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PAPER

Ten crucial unknowns in atmospheric chemistry in the cold

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The Southern Ocean, wintertime cities, the upper troposphere, the Arctic and Antarctica, and alpine mountains are places where atmospheric chemistry impacts human health, air quality, climate, or geochemical cycles and that are characterized by low temperatures where ice or snow can be present. The atmospheric impact is evident from the role of polar biogenic sulphur emissions on aerosol formation, multiphase nitrogen and sulphur chemistry on wintertime haze, and industrial emissions in snow-covered areas on the ozone budget. The Cryosphere and ATmospheric CHemistry community (CATCH) addresses the environmental processes within these coupled cryosphere–atmosphere systems, and here we present open research questions specific to the cold environments, focusing on the unique interplay of chemistry and physics. These research needs call for interdisciplinary approaches to address atmospheric science in a warming climate with changing human impact in Earth's cold regions.

Widespread regions of Earth's atmosphere are characterized by low temperatures, including the Arctic, Antarctic, and Southern Oceans; the upper troposphere; the Himalayan plateau; high alpine mountains; and most of the Northern Hemisphere in winter. Cold conditions introduce ice in clouds, sea ice, snow, and frozen ground as part of the hydrosphere. Importantly, cold temperatures in the cryosphere introduce unique interfaces and chemical environments, impacting reaction mechanisms, atmospheric composition, geochemical fluxes, fate of toxins, and physicochemical properties different to those in moderate atmospheres.[1,2]

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Multidisciplinary research approaches combining chemistry, biology, and physics within the coupled cryosphere–atmosphere system are crucial to progressing our understanding.[3–5] The cryosphere includes snow on the ground, lake and river ice, frozen ground and permafrost, ice sheets, ice caps, glaciers, ice shelves and icebergs, and sea ice. In this work, we pragmatically expand the definition of the cryosphere to include ice clouds as well. The iconic research that moved environmental cryospheric science to the political and economic agenda, the scientific understanding of the stratospheric ozone depletion, is a pivotal example of where chemistry, atmospheric science, meteorology, and satellite technology worked together.[6–8] Other examples include the recognition of surface snow as a temporal reservoir of toxins and multiphase chemistry pioneered by Honrath's observation of nitrogen oxide production from illuminated snowpack[9] bringing physicists, chemists, and atmospheric scientists together.[10–13] Recently, severe and persistent wintertime air pollution events in Asia and urban areas globally have received great publicity due to their adverse impact on atmospheric visibility, air quality, and human health[14,15] and included health and social science.[16] Last but not least, research in the Southern Ocean illustrates the need to include oceanographers and biologists: microorganisms in the sea ice, polar oceans, glaciers, and permafrost have been shown to impact aerosol composition, and particularly aerosols that serve as seeds for cloud droplet and ice crystal formation – cloud condensation nuclei (CCN) and ice nucleating particles (INPs), respectively.[17–20]

The Biogeochemical Exchange Processes at Sea-Ice Interfaces (BEPSII),† Coupling of ocean–ice–atmosphere processes: from sea-ice biogeochemistry to aerosols and clouds (SCOR working group #163, CIce2Clouds‡), and the Cryosphere and ATmospheric CHemistry (CATCH)§ working groups of the Scientific Committee of Ocean Research (SCOR), the Surface Ocean Lower Atmosphere Study (SOLAS), the International Arctic Science Committee (IASC), and the International Global Atmospheric Chemistry (IGAC) have formed to foster such multidisciplinary research and develop research priorities for their specific science targets. Here, we present 10 key questions and research priorities from these communities.

How does the cold impact emissions?

Wintertime pollution in cities and densely populated areas has become a recent research focus.[16,21] Both natural and anthropogenic emissions differ significantly in winter compared to warmer atmospheric conditions. For instance, homes are heated – usually through the combustion of wood, oil, coal, or natural gas – which is a significant source of local emissions. It remains a challenge to derive a source apportionment and fully understand meteorology and transport's role on observed pollution levels.[16] Further, emissions by soil and plants show distinct changes with temperature that are still a challenge to account for.[22–24]

In the Southern Ocean, considerable efforts have focused on wind-driven sea spray aerosol and trace gas emissions.[20] For example, an ice-covered surface ocean

† https://sites.google.com/site/bepsiiwg140/home.

‡ https://www.cice2clouds.org.

§ https://www.catchscience.org.

impacts the reactive nitrogen budget as well as the oxidative capacity of the marine boundary layer.[25] Although there are generally uncertainties in the emission of well-studied compounds like dimethylsulfide (DMS), we might also be critically underestimating the importance of other reactive trace gases and biological particles. A key example is the emission of methanethiol, a previously neglected co-emission with DMS, which is oxidised rapidly in the atmosphere and may be a particularly important, but poorly constrained, biogenic sulfur emission from high latitude oceans.[26]

How do behavioural change and technical innovation impact the cryosphere?

