Chromites: geochemical insights provided by statistics. A case study

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Abstract

The present study aims to identify the extent to which the statistical approach can provide insights into the geochemical properties of chromite. For this purpose, chemical analyses referring to metasomatic chromites, chromites hosted by arc-related ophiolites, and chromites associated with Precambrian ophiolitic mafic-ultramafic bodies were considered. The results of the statistical approach showed that Principal Component Analysis and Pearson correlation coefficients can help identify the geochemical properties that individualize the three types of chromites. Also, the strong negative correlations noticed between Mg and Fe^{2+}, Cr and Al, (Mg + Cr) and (Fe^{2+}+Al), and (Mg+Al) and (Fe^{2+}+Cr) indicate the geochemical changes that may occur during chromite crystallization, regardless of their geological setting. They reveal the nature of either metasomatic substitutions (regarding metasomatic chromites) or changes in the chemistry of the crystallization environment (when chromites are related to ophiolites). Similar processes, but involving different geochemical changes, take place at the scale of each occurrence. That is supported by the strong negative correlations between Cr and (Mg+Al), Al and (Mg+Cr), Cr and (Fe^{2+}+Al), as well as Al and (Fe^{2+}+Cr), identified especially in metasomatic chromites and those hosted by arc-related ophiolites. The interdependency between the degree of occupancy of the R1 and R2 structural sites is more pronounced in the metasomatic chromites.

Keywords: chromite, univariate statistics, bivariate statistics, Principal Component Analysis, Pearson correlation matrix.

1. Introduction

Statistical analysis is widely used across scientific fields such as physics, chemistry, biology, and engineering, as well as in social, political, or economic sciences. Depending on the type of information being sought and the complexity of the problem, studies run univariate, bivariate, or multivariate statistical anal-

yses. Univariate analysis involves examining a single variable, while bivariate analysis involves comparing two variables to establish their relationship. In natural and geological phenomena, multiple variables are often involved, necessitating the use of multivariate statistical tools. Principal Component Analysis (PCA) is commonly utilized in such cases, as it can reduce the dimensionality of large datasets, resulting in smaller and more manageable ones. While this reduction may lead to some loss in accuracy, it simplifies the interpretation of the statistical information.

Statistical tools are increasingly used in various fields of geology, such as, among many others the mobility of chemical elements in riverbed sediments and the assessment of the environmental impact (e.g., Martín-Crespo et al., 2012; Shumilin et al., 2015), the soil contamination (e.g., Fernández-Caliani et al., 2009; Ferreira da Silva et al., 2013), characterization of mining tailings and selective chemical extraction of heavy metals (e.g., Favas et al., 2011; Ceniceros-Gómez et al., 2018) or geochemical properties of minerals of pegmatite-hydrothermal origin (e.g., Marks et al., 2013; Roda-Robles et al., 2015).

The aim of this study is to determine how statistical analysis can be used to gather information about the geochemistry of chromites from various geological settings. Univariate, bivariate, and multivariate statistical tools were employed for this purpose. The statistical analyses utilized published chemical data from three different chromite occurrences:

metasomatic chromites from the Finero complex in Italy (Grieco et al., 2004), chromites associated with the arc-related ophiolites of the Dalabute belt in China (Zhou et al., 2001a), and chromites hosted by the mafic-ultramafic bodies from Lipovenki in Ukraine (Gornostayev et al., 2004).

2. Geological framework

2.1 Chromites-PGE in the Finero Complex (Italy)

According to Grieco et al. (2004), chromites of the Finero Complex form decimeter to meter size chromitites pods that occur in the phlogopite peridotites unit of the complex. The latter comprises four units as follows: (1) phlogopite peridotites, phlogopite-bearing harzburgites, and dunites containing the chromitite bodies; (2) complex of layered rocks consisting of alternating cm- to dm-thick cumulates of gabbro, websterite, clinopyroxenite, hornblendite, and anorthosite; (3) amphibole peridotite unit, comprising amphibole lherzolites as cumulate layers (minor dunites and wherlites were also recorded); (4) amphibole- and garnet-bearing gabbros with occasional layers of anorthosites.

