Computers and Geosciences 115 (2018) 56–65

Contents lists available at ScienceDirect

Computers and Geosciences

journal homepage: www.elsevier.com/locate/cageo

Research paper

Quantitative X-ray Map Analyser (Q-XRMA): A new GIS-based statistical approach to Mineral Image Analysis

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ARTICLE INFO

Keywords:
Numerical petrology
Linear regression
Mineral formula quantification
ArcGIS®
Python

ABSTRACT

We present a new ArcGIS®-based tool developed in the Python programming language for calibrating EDS/WDS X-ray element maps, with the aim of acquiring quantitative information of petrological interest. The calibration procedure is based on a multiple linear regression technique that takes into account interdependence among elements and is constrained by the stoichiometry of minerals. The procedure requires an appropriate number of spot analyses for use as internal standards and provides several test indexes for a rapid check of calibration accuracy. The code is based on an earlier image-processing tool designed primarily for classifying minerals in X-ray element maps; the original Python code has now been enhanced to yield calibrated maps of mineral end-members or the chemical parameters of each classified mineral. The semi-automated procedure can be used to extract a dataset that is automatically stored within queryable tables. As a case study, the software was applied to an amphibolite-facies garnet-bearing micaschist. The calibrated images obtained for both anhydrous (i.e., garnet and plagioclase) and hydrous (i.e., biotite) phases show a good fit with corresponding electron microprobe analyses. This new GIS-based tool package can thus find useful application in petrology and materials science research. Moreover, the huge quantity of data extracted opens new opportunities for the development of a thin-section microchemical database that, using a GIS platform, can be linked with other major global geoscience databases.

1. Introduction

Advanced geological and petrographic investigations are increasingly supported by a collection of quantitative information. In this context, quantitative X-ray mapping is emerging as an innovative tool that can be applied in many fields, from basic scientific research to mining industry, cultural heritage and forensic geology, as demonstrated by the large investments made by manufacturers of microanalytical devices. However, in contrast to single spot analyses, X-ray maps are characterised by semi-quantitative raw data that is not corrected, for instance, for the mean atomic number, absorption or fluorescence (ZAF) effects (Lanari et al., 2018 and references therein). In this context, to derive fully quantitative data on element concentrations, good analytical standardization procedures are crucial in overcoming the intrinsic limits of X-ray mapping (Lanari et al., 2018 and references therein), such the previously mentioned ZAF effects, background noise and possible peak overlap in energy-dispersive-spectrometer (EDS) X-ray maps, or the volatilisation of light elements due to prolonged exposure under a finely focused electron beam.

Taking into careful consideration the problems above, a new performing multiple linear regression technique was applied for the first time to calibrate the unprocessed wavelength-dispersive-spectrometer (WDS) or EDS X-ray maps using high-precision spot analyses as internal standards (De Andrade et al., 2006). The developed procedure, which largely modifies the pre-existing ArcGIS®-based image processing platform by Ortolano et al. (2014b) (i.e., X-ray Map Analyser), aims to calibrate sets of X-ray maps for each classified phase.

In general, image processing in materials science is mainly based on the multivariate statistical analysis of raw X-ray intensities in EDS/WDS matrices or of images with multiple raster bands (typically 8-bit images with 256 intensity levels) recording the distribution of chemical elements. This is one of the techniques applied routinely in the investigation of many geo-petrological processes (e.g., Airaghi et al., 2017; Belfiore et al., 2016; Coutelas et al., 2004; Lanari et al., 2013; Loury et al., 2016; Marmo et al., 2002; Mészaros et al., 2016; Ortolano et al., 2014a; Vidal et al., 2006). The technique has been adopted also thanks to the development of semi-automated tools for classifying mineral phases and quantifying modal parameters from selected thin section micro-domains

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https://doi.org/10.1016/j.cageo.2018.03.001
Received 23 June 2017; Received in revised form 23 February 2018; Accepted 2 March 2018
Available online 6 March 2018

(e.g., Friel and Lyman, 2006; Gu, 2003; Lanari and Engi, 2017; Launeau et al., 1994; Ortolano et al., 2014b; Tarquini and Favalli, 2010; Tinkham and Ghent, 2005).

Others tools use a very efficient clustering algorithm (i.e., K-means) to obtain several homogeneous groups of pixels automatically bounded by a mask (e.g., Cossio and Borghi, 1998; Cossio et al., 2002; Lanari et al., 2014). In particular, software such as Petromod (Cossio et al., 2002) and XMapTools (Lanari et al., 2014) treat each of these groups of pixels as an array of X-ray intensity values representing a single mineral phase, where the element pixel array is a function of chemical concentration. Such software use linear regression functions to obtain new sets of calibrated X-ray images, converting qualitative raster images into calibrated grid formats (i.e., an array of equally-sized, square grid points storing concentrations as absolute numeric values) (Cossio and Borghi, 1998; Cossio et al., 2002; De Andrade et al., 2006; Lanari et al., 2014; Togami et al., 2000).

In this context, calibration techniques are obviously a crucial step in the quantitative analysis of raster imagery. Careful control of the prediction model is required to obtain physically significant results.

