Bulletin of Chinese Academy of Sciences 院刊

Volume 35 | Issue 4 Article 8

April 2020

Cryospheric Chemistry: Fingerprint to Decipher Climate/ Environmental Changes and Anthropogenic Activities

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Abstract

Cryospheric chemistry is a new discipline and one of important branch of cryospheric science. Pollutant emissions from anthropogenic activities have greatly altered the status of chemical components in the cryospheric environment since the Industrial Revolution. When this scenario is coupled with the rapid shrinkage of cryosphere under global warming, both anthropogenic activity and global warming have cooperatively influenced the biogeochemical cycling in the cryosphere and even on a global scale, which results in significant effects on climate and environment. In this paper, firstly, the role of cryospheric chemistry in the cryospheric science is introduced, and the discipline framework and research focuses of cryospheric chemistry are summarized. Secondly, the coupling relationship among cryospheric chemistry and climate/environmental changes, and anthropogenic activities, with emphasis on this relationship in the research field of global change, was exemplified. Lastly, we summarize a review and prospect of hot topics in the current research of cryospheric chemistry. The rapid development of cryospheric chemistry will provide important scientific and technological support for tackling the climate and environmental issues that challenge human survival and development.

Keywords

cryospheric chemistry; climate change; environmental pollution; anthropogenic activities; biogeochemical cycling

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Citation: KANG Shichang, HUANG Jie, MU Cuicui, ZHANG Yulan, XU Jianzhong, DONG Zhiwen, DU Wentao. Cryospheric Chemistry: Fingerprint to Decipher climatic/environmental Changes and Anthropogenic Activities [J]. Bulletin of Chinese Academy of Sciences, 2020 (4): 456–465.

Cryospheric Chemistry: Fingerprint to Decipher climatic/environmental Changes and Anthropogenic Activities

KANG Shichang[1,2,5], HUANG Jie[2,3], MU Cuicui[4], ZHANG Yulan[1,2], XU Jianzhong[1], DONG Zhiwen[1,2], DU Wentao[1]

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Abstract: Cryospheric chemistry is a new discipline and an important branch of cryospheric science. Pollutant emissions from anthropogenic activities have greatly altered the status of chemical components in the cryospheric environment since the Industrial Revolution. When this scenario is coupled with the rapid shrinkage of cryosphere under global warming, both anthropogenic activities and global warming have cooperatively influenced the biogeochemical cycling in the cryosphere and even on a global scale, which results in significant effects on climate and environment. In this paper, firstly, the role of cryospheric chemistry in the cryospheric science is introduced, and the discipline framework and research focuses of cryospheric chemistry are summarized. Secondly, the coupling relationship of cryospheric chemistry with climatic/environmental changes and anthropogenic activities, with emphasis on this relationship in the research field of global change, is exemplified. Lastly, we summarize a review and prospect of hot topics in the current research of cryospheric chemistry. The rapid development of cryospheric chemistry will provide important scientific and technological support for tackling the climate and environmental issues that challenge human survival and development. **DOI:** 10.16418/j.issn.1000-3045.20200302006-en

Keywords: cryospheric chemistry; climate change; environmental pollution; anthropogenic activities; biogeochemical cycling

Since the Industrial Revolution, anthropogenic activities have been increasingly intensified, which has a profound impact on the earth environment. Especially with the rapid increase of population since the mid-20th century, human beings are facing serious problems of resources, environment and social development. In the 1980s, scientists put forward the concept of global change, gradually bringing the atmosphere, hydrosphere, biosphere, lithosphere, cryosphere and anthroposphere into the category of this concept, and highlighting the multi-sphere interaction and environmental change of the earth [1].

In the context of global warming, the cryosphere has been shrinking rapidly in recent decades [2]. The rapid shrinkage of cryosphere has changed biogeochemical cycling in the cryosphere region, resulting in changes in the chemical components and process of the cryosphere, which has impacted regional and global climate/environment. As one of the core bonds linking the interactions among the spheres, cryospheric chemistry can provide a basis for the research on anthropogenic activities, environmental pollution, climate change and biogeochemical cycling. The understanding of the chemical component characteristics, spatial-temporal pattern, migration, transformation and fate of cryospheric elements, and their response and feedback to climatic/environmental changes can provide scientific support for the sustainable development of human economy and society. Therefore, under the general framework of cryospheric science, cryospheric chemistry, which involves physics, chemistry, biology, atmospheric science, ecology, climatology and environmental science, emerges at the right moment.

