Application of Airborne Gamma-Ray Spectrometry and Remote Sensing to Map Contamination by Fertilizers

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Fertilizers are widely used in agriculture, particularly in tropical regions, where weathering of soil nutrients is more intense. Uranium is commonly present in phosphate rocks and, consequently, in fertilizers. The exploitation, processing and persistent use of large amounts of phosphate fertilizers lead the environment to be contaminated by U, Th and other trace elements resulting from radioactive decay. In the present study, airborne gamma-ray spectrometry, remote sensing, digital imagery and geographic information systems were used to integrate geophysical, geological, topographical, pedological and drainage data in order to interpret radiometric (K, U and Th) anomalies in the region of Araras, State of São Paulo, Brazil. These anomalies occur in soils developed from basic rocks (diabase sills), which are known to normally contain low concentrations of radionuclides. Owing to their high fertility, these soils have been historically used for important crops such as coffee and sugar cane, but an increasing amount of fertilizers has been used there over the last thirty years. The radiometric anomalies (U, Th) are attributed to impurities in phosphates, whereas the ^{40}K anomalies result both from the use potash and agricultural residues like those from the sugar cane industry, in which up to 0.7g/l of K_2O is present.

INTRODUCTION

Coincidence of K, U and Th aerogamma-spectrometric anomalies and diabase sills was detected over the area delimited by 22°00'/22°30' S and 47°00'/47°30'W by geoprocessing airborne geophysical survey data collected between 1978 and 1979 as part of the *Projeto Aerogeofísico São Paulo - Rio de Janeiro* (Anjos & Mourão, 1988). Since airborne gamma-spectrometry can detect radiation generated from as deep as 30cm into the earth's surface and soils developed from weathering of basic sills are known to normally contain low concentration of radionuclides, the high concentrations of K, U and Th would not be expected in the area of study (Ferreira, 1991).

In the present study, geographical information systems and digital processing of aerogamma-spectrometric and remote sensing imagery are used to integrate geophysical, geological, superficial formation, topographical, pedological and drainage parameters. Chemical and physical analysis (grain size) was performed to compare soil samples collected from areas geophysically anomalous with samples from primary forest areas. Radiochemical analysis for K, U and Th and research into the spatial relationship of geophysical, geological and agronomic parameters are being done in soils of those areas. The purpose of such integration is a better understanding of geophysical anomalies as a result of massive, continued use of fertilizers in sugar-cane crops.

RADIOACTIVITY IN FERTILIZIERS

The modern agriculture, in the process of increasing its efficiency in food prodution, has given subsidies to the culture through soils preparation practices, genetic improvement, use of agrochemicals, manufacturing and use of fertilizers. In order to enable the modernization process, the costs should not surpass the benefits. Considering man as part of the ecosystem, the negative effect which the agricultural practices are causing to the environment should be evaluated in a cost-benefit scheme. Despite the advantages of using fertilizers, the massive application of such products causes heavy-metals and radioactive elements to be released to environment (Guimond, 1978).

Phosphate rocks are treated with sulfuric acid (normal superphosphate with 18-20% P_2O_5) to produce phosphoric acid, which is then combined with ammonia or some phosphate rock into ammonium phosphates (46-48% P_2O_5) or concentrated (triple) superphosphates respectively. The resulting fertilizers contain nitrogen, phosphorous and potassium (NPK). Normally, nitrogen is originated from nitric or ammoniac sources; phosphorous is obtained from phosphate rocks and potassium is originated from potassium salts (potassa). Nitrogen contains negligible amounts of radionuclides, whereas phosphates contain significant concentrations of U, Th and Ra. Potassa contains ^{40}K according to the normal normal concentration when comparated to the total K in the earth´s crust (Guimond & Hardin, 1989).

The radioactive elements found in the Brazilian igneous deposits of phosphate (Jacupiranga SP, Araxá and Tapira MG), from which fertilizers are produced, occur either as pyrochlore or as a complex niobium-tantalotitanate compound of rare earths with some concentration of radionuclides (Cullen & Paschoa, 1978). Within these deposits, significant concentrations of U_3O_8 can be found: 180 ppm in Tapira (Cullen & Paschoa, 1978), 30 ppm in Jacupiranga, and 100 to 160 ppm in Araxá (Lapido-Loureiro, 1986). The average concentration of U_3O_8 in sedimentary deposits of phosphate in the Northeast of Brazil is 150 ppm, one of the highest concentrations of this type the of ore minerals in the world (Saad, 1974).

Every soil is radioactive to some extent and emits gamma radiation in smaller or bigger grade, according to the substract´s geological nature. Igneous rocks show a marked variation in potassium, uranium and thorium content. The most abundant of these elements is potassium, which is most commonly found in alkali-feldspars and mica. Uranium and thorium, when present in igneous rocks, are mainly associated with moderately radioactive accessory minerals, weakly radioactive essential mineralls within mineral intertices and may also occur in fluid inclusions. In general, minerals containing higher concentrations of uranium and thorium form in the late stages of magma segregation, i.e., when the amount of silica is also higher. Therefore, the average content of radionuclides in acid igneous rocks (3.5% K, 4.1 ppm U and 21.9 ppm Th) is much higher than that of basic-ultrabasic rocks (0.01-1% K, 0.007-0.8 ppm U and 0.02-3.4 ppm Th) (Saunders & Potts, 1976). As a result of this variation in concentration of radionuclides linked with silica content, soils derived from acid igneous rocks like granites are more radioactive than those derived from basic rocks like diabase. In these one, it is expected a low natural radioactivity and not high concentrations of radionuclides as it was observed in the area of study.

