


# On the Role of Starchy Grains in Ice Nucleation Processes

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**ABSTRACT:** Little is known about the role of starchy food on climate change processes like ice nucleation. Here, we investigate the ice nucleation efficiency (INE) of eight different starchy food materials, namely, corn (CO), potato (PO), barley (BA), brown rice (BR), white rice (WR), oats (OA), wheat (WH), and sweet potato (SP), in immersion freezing mode under mixed-phase cloud conditions. Notably, among all these food materials, PO and BA exhibit the highest ice nucleation efficiency with ice nucleation temperatures as high as  $-4.3\text{ }^{\circ}\text{C}$  ( $T_{50} \sim -7.0 \pm 0.5\text{ }^{\circ}\text{C}$ ) and  $-6.5\text{ }^{\circ}\text{C}$  ( $T_{50} \sim -7.2 \pm 0.2\text{ }^{\circ}\text{C}$ ), respectively. We also explore the effect of environmentally relevant physicochemical conditions on ice nucleation efficiency, including different pH, temperature, UV/O<sub>3</sub>/NO<sub>x</sub> exposure, and various cocontaminants. The change in shape, size, surface properties, hydrophobicity, and crystallinity of materials accounted for the altered INE. The increase in shape, size, and hydrophobicity of the sample generally reduces the INE, whereas an increase in crystallinity enhances the INE of the sample under our experimental conditions. The results suggest that environmentally relevant concentrations slightly alter INE, indicating their role as catalysts in environmental matrices. The outcome of studies on the ice nucleation properties of these food-containing aerosols might help in the physicochemical understanding of other biomolecule-induced ice nucleation, which is still an underdeveloped research area.

**KEYWORDS:** ice nucleation, starchy food materials, environmentally relevant, crystallinity, catalyst

## INTRODUCTION

The ice nucleation process in the atmosphere is a crucial and complex phenomenon with wide-ranging implications for the Earth's climate.<sup>1</sup> Atmospheric clouds form when aerosol particles are activated into liquid water and supercooled droplets or ice crystals in the atmosphere. The ice present in the atmosphere changes the radiative properties of aerosol influencing the aerosol–cloud interaction. These alteration in aerosol–cloud interaction have an effect on the climate change.<sup>1</sup> The precipitation occurs primarily via the ice phase,<sup>2</sup> making atmospheric ice nucleation extremely important in the global hydrological cycle.

Our planet experiences the phenomenon of ice nucleation frequently in various scenarios, from climate change to intracellular freezing.<sup>3–5</sup> Pure water can be supercooled to about  $-40\text{ }^{\circ}\text{C}$  without freezing.<sup>6</sup> The mechanisms driving efficient ice nucleation remain complex, primarily due to the diverse nature of nucleating materials, which range from biological compounds to nanoparticles and crystalline.<sup>7</sup> Since the last few decades, considerable research (both experimental and theoretical) has been focused on the relevant pathways and establishing an unambiguous driving force for efficient ice nucleation. However, the understanding of the role of food materials as ice nuclei remains poorly understood, and its physicochemical complexity has yet to be decoded.

Starch is a polysaccharide produced by plants and serves as energy storage in an osmotically inert form. Seeds, fruits, roots, and tubers are plants' primary sources of starch. Still, starch granules can also be present in pollen grains, showing the evolution patterns regarding the pollen size and pollination strategy.<sup>8</sup> Pollen grains rupture from the plant cells, releasing starch granules into the atmosphere and becoming airborne.

Laurence et al. used the term “starch grain” for airborne starch while describing notable contamination of archeological artifacts.<sup>9</sup> During thunderstorms, starch granules are released from the pollen grains by osmotic shock, which impacts environmental health as thunderstorm-triggered asthma.<sup>10,11</sup> Production and processing of cereal grains can cause the emission of airborne dust<sup>12,13</sup> that contains over 50% starch.<sup>14</sup> Starch dust coming out from anthropogenic activity, such as grain handling, can be hazardous to humans and animals because of its high organic contents and physical properties,<sup>15</sup> which makes it explosive.<sup>16</sup> Previous report suggest exposure to airborne starch have resulted in subclinical inflammation and an accumulation of eosinophils.<sup>17</sup> A large number of bioactive substances come out of the starch granules released from the grain handling process. It contains bioactive remains of plant cells, which, if inhaled, can cause baker's asthma, one of occupational energy's most critical and recurrent manifestations.<sup>18,19</sup>

Food and Agricultural Organization (FAO) estimated that 1.3 billion tons of food are wasted every year globally, about one-third of the annual production harvested for the consumption of human beings.<sup>20</sup> Agrifood waste emerges all around the food supply chain, from production to postharvesting, distribution, consumption, and domestic and industrial processing, with wastage amounts differing among phases and

