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Modeling background radiation using geochemical data: A case study in and around Cameron, Arizona

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ABSTRACT

This study compares high resolution forward models of natural gamma-ray background with that measured by high resolution aerial gamma-ray surveys. The ability to predict variations in natural background radiation levels should prove useful for those engaged in measuring anthropogenic contributions to background radiation for the purpose of emergency response and homeland security operations. The forward models are based on geologic maps and remote sensing multi-spectral imagery combined with two different sources of data: 1) bedrock geochemical data (uranium, potassium and thorium concentrations) collected from national databases, the scientific literature and private companies, and 2) the low spatial resolution NURE (National Uranium Resource Evaluation) aerial gamma-ray survey. The study area near Cameron, Arizona, is located in an arid region with minimal vegetation and, due to the presence of abandoned uranium mines, was the subject of a previous high resolution gamma-ray survey. We found that, in general, geologic map units form a good basis for predicting the geographic distribution of the gamma-ray background. Predictions of background gamma-radiation levels based on bedrock geochemical analyses were not as successful as those based on the NURE aerial survey data sorted by geologic unit. The less successful result of the bedrock geochemical model is most likely due to a number of factors including the need to take into account the evolution of soil geochemistry during chemical weathering and the influence of aeolian addition. Refinements to the forward models were made using ASTER visualizations to create subunits of similar exposure rate within the Chinle Formation, which contains multiple lithologies and by grouping alluvial units by drainage basin rather than age.

1. Introduction

1.1. Aerial gamma-ray surveys

Aerial gamma-ray surveys are one of the primary tools for tracking the dispersal of radioactive contamination in the environment (Pitkin et al., 1964; Sanderson et al., 1993; Lyons and Colton, 2012; Sanada et al., 2014) and are used for a variety of homeland security purposes as well as in disaster response and mitigation (cf. Blumenthal and Musolino, 2016; Blumenthal et al., 2103). For any gamma-ray survey made over land, a principal component of the naturally occurring radiological background comes from rocks and soils because earth materials inevitably contain some level of radionuclides, in particular potassium-40 (^{40}K), uranium-238 (^{238}U), thorium-232(^{232}Th) and their short-lived daughter products. During the recent disaster at the Fukushima Daiichi nuclear power plant in Japan, it became clear that for emergency response purposes, simple, rapid methods of separating the contaminant plume from natural background radiation coming from geologic sources are needed (Lyons and Colton, 2012). Past studies have focused on interpreting gamma-ray survey data in terms of bedrock or soil type (e.g. Griscom and Peterson, 1961; Pitkin et al., 1964; Galbraith and Saunders, 1983; Harris, 1989; Schetselaar and Rencz, 1997; Schetselaar et al., 2000; Wilford, 2002; Martelet et al., 2006; Rawlins et al., 2007; Dierke

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and Werban, 2013; Beamish, 2013), prospecting for mineral resources (e.g. Dickson and Scott, 1997) or identifying fallout from nuclear weapons testing (Books, 1962). In contrast, our study attempts to use the measured geochemistry of the bedrock to forward model the observed natural gamma-radiation background. A successful method for modeling the background will be useful for a variety of homeland security, nuclear disaster response and resource exploration purposes. An ideal model would predict the gamma-radiation background point by point across the land surface, with each location modeled independently from each adjacent point. However, such a model would be impractical to construct. Instead, we seek to segment the region over which the model extends into geographic units over which average characteristics can be assigned. For the sake of discussion, we will refer to these geographic areas as 'background radiation units'. For reasons that will be apparent below, in this study we have chosen to define background radiation units using geologic maps and remote sensing imagery.

Airborne gamma-ray spectroscopy measures the gamma-radiation emitted at or near the surface of the Earth. Gamma-radiation is able to penetrate about 30 cm of rock or soil and a few hundred meters of air. ^{238}U, ^{40}K and ^{232}Th are the only naturally occurring radioactive isotopes that exist in large enough quantities, and produce gamma-rays (either directly or in their decay chains) with high enough energies to be measured during an aerial survey (Minty, 1997), which is typically conducted a few hundred meters off the ground. The energy released during radioactive decay is different for each isotope (Dickson and Scott, 1997). Potassium, a common element in rocks and soils, makes up approximately 2% of the Earth's crust. 0.012% of all potassium is its radioactive isotope, ^{40}K. Potassium is most abundant in K-feldspars and micas, and when these K-rich minerals are weathered the potassium may be incorporated into new clay minerals (Dickson and Scott, 1997). ^{232}Th is a radioactive isotope that occurs as a common trace element in rocks and soils, with a decay series that releases a number of gamma-rays that can be detected by aerial gamma-ray surveys. ^{232}Th itself does not release any high energy, high intensity gamma-rays; instead, its subsequent daughters release these gamma-rays which are used to calculate an equivalent Th (eTh) concentration. Th, with a crustal abundance of 12 ppm, occurs in significant quantities in minor minerals such as allanite, monazite, xenotime and zircon (Dickson and Scott, 1997). When Th is weathered out of a mineral, it tends to stay in place due to its low solubility. It can be taken up by clays or iron oxides (Koons et al., 1980), and if adsorbed by colloidal clays, can be transported out of the system. U is another important contributor to the gamma-radiation background. It occurs in many of the same environments, minerals, and rocks as Th when in its reduced state (U^{4+}). Unlike Th, U also has an oxidized state (U^{6+}) that is soluble and therefore mobile. Like ^{232}Th, ^{238}U does not release high energy, high intensity gamma-rays, but its daughters release these gamma-rays, which are used to calculate an equivalent U (eU) concentration. U occurs in the Earth's crust at a concentration of about 3 ppm, and has two major isotopes: ^{238}U and ^{235}U, with ^{238}U being far more abundant. U occurs in minor quantities in oxides and silicates and along grain boundaries, and in higher concentrations in zircon, monazite and xenotime (Dickson and Scott, 1997).