The Finero Complex is located within the Ivrea–Verbano Zone, which is pelitic and shows a variable lithology, i.e., from rocks specific to the amphibolite facies to rocks of the granulite facies. The Ivrea–Verbano Zone comprises lower crustal rocks and seems to be a segment of the African plate accreted onto the European plate during the Alpine orogenesis.

Chromites within the phlogopite peridotites of Finero Complex are chromif-

erous spinels that belong to three distinct geochemical types (Grieco, 1998; Grieco et al., 2001): (1) disseminated chromites in harzburgites; (2) massive chromites in chromitites; (3) chromites associated with phlogopite in symplectites. All three types of Cr-spinels are accompanied by PGE.

The presence of phlogopite in the peridotites of the Finero Complex led Grieco et al. (2004) to conclude that they were metasomatized. Various models have been proposed to explain the mechanisms behind these metasomatic processes. For the purpose of this paper, only one of these theories will be briefly outlined. Grieco et al. (2001) identified two distinct and successive metasomatic events. The first event led to the formation of chromitite pods, while the second event, unrelated to the first, involved K metasomatism and the crystallization of phlogopite.

2.2 Chromites of Sartohay deposits in Dalabute ophiolites (China)

Zhou et al. (2001a) studied the Dalabute ophiolite belt and found that it formed from the accumulation of arc-related terranes during the Paleozoic era. The ophiolite belt stretches nearly 100 km along the Dalabute fault in a northeast-southwest direction. The main components of the ophiolites are mantle peridotites and lavas. The volcanic sequences within the ophiolites are elongated in shape and are found alongside the peridotites.

In the Sartohay area, there is no complete section of the ophiolite sequence. However, Zhou et al. (2001a) consider that peridotites seem to come from the mantle located in a transition zone below the crust.

This mantle comprises mostly harzburgites and lherzolites in which bands and lenses of dunites occur. The latter appear mainly as envelopes of the chromitite ore bodies.

Although the chromitites are hosted in harzburgites, being enveloped in dunites, they are also frequently associated, as podiform bodies, with troctolite dykes. For the most part, the chromitites bodies have tabular or lens shapes, being concordant with the foliation of the serpentinized peridotites; their texture is mainly massive or disseminated. Some chromite bodies occur as veins, fracture fillings, and linear or planar segregations.

2.3 Chromites of the Lipovenki mafic-ultramafic massif (Ukraine)

As stated by Gornostayev et al. (2004), there are around 60 mafic-ultramafic massifs related to a North-South-trending suture zone (Golovanevsk suture) developed within the Ukrainian Shield. The evolution over time of the latter produced granulite-gneiss and granite-greenstone terrains as well as intracratonic basins and troughs.

The ultramafic rocks related to the Golovanevsk suture were classified into two associations: dunites-harzburgites and dunites-peridotites-gabbros-norites. The dunites-harzburgites association comprises serpentinized harzburgites and lherzolites, serpentinites, and piroxenites. These are the main rocks of the Lipovenki massif that host important chromite deposits (Gornostayev et al., 2004). The dunites-peridotites-gabbros-norites association appears as mafic-ultramafic intrusions resulting from magmatic differentiation.

In the Lipovenki massif, three separate ultramafic bodies have been identified; they occur as sub-vertical, fault-bonded bodies, and lenses. They mainly consist of highly serpentinized dunites-harzburgites, on top of which a lateritic nickeliferous cover developed. Chromitites ores are hosted within the western one of the three ultramafic bodies. The main ore body consists of massive, nodular, and schlieren-type chromitites. Gornostayev et al. (2004) consider that the high ratio between ore body and host rocks resembles that identified by Zhou et al. (2001b) for the ophiolitic complexes.