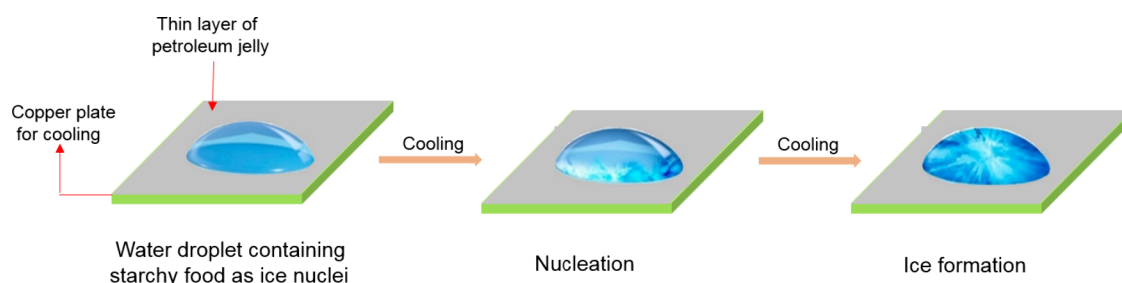
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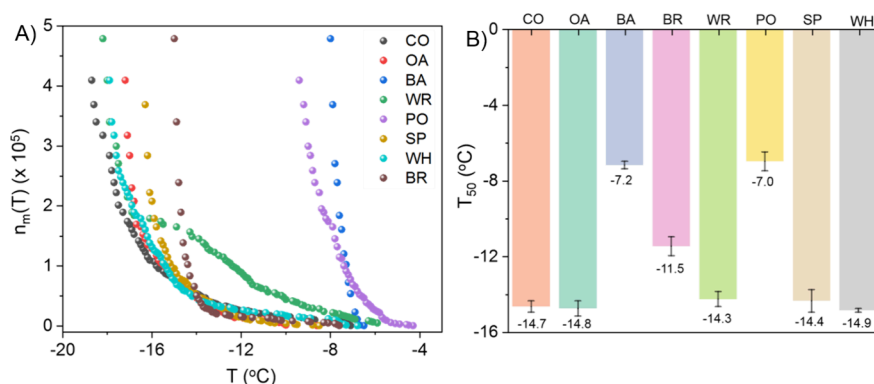
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**Figure 1.** Schematic representation of heterogeneous ice nucleation process where starch-containing food materials were used as heterogeneous ice nuclei.



**Figure 2.** (A) Ice nucleation spectra of different starch-containing food materials. (B) Comparison of  $T_{50}$  values of different starchy food materials used for ice nucleation experiment.

food commodities.<sup>21,22</sup> The anticipation is that the combination of increasing population and consumption on a global scale will give rise to heightened demand for food in the near future, thereby necessitating the intensified exploitation of natural resources, notably land, water, and energy.<sup>23</sup> As a result, global food consumption and global food waste generation will increase aggressively. The generation of a substantial quantity of food waste has additional implications for the atmosphere, particularly in terms of airborne aerosols<sup>24</sup> resulting from decomposed or undecomposed food waste, which consequently impacts atmospheric ice nucleation and cloud formation.<sup>25</sup> Disposal of undecomposed foods (starchy) and other decomposed organic waste (formed when bacteria and other microorganisms use these compounds as a source of food) in an open environment causes the formation of starchy bioaerosols that often get mixed with the soil dust and transported by air, resulting in atmospheric aerosols impacting human health and climate change.<sup>26</sup>

During the last few decades, various inorganic and organic materials have been listed to contribute to ice nucleation.<sup>27–31</sup> Till now, a Gram-negative bacteria, *Pseudomonas syringae*, is known to be the best ice nuclei, which freeze around  $-2^{\circ}\text{C}$ .<sup>27</sup> However, very little has been explored on the ice nucleating capabilities of starch-containing food materials. In this study, the ice nucleation ability of various starch-containing foods relevant to the atmosphere and atmospheric surfaces was investigated, employing immersion freezing experiments to evaluate their efficiency. The outcome of this study will contribute to our understanding of the ice nucleation efficiency of food materials in diverse environments. We also explored the factors responsible for forming highly efficient ice-nucleating particles and their environmental impact.

