Integrating of airborne gamma-ray survey and geological data for environmental radioactivity map construction

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Abstract The airborne γ -ray spectrometry method (AGRS) is widely considered as an important tool for mapping environmental radioactivity both in geosciences studies and for purposes of emergency response. AGRS is a powerful tool with respect to in-situ gamma-ray measurements for monitoring large areas, especially if it is handled with informations brought by geological maps.

Using a typical cluster of Nal crystals (16 L) mounted on microlight, we performed the first airborne gamma-ray survey on Elba Island (about 224 square km) with the purpose to measure the K-40, U and Th content in the top soil.

The results provide comprehensive records of environmental radioactivity concentrations, by considering the geological setting of the area. In this study we advance a new multivariate approach by using Collocated CoKriging (CCoK) interpolator with geological constrains, in order to produce airborne gamma-ray maps. The very complex geological setting of Elba Island makes the indicator approach almost inapplicable, which would have been the more logical choice if the number of categories would have been relatively low. In fact, the most actual geological database is composed by a relatively large number of different geological formations (73 different geological formations mapped by a detailed field survey at scale 1:10,000), very high regarding to the usual application of indicator estimators.

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However, we overcame the categorical character of geological database by randomly assigned a number to each category (geological formation) of the polygons of the geological map. The non-dependency of the maps on the random numbering process has been tested and leads to satisfactory results (differences between pairs of estimated maps show the mean close to zero). The efficient use of CCoK shows comprehensive correlation between natural radioactivity concentrations in the top soil and geological constrain: it confirms the potentiality of this method for further applications.

Introduction

Airborne γ -ray spectrometry (AGRS) for ground concentrations of potassium, uranium and thorium measuring is a well-established monitoring technique (I.A.E.A., 1991, 2003, 2010). This is mainly because AGRS method is highly appropriate for large scale environmental surveys. In fact, AGRS method can reduce time consumption in monitoring wide areas, even if it restricts the spatial resolution of measurement, which averages the signal over the area covered by the time slice (typically of the order of I km²). We utilized four Nal(TI) detectors of four liters volume each (I.A.E.A., 2003, 1991). By introducing Full Spectrum Analysis (FSA) with the Non Negative Least Square (NNLS) constraint in the γ -spectra analysis, the quality of the analysis performed by the detectors was increased (Caciolli et al., 2012).

Since the AGRS measurements refer to spot areas only, they have to be interpolated in order to create the radioactivity map. Natural radioactivity is strictly connected to geological features of the region, and we checked the performances of the method in a suitable testing site, the Elba Island (a relatively small island of 224 km² in Northern Tyrrhenian Sea, Italy). This is a sector of the northern Apennines, with a large number of different geological formations and lithologies. The geological distinctive features of this island are linked to its complex tectonic pile of nappes and to the well-known Fe-rich ores, as well as to the well-exposed interactions between Neogene magmatic intrusions and tectonics, as you can see for instance in Trevisan, 1950, 1951 and Bortolotti et al., 2001. The former described the tectonic building of Elba Island as a structural pile of at least five main units called "Complexes" of Trevisan", TC, the lowermost of them belong to the three Tuscan Domain, while the uppermost two relate to the Ligurian Domain. The latter proposed a new stratigraphic and tectonic model in which the five TC were reinterpreted and renamed. Moreover, we utillized a recent fine continuos geological map coming from a detailed fieldwork.

In our study geological informations are coupled to airborne γ -ray data in a multivariate analysis to obtain the first radioactivity map of Elba Island. In order to reach this goal, we shall make a non-conventional use of a well known

multivariate geostatistical interpolator, the Collocated Co-Kriging (CCoK) described by Xu et al. (1992). In fact, the overall objective is to develop a new breakthrough cost-effective solution for environmental variables interpolating, especially when measurements are performed either under difficult conditions or in extreme environments (Figure 1).



Figure I. geological map of Elba Island (taken from the new Geological Map of Tuscany region realized at 1:10,000 scale, see C.G.T., 2011): the western sector is mainly characterized by intrusive igneous rocks (magenta), the central and eastern parts of the island are characterized by a wide lithological variation (green, purple and pink), while the south- eastern outcrop almost exclusively consists of metamorphic rocks (Mt. Calamita). For the official legend of all the geological formations see http://www.geologiatoscana.unisi.it . Coordinate system is UTM WGS84 Zone 32 North.

Methods

The AGRS system is a modular equipment composed by four Nal(TI) detectors with a total volume of 16 liters mounted on an ultralight vehicle (autogyro). As survey strategy we planned to be as perpendicular as possible to the main North-South style of geological structures of the area. On June 3 2010 the autogyro flew on Elba Island in two hours collecting 803 spectra referred to an averaged spot area of the order of about 0.25 km² (area from which comes 90% of the signal). The average height of flight was 140±50 m: at this height 90% of gamma

radiation from ²¹⁴Bi at 1760 keV is originated at about 20 cm of topsoil. Performing a post-processing procedure, we associated homogenous K (%), eTh (ppm), and eU (ppm) abundances to each spot area.

