

CHEMICAL PROPERTIES OF GLACIAL AND GROUND ICE

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Contents

1. Ionic composition in glaciers
 2. Ice and snow chemistry
 3. Ion migration in ice and frozen soils
 4. Methane hydrate
 5. Chemical physics of air clathrate hydrates in ice core
 6. Chemistry of ice in dependence of electrical conductivity
 7. Ionic composition in ground ice
 8. Subpermafrost water geochemistry
 9. Heavy metals in glaciers
 10. Heavy metals in ground ice
 11. Stable oxygen and hydrogen isotope of the ice
 12. Temporal variations of isotopic composition of glacial-river water during summer;
oxygen isotope composition of water sources
 13. Stable isotope composition in glaciers
 14. Stable isotopes in ground ice
 15. Isotope composition of ice-wedge ice
- Acknowledgements
Glossary
Bibliography
Biographical Sketch

Summary

Marine aerosols are the main source of Cl, Mg, Na, K, Mg, SO₄ in ice sheets of Greenland and Antarctic. Marine salts accumulate along the coastline, their concentration decreases sharply away from the coastline. Concentration of elements of continental origin is independent on the distance from coastline. In Greenland dust concentrations in ice-age ice are 3 to 70 times those in Holocene ice. The ice-age dust contains a strong component of calcium carbonate. This neutralized acid aerosols in atmosphere so that in contrast to Holocene ice, nearly all ice-age ice is alkaline. Air clathrate hydrates are observed in deeper ice sheets. Mineralization of ground ice is an important indicator of ice origin; ground ices are classified according to their salinity. Ice with mineralization 0.02– 0.1 g/l is predominant in Holocene ice wedges of Northern Yakutia. The mineralization of Pleistocene ice wedges is 60 – 478 mg/l. Salt concentration ranges in dependence of distance from the sea. Most ice-wedge ice is fresh. Maximum concentration of heavy metals in Greenland ice cores Pb –151 pg/g, Cu – 238 pg/g, Zn – 644 pg/g, Cd – 3.0 pg/g and in Vostok ice core: Pb –38 pg/g, Cu – 33 pg/g, Zn – 101 pg/g,

Cd – 3.0 pg/g. Because the $\delta^{18}\text{O}$ composition of water infiltrating through the snow layer reflects the isotopic composition of the snow, the isotopic composition of modern ice wedges also corresponds with that of winter snow. For example, the mean values of $\delta^{18}\text{O}$ composition of snow from three localities in the permafrost zone of Northern Eurasia are similar to or isotopically lighter than $\delta^{18}\text{O}$ values in modern adjacent ice wedges. The isotope record of ice wedges is averaged oxygen isotope composition of snow precipitation for many years. The oxygen isotope composition of ice–wedge ice varies spatially along the northern coastline of Siberia and temporally during the last 40 ka. This indicates a dependence of the $\delta^{18}\text{O}$ composition of ice wedges upon climatic conditions.

1. Ionic Composition in Glaciers

Chemical composition of glacier ice and snow depends on atmospheric processes connected with precipitation and chemical composition of underlying rocks in case of mountain glaciers. The concentration of SO_4 , Pb and Zn increases due to pollution in mountain glaciers in the recent times. Typical concentration of macro-elements in snow cover of Polar Ice Sheets and Ice Caps show the difference between recent impurities accumulation (Table 1).

Area	Na	Cl	Mg	K	Ca	Fe	Al
Antarctic continent	28	80	5	3.1	2.2	2.9	0.7
Greenland	8	22	7.7	3.8	9.2	10.9	10.4
Devon	29	–	11.3	4.9	8.2	10.1	8.2
Svalbard	–	1100	78	–	139	–	–

Table 1: Typical concentration of macro–elements in snow cover of Polar ice sheets and ice caps (10^{-7} %) (after Murozumi et al., 1969)

Marine aerosols are the main source of Cl, Mg, Na, K, Mg, SO_4 in the ice sheets of Greenland and Antarctica. Marine salts accumulate along the coastline, their concentration decreases sharply away from the coastline. Concentration of elements of continental origin is independent on distance from coastline. In the inland areas of Antarctica and Greenland marine and continental elements are approximately equal.

