

## CRITICAL REVIEW

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## Snow particles physiochemistry: feedback on air quality, climate change, and human health

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During the last several decades, numerous researchers have provided evidence that physical and biogeochemical processes at air-snow/ice-water interfaces are very complex, and, in many cases, interlinked. This review focuses on the current state of knowledge regarding snow-borne particles. It integrates snow science from different angles: from the formation of snow and precipitation to transformations through natural and anthropogenic processes and impacts and snow management in urban areas sites. We discuss the physical, chemical, and biological characteristics of particles in snow, such as their composition, abundance, size distribution, ice nucleation properties, genomic features, and microphysical processes, in urban settings, remote areas of the Arctic, and remote industrial regions (oil sands). We explore physicochemical processes of snow particles: from microbial to emerging contaminants, like nano/microplastics, light-absorbing carbonaceous organics, halogenated and nanometals particles. We review the possible contributions of snow particles to atmospheric radiation and climate, biogeochemistry, human health, and urban snow management. We propose further research directions to improve understanding of air-snow feedback, and sustainable snow management in urban areas, in the age of emerging contaminants in a changing climate.

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### Environmental significance

Fresh snow is a snapshot of atmospheric processes. Airborne particles or aerosols are pivotal for heterogeneous ice nucleation, ice growth, accumulation of ice crystals, and snow precipitation. Snow can accommodate and uptake particles (natural, anthropogenic including emerging contaminants) and gaseous species, as it falls. In this paper, we explore the selected physicochemical characterization of snow-borne particles and their roles in complex snow transformations, biogeochemical processes, microphysics, and snow reflectivity or albedo. We discuss the role of snow particles on freezing-melting cycles and their impact on snow interactions with the Earth's atmosphere, lithosphere, hydrosphere, and cryosphere.

## 1. Introduction

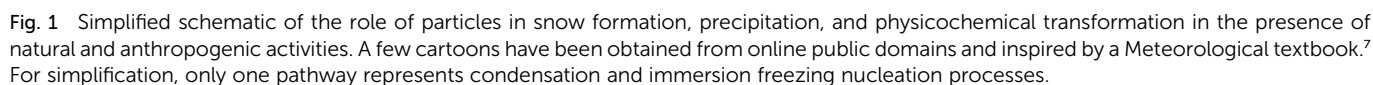
**Overview:** Fresh snow is a snapshot of atmospheric processes.<sup>1</sup> Airborne particles or aerosols are pivotal for heterogeneous ice nucleation, ice growth, accumulation of ice crystals, and snow precipitation. Airborne particles are also important factors in determining the type, intensity, and frequency of precipitation.<sup>2</sup> Snow can accommodate and uptake particles and gaseous species, as it falls. In this paper, we explore the selected physicochemical characterization of snow-borne particles and their roles in complex snow physicochemical transformations, biogeochemical processes, microphysics, and snow reflectivity or albedo (Fig. 1). We also discuss the role of snow particles on freezing-melting cycles and their impact on snow interactions with the Earth's atmosphere, lithosphere, hydrosphere, and cryosphere.<sup>3</sup>

Snow-borne particle characterization provides insights into the understanding of aerosol-cloud interactions, which is a research priority identified by the Intergovernmental Panel on Climate Change (IPCC).<sup>4</sup> The lack of understanding of aerosols and their interactions in cloud processes has been identified as an important uncertainty in the formation of precipitation. To address this challenge, the physical and chemical properties of aerosols, such as size, composition, contact angle – the liquid-vapor interface angle with a solid surface, hygroscopicity, surface properties, and photochemistry should be understood.<sup>5</sup> Interestingly, the health effects of aerosols are also determined by these physicochemical properties. The World Health Organization (WHO)<sup>6</sup> has considered airborne particles, particularly nanoparticles (<100 nm diameters), as a top research priority and a significant cause of premature human death. In this paper, we also explore how snow particles originating from the atmosphere can affect climatic processes and others that have also the potential for adverse health effects.

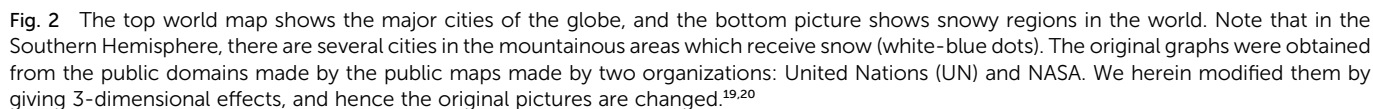
**Urban snow:** The urban areas cover only about 3% of the World's land surface.<sup>8</sup> Yet, urban atmospheric emissions are responsible for ~50% to 80% of the total global greenhouse gas

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gases and particles.<sup>13,14</sup> In polluted urban regions, snow has been shown to be effective for uptake of particulate matter, including airborne nanoparticles.<sup>15</sup> Yet, after snow events and upon melting-freezing cycles, the emission of airborne nanoparticles to the atmosphere is observed.<sup>15</sup> In urban regions, vehicle combustion particulate emission has been shown to alter snow composition.<sup>14,16</sup> Furthermore, the usage of salt





sites, in comparison to remote regions. This trend is not universally valid, as there are sources of anthropogenic activities in remote sites too. Thirdly, snow accommodates various particles and gaseous pollutants. Table 2 collectively illustrate that the same snow samples when exposed to gasoline exhibit much higher level of PAHs and BTEX in particulate forms. Fourthly, as shown in Table 3, in contrast to mostly anthropogenic organic compounds, snow concentrations of alkali, post-transition, transition, and rare metals are not always the highest in polluted regions. For instance, the highest concentration of iron is observed in Arctic sites due to the abundance of the

Compound	Type of snow	Concentration ( $\mu\text{g L}^{-1}$ )	Technique	Ref.
Benzene	Arctic	0.437–3.76	SPME/GC-FID	79
	Arctic	4.17	SPME/GC-MS	80
	Urban	1.48	SMPE/GC-MS	14
	Urban	6.48	SPME/GC-MS	80
	Alpine	0.069–0.148	HS-SPDE/GC-FID	81
Toluene	Arctic	0.271–2.81	SPME/GC-FID	79
	Arctic	43.6	SPME/GC-MS	80
	Urban	2.18	SMPE/GC-MS	14
	Urban	22.6	SPME/GC-MS	80
	Alpine	0.397–0.651	HS-SPDE/GC-FID	81
Chlorobenzene	Arctic	0.104–1.08	SPME/GC-FID	79
	Arctic	2.74	SPME/GC-MS	80
Ethylbenzene	Arctic	0.008–0.132	SPME/GC-FID	79
	Arctic	2.65	SPME/GC-MS	80
	Arctic	1.0–1.1	PTI/GC-MS	82
	Urban	0.13–2.7	GC-MS	83
	Alpine	0.074–0.29	HS-SPDE/GC-FID	81
<i>m</i> - <i>p</i> -Xylene	Arctic	0.162–1.89	SPME/GC-FID	79
	Arctic	0.65	SPME/GC-MS	80
	Urban	8.59	SPME/GC-MS	80
	Arctic	3.0–3.3	PTI/GC-MS	82
	Urban	0.17–7.4	GC-MS	83
	Alpine	0.026–0.169	HS-SPDE/GC-FID	81
	Arctic	3.46–39.0	SPME/GC-FID	79
Styrene	Arctic	0.22	SPME/GC-MS	80
	Urban	7.82	SPME/GC-MS	80
	Arctic	0.031–1.16	SPME/GC-FID	79
Trimethylbenzene	Arctic	0.4–1.3	PTI/GC-MS	82
	Alpine	0.030–0.477	HS-SPDE/GC-FID	81
	Arctic	7.18–20.1	SPME/GC-FID	79
Dichlorobenzene	Arctic	4.35	SPME/GC-MS	80
	Arctic	0.16	SPME/GC-MS	80
1,2-Dimethylbenzene	Urban	4.59	SPME/GC-MS	80
1,2,4-Trimethylbenzene	Arctic	5.41	SPME/GC-MS	80
Acetophenone	Arctic	1.25	SPME/GC-MS	80
1-Methylnaphthalene	Alpine	0.838	HS/GC-MS	84
Biphenyl	Alpine	1.05	HS/GC-MS	84
Fluorene	Alpine	1.11	HS/GC-MS	84
	Urban	0.001	SPME-GC-MS	14
Phenanthrene	Urban	0.208	SMPE/GC-MS	14
	Alpine	5.65	HS/GC-MS	84
Fluoranthene	Urban	0.417	SMPE/GC-MS	14
	Alpine	0.319	HS/GC-MS	84
Pyrene	Urban	0.097	SMPE/GC-MS	14
	Alpine	0.146	HS/GC-MS	84
Naphthalene	Urban	0.09	SPME/GC-MS	14
Acenaphthylene	Urban	0.223	SPME/GC-MS	14



**Table 2** Quantification of BTEX and PAHs in snow (in mg L<sup>-1</sup>) upon exposure of snow to exhaust engine emissions.<sup>14</sup> They were observed in melted snow, and they were condensed. Both BTEX and PAH have been shown to form droplets<sup>105</sup>

Pollutant	Unexposed snow	Exposed snow
Benzene	<0.001	0.052 ± 0.005
Toluene	<0.001	0.380 ± 0.038
Ethylbenzene	<0.010	0.243 ± 0.024
Xylene	<0.010	0.238 ± 0.024
Naphthalene	0.090 ± 0.010	0.485 ± 0.050
Acenaphthylene	0.004 ± 0.005	0.223 ± 0.025
Acenaphthene	0.010 ± 0.005	<0.001
Fluorene	<0.001	<0.001
Phenanthrene	0.017 ± 0.005	0.208 ± 0.023
Anthracene	<0.001	<0.001
Fluoranthene	0.004 ± 0.005	0.417 ± 0.046
Pyrene	0.002 ± 0.005	0.097 ± 0.011
Benz[a]anthracene	0.001 ± 0.005	0.089 ± 0.010
Chrysene	0.001 ± 0.005	0.029 ± 0.005
Benzo[b]fluoranthene	<0.001	0.074 ± 0.008
Benzo[k]fluoranthene	<0.001	0.338 ± 0.037
Benzo[a]pyrene	0.001 ± 0.005	0.058 ± 0.006
Dibenz[a,h]anthracene	<0.001	<0.001
Indeno[1,2,3-cd]pyrene	<0.001	0.043 ± 0.005
Benzo[ghi]perylene	0.001 ± 0.005	0.005

Arctic dust, partly transported from the Asian deserts and deposited in the Arctic snow,<sup>27</sup> whereas the highest concentration of cobalt is observed in snow in the Athabasca oil sands region (Alberta, Canada), as cobalt is commonly used in oil industries.<sup>40</sup>

Complementary to the chemical characterization and quantification in Tables 1–3, Fig. 3 illustrates various types of snow particle morphology, as well as particle size and number density distribution, respectively.