1 Cryospheric science and cryospheric chemistry

Cryosphere refers to the negative temperature sphere with

Received: 2020-4-2
Supported by: National Natural Science Foundation of China (41721091); The Second Tibetan Plateau Scientific Expedition and Research Program (2019QZKK0605); Strategic Priority Research Program of Chinese Academy of Sciences (XDA20040500)

a certain thickness and continuous distribution on the earth surface [3]. It is composed of glacier (including ice sheet), frozen soil (including permafrost and seasonally frozen soil), snow cover, river ice and lake ice of terrestrial cryosphere, ice shelf, iceberg, sea ice and subsea permafrost of marine cryosphere, as well as frozen water body (such as ice crystal, ice nucleus and hail) of atmospheric cryosphere. Cryospheric science is a new interdiscipline that studies the process and mechanism of the formation and changes of cryospheric elements, the interaction between cryosphere and other spheres of the climate system, and the influence and adaptation of cryosphere changes [3,4]. Cryospheric chemistry is a discipline that studies the spatial-temporal pattern, sources, migration, transformation, fate of chemical components of cryospheric elements and their impacts on climate and environment [5]. This discipline involves regional characteristics, source and sink characteristics and biogeochemical cycling of chemical components in the cryosphere, as well as chemical processes at the interface between cryosphere and other spheres.

Cryosphere is the most sensitive sphere in the climate system and an amplifier of global change. With the main distribution in polar and alpine regions, cryosphere is rarely affected by local anthropogenic activities. Therefore, cryospheric chemistry, as fingerprint, can reflect the climatic/environmental changes on the regional and even global scale, which may reveal the evolution process of different climatic and environmental factors. The unique mass-energy exchange and rapid phase transition of cryosphere are extremely sensitive to climate warming and anthropogenic activities, which makes cryosphere one of the important spheres participating in the global biogeochemical cycling. Global warming and anthropogenic activities have cooperatively influenced the biogeochemical cycling and chemical components in the cryosphere, which have resulted in marked effects on the climate and environment.

Although being a new research field in cryosphere science, cryospheric chemistry will support the development of cryospheric science with the deepening discipline construction, development and research. In addition, cryospheric chemistry can uncover the history and mechanism of global climatic/environmental changes in the past by revealing the biogeochemical cycling patterns of multiple chemical indicators, and predict future changes to serve human development, which has important scientific significance and application prospects.

2 Discipline framework of cryospheric chemistry

As shown in Figure 1, the research of cryospheric chemistry involves terrestrial cryosphere, marine cryosphere and atmospheric cryosphere. The main research objects include trace gases, inorganic and organic chemical components, stable and radioactive isotopes, and microorganisms in the cryosphere. The research time range covers different dimensions such as hours, days, seasons and years. The research content of cryospheric chemistry mainly includes the following three aspects.

(1) Basic physical, chemical and biological processes related to cryospheric chemistry. The basic physical processes mainly include dry and wet deposition and scavenging processes of atmospheric components, ionic pulse of snowmelt runoff, leaching of permafrost and desalination of sea ice. The chemical processes mainly include isotopic fractionation, photochemical effect, and redox reaction, The biological processes include methylation, microbial process of methane, nitrification and denitrification.

(2) Spatial-temporal pattern and sources of chemical components in the cryosphere. The relevant studies focus on the spatial-temporal pattern, transmission and sources of inorganic components (chemical ions, insoluble particles, elements, black carbon, etc.) and organic components (organic matter, persistent organic pollutants, etc.) in the atmospheric cryosphere, terrestrial cryosphere and marine cryosphere, exploration of the natural and man-made sources of chemical components on the basis of fingerprint characteristics of isotopes, and the unveiling of the history of pollutant emissions from anthropogenic activities according to snow and ice records.

(3) Impact of biogeochemical cycling in the cryosphere. The relevant studies focus on the key biogeochemical processes of different elements in the cryosphere and the climate and environmental effects of biogeochemical cycling on the cryosphere in the context of climate warming and intensified anthropogenic activities, which provide scientific support for the response to climatic and environmental changes in the future.

