

The concept of mineral systems and its application to the study of mineral diversity and evolution

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Abstract: The chemical diversity of minerals can be analysed in terms of the concept of mineral systems, defined by the set of chemical elements essential for the definition of a mineral species. Only species-defining elements are considered as essential. According to this approach, all minerals are classified into ten types of mineral systems with the number of essential components ranging from 1 to 10. For all the minerals known today, only 70 chemical elements act as essential species-defining constituents. The number of minerals of different chemical elements are calculated as follows (number of mineral species is given in parentheses): oxygen (4138), hydrogen (2814), silicon (1479), calcium (1182), sulfur (1064), aluminum (989), sodium (953), iron (953), copper (643), arsenic (601), phosphorus (599), and magnesium (576). The distribution of the majority of the species-defining elements among mineral systems submits to a normal distribution. Using the concept of mineral systems, different geological objects can be compared from the viewpoint of their mineral diversity as exemplified by alkaline massifs (Khibiny, Lovozero, Russia, and Mont Saint-Hilaire, Canada), evaporite deposits (Inder, Kazakhstan, and Searles Lake, USA) and fumaroles at active volcanoes (Tolbachik, Kamchatka, Russia, and Vulcano, Sicily, Italy). The concept of mineral systems can be applied to mineral evolution overall by calculating the mean number of elements for the first three stages in the evolution of minerals as proposed by R.M. Hazen and co-authors in 2008, plus a fourth period corresponding to Hazen's stages 4–10, as follows: 2.08 ± 0.45 (I: ur-minerals); 2.68 ± 0.13 (II: minerals of chondritic meteorites); 3.86 ± 0.07 (III: Hadean minerals); 4.50 ± 1.47 (IV: post-Hadean minerals).

Keywords: mineral species; species-defining constituent; crystal-chemical formula; mineral systems; fumaroles; volcanoes; alkaline massifs; mineral evolution.

1. Introduction

Recent contributions to the studies of mineral evolution on the Earth (Hazen *et al.*, 2008, 2009, 2011, 2012, 2014; Hazen & Ferry, 2010; Krivovichev, 2013a and b; Grew & Hazen, 2014) have attracted considerable attention to the problems of mineral diversity and mineral evolution through space and time.

The concept of mineral evolution was applied to clay minerals (Hazen *et al.*, 2013a) and the minerals containing essential U and Th (Hazen *et al.*, 2009), Hg (Hazen *et al.*, 2012), C (Hazen *et al.*, 2013b), Be (Grew & Hazen, 2014) and B (Grew *et al.*, 2016), leading to the formulation of “mineral ecology” as a field of mineralogy dealing with the study of the distribution, diversity, complexity, and abundance of minerals, including the interactions of minerals with the environment. Mineral ecology applies statistical analysis of large mineralogical

data resources to estimating potential diversity, not only on Earth, but also on other planetary bodies (Hazen *et al.*, 2015a and b; Hystad *et al.*, 2015a and b).

Hazen *et al.* (2008) defined three eras and ten stages of mineral evolution in the Earth's history and reported three lists of minerals (Hazen *et al.*, 2008; Hazen, 2013) to estimate evolution of minerals on the basis of mineral diversity. The law of increasing mineral diversity (stated by Tschermak and developed by Yushkin, 1982) has now been supported by the large sets of mineralogical data accumulated over the last two hundred years: “...owing to the continuing transformation of minerals, the diversity of the Earth's crust composition is increasing... The permanent increasing diversity could be called the development of the mineral kingdom” (Yushkin, 1982). Yushkin (1982) identified as an important task “...a search and use in a comparative-historical analysis of specific indicators, which, by characterization of complex mineralogical

objects, would contain truly mineralogical information”. Therefore, it becomes necessary to elaborate on quantitative methods for the estimation of mineral diversity. One of the possible methods is to evaluate the number of different mineral species that occur in a particular geological object such as vein, deposit, massif, and Earth’s crust. However, the mere numbers of different mineral species do not provide enough information on chemical diversity and complexity of mineral associations.

An alternative and more elaborate approach was recently proposed by Krivovichev & Charykova (2013a and b) and Grew *et al.* (2016) on the basis of the concept of mineral systems. According to this approach, any mineral can be assigned to a particular “mineral system”. This is the chemical system consisting of the set of the minimum number of chemical elements (essential constituents of its formula) required to stabilize the crystal structure of the mineral. Using the concept of mineral systems makes it possible to compare different geological objects according to their mineralogical compositions and to evaluate their mineral and chemical diversity (Krivovichev & Charykova, 2015, 2016, 2017) by means of numerical characteristics with reasonably strong discriminative power.

In this paper, we provide an outline of the concept of mineral systems from basic definitions to possible applications and directions for further development.

2. Basic definitions and methods

2.1. Mineral species

In this study we consider only mineral species *sensu stricto*, *i.e.*, the “elementary substances or chemical compounds crystallized as a result of geo- or cosmochemical process in natural physicochemical systems” (Bokii, 1997).

In order to have an appropriate database at hand, we have compiled a list of all mineral species (5072) known up to 2016 using continuously updated lists published by Pasero (2016) at the website of Commission on New Minerals, Nomenclature and Classification, International Mineralogical Association (CNMNC IMA) and CNMNC IMA Newsletters (*e.g.* Hålenius *et al.*, 2017). The crystal chemical formulae of these minerals have been rewritten in a canonical way in order to determine the minimal set of species-defining constituents (*i.e.*, chemical elements that are dominant in particular sites in a crystal structure). This point is of crucial importance, because crystal chemical formulae of the same mineral species are written differently in different texts, reference books and the IMA list. Unfortunately, this problem occurs in most of known mineral databases (*e.g.* <http://www.mindat.org>, <http://www.webmineral.com>) that frequently provide mineral formulae taken from the literature without critical consideration.