Several technologies have been used for chemical characterization of snow. Fig. 5 depicts the analysis using electrospray chemical ionization and matrix-assisted laser-induced ablation mass spectrometry spectra of biological material in snow. High-resolution electron microscopy images identify the existence of likely bio-organic particles in melted snow samples in Alert due to the optical contrasts in the image. Electron dispersive imaging (Fig. 5) indicates that the identical particles also contain various metals such as Al, Fe, and Mg, which are consistent with the ultra-trace metal analysis in Table 3, using triple quadruple inductively coupled plasma mass spectrometry (QQQ-ICP/MS/MS) for metals. Since there are various chemical compounds in snow as particles or attached to a particle, in the following section, we only focus on a few categories of snow particles, to better understand their characteristics and impacts.

### 2.1. Instrumental analytical analysis

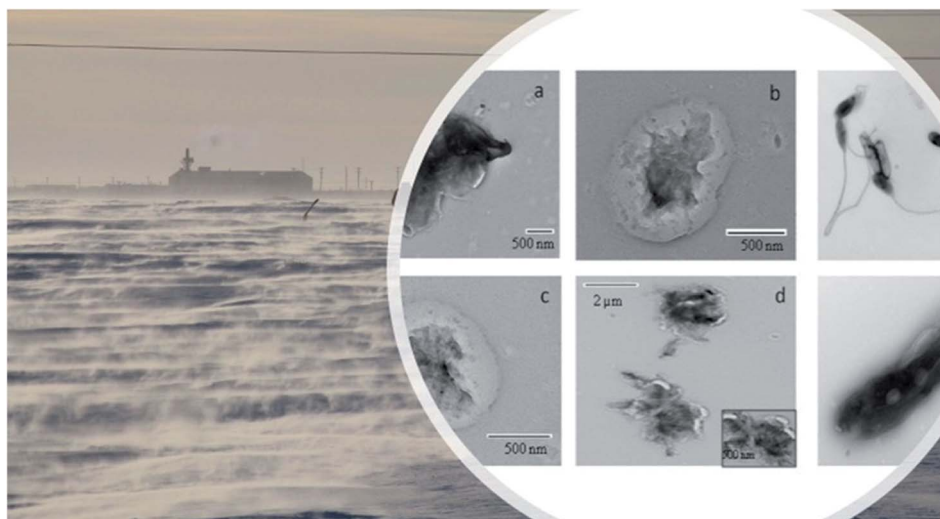
Characterization of particles in snow include the study of their chemical composition and physical properties like size and morphology, some of these include the following.

Mass spectrometry (MS): this technique is the most widely used to obtain the chemical composition of particles in snow.

**Table 3** Reported field data concentrations of alkali, post-transition, transition, and rare metals in snow/ice matrices. Different types of metals are in different colors

Element	Type of snow	Concentration (mg m <sup>-2</sup> )	Technique	Reference
Ba	Arctic	0.739	ICP/MS-MS	27
	Urban	0.779	ICP/MS-MS	27
	Urban	1.66–4.27	HR-ICP-MS	121
	Urban	0.25–2.49	HR-ICP-MS	121
	Oil sands	87.2	ICP/MS-MS	37
Sr	Arctic	3.622	ICP/MS-MS	27
	Urban	1.676	ICP/MS-MS	27
	Oil sands	29.0	ICP/MS-MS	37
Al	Urban	1.36–4.58	HR-ICP-MS	121
	Urban	0.70–2.50	HR-ICP-MS	121
	Oil sands	>100	ICP/MS-MS	37
As	Arctic	0.054	ICP/MS-MS	27
	Urban	0.181	ICP/MS-MS	27
	Urban	0.39–0.69	HR-ICP-MS	121
	Urban	0.09–0.23	HR-ICP-MS	121
	Urban	0.08–0.47	ICP-MS	122
	Oil sands	2.57	ICP/MS-MS	37
Cd	Urban	0.053	ICP/MS-MS	27
	Urban	0.004–0.012	HR-ICP-MS	121
	Urban	0.0013–0.004	HR-ICP-MS	121
	Urban	0.03–0.62	ICP-MS	122
	Oil sands	0.07	ICP/MS-MS	37
	Arctic	0.0083 ± 0.0032	ICP/MS	123
Co	Arctic	0.022	ICP/MS-MS	27
	Urban	0.447	ICP/MS-MS	27
	Oil sands	4.19	ICP/MS-MS	37
Cr	Arctic	0.034	ICP/MS-MS	27
	Urban	0.496	ICP/MS-MS	27
	Urban	1.49–3.22	HR-ICP-MS	121
	Urban	0.03–0.27	HR-ICP-MS	121
	Urban	0.01–4.29	ICP-MS	122
	Oil sands	15.7	ICP/MS-MS	37
Cu	Arctic	3.717	ICP/MS-MS	27
	Urban	1.267	ICP/MS-MS	27
	Urban	0.71–1.10	HR-ICP-MS	121
	Urban	0.33–2.65	HR-ICP-MS	121
	Oil sands	8.54	ICP/MS-MS	37
	Arctic	34.469	ICP/MS-MS	27
Fe	Urban	18.051	ICP/MS-MS	27
	Urban	0.85–30.53	HR-ICP-MS	121
	Urban	0.72–2.71	HR-ICP-MS	121
	Oil sands	>100	ICP/MS-MS	37
	Arctic	30.630	CVAFS	27
	Urban	3.294	CVAFS	27
Hg	Urban	0.062	ICP-MS	122
	Arctic	1.868	ICP/MS-MS	27
	Urban	2.392	ICP/MS-MS	27
Mn	Urban	3.63–30.9	ICP-MS	122
	Oil sands	>100	ICP/MS-MS	37
	Arctic	0.078	ICP/MS-MS	27
Ni	Urban	0.773	ICP/MS-MS	27
	Urban	0.33–0.92	HR-ICP-MS	121
	Urban	0.21–11.97	HR-ICP-MS	121
	Urban	0.37–3.71	ICP-MS	122
	Oil sands	39.3	ICP/MS-MS	37
	Arctic	0.245	ICP/MS-MS	27
Pb	Arctic	0.2743 ± 0.0104	ICP/MS	123
	Urban	0.743	ICP/MS-MS	27
	Urban	0.17–1.57	HR-ICP-MS	121
	Urban	0.02–0.15	HR-ICP-MS	121
	Urban	0.20–30.1	ICP-MS	122
	Oil sands	4.97	ICP/MS-MS	37
Zn	Urban	11.36–27.14	HR-ICP-MS	121
	Urban	7.30–20.56	HR-ICP-MS	121
	Urban	8.25–545.4	ICP-MS	122
V	Arctic	0.2336 ± 0.0033	ICP/MS	123
	Arctic	0.090	ICP/MS-MS	27
	Urban	0.144	ICP/MS-MS	27
Y	Arctic	0.016	ICP/MS-MS	27
	Urban	0.014	ICP/MS-MS	27
La	Arctic	0.014	ICP/MS-MS	27
	Urban	0.020	ICP/MS-MS	27
Ce	Arctic	0.027	ICP/MS-MS	27
	Urban	0.029	ICP/MS-MS	27
Pr	Arctic	0.004	ICP/MS-MS	27
	Urban	0.004	ICP/MS-MS	27
Nd	Arctic	0.017	ICP/MS-MS	27
	Urban	0.015	ICP/MS-MS	27





**Fig. 3** High Resolution Transmission Electron Microscopy (HR-TEM) images of selected biological nano-size and micron-size particles in the Arctic snow samples in an Arctic site (modification of Mortazavi *et al.*, 2015 (ref. 29)).

Common techniques using MS include GC-MS for SVOC and combustion products like PAHs.<sup>64–66</sup> Matrix-assisted laser desorption/ionization time-of-flight (MALDI-TOF-MS) for large molecules like polymers and biopolymers including DNA, proteins, and carbohydrates.<sup>1,47</sup>

**Induced Coupled Plasma (ICP):** For the analysis of metals in snow, ICP coupled with MS,<sup>27,38,67–69</sup> optical emission spectrometry (ICP-OES)<sup>41,70</sup> and atomic emission spectroscopy (ICP-AES)<sup>71</sup> are the primary methods used.