3. Analytical techniques and processing of geochemical data

The present section provides information regarding the analytical techniques used by Grieco et al. (2004), Zhou et al. (2001), and Gornostayev et al. (2004) to determine the chemical composition of chromites identified in the three occurrences.

Chromites of the Finero Complex (Italy) were analyzed with the help of an ARL-SEMQ microprobe under the following operating conditions: 15 kV acceleration voltage, a beam current of 20 nA, and 3 μm spot size (Grieco et al., 2004).

The chemical composition of chromites within the Dalabute massif (China) was determined using a Jeol-JXL-7336 microprobe at an accelerating voltage of 15 kV and a beam current of 10 nA (Zhou et al., 2001a).

As for chromites of the Lipovenki massif (Ukraine), Gornostayev et al. (2004)

only specify that they were performed using a Jeol-JCXA-733 electron microprobe without providing the operating parameters.

The univariate and multivariate statistics of geochemical data were conducted using the XLSTAT 2019.2.2 software (Addinsoft, 2024).

To assess the geochemical properties of chromites from each occurrence, univariate statistics have been used, including the determination of central tendency (mean) and dispersion (range, standard deviation, and coefficient of variation).

The geochemical properties of chromites from the three occurrences were analyzed using Principal Component Analysis (PCA). This statistical method helps to simplify large datasets and identify the most important variables (Hou et al., 2017; Cortada et al., 2018; Stumbea et al., 2019). Before conducting PCA, the variables were log-transformed to ensure a normal distribution, and then standardized using z-scores to give them equal weight in the analysis (Güler et al., 2002; Cortada et al., 2018). The contribution of each principal component to the geochemical variables was calculated as part of the analysis.

## 4. Results and discussion

4.1 Univariate statistics

Table 1 shows the univariate statistical parameters calculated for the cations ratio within the crystal chemical formula of the studied chromites. In this analysis, the chromite standard formula provided by the American Mineralogist Crystal Structure Database (2019) was considered:

Tab. 1 Statistics on major elements* in chromites

	Ti	Al	Cr	Fe^{3+}	Fe^{2+}	Mg	Mn	R1	R2
Metasomatic chromites – Finero (*n = 36*)									
Min.	0.004	0.342	0.976	0.038	0.425	0.429	0.007	0.860	1.361
Max.	0.017	0.845	1.562	0.157	0.570	0.578	0.011	1.159	2.581
Mean	0.010	0.596	1.262	0.121	0.478	0.525	0.008	1.011	1.989
SD	0.004	0.093	0.109	0.032	0.031	0.033	0.001	0.065	0.238
CV	0.34	0.16	0.09	0.26	0.07	0.06	0.14	0.27	0.85
Chromites in arc-related ophiolites– Sartohay (*n = 13*)									
Min.	0.001	0.788	0.803	0.040	0.291	0.559	0.003	0.853	1.632
Max.	0.010	1.142	1.163	0.104	0.436	0.703	0.016	1.156	2.419
Mean	0.006	0.945	0.971	0.075	0.353	0.642	0.008	1.003	1.996
SD	0.003	0.094	0.090	0.021	0.059	0.058	0.004	0.120	0.208
CV	0.52	0.10	0.09	0.29	0.17	0.09	0.47	0.72	0.99
Chromites in mafic-ultramafic bodies – Lipovenki (*n = 12*)									
Min.	0.002	0.659	1.086	0.024	0.404	0.389	0.009	0.802	1.771
Max.	0.005	0.796	1.255	0.137	0.596	0.588	0.014	1.198	2.194
Mean	0.004	0.747	1.155	0.094	0.462	0.525	0.010	0.998	2.000
SD	0.001	0.041	0.057	0.043	0.049	0.050	0.001	0.100	0.142
CV	0.26	0.05	0.05	0.46	0.11	0.10	0.14	0.34	0.83