It should be noted that the assignment of an element as species-defining (*cf.* Hawthorne, 2002; Hatert & Burke, 2008; Krivovichev & Charykova, 2013a; Christy, 2015; Grew *et al.*, 2016) is determined by the rules of new mineral species definition (Mandarino *et al.*, 1984; Nickel &

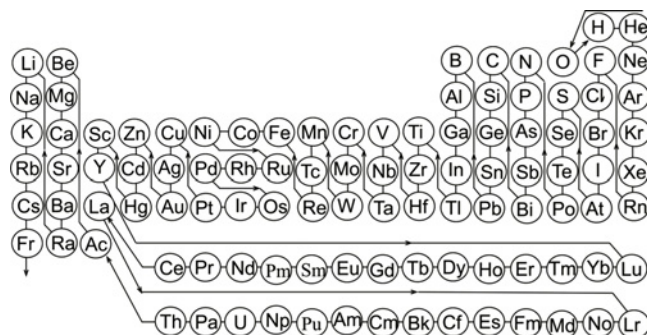


Fig. 1. Thermochemical sequence of chemical elements and corresponding single-component systems.

Mandarino, 1987; Nickel, 1992, 1995; Nickel & Grice, 1998). In isovalent and heterovalent series, we use representative end-member compositions to indicate the chemistry of mineral species, bearing in mind that this is a slight oversimplification in cases where species-end-member mapping is not one-to-one (*cf.* the pyrochlore nomenclature report, Atencio *et al.*, 2010). For example, in the forsterite–fayalite solid-solution series, two mineral species are considered: forsterite, $\text{Mg}_2(\text{SiO}_4)$, and fayalite, $\text{Fe}_2(\text{SiO}_4)$. By analogy, in the albite–anorthite heterovalent solid-solution series, only two mineral species are considered, *i.e.* albite, $\text{Na}(\text{AlSi}_3\text{O}_8)$, and anorthite, $\text{Ca}(\text{Al}_2\text{Si}_2\text{O}_8)$. It is noteworthy that only species-defining elements were taken into account in the attribution of a mineral to a certain mineral system (see below), whereas other isomorphic elements, albeit perhaps geochemically important, were omitted. In some cases, when charge-balance considerations requires two elements with different valence to be present in the same site, as for some end-members discussed by Hawthorne (2002) and Hatert & Burke (2008), both have been regarded as essential.

2.2. Mineral systems

Any mineral can be assigned to a particular system, the components of which are the species-defining chemical elements required to stabilize the crystal structure of the mineral. We note that a very similar approach has been used in thermodynamics in order to systematize, store, and search thermochemical information on different chemical substances. The main rule of coding of minerals is to determine the sequence of chemical elements that are essential in their formulae. There are definite rules to denote (encode) thermodynamic systems, which determine the consecutive recording of element symbols. For example, in most basic reference books on thermodynamic data, the systematics are based on the thermochemical sequence of chemical elements and corresponding single-component systems (Fig. 1). Notations (formulae) of two-, three-, four-component and other complex systems are determined by their thermochemical sequence. For example, the crystal chemical formulae of anorthite, $\text{Ca}(\text{Al}_2\text{Si}_2\text{O}_8)$, or phlogopite, $\text{KMg}_3(\text{AlSi}_3\text{O}_{10})(\text{OH})_2$, corre-

Table 1. Examples of mineral systems.

n^*	Mineral system	Mineral	Formula
1	Bi	Bismuth	Bi
2	AlCu	Cupalite	CuAl
3	OSiAl	Mullite	$\text{Al}_6\text{Si}_2\text{O}_{13}$
4	OHCIFe	Akaganeite	$\text{Fe}_8[(\text{OH})_9\text{O}_7]_{\Sigma 16}\text{Cl}\cdot n\text{H}_2\text{O}$
5	OAsSbFeCa	Stenhuggarite	$\text{CaFe}_3\text{Sb}_3(\text{AsO}_3)_2\text{O}$
6	OCISeCuCdK	Burnsite	$\text{KCdCu}_7(\text{SeO}_3)_2\text{O}_2\text{Cl}_9$
7	OFPSiTiCaNa	Polyphite	$\text{Na}_9\text{Ca}_2\text{Ti}_2(\text{Si}_2\text{O}_7)(\text{PO}_4)_3\text{O}_2\text{F}_2$
8	OHFPCeCaSrNa	Deloneite	$(\text{NaCe})_{\Sigma 2}\text{Ca}_5\text{Sr}_3(\text{PO}_4)_6\text{F}(\text{OH})$
9	OHFCISCYBaNa	Mineevite-(Y)	$\text{Na}_{25}\text{BaY}_2(\text{CO}_3)_{11}(\text{HCO}_3)_4(\text{SO}_4)_2\text{F}_2\text{Cl}$
10	OHSCSiZrNbFeCaNa	Golyshevite	$\text{Na}_{10}\text{Ca}_9\text{Fe}_2\text{Zr}_3\text{SiNb}(\text{Si}_{24}\text{O}_{72})(\text{CO}_3)(\text{OH})_3\cdot\text{H}_2\text{O}$

* Number of species-defining chemical elements per mineral system.

spond to the OSiAlCa and OHSiAlMgK systems, respectively (Krivovichev & Charykova, 2013a). Based on the sets of essential, species-defining chemical elements in minerals, n -component mineral systems are allocated (where $n = 1-10$) for all known mineral species (5072 in our dataset). It should be noted that Rieder (2016) reported that "...the number of elements in end-member mineral formulae ranges from one (native elements) to sixteen (hellandite-(Ce))". However, this cannot be considered as a correct formulation, as the crystal-chemical formula of hellandite-(Ce) should be written as $(\text{Ca}_3\text{REE})_{\Sigma=4}\text{Ce}_2\text{Al}\square_2(\text{B}_4\text{Si}_4\text{O}_{22})(\text{OH})_2$ (Oberti *et al.*, 1999, 2002). Hence the mineral system of hellandite-(Ce) is OHSiBaICeREECa, *i.e.* $n = 8$ (or 7 if REE = Ce).

