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Do snow and ice alter urban air quality?

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Air pollution adversely affects the people and the Earth's ecosystems (Prüss-Ustün et al., 2016; Vallero, 2014). According to different estimates, air pollution is linked to between 5.5 and 9 million premature deaths annually (Brauer, 2016; Prüss-Ustün et al., 2016; WHO2014; Landrigan et al., 2018), worldwide. Several decades of research demonstrate that physical and chemical processes, notably meteorological events and atmospheric photochemical transformations of emitted pollutants, influence air quality (Grannas et al., 2007; Radke et al., 1980). For instance, many gaseous air pollutants such as volatile hydrocarbons and nitrogen oxides ($NO_x = NO + NO_2$) can lead to ozone formation (Brown and Stutz, 2012). Together with airborne particulate matter (aerosols), they affect not only the atmospheric environment and climate but also human health (Vallero, 2014).

The majority of our planet's human populations live in the cities. Urban regions are also major emitters of air pollutants. Urban emission of greenhouse gases (GHGs) contributes approximately 50%–80% to the total global greenhouse gas emissions (Satterthwaite, 2008). Urban and suburban locations are also significant emission sources of aerosols and airborne emerging contaminants. Snow is ubiquitous in many densely populated parts of the world, as well as more remote yet environmentally important polar, sub-polar and mountainous regions. Large parts of North America, Europe, Asia and parts of South America experience snowy seasons, or a permanent or near-permanent snow- cover. Numerous cities in the world receive some form of frozen precipitation annually. However, research is only emerging to answer the question whether urban air quality is affected by snow and ice. Should we revisit what we think determines air quality in cold and snowy urban sites?

The importance of snow and ice in aerosol-cloud interactions, atmospheric chemistry, biogeochemistry, climate change and polar sciences has long been recognized in the context of remote regions. For instance, in the Arctic, a large body of research points to the significance of frozen precipitation and ice surfaces on reflectivity (albedo), (photo) physical, chemical, and biogeochemical transformation of a wide range of organic, metallic, nitrogenized, halogenated, sulfur- containing species, and particulate matter (e.g., Albert et al., 2002; Grannas et al., 2007; Ariya et al., 2011). Yet, little is known about the role of snow and ice physics and chemistry in urban regions, where many air-pollutant emission sources are located (Fig. 1).

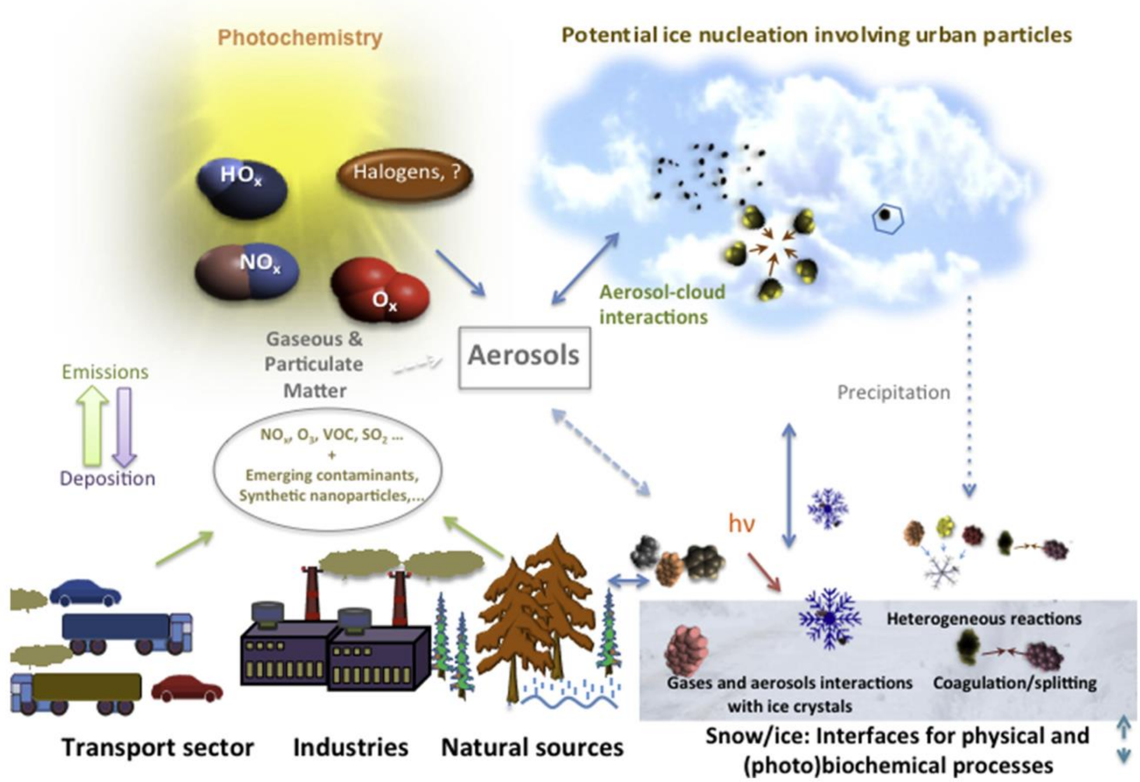

Fig. 1. Simplified schematic of chemical and physical transformations and interactions of gaseous and particulate air pollutants, including emerging contaminants, with snow and ice in urban areas. The arrows point to unknown effects of snow and ice in air quality, aerosol-cloud interactions and further chemical and physical transformations at snow/ice-air interfaces.