The new procedure differs substantially from the previous one (i.e., X-ray Map Analyser: Ortolano et al., 2014b), which has been completely revised and now includes: (a) an additional analytical cycle based on a multiple linear regression that is used to produce X-ray grid images of each identified mineral phase; (b) a third analytical cycle that uses calibrated maps to extract the zoning patterns of complex solid-solution mineral phases. The third cycle is useful in visualising the progressive changes in end-member concentrations, which are generally related to multivariant chemical reactions in complex systems.

2. Algorithms

Ortolano et al. (2014b) used the Python programming language to develop a semi-automated image processing procedure that was implemented in ArcGIS®. This procedure, based on the multivariate statistical analysis of X-ray maps, was designed for classifying chemically homogeneous zones and extracting quantitative information such as mineral modes (e.g., Belfiore et al., 2016; Ortolano et al., 2014a). The Python code of this procedure was implemented in a first analytical cycle (Fig. 1) used to classify all recognizable phases within the entire selected domain and in a second cycle for a more in-depth analysis of the previously identified phases and the detection of any sub-phases such as those related to mineral zoning. Both cycles use principal component analysis together with a maximum likelihood classification algorithm to generate graphical outputs. Although this image processing procedure yields a significant amount of information, it lacks an algorithm able to transform the derived raster X-ray image of identified phases into a standard grid format.

A new Python code was therefore implemented to cyclically calibrate the sets of X-ray maps for each classified phase using the ArcGIS® licensed software available on demand at the Geoinformatics and Image Analysis Lab of the Biological, Geological and Environmental Sciences Department at the University of Catania.

The second cycle (Fig. 1) has been completely modified. Based on microprobe spot analyses, it introduces a multiple linear regression technique to predict element concentration values for each pixel (expressed as atoms per formula unit, a.p.f.u) within a specific phase. This is a crucial step for implementing a third analytical cycle (Fig. 1) used to derive maps of end-member proportions and to highlight chemical variations within the investigated phases.

The following sections describe the main steps of the new analytical cycles, focusing on the adopted multiple linear regression technique. Further specific insights are also provided in the user guide available in the supplementary material within the software (i.e., Q-XRMA).

2.1. Second cycle: calibration algorithm

We chose a multiple linear regression algorithm (Hosmer and Lemeshow, 1989; Marquardt, 1980; Neter et al., 1996) because it can

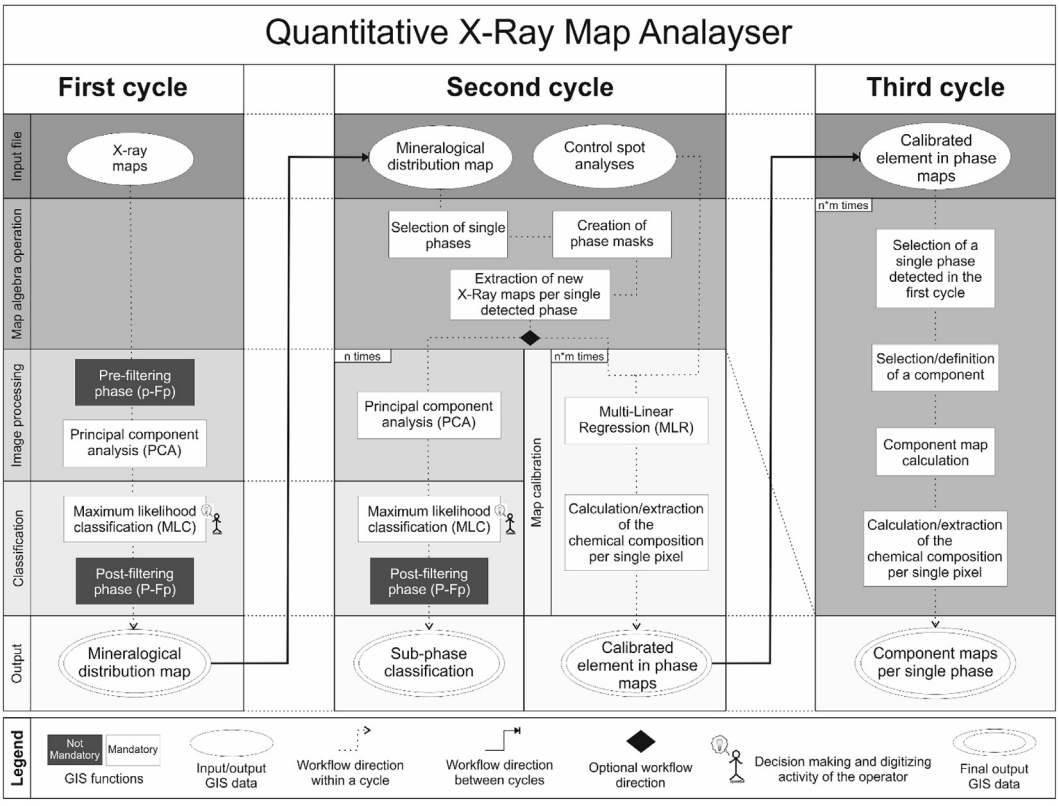

Fig. 1. Simplified flow charts of the GIS-based image-processing procedure.
The procedure is subdivided into three analytical cycles.