**Particle sizers:** these techniques give the particle size distribution in a sample. For some of these techniques, snow needs to be melted prior to the analysis. Dynamic Light Scattering (DLS)<sup>72</sup> and Nanoparticle Tracking Analysis (NTA)<sup>1,72</sup> are techniques that can calculate the size of a particle in a liquid based on their Brownian motion. Other techniques include the aerosolization of particles and the measure their sizes in air, these include Scanning-Mobility Particle Sizer (SMPS) Optical Particle Sizer (OPS).<sup>1,73</sup>

**Microscopy:** microscopy techniques such as scanning electron microscopy (SEM), and transmission electron microscopy (TEM) have been used to study particles after melting and drying<sup>74</sup> of snow samples.<sup>26,75,76</sup> While these techniques can analyze particles from a variety of chemical composition and sizes, the samples preparation prior to analysis can alter the particles and the distribution of particles within the samples, not to mention that obtaining particle distributions with these techniques is often challenging.<sup>77</sup>

## 2.2. Examples for snow-particle interactions with the earth ecosystem

**2.2.1. PAHs.** Polycyclic Aromatic Compounds (PACs) contain at least two fused aromatic rings and can contain other elements other than carbon like nitrogen and sulfur, however PACs that only contain carbon are more abundant, these are known as PAHs. The primary source of anthropogenic organic

aerosols found in snow is the combustion of fossil fuels and biomass burning, PACs being a common product of this combustion,<sup>85</sup> with an estimated production of 33.2  $T_g$  of soot particles containing PACs per year.<sup>86</sup> Low-molecular-weight PACs occur in the atmosphere mainly in the gas phase, whereas multi-ringed PACs are bound mainly to particles.<sup>87</sup> Studies suggest that the main source of PACs in snow is airborne particulate matter generated from local combustion,<sup>88</sup> namely diesel engine exhaust, and coal and biomass combustion.<sup>89</sup> PACs containing sulfur are dibenzothiophene and its derivatives are commonly found in bitumen and petroleum products indicating petrogenic activities as their origin. Canadian oil sands regions are common areas where these products are generated.<sup>39,90–92</sup> Thia- and aza-arenes are PACs containing nitrogen that can be found in bitumen, but also can be generated by combustion and mining of oils sands.<sup>39,90,93,94</sup>

The EPA considers 16 PAHs as priority pollutants, however, pyrene, phenanthrene, naphthalene, and fluoranthene have been identified as the most common PAHs in snow,<sup>89</sup> and several of these are known as carcinogenic compounds.<sup>95</sup> Other studies have indicated that many of the PAHs found in snow may end up in rivers and other water bodies after snowmelt in spring as part of the natural hydrological cycle of snow.<sup>96</sup> PAHs can also have toxic effects on aquatic organisms or be absorbed by plants and consumed by terrestrial animals making their way up in the food chain.<sup>49</sup> Upon snow melt, the release of airborne particles has been observed.<sup>15</sup> Human exposure is through inhalation of air and re-suspended soil and dust, consumption of food and water, and dermal contact with soil and dust.<sup>15</sup>

**2.2.2. Secondary organic aerosols.** Organic aerosols can be produced from volatile and semi-volatile organic compounds *via* atmospheric photo-oxidation and condensation processes. These particles are called Secondary Organic Aerosols (SOA). They can be further deposited on snow.<sup>97</sup> Table 1 shows reported data of volatile and semi-volatile compounds found in snow. Precursors of SOA can be anthropogenic and biogenic in



origin, however, biogenic sources produce 5.5 times more SOA precursors than anthropogenic sources. A large portion of these precursors are terpenes, which are usually oxidized by ozone,  $\text{NO}_3$ , and hydroxyl radicals among other oxidants.<sup>98,99</sup> It is also considered that many SOA are formed when partially oxidized organic vapors condense on ice particles and undergo chemical reactions. These are removed from the atmosphere when the snow precipitates.<sup>100</sup> SOA can act as ice-nucleating particles (INPs), but also water can condense on their surface to form water droplets and then act as INP.<sup>101</sup>

**2.2.3. Water soluble organic particles.** Water-soluble organic particles are released in high concentrations during the early stages of the snow melting process, whereas hydrophobic compounds and particle-bound compounds are released at later stages.<sup>102,103</sup> Depending on the snowpack depth, the release process efficiency differs. Shallow snowpack, for instance, experiences an irregular melting process (numerous melting episodes over winter), and thus releases small quantities of chemicals and particles throughout wintertime.<sup>104</sup>

**2.2.4. Biological particles.** Another type of organic aerosols of biogenic origin are bioaerosols, which include microorganisms and remnants of biological activities such as reproductive material (*e.g.*, pollen) and decaying matter.<sup>106</sup> Forested areas represent the major source of bioaerosols inland, yet, oceans are proposed to be the major sources of bioaerosols overall. It is believed that bubble-bursting in the ocean surface layer is a predominant pathway in which bioaerosols are released from the oceans into the atmosphere.<sup>107</sup> It has been revealed that up to 25% of insoluble aerosols are bioaerosols,<sup>108</sup> which can travel long distances and end up on snow.<sup>109–111</sup> Fig. 3 and 5a–d show some examples of particles, including biological particles of different sizes, from a few nanometers to a few micrometers, in Arctic snow samples.

Next generation sequencing (NGS) technology or massively parallel sequencing approach to DNA sequencing,<sup>27</sup> has allowed the rapid sequence of the entire genome of a species like humans. NGS has also evolved in environmental microbiology genomic research significantly. During the last decade, the usage of NGS paved the way for further understanding of the large types of biological particles in snow, such as bacteria, in urban and remote sites.<sup>29</sup> Most environmental microbes cannot be cultured.<sup>28,112</sup> NGS provides an opportunity to identify all potential bacteria communities or other microorganisms in a given snow sample. NGS analysis may allow distinction between bacterial communities in the Arctic and Urban snow.<sup>27,113</sup> For instance, in one study, the snow genomic analysis known phyla or candidate divisions were detected with the majority of sequences related to one of the five major phyla: Proteobacteria, Actinobacteria, Bacteroidetes, Firmicutes, and Cyanobacteria. As shown in Fig. 3, the morphology of biological particles was obtained using high resolution electron microscopy with energy-dispersive X-ray. NGS analysis led to the identification of metal interacting bacteria particles in all types of snow and frost flowers in the Arctic.<sup>27</sup> The combination of various cutting-edge mass spectrometry techniques, cryo-high-resolution electron microscopy and various optical sizers, along with NGS and culture methods, may allow much more

comprehensive understanding of the role of snow biological particles in the future.

**2.2.5. Inorganic particles and mineral dust.** Like organic aerosols, inorganic aerosols in snow can be of anthropogenic and natural origins. Mineral dust particles are one of the largest contributors to the formation of snow.<sup>114</sup> Mineral dust particles are generated from soil and ground by the effect of rapid winds. The common elemental composition of mineral dust includes metal elements like alkaline metals and transition metals (Fig. 5e and f). Deserts such as the Saharan desert in Africa and the Gobi and Takla-Makan deserts in China, are abundant sources of mineral dust in the Northern Hemisphere.<sup>115</sup> In the following sections, we discuss how dust and black carbon particles in snow can affect snow albedo and physicochemical processes such as melting.

**2.2.6. Other snow particles like sea spray and volcanic particles.** In the Arctic and sub-Arctic regions, sea spray, volcanic activities, and forested areas are among the most critical sources of particles. There is, consequently, a wide diversity in size distributions of particles in snow in the Arctic and sub-Arctic regions, as shown in Fig. 4,<sup>116</sup> which depicts the size-aggregated particles, including nanoparticles in snow, that exchange with the atmosphere.<sup>15</sup> Long-range transport of mineral dust, organic particles, and soot, among other particles, to the Arctic region further enriches the chemical diversity of particles that can be deposited on snow.<sup>117</sup>

Most of the aerosols generated by sea spray, volcanic activities, and forested areas are salts (like  $\text{Cl}^-$ ,  $\text{Na}^+$ ,  $\text{Mg}^{2+}$ ,  $\text{Ca}^{2+}$ ,  $\text{K}^+$ ,  $\text{NO}_3^-$ , and  $\text{SO}_4^{2-}$ ), along with organic components including biological compounds or organic remnants of biological processes like microorganisms' debris, membrane proteins or organelles.<sup>118</sup> Forested areas and vegetation are also among those generating biological particles (Fig. 2).<sup>119</sup> In general, unless the Arctic region is close to a significant source of particles, such as mining activities, the concentration of chemicals is smaller in contrast to urban regions (Table 1). Particles in the range of <200 nm are the most abundant particle sizes in snow and comprise 38–78% of the total number of particles (by mass, smaller particles make up a small fraction of all particles), with 11–19% under 100 nm.<sup>1</sup> Atmospheric aerosol size distribution systematically shows that nano-size particles (<200 nm) are the most abundant particles in the air in cold climate.<sup>15</sup> Hence, there is a similar trend with the abundance and size distribution of aerosols in the atmosphere and fresh snow, confirming that most upper layers of snow exchange efficiently with the atmosphere. Clearly, aged snow particles are expected to influence on surfaces such as soil and water, and upon freezing-thawing periods.<sup>120</sup>

**2.2.7. Importance of snow particles in selected photochemical processes.** There are several field observations as well as experimental and modelling research pointing to the importance of photochemical and/or heterogeneous reactions in the presence of snow borne particles.<sup>124,125</sup> For example, researchers have found that not only the snow albedo but also the photochemical processes can occur as a function of the black carbon content in snow.<sup>124</sup> Snowpack is an effective venue for photochemistry.<sup>55,56,126</sup> It has also been shown that



increasing black carbon concentration in the snow reduces the rate of photolysis of impurities and pollutants in the snowpack such as nitrate.<sup>127–129</sup> Furthermore, the redox reactions of toxic compounds such as mercury are shown to be affected by snow particles and biological particles.<sup>24,130</sup> Further research is recommended to evaluate the complex (photo)chemical reactions in snow matrices in the presence of a variety of particles.

### 2.3. Why do we care about snow-borne particles?

Here we summarize a few processes that make particle-bound species in snow important to understand in the Earth's ecosystem. The role of particles in the ice nucleation and potential implication on freezing–melting processes are discussed separately in Section 3.