2. Ice and Snow Chemistry

The impurities in polar ice are either introduced directly into atmosphere (so called primary aerosols such as sea salt and dust emitted by the wind from marine and continental surfaces), or produced within the atmosphere along various oxidation processes, involving numerous trace gases mainly derived from sulfur, nitrogen, halogen, and carbon cycles. While aluminum present in polar precipitation can be traced back to continental primary emission.

Various soluble impurities are expected to be trapped in polar snow layers and their corresponding origins and sources. Primary aerosol of sea salt is mainly Na^+ , Cl^- , and some Mg^{2+} , Ca^{2+} , SO_4^{2-} , and K^+ . These impurities originate from oceanic waves and winds. Primary aerosol of terrestrial salts is: Mg^{2+} , Ca^{2+} , CO_3^{2-} , SO_4^{2-} , aluminosilicates.

These impurities originate from aridity and winds from continents and shelves. Secondary aerosol and gases which occur in polar ice are as follows: H^+ , NH_4^+ , Cl^- , NO_3^- , SO_4^{2-} , CH_2SO_3^- , F^- , HCOO^- and other organic compounds. The sources of secondary aerosol are biogenic and anthropogenic gas emissions, SO_2 , $(\text{CH}_3)_2\text{S}$, H_2S , NO^{2+} , NH^{3+} , hydrocarbons and halocarbons (Legrand and Delmas, 1994).

As to other ions the problem is more complicated. The presence of sulfate in ice can be linked to primary marine (sea salt) or continental (CaSO_4) inputs. It can also be due to the presence of H_2SO_4 produced during the atmospheric oxidation of SO_2 itself being directly introduced in the atmosphere during volcanic eruptions or by human activities or produced by oxidation of various S compounds emitted from the biosphere.

In coastal areas due to large contribution of sea salts input Na^+ , Cl^- , and some Mg^{2+} , Ca^{2+} , K^- and part of SO_4^{2-} , represent a dominant part of the ionic budget (more than 80%) of the ice. Further inland and for present climate, this sea salt input is strongly decreased and other contributions of the ionic composition of snow become dominant (65–80%, depending of sites). The Antarctic Na^+ content corresponds to the sum of two fractions: the first, mainly marine in origin, is soluble in water, the second coming along with dust is not dissolved during the melting step. It is therefore necessary to correct Na data from this dust contribution using Al content of the sample. Among the trace substances in the polar ice cores the strong acids deserve the special interest, because their concentration in time is of particular importance. In the existing deep ice cores acids can be traced at least 35000 years back in time. The yearly average value of acid concentration exhibits remarkably little variability over long time periods: less than 30% in periods with no major volcanic activity. Apparently stratospheric HNO_3 constitutes a major part of the non-volcanic acid composition. Individual volcanic eruptions, which contributed strongly to the chemistry of the snow falling of the ice sheet for a few years after the eruption, can be identified in ice cores. The chemical composition of the acids is mainly HNO_3 and some H_2SO_4 in years with little volcanic activity and H_2SO_4 , HCl , and HF in volcanic fallout. The chemical composition of the volcanic acids is dependent on the eruption, but H_2SO_4 is often an important or dominant component. The contribution of insoluble species can be estimated from aluminum determinations using a composition of mean crust. As summarized in Table 2, under present climatic conditions, soluble species dominate the mass of impurities present in Antarctic snow deposits with a main contribution from the sea salt and increasing contribution of mineral acids (HCl , HNO_3 , H_2SO_4) further inland. In Antarctic ice corresponding to glacial conditions insoluble species become predominant, representing almost half of the total mass. Similarly, terrestrial salts, which are present at insignificant levels under present climatic conditions, represent some 25% of the total ionic budget of glacial Antarctic ice.

The bulk impurity composition (Table 3) of the polar ice sheets do show some regional differences, but are on the average remarkable similar during the Holocene, even for the Greenland Ice Sheet and for Antarctica.

Greenland and Antarctic profiles, covering the last 200 years have provided useful information of the impact of human activities on the chemical composition of the northern and southern hemispheres respectively. For instance, in contrast to the Antarctic sulfate level, the Greenland one reveals the increasing SO_2 fossil fuel burning emissions

of the Northern Hemisphere. As summarized in Table 4 human activities also influence the natural budget of other species including fluoride in relation with growing coal burning, excess–chloride, HCHO and H₂O₂.