In our study, we used the collocated cokriging (CCoK) as multivariate estimation method for the interpolation of airborne gamma-ray data (the primary undersampled variable, Z1, natural abundances of K, eTh, or eU measured via airborne γ -ray spectrometry) on the basis of the geological map (the secondary variable Z2 that refers to the continuous distribution of the geological formations, i.e. the detailed geological map at 1:10,000 scale, following C.G.T., 2011).

Since the geological map used lists 73 different geological formations, the approach based on categorical variables (see for instance Hengl et al., 2007; Pardo-Iguzquiza and Dowd, 2005; Goovaerts, 1997) should have required a long time for processing and interpretations. Therefore, we had to consider the geological qualitative (categorical) map as a quasi-quantitative constraining variable, by means of a randomly assigning progressive numbers in different ways to each category of geological map. This choice does not affect the final interpolation results.

The measured radionuclides show different distributions: K (%) abundances have a quasi-Gaussian distribution, while eTh (ppm) and eU (ppm) abundances distributions tend to be positively skewed (Table I), Linear correlation is high between pairs of abundance variables (Table 2). On the ground of the previous assumptions, the linear correlation coefficient between radioactivity measures and values randomly assigned to geological formations is meaningless.

| Parameter | K (%) | eTh (ppm) | eU (ppm) |
|-----------------------|-------|-----------|----------|
| Count | 803 | 803 | 803 |
| Minimum | 0.0. | 0.70 | 0.00 |
| Maximum | 6.73 | 38.52 | 30.30 |
| Mean | 2.74 | 13.01 | 6.87 |
| Standard Deviation | 1.27 | 6.87 | 4.87 |
| Variance | 1.63 | 47.15 | 23.72 |
| Variation Coefficient | 0.47 | 0.53 | 0.71 |
| Skewness | 0.14 | 0.46 | 1.12 |
| Kurtosis | 2.64 | 2.54 | 4.67 |

Table 1 descriptive statistical parameters of airborne γ -ray values.

Experimental semi-variograms (ESV) and experimental cross-semi-variograms (X-ESV) allowed interpreting the models by taking into account factors conditioning the spatial distribution of these regionalized variables. Analysis was performed on raw data, even if they do not have exactly Gaussian distribution.

This is due to the substantial absence of outliers in abundance distributions, as well as non-erratic styles of both ESVs and X-ESVs.

Table 2 Correlation matrix for abundance variables (Pearson's linear correlation coefficient value for each bivariate distribution), and the statistical significance testing scores for each correlation test (**** indicates high significance of test).

| | K (%) | eTh (ppm) | eU (ppm) |
|-----------|-------|-----------|----------|
| K (%) | I | *** | *** |
| eTh (ppm) | 0.853 | I | *** |
| eU (ppm) | 0.813 | 0.865 | I |

The directional X-ESVs show an erratic behavior. Therefore we modeled the experimental co-variability as isotropic and an omnidirectional linear model of coregionalization has been fitted by means of trials and errors procedure (Wackernagel, 2003).

Results

Both the high density of radioactivity data and the highly refined geological map allowed to construct a well checked linear coregionalization model. The ESVs models built up for the AGRS measurements show regular structures with low variabilities at small distances and generally higher at the spherical structures.

In CCoK estimations for all groups of variables (one of each is composed by one radionuclide and the categorical geological map) we did not consider the experimental uncertainties of airborne γ -ray measures. The estimated natural radioactivity maps (an example is shown in Figure 2 for eTh estimated abundances) are calculated with a spatial resolution in accordance with the choice of the geological map at 1:10,000 scale. Note that we considered the standard deviation of CCoK estimations normalized respect to the CCoK estimated values of the abundances (normalized standard deviation, NSD), with an average NSD of 21.6%, 25.8% and 25.5% for K, eU, eTh respectively.



Figure 2. Estimation map of Th (ppm) abundance and normalized estimation errors

Conclusions

The AGRS method was tested to be a powerful investigation tool for mapping the environmental radioactivity of large areas, gathering a relatively high sample density (average 3.5 sample/km2 for about 2h survey). This fact assures higher accuracy in using spatial interpolators.