**2.3.1. Albedo (reflectivity).** In brief, the importance of snow particles such as black carbon on radiative properties (albedo), as well as changing properties and air–snow interactions have been studied.<sup>131</sup> Previous research has found a significant correlation between black carbon in snow and spectral surface albedo.<sup>131</sup> For example, the addition of even 10 ng per gram of equivalent black carbon in snow lowered the snow's albedo by 0.004, noting the recorded concentrations of black carbon in ice cores usually range from 0.01 to >20 ng g<sup>−1</sup>.<sup>131,132</sup> Increasing the amount of the equivalent black carbon by an order of

magnitude further results in the albedo reduction effect increasing 5-fold.<sup>131</sup> Current atmospheric models fail to predict with high accuracy the real distribution of particles in the atmosphere, including the transport of particles from their sources to snow-covered areas and their deposition, as well as their effects on snow albedo, snow grain shape, and the particle–snow internal mixing.<sup>133,134</sup> Particles that change snow albedo can also modify the melting and freezing processes of snow.<sup>27</sup> Black carbon reduces snow albedo, which increases the absorption of radiation, resulting in early snowmelt.<sup>135,136</sup> Research has also shown that trace amounts of dust, HULIS (HUMic Like Substances) and black carbon deposited on or within a snowpack can reduce its albedo relative to pure snow.<sup>137,138</sup>

**2.3.2. Ecosystem and health.** As mentioned earlier, certain particles in snow are hazardous to ecosystems and human health (such as PAHs and toxic metals).<sup>121</sup> Furthermore, with the development of newer cutting-edge technologies, emerging contaminants such as nano- and micro-plastics are being detected in the Arctic and urban snow.<sup>52</sup> During the precipitation, melting, and evaporation cycle of snow, these contaminants are released back to the air and surrounding surfaces like soil. More water-soluble species can also be released into aquatic systems especially as snow melts in spring. This results in the discharge of pollutants stored in the snowpack into the

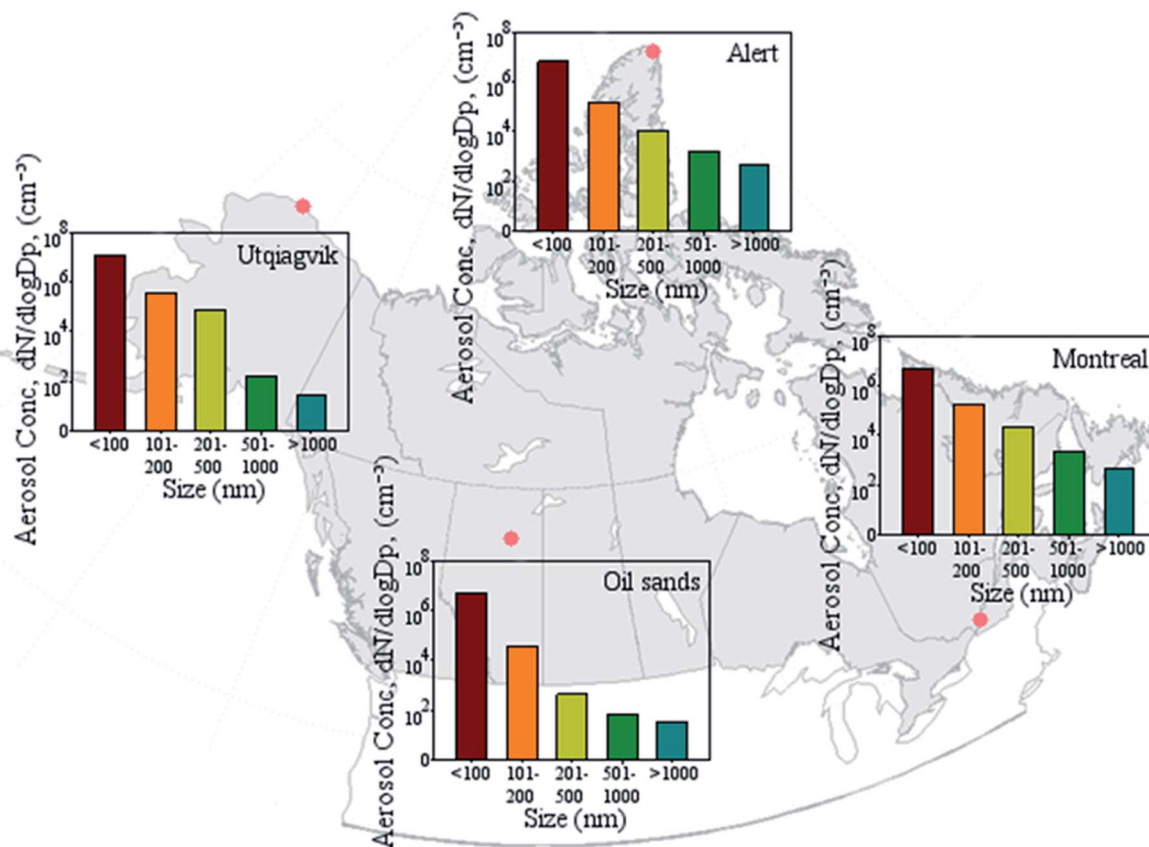


Fig. 4 Abundance of different size fractions in North American snow. Data obtained from references.<sup>1102</sup> The data was obtained using a scanning mobility particle sizer and an optical particle sizer with size limits from 11.5 to 9033.88 nm. Each color represents a different size fraction, which is on x-axis.







Fig. 5 Images (a and b) show electrospray chemical ionization and matrix-assisted laser-induced ablation mass spectrometry spectra of biological material in snow, and images (c and d) are high resolution electron microscopy images identifying the existence of likely bio-organic particles in melted snow samples in Alert due to the optical contrasts in the image. Images (e and f) show electron dispersive imaging indicating that the same particles also contain various metals such as Al, Fe, and Mg. The analysis was done on a copper grid<sup>1</sup> and thus the Cu signal is predominantly from the grid itself.

environment. Exposure to PACS and PAHs from the snowmelt can lead to toxic effects like reduction and delay of reproduction of aquatic lifeforms, increased physiological sensitivity, and reduced physical abilities like swimming and aerobic capacity.<sup>139,140</sup> Other effects include increased indices of cancer and death.<sup>94</sup> Toxic effects of metals in snowmelt contaminated by oil sands include increased sensitivity of fish and amphibian species and delays in some developmental stages.<sup>141,142</sup>

**2.3.3. Importance of addressing the shortcomings of current air quality and aerosol.** Numerical models currently provide regional and global understanding of particle distribution in snow, aerosol-cloud interactions, and reactions at snow/ice surfaces.<sup>143</sup> These models use simplified schemes to investigate the role aerosols and aerosol-cloud interaction play in air pollution.<sup>133,143</sup> However, uncertainties make the

prediction of air quality, cloud processes, and climate change challenging.<sup>4</sup> Furthermore, blowing snow transport and sublimation can further change the surface distribution of snow, redistribute ice particles vertically, and cool and moisten the atmospheric boundary layer.<sup>144</sup> In a northern urban setting, pollution from anthropogenic activities, namely local traffic and industry combined with long-range transport, adds to the complexity of snow composition and makes the understanding of snow processes more difficult.<sup>145</sup> Yet, they should be understood, as they are required for regional and global air quality and the climate processes.

**2.3.4. Snow as a reaction medium: scavenging and transformation of particles.** During or after snow formation, aerosols can be scavenged in clouds. Scavenging of aerosols can also occur after snow precipitates from the clouds. Snow crystals and

**2.3.5. The potential impact of salt particles on air quality.** In the Arctic, it has long been established that there is halogen exchange between the upper layer of snow and the atmosphere.<sup>59,156–158</sup> In urban regions, during the cold seasons,

In a laboratory experiment done in an environmental chamber (Fig. 6), it was found that both snow and exhaust emissions affected each other. Snow modified the size distribution (a shift of the modes to lower sizes) of exhaust, presumably by limiting its growth.<sup>16</sup> Additionally, exhaust exposure changed the chemical composition of snow. The



Fig. 6 At LHS, a simplified schematic of snow chamber-exhaust facility is shown.<sup>16</sup> At RHS, high-resolution transmission electron microscopy (HR-TEM) with energy dispersive spectroscopy (EDS) of duplicate melted snow samples in a site before and after exhaust exposure, are illustrated. An increase of particles in snow upon gasoline exposure was also observed.

concentrations of benzene, toluene, ethylbenzene, xylene (BTEX), and some PAHs were higher in snow samples subjected to exhaust emissions compared to fresh snow, as shown in Table 2.<sup>16</sup>

### 3. The role of particles in ice nucleation processes: impacts on freezing-melting process

Clouds are required for snow precipitation, but not all clouds make precipitation.<sup>7</sup> Indeed, only a tiny fraction of clouds precipitate, such as nimbostratus clouds that can produce snow (Fig. 1).<sup>7</sup> Several physicochemical processes are needed, from the formation of ice nuclei and their growth, to snow precipitation. In this section, we explore a brief background about the role of particles in heterogeneous nucleation processes, and examples of different compositions and sizes of ice nuclei.

#### 3.1. Heterogeneous ice nucleation: a brief survey of theory

Homogeneous nucleation is a process in which water vapour molecules become solidified or frozen, in absence of a foreign phase (aka a nucleating agent), at low temperatures ( $\leq 37^\circ\text{C}$ ).<sup>7</sup> Heterogeneous nucleation follows almost the same concept of homogeneous nucleation. However, this type of nucleation involves preferential sites such as phase boundaries, surfaces,

or impurities like aerosols such as dust.<sup>164</sup> Aerosols serve as nucleating agents on which ice crystal growth occurs. In heterogeneous nucleation, due to lower surface energy, the free energy barrier reduces and facilitates nucleation at these preferential sites. The existence of aerosols renders heterogeneous nucleation to require less activation energy than homogeneous nucleation. As such, the freezing process occurs at a higher temperature in heterogeneous nucleation processes in comparison to homogeneous nucleation processes.

There are four different types of heterogeneous nucleation: these are (a) deposition freezing, (b) condensation freezing, (c) contact freezing, and (d) immersion freezing, as shown in Fig. 1.