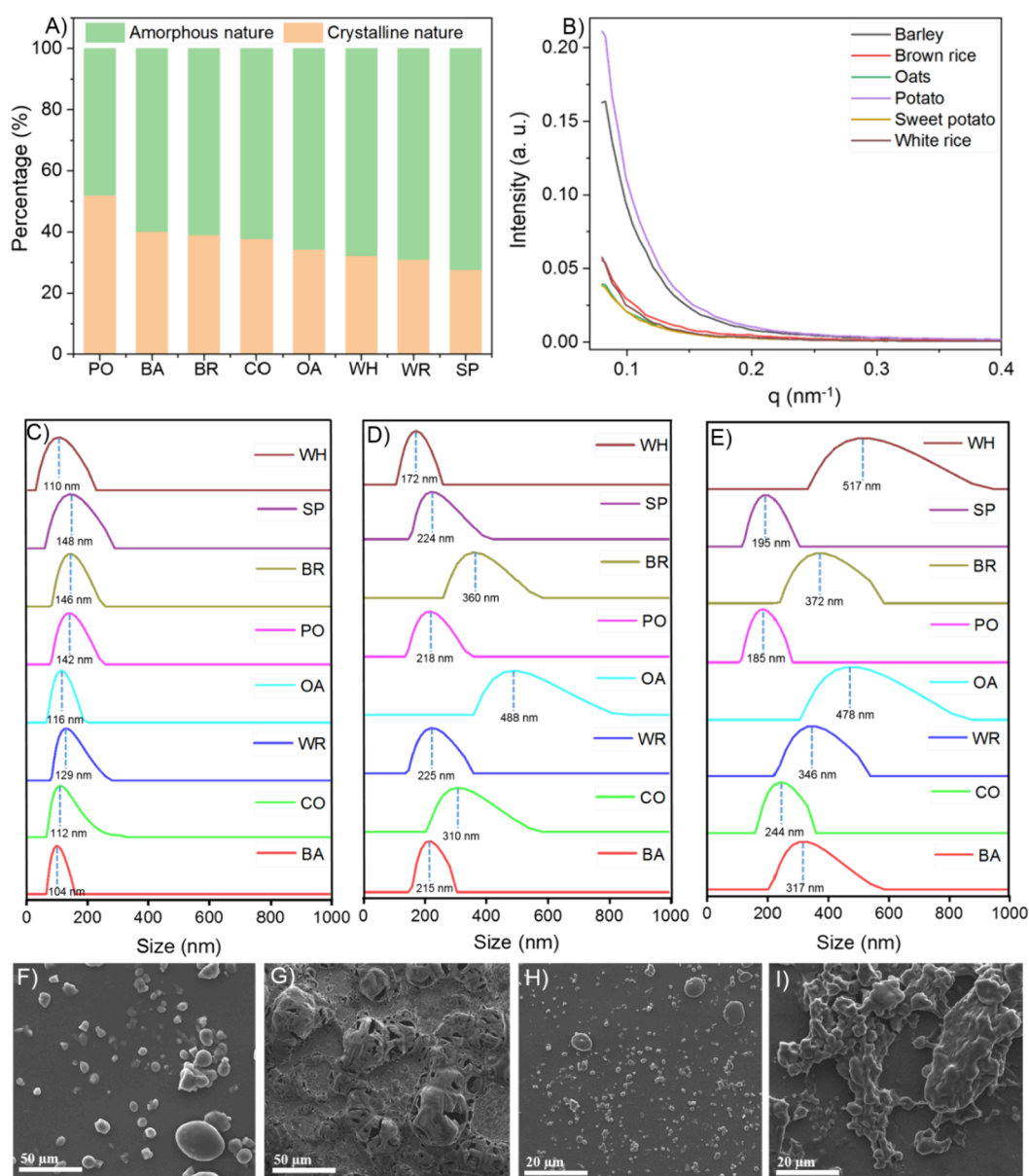
## RESULTS AND DISCUSSION

### Objective and Significance of Present Research.

Usually, primary food ingredients are considered biodegradable and pose less threat to the environment in terms of pollution and health hazards. However, their effect on climate change, especially their role in atmospheric cloud condensation, needs to be developed in research. Studying ice nucleation is extremely important because the formation of icy clouds at appropriate altitudes can scatter the incoming sunlight that might offset the global warming associated with the increased concentration of greenhouse gases. Besides, cloud formation is associated with rainfall, essential for growing vegetation and farming.

Previous reports suggest that starch is known to cause subclinical inflammation and asthma, indicating the presence of starch as an aerosol in the environment.<sup>17</sup> However, the role of these starchy aerosols in the climate in ice nucleation has yet to be investigated. A significant portion of the primary food ingredients consists of starch, which may end up in the environment as aerosols generated from food waste. These starchy aerosols get mixed with dust and transported by air, resulting in atmospheric aerosols that could impact the ice nucleation. Therefore, a few cereal grains containing the most common food staples are chosen to study their ice nucleation microphysics and evaluate whether they may affect climate processes. Food waste, especially starchy food materials, can be an essential source of atmospheric particles and might play a crucial role in climate anomalies in various regions, affecting the precipitation processes, like rainfall and human living.

**Ice Nucleation Studies of Different Starch-Containing Food Materials.** The schematic representation of the heterogeneous ice nucleation, where starch-containing food materials act as ice nuclei, is presented in Figure 1. We have



**Figure 3.** (A) Percentage of crystallinity and amorphous behavior of the starchy food materials calculated from their XRD spectra. (B) SAXS intensity distribution for colloidal sample solutions of different starchy food materials. Dynamic light scattering (DLS) performed on starchy food solutions (C) before acid treatment or heating, (D) after acid treatment, and (E) after heating at 60 °C. (F) SEM image of PO before acid treatment; (G) SEM image of PO after acid treatment showing aggregation of the sample. (H) SEM image of PO before heating; (I) SEM image of PO after heating at 60 °C resulted in aggregation of the sample.

investigated the ice nucleation efficiency of several starch-containing foods, that is, brown rice (BR), white rice (WR), barley (BA), potato (PO), corn (CO), oats (OA), wheat (WH), and sweet potato (SP), for the ice nucleation experiment. The ice nucleation process predominantly depends on the physicochemical properties of the ice nucleating particles (INP), as it provides sufficient enhancement of active sites for nucleation.<sup>32</sup>

To neutralize the effect of concentration, a similar concentration for each of food material was selected. We have observed that the BA and PO exhibit the most efficient INP among all the other tested contaminants with  $T_{50}$  values of  $-7.2$  °C (freezing range  $-6.5$  to  $-8.1$  °C) and  $-7.0$  °C (freezing range  $-4.3$  to  $-9.5$  °C), respectively (Figure 2A,B). In contrast, BR exhibits a slightly lower ice nucleating temperature with a  $T_{50}$  value of around  $-11.5$  °C (freezing

range  $-5.9$  to  $-18.5$  °C). On the other hand, WR, CO, OA, WH, and SP are not as efficient ice nuclei as BA and PO. WR, CO, OA, WH, and SP froze at much cooler temperature that shows  $T_{50}$  of  $-14.3$  °C (freezing range  $-7.6$  to  $-15.2$  °C),  $-14.7$  °C (freezing range  $-7.1$  to  $-18.7$  °C),  $-14.8$  °C (freezing range  $-8.6$  to  $-17.2$  °C),  $-14.9$  °C (freezing range  $-6.8$  to  $-17.9$  °C), and  $-14.4$  °C (freezing range  $-8.5$  to  $-16.3$  °C), respectively.