Our approach allows to organize mineral species using their chemical composition and to arrange the existing data into coherent frameworks emphasizing changes in mineral diversity and composition through space and time. The concept of mineral systems can be used to assess mineral diversity of geological objects in more detail than the simple number of different mineral species present in a particular association. Examples of mineral systems are given in Table 1.

3. Results

3.1. Classification of mineral systems

According to the number of species-defining elements, all mineral systems can be subdivided into one-, two-, three-, and n -component mineral systems. For example, all native elements (diamond, graphite, and platinum) belong to one-component mineral systems; minerals consisting of two elements (quartz, corundum, hematite, and pyrite) are assigned to two-component mineral systems, whereas minerals consisting of three elements (forsterite, enstatite, spinel, and chromite) belong to three-component mineral systems.

Table 2 shows the distribution of minerals among different mineral systems depending on the number n of species-forming elements. The maximum value of n is ten, and there are only eleven mineral species that belong to the ten-component system. The distribution (Table 2, column 2) follows the Gauss law (Fig. 2) with a significant

determination coefficient ($R^2 = 0.982$). The highest number of minerals corresponds to four- and five-component systems (1362 and 1437 mineral species, respectively). Note that all minerals composed of six or more chemical elements are oxygen-bearing; among five-element minerals, 90% are oxygen-bearing; and among four-element minerals, they amount to 80%. All systems could be divided into two large groups: (i) non-recurrent or monomineral systems that contain one mineral only (Table 2, column 3) and (ii) polymineral systems that contain more than one mineral (Table 2, column 4). For example, the OHSiAlCa system is polymineral and contains 34 mineral species (Table 3). In order to distinguish these minerals, stoichiometric formulae are obviously needed. It should be noted that, in general, the structure type should be considered for minerals that occur in several polymorphic modifications, which should be distinguished from the minerals that have no polymorphs (*e.g.*, amstallite and scolecite with the same atomic Ca:Al:Si:H:O ratio).

According to this approach, all minerals can be classified uniquely forming natural thermodynamic classification of mineral species (Bokii, 1997) complementary to traditional classification schemes (Strunz & Nickel, 2001; Mills *et al.*, 2009; Bulakh *et al.*, 2014). A certain advantage of this scheme is its formal and unambiguous character, though we are quite aware that certain amount of essential information is lost.

3.2. Essential constituents of known mineral species

The number of mineral species formed by a particular chemical element as a species-defining component may serve as a measure of the capability of this element to form minerals (Povarennykh, 1966; Ivanov & Yushko-Zakharova, 1989; Yaroshevskiy, 2008; Nikolaev, 2009; Krivovichev & Charykova, 2013a, 2015; Christy, 2015). Using the criteria outlined above, 70 elements (Table 4) were found to be essential constituents of known mineral species (Krivovichev & Charykova, 2013a, 2015; Christy, 2015). No minerals are known for He, Ne, Ar, Kr, Xe, Rn, Tc, Pm, Po, At, Ra, Ac, Pa, Pr, Pm, Eu, Tb, Dy, Ho, Er, Tm, Lu, and therefore these elements have been excluded from the current study. Rieder (2016) reported another six

Table 2. Distribution of minerals according to the number of the species-forming elements.

n^*	NM**	NM that belong to monomineral systems	NM that belong to polymineral systems
1	44	23	21
2	405	103	302
3	726	299	427
4	1362	351	1011
5	1437	558	879
6	645	410	235
7	295	226	69
8	119	99	20
9	28	22	6
10	11	9	2
Total	5072	2105	2967

* Number of species-defining chemical elements per mineral system.

** Number of minerals.

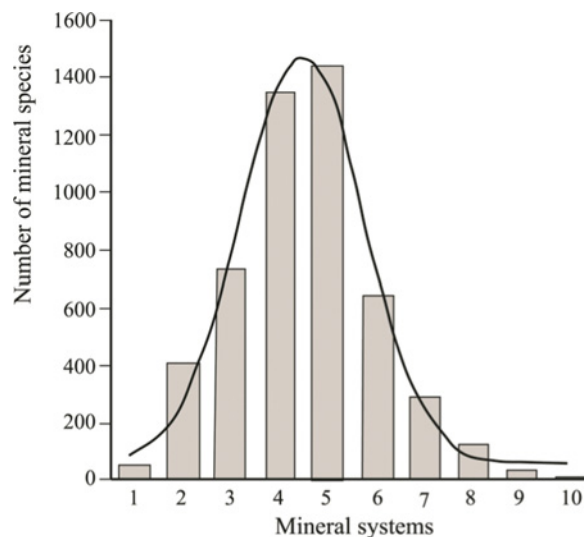
Fig. 2. Total number of mineral species *versus* number of mineral systems (species-defining elements).

Table 3. The OHSiAlCa system comprising 34 mineral species.