To better understand heterogeneous nucleation, we consider a hypothetical scenario that the nucleation proceeds *via* immersion freezing. When a spherical nucleus is formed at a preferential active site (considered from here on a surface), the spherical surface is reduced, and thus the area of the interface between the nucleus and the surrounding fluid, for instance, supercooled liquid water, is less than  $4\pi r^2$ . This geometric reduction decreases the surface area term in eqn (1) and thus lowers the nucleation activation barrier.<sup>165</sup>

$$\Delta G_{\text{Homo}} = -\frac{4}{3}\pi r^3 \Delta G_V + 4\pi r^2 \gamma \quad (1)$$

Here  $\Delta G_V$  is the difference in Gibbs free energy per unit volume between the thermodynamic phase where ice nucleation is



taking place and the phase that is nucleating,  $r$  is the ice nucleus radius, and  $\gamma$  is the surface energy/tension between the ice nucleus and its surroundings. At very low values of  $r$ , the surface term ( $r^2$  term) dominates, and the free energy is positive, and at higher values, the volume term ( $r^3$  term) dominates, and the Gibbs free energy is negative.

In the Earth's atmosphere, heterogeneous nucleation is achieved using aerosols as seeds for heterogeneous ice nucleation (Fig. 1). For heterogeneous ice nucleation, eqn (1) can be corrected by a factor to compensate for the loss of surface area and volume.

$$\Delta G_{\text{Heter}} = \Delta G_{\text{Homo}} \times f(\theta) \quad (2)$$

where  $f(\theta)$  is a function of the contact angle between the particle and the ice embryo.<sup>166</sup>

$$f(\theta) = \frac{1}{4}(2 + \cos \theta)(1 - \cos \theta)^2 \quad (3)$$

The contact angle ( $\theta$ ) is the degree between the interactions of the ice embryo with the INP. It should not be confused with the contact angle used to measure the wettability of a surface that is commonly used in material sciences. Geometric observations show that as the contact angle decreases, and so does the spherical surface area. Therefore, the smaller the contact angle, the lower the activation barrier and hence, the faster the nucleation rate.<sup>165</sup>

In laboratory studies, the contact angles of ice nucleating materials, including those found in snow, are usually calculated as described in Marcolli *et al.* (2007).<sup>167</sup> Contact angles ( $\alpha$ ) were calculated from the compatibility function  $f_{\text{het}}$  for supercooled water.

$$f_{\text{het}} = \frac{1}{4}(2 + \cos \alpha)(1 - \cos \alpha)^2 \quad (4)$$

Which is responsible for the reduction of the Gibbs free energy  $\Delta G$  barrier in the presence of an ice nucleating particle.

$$\Delta G(T) = \frac{16\pi}{3} \frac{v_{\text{ice}}^2(T)\sigma_{\text{sl}}^3(T)}{[kT \ln S(T)]^2} \quad (5)$$

where  $v_{\text{ice}}$  is the volume of a water molecule in ice,  $\sigma_{\text{sl}}$  is the interfacial tension between water and the ice embryo, and  $S$  is the ice saturation ratio. These parameters were calculated as in Zobrist *et al.* (2007)<sup>168</sup> using the heterogeneous ice nucleation rate coefficient  $j_{\text{het}}$  and the surface area of the ice nucleating particle in a droplet of water.

Particles with small contact angles tend to be more efficient at nucleating ice than large contact angle particles. Water suspensions of various environmental particles, including those found in snow, have been shown to exhibit strong properties to act as effective ice nuclei.<sup>169,170</sup> For example, long-term studies of snow in the Arctic and sub-Arctic and mid-latitude sites indicate that melted snow freezes above  $-10^\circ\text{C}$ ,<sup>1,171</sup> even after removing particles larger than 200 nm, indicating that snow nano-size particles, including nanoparticles can be good

ice nuclei. These agglomerates of nanoparticles, still nanosized, have a high surface-to-volume ratio and sufficient curvature, to have adequate contact angle for ice nucleation. A few laboratory studies have shown that such nano-size agglomeration of different materials can lead to high ice nucleation capability.<sup>169,170,172</sup>

Diverse types of particles are suggested to act as INP in the atmosphere, *e.g.*, metal containing particles (*e.g.*,  $\text{Fe}_2\text{O}_3$ ), mineral dust, soot, and biological particles (*e.g.*, airborne bacteria and viruses), and emerging contaminants such as nano- and micro-plastics.<sup>97,172</sup> INP can also nucleate ice directly from water vapor or act as cloud condensation nuclei to form a water droplet and then freeze it.<sup>173</sup> Three factors are predominantly responsible for enhancing the rate of nucleation: (i) an overlayer of water acting like a template material, having a similar crystalline structure to ice;<sup>169</sup> (ii) the appearance of a contact layer, being buckled in an ice-like fashion, and (iii) improved nucleation on compact surfaces, having very strong adsorption energy.<sup>174</sup>

### 3.2. Examples of particles' impact on ice nucleation and freezing-melting processes

Previous studies have confirmed the presence of different microorganisms (*e.g.*, bacteria, algae, and viruses) on snow and glacier ice.<sup>175</sup> For example, some groups have investigated the genomics of bacterial populations in snow,<sup>26</sup> identifying five major phyla: Proteobacteria, Actinobacteria, Bacteroidetes, Firmicutes, and Cyanobacteria, several of which have properties like ice nucleating.<sup>176</sup> Beside bacteria, several other types of microorganisms have been found in Arctic snow which seem to have resistance to heavy metals.<sup>27</sup> Fig. 3 and 4 illustrate the presence of biological material in Arctic snow samples and particles containing various metals, demonstrating how diverse particles in snow are and how biological matters coexist with various types of particles, including nanoparticles, which are abundant in snow.

During the last two decades, research has been focusing on understanding how some of the most efficient INPs work, namely the bacterial ice-nucleating protein (InaZ),<sup>177</sup> and how some processes affect immersion freezing of well-known INP (*e.g.* the oxidative polymerization of catechol on dust particles<sup>178</sup>). There has also been researches on novel materials that can act as INPs like graphene-graphene oxide nanoparticles<sup>179</sup> and inorganic composite particles.<sup>170</sup> Recent studies indicate that several types of anthropogenic particles, including emerging compounds such as nano- and micro-plastics and synthetic complex inorganic materials,<sup>169,172</sup> in addition to dust and biological particles, can serve as effective ice nuclei.

Human activities continuously create new materials, including nano- and microparticles, which are released in the environment. Such particles are detected in the air,<sup>43</sup> and they are deposited<sup>15</sup> in snow. As such, there will be likely new ice freezing nuclei and anti-freeze materials, which will be released in the Earth's ecosystem, facilitating freezing or melting processes in the future.





## 4. Emerging contaminants in snow

Emerging contaminants refer to synthetic or natural compounds that are not monitored in the environment regularly. Yet, these compounds are known (or suspected) to have adverse effects on the ecosystem or/and on human health.<sup>180</sup> Emerging contaminants are considered to be of serious concern due to their potential health effects. Recent scientific and technological advances allow ultra-trace quantification of selected contaminants in the environment. Recent innovations in medical field allow researchers to assess health impacts of selected emerging contaminants. Such pollutants are everywhere, from microplastics in oceans to nanometals in electronic waste.<sup>181</sup> These emerging contaminants have also been observed in various types of snow, in Arctic regions, industrial sites, and Urban settings.<sup>158</sup>

Emerging contaminants include synthetic or natural micro-organisms and their products, which can adversely affect ecosystems and human health. These contaminants are widely distributed in the environment, from microplastics in oceans to nanometals in landfills. An example of this is the Athabasca Oil Sand Region in Alberta, Canada, where the extraction and processing of oil sand generate aerosols that can be deposited in snow. Snow from this region contains high concentrations of C, Si, Al, and Fe.<sup>37,39,182</sup>

The concentrations of metallic species in snow, compiled from literature, are shown in Table 3. These species can be of lithogenic origin. They are released into the environment by activities and processes such as mining, land clearing, and wind-blown dust.<sup>182</sup> Metallic species that are recognized as priority pollutants by U.S. EPA and WHO (Cr, Ni, Cu, As, Se, Cd, and Pb) have also been detected in snow from oil sand regions<sup>54</sup> at concentrations that are 10 to 25 times higher than those in urban areas.

Snowmelt runoff can introduce these metals into aquatic bodies, which ultimately become concentrated in aquatic life and irrigated food crops. Consumption of contaminated fish and crops increases human exposure to these toxicants.<sup>183–185</sup> Recent research confirms the existence of nano- and microplastics (<5 µm) in snow.<sup>44,186,187</sup> These particles are in low concentrations but are present globally – from the Arctic to the Alps and cities and Arctic sea ice.<sup>44,188,189</sup> The sources of these particles are suspected to be the protective coatings on ships, vehicles and buildings, containing various plastic materials such as polyethylene and polystyrene.<sup>44</sup>

In recent decades, there has been rapid development of analytical techniques (Table 3) and remediation technologies relevant to emerging contaminants. For instance, several techniques have been used to detect microplastics in environmental samples like Raman spectroscopy and thermal assisted mass spectrometry.<sup>190,191</sup> However, pre-concentration methods are required, especially in the case of snow where concentrations can be in the ultra-trace level. New techniques are being developed and applied to the ultra-trace analysis of microplastics like nanostructured laser desorption/ionization time-of-flight mass spectrometry (NALDI-TOF-MS).<sup>47</sup> This technique

allows direct micro-/nano-plastic analysis in different environmental matrices like snow, without requiring pre-treatment. It is beneficial to continue with the research and development of tools to detect and quantify microplastics.

