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Reprint of "Comparison of datasets obtained by global-scale geochemical sampling in Australia, China and Europe"☆

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ABSTRACT

Catchment/floodplain sediment geochemical datasets from three continental-scale geochemical projects in Australia (AU), China (CH) and Europe (EU) were studied by comparing sample media, sample preparation, elements determined, analytical methods, detection limits and proportions of reportable values, and certified reference materials used. Twenty six elements (Ba, Ce, Co, Cr, Cu, Mo, Nb, Ni, Pb, Rb, Sr, Th, V, Y, Zn, Zr, Al_2O_3, CaO, K_2O, MgO, MnO, Na_2O, P_2O_5, SiO_2, Fe_2O_3 (total), TiO_2) were considered comparable among the 35 determined by all three projects, according to the criteria for global comparability of elements proposed in this paper. The median concentrations of all elements (except Co, Y, Zr, SiO_2 and TiO_2) in the three datasets increase in the following order: AU < EU ≤ CH, and the median CH/EU ratios for all elements range from 0.98 to 1.94, while the median CH/AU and EU/AU ratios for elements, such as CaO, Na_2O, MgO and Zn are generally greater than 2 (and up to 7), which could be explained partly by protracted weathering in Australia, and partly by the smaller grain size fraction analysed of the CH dataset (<1 mm) compared to AU and EU (<2 mm). In conclusion, a unified sample medium should be collected and unified sample preparation techniques should be followed. Key elements related to mineral resources and the environment should be determined and international or exchanged internal standard materials inserted in new national or global geochemical mapping projects in order to generate globally comparable datasets for establishing global geochemical baselines.

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1. Introduction

Ninety-four elements in the periodic table occur in nature, and their abundance in the Earth's upper continental crust has been studied for the past 120 years, beginning with the first estimation by Clarke (1889). Little is known, however, of their baseline concentrations and spatial distributions in the Earth's surface or near-surface environment (Wang et al., 2006). To know and to understand the current geochemical baselines are the first essential steps to quantify the future geogenic and human-induced changes, and to evaluate the past evolution due to geological processes (Darnley et al., 1995; Reeder, 2007; Smith et al., 2012; Wang, 2012; Zoback, 2001). Geochemical maps display the spatial distributions of chemical elements in Earth's surficial materials, facilitating the solution to problems related to natural primary resources, agriculture, forestry, environmental issues, human health, land use, and in many other fields (Darnley et al., 1995).

To date, a systematic geochemical mapping project covering the whole terrestrial surface of the Earth has not yet been carried out. Systematic regional geochemical mapping projects have been supported, however, by forward-thinking national governments and funds made available to state institutions to carry out this work. Since the 1970s, more than 50 countries have launched regional-scale geochemical mapping projects, covering a little over a fifth of the Earth's terrestrial surface (≈148,940,000 km^2). Starting from the 1990s, since the implementation of IGCP 259 'International Geochemical Mapping' (1988–1992) and IGCP 360 'Global Geochemical Baselines' (1993–1997), significant progress has been made on global-scale or continental-scale geochemical mapping projects, covering a total area of approximately 32,000,000 km^2 in about 30 countries (≈22% of the Earth's land surface). Several valuable datasets have been generated to study continental-scale to global-scale geochemical patterns.

In order to establish a global geochemical database of permanent value, the report of IGCP 259 project outlined detailed recommendations and proposed the following seven basic requirements (Darnley et al., 1995):

(1) *commonly available representative sample media, collected in a standardised manner;*
(2) *continuity of data across different types of landscape;*

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(3) *adequate quantities of the designated sample media for future reference and research requirements;*
(4) *analytical data for all elements of environmental or economic significance;*
(5) *the lowest possible detection limits for all elements;*
(6) *determination of the total amount of each element present, and*
(7) *tight quality control at every stage of the process.*

Several continental-scale geochemical surveys have been conducted in Australia, China, Europe, India, Mexico, and the United States, partly according to the recommendations of the IGCP 259 report (Darnley et al., 1995). However, analytical variation still exists in these datasets, because of different analytical techniques. Separating analytical variation from natural variation (resulting from lithology, geological or pedological process, climate, human activities, etc.) in a reliable way is crucial prior to application of the analytical data to global comparability.

Yao et al. (2011) compared analytical results generated in Chinese and European laboratories for the same sub-soil samples of FOREGS (Forum of European Geological Surveys, now EuroGeoSurveys) Geochemical Atlas of Europe project to evaluate between-laboratory bias. It was demonstrated that comparable analytical data of certain elements can be achieved by different laboratories and at different mapping stages. Reimann et al. (2012) selected elements with direct comparability from the NGSA (National Geochemical Survey of Australia) and GEMAS (EuroGeoSurveys Geochemical Mapping of Agricultural and Grazing Land Soil project) based on exchanged standards analysed by both surveys. Reimann et al. (2012) concluded that to produce directly comparable datasets at the continental (or even country) scale is a difficult undertaking; they stated that even small differences in sample material, sampling method, sample preparation or analytical procedures can have a major impact on the observed element concentrations, and such effects have in the past often led to country borders being visible on geochemical maps rather than the true element spatial distribution patterns. According to these authors, the most crucial point appears to be analytical quality and the direct comparability of analytical results, and that total element concentrations are usually more comparable than values from a weaker extraction, such as aqua regia.