No.	Code of mineral	Mineral species	Crystal-chemical formula
1	O ₅ HSiAlCa	Vuagnatite	CaAl(SiO ₄)(OH)
2	O ₈ H ₂ SiAl ₂ Ca ₂	Bicchulite	Ca ₂ (Al ₂ SiO ₆)(OH) ₂
3	O ₈ H ₂ SiAl ₂ Ca ₂	Kamaishilite	Ca ₂ (Al ₂ SiO ₆)(OH) ₂
4	O ₁₀ H ₄ Si ₂ Al ₂ Ca	Lawsonite	CaAl ₂ (Si ₂ O ₇)(OH) ₂ ·H ₂ O
5	O ₁₂ H ₂ Si ₂ Al ₄ Ca	Margarite	CaAl ₂ (Al ₂ Si ₂ O ₁₀)(OH) ₂
6	O ₁₂ H ₂ Si ₃ Al ₂ Ca ₂	Prehnite	Ca ₂ Al(AlSi ₃ O ₁₀)(OH) ₂
7	O ₁₂ H ₄ Si ₂ Al ₂ Ca ₃	Holtstamite	Ca ₃ Al ₂ (SiO ₄) ₂ (OH) ₄
8	O ₁₃ H ₆ Si ₃ Al ₂ Ca	Amstallite	CaAl(AlSi ₃ O ₈)(OH) ₄ ·H ₂ O
9	O ₁₃ H ₆ Si ₃ Al ₂ Ca	Scolecite	Ca(Al ₂ Si ₃ O ₁₀)·3H ₂ O
10	O ₁₃ HSi ₃ Al ₃ Ca ₂	Clinozoisite	Ca ₂ Al ₃ (SiO ₄)(Si ₂ O ₇)O(OH)
11	O ₁₃ HSi ₃ Al ₃ Ca ₂	Zoisite	Ca ₂ Al ₃ [SiO ₄][Si ₂ O ₇]O(OH)
12	O ₁₄ H ₃ Si ₃ Al ₃ Ca ₂	Pumpellyite-(Al)	Ca ₂ AlAl ₂ (SiO ₄)(Si ₂ O ₇)(O(OH))·H ₂ O
13	O ₁₄ H ₄ SiAlCa	Wairakite	Ca(AlSi ₂ O ₆) ₂ ·2H ₂ O
14	O ₁₆ H ₁₂ Si ₃ Al ₂ Ca	Cowlesite	Ca(Al ₂ Si ₃ O ₁₀)·6H ₂ O
15	O ₁₆ H ₈ Si ₄ Al ₂ Ca	Laumontite	Ca(Al ₂ Si ₄ O ₁₂)·4H ₂ O
16	O ₁₇ H ₈ Si ₆ Al ₂ Ca	Yugawaralite	Ca(Al ₂ Si ₆ O ₁₆)·4H ₂ O
17	O ₂₁ H ₁₀ Si ₄ Al ₄ Ca ₂	Parthéite	Ca ₂ (Al ₄ Si ₄ O ₁₅)(OH) ₂ ·4H ₂ O
18	O ₂₁ H ₁₀ Si ₆ Al ₂ Ca	Goosecreekite	Ca(Al ₂ Si ₆ O ₁₆)·5H ₂ O
19	O ₂₄ H ₁₆ Si ₄ Al ₄ Ca ₂	Gismondine	Ca ₂ (Al ₄ Si ₄ O ₁₆)·8H ₂ O
20	O ₂₄ H ₁₆ Si ₆ Al ₂ Ca	Tschernichite	Ca(Al ₂ Si ₆ O ₁₆)·8H ₂ O
21	O ₃₅ H ₂₂ Si ₈ Al ₄ Ca ₂	Gmelinite-Ca	Ca ₂ (Al ₄ Si ₈ O ₂₄)·11H ₂ O
22	O ₃₇ H ₂₆ Si ₈ Al ₄ Ca ₂	Chabazite-Ca	Ca ₂ (Al ₄ Si ₈ O ₂₄)·13H ₂ O
23	O ₃₇ H ₂₆ SiAl ₂ Ca	Chantalite	CaAl ₂ (SiO ₄)(OH) ₄
24	O ₄₄ H ₂₄ Si ₁₀ Al ₆ Ca ₃	Phillipsite-Ca	Ca ₃ (Al ₆ Si ₁₀ O ₃₂)·12H ₂ O
25	O ₄₆ H ₅₆ Si ₅ Al ₈ Ca ₈	Vertumnite	Ca ₈ Al ₄ (Al ₄ Si ₅ O ₁₂)(OH) ₃₆ ·10H ₂ O
26	O ₅₃ H ₃₄ Si ₁₂ Al ₆ Ca ₃	Lévyne-Ca	Ca ₃ (Al ₆ Si ₁₂ O ₃₆)·17H ₂ O
27	O ₅₇ H ₅₈ Si ₄ Al ₈ Ca ₈	Strätlingite	Ca ₈ Al ₄ (Al ₄ Si ₄ O ₈ (OH) ₁₆)(OH) ₂₄ ·9H ₂ O
28	O ₆₁ H ₂₆ Si ₂₀ Al ₄ Ca ₂	Dachiardite-Ca	Ca ₂ (Al ₄ Si ₂₀ O ₄₈)·13H ₂ O
29	O ₆₉ H ₃₆ Si ₁₈ Al ₂ Ca ₁₂	Tacharanite	Ca ₁₂ (Al ₂ Si ₁₈ O ₃₃)(OH) ₃₆
30	O ₈₀ H ₃₂ Si ₁₈ Al ₅ Ca ₃	Epistilbite	Ca ₃ (Al ₅ Si ₁₈ O ₄₈)·16H ₂ O
31	O ₉₂ H ₄₀ Si ₃₀ Al ₄ Ca ₃	Clinoptilolite-Ca	Ca ₃ (Al ₆ Si ₃₀ O ₇₂)·20H ₂ O
32	O ₁₀₂ H ₆₀ Si ₂₆ Al ₁₀ Ca ₅	Erionite-Ca	Ca ₅ (Al ₁₀ Si ₂₆ O ₇₂)·30H ₂ O
33	O ₁₁₈ H ₆₈ Si ₃₂ Al ₁₀ Ca ₅	Paulingite-Ca	Ca ₅ (Al ₁₀ Si ₃₂ O ₈₄)·34H ₂ O
34	O ₁₆₀ H ₁₂₈ Si ₈ Al ₁₄ Ca ₇	Faujasite-Ca	Ca ₇ (Al ₇ Si ₁₇ O ₄₈) ₂ ·64H ₂ O