### 4.1. Potential health impacts of microplastics

Large particles of plastics or macro-plastics have shown to have adverse health effects on the Earth's ecosystem. For instance, upon ingestion of plastics, certain wildlife starve to death or suffocate by plastics.<sup>192</sup> During the last decade, several studies suggested the potential adverse health effects of nano- and micro-plastics in ecosystem and human health.<sup>193</sup> There are a couple of evidence of health impacts in other animals.<sup>194–197</sup> One of these studies has shown that microplastic may affect the genome by inducing transcriptional change, immune response, and behaviour alteration in adult zebrafish.<sup>196</sup> In the second study,<sup>197</sup> the analysis of juvenile *Daphnia* revealed a variety of subtle responses of morphological traits. Yet, in adult *Daphnia*, alterations in the expression of genes related to stress response genes involved in body function and body composition were observed already 48 h after exposure. As for human exposure research, a recent study has evaluated microplastic consumption by humans.<sup>48</sup> They estimated that Americans consume 39 000 to 52 000 microplastic particles per year, depending on age and sex. These estimates increased to 74 000 and 121 000 when inhalation was considered. In brief, macro-plastic adverse health effects are well-established. However, there is growing evidence for other animals, and the adverse effect of microplastic on human and ecosystem health are likely, but further research is required to further substantiate it.

## 5. Light-absorbing carbonaceous particles in snow

### 5.1. Sources of LACs

Light-absorbing carbonaceous particles (LACs) consist of black carbon (BC) and light-absorbing organic carbon (*i.e.*, brown carbon, BrC). The primary source of BC are fossil fuel combustion and biomass burning. The dominant component of BC is elemental carbon (EC) which makes it strongly absorb solar radiation ranging from UV to near infrared. However, the sources of BrC are more diverse. BrC primarily comes from biomass burning and fossil fuel combustion, whereas the secondary sources include photochemical reactions of anthropogenic or biogenic secondary organic aerosol precursors,<sup>209–212</sup> and heterogenous reactions with NO<sub>x</sub>.<sup>210–213</sup> The complex matrices of BrC make it difficult to determine every component of BrC, but there are several representative compounds that have been proved to be BrC, such as HULIS, nitroaromatics, PAHs, and vanillin.<sup>210–212,214</sup>

LACs can undergo long-range atmospheric transport (LRAT) and then deposit on the surface of snow and glaciers. During this process, both wet deposition and dry deposition can take place.<sup>201</sup> For dry deposition, when the size and weight of LACs gradually increase, it would be easier for LACs to be deposited by wind. For wet deposition, which is the main deposition of



LACs, there are more ways to deposit LACs. For instance, LACs can serve as ice nuclei or cloud condensation nuclei to form ice, cloud, or rain. They can also be scavenged by precipitation. Although there are very limited human activities in the cryosphere like Arctic,<sup>215–218</sup> Tibetan Plateau<sup>219</sup> and Alps,<sup>220–222</sup> LACs can be deposited in cryosphere by LRAT, affecting regional and global climate in the end.

## 5.2. Optical properties of LACs

Absorption coefficient (Abs) is usually used to describe the absorption of LAC particles in a power-law relationship with absorption Angström exponent (AAE),

$$\text{Abs}(\lambda) = K \times \lambda^{-\text{AAE}} \quad (6)$$

where Abs is with the unit of  $\text{Mm}^{-1}$ ;  $K$  is a constant;  $\lambda$  is the wavelength of light with the unit of nm. Once the mass concentration of LACs is determined, mass absorption cross-section (MAC) or mass absorption efficiency (MAE) at a specific wavelength can be calculated by eqn (7),

$$\text{MAC}(\lambda) \text{ or } \text{MAE}(\lambda) = \frac{\text{Abs}(\lambda)}{C} \quad (7)$$

where MAC or MAE is with the unit of  $\text{m}^2 \text{g}^{-1}$ ;  $C$  is the mass concentration with the unit of  $\mu\text{g m}^{-3}$ . MAC is usually used to describe the optical property of BC or BrC particles, whereas MAE is used to describe the optical property of BrC solution.<sup>211</sup>

AAE of BC is close to unity. Consequently, BC shows less dependence on wavelength, strongly absorbing light from UV to near infrared. However, AAE of BrC varies from smaller than 1 to as great as 8,<sup>230</sup> indicating high wavelength dependence. As a result, the absorption of BrC sharply decreases with increasing wavelength, with strong absorption on UV but weak absorption on short-wavelength visible light. The value of AAE of BrC can be calculated in a selected range of wavelength by eqn (8),

$$\text{AAE} = -\frac{\ln(A_{\lambda_1}/A_{\lambda_2})}{\ln(\lambda_1/\lambda_2)} \quad (8)$$

where  $A_\lambda$  is the absorbance of BrC at wavelength  $\lambda$ , which can be measured by UV-vis spectrophotometer.

The measurement of BC absorption has been well summarized by Bond *et al.*<sup>223</sup> Briefly, there are various instruments based on several methods to measure the absorption of BC: (1) photoacoustic method, such as photoacoustic extintometer (PAX); (2) filter-based optical absorption method, such as aethalometer; (3) thermal-optical method, such as thermal optical reflectance (TOR); (4) laser-induced incandescence method, such as single particle soot photometer (SP2). Compared with the diverse methods for BC, to our best knowledge, there is no commercial instrument designed specifically for BrC. Although PAX has a short-wavelength laser module, it cannot distinguish mineral dust from LACs. Absorption measured by aethalometer can be used to estimate the contribution of BrC absorption only if the value of AAE is determined, whereas AAE for BrC in the ambient air will not be constant. Current methods to determine the absorption, mass concentration, and chemical constitutions of BrC are filter-

based. Generally, samples are collected on one filter and then are subtracted by water or methanol to do further analyses, including total organic carbon (TOC) analysis, UV-vis spectrophotometry, and GC-MS or LC-MS. The filter-based methods will not change the chemical properties of BrC. However, they may change the size distribution and morphology of BrC, causing bias in measuring the optical properties of BrC.

Fresh BC particles are nanosized spheres with sizes less than 100 nm.<sup>223</sup> Once emitted, BC will undergo internal or external mixing with other components in the air, including BrC, to form aggregates with sizes ranging from nanometers to micrometers.<sup>223</sup> Internal mixing or external mixing with BrC would also modify the optical properties of BC to cause enhanced absorption, which is termed as “lensing effect”.<sup>211,224,225</sup> In turn, the size and morphology of BC can affect light absorption of BrC, causing overestimation on MAC of BrC.<sup>226,227</sup> In addition, the effects of atmospheric evolutions of BrC are more complex. On the one hand, photochemical or heterogeneous reactions could facilitate the formation of new BrC components and lead to absorption enhancement.<sup>210,211</sup> On the other hand, these reactions may cause photobleaching of BrC,<sup>210,211</sup> decreasing the absorption contributed by BrC. The complex matrices of BrC make it difficult for researchers to comprehensively evaluate the overall impacts of various atmospheric processes on BrC, which, in the end, hinders the work on accurately modeling the climate effect of BrC.

## 5.3. The radiation impacts of LACs deposited on snow

The radiation impacts of BC have been comprehensively investigated and evaluated by other researchers.<sup>201,215,216,223,228</sup> To be brief, BC deposited in the cryosphere mainly comes from anthropogenic sources like fossil fuel combustion in winter and biomass burning in summer.<sup>201</sup> The deposition of BC in snow can cause radiative forcing (RF) with a few watts per square meter in fresh snow. Nevertheless, as snow ages, RF induced by BC will gradually increase to even as large as hundreds of watts per square meter, leading to the acceleration of glacier melt.<sup>114</sup>

Unlike BC, although BrC in snow has drawn more attention in the past two decades, there are only a few studies which investigated and evaluated the optical properties and RF of BrC in the cryosphere. Wu *et al.* summarized the knowledge of MAE and AAE of BrC but did not include the data of RF.<sup>225</sup> Table 4 provides a summary of up-to-date knowledge of the optical properties and RF of BrC in the cryosphere. As shown in Table 4, the RF of BrC is sometimes described as a ratio of radiative forcing of BC, which is due to lack of knowledge of precise components of BrC. As discussed above, the complex constitutions of BrC make it challenging to determine the mass concentration of BrC, and thus MAC of BrC can be either overestimated or underestimated. The RF of BrC can only be estimated as a ratio of RF of BC. And for the cases which provide RF in the form of numbers, the values of RF of BrC are not accurate either. Additionally, it is evident that AAE of BrC varies more than AAE of BC, which confirms the strong wavelength dependence of BrC. Moreover, the variation of AAE indicates BrC consists of both strong absorbers and weak absorbers,



**Table 4** The optical properties and RF of BrC in the cryosphere. <sup>a</sup>Indicates the measurement was done on dissolved organic carbon (DOC). <sup>b</sup>Indicates middle altitude of Asia. <sup>c</sup>Indicates the measurements were done on water-soluble BrC. <sup>d</sup>Indicates the values were reported as MAE; <sup>e</sup>indicates the measurements were done on water-soluble organic carbon

Site	Sample type	Method	MAC (m <sup>2</sup> g <sup>-1</sup> )	AAE	RF (W m <sup>-2</sup> )	Ref.
Tibetan plateau	Snow	UV-vis spectrophotome-ter	1.4 ± 0.4 (365 nm)	5.0 ± 5.9	0.43	248
Laboratory	Snow	UV-vis spectrophotome-ter	N/A	4.12–6.28 (330–400 nm)	1.23 per ppm of BrC	249
Altai mountain <sup>a</sup>	Snow	UV-vis spectrophotome-ter	0.45 ± 0.35 (365 nm)	2.59 ± 1.03	0.11	250
Global Arctic Asia <sup>b</sup>	Simulated in snow	GEOS-Chem, MERRA2, RRTMG	0.46–1.7 (300–700 nm)	N/A	0.0064, 0.13 (Arctic summer) 0.24 (Asia winter)	215
Arctic	Snow	Integrating-sandwich spectrophotome-ter, SP2	N/A	3.5–7.0	N/A	216
Arctic	Aerosol <sup>c</sup>	UV-vis spectrophotome-ter	0.10–0.98 (365 nm) <sup>d</sup>	8.5 ± 4.2	5–34% of BC	217
Arctic	Aerosol <sup>e</sup>	UV-vis spectrophotome-ter	0.70 ± 0.44 to 1.54 ± 0.75 (365 nm) <sup>d</sup>	2.91 ± 1.02, ~4.76 ± 2.28	N/A	218

making it hard for model researchers to parameterize the optical properties of BrC, causing deviation in modeling RF of BrC in the end.