**Effect of Crystallinity on Ice Nucleation. X-ray Diffraction (XRD) Analysis.** All the tested food materials used for the experiments are rich in starch (60–80%), and their mean freezing temperature variations are observed. The chosen food materials contain a high quantity of starch and some quantities of proteins, fats, phospholipids, metals, and so forth.<sup>33</sup> However, these have minimal impact on the ice nucleation of the food materials. For further confirmation, we

have investigated the ice nucleation efficiency of commercially purchased pure starch (Sigma-Aldrich), which exhibited similar ice nucleation properties of some of the food materials (Figure S2), with BA and PO being the exceptions, reinforcing the idea that starch content predominantly dictates ice nucleation efficiency.

The presence of protein such as gluten does not significantly impact the ice nucleation of starchy food material, and the impact of other ingredients remains minimal. The ice nucleation depends on the properties of the material present on the surface. Due to the higher content of starch in the food materials, the surface of the liquid droplet is more likely to contain starch. The proteins are likely hindered by the starch that showed similar ice nucleation efficiency of these materials with pure starch, under experimental conditions.

To understand their different behavior as ice nuclei (difference in  $T_{50}$ ), their physical properties using XRD (Figure S3) and SAXS analysis were investigated. The XRD analysis revealed that the crystallinity of PO and BA was notably higher, at 52 and 40.2%, respectively, compared to the rest of the materials (Figure 3A). This enhanced crystallinity in PO and BA may be linked to their ice nucleation efficiency, as these materials outperformed others (Figure 3A). The percentage of crystallinity was found to be for BR, CO, OA, WH, WR, and SP, ranging from 27.7 to 39.1%.

All starchy food materials are made up of mainly two types of starches, that is, amylose and amylopectin.<sup>34</sup> PO is known to be a B-type (waxy starch) where the amylopectin structure is more branched and responsible for high crystallinity in these materials. On the other hand, cereal grains, including BA, are known as A-type starch. Two different features in XRD indicate the difference, like amylose and amylopectin present in both systems, which is directly related to the percentage of crystallinity in these materials. Thus, the higher crystallinity of PO and BA contributed to their enhanced ice nucleation efficiency compared to other materials (Figure 2B).

**Small-Angle X-ray Scattering (SAXS) Analysis.** Small-angle X-ray scattering (SAXS) measures the nanoscale density difference in aqueous and solid samples.<sup>35,36</sup> These density differences in samples can lead to determining the nanoparticle size distribution, resolve shape and dispersity (mono or poly) of macromolecules, pore size, local crystallinity, and so forth.<sup>37</sup> The scattering curves in SAXS contain information on the particle's size, shape, orientation, squared contrast, weighted concentration, and volume of the particles.<sup>37</sup> The obtained scattering curves are the summation of all the above factors, resulting in an averaged form factor that no longer shows sharp minima.

Here, we performed the correlation function analysis using sasview5.2 software to estimate the changes in phase or orientation of particles within the sample, which might cause an alteration in the local crystallinity (phase) of the materials.<sup>38</sup> Table S1 represents the local crystallinity of the sample during the specific phase change in a particular area of the material. Polydispersity measures the width of the size distribution of correlation function decay rates and not the particle size distribution. The change in phase (local crystallinity) and polydispersity of material significantly altered the ice nucleation capability of materials.<sup>39</sup>

To understand the physical characteristics that influence the ice nucleation properties of the different starch-containing food materials, several samples using SAXS were analyzed. These materials included BA, PO, BR, WR, SP, and OA. Among

them, some starchy materials were observed with good (BA, PO), moderate (BR), and poor (WR, SP, OA) ice nuclei. The SAXS data indicated that a combination of high polydispersity and local crystallinity is likely the reason for efficient ice nucleation for naming materials compared to others.<sup>39</sup>

Figure 3B shows the scattering intensity counts of the colloidal solutions at a given concentration of the experimented starchy food samples. Table S1 provides the polydispersity and crystallinity data for all the pieces. From Figure 3B, it was observed that PO and BA scattering intensity counts are higher than other ice nucleating materials such as BR, WR, SP, and OA. It indicates a combination of higher local crystallinity and polydispersity, leading PO and BA solutions to freeze at warmer temperatures than the rest. The overall effectiveness of the ice nucleation depends on the combined effect of local crystallinity and polydispersity. Higher polydispersity with lower crystallinity or vice versa may not always show effective ice nucleation. For example, in our case, BR and WR show higher local crystallinity but lower polydispersity values than BA or PO and end up showing ice nucleation at much cooler temperatures. Similarly, SP has higher polydispersity but lower crystallinity than BA or PO, resulting in freezing at a much-reduced temperature.

