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Mapping potassium and thorium concentrations in Belgian soils

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ABSTRACT

The European Atlas of Natural Radiation developed by the Joint Research Centre (JRC) of the European Commission includes maps of potassium K and thorium Th. With several different databases available, including data (albeit not calibrated) from an airborne survey, Belgium is a favourable case for exploring the methodology of mapping for these natural radionuclides. Harmonized databases of potassium and thorium in soil were built by radiological (not airborne) and geochemical data. Using this harmonized database it was possible to calibrate the data from the airborne survey. Several methods were used to perform spatial interpolation and to smooth the data: moving average (MA) without constraint, or constrained by soil class and by geological unit. Overall, there was a reasonable agreement between the maps on a $1 \times 1\,km^2$ grid obtained with the two datasets (airborne data and harmonized soil data) with all the methods. The agreement was better when the maps are reduced to a $10\,km \times 10\,km$ grid used for the European Atlas of Natural Radiation. The best agreement was observed with the MA constrained by geological unit.

1. Introduction

Terrestrial radioactivity is mostly caused by uranium (^{238}U, ^{235}U) and thorium (^{232}Th) radioactive families together with potassium (^{40}K) (UNSCEAR, 2008). U mapping in Belgian soils was considered in a previous paper (Cinelli et al., 2017). The present contribution is devoted to K and Th, following similar methods. Maps of K and Th concentrations in soil are included in the European Atlas of Natural Radiation (EANR) developed by the Joint Research Centre (JRC) of the European Commission. The EANR is a collection of maps of Europe displaying the levels of natural radioactivity caused by different sources of radiation. The digital version is now available online at: https://remon.jrc.ec.europa.eu/About/Atlas-of-Natural-Radiation.

^{40}K and ^{232}Th contents in soil could be estimated through geochemical or radiological analysis. The isotopic abundances of ^{40}K (0.0117%) and ^{232}Th (100%) being very stable, the elemental concentrations can be directly deduced from the activity concentrations. In radiological analysis, ^{40}K is measured by gamma spectrometry, whereas ^{232}Th could be measured directly (through alpha spectrometry) or indirectly by considering its progenies (by gamma spectrometry) (Knoll, 2000; Gilmore, 2008). Gamma spectrometry could be performed in situ, on an airborne platform or in the laboratory. ^{208}Tl is mostly used in gamma spectrometry for measuring ^{232}Th assuming the secular equilibrium in its decay chain. In the environment, this condition may not be perfectly achieved due to the mobilization of the radionuclides. However, disequilibrium is expected to be less marked than in the uranium decay chain, due to the lack of long-lived progeny.

Geochemical K and Th data in soil were collected for all of Europe through the Geochemical Atlas of Europe (FOREGS) and Geochemical Mapping of Agricultural and Grazing Land Soil (GEMAS) projects (Reimann et al., 2014a; 2014b). Radiological data are available at national to regional scale. In many European countries, these data were not collected with a density high enough to perform local statistics on the $10 \times 10\,km^2$ square grid used for the EANR, and some kind of interpolation and smoothing is necessary.

It is reasonable to assume that K and Th concentrations in the soil are correlated to other soil properties (Ferreira et al., 2018). This makes it possible to use European soil maps (ESDB, 2004) as a framework in which K-Th mapping could be inserted. The relation with geology is a bit less direct, but many soil types are expected to include material derived from the alteration of the local bedrock.

Belgium is a favourable case for testing various possible mapping options, because of the availability of detailed airborne radiological surveys that could be used as the reference map. We present a study of different options to map K-Th concentrations in soil in Belgium using geochemical and radiological data. While the results have intrinsic interest for Belgium, this work can also be considered as a contribution for the development of the EANR, i.e. at the European level.

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This paper is structured as follows: In Section 2, we present the data available on K and Th in soil and the soil and geological classifications applicable in Belgium, as well as the software packages used in the present work. Then, three important methodological aspects will be examined. First, in Section 3, we examine harmonization of the input data, as they stem from different studies, each with its own methods. Second, statistical tools, such as analysis of variance (ANOVA) and variograms, will be used to select the mapping methods in section 4. Finally, in Section 5, the different mapping methods are applied to K and Th data: (a) interpolation/smoothing/averaging of measured data, without considering any other factor; or (b) mapping separately the data belonging to distinct classes, such as soil or geological classes. The impact of the mapping resolution is examined, and the method giving the best agreement between the maps of harmonized data and of airborne data is determined.

2. Input data and software

This section is limited to a synthetic presentation, because the databases and methods used in the present paper are very similar to those used in our previous work devoted to U mapping (Cinelli et al., 2017), to which the reader should refer for more detail.

Belgium may be divided into three regions of progressive elevation: in the North, a low-altitude plain with Cenozoic grounds; in the Centre, a low Meso-Cenozoic plateau often covered with quaternary loess; in the South, higher Paleozoic massifs, except a small Mesozoic area in the extreme South.