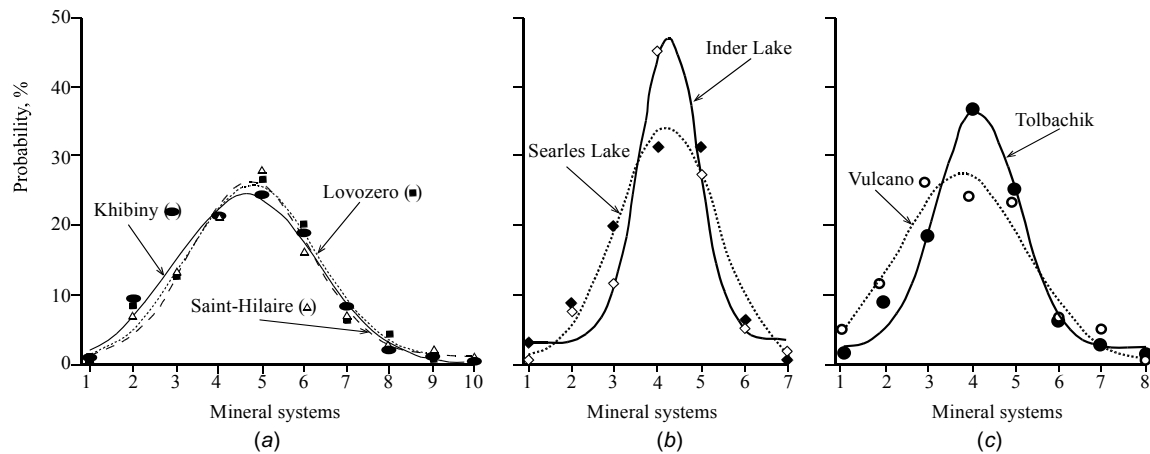


Fig. 3. Distribution of mineral species by mineral systems based on number of species-defining elements in peralkaline plutons (a), in evaporite deposits (b) and in the products (minerals) of fumarole activity (c).

Table 4. Number of mineral species (NM) and crustal abundance for 70 essential species-defining chemical elements.

No.	Symbol	NM	NM (in %)	Crustal abundance by atoms (ppm)*	No.	Symbol	NM	NM (in %)	Crustal abundance by atoms (ppm)*
1	O	4115	81.58	599 000	36	Nb	111	2.20	4.5
2	H	2800	55.51	28 900	37	Sr	112	2.22	88
3	Si	1471	29.16	209 000	38	N	100	1.98	28
4	Ca	1167	23.14	21 500	39	Hg	92	1.82	0.0088
5	S	1056	20.94	227	40	Sn	92	1.82	0.40
6	Al	985	19.39	63 400	41	Cr	23	0.46	41
7	Na	949	18.81	21 300	42	Pd	65	1.29	0.0029
8	Fe	945	18.73	21 000	43	Tl	63	1.25	0.086
9	Cu	636	12.61	20	44	Mo	58	1.15	0.26
10	P	597	11.84	705	45	Co	58	1.15	8.8
11	As	594	11.78	0.50	46	Ta	49	0.97	0.23
12	Mg	571	11.32	19 900	47	W	40	0.79	0.14
13	Mn	548	10.86	359	48	La	38	0.75	5.8
14	Pb	488	9.67	1.4	49	Pt	28	0.55	0.00053
15	K	418	8.29	11 100	50	Ge	29	0.57	0.43
16	C	389	7.71	346	51	Au	29	0.57	0.00042
17	F	352	6.98	585	52	Cd	25	0.50	0.028
18	Cl	353	7.00	145	53	I	24	0.48	0.074
19	Ti	314	6.22	2450	54	Nd	23	0.46	6.0
20	B	276	5.52	19	55	Th	22	0.44	0.86
21	U	242	4.80	0.24	56	Cs	22	0.44	0.47
22	Sb	230	4.56	0.034	57	Sc	16	0.32	10.2
23	Zn	216	4.28	22	58	Rh	13	0.26	0.0002
24	Ba	215	4.26	64	59	Ir	13	0.26	0.00011
25	V	215	4.26	49	60	In	12	0.24	0.045
26	Bi	208	4.12	0.00085	61	Br	8	0.16	0.62
27	Te	159	3.15	0.00016	62	Ga	5	0.10	5.7
28	Ag	160	3.17	0.014	63	Ru	5	0.10	0.0002
29	Ce	135	2.68	9.9	64	Yb	5	0.10	0.38
30	Ni	130	2.58	30	65	Os	4	0.08	0.00016
31	Be	121	2.42	6.5	66	Rb	3	0.06	22
32	Se	117	2.32	0.013	67	Sm	2	0.04	0.97
33	Y	114	2.26	7.7	68	Re	2	0.04	0.000078
34	Zr	114	2.26	38	69	Hf	1	0.02	0.35
35	Li	109	2.16	60	70	Gd	1	0.02	0.82

* Number of atoms in parts per million according to Christy (2015).

essential, species-defining constituents (the number of minerals is given in parentheses): Dy (9), Pr (6), Er (6), Lu (3), Ho (2), Tm (2), whereas Hazen (2013) and Hazen *et al.*

(2015a) gave only Dy (1) and Er (1) in addition to the 70 elements in our list. These differences are due to the fact that Dy, Pr, Er, Lu, Ho, Tm are included in the IMA-

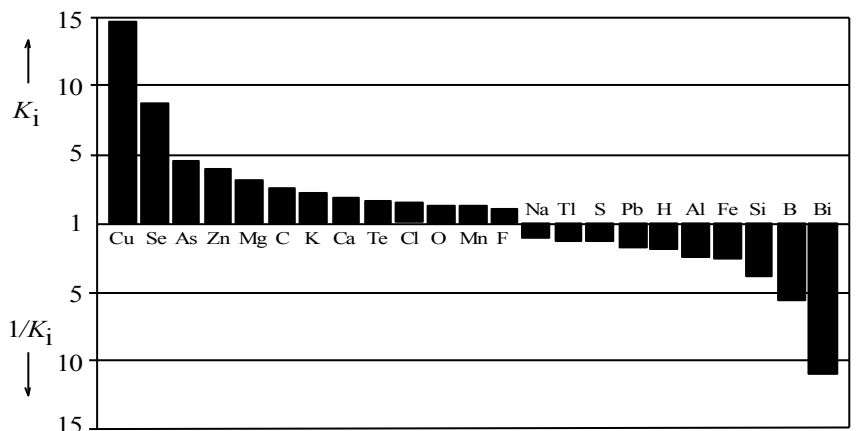


Fig. 4. The concentration coefficient $K_i = \text{NM (in \% (Tolbachik))} / \text{NM (in \% (Vulcano))}$ (see Table 6).