Furthermore, neither experimental methods nor modeling methods could provide a relatively precise concentration of BrC in the atmosphere or snow/ice. For experimental methods, both water extraction and methanol extraction methods would discard insoluble organic carbon which may contain a fraction of BrC. The following absorption measurement by UV-vis spectrophotometer cannot reflect the contribution from insoluble BrC. For modeling methods, currently, the mass concentration of BrC is estimated by using a certain percentage of organic carbon,<sup>229,230</sup> which no doubt would cause deviation in the radiative impacts of BrC.

#### 5.4. Ice nucleation of LACs

**5.4.1. Ice nucleation of BC.** The ice nucleation of BC has been widely investigated in the past decades. However, the question whether BC particles are important and efficient INPs in the atmosphere is still under hot debate because there are many conflicting results from both laboratory experiments and field campaigns.<sup>223,231</sup> For example, some early research indicates BC can nucleate ice below −20 °C,<sup>223,232,233</sup> and can be as ice active as mineral dust.<sup>233</sup> However, later experiments found contradictory results, suggesting BC particles are poor ice nuclei.<sup>223,234–236</sup> Even for field campaigns from the same site, results can be contradictory. Cozic *et al.* reported BC enrichment in ice particle residuals, and therefore BC may be able to nucleate ice efficiently in the mixed-phase cloud at Jungfraujoch, Switzerland.<sup>220</sup> However, at the same site, Kamphus *et al.* and Kupiszewski *et al.* found that BC was depleted in ice residuals, indicating BC was unimportant in mixed-phase cloud ice nucleation.<sup>221,222</sup> One of the reasons for controversial results of ice nucleation abilities of BC is that BC may undergo different modes of heterogeneous ice nucleation. For instance, BC particles may be good ice nuclei under contact mode and immersion mode,<sup>198</sup> but poor ice nuclei under deposition mode

at homogeneous IN temperature.<sup>199</sup> Another factor which can affect the ice activation of BC is the mixing state of BC particles. Mixing with OC would modify the surface of BC particles, decreasing the deposition ice nucleation of BC.<sup>200</sup>

Although there are already several articles reviewing ice nucleation of BC, this work includes recent advances in Table 5 which can complement previous work. Recent work by Nichman *et al.*, Mahart *et al.*, and Zhang *et al.* applied the pore condensation and freezing (PCF) mechanism to explain the results they obtained on soot (BC), and PCF did fit better with their experiments than classic nucleation theory, which provide insights on future research.<sup>226,237,238</sup> PCF was firstly proposed to predict ice formation on porous materials.<sup>239</sup> The porous surface of soot or BC makes it suitable to apply PCF to predict the ice nucleation ability of BC.

Table 5 also gives clues of why the results of BC ice nucleation are contradictory. In these selected studies, materials chosen by researchers are usually different, such as lamp soot, diesel soot, and carbon black. Sources and combustion conditions strongly affect size, morphology, and chemical constitutions of BC particles, and eventually, results of experiments. Moreover, there is a need to prove the atmospheric relevance of these materials. For example, commercial carbon black particles used by Nichman *et al.*<sup>237</sup> and Zhang *et al.*<sup>226</sup> are manufactured for coating or painting, which are not considered as one of important sources of atmospheric BC, although the advantage of manufactured carbon black is that physicochemical properties of these particles can be controlled.

Consequently, it should be taken into consideration that to what extent, ice nucleation experiments based on these commercial materials or homemade combustion products can suggest actual processes of BC in the atmosphere. As such, one of the challenges in studying ice nucleation of BC is to generate BC particles with controllable physicochemical properties, including size, morphology, coating, and so on. Furthermore, these BC particles should be comparable with atmospheric BC so that conclusions based on these BC particles have



**Table 5** Selected laboratory experiments on ice nucleation of BC. RH<sub>i</sub> indicates relative humidity to ice, and SS<sub>i</sub> indicates supersaturation with respect to ice. RH<sub>i</sub> and SS<sub>i</sub> are describing the same physical condition. And *T* indicates temperature

Material	Size	Nucleation mode	<i>T</i>	Active fraction density	RH <sub>i</sub> /SS <sub>i</sub>	Ref.
Lamp soot	240 nm	Deposition	213–233 K	N/A	N/A	232
Soot	100–250 nm	Deposition	216–233 K	N/A	135–154%	251
Soot with OC coating	N/A	Deposition	223–226 K	N/A	1.22–1.70	244
Diesel soot	100–270 nm	Deposition	243 K	N/A	137%	240
Diesel soot	120–280 nm	Deposition	223 K	N/A	143%	252
			228 K		142%	
			233 K		136%	
Carbon black	800 nm	PCF	217–235 K	$0.6 \times 10^6$ to $1.2 \times 10^6$ cm <sup>-2</sup>	100–150%	237
Soot graphite	N/A	Deposition	228–258 K	$3.7 \times 10^{-2}$ to $5.2 \times 10^{-2}$ cm <sup>-2</sup>	1.0–1.35	241
				$6.4 \times 10^{-2}$ cm <sup>-2</sup>		
Soot	400 nm	PCF	218 K	N/A	100–180%	238
			233 K			
Carbon black	100–400 nm	PCF	227–235 K	N/A	1.15–1.45	226

atmospheric significance. Some researchers use miniature combustion aerosol standard (miniCAST) to generate BC particles in a stable and reproducible way,<sup>231,234,238,240–242</sup> which may indicate a feasible methodology to generate various BC particles. Whereas the selection of specific sizes has been achieved by using differential mobility analyzer (DMA).

Another challenge is how the mixing state will impact ice nucleation ability of BC. Fresh BC particles are nanosized spherules but will grow to aggregate and then agglomerate when aging in the atmosphere.<sup>223</sup> During this process, not only will the sizes grow, but also other aerosols, such as organic aerosols and dust, will be internally or externally mixed with BC particles, complicating the physicochemical properties of BC particles.<sup>201,223,226,238,243</sup> For example, besides the aforementioned “lensing effect”, the coating of organic carbon on BC particles can change the ice nucleation ability of BC, either enhancing<sup>232</sup> or, in most of cases, decreasing ice nucleation of BC.<sup>234,240,244</sup>

Externally mixing with other aerosols may also alter the surface of BC, affecting hydrophilicity, hygroscopicity and pores or cavities on the surface.<sup>223</sup> However, more research shall be done to investigate how these surface properties affect ice nucleation ability of BC. For instance, according to molecular dynamic simulation by Lupi *et al.*, hydrophilicity, simulated by introducing hydroxyl group on the surface of graphite, cannot generally predict ice nucleation ability of BC.<sup>245</sup> Yet, a recent study by Xue *et al.* indicates that the introduction of hydroxyl group on the surface of BC can greatly enhance ice nucleation ability.<sup>246</sup> It should be noted that Lupi *et al.* did simulation on the surface of graphite,<sup>245</sup> whereas Xue *et al.* did their experiments on the surface of graphene,<sup>246</sup> which may, to some extent, lead to the discrepancy between their results. Thus, more studies are favored on these properties to draw firm conclusions.

**5.4.2. Ice nucleation of brown carbon (BrC).** To our best knowledge, currently, there is no review on the ice nucleation of BrC. Although Laskin *et al.* and Hems *et al.* reviewed chemical processes of atmospheric BrC,<sup>210,211</sup> ice nucleation ability of BrC and the impacts of aging process on ice nucleation ability were not discussed. Knopf *et al.* summarized both experimental

research and field study on ice nucleation of organic carbon, which covers the components of BrC such as HULIS and PAHs.<sup>91</sup> BrC was not discussed specifically in their work, which may further cause discrepancy when modeling the effects of organic carbon on climate. Most of the components of organic carbon are transparent to light and have cooling effect on climate.<sup>201</sup> Whereas BrC particles can not only serve as ice nuclei but also absorb radiation, which makes them have more complex effects on the climate,<sup>211,212,225</sup> calling for discriminating BrC from other organic carbon when comprehensively evaluating the impacts of organic carbon on climate.

Moreover, it is necessary to bridge the gap between ice nucleation ability and optical properties of BrC. A recent study by Chen *et al.* reveals that HULIS entities are efficient ice nuclei, comparable with mineral dust and bioaerosols, under mixed-phase cloud conditions, demonstrating the potential impacts of atmospheric BrC in ice nucleation.<sup>247</sup> Conjugating optical properties and ice nucleation ability of BrC may facilitate the prediction of ice nucleation ability of BrC by measuring its optical properties. The change of optical properties can reflect the modification of physicochemical properties in molecular level and thus the change of ice nucleation ability.

A major challenge in evaluating ice nucleation of BrC is still the complexity of BrC constitutions discussed above. Knopf *et al.*<sup>91</sup> illustrated that amorphous organic carbon could exist in different phases depending on RH and temperature, from liquid phase to glassy phase.<sup>91</sup> The phase of organic carbon governs the mode of ice nucleation that it will undergo in the atmosphere. For example, HULIS, depending on its phase, could serve as immersion IN in supercooled droplets or as deposition IN under lower supersaturation.<sup>201</sup> As such, it is likely that the various components of BrC will serve as efficient INPs like HULIS in mixed-phase clouds, or poor INPs under other conditions. More experiments and field campaigns shall be done to provide more details on ice nucleation ability of BrC and the related conditions. Furthermore, another challenge, as illustrated above, is to discriminate ice nucleation of BrC from that of BC, so that the contribution from BrC to ice nucleation and RF can be assessed.





A significant and challenging task is presently to develop an instrument which can specifically detect BrC in the atmosphere. Such data will be pivotal to assess the contribution of BrC to climate change, considering the proportion of BrC relative to BC in the atmosphere may increase if fossil fuel combustion is constrained. In addition, an open-access database on BrC components would greatly facilitate the research on BrC. Further machine learning methods may be developed on the database to discriminate new components of BrC, in near future.