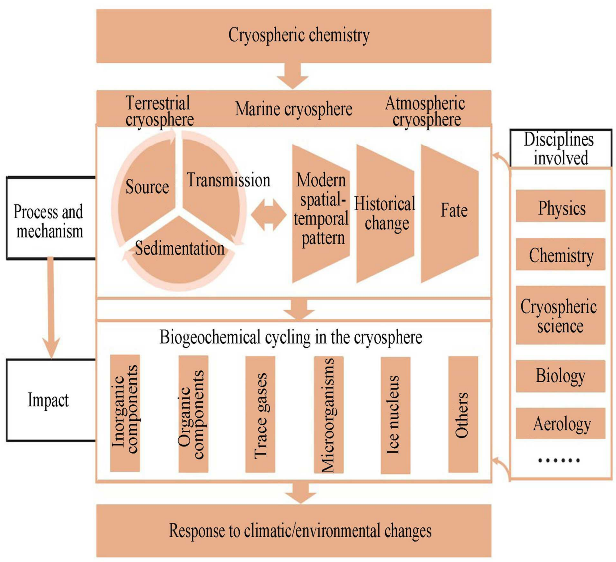

Figure 1 The disciplinary framework of cryospheric chemistry

Modified according to Reference [5].

3 Cryospheric chemistry, climatic/ environmental changes and anthropogenic activities

The chemical components in the cryosphere are associated with anthropogenic activities and environmental changes. In the case of little interference from anthropogenic activities, the chemical components in the cryosphere are mainly influenced by environmental processes. In the case of intense anthropogenic activities, however, the chemical components in the cryosphere are mainly affected by pollutant emissions from anthropogenic activities. The spatial-temporal pattern, migration, transformation and fate of chemical components in the cryosphere are directly related to climate and environment, which have reshaped the biogeochemical cycling of chemical components in the cryosphere. Cryospheric chemistry, involving the climatic/environmental changes, plays an important supporting role. In this paper, the coupling relationship of cryospheric chemistry with climatic/ environmental changes and anthropogenic activities is exemplified.

3.1 Cryospheric chemistry and climatic/ environmental changes

Environmental media of the cryosphere, such as glaciers, are important carriers of global climate change information. The chemical records of the environmental media, as a unique alternative of climatic change data, have been widely used in the study of global climate change. With these data, some important discoveries in climate change have been achieved. Scientists have found in the 1950s that there is a significantly positive correlation between stable isotopes in precipitation and air temperature in the high-latitude cryosphere [6], which marks a milestone of climate change research. The results of a number of reconstruction studies on the basis of hydrogen and oxygen stable isotopes in ice core are consistent with those of the measured data in recent hundred years, indicating that the global climate is warming rapidly [7–10].

The mass-independent fractionation (MIF) of oxygen and sulfur isotopes in cryospheric chemistry has received great attention in the study of climatic/environmental changes. Isotopes have shown strong tracing ability in the study of oxidation ability and oxidation process in the atmosphere (including paleoatmosphere), genesis of mineral deposit, impact of volcanic activity on climate and sulfur cycle [11,12]. In recent years, the law of MIF has been applied to the research of cryosphere. The change of atmospheric oxidation ability in the past 100 000 years was reconstructed with nitrate $\delta^{17}O$ in the deep ice core of Greenland Ice Sheet Project 2 (GISP2), which revealed the key information of atmosphere (paleoatmosphere) oxidation ability with climate change [11]. With the analysis on the sulfate $\delta^{33}S$ and $\delta^{36}S$ in the sediments of Gokyo Lake in the south slope of Mount Everest, the sulfur cycle history in this region in the past 200 years was reconstructed, which enriched the understanding on the environmental change pattern in high-altitude areas [12].

In addition, dust and other chemical components in ice cores record not only the natural processes such as drought but also the changes of atmospheric circulation mode and intensity sensitively, which well reflect the evolution of climate system. The chemical records of dust in the ice core of global cryosphere reflect the historical differences of dust load and circulation of atmosphere. For example, the dust records of ice cores show that the dust activity has been weakening due to the change of circulation intensity in the central and southern Tibetan Plateau since the 20th century [14].

3.2 Cryospheric chemistry and anthropogenic activities

The chemical component records of cryosphere, like a historical book, reflect the chemical status of snow and ice in different periods, thus providing the basis for interpreting the climatic/environmental changes in the past. Anthropogenic activities have not only accelerated the social development but also caused severe damage to the environment since the Industrial Revolution, which gradually becomes a major factor affecting the redistribution of chemical elements in the environment. Chemical pollutants are ubiquitous with low content in the nature, but the increased pollutant emissions have caused environmental pollution worldwide. The cryosphere is mainly in remote regions, with a small population and long distance from the source of industrial and agricultural discharge, receives rare direct disturbance of anthropogenic activities. Therefore, the process of pollutant emission from anthropogenic activities in the polar regions and alpine glaciers can be taken as a substitute index to evaluate the impact of anthropogenic activities on the atmospheric environment.