The materials with high crystallinity and higher polydispersity tend to have a polymorph surface. These surfaces lower the free energy, local structuring upon cooling, and preparing crystallization in a supercooled state.<sup>40,41</sup> Hence, it makes the droplets freeze at warmer temperatures. Density, poly surfaces, and polydispersity are control parameters to induce crystallization and favor structures acting as precursors for the ice nucleation process. However, crystallinity and polymorph surfaces are not the only factors favoring the condition for efficient ice nuclei; a more detailed understanding is required to understand this phenomenon.<sup>39</sup>

**Effect of Physicochemical Parameters. Effect of pH.** Starch is widely used in food, chemical, textile, medicine, and other industries. In the process of starch production, wastewater discharge is very large, each production of 1 ton of starch will produce 10–20 m<sup>3</sup> of wastewater. This wastewater mainly contains dissolved starch, a small amount of protein, organic acid, dust, minerals, and a small amount of oil and fat, easy to corrupt fermentation, making the water black and smelly; discharge into rivers will consume dissolved oxygen in the water, promoting algae and aquatic plant reproduction. When the amount of wastewater is large, the river will suffer from serious hypoxia, anaerobic corruption, and odorous smell, and marine animals may suffocate and die, thus threatening the human living environment.<sup>42</sup>

A wide range of pH values are observed in Earth's ecosystem. Soils, anthropogenic environments, and lentic water are acidic in nature<sup>43</sup> whereas the oceanic environments have an alkaline pH.<sup>44</sup> When starch is disposed in aquatic bodies, it can be subjected to a diverse range of pH. Therefore, the effect of pH was evaluated by performing experiments in all of the eight studied food materials at three different pH values: pH 4, pH 6, and pH 8, representing the most common pH values in natural water.

Figure S4 displays the ice nucleation spectra of PO, BA, CO, OA, BR, WR, WH, and SP at three different pHs, that is, at pH 4, 6, and 8. All the food materials show a similar trend of ice nucleation spectra in acidic and basic conditions, with significantly lower freezing temperatures than neutral pH (Figure S4). The  $T_{50}$  values of BA and PO at pH 6 were

**Table 1. Mean Freezing Temperature ( $T_{50}$ ) of Different Starchy Food Materials in the Presence of ZnO, Kaolin, Phenanthrene, and Micro/Nanoplastics as Cocontaminants and Cochemicals**

material	$T_{50}$	material	$T_{50}$	material	$T_{50}$	material	$T_{50}$
ZnO + BA	-6.9	kaolin + BA	-7.0	Phen + BA	-7.0	MNPs + BA	-10.0
ZnO + PO	-6.8	kaolin + PO	-6.0	Phen + PO	-6.7	MNPs + BA	-9.5
ZnO + OA	-14.3	kaolin + OA	-14.4	Phen + OA	-14.4	MNPs + BA	-15.8
ZnO + CO	-14.4	kaolin + CO	-14.4	Phen + CO	-14.5	MNPs + BA	-16.1
ZnO + WR	-14.0	kaolin + WR	-14.0	Phen + WR	-14.0	MNPs + BA	-15.1
ZnO + BR	-11.3	kaolin + BR	-10.9	Phen + BR	-9.9	MNPs + BA	-11.5
ZnO + WH	-14.5	kaolin + WH	-14.5	Phen + WH	-12.3	MNPs + BA	-16.2
ZnO + SP	-14.0	kaolin + SP	-14.1	Phen + SP	-14.3	MNPs + BA	-15.3

observed at  $-7.3$  and  $-7.2$  °C, whereas pH 4 shows  $T_{50}$  at  $-10.7$  and  $-9.5$  and pH 8 at  $-11.4$  and  $-8.6$ , respectively. Similarly, for CO, WR, BR, OA, WH, and SP at pH 4, the  $T_{50}$  values recorded are  $-16.8$ ,  $-16.3$ ,  $-14.3$ ,  $-17.7$ ,  $-15.6$ , and  $-16.7$  and at pH 8, the values are  $-16.2$ ,  $-16.4$ ,  $-15.1$ ,  $-17.5$ ,  $-15.4$ , and  $-16.7$ , respectively. The  $T_{50}$  values at pH 6 are  $-14.8$ ,  $-14.4$ ,  $-11.3$ ,  $-14.7$ ,  $-14.8$ , and  $-14.3$ , significantly higher than those of acidic or basic solutions. This observed decrease in ice nucleation efficiency under acidic and basic conditions can be attributed to several factors. Acidic conditions lead to the hydrolysis of starch molecules, breaking hydrogen bonds between starch molecules and increasing the number of free hydroxyl groups. This results in the generation of more hydrophilic functional groups that decrease the ice nucleation efficiency.<sup>45–47</sup> Furthermore, acid treatment of starch materials causes a reduction in the crystallinity of starch molecules, considering starch is a semicrystalline molecule that contains both amorphous and crystalline regions, and acid hydrolysis affects both, leading to further reduction of overall crystallinity.<sup>46</sup> This, in turn, contributes to the decrease in ice nucleation efficiency.