The accumulation of pollutants in the global cryosphere from where they may enter the aquatic ecosystem and the food web is now well documented.[27,28] Yet, continuing studies on the biogeochemical cycle of long-known pollutants reveal insights on the specific cryospheric fate, for example, how the biodegradation of PAH is impacted by snow melting.[29] An additional concern arises from the appearance of new pollutants driven by innovation in consumer products, changes in traffic routes, or shifts in emission and atmospheric transport regimes. For example, organophosphate esters used in flame retardants have recently been identified in the Arctic, some of which possess toxic properties for the aquatic environment.[30]

What do we know about historic atmospheres?

Environmental archives such as glaciers have added to the understanding of atmospheric chemistry by providing information on the composition of trace gases and aerosol starting in pre-industrial times.[31] Recently, the first nontarget screening record of secondary organic aerosol species preserved in a Siberian ice core ranging from 1800 to 1980 was presented, showing changes in aerosol composition since the 1950's with the appearance of nitrogen-containing organic molecules and a general higher oxidation state suggesting an increased oxidative capacity of the atmosphere.[32]

A key difficulty in interpreting ice core records is accounting for potential changes by chemical reactions in the snow before the species is preserved in the ice. Bromine is an example of such a reactive species in snow.[33] Records of bromide have been interpreted to reflect changes in sea ice extent[34,35] and anthropogenic bromine emissions.[36] However, in low snow accumulation sites, where the transformation of snow to ice takes a long time, bromide chemistry in snow needs to be better understood to interpret ice cores.[37]

How is chemistry in snow impacting the air?

A quarter century after the pioneering discoveries of nitrogen oxide release to the atmosphere from illuminated snowpack, we still lack a predictive understanding of the underlying snow chemistry.[9,38] This lack of knowledge is critical as chemistry in snow impacts regional atmospheric composition, the chemical fate of trace gases and aerosol components, geochemical cycles and distribution of

toxins.[11,27,33,39,40] Recently, snow in polluted cities has gained attention as a reactive medium impacting air composition and a transfer medium impacting meltwater toxicity.[41,42]

A central unknown is where reactions occur in snow and ice. This is important as the individual reaction compartments in the snow show different local physical and chemical properties.[1] For example, the photolysis of nitrate has been found to differ at the air–ice interface and within liquid pockets.[43] While thermodynamics predicts the presence of liquid over a wide range of environmental temperatures,[44,45] the size, location, and chemistry of this liquid is currently unclear.[46–48]

What will happen in a warming climate?

The warming climate imposes drastic changes on cryospheric environments. Seasonal snowmelt occurs earlier, snow cover and sea ice extent, and thickness are reduced. Its impact on groundwater cycles in mountain areas has recently been highlighted.[49] Loss of snow has profound impacts on atmospheric chemistry, such as recently demonstrated for halogens, where sea salt aerosol increase with the reduction of sea-ice[50] and where reactive bromine species in the air ceased once the snow melted.[51] In contrast, Wang *et al.* reported efficient nitrous acid (HONO) production in snow melt, indicating that meltwater might be a potential reaction medium as well.[52] Meltwater may corrupt the chronology in ice core archives[53,54] and flush pollutants to the environment.[28] Further studies are needed on how the formation, evolution, and melt of sea ice and snow cover in the polar oceans and on land impact the emission and deposition of climatically and biogeochemically active materials.

How is the cold relevant to multiphase chemistry?

Multiphase processes in clouds and aerosol have long been identified as key contributors to air quality and the chemical cycling between the atmosphere and the hydrosphere.[10,24,55] The current key challenge lies in extrapolating the physical chemistry, such as rate constants and solubility data, to the wintertime dry and cold atmospheric conditions.[14] This question has been a priority in the last decade, driven by severe pollution events in polluted wintertime urban areas such as the Asian Haze or Arctic cities.[15,16] New multiphase chemistry routes and mechanisms under these cold and hazy conditions have been identified with impact on aerosol formation and health, such as for the oxidation of SO_2.[56–58] Further work is urgent to improve our knowledge and concepts of fundamental parameters such as trace gas solubility,[59,60] acid–base chemistry[61,62] or phase separation.[63]

Atmospheric relevant reactions have been found to be accelerated in small droplets,[64–71] including observing a spontaneous H_2O_2 formation.[72,73] Initially, interfacial electric fields were proposed as key drivers.[2] However, recent literature reveals more complexity. Key, highly debated uncertainties are the magnitude of these fields, their impact on chemistry, experimental artifacts (*e.g.*, gas phase ion chemistry), the role of interfacial solvation, and other factors potentially accelerating multiphase chemistry.[74–81] Further studies that carefully control for confounding experimental factors are urgently needed.[82] Interestingly, charges on

static ice films have not been reported but are known during ice growth and friction in blowing snow, their impact on atmospheric chemistry is unclear.[83,84]

Why are hydrogen bonds important?