Location	Sea salt	HCl	HNO ₃	H ₂ SO ₄	Terrestrial salts	Insoluble (% in mass)
Coastal areas (recent years)	85	N	5	10	N	2
Central area (present climate)	36	22	2	36	N	6
Central area (glacial age)	54	N	2	16	26	62

Table 2: Partitioning of soluble species expressed in % the total ionic budget and concentration of insoluble species to the total mass of impurities found in Antarctic ice (after Legrand and Delmas, 1994)

Cations (µg/kg of ice)		Anions (µg/kg of ice)	
H ⁺	1.2	NO ₃ ⁻	1.0
NH ₄ ⁺	0.3	SO ₄ ²⁻	0.5
Na ⁺	0.4	Cl ⁻	0.5
(Mg ²⁺ , Ca ²⁺ , K ⁺),	0.1	Sum of anions	2.0
Sum of cations	2.0	Dust (µequiv/kg of ice)	50

Table 3: Typical bulk impurity composition of the Greenland ice sheet (after Hammer, 1983)

Species	Pre-industrial level	Recent level (time period)
Nss–SO ₄ ⁻	26 ng g ⁻¹	85 ng g ⁻¹ (1950–1985)
NO ₃ ⁻	68 ng g ⁻¹	120 ng g ⁻¹ (1950–1985)
Nss–Cl ⁻	4 ng g ⁻¹	9 ng g ⁻¹ (1950–1985)
F	0.06 ng g ⁻¹	0.19 ng g ⁻¹ (1971–1989)
Pb	1 pg g ⁻¹	250 pg g ⁻¹ (1960's)
HCHO	2–3 ng g ⁻¹ (1700–1900)	5 ng g ⁻¹ (1980's)
H ₂ O ₂	4 µM (1750–1960)	5 µM (1960–1989)

Table 4: Recent concentration changes of several species over the last century recorded in a Summit (Central Greenland) ice core (after Legrand and Delmas, 1994)

Hinkley (1997) has investigated areas that are protected from the influences of local dust surface to follow chemical composition of dust transported over long distances by the atmosphere and preserved in Northern Hemisphere snow: They are Klutlan Glacier of St. Elias range in Alaska and 20–D site, 40 km west–southwest of Dye–3 in central south Greenland. These points are surrounded by continuous snow and ice cover and isolated from dust sources. The three heavy alkali metals K, Rb, and Cs and the three heavy alkali earth metals Ca, Sr, and Ba have been measured. The proportions of metals measured in the sites (Table 5 and Table 6) of incremental snow samples indicate that the sources of the metals are blown rock and soil dust at both sites. The similarity of the dusts from

Greenland and Alaska, geographically different from each other indicates that these dusts may be an approximation of the background atmospheric load of dusts. The compositions of the dust are similar to that of snow at the Mizuho Plateau site in Antarctica. The body of data suggests that the atmospheric dust load is chemically homogenous on a broad regional or hemispheric scale.

Depth, cm	K	Rb	Cs	Ca	Sr	Ba
Snow of St.Elias Range, Alaska						
Surface	0.850	0.0029	0.00032	4.04	0.026	0.023
Snow, 0–4	1.75	0.0067	0.00058	4.95	0.033	0.058
36–71	760	2.8	0.145	5.320	31.5	29.2
71–101	19.7	0.0800	0.00811	95.5	0.524	0.558
198–231	1.17	0.0045	0.00100	5.2	0.040	0.027
Snow from central south Greenland						
Summer top crust	2.1	0.0054	0.00093	9.3	0.059	0.33
Early winter	2.1	0.0060	0.00075	4.6	0.032	–
Autumn	2.6	0.066	0.00046	2.9	0.023	≤0.09
Spring	9.0	0.031	0.0019	25	0.19	0.24
Later winter	0.96	0.0016	–	1.3	0.014	≤0.04

Table 5: Metal concentrations in snow of St. Elias Range, Alaska and in snow from central south Greenland (parts metal per 10⁹ parts snow, after Hinkley, 1997)