Table 5. The number of minerals of Be and B in highly alkaline complexes (Mont Saint-Hilaire, Lovozero, and Khibiny) and in the Earth crust (NM = number of minerals).

Element	Complexes						The Earth's crust (see Table 4)	
	Mont Saint-Hilaire (400 species)		Lovozero (385 species)		Khibiny (518 species)		NM	NM (in %)
	NM	NM (in %)	NM	NM (in %)	NM	NM (in %)		
Be	20	5.0	13	3.38	14	2.70	121	2.42
B	14	3.5	5	1.30	3	0.58	276	5.52

approved mineral formulae, though they are chemically subordinate, *i.e.* play the role of isomorphic admixtures not reaching 50% occupancy in particular structural sites.

Seventy species-defining chemical elements are distributed among the mineral systems as follows (system type in *italic*, number of elements in bracket): *1* (29), *2* (62), *3* (68), *4* (61), *5* (61), *6* (55), *7* (49), *8* (38), *9* (28), and *10* (19). The numbers of mineral species formed by these elements are given in Table 4, which provides important information on the ability of chemical elements to form mineral species. For example, oxygen, hydrogen and silicon are the species-defining elements for 81.6%, 55.63%, and 29.24% of all mineral species known today. These data are substantially different from global atomic values that show the relative abundances of elements in the Earth's crust as relative numbers of atoms (% or ppm) calculated from the mean contents of elements in wt%. Hence, in the case of global atomic values, it is not important whether the element is species-defining or an isomorphic admixture. Therefore, the number of mineral species does not really coincide with the mean atomic value of certain elements. For example, the global atomic values of S and Al are drastically different (227 and 63 400 ppm, respectively), whereas the global mineral values are similar (20.94% and 19.39%, respectively). These mismatches result from many causes and, probably, there are many factors limiting the number of minerals in nature with common positive trend between the crustal abundance of an element and the number of mineral species incorporating that element (Urusov, 2010; Christy, 2015; Hazen *et al.*, 2015a).

4. Discussion

4.1. Comparative analysis of the mineral systems distribution in various settings

It is of special interest to compare mineral distributions in various geological objects on the basis of the concept of mineral systems outlined above. Below we provide several examples of such an analysis and demonstrate the usefulness of the proposed approach to evaluate mineral diversity of specific geochemical environments.

4.1.1. Highly alkaline massifs: Khibiny, Lovozero and Mont Saint-Hilaire

Alkaline complexes of the Kola Peninsula (Khibiny and Lovozero) and Quebec (Mont Saint-Hilaire) are composed of peralkaline igneous rocks and their products, which are pegmatites and hydrothermal assemblages abnormally enriched in rare lithophile elements (Nb, Zr, REE, Sr, Ba) and volatiles (Krivovichev & Charykova, 2015). Pekov (2001) considered similar alkaline complexes as champion localities, which have extensive lists of different minerals, including mineral species discovered for the first time within these plutons. In total, 761 mineral species have been described from these complexes (Table S1 in the Supplementary Material, freely available online as supplementary material linked to this article on the GSW website of the journal, <http://eurjmin.geoscienceworld.org>). The distributions of mineral species among various mineral systems within Khibiny,

Table 6. The number of minerals of the essential, species-defining chemical elements and K_i values in the products (minerals) of fumarole activity (Tolbachik and Vulcano) (NM=number of minerals).

Element	Locality				K_i^{**}
	Tolbachik		Vulcano		
	NM	NM (in %)*	NM	NM (in %)	
O	133	89.86	78	66.67	1.35
Cu	75	50.68	4	3.42	14.82
S	58	39.19	60	51.28	0.76
K	49	33.11	17	14.53	2.28
Cl	49	33.11	27	23.08	1.43
H	39	26.35	57	48.72	0.54
Na	28	18.92	21	19.27	0.98
Mg	24	16.22	6	5.13	3.16
As	23	15.54	4	3.43	4.53
Ca	18	12.16	8	6.84	1.78
V	15	10.13	0	0	–
F	14	9.46	11	9.40	1.01
Al	13	8.78	25	21.37	0.41
Fe	12	8.11	26	21.10	0.38
Se	11	7.43	1	0.85	8.74
Pb	10	6.76	14	11.97	0.56
C	10	6.76	3	2.56	2.64
Zn	10	6.76	2	1.71	3.95
Tl	4	2.70	4	3.42	0.79
Si	3	2.03	4	7.69	0.26
Mn	3	2.03	2	1.71	1.19
Mo	3	2.03	0	0	–
Ti	2	1.35	0	0	–
B	2	1.35	9	7.69	0.18
Bi	2	1.35	17	14.53	0.09
Te	2	1.35	1	0.85	1.59
Cd	1	0.68	0	0	–
P	0	0	2	1.71	0
Sb	0	0	1	0.85	0
Ba	0	0	1	0.85	0
N	0	0	7	5.98	0
Sn	0	0	2	1.71	0
I	0	0	1	0.85	0
Br	0	0	1	0.85	0
Au	0	0	1	0.85	0

* NM (in %)=[NM of a given element at Tolbachik (or Vulcano)/total number of minerals at Tolbachik (or Vulcano)] (in %).

** The concentration coefficient K_i = NM (in %) (Tolbachik)/NM (in %) (Vulcano).

Lovozero and Mont Saint-Hilaire are only slightly different (Fig. 3a) and, for all three plutons, they fit the normal law.