The properties of ice, water, and aqueous solutions are fundamentally linked to the hydrogen bonding network.[60] The structure and physical properties of the hydrogen bonding network at the surface of ice are of ongoing interest.[1,85] Relevance to atmospheric research comes from the far-ranging environmental impacts of the air–ice interface, such as in thunder formation[86] and the acceleration of heterogeneous reactions and product releases to the atmosphere.[87]

While it is clear that these unique properties are rooted in the hydrogen bonding network being more flexible at the air–ice interface compared to the bulk crystal,[1,88] an open question is how to link the physics at the interface to chemical reactivity precisely and to develop a predictive understanding,[1] interfering chemical properties from physical observations is controversial.[1] A way forward might be the study of chemical reactions, such as acid–base chemistry, directly on ice surfaces.[89–93] This line of research links to the broader chemistry community beyond atmospheric chemistry, where the role of water is discussed in heterogeneous catalysis.[2,94]

Are there fluxes through ice & snow?

Trace gas exchanges between the atmosphere and the ocean or ground in winter often proceed *via* sea ice or snow. We are far from a physically sound understanding and description of the permeability of these matrixes for gases.[12] In particular, the role of wind and wind-induced pressure fields is not yet at a level that would allow predictions.[95] Addressing the transport through snow is particularly challenging because snow is known to evolve with time (*e.g.* snow metamorphism) and these dynamics impact the fate of trace gases.[96,97] A current example illustrating the lack of knowledge is polar iodine chemistry: The potential of ozone destruction from iodine is an active research area and one major question is whether iodine is transported *via* the sea-ice to the atmosphere or originates from the overlying snow.[98,99]

How do particles grow in wintry conditions?

Our community is just at the beginning of understanding how clouds form in cold regions of the atmosphere, such as the Southern Ocean and the Arctic Ocean and wintertime cities or the upper troposphere. Knowledge of the precursor tracegases and vapours that grow the particles is continuously increasing. Controlled studies at the CERN CLOUD chamber have significantly contributed to the current understanding, that is still incomplete under dry and cold conditions. One example is the key finding that iodic acid drives particle formation in Antarctic conditions.[100] Such observations highlight the important role of gas-phase chemistry in controlling the aerosol population. Further challenges are posed by the interplay of gas-phase chemistry with particle nucleation and growth at cold temperatures, as for example shown by recent studies on ozone-initiated oxidation of alpha-pinene on the formation of organic aerosol. Small particles in

the Aitken-mode down to a few tens of nanometers have recently been observed directly within cloud residuals, suggesting that they are an important source of cloud condensation nuclei (CCN).[101]

How do aerosols impact a changing climate?

The physicochemical properties of aerosols in cold environments determine their fate as directly interacting with radiation and the surface energy budget and as cloud condensation nuclei (CCN) and ice nucleating particles (INPs) that influence cryospheric cloud properties. For example, sea spray aerosol from polar oceans scatters solar radiation[102] while combustion aerosols absorb energy both in the atmosphere and when deposited on the high-albedo frozen surface.[103,104] After deposition, multiphase and photochemical reactions on the snow surface result in changes in snow chemical composition and the release of reactive halogen gases.[39,98,105] CCN in cryospheric regions are typically tied to aerosol number concentration and originate from both (1) anthropogenic pollutants from local combustion and long-range transported pollution[106] and (2) natural aerosols produced from biogenic emissions.[107,108] INP sources and abundances, on the other hand, are less known than CCN. However, key studies indicate that INPs from primary biological materials and/or dust from high-latitude and long-range transported sources can impact INP populations in the Arctic,[109,110] Antarctic,[111] Southern,[112] and high mountain regions.[113]

While there have been substantial research efforts focused on characterizing CCN and INP sources and impacts on clouds in cryospheric regions, we still have a limited understanding of these special types of aerosols compared to lower latitudes. New sources are emerging and/or changing, such as melting sea and glacial ice and thawing permafrost, yet these sources are only in their infancy of being studied. Additionally, ground-based measurements, which are more common in the past, may not be representative of CCN and INPs at cloud level, thus, more routine vertically resolved observations are needed *via* platforms such as uncrewed aerial vehicles, tethered balloon systems, and traditional crewed aircraft. Further complications include the inadequacies of models of all scales to reproduce realistic cloud microphysics (*e.g.* ref. 114), thus, more observations and model improvements are needed to better predict the impacts of aerosols on cloud formation, lifetime, radiative properties, and precipitation production.

Data availability

No primary research results, software or code have been included and no new data were generated or analysed as part of this review.

Author contributions

All authors discussed and contributed to the content and added to the writing and editing the manuscript.

Conflicts of interest

There are no conflicts to declare.

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