## 6. Snow management in cities in the age of increasing emerging contaminants

Snow removal is important municipal management for cold regions which receive a large amount of snow every year. Table 6 summarizes the situations in selected areas in the world, including annual snowfall and the amount of road salts used for snow removal. In urban areas, accumulated snow on pavements and roads will hinder pedestrians' walking and transportation. When ice forms on the ground, it will pose an even serious threat on vehicles and pedestrians. Furthermore, snowpack in urban areas will take part in several heterogeneous processes shown in Fig. 8, causing accumulation, transformation, and transport of particles and metals.

There are several methods to do snow removal worldwide, depending on local situations and municipal budgets. One of the most prevalent methods is using snowplows that can efficiently remove snow and ice after snowfalls. However, high labor costs and high rentals for snowplows make it necessary to combine other methods to lower the expenses. Another prevalent but cost-effective method is to spread deicers, such as NaCl, MgCl<sub>2</sub>, and CaCl<sub>2</sub>.

Table 7 shows several common deicers, their costs, and their lowest effective temperatures. NaCl is the most widely used deicing materials among these deicers because of its good availability and low cost. However, there are increasing concerns about using NaCl.<sup>279–283</sup> First, using NaCl will increase the salinity of fresh water when snow or ice melts, threatening the biota in fresh waters. Secondly, soil which absorbs

snowmelt with high salinity cannot support vegetation growth. Li *et al.* found that shrubs were severely damaged, due to deicing salt pollution of the soils, whereas arbors were less vulnerable.<sup>284</sup> More importantly, extra NaCl entering ecosystems will increase the mobilization of heavy metals such as Cu, Cd, and Pb (Fig. 8).

Mahrosh *et al.* reported that the application of road salts (NaCl) caused an increment of Cu in freshwater systems. The high concentration of NaCl mixed with Cu could lead to less egg survival, delayed hatching, and high percentage of deformities of Atlantic salmon.<sup>285</sup> Whereas Cl<sup>−</sup> may facilitate the dissolution of Pb in soils. And when water with a high concentration of Cl<sup>−</sup> enters municipal water supplies, the water plumbing may undergo dezincification and galvanic corrosion, thinning pipe walls and releasing Pb into drinking water, which severely threatens human health of residents.<sup>282</sup>

The atmosphere may also be affected from salt addition to snow in the cities. High concentrations of chloride found in urban cities in winter period, and the concentration of photo-active chlorine in the atmosphere showed a positive correlation with the concentration of chloride.<sup>18</sup>

*Economic costs of de-icing:* Snowmelt with high salinity causes corrosion of infrastructures and vehicles, bringing extra economic costs of repair and maintenance of infrastructures and vehicles. For instance, Vitaliano *et al.*, estimated that the damages caused by road salts in the US cost at least \$615 per ton for bridge repairs and highway maintenance. And the estimation on vehicular corrosion was \$113 per ton.<sup>286</sup> These costs may have been increasing in the past three decades, considering the expansion of the transportation network and the increment of vehicle ownerships.<sup>202</sup>

The current threshold recommended by USEPA for NaCl in the environment has been questioned and needs to be updated, considering the usage of road salts has been increasing in the past decades.<sup>203</sup> Beside NaCl, there are also some other de-icers that are used in the world, such as calcium magnesium acetate, urea, beet juice, salt brine, and abrasives.<sup>204</sup> However, due to the problem of costs and effectiveness, these de-icers are not used as widely as NaCl.

Another method for snow removal is to use anti-icers before snow, such as salt brine. And in aviation, organic fluids like

Table 6 Selected areas which receive snowfall during a year

Location	Snowfall	Annual budget for snow removal	Amount of road salts	Ref.
Montreal Canada	235.6 cm	166 m CAD	More than 200 kilotons	253 and 260
Calgary Canada	103.6 cm	40.9 m CAD	30 to 40 kilotons	254, 261 and 268
Helsinki Finland	74.0 cm	24 m EURO	80 to 100 tons	255, 262, 264 and 269
Oslo Norway	87.2 cm	5.3 m EURO	250 kilotons (nationwide)	256, 263 and 270
Stockholm Sweden	99.8 cm	190 m SEK	300 kilotons (nationwide)	257, 264 and 276
Sapporo Japan	437 cm	190 m USD	N/A	258 and 265
Ohio US	N/A	65 m USD	600 kilotons	259, 271 and 281
Illinois US	100.0 cm	100 m USD	522 kilotons	259, 266, 268 and 272
New Jersey US	N/A	92.5 m USD	375 kilotons	273
New York city US	93.7 cm	92.3 m USD	About 407 kilotons	259, 267 and 274





Fig. 7 Electron microscopy of frozen snow with embedded particles using high resolution Leica EM VCT100 cryo-stage on the Dual Beam (FIB-SEM; courtesy of McGill microscopy facilities). The person in a clean suit is Rodrigo Rangel who was collecting snow samples to be analyzed in Montreal, QC, Canada.

ethylene glycol are usually applied on the surface of airplanes for anti-freezing, but they are seldomly used in urban areas because of the cost and impacts on the environment.

Although the side effects of using NaCl have been discussed above, it is still the most common deicer in the world. At present, there is no cost-effective and environmental-friendly alternative for NaCl.<sup>282</sup> Thus, most efforts in snow management are focused on optimizing snow removal strategies to lower the cost of snowplows and reduce the usage of deicers.<sup>282,287</sup> However, it should be noted that, as discussed in

Sections 2 and 4, snowpack on the roads can serve as a good reservoir for vehicular exhausts and emerging contaminants. Even after snow removal by snowplows, it is necessary to treat those snow properly. Dumping snow into local rivers or streams would still introduce emerging contaminants and road salts to local water systems, leading to health concerns for residents. It might be a good idea to store snow in deposit sites built with impermeable materials and then treat snow or snowmelt to make sure it is clean and safe enough to enter the city's drainage or rivers.



Fig. 8 Air-snow particle interaction in urban areas.



**Table 7** The costs and lowest effective temperature of common de-icers. Temperatures were reported in Fahrenheit and converted into Celsius in this table. It should be noted that potassium acetate (Kac.) is usually used as solution with 50% concentration

Deicer	Cost	Lowest effective temperature (°C)	Ref.
NaCl	\$26 per ton	−9.4	275 and 277
MgCl <sub>2</sub>	\$95 per ton	−23.3	275
CaCl <sub>2</sub>	\$294 per ton	−28.9	275 and 277
Calcium magnesium acetate (CMA)	\$670 per ton	−6.7	275 and 277
Potassium acetate (Kac.)	\$4.50 to \$5.12 per gallon	−26.1	275, 277 and 278

## 7. Future directions

We need to better understand the influence of snow and ice surfaces on the processes and life cycle of air pollutants, with field work, laboratory research and modelling studies. We specifically require evaluating the chemical, physical and biological processes occurring on snow and ice surfaces from urban, rural, industrial, as well as remote sites and synthesize an in-depth understanding of the biogeochemical cycling and toxicological effects of snow-borne particles on living ecosystems as well as human health impacts.

Future research direction should address the existing challenges, namely:

- There is a need to better characterize the chemical composition, speciation, and abundance of organic volatile, semi-volatile compounds (including bio-organic), metal pollutants, and particulate matter in snow samples, in urban, rural, industrial, and remote areas. To do so, improvements in analytical methods and instruments are required.

- To advance the understanding of the impact of ice nucleation in snow/ice processes, we recommend: (a) to develop instruments that can generate various BC particles in a stable and reproducible way, like miniCAST mentioned above, so that results from different research groups are more comparable; (b) more studies shall be done on mixing states of BC by using various materials, especially organic carbon, to figure out how BC will interact with these materials and what sort of materials can enhance the ice nucleation ability of BC; (c) more characterizations on BC particles used in experiments shall be done to provide details and impacts of physicochemical properties of BC; (d) more research on the size distribution of BC particles are recommended, since size-resolved information of BC particles are significant for more precise modelling study and predictions.

- There are a couple of technologies which have a potential to contribute to future snow research. The first one is cryo-stage high-resolution microscopy that allows snow samples at their original temperature to be studied. Fig. 7 depicts a snow sample taken in an urban park (Mont Royal, Montreal, Canada) and

analyzed with such a unit (Leica EM VCT100 cryo-stage on the DualBea). This technique may obtain insights into structure particle distribution and morphology within frozen snow. Another promising technique is Digital Inline Holography Microscope (DIHM),<sup>205</sup> which has been used for atmospheric research<sup>205</sup> and marine research.<sup>206</sup> Both these technologies hold promise for future physicochemical snow research, particularly with the elements of robotics and automation.

- There is evidence of reactive urban grime, which refers to films deposited on urban surfaces, including snowy cities.<sup>207</sup> Potential biological effects on film extracts, using zebrafish embryo essays, evaluated a dose-dependent syndrome of abnormalities including cardiovascular, hematopoietic, and behavioral defects.<sup>207</sup> These films are composed of a complex mixture of organic and inorganic compounds, containing significant levels of inorganic nitrate and sulfate.<sup>14,16,208</sup> Yet, upon photolysis, they release air pollutants such as NO<sub>x</sub> (NO + NO<sub>2</sub>) and HONO in urban areas. There is not much known about the interactions of snow and urban grime, yet some components of urban grime such as PAHs have been observed in snow. Hence further research in the domain is recommended.

- We must answer fundamental questions such as: What is the fate of snow-borne compounds after photochemical aging? Do the aging processes change the emission fluxes of chemical compounds to the gas phase? How do chemical compounds of anthropogenic origin change the distribution and composition of nanoparticles in the air and snow at the snow grain level? What types of interactions do snow-borne anthropogenic materials have with the atmosphere, water, and soil? Can we use snow as a probe for new emerging contaminants, as some contaminants have been shown to be more concentrated in snow than rain? What is the life cycle of emerging contaminants in snow? What are the effects of the cycles of melt, precipitation, and freezing on chemical compositions and distribution of emerging contaminants?

## Conflicts of interest

There are no conflicts to declare.

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