Glaciers (ice sheets) and alpine lakes are important components of the cryosphere. Their chemical components, mainly from dry and wet deposition, are the natural archives of atmospheric composition. Compared with other data, the research data of glaciers and alpine lakes are characterized by continuous recording, high resolution, strong fidelity and slight change after deposition, which can accurately depict the history of pollutant discharge by human beings.

The chemical component records of ice cores and alpine lakes in the cryosphere are significant fingerprint for deciphering historical changes of pollutant discharge by human beings. For example, there are many historical records of Hg in ice cores and lake cores around the world [15–18]. As shown in Figure 2, the atmospheric mercury deposition in the global cryosphere has been rising rapidly since the Industrial Revolution, which is closely associated with the mass production of mercury and the rapid release of mercury pollutants by anthropogenic activities [19]. With the enhancement of human environmental protection awareness, developed countries in

Europe and North America have taken strong measures to reduce emissions in recent decades, and the mercury pollutants released from anthropogenic activities in these areas tend to decrease [15,16] (Figure 2). However, the mercury concentration in the Altai Mountains and the winter ice core of Gêladaindong Peak in Asia has been increasing significantly in recent decades [17,18], which is closely related to the rapid economic and industrial development in Asia. The research demonstrated that Asia has become the major source of mercury emissions from anthropogenic activities, which account for more than half of the global total emissions [20].

The ice cores and lake cores of the Tibetan Plateau record the rapid increase of atmospheric mercury deposition flux since the Industrial Revolution, especially after World War II. The records correspond to the recent increase of mercury emissions from anthropogenic activities in South Asia, which reveals that the atmospheric mercury background and deposition flux on the Tibetan Plateau is mainly affected by pollutant emissions from anthropogenic activities in South Asia [21]. The above facts mean that the pollutants have impacted the global environment via air transmission. Therefore, the cryosphere has become an ideal place to evaluate the pollution degree of anthropogenic activities and historical changes. On the basis of the facts recorded by the cryosphere media, governments can be warned to strictly control and reduce the emissions of air pollutants.

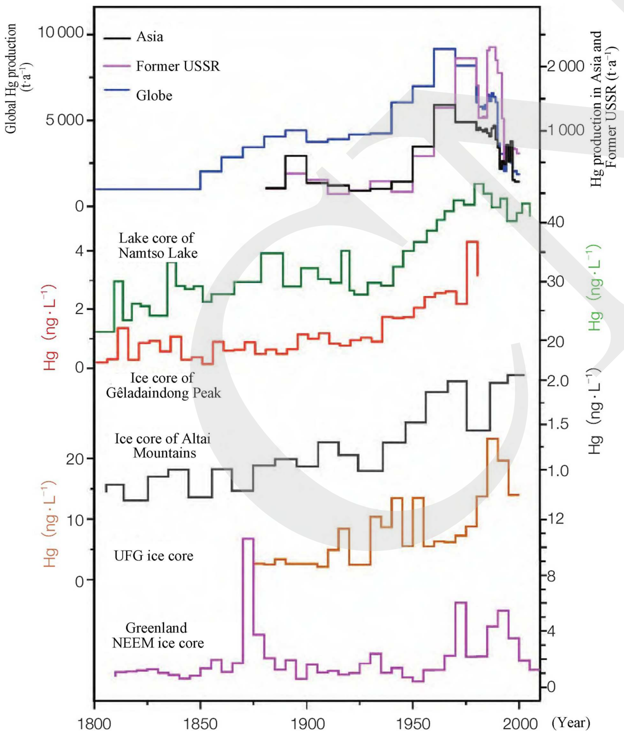

Figure 2 Historical records of mercury in ice cores and lake cores and variation of mercury production in different cryosphere regions

Data sources: Lake core of Namtso Lake and ice core of Gêladaindong Peak [8]; ice core of Altai Mountain [9]; Upper Fremont Glacier (UFG) ice core [6]; Greenland NEEM ice core [7].

