Uranium as a pollutant from fertilizer industries located on the Black Sea shore and upstream from the Danube Delta

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Introduction

It is known that hazards can arise from the industrial plants using raw phosphorus materials to prepare fertilizers for agriculture purposes, due to release of dust and polluted waters into the environment. This waste contains not only toxic stable micro-elements such as As, Cd, Cr, Hg, Zn, etc., but also radioactive elements like U and Th in different concentrations. SALAGEAN *et al.* (1988) investigated U, Th and characteristic micro-elements in biota from the Romanian Black Sea shore, while others (FRONTASIEVA *et al.*, 1991) pointed out that phosphorus fertilizers contain some elements whose accumulation in vitally important media such as water, soil and food are undesirable from the medical-hygienic point of view. In the present work, we investigated only the U content of phosphorus raw materials and finite fertilizers obtained by nitric and sulphuric acid procedures in industrial plants located along the Romanian Black Sea coast and upstream from the Danube River Delta. Owing to contaminated radioactive water entering the sea either from direct discharge of the water or via migration through soil, we can gain information on the uranium concentration in the biota living in these waters.

Materials and Methods

Raw phosphorites imported from Algeria, Morocco, Tunisia, Jordan, Israel, U.S.A. (Florida) and apathite from the Kola Peninsula (Russia), as well as the finite fertilizers derived by nitric or sulphuric acid procedures were obtained as samples. About 100 g dry of each powdered sample was placed in a plastic bag and measured for uranium activity by gamma-spectrometry. All the samples were counted in the same geometry using a Phoswich (USA) detector of 127 mm dia. [3 mm NaI(T1)+50 mm CsI(T1)] coupled to a multichannel analyzer and personal computer. This system could detect either the 63 keV and 93 keV gammas from the Th-234 daughter of U-238 or the 186 keV from U-235.

Results and Discussion

The experimental data are shown in Tables 1 and 2. A significant quantity of uranium is noted in the final products. By correlating the chemical and radiological limits with the U content in the finite fertilizer products, it has generally been recommended to avoid the maximum possible uranium from all fertilizers. However, in our fertilizers U is not considered dangerous for several reasons. If we consider that 2-3 kg (case 1) or at maximum 200-300 kg (case 2) of finite fertilizer NPK2 nitric product (Table 2) with the highest U content (i.e. 28.85 mg/kg corresponding to an activity of 721.25 Bq/kg) is sprayed on 10,000 m^2, it means a variation of U activity ranging between 0.1442 to 0.21637 Bq/m^2 (case 1) or 14.42 to 21.637 Bq/m^2 (case 2). Upon the recommendations of ICRP-30 (1979), the Annual Limit on Intake (ALI) of natural U in critical organs (lungs, kidneys), is 1.5×10^3 Bq which corresponds to 60 mg natural U. However, an important quantity of U is retained by chemically complexed compounds such as humic acids in the soil and sediments of the Danube or Black Sea. Previous investigations by SALAGEAN *et al.* (1988) on U content in biota from the Black Sea have found 0.6 ppm in *Enteromorpha linza*, 2.8 ppm in *Ceramium rubrum* and <6 ppm (soft tissue) and <4 ppm (byssus) in *Mytilus galloprovincialis* from the Danube Delta near Sulina.

Table 1. Uranium content in raw samples

Sample	mg/Kg	Bk/kg*
Phosphorite Morocco	131.07	3276.75
Phosphorite Tunisia	40.42	1010.50
Phosphorite Jordan	73.21	1830.25
Phosphorite calcinated Morocco	148.58	3714.50
Phosphorite calcinated Algeria	56.37	1409.25
Phosphorite calcinated Israel	272.7	6817.50
Phosphorite calcinated U.S.A.	15.99	399.75
Apathite Kola (Russia)	141.46	3536.50

Table 2. Uranium content in fertilizers (finite products)

Fertilizer	mg/kg	Bk/kg*
NPK_2 nitric product	13.84	346.00
NPK_1 sulphuric product	15.27	381.75
NPK_2 nitric product	28.85	721.25
NPK_1 sulphuric product	27.30	682.50
NPK_2 nitric product	10.34	258.50
NPK_2 nitric product	2.82	70.50

* 1 mg U generates 25 Bq for natural uranium (88 - EHD - 139 Bioassay Guideline 4. Guidelines for U Bioassay. Health and Welfare - Canada)

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