Nevertheless, for most completed and on-going continental/national-scale geochemical surveys, the between-laboratory bias has not been estimated (as Yao et al., 2011 did), and exchanged standard materials have not been analysed within the analytical stream of each project (as Reimann et al., 2012 did). How to assess the analytical variation among these datasets is a crucial issue that requires special attention.

In this study, six aspects including Sample media, Sample preparation, Elements determined, Analytical methods, Detection limits and proportions of reportable values, and Certified reference materials are considered in order to assess data comparability of three completed continental-scale geochemical mapping projects: the National Geochemical Survey of Australia (NGSA) (de Caritat and Cooper, 2011a), the Environmental Geochemical Monitoring Networks project of China (EGMON) (Cheng et al., 1997; Xie and Cheng, 1997; Xie et al., 1996, 1997), and the FOREGS Geochemical Baseline Mapping Programme in Europe (De Vos et al., 2006; Salminen et al., 1998, 2005).

2. Overviews of the three projects

2.1. The NGSA project

The National Geochemical Survey of Australia (NGSA) project was conducted in collaboration with all State and Northern Territory geosciences agencies in Australia between 2006 and 2011 (Johnson, 2006). This project aimed to provide a pre-competitive geochemical database and to improve the national information of concentrations and abundances of energy-related elements, such as U and Th, by determining the chemical composition of transported regolith samples from the outlet of large catchments (de Caritat and Cooper, 2011a).

Sampling was conducted from 2007 to 2009. The sampling method suitable to Australia landscape and climate conditions is described in detail in the NGSA Field Manual (Lech et al., 2007). In total, 1186 catchments covering about 81% of Australia were sampled, representing an average density of approximately 1 site per 5200 km^2. Only six of the largest catchments were sampled at two widely separated locations, and 123 catchments (>10%) were sampled in duplicate for quality control purposes, totalling 1315 sample sites. At each site, two samples were collected: a Top Outlet Sediment (TOS) from 0 to 10 cm and a Bottom Outlet Sediment (BOS) from 60 to 80 cm on average. Both samples were taken as composite samples either from a shallow ~1 m^2 soil pit (TOS) or from generally at least 3 auger holes within an area of ~100 m^2 (BOS). The weight of each sediment sample was about 9 kg (de Caritat and Cooper, 2011a).

The parameters determined in the NGSA project included total concentrations of 60 elements (the subject of this paper), aqua regia soluble concentrations of 60 elements, Mobile Metal Ion™ (MMI) concentrations of 54 elements, FeO, Loss On Ignition (LOI), pH (field and lab in 1:5 soil:water slurries), electrical conductivity (in 1:5 soil:water slurries), laser particle size analysis, and visible–near-infrared spectroscopy. All analytical methods are described in de Caritat et al. (2010) except for spectroscopy, which are in Viscarra Rossel et al. (2010, 2011).

For the generation of geochemical data of high quality and integrity, the NGSA project followed a strict quality control programme involving randomised sample numbers, field duplicates, analytical replicates, internal standards and several certified reference materials. The quality control programme is discussed in the NGSA Data Quality Assessment report (de Caritat and Cooper, 2011b).

2.2. The EGMON project

The Environmental Geochemical Monitoring Networks (EGMON) project in China was conducted from 1992 to 1997 with Professor Xie Xuejing as project leader. This project was part of the IGCP 259 (International Geochemical Mapping) pilot study to find a suitable sampling medium for continental-scale geochemical surveys, using a grid cell of 160 × 160 km^2. Floodplain sediment was tested as the first choice for a global sampling medium (Cheng et al., 1997; Xie and Cheng, 1997; Xie et al., 1996, 1997), following the successful results of the Western European Geological Surveys' project on Geochemical Mapping of Western Europe towards the Year 2000 conducted by the Working Group on Regional Geochemical Mapping under the leadership of Professor Bjørn Bølviken (Bølviken et al., 1990, 1993, 1996; Demetriades et al., 1990; Ottesen et al., 1989).

Floodplain sediments were taken from 532 sample sites covering the central and eastern part of China in 1994. The sampling sites were mostly located on the floodplains of large-catchment basins ranging from 1000 to 10,000 km^2 and at least 500–1000 m from the confluence point with other rivers. Two samples were taken at each site using a Luoyang shovel, i.e., (i) a surface floodplain sediment from a depth of 5–25 cm, and (ii) a deep floodplain sediment from a depth of 80–120 cm. Additional 314 stream/overbank sediment samples were collected at river beds in 1996 in Tibet plateau and Xinjiang province, where floodplain sediments are not developed. A total of 846 samples were taken in EGMON project to cover approximately 90% of China, corresponding to a sampling density of about 1 sample/10,000 km^2. The minimum weight of each sample was 2.5 kg (Cheng et al., 1997; Xie et al., 1996).

Total concentrations of 50 elements were determined in the EGMON project at the central laboratory of the Institute of Geophysical and Geochemical Exploration (Cheng et al., 1997; Xie et al., 1996); Pt and Pd were analysed at the central laboratory of the Henan Institute of Noble Metals, which is affiliated to the Ministry of Geology and Mineral Resources. The standard reference samples GSD 1–9 (Xie et al., 1985a),

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