The acid treatment of starch corrodes the surfaces. This roughening occurs because crystalline regions are located at the surface of the starch particles, while amorphous regions are located within the particles. Both regions were acid-hydrolyzed to a particular extent to give rough surfaces.<sup>46</sup> In our case, we observed an increase in roughness and a decrease in the ice nucleation temperature.<sup>45,48,49</sup> The colloidal dispersions' hydrodynamic size before and after acid treatment was tested. Before acid treatment, the sizes of all these particles are below 150 nm (Figure 3C). We have observed that after acid treatment, the particles' size increases (Figure 3D). An increase in the size of the particles causes a decrease in the surface area, likely resulting in the diminution of INE. Figure 3F,G shows the SEM images of PO before and after acid treatment, respectively. After acid treatment, it was observed that surface roughness increased. Similar SEM images were obtained for BA up on acid treatment (Figure S5). As discussed earlier that the increase in surface roughness, in this case, is due to the etching of crystalline region present at the surface. As a result, the crystallinity of the PO decreases, and surface roughness increases, resulting in a decrease in INE (Figure S6). Crystallinity, surface roughness, and particle aggregation contribute to the alteration of INE. A similar effect was observed in the base hydrolysis of starch, which reduces the crystallinity and increases the hydrophilicity and gelatinization properties,<sup>50</sup> resulting a decrease the ice nucleation efficiency.

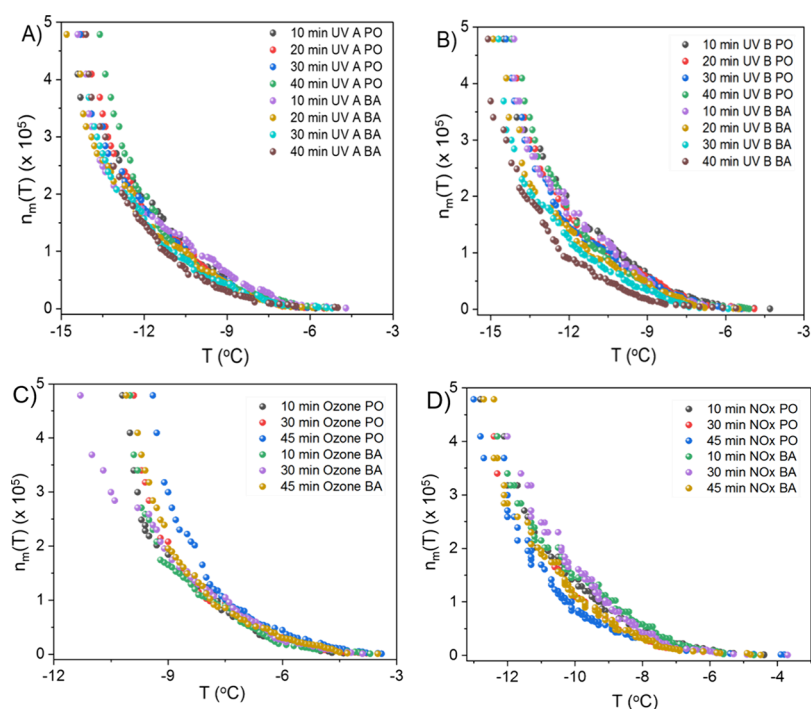
**Effect of Temperature.** Temporal variability is a significant physical factor affecting living beings.<sup>51</sup> Temperature often changes the materials' phase and composition, affecting the ice

nucleation ability. Our study investigated how temperature impacts the ice nucleation efficiency of all the starchy food materials (BA, PO, OA, BR, WR, WH, CO, and SP). We aged these materials at three different temperatures, that is, 25, 45, and 60 °C. The chosen temperatures approximatively depict the observed temperature around the globe.

Figure S7 shows the ice nucleation spectra of PO, BA, CO, OA, BR, WR, WH, and SP at three different temperatures, that is, at 25, 45, and 60 °C. It was observed that a gradual increase in temperature from 25 to 60 °C caused a decrease in the mean freezing temperature, suggesting that with an increase in temperature, the ice nucleation efficiency decreased (Figure S8). A similar trend was observed for all the starchy food materials. The  $T_{50}$  values of all the food materials at varied temperatures are shown in Figure S8. To quantify the heating impact on starchy food ice nucleation efficiency, the size analysis using the dynamic light scattering (DLS) method was performed, which suggested that heating the colloidal dispersions resulted in particle aggregation (Figure 3E). This observation was further supported by scanning electron microscopy (SEM) images, which displayed increased aggregation after heating compared to unheated samples (Figure 3H,I). Similar aggregation was noticed for BA samples on heating (Figure S9). The aggregation of particles reduces the active surface sites for ice nucleation, leading them to nucleate at cooler temperatures.

Many mechanistic pathways likely influence the ice nucleation efficiency of the starch molecules on temperature variations. First, heating can unwind the double-amylose helix and improve the bending degree of starch molecules, indicating that heating treatment is destructive to the crystalline region.<sup>52</sup> Second, starch molecules become more unstable with the increase in temperature, which is the key factor of heating-induced starch gelatinization, thereby reducing the crystallinity<sup>53</sup> and lowering the INE. Third, heating increases the intramolecular hydrogen bonding and decreases the starch–water hydrogen bonds.<sup>52</sup> The hydrogen bonds between heterogeneous nuclei and water enhance ice nucleation efficiency.<sup>54</sup> Reduction in the starch–water hydrogen bonds with the increase in temperature could be one of the reasons for the lowering of ice nucleation efficiency of the materials.