4 Hot topics in cryospheric chemistry

4.1 Permafrost degradation and carbon cycle

The organic carbon stock in permafrost regions of the Northern Hemisphere is 1 400–1 850 Pg C, accounting for about 50% of the global soil carbon stock, which is over twice of the atmospheric carbon stock [22]. The rapid global warming is aggravating the degradation of permafrost, leading to the melting and decomposition of organic carbon originally frozen and the massive release of greenhouse gases (such as CO_2 and CH_4) into the atmosphere. The increased greenhouse gases in the atmosphere further accelerate global warming. Therefore, the permafrost degradation has a strong positive feedback effect on climate change [23] (Figure 3). However, the carbon source and carbon sink effects of permafrost show great differences in different regions, which may lead to large deviation of the prediction. Specifically, the assessment of soil carbon emission, especially deep soil carbon emission, caused by climate warming is quite different [23]. After the degradation of permafrost in the Arctic region, it is possible to form thermokarst lake and increase CH_4 emissions. However, the mechanism of climate effect caused by such surface change is still unclear [24]. Although global warming increases the release of soil carbon in local areas, it promotes the growth of plants which absorb more carbon [25]. Therefore, the lack of understanding on these processes leads to great uncertainty in the assessment of future carbon cycle and climate change.

The biogeochemical cycling of carbon in permafrost is a significant part to develop and improve the earth system model. The available studies on the earth system model primarily focus on the slow warming of permafrost. In the permafrost areas that are rich in ice, the degradation of permafrost would cause rapid collapse of the surface and form thermokarst [13]. However, since this process is complicated and not fully studied, it is not included in the coupling model, which leads to the uncertainty of carbon cycle assessment. Climate warming accelerates the collapse of permafrost, which makes the ecosystem change from net carbon sink to net carbon source, and the plant growth can partially offset carbon source [26]. In addition, the dissolved organic carbon transports across regions with runoff and changes its bioavailability. The response of the release flux to permafrost degradation is also one of the uncertain factors for evaluation of the feedback potential of carbon in the permafrost [27].

In addition to the terrestrial permafrost, the subsea permafrost also influences climate change. Because the subsea permafrost distribution, CH_4 hydrate storage and permeation process [28], and biogeochemical mechanism of organic carbon stock and decomposition in sediments are unclear, it is difficult to systematically evaluate the climate effect of the carbon sink of subsea permafrost. To accurately evaluate the feedback of permafrost degradation to climate change in the context of climate warming, we should unveil the chemical

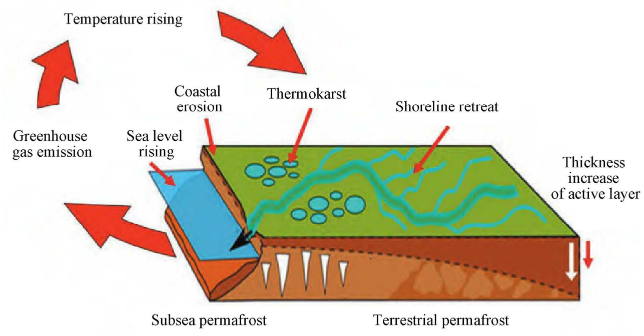

Figure 3 Positive feedback effect of permafrost degradation on climate change

mechanism of carbon decomposition and stabilization in the process of terrestrial and subsea permafrost degradation, and the balance of greenhouse gas release rate and mode with plant carbon sequestration in the process of slow warming and rapid collapse of permafrost.

4.2 Environmental effect of cryosphere shrinkage

Pollutants from anthropogenic activities are transmitted to distant cryosphere through atmospheric circulation and can be sealed in glaciers and permafrost [21,29–31]. Therefore, the cryosphere is also a "repository" for air pollutants [31–32]. With the aggravating global warming and rapid cryosphere melting, the toxic pollutants stored in the cryosphere will be released in a near future [32–34]. That is, the secondary release of pollutants. The hazards of glacier melting and permafrost degradation to the environment have been well recognized [35]. In the past four decades, the glaciers in western China have released about 2 500 kg mercury pollutants through glacier meltwater, which enter the downstream ecosystem [32]. Moreover, the cryoconite accumulation areas on the glacier surface in the context of warming may become a new place for mercury methylation [29]. There is also much mercury stored in the Arctic permafrost region [31]. The permafrost degradation caused by climate warming increases the release and migration of mercury in permafrost. In the Arctic region, about 20 000 kg mercury pollutants enter rivers and into the Arctic Ocean each year [34]. The mercury pollutants and persistent organic pollutants released from glaciers and permafrost will enter the downstream ecosystem via surface runoff, which may pose potential impact on the ecological environment of the downstream area of the river which is supplied by the melting water of the cryosphere. Therefore, the caused environmental pollution risk should not be underestimated.