The numbers of minerals of species-defining elements from the three plutons under consideration are given by Krivovichev & Charykova (2015), who demonstrated that, for the three plutons, Si, Na, K, C, F, Ti, Ce, Zr, Nb, Sr, and Th can be considered as excess species-defining elements as compared with the relative number of minerals in the Earth's crust, whereas Mg, Mn, Ca, S, Cu, Pb, Cl, Te, Ag, and Ni are deficient. The opposite is true for average values of some petrogenetic elements of alkaline rocks in the Kola Peninsula. According to Kukharensko *et al.* (1965), the concentration of Si, Al, and Na are significantly less than

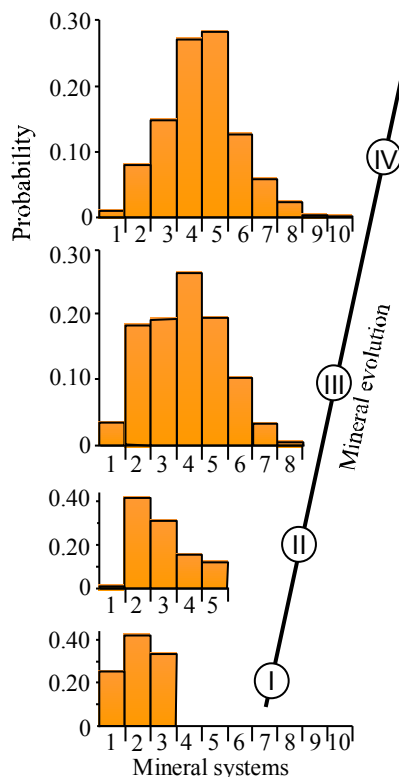


Fig. 5. Distribution of mineral species in the four groups of minerals (I–IV). (Online version in color.)

crustal abundance for these elements, whereas Mg, Mn, and Ca are excess elements. These differences are less evident for other petrogenetic elements.

Special attention was paid to the diversity of Be and B minerals, which was recently discussed by Grew & Hazen (2014) and Grew *et al.* (2016), respectively. The two papers are of particular relevance for our study, because the minerals in question are from the same localities (Mont Saint-Hilaire, Lovozero, Khibiny). The data obtained show that the relative number of Be minerals in the three complexes is higher than the relative number of Be minerals in the Earth's crust (Table 5). For B minerals, the opposite is true, the relative content of B minerals in the highly alkaline complexes is much less than in the Earth's crust as a whole (Table 5).

4.1.2. Evaporites: Inder, Kazakhstan, and Searles Lake, USA

The comparative analysis of the distribution of mineral species in evaporite deposits (Inder, Kazakhstan, and Searles Lake, USA; Table S2) was reported by Krivovichev & Charykova (2016). The data (Fig. 3b) demonstrate that these deposits are very similar and the distributions again fit the normal law.

It can be shown that Ca, K, Na, Cl, B, and S are the “superfluous” essential elements present in minerals of these evaporites in increased amounts, whereas Si and Al are scarce. Krivovichev & Charykova (2016) reported that the analysis of mineral composition of evaporites by means of

Table 7. (continued).

Element	I		II		III		IV	
	Ur-minerals (12 species)		Chondrite minerals (60 species)		Hadean minerals (425 species)		Post Hadean minerals (5072 species)	
	NM	NM, %	NM	NM, %	NM	NM, %	NM	NM, %
Br	0	0	0	0	0	0	8	0.16
Ru	0	0	0	0	0	0	5	0.10
Yb	0	0	0	0	0	0	5	0.10
Ga	0	0	0	0	0	0	5	0.10
Os	0	0	0	0	0	0	4	0.08
Rb	0	0	0	0	0	0	3	0.06
Sm	0	0	0	0	0	0	2	0.04
Re	0	0	0	0	0	0	2	0.04
Gd	0	0	0	0	0	0	1	0.02
Hf	0	0	0	0	0	0	1	0.02

the mineral system approach can be used as a basis for the construction of quantitative physico-chemical models of evaporite formation. Moreover, our approach allows to better understand the conditions of formation of some solid phases (in particular, crystalline hydrates), which are synthesized in “water-salt systems” under laboratory conditions, but have never been found in nature.

4.1.3. Fumaroles of active volcanoes: Tolbachik, Russia, and Vulcano, Italy

Krivovichev & Charykova (2017) demonstrated that the distributions of mineral species among mineral systems in the products of fumarolic activity of Tolbachik, Kamchatka, Russia, and Vulcano Island, Sicily, Italy (Table S3), are very similar (Fig. 3c) and fit the normal law as well. Table 6 and Fig. 4 show the differences in mineralogy between Tolbachik and Vulcano fumaroles.

The values of K_i above the 1:1 line indicate those species-defining elements that have proportionally more species in Tolbachik fumaroles than at Vulcano. These elements include Cu, Se, As, Zn, V, Cl, and F. Accordingly, the values of $1/K_i$ below the 1:1 line refer to the elements with a higher proportion of species in Vulcano fumaroles than in Tolbachik: Bi, Pb, B, Tl, and S. These data are in good agreement with the reported compositions of high-temperature volcanic gases in these two volcanoes (Oppenheimer *et al.*, 2014). Cheynet *et al.* (2000) reported that the most typical components in the Vulcano fumaroles are Pb, Bi, As, Zn, Tl, and Cd. The thermochemical modeling (experimental and numerical) have shown that crystallization of these gases results in the formation of sulfides and sulfosalts mainly of Bi and Pb, whereas As remains mainly in the gas phase (Cheynet *et al.*, 2000). Indeed, while no sulfides or sulfosalts have been reported from Tolbachik fumaroles, they are quite widespread at Vulcano (Garavelli *et al.*, 1997; Campostrini *et al.*, 2010; Weiss, 2010) in addition to recently discovered Bi sulphohalogenides such as demicheleite-(Br), BiSBr (Gramaccioli *et al.*, 2008), demicheleite-(Cl), BiSCl (Demartin *et al.*, 2009a), demicheleite-(I), BiSI (Demartin *et al.*, 2010), vurroite, $Pb_{20}Sn_2Bi_{22}S_{54}Cl_6$ (Garavelli *et al.*, 2005), and mozgovaite, $PbBi_4(S,Se)_7$ (Vurro *et al.*, 1999).