Besides particle scavenging by ice-nucleation and riming processes, gas-phase species also partition directly to the snowflake surface through adsorption processes (Bartels-Rausch et al., 2012). The enhanced partitioning of gas-phase chemicals to the snow surface at cold temperatures, combined with the large surface areas of snowflakes, results in snow being an excellent scavenger of semi-volatile organic chemicals from the atmospheric gas phase (Lei and Wania, 2004). Snow and ice surfaces modify boundary layer chemistry through the ex- change of contaminants by deposition and revolatilization processes. Snow can serve as an efficient scavenger for contaminants, and the porous nature of snow and the large extent of the snow cover during winter provide for a substantial exchange of organic chemicals between snow and the overlying atmosphere affecting atmospheric chemistry (Grannas et al., 2007).

Revolatilization of chemicals from the snowpack to the atmosphere may be driven thermodynamically in aging snow, and kinetically by wind ventilation (Hansen et al., 2006). In a modeling study, Stocker et al. (2007) found that, depending on the contaminant's physicochemical properties, and atmospheric and cryospheric conditions, snow and ice can either act as a barrier to deposition of contaminants to water, soil and vegetation or as a transfer medium, allowing for atmospheric uptake with subsequent release to water or soil during snowmelt. While the transfer rate between air and snow depends on the chemical's partitioning properties and the physical processing of contaminants in ice/snow, field and laboratory investigations have un- ambiguously demonstrated that organic-inorganic compounds can undergo chemical changes in ice or snow (Bartels-Rausch et al., 2012). Subsequent release of the products may significantly impact the com- position and chemistry of the overlying atmosphere, thus, the air quality. Recent microphysical studies propose the potential significance of some emerging nanoparticle contaminants in nucleation processes (Ganguly et al., 2018). Measurement of snow-air and snow-ice partition coefficients, and a comprehensive understanding of snow photo- chemistry and snowmelt-induced contaminant pulses are needed to adequately model these processes and assess their impacts on urban air quality.

Recent field and experimental studies provide evidence that urban snow and ice directly participate in retention, transport and transformation of a wide range of pollutants, and can affect urban air quality (Bućko et al., 2011; Kuoppamäki et al., 2014; Meyer and Wania, 2008; Nazarenko et al., 2017; Osipova et al., 2015; Vasić et al., 2012; Wei et al., 2017). A wide range of toxic and/or carcinogenic (semi)volatile organic compounds such as BTEX (benzene, ethylbenzene, toluene, and xylenes), and PAHs (polycyclic aromatic hydrocarbons) have been de- tected at elevated concentrations in snow after a short exposure to urban exhaust emissions (Nazarenko et al., 2017; Pirjola et al 2016). In the presence of snow and at cold temperatures, the general trends in exhaust-derived aerosol size distribution and abundance have also been observed to shift towards smaller particles, including nanoparticles (Nazarenko et al., 2017). The World Health Organization identified airborne nanoparticles as a major health concern, and basic research on the toxicity of particles shows substantial differences in adverse biological effects depending on the size of such particles (Chen et al., 2016; Slezakova et al., 2013). Yet, further research is needed to evaluate whether the impact of snow and ice on air pollution is significant.

In urban and suburban regions, both natural and anthropogenic environmental factors likely have the power to substantially affect atmospheric and interfacial air-snow processes. The factors at play include atmospheric oxidation potential, aerosol transformations, cloud nucleation, melting and freezing cycles, radiation and atmosphere- snow-ice-water-soil-building interactions.

The major modeling gaps contributing to wintertime urban boundary layer air pollution where research is lacking are: ice-cloud nucleation (the understanding is still very poor to model it), pollution scavenging and photochemistry on frozen hydrometeors, the modeling of dynamics, extent, depth and characteristics of surface snow and ice, and modeling of deposition and revolatilization processes, which are related to proper modeling of snow dynamics and characteristics, and chemistry in snow. Air-surface exchange processes of anthropogenic emissions are more dynamic and important in wintertime in comparison to the other seasons. In the presence of snow surfaces, the air- surface exchange processes can dramatically influence near-surface air quality, particularly in the presence of a persistent temperature inversion in the atmospheric boundary layer. Persistent low-level clouds have been suggested to impact air quality during wintertime cold conditions (VanReken et al., 2017). In winter, inversions can last a few days to several weeks, being triggered and maintained by anticyclonic conditions at synoptic scale, accounting for the sustained increase of pollutants in urbanized complex terrain during the inversion period (Largeron and Staquet, 2016a,b). Pollutants emitted by traffic and industry can be trapped in the inversion layer and experience prolonged interaction with the snow surfaces.

In this context, there is an emerging need for the field, experimental and modeling research addressing questions including:

1. Analytical capabilities for observation: Do we have all adequate technologies, preferably sustainable technologies, to study complex air-snow transformation processes at low temperatures and frozen environmental conditions in urban environments?
2. Observations: Can we identify the most harmful and persistent anthropogenic pollutants as they cycle and undergo transformations in the air, snowpack, meltwater, and soil? What are the effects of seasonality and snow coverage on

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pollutant concentrations and distributions in cities? What is the effect of multiple melting, freezing and precipitation cycles on pollutant concentrations and constant modification of the urban snowpacks by humans?

3. Processes: What kinds of physicochemical transformations occur within the snowpack in urban regions? What is the effect of snow transformations on concentrations, size distributions, and phases of pollutants? Can key emerging contaminants be targets for reduction, for instance in formulations of gasoline, and in the optimization of engines or optimization of the urban green environment?

Integration and decision-making: What are the most logical ways to model complex atmosphere-snow air quality processes in cold urban settings? What are different scenarios for combined effects of low temperatures and various types of frozen precipitation in the context of diverse polluted urban environments around the world? How can we properly evaluate air quality impacts on weather, cli- mate and health processes, locally, regionally and globally? How can we use the results of such evaluations to advise policy?

The research to answer these questions will strengthen the scientific basis for reduction of anthropogenic pollutants, which will contribute to the betterment of the environment and the human health.

Author contributions

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