* a.p.f.u. calculated based on analyses published by Grieco et al. (2004) (Finero); Zhou et al. (2001a) (Sartohay); Gornostayev et al. (2004) (Lipovenki); R1 = Fe^{2+}+Mg+Mn; R2 = Cr+Al+Fe^{3+}+Ti; SD – standard deviation; CV – coefficient of variation

$$(Fe^{2+}, Mg, Mn) (Cr, Al, Fe^{3+}, Ti)_2 O_4$$
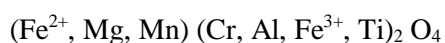
The low coefficients of variation in Table 1 indicate that the means calculated for each statistical population are representative. Data also show that all chromites in the present study have similar cation ratios in their crystal-chemical formula. The exception is aluminum, which has a higher mean in the chromites hosted by arc-related ophiolites (Sartohay) comparing the metasomatic ones (Finero).

4.2 Geochemical correlations by occurrence

The correlations between cation ratios in the chromites of each of the three occurrences were tested with the help of correlation diagrams (Fig. 1). A summary of the ensuing results is presented in Table 2.

As an overview, the diagrams in Figure 1 show negative correlations between either cations or groups of cations, regardless of the occurrence of chromites. In the case of the metasomatic
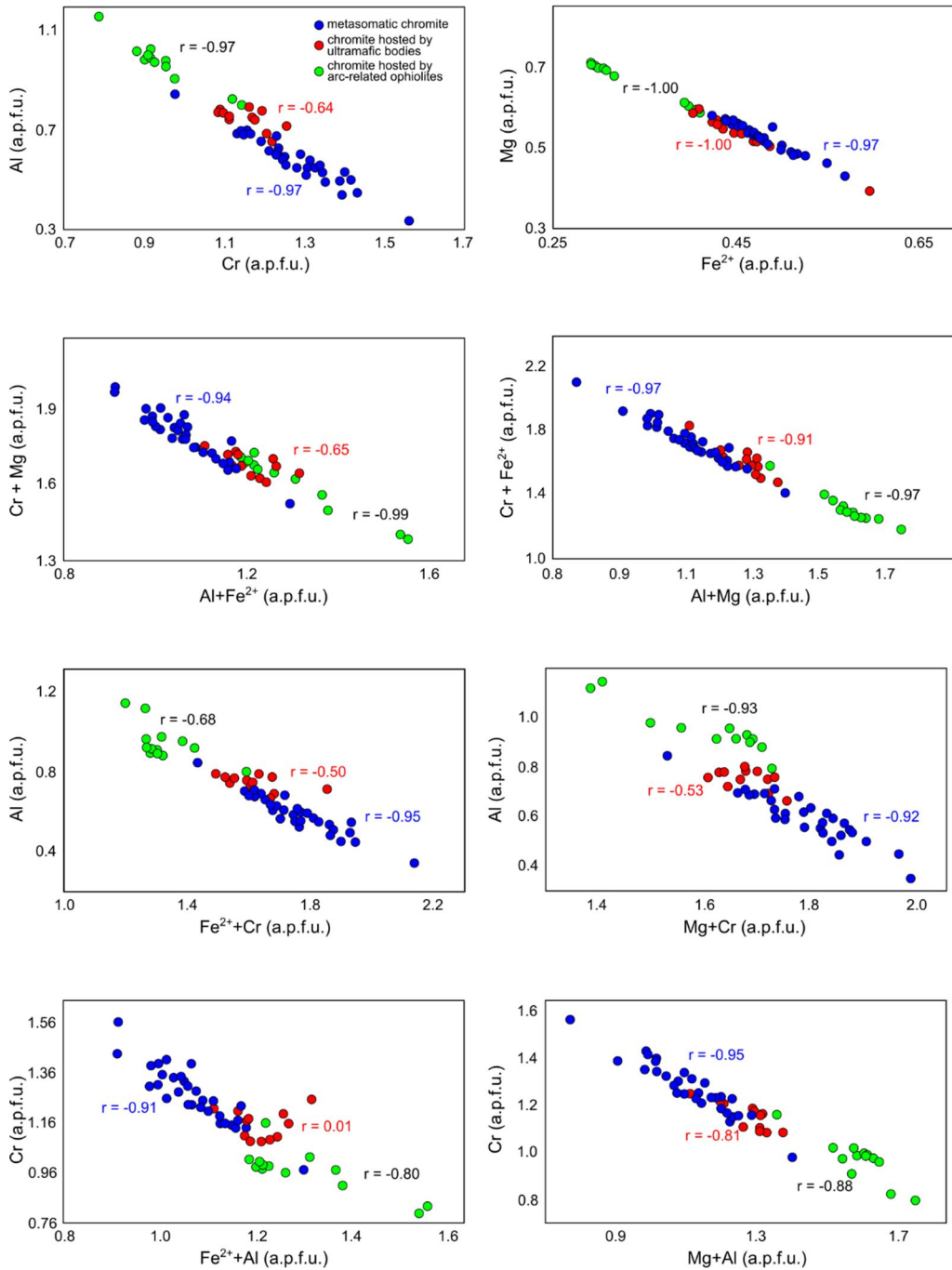
Fig. 1 Diagrams showing geochemical correlations by occurrence.