**Starch Mixing with Cocontaminants: Impact on Ice Nucleation Efficiency.** As starch is present in considerable quantities in the atmosphere, it can mix with other aerosols and affect the efficiency of ice nucleation. We have chosen four very common atmospheric particles that are also considered contaminants: (a) zinc oxide (ZnO) is an emerging metal contaminant; (b) kaolin is a clay material; (c) phenanthrene is a polycyclic aromatic hydrocarbon; (d) polyethylene-derived



**Figure 4.** Ice nucleation spectra of PO and BA at different exposure times upon exposure with (A) UV A, (B) UV B, (C) ozone, and (D) NO<sub>x</sub>.

micro/nanoplastics as emerging contaminants of global significance.

**Effect of ZnO.** ZnO is a commonly used nanomaterial in diverse applications such as drug delivery, cosmetics, sunscreen, sports, food additives, clothing, electronics, and other products.<sup>55</sup> Extensive use of ZnO causes toxicity and affects human life.<sup>56</sup> The effect of ZnO on the ice nucleation ability of all the starchy food materials (BA, PO, OA, CO, WR, BR, WH, and SP) was explored. Table 1 summarizes  $T_{50}$  values for pure starchy food materials and those mixed with ZnO. When these starchy food materials were mixed with ZnO, we observed changes in their ice nucleation properties, as reflected in their respective  $T_{50}$  values (Table 1). Notably, mixing ZnO resulted in slightly higher  $T_{50}$  values than pure samples.

Figure S10A shows their corresponding ice nucleation spectra. Ganguly and Ariya have previously shown that ZnO as a cocontaminant reduces the ice nucleation efficiency of inorganic materials.<sup>57</sup> In contrast, it was observed that the presence of ZnO increases the ice nucleation efficiency of all the starch-containing food materials. ZnO is a highly crystalline material, and adding ZnO into the starch solution (semi-crystalline) increases the number of crystallite molecules in the solution. As a result, active nucleation sites increase, which allows droplets to cool at warmer temperatures. Previous reports suggest that FeO increases the ice nucleation ability due to its low lattice mismatch with the hexagonal ice.<sup>58</sup> Similarly, ZnO could (wurtzite hexagonal structure) increase the ice nucleation activity in the solution due to its low lattice mismatch with the hexagonal ice.

**Effect of Kaolin.** Kaolin is one of the most widely abundant clay minerals. Kaolin was selected as a cocontaminant because of its wide availability and its presence in the atmosphere as clay-based dust particles. Airborne dust is a typical aerosol and significantly influences the ice nucleation processes.<sup>30,59</sup> When kaolin was mixed with starchy materials, we observed increased ice nucleation efficiency of all the starchy food materials (Table

1). Figure S10B presents the ice nucleation spectra of the kaolin-mixed samples. Adding kaolin increases the active sites for nucleation in the solution,<sup>29–31</sup> possibly the reason for the enhancement of the ice nucleation efficiency of all the starchy food materials.

**Effect of Phenanthrene.** Polycyclic aromatic compounds (PAHs) are a class of organic compounds composed of fused aromatic rings. They are generated from natural as well as anthropogenic activities such as bushfires, coal tar pitch, manufacturing of carbon black, vehicular emission, combustion of fossil fuels, asphalt, and so forth.<sup>60</sup> The United States Environmental Protection Agency (US EPA) prioritizes 16 PAHs in its list of photomutagenic chemicals, and phenanthrene is one of them.<sup>61</sup> When phenanthrene is mixed with starchy food materials, the ice nucleation efficiency is increased (Table 1). The ice nucleation spectra of phenanthrene mixed starchy food samples are shown in Figure S9C. The corresponding  $T_{50}$  values of BA, PO, CO, OA, WR, BR, WH, and SP are  $-7.0$ ,  $-6.7$ ,  $-14.4$ ,  $-14.5$ ,  $-14.0$ ,  $-9.9$ ,  $-12.3$ , and  $-14.3$  °C, respectively (Table 1). When phenanthrene was added to the starch solution, the number of active sites for nucleation and hydrophobicity of the solution increased. These factors enhance the ice nucleation efficiency of the various starchy food solutions.<sup>45,47,62</sup>

**Effect of Micro/Nanoplastics.** The development of synthetic plastics has brought revolution in a wide range of industries. The inexpensive and handy nature of plastics led to an exponential growth in their use and reached 335 million tonnes per year worldwide in 2016.<sup>63</sup> Extensive use of plastics caused the development of micro and nanoplastics, an emerging global contaminant. Nanoplastics are small enough to be ingested by marine organisms and may cross the biological barrier due to the disposal of nonbiodegradable plastics in nearby water bodies that lead to excessive intake of nanoplastics by marine organisms.<sup>64</sup> As a result of bioaccumulation and biomagnification, research has been

growing on the potential toxicity of micro and nanoplastics.<sup>65</sup> Micro and nanoplastics also affect the atmosphere and atmospheric surfaces by influencing the ice nucleation,<sup>57</sup> which is important for the global hydrological cycle.