From the geochemical point of view, of interest are ammonium-bearing halogenides such as argesite, $(NH_4)_7Bi_3Cl_{16}$ (Demartin *et al.*, 2012), barberiite, NH_4BF_4 (Garavelli & Vurro, 1994), brontesite, $(NH_4)_3PbCl_5$ (Demartin *et al.*, 2009b), and panichiite, $(NH_4)_2SnCl_6$ (Demartin *et al.*, 2009c).

In contrast, Tolbachik fumaroles show a predominance of Cu, Se, Zn, K, As, and V minerals (Vergasova & Filatov, 2016). More than seventy fumarolic copper minerals have been first described from Tolbachik fumaroles, including recent discoveries of nicksobolevite, $Cu_7(SeO_3)_2O_2Cl_6$ (Vergasova *et al.*, 2014), grigorievite, $Cu_3Fe_2Al_2(VO_4)_6$ (Pekov *et al.*, 2014), shchurovskyite, $K_2CaCu_6O_2(AsO_4)_4$, dmsokolovite, $K_3Cu_5AlO_2(AsO_4)_4$ (Pekov *et al.*, 2015a), and popovite, $Cu_5O_2(AsO_4)_2$ (Pekov *et al.*, 2015b). A similar pattern is observed for selenium: in Vulcano, only native selenium was found (Garavelli *et al.*, 1997) whereas, in Tolbachik fumaroles, more than ten selenites were discovered, including chloromenite, $Cu_9(SeO_3)_4O_2Cl_6$ (Vergasova *et al.*, 1999), and allochalcocellite, $CuCu_5Pb(SeO_3)_2O_2Cl_5$ (Vergasova *et al.*, 2005). No vanadium minerals have been found in Vulcano, whereas several V species have been described in Tolbachik, including koksharovite, $CaMg_2Fe_4(VO_4)_6$, and grigorievite, $Cu_3Fe_2Al_2(VO_4)_6$ (Pekov *et al.*, 2014). The same applies to titanium [katiarsite, $KTiO(AsO_4)$ (Pekov *et al.*, 2016)] and molybdenum [vergasovaite, $Cu_3(MoO_4)(SO_4)O$ (Bykova *et al.*, 1998) and cupromolybdate, $Cu_3O(MoO_4)_2$ (Zelenski *et al.*, 2012)] minerals, which have been found exclusively at Tolbachik. It should be stated that the absence of sulfides in Tolbachik fumaroles and that of oxysalts such as arsenates, selenites and vanadates in Vulcano correlates with the relatively higher value of $f(O_2)$ in Tolbachik gases and higher value of $f(S_2)$ in Vulcano, as well as with the K_i values for O and S in Table 6. The major reason for the absence of As minerals at Vulcano *versus* the abundant and diverse As mineralogy at Tolbachik is the that, at Vulcano, As exists as volatile molecular sulfides and the vent temperature is too high for these to condense, whereas, at Tolbachik, As forms relatively refractory (oxy) arsenates, which condense readily.

4.2. Mineral evolution of the Earth crust

The topic of global mineral evolution has emerged as a new paradigm in mineralogical sciences (Hazen *et al.*, 2008, 2009, 2011, 2012). According to Hazen *et al.* (2008) and Hazen (2013), mineral evolution of Earth can be subdivided into four partially overlapping stages, each of which saw the expansion of mineralogical diversity and/or variation in relative mineral abundances. The starting point of mineral evolution is that of the “ur-minerals”, the twelve earliest mineral phases to appear in the pre-solar nebulae (I). Chondritic meteorites incorporate about 60 primary mineral phases, which constitute the second phase (II). For the Hadean Eon, Hazen (2013) estimated 425 mineral species (III), whereas post-Hadean processes were responsible for the appearance of more than 5000 mineral species known today (IV).

Quantitatively, the evolution of mineral diversity can be assessed by the concept of mineral systems. The data obtained show that the four selected groups of minerals (I–IV) distinctly differ from each other in the number of essential, species-defining elements per species (*i.e.* n , the mineral-system type). Thus, the mean value of the mineral-system type (\bar{n} , the confidence limits are $\pm t_{0.5} s_x$, where $t_{0.5}$ is t -statistic for a probability of 0.95 and s_x is standard error (Wall, 1986)) increases from “ur-minerals” (group I; $n=1-3$; $\bar{n}=2.08 \pm 0.45$) to the chondritic meteorites (group II; $n=1-5$; $\bar{n}=2.68 \pm 0.13$), then to the Hadean minerals (group III; $n=1-8$; $\bar{n}=3.86 \pm 0.07$) and reaches the highest value (and maximum range) for minerals produced by post-Hadean processes (group IV; $n=1-10$; $\bar{n}=4.50 \pm 1.47$) (Fig. 5).

The number of essential, species-defining elements in minerals (in parentheses) also increases from “ur-minerals” (I: 7) to the chondritic meteorites (II: 24), then to the Hadean Eon minerals (III: 35) and is maximum for minerals produced by post-Hadean processes (IV: 70) (Table 7).

The general trend of mineral evolution corresponds to the accumulation of species-defining elements in terms of diversity of the mineral world.

5. Conclusions

The analysis presented above shows that the mineral diversity of different geological objects can be estimated not only by the number of mineral species, but also by the number of the essential species-defining elements that define a mineral system, and their average values. The approach is applicable to different geological objects and allows the analysis of their chemical similarities and differences. It also allows to consider evolution of mineral diversity in terms of the chemical variations of mineral species. In particular, mineral-system analysis of the Hazen’s lists of minerals for different geological epochs provides compelling quantitative evidence for the increasing chemical diversity of mineral species that supports the view that one of the major driving forces of mineral evolution is a chemical differentiation of matter.

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