Tab. 2 Summary of geochemical correlations by occurrence

Correlation	Metasomatic chromites (Finero)	Chromites in arc-related ophiolites (Sartohay)	Chromites in mafic-ultramafic bodies (Lipovenki)
Mg – Fe^{2+}*	●●	●●	●●
(Cr+Fe^{2+}) – (Al+Mg)*	●●	●●	●●
Cr – (Mg+Al)	●●	●●	●●
Al – Cr*	●●	●●	●
(Cr+Mg) – (Al+Fe^{2+})*	●●	●●	●
Al – (Mg+Cr)*	●●	●●	●
Cr – (Fe^{2+}+Al)*	●●	●●	–
Al – (Fe^{2+}+Cr)*	●●	●	●

* negative correlation; ●● – strong correlation; ● – moderate correlation; – no correlation

chromites, the negative correlation can result from specific substitution processes. The ophiolite-related chromites may show negative correlations because of either the growth of chromite crystals in an environment whose chemistry is variable over time or possible substitution processes.

Considering the strong negative correlations (Fig. 1, Tab. 2), the crystallization of the metasomatic chromites (Finero) involved intense substitution processes, as expected. Both simple (e.g., Mg–Fe^{2+}, Al–Cr) and complex substitutions (e.g., [Cr+Fe^{2+}]–[Al+Mg], Al–[Mg+Cr], [Cr+Mg]–[Al+Fe^{2+}]) may be assumed.

Moreover, except for the moderate negative correlation between Al and (Fe^{2+}+Cr), chromites hosted by arc-related ophiolites (Sartohay) show strong negative correlations as well (Fig. 1, Tab. 2). As stated, they can be explained by changes in the composition of the crys-tallization environment. However, small-scale substitution processes cannot be excluded either.

In turn, the only strong correlations identified in chromites hosted by mafic-ultramafic bodies of Lipovenki are Mg–Fe^{2+}, (Cr+Fe^{2+})–(Al+Mg), and Cr–(Mg+Al) respectively (Fig. 1, Tab. 2). The other cations or groups of cations only have a moderate correlation ($0.50 < r < 0.65$) or even a lack of correlation as in the case of Cr – (Fe^{2+}+Al). It can be assumed that the peculiar geochemical properties of these chromites compared to those of the other two occurrences are related to the serpentinization processes of ophiolitic rocks, as described by Gornostayev et al. (2004).