Glaciers and permafrost not only contain the above-mentioned toxic pollutants but also are likely to contain ancient microorganisms. These ancient lives were frozen in the cryosphere before climate warming and did not migrate between cryosphere and other spheres. However, once the glacier melts and the permafrost degenerates, these microorganisms are very likely to be released and enter the human living environment. Such extreme consequences may be the next disaster that human beings cannot bear. It has been found that there are ancient viruses in the samples of deep ice cores from the Tibetan Plateau, among which 28 are novel viruses [36]. Similarly, microorganisms are found in unfrozen water in permafrost, which may contain ancient viruses. A virus, which has been preserved for over 30 000 years [37], was extracted from the permafrost. Moreover, this virus could still be activated rapidly after being reheated in laboratory. In other words, these unknown pathogens frozen in the permafrost may revive due to climate warming. With the rapid shrinkage of cryosphere, the effects of reactivation of unknown viruses dormant in the cryosphere on the environment and health are worthy of further study.

4.3 Feedback effect of cryospheric chemistry on climate

The feedback effect of light-absorbing components (black carbon, organic carbon, dust, etc.), as an important component of cryospheric chemistry, on climate has received great attention. The light-absorbing components in snow and ice can directly reduce the albedo of snow and ice. That is, they can absorb more solar radiation after the surface of snow and ice darkens, which strengthens the warming and melting of snow and ice [38,39].

In the alpine cryosphere of Asia, the albedo feedback of light absorbing components deposited in snow and ice can increase the surface temperature by 0.1°C–1.5 °C [40]. It is the important climate forcing factor only second to CO_2 with short life cycle [41]. The contribution ratio of black carbon and dust to the reduction of albedo in the glacier melting period on the Tibetan Plateau can reach 20%–50%, and the radiative forcing can reach 100 $W\cdot m^{-2}$. Due to the climate feedback of black carbon and dust in snow and ice, the glacier and snow melting on the Tibetan Plateau increased by about 20% and 5–25 mm·w.e., respectively, and the duration of snow cover was shortened by 3–4 days [21]. In recent years, the temperature increase in the Arctic region has reached more than twice of the global average. Because of the high albedo and strong feedback on the ice and surface, the climate in the Arctic is significantly affected by the light absorbing components in the snow and ice, while the light absorbing components are an important factor for climate warming in the Arctic in addition to greenhouse gases [42]. The radiative forcing caused by black carbon in Arctic snow cover can reach 0.17 $W\cdot m^{-2}$ [43], and the temperature rise range can reach 0.24 °C [44]. The light absorbing components can result in an annual sea ice reduction of about 1% from July to September in the region north of 66.5°N [44] and an increase of about 8 $GT\cdot a^{-1}$ (about 6.8% of the total ablation volume) in Greenland ice sheet ablation [45]. The continuous melting of the cryosphere, especially the glacier, results in the enrichment of light absorbing components on the surface of the glacier, which further accelerates the melting of glaciers. Therefore, the climate effect of the light absorbing components in snow and ice will be increasingly obvious. It is an urgent scientific problem to evaluate such effect and hydrological and water resource effect on a global scale.

5 Conclusions

With the improvement of laboratory analysis and field automatic monitoring technology, the research on cryospheric chemistry has developed rapidly. The traditional research focus of cryospheric chemistry is mainly the migration, transformation and fate of chemical components in the cryosphere, which, with the interdisciplinary integration, has gradually been integrated with climatic and environmental effects. Undoubtedly, glacier (ice sheet), permafrost, river ice, lake ice and sea ice are unique media recording climatic/environmental changes and anthropogenic activities, which has irreplaceable advantages. With the improvement of technology, more chemical components in the cryospheric environment will be determined. Therefore, the research content of cryospheric chemistry will become increasingly rich.

Cryospheric chemistry has gradually become an important branch of cryospheric science. The construction, connotation and extension of cryospheric chemistry need to be improved and developed in the research and practice. It is urgent to obtain the first-hand data of chemical components and their migration and transformation in the cryosphere by establishing a global three-dimensional observation network. These data, together with laboratory test and simulation, can aid in the elaboration on the migration and transformation mechanism of chemical components in the environmental media of the cryosphere. Further, the changes of chemical components in the cryosphere, the natural and anthropogenic processes, mechanism and impact of biogeochemical cycling, as well as the climate and environmental effect in the context of global warming and anthropogenic activities can be revealed, which provides scientific support for the response to global climate change and regional sustainable development.

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