Depth, cm	K/Rb	K/Cs	Ca/Sr	Ca/Ba	K/Ca
Snow of St.Elias Range, Alaska					
Surface	295	2.650	156	290	0.210
Snow, 0–4	260	3.010	147	86	0.35
36–71	280	5.220	169	182	0.142
71–101	245	2.430	182	1171	0.206
Snow from central south Greenland					
Summer top crust	390	2.200	160	30	0.22
Early winter	370	2.900	80	100	0.65
Autumn	400	5.700	130	–	0.90
Spring	250	2.900	150	85	0.35
Later winter	340	3500	140	80	0.60

Table 6: Metal ratios in snow of St.Elias Range, Alaska and in snow from central south Greenland (mass basis, after Hinkley, 1997)

Key points to interpretation of ratios involving the six metals are the following. The K/Ca in sea salt is consistently near unity, whereas the K/Ca ratio in rocks and their derived dusts is most commonly smaller, values of 0.5 or smaller are typical of ferromagnesian, silica-poor rock types. Only in silica-rich rocks K/Ca ratios approach or exceed unity, and such rocks seldom dominate large geographical areas. Cs (and to a smaller degree Rb), and Ba (and to a smaller degree Sr) have very small relative concentration in the sea salt, compared to rocks and their derived dusts. K/Rb ratios are 300 in continental rocks and dusts, vs. 3000 in sea solute; K/Cs ratios, 5000–10000 vs. 10⁶, Ca/Sr ratio, 25–100 vs. 50, Ca/Ba ratio, 25–100 vs. 1500. The large alkaline earth ion is present in the ocean even smaller concentrations than would be allowed by its small solubility in the presence of

sulfate. Both of the large alkali ions Rb and Cs are removed from the seawater by formation and alteration of clay minerals. They are enriched in the clay and mica minerals derived from various type of rocks and the degree of chemical weathering or/and the degree of winnowing during atmospheric uptake and transport can strongly affect the metal composition of dusts from a given rock terrain. Fractionation of earth materials that occurs during atmospheric transport, especially those involving preferential transport of clays and micas, may be indicated by changes in proportions of the alkali and alkaline earth metals. Therefore impurities in snow show a signature of the rocks and their degradation products.

The proportions of metals measured at the sites of incremental snow samples indicate that the sources of the metals are blown rock and soil dusts at both sites. The K/Ca ratios measured are all far smaller than the ocean solute value of unity. The rock types of the sources of the dusts change moderately through depositional year, but these changes in composition of the dusts are neither systematic nor clearly related to the distinct seasonal pattern of changes of amounts of dust in the snow. The differences in the metal ratios in the snowpack appear to reflect alternation between different dust-source terrains that have distinctive rock and soil types or that have been weathered to different degrees. Possibility of the uptake of dust by the atmosphere with differing degrees of particle type fractionation and winnowing is due to such conditions as different wind energies. K/Rb and K/Cs ratios vary sympathetically, as expected indicating different magnitudes of the clay and mica components (clays and micas are indicators of advanced degrees of weathering). These modern dusts are very different from the dusts in older Greenland ice; they have different composition and different sources. Also different rock signatures are observed in other localities. The Sierra Nevada snows have small K/Cs and Ca/Ba ratios reflecting the high-Cs micas, clays and forest soils, and high-Ba feldspars known to be present in the surrounding plutonic terrain.

Insoluble impurities in polar ice sheets consist largely of “dust”, that is, particles in the size range 0.1 to 2.0 μm carried from the continents by wind. Paterson (1991) compares dust concentration in Arctic and Antarctic polar ice sheets in Holocene and Pleistocene. The important conclusions are follows:

1. At Greenland dust concentrations in ice-age ice are 3 to 70 times those in Holocene ice. The mean factor is 12.
2. The concentration is also higher in the ice-age-ice at Byrd station, but only by a factor of 3 (Thompson and Mosley-Thompson, 1981).
3. The average ice concentration at Byrd station (55 particles/ μl) is approximately equal to the Holocene concentration in Greenland (51 particles / μl).
4. Highest concentrations occur in the last part of the ice age (about 30 000 to 15 000 yr. BP at Byrd which is the best-dated record).
5. In Greenland and Canada, the ice-age dust contains a strong component of calcium carbonate. This neutralized acid aerosols in the atmosphere so that in contrast to Holocene ice, nearly all ice-age ice is alkaline. In Antarctica, the major ice-age increases are in aluminum and silicon, rather than calcium and all the ice is acidic (Hammer et al., 1985).
6. Concentrations in Devon and Agassiz ice Caps appear to be less than in Greenland, but this is probably because the lower limit of counting was 1 μm as against 0.6 μm

elsewhere. Calcium and aluminum concentrations at Vostok are comparable with those in Greenland rather than those at Byrd station (Table 7).

Approximate age interval (thousand yr. BP)	Station	Concentration(ng/g)	
		Al	Ca
0–12	Camp Century	10±9	5.1±3.4
	Byrd Station	1.5±0.6	3.4±1.7
	Vostok	3.0±1.5	4±2
12–30	Camp Century	110±52	162±74
	Byrd Station	11±7	8.1±3.0
	Vostok	97±37	100±36
30–60	Camp Century	47±35	50±43
	Byrd Station	5.3±4.4	4.8±3.0
	Vostok	31±20	36±20

Table 7: Concentration of aluminum and calcium in Greenland and Antarctica (after Paterson, 1991)

Soluble impurities may influence the mechanical properties of ice, and for which concentration data are available, are sodium, chloride, sulfate and nitrate ions. Sea salts is the main sources of chloride in both hemispheres in both ice age and at present.

Mean Holocene and Late Pleistocene concentrations of chloride, sulfate, and nitrate are given in Table 8.

Period	Station	Mean concentration		
		Cl ⁻	SO ₄ ⁻²	NO ₃ ⁻
Holocene	Camp Century	34	34	73
	Dye 3	19	22	58
	Barnes Ice Cap	<50	–	–
	Byrd	48	40	41
	Vostok	18	134	16
Ice age	Camp Century	95	168	40
	Dye 3	73	104	45
	Barnes Ice Cap	100	–	–
	Byrd	80	56	46
	Vostok	101	204	56

Table 8: Concentrations of soluble impurities

In contrast to the case of insoluble impurities, concentrations of chloride and nitrate are similar at all stations. For sulfate, the concentration at Vostok is about twice and that at Byrd Station about half, the value of Greenland. The ice-age concentration of nitrate is greater than Holocene concentration at Vostok and less than in Greenland. The two are approximately equal at Byrd Station. The ratio of Late Pleistocene to Holocene concentrations of sulfate is about 5 in Greenland and 1.5 in Antarctica. For chloride, the ratio is 3.3 in Greenland, 11.7 at Byrd Station, and 5.6 at Vostok. Ice-age ice deforms more readily than Holocene because, (1) the concentration of chloride, and possibly

sulfate ions is high relative to Holocene concentrations; (2) the temperature at the time of deposition was about -15°C or lower; (3) the predominant stress is shear parallel to the bed.

The initial chemical composition of glaciers is determined through the chemical composition of solid atmospheric precipitation. Chemical composition of atmospheric precipitation brought to the glaciers under investigation of air masses undergoes substantial changes due to influence of the vast territories over which they pass. It also depends to some extent, on the burning of different kinds of fuel, in the processes of which enormous quantities of SO_2 and NO are emitted into the atmosphere. The content of SO_4 and NO_3 increases considerably as a result of their transformation into acids in precipitation. NH_4 , Cl , Na are added.

The regions of Tien Shan and Eastern Pamirs area differ by hydrochemical types of precipitation. Precipitation rich in Na_2SO_4 , Na , Cl , is peculiar to the glaciers of the Southern Tien Shan, situated closer to the vast desert territories and industrial regions. Thus, the following ratio $\text{SO}_4+\text{Cl} > \text{HCO}_3$ is typical for solid precipitation on the glaciers of this group.

Away from industrial regions the mineralization type of precipitation falling on the glaciers changes in Eastern Pamirs, being total mineralization negligibly small. Investigations of the chemical composition of glacier water and the level of atmospheric precipitation pollution was carried out on the Aksu glacier situated on the northern slope of Turkestan'skiy Range (Southern Tien Shan).