4.3 Multivariate statistics

The geochemical distribution of cations within the chromite structure has also been interpreted using the Principal Component Analysis (PCA) (Fig. 2) and Pearson's correlations matrix (Tab. 3).
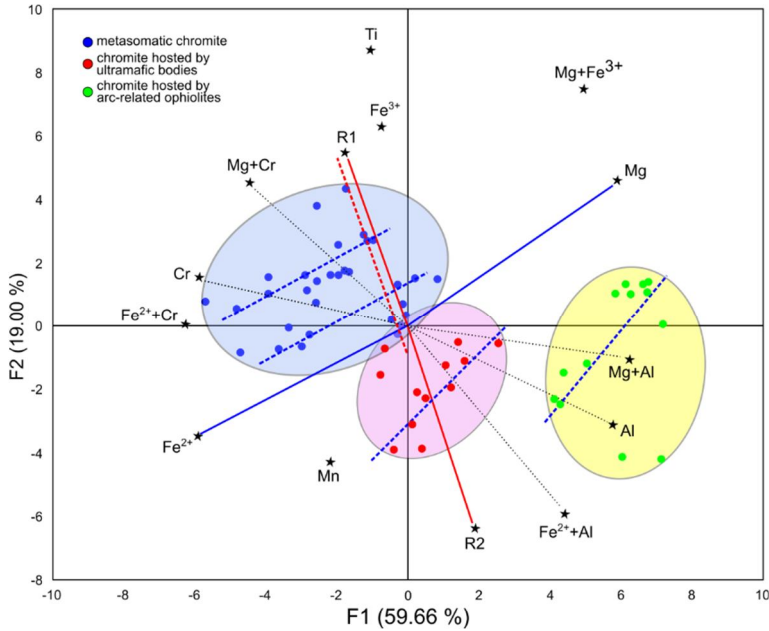
Fig. 2 PCA of chromite geochemical data, using the loadings of 14 variables

PCA considers 14 geochemical variables and used 61 observations; it accounts for 78.66% of all data variations. The first principal component (F1) refers to 59.66% of the total variance, while the second (F2) explains 19.00%.

Component F1 seems to reflect the variation in the ratios $Cr : Fe^{2+}$ and $Al : Mg$ within the chromite structure. The F2 axis discriminates between chromite abundant in Cr and Mg (positive F2) and chromite with a higher ratio of Fe^{2+} and Al (negative F2). At the same time, F2 accounts for the Fe^{3+} abundance and the completion degree of the R1 structural site (positive F2). On the other hand, negative F2 may also point out to Mn-rich chromite, having a more complete R2 structural site.

From this perspective, Figure 1 indicates distinct geochemical properties for the three types of chromites under study. Thus, the metasomatic chromites from the Finero complex show a high abundance of Cr and seemingly Fe^{3+}, as well as a tendency towards a more complete occupancy of the R2 structural site. On the contrary, the geochemistry of chromites associated with mafic-ultramafic bodies of Lipovenki and especially those hosted by the arc-related ophiolites of Sartohay seems to be controlled by Al. In the structure of some of the latter chromites, Mg may have a greater weight.

PCA performed on the geochemical data of chromites, regardless of the occurrence from which they originate (Fig. 2), also suggests some negative

Tab. 3 Pearson's correlation matrix regarding all chromite geochemical data

	Al	Cr	Fe^{2+}	Mg	Fe^{2+}+Cr	Mg+Al	Fe^{2+}+Al	Mg+Cr	Fe^{2+}+Ti	Mg+Ti	Mn+Cr	Mn+Al	R1	R2
Al	1													
Cr	-0.970	1												
Fe^{2+}	-0.705	0.721	1											
Mg	0.673	-0.700	-0.991	1										
Fe^{2+}+Cr	-0.947	0.975	0.857	-0.839	1									
Mg+Al	0.975	-0.960	-0.843	0.821	-0.984	1								
Fe^{2+}+Al	0.931	-0.884	-0.398	0.362	-0.785	0.828	1							
Mg+Cr	-0.894	0.918	0.393	-0.360	0.809	-0.799	-0.955	1						
Fe^{2+}+Ti	-0.725	0.735	0.998	-0.984	0.867	-0.856	-0.425	0.415	1					
Mg+Ti	0.647	-0.680	-0.984	0.998	-0.821	0.800	0.331	-0.335	-0.973	1				
Mn+Cr	-0.969	1.000	0.723	-0.703	0.976	-0.960	-0.882	0.917	0.737	-0.683	1			
Mn+Al	1.000	-0.969	-0.702	0.670	-0.946	0.974	0.933	-0.894	-0.722	0.643	-0.967	1		
R1	-0.349	0.296	0.266	-0.136	0.306	-0.310	-0.314	0.312	0.297	-0.102	0.296	-0.348	1	
R2	0.386	-0.320	-0.248	0.117	-0.317	0.333	0.372	-0.353	-0.287	0.075	-0.319	0.386	-0.985	1