Methods

The geological (1:250.000 - *Instituto de Pesquisas Tecnológicas do Estado de São Paulo IPT*), pedological (1:100.000 - *Istituto Agronômico de Campinas IAC*) and Superficial Formation (1:50.000 - *Instituto Geológico - IG*) maps of Araras region were digitized, adjusted and had their elements' topology constructed in the *SGI/SITIM* geographical information system. 20 m equidistant contour lines and drainage systems were digitized in *AutoCad12* from cartographic maps on the scale of 1:50.000 published by *IBGE* (*Instituto Brasileiro de Geografia e Estatística*). A digital terrain model was constructed in *SGI/SITIM* for the region. K, U and Th aerogammaspectrometric data was processed with the *Geosoft* Package (Geosoft, 1994) . A regular grid of 125 x 125 m cells was built from N-S, 1 km equidistant flight lines. *Idrisi for Windows* was used to integrate the geophysical, pedological, geological and topographical variables involved. The grid of K, U and Th values was transformed into a raster image (8 bits), each pixel of which being represented by one of 256 shades of gray, which resulted in a compression of the original spectral resolution of the geophysiscal variables. The R, G and B channels of a composite image were assigned to K, U and Th bands, respectively. The unsupervised classification of the composite image resulted in 25 different patterns of K, U and Th spectral response. The less significant groups were ommited. The same procedure was followed for the 5 (R), 4 (G) and 3 (B) LANDSAT/TM bands. The cluster of coincidence of K and Th obtained from the unsupervised classification was superimposed to a Digital Elevatiom Model (DEM). By assigning 1 to the cluster that resulting from the classification of the composit image, new layers were generated which allowed queries to be performed on the geological, pedological and superficial formation data. Intersection area calculations were also made for the variables.

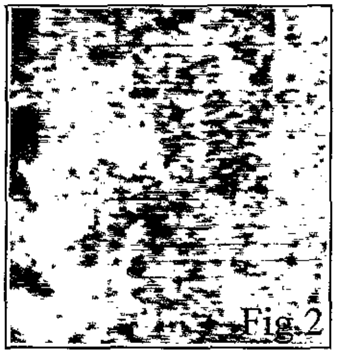
Geology

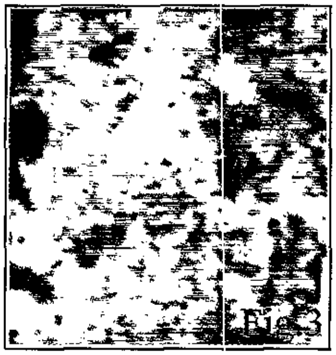
Araras region is located in Paraná Basin. The geology of area comprises formations ranging in ages from the Paleozoic up to the Cenozoic. The Palaeozoic is represented by Itararé Group and Aquidauana Formation (late Carboniferous to early Permian), Tatuí Formation (middle-late Permian), Passa-Dois Group (late Permian Irati and Corumbataí formations). The Mesozoic rocks of Araras region correspond to São Bento Group (Pirambóia Formation and the basic sills of the Serra Geral Formation). The Cenozoic sediments correspond to the Rio Claro and Piraçununga formations and to alluvionar sediments. The geology of area involves: **Qa** Alluvionar sediments; **TQp** Piraçununga Formation; **TQr** Rio Claro Formation; **JKβ** basic sills; **TRjp** Pirambóia Formation; **Pc** Corumbataí Formation; **Pi** Irati Formation; **Ptt** Tatuí Formation; **CPi** Itararé Group, undivided; **CPs** Aquidauana Formation.