Disequilibrium can affect the decay series of U and Th, causing eU and eTh to differ from actual concentrations. eU is mainly calculated from the spectra of the ^{238}U daughter bismuth-214, while eTh is mainly calculated from the spectra of the ^{232}Th daughter thallium-208 (Minty, 1997). In a closed system, equilibrium occurs after 10 half-lives of the longest-lived daughter, which is about 40 years for Th and 1.5 million years for U (Dickson and Scott, 1997). Disequilibrium can also occur through the preferential removal of either the parent or daughter isotopes. Due to the low mobility of Th and its daughters in aqueous environments as well as the short half-lives of its daughters, the Th decay chain is less susceptible to disequilibrium. In contrast, due to the higher solubility of U^{6+} as well as the volatility of its daughter radon-222 (Rn), equilibrium in the U decay chain cannot be taken for granted. However, studies have shown that at least in arid environments, aerial gamma-ray survey interpretation is not affected by disequilibrium (Dickson, 1995).

1.2. Past studies

Aerial gamma-ray surveying of the US began in the late 1940s for the purpose of U exploration. With the advent of commercial nuclear power plants, aerial gamma-ray surveys of areas around nuclear facilities were employed to create a baseline against which to detect future anomalies (Pitkin et al., 1964). Even in these initial studies, the relationship between background radiation and bedrock geology was apparent. For example, Pitkin et al. (1964) proposed that faults could be detected by higher exposure rates due to leaking radon. Using an aerial gamma-ray survey done of the Maryland Piedmont, Griscom and Peterson (1961) found previously unknown mafic intrusions. Moxham (1963) concluded that surface radiation is dominated by contribution from rock, not soil in the northeastern US. Aerial gamma ray surveys are now used for a wide variety of applications including environmental monitoring (Jones, 2004; Sanderson et al., 2004), soils research (Beamish, 2013; Dierke and Werban, 2013), surficial processes research (Martz and deJong, 1990; Pickup and Marks, 2000) and as an aide to geologic mapping in remote areas (Campbell et al., 2007; Martelet et al., 2006; Ford et al., 2008) as well as for U, Th, and precious metals exploration (Mernagh and Miezitis, 2008; Dickson and Scott, 1997; Dickson, 1995; Wilford, 2002; Ford et al., 2007).

Although early studies measured radiation only in terms of counts per second, by the 1970s radiation was characterized in terms of exposure rate in units of μR/hr (micro-roentgen per hour). Exposure rate is a measure of ionizations in a mass of air generated by gamma-rays per unit of time. It can be determined by integrating the number of counts per second between approximately 0.4 MeV and 3.0 MeV. Using computational models of photon emission, Beck et al. (1972) and Løvborg and Kirkegaard (1974) proposed that exposure rates could be calculated from a linear combination of K, U, and Th concentrations. Grasty et al. (1984) compared the resulting coefficients to their own calculations and proposed average values of the coefficients, which have been cast in terms of dose by Duval et al. (2005) and used here assuming a factor of 0.1 between nGy/h and μR/h:

$$\dot{X} = 1.32\ \mathrm{K} + 0.548\ \mathrm{eU} + 0.272\ \mathrm{eTh} \quad (1)$$

Where $\dot{X}$ is exposure rate in μR/hr, K is weight percent potassium, eU is ppm uranium, and eTh is ppm thorium. From 1973 to 1980, the Atomic Energy Commission conducted an aerial gamma-ray survey of the continental United States known as the NURE (National Uranium Resource Evaluation) survey for the purpose of evaluating U resources (Duval et al., 2005). The data, made available by the USGS, are reported as K, eU and eTh concentrations, and exposure rates may be calculated using Equation (1).

Although relationships between bedrock geochemistry and the spatial distribution of exposure rates are widely observed, the relationship is not so straightforward that a gamma-ray survey can replace a geologic map. Average radioactive isotope contents for a wide variety of rock types have been determined (Mernagh and Miezitis, 2008; Dickson and Scott, 1997), but the ranges of radioactive isotope concentrations overlap (c.f. Fig. 6 in Beamish, 2013)