correlations between the abundance of either cations or groups of cations; they agree with the information provided by the Pearson correlation coefficients (Tab. 3): Mg–Fe^{2+} (r = –0.991); Cr–Al (r = –0.970); (Mg+Cr)–(Fe^{2+}+Al) (r = –0.955); (Mg+Al)–(Fe^{2+}+Cr) (r = –0.984); R1–R2 (r = –0.985).

Although correlations occur for all three types of chromites, their meaning appears to be different. Thus, regarding the metasomatic chromites, the correlations can be ascribed to substitutions between cations or groups of cations during the crystallization process. The negative correlations identified for the chromites associated with the arc-related ophiolites (Sartohay) and the Lipovenki ultramafic bodies, respectively, suggest the variant quantitative ratios between the chemical elements during the crystallization from the parental magmas. The correlations also indicate that, regardless of genetic processes, the degrees of occupancy of the structural sites R1 and R2 within chromites are closely related.

In addition, Figure 2 indicates that the negative correlation between Fe^{2+} and Mg occurs for each of the three populations of chemical data. As for the chromites hosted by the arc-related ophiolites (Sartohay) and those associated with mafic-ultramafic bodies (Lipovenki), the negative correlation may indicate that the variation of the Fe^{2+} : Mg ratio occurs at higher Al contents of the parental magmas compared to metasomatic chromites. The latter seems to have suffered at least two events of metasomatic Fe^{2+}–Mg substitutions generated by fluids depleted in Al at

different Cr ratios. At the same time, metasomatic chromites show a greater interdependence of the degree of occupation of the R1 and R2 structural sites compared to the other two types of chromites.

5. Conclusions

The present study aimed to identify the extent to which the use of univariate and multivariate statistics provides information on the geochemical properties of chromites through the participation of constituent cations in the structure of the mineral. The informations brought by this approach are:

(i) The univariate statistics show no apparent large geochemical differences between the metasomatic chromites (Finero complex), those associated with arc-related ophiolites (Sartohay deposits), and chromites hosted by mafic-ultramafic bodies of Lipovenki;

(ii) However, PCA shows the higher control of Cr over the composition of metasomatic chromites, while Al is controlling at different ratios the structure of the other two types of chromites;

(iii) Chromites are defined by negative correlations between several cations or groups of cations. These correlations are strong in the case of metasomatic chromites and those hosted by arc-related ophiolites, respectively moderate in the case of chromites associated with mafic-ultramafic ophiolitic bodies;

(iv) PCA and Pearson correlation coefficients suggest that, regardless of their geological settings, chromites are charac-

terized by the following negative correlations: Mg–Fe^{2+}; Cr–Al; $(Mg+Cr)$–$(Fe^{2+}+Al)$; $(Mg+Al)$–$(Fe^{2+}+Cr)$. As regards the metasomatic chromites, the meaning of these correlations can be explained by simple (between singular cations) or complex (between groups of cations) metasomatic substitution processes. On the other hand, the negative correlations mentioned above seem to suggest the growth of chromite crystals from magmas with variable chemistry in the case of ophiolite-related chromites. In addition to that, either metasomatic or ophiolite-related chromites occur under the variant Cr : Al ratio.

(v) The metasomatic processes strengthen the relationship between the degrees of occupation of the R1 and R2 structural sites of chromites.

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