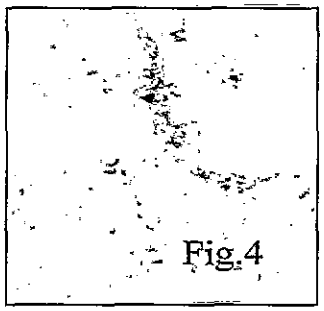
Soils

The following types of soil are present: **LR** ***(latossolo roxo* - oxisol):** mineral soils derived from basic rocks, with oxic B horizon and considerably high content of Fe_2O_3 (18%-40%), MnO and TiO_2 and strong magnetic attraction; clay loam and clay; **LE *(latossolo vermelho escuro* - oxisol):** mineral soils with B oxic horizon, Fe_2O_3 content of 18% where clayey and usually lower than 8% where the textural is loam; low to null magnetic attraction; **LV *(latossolo vermelho amarelo* - oxisol):** mineral soils with oxic B horizon; Fe_2O_3 equal or lower than 11% and higher than 7% where clay but not concretionary (where concretionary, a higher content of Fe_2O_3 may be observed); where texture is medium, the ratio Al_2O_3/Fe_2O_3 is greater than 3.14; virtually no magnetic attraction; **PV *(podzólico vermelho amarelo* - ultisol):** mineral soils with textural B horizon and low CEC; **TR *(terra roxa estruturada* - alfisol):** mineral soils derived from basic rocks; relatively high content of Fe_2O_3 (minimum 15%) and 1.5% TiO_2; clay loam or clay; low textural gradient with textural B horizon and low activity clay; **QA *(areias quartzosas profundas* - quartzipsamments):** deep, very friable soils with sandy grain size along the profile (minimum 2m below surface); virtually no primary minerals; easily weathered where coarse; **Li *(solos litólicos* - orthents):** soils normally thinner than 30 cm; A-R horizons; underlying rock usually altered and weak; varied textures closely related to the substratum's nature; **Hi *(solos hidromórficos* - entisols):** hydromorphism at depths shallower than 120 cm.

RESULTS AND DISCUSSION







The light shades of gray in figures 1, 2 and 3 correspond to high counts of K, U and Th, respectively. An important aspect to notice is the high concentrations of K (Figure 1) along the drainage system as a result from the discharge of vinhaça, a by-product of the processing of sugar cane that contains approximately 0.7 g/l of K_2O. The same can be observed in Figure 4, that represents the K-Th cluster that resulted from the unsupervised classification of the image. Uranium is present over the latosols in the higher portions of the terrain (Souza ***et al.*** 1997), over which high amounts of phosphate-rich fertilizers were are used. An interesting relationship between the variables involved in the

present study can be observed in Figure 5, which represents the LANDSAT/TM band 5, layers the *latossolo roxo* - a (pedological map), diabase sills - b (geological map), the diabase sill cluster - c (resulting from the classification of the RGB composite of 5, 4 and 3 LANDSAT/TM bands) and the cluster of high cps of K, U and Th - d (resulting from the classification of the composite of gamma-ray imagery). These relationship reveals that the soils developed from diabase sills, in which sugar cane is cropped, contain high concentrations of radionuclides as indicated by treatment of imagery digital. Considering that 700kg/alq of NPK composite with concentrations of 4-20-20 and/or 5-25-25 are applied to to sugar cane crops and, in some areas, additional 200kg/alq of superphosphate are applied, the high concentration of U can be explained by the massive, continued use of phosphate fertilizers. When the production system is of the "*cana-soca*" ("*rebrote*") type, formulations with low concentrations of P and high concentrations of N and K, like 14-07-28, 18-00-36, 20-05-20, which can explain the low concentrations of U and the high concentrations of K^{40}. Radiochemical analysis of soil and fertilizers is being carried out to confirm the hypothesis put forward in the present paper.

REFERENCES

ANJOS, I. L. S. and MOURÃO, L. M. F. 1988. Projeto Acrogeofísico São Paulo - Rio de Janeiro, Relatório Final, Processamento dos Dados, Parte I - São Paulo. Companhia de Pesquisa de Recursos Minerais (CPRM), vol. 2, 29p, (restricted circulation).

FERREIRA, F. J. F. 1991. Aerogamaespectrometria e Aeromagnetometria de um Trato Ocidental do Pré-Cambriano Paulista, Tese de Doutoramento, Instituto de Geociências da Universidade de São Paulo (IG-USP), 160p.(restricted circulation).

GUIMOND, R. J. 1978. The Radiological Aspects of Fertlizer Utilization, Radioactivity in Consumer Products, NUREG/CP-0001, U. S. Nuclear Regulatory Comission, Washington, D.C., p.380- 93.

GUIMOND, R. J.; HARDIN, J. M. 1989. Radioactivity Released from Phosphate-Containing Fertlizers and from Gypsum, Radiat. Phys. Chem., vol. 34, p. 309-315.

CULLEN, T. L. and PASCHOA, A. S. 1978. Radioactivity in Certain Products in Brazil, Radioactivity in Consumer Products, NUREG/CP-0001, U. S. Nuclear Regulatory Comission, Washington, D.C., p.376-379.

LAPIDO-LOUREIRO, F. E. 1986. Alkaline Rocks and Carbonatites, Correlation of Uranium Geology Between South America and Africa, Technical Reports Series n°270, International Atomic Energy Agency, Vienna, p. 345-415.

SAAD, S. 1974. Aspectos Econômicos do Aproveitamento do Urânio Associado do Nordeste, Boletim n°7, Comissão Nacional de Energia Nuclear (CNEN).

SAUNDERS, D. F. and POTTS, M. J. 1976. Interpretation and Application of High-Sensitivity Airborne Gamma-Ray Spectometer Data, Exploration of Uranium Ore Deposits, Proc. Series, International Atomic Energy Agency, Vienna, p. 107-125.

SOUZA, J. L., FERREIRA, F. J. F., ROCHA, H. O., MANTOVANI, L. E. 1997. Soil Radioactivity and its Possible Relationship with Fertilizers in Araras Region, Brazil. *In:* 4'th Meeting on Nuclear Application (in press).