mixtures of specimens of different batches, and those from the warehouses were the mixtures of the specimens collected from various sites. A sample was a mixture of at least 4 specimens uniformly mixed. The samples of SSP fertilizer, 20 in number, were assembled for this study.

Around one kg each of PR and PF sample was put into the cloth bag with proper marking. The samples were shifted to Geochemistry Laboratory of Pakistan Institute of Engineering and Applied Sciences (PIEAS). The rock samples were washed to remove soil dust and dried in the sun. The rock and fertilizer samples were blended and ground to make powder. The powder was heated at 100 °C for more than 20 h to remove moisture if any and passed through a sieve of mesh size 200 μm.

Reference material RG1 set was obtained from Analytical Quality Control Services (AQCS) of International Atomic Energy Agency (IAEA) for calibration of gamma ray spectrometer. The RG1 set consisted of uranium, thorium, and potassium with certified concentration of these elements. The samples and reference material were sealed in radon impermeable plastic containers of dimensions: thickness 2 mm, diameter 65 mm and height 75 mm. Density of the study samples and that of reference material were kept identical in packing containers. The samples and the reference material were stored for 40 days to achieve secular equilibrium between ²²⁶Ra and ²²²Rn. The details regarding sample management are given elsewhere [11–13].

2.3. Radiometric assay

Specific activity measurement of the NORM in the samples of PR and PF were performed in the Low Level Activity Measurement (LLAM) laboratory of PIEAS. The assay of the samples was carried out with a gamma ray spectrometry system consisting of a high purity germanium (HPGe) detector coupled with a PC-based multichannel analyzer (MCA) along with APTEC software installed in the PC. The detector was kept in a 30 cm × 30 cm × 30 cm cavity shielded with 10 cm lead with 2 mm each successive inner lining of Cu, Al and Perspex to reduce background in gamma ray spectra [12]. Head of the detector was placed nearly at the centre of the cavity. Energy calibration of the system was done initially with ²⁴¹Am, ⁵⁷Co, ¹³⁷Cs, ²²Na and ⁶⁰Co gamma ray point sources and subsequently with the reference materials. Resolution (FWHM) of the system was 1.9 keV at 1332.5 keV gamma peak of ⁶⁰Co point source kept at a distance of 25 cm in front of the detector face.

Efficiency calibration of the detection system was performed by using the reference material RG1 set. Efficiency vs. energy curve was produced from the spectra of reference materials and a polynomial was fitted to the efficiency curve that was stored in the PC. Specific activities (activity mass concentrations) of ⁴⁰K, ²²⁶Ra and ²³²Th were determined in the samples of PR and PF under study. The details regarding efficiency calibration and activity measure-

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