Several stages of element concentration were considered, such as atmospheric precipitation, old snow and ice. Due to melting, washing away, secondary freezing, and also lixiviation, water–rock exchange, the initial chemical composition of precipitation is differed from that in the glacier ice.

The ice is tended to be accumulated microelements. Mean content of Al in the rain is 47 mg/dm^3 , in the snow – 73 mg/dm^3 , in the ice 128 mg/dm^3 , in the melt water – 122 mg/dm^3 . Mean content of Fe in the rain is 24 mg/dm^3 , in the snow – 98 mg/dm^3 , in the ice 206 mg/dm^3 , in the melt water – 138 mg/dm^3 .

Mean content of Mn in the rain is 3.2 mg/dm^3 , in the snow – 3.0 mg/dm^3 , in the ice 18.6 mg/dm^3 , in the melt water – 2.7 mg/dm^3 . Mean content of Zn in the rain is 3.6 mg/dm^3 , in the snow – 14 mg/dm^3 , in the ice 15.4 mg/dm^3 , in the melt water – 4.8 mg/dm^3 . Mean content of Cu in the rain is 1.93 mg/dm^3 , in the snow – 2.57 mg/dm^3 , in the ice 3.08 mg/dm^3 , in the melt water – 1.76 mg/dm^3 .

Mean content of Ni in the rain is 0.16 mg/dm^3 , in the snow – 1.08 mg/dm^3 , in the ice 1.97 mg/dm^3 , in the melt water – 0.08 mg/dm^3 . Mean content of Co in the rain is 0.05 mg/dm^3 , in the snow – 0.17 mg/dm^3 , in the ice 1.11 mg/dm^3 , in the melt water – 0.21 mg/dm^3 . In the melt water flowing from the glacier, there is decrease of heavy metal concentrations in comparison with ice, but Al and Fe remains predominant elements. This can be explained by the presence of aluminum in minerals composing the moraine of Aksu Glacier (Kotlyakov, Gordienko, 1982).

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Biographical Sketch

Prof. Vasil'chuk Yuriy Kirillovich was born in 1954 in Lazo, (Moldova). He graduated Lomonosov's Moscow State University with excellent degree in geocryology and glaciology in 1975. He received a PhD in 1982 and Doctor of Sciences degree in 1991. He is an Academician of the Russian Academy of Natural

Sciences since 2004. He is the head of Glaciology and Geocryology Data Centre of Theoretical Problems Department of Russian Academy of Sciences since 1992, and of Regional Engineering Laboratory of Engineering and Ecological Geology Department of Geology faculty of Lomonosov’s Moscow State University since 1997. He is also a professor of Cryolithology and Glaciology Department of Geography faculty of Lomonosov's Moscow State University since 1996. His principal scientific interests are in area of isotope geochemistry, geochronology, Quaternary Geology, stratigraphy, geocryology, glaciology and geomorphology. He undertook field investigations in nearly all permafrost regions of Eurasia, such as Gydan and Yamal Peninsulas in the North of Western Siberia, Central and Northern Yakutia, Chukotka, Magadan region, Trans-Baikal region and Arctic Islands. Yu.K.Vasil’chuk is the author of over 200 publications, from them there are 7 monographs, such as: “Oxygen-Isotope Composition of Ground Ice” (Application to paleogeocryological reconstructions) 2-volum issued in 1992 and the textbook “Principles of Isotope Geocryology and Glaciology” (coauthored with Academician RAS V.M.Kotlyakov) issued in 2000 et al., about 20 papers he has published in “Transactions of Russian Academy of Sciences” and more than 25 ones in the International Journals, such as *Radiocarbon*, *Permafrost and Periglacial Processes*, *Nuclear Instruments and Methods in Physics Research B*, *Earth and Planetary Science Letters* etc. His recent textbook, "Soil Engineering» (2005, Lomonosov’ Moscow University Press), was co-authored with V.T.Trofimov et al. This textbook characterized the ground ice as a base for constructions. Currently he prepared the new book “Ice wedge: Heterocyclicity, Heterogeneity, Heterochroneity”.