2.1. Databases of K and Th in Belgian soils

The positions of the measurement sites are plotted in Fig. 1, and the main features of the different datasets are summarized in Table 1.

2.1.1. Geochemical data

FOREGS is a European database developed for the Geochemical Atlas of Europe. Topsoil and subsoil samples were taken at the same sites (http://weppi.gtk.fi/publ/foregsatlas/index.php).

The GEMAS European database (Reimann et al., 2014a,b; http://gemas.geolba.ac.at/) includes samples of agricultural soil (Ap, 0–20 cm) and of grazing land (Gr, 0–10 cm).

2.1.2. Radiological data

The Study Centre for Nuclear Energy (SCK-CEN) and the Institute of Public Health (IHE) (Gillard et al., 1988; Deworm et al., 1988) collected soil samples and measured their activity by gamma spectrometry. Similar measurements were done by the Institut Supérieur Industriel de Bruxelles (ISIB), for ^{232}Th in soil samples taken at a few sites close to Brussels.

The University of Ghent (GENT), collected data by in-situ gamma spectrometry in air, at 1 m height (Uyttenhove et al., 2000).

2.1.3. Airborne measurements of K and Th progeny

The Belgian Geological Survey organised an airborne campaign of radiometric measurements (BGS, 1995), using a multi-crystal NaI detector and a 256-channel spectrometer, considering three energy windows for gamma rays, corresponding to lines of ^{40}K (1.35–1.59 MeV), ^{214}Bi (^{238}U/^{222}Rn progeny, 1.63–1.89 MeV) and ^{208}Tl (^{232}Th progeny, 2.42–2.81 MeV). The results were reported as counts per second (cps), without calibrating concentrations of the elements considered. Data were acquired along parallel lines at 1 km from each other at 120 m altitude. The data were interpolated at the nodes of a 100 × 100 m^2 grid. We only used these data on a kilometric grid. For each node of our grid, we linearly interpolated the 4 nearest values of the 100 × 100 m^2 grid. It is important to realize that the airborne spectrometer "saw", from the altitude of 120 m, an area of about 1 km^2, and thus reducing the dataset from the 100 × 100 m^2 grid to the 1 × 1 km^2 grid does not imply a real loss of information.

2.2. Soil and geological classifications

2.2.1. European soil maps

The European Soil Database – ESDB (ESDB, 2004; Panagos, 2006) provides EU-wide data for 73 soil attributes. The datasets used in the current work were: (a) WRBLV1 containing the soil reference group code of the Soil Typological Units (STU) from the World Reference Base (WRB) for Soil Resources, (b) TXSRFDO contains the dominant surface textural class of the STU. The corresponding maps are available to be downloaded at ESDAC (ESDAC: https://esdac.jrc.ec.europa.eu/content/european-soil-database-v20-vector-and-attribute-data.

2.2.2. Geological maps

An old set of 1:40,000 geological maps (BGS, 2015), still the only set of geological maps to cover the entire territory of Belgium, is available

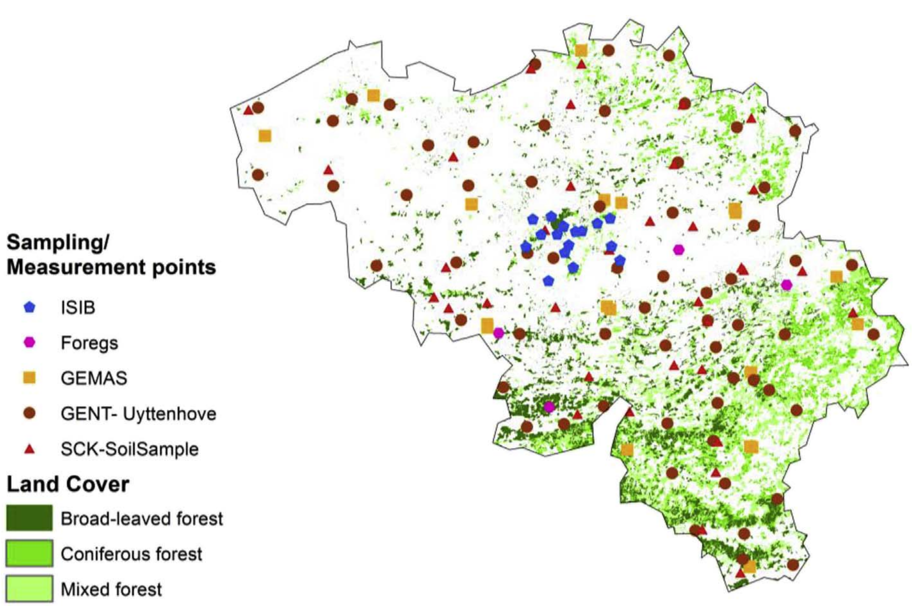

Fig. 1. Map showing sampling points for the datasets used in the present work. The areas covered by forest are displayed (source: CORINE Land Cover map, Silva and Lavalle, 2011).