In this work, the well-known multivariate analysis technique of collocated cokriging CCoK was applied in a non-conventional way considering the well-sampled geology composed by 73 geological formations as a quasi-quantitative variable and constraining parameter. Conventionally, categorical variables (like geology) are treated through the indicator estimators, which is a well-tested interpolation method for a relatively low number of categories (for instance, Hengl et al., 2007 utilized indicators methods to interpolate 12 possible classes of soil texture). We overcame the categorical feature of the geological database, by randomly assigning a number to each category of the geological formations. This approach gives a well-structured linear model showing a good spatial covariability in the omnidirectional co-regionalization ESV model.

As a result of this study, was realized the first detailed map of potassium, uranium and thorium of Elba Island at 1:10,000 scale by combining smoothing

effects of probabilistic interpolators, such as CCoK, and the abrupt characteristics of the geological map. Indeed, polygons of geological formations are assumed to be homogeneous in terms of abundance of radioactive elements, but separated from each other by sharp contact lines.

Finally, the geological implications of this method lead to the following results. The main patterns of geological maps show a distinct correlation between the geological formation and radioactivity content, as well as relatively high abundances in the intrusive granitic complex of Mt. Capanne (Western part of Elba Island) and the metamorphic complex of Mt. Calamita (South-East of Elba Island). Furthermore, evidences of anomalous areas of different abundances are marked for instance around Porto Azzurro village (Elba's Eastern coast). However, in this part of Elba Island an homogeneous granitic pluton is shown by geological map. Thus, these anomalies in CCoK estimated maps could be geologically explained by considering this area as constituted by small emplacements of granitic plutons and aplitic intrusions, which are out of the spatial resolution of 1:10,000 scale. These evidences testify the accuracy and potentiality of the AGRS survey as a tool for radioelement mapping and as a precious integrative support for geological mapping. Finally, In fact, we reached our overall objective by utilizing a new breakthrough cost-effective solution for environmental variables interpolating measured in a relatively few locations such as airborne γ -ray.

Bibliography

- Bortolotti, V., Fazzuoli, M., Pandeli, E., Principi, G., Babbini, A., and Corti, S. (2001). Geology of central and eastern Elba Island, Italy. *Ofioliti*, 26(2a): 97–150.
- Caciolli, A., Baldoncini, M., Bezzon, G., Broggini, C., Buso, G., Callegari, I., Colonna, T., Fiorentini, G., Guastaldi, E., Mantovani, F., Massa, G., Menegazzo, R., Mou, L., Rossi Alvarez, C., Shyti, M., Xhixha, G., and Zhanon, A. (2012). A new fsa approach for in situ gamma-ray spectroscopy. *Science of The Total Environment*, 414:639–645.
- C.G.T. (2011). Accordo di programma quadro ricerca e trasferimento tecnologico per il sistema produttivo - c.l. geologia e radioattivita' naturale sottoprogetto a: Geologia (Regional Framework Program for research and technological transfer to industry, C.l. Geology and Natural Radioactivity, Sub-project A: Geology). Technical report, CGT Center for GeoTechnologies, University of Siena; Tuscany Region: Italian Ministry of Education, University and Research.
- Goovaerts, P. (1997). Geostatistics for natural resources evaluation. Applied geostatistics series. Oxford University Press, New York; Oxford. Geology Statistical methods.

- Hengl, T., Toomanian, N., Reuter, H. I., and Malakouti, M. J. (2007). Methods to interpolate soil categorical variables from profile observations: Lessons from Iran. Geoderma, 140(4):417–427.
- I.A.E.A. (1991). Airborne gamma-ray spectrometry surveying. Technical Report Series 323, International Atomic Energy Agency, Vienna.
- I.A.E.A. (2003). Guidelines for radioelement mapping using gamma-ray spectrometry data. *Technical Document IAEA-TECDOC-1363*, International Atomic Energy Agency, Vienna.
- I.A.E.A. (2010). Radioelement mapping. Technical Document. Nuclear Energy Series IAEA NES No.NF-T-13, International Atomic Energy Agency, Vienna.
- Pardo-Iguzquiza, E. and Dowd, P. A. (2005). Multiple indicator cokriging with application to optimal sampling for environmental monitoring. *Computers and Geosciences*, 31(1):1–13.
- Trevisan, L. (1950). L'Elba orientale e la sua tettonica di scivolamento per gravità. Mem. Ist. Geol. Univ. Padova, 16:5–35.
- Trevisan, L. (1951). La 55^a riunione estiva della Società Geologica Italiana. Isola d'Elba, 18-23 settembre 1951. Boll. Serv. Geol. It., 70:435–438.
- Wackernagel, H. (2003). Multivariate Geostatistics: An Introduction With Applications. *Springer-Verlag*, Berlin, Germany, 3rd edition edition.
- Xu, W., Tran, T., Srivastava, R., and Journel, A. (1992). Integrating seismic data in reservoir modeling: The collocated cokriging alternative. In SPE Annual Technical Conference and Exhibition, Washington, D.C. Society of Petroleum Engineers Inc.