The effect of micro/nanoplastics on the ice nucleation of starchy foods has been studied, and it is found that the addition of micro/nanoplastics led to a decrease in the ice nucleation efficiency of all the starchy foods (Table 1). The  $T_{50}$  values of BA, PO, CO, OA, WR, BR, WH, and SP are  $-10.0$ ,  $-9.5$ ,  $-15.8$ ,  $-16.1$ ,  $-15.1$ ,  $-11.5$ ,  $-16.2$ , and  $-15.3$  °C, respectively. Corresponding ice nucleation spectra are presented in Figure S10D. Control studies were performed to check the ice nucleation of THF solvent with milli-Q water required to disperse the MNPs (Figure S11). Ice nucleation spectrum shows a  $T_{50}$  value around  $-20.8 \pm 0.3$  °C, indicating no interference due to THF in the ice nucleation studies when MNPs (dispersed in THF) were mixed with the starchy food solutions (dispersed in H<sub>2</sub>O). The low crystallinity of nano/microplastics<sup>57</sup> and less availability of active sites for nucleation due to the hindrance from nano/microplastics could be the reason for the lowering in ice nucleation efficiency of starchy foods.

#### Effect of Light Exposure and Key Gaseous Species.

**Effect of Exposure to UV Light.** Several studies have shown that selected organic and inorganic molecules undergo chemical transformations upon UV exposure. Ganguly et al. have demonstrated that the exposure of UV rays to the FeHg increases the ice nucleation efficiency.<sup>66</sup> Besides, upon exposure to UV light, biological molecules cause a change in ice nucleation due to the alteration in the protein structure of physical–biological entities. The introduction section has mentioned previously several ways starch molecules may end up in the atmosphere. Being an organic molecule, it can undergo structural changes on exposure to UV radiation, thereby affecting the atmospheric ice nucleation.

To study the effect of UV radiation on starchy foods, BA and PO were irradiated with UV A (315–400 nm), UV B (285–315 nm), and visible (400–800 nm) light. BA and PO were chosen for these studies due to their high INE over other starchy food materials. All the samples were irradiated with UV A, UV B, and visible light for 10, 20, 30, and 40 min to check any observable change in the ice nucleation properties (Figure 4A,B). Table S2 shows the ice nucleation of BA and PO were greatly influenced by the exposure to UV rays, resulting in a significant change in their  $T_{50}$  values. When the starchy foods were exposed to UV irradiation, it was observed that the  $T_{50}$  values were shifted toward cooler temperatures (Table S2). The DLS analysis followed for the starchy foods before UV treatment shows a size  $<150$  nm for all the food materials (Figure 3C).

After UV treatment, the particles produced a broad size distribution in 150–350 nm (Figure S12). As the size of the particles increases, there is a decrease in the surface area of the particles. The SEM image of the BA particles after UV treatment shows the light-induced aggregation that causes enhancement in the size (Figure S13). The decrease in the surface area of the particles after UV treatment reduces the active sites for nucleation via aging under UV due to surface modification. The ice nucleation spectra when samples were irradiated with visible light are displayed in Figure S14. Visible light shows features similar to UV light and decreases the INE with increased exposure time.

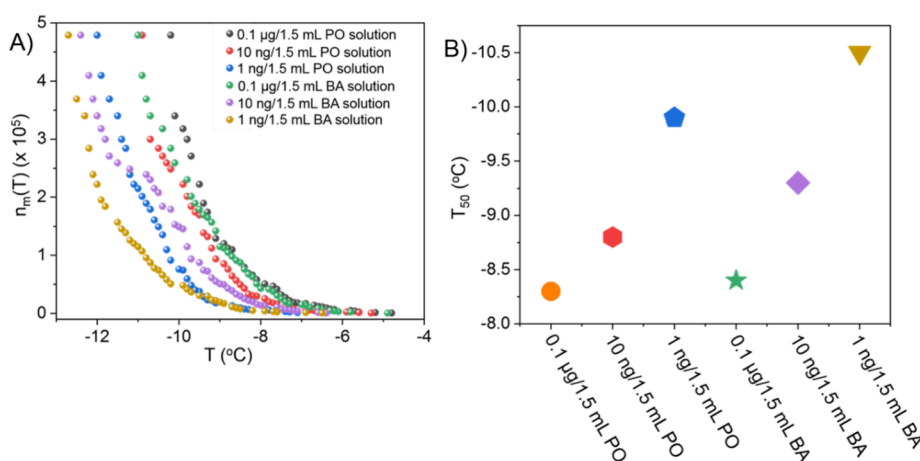
**Effect of Treatment of Ozone.** Ozone photochemical secondary processes produce several volatile and semivolatile organic compounds, which are emitted from natural and anthropogenic sources upon photochemical reactions.<sup>67</sup> Ozone is known to start a series of primary and secondary reactions from active radicals like OH and HO<sub>2</sub>, in the atmosphere.<sup>68</sup> Previous literature suggests that ozone exposure changes drastically ice nucleation properties of certain chemicals.<sup>69</sup>

O<sub>3</sub> was bubbled through a 10 mL sample solution for 10, 30, and 45 min in an open container, and its effect on the ice nucleation properties after O<sub>3</sub> exposure was observed (Figure 4C). It was found that the  $T_{50}$  values of BA and PO were reduced with the increase in exposure time (Table S3). The values of  $T_{50}$  for PO at 10, 30, and 45 min are  $-6.9$ ,  $-7.0$ , and  $-7.3$  °C and for BA are  $-7.1$ ,  $-7.2$ , and  $-7.2$  °C, respectively. The reduction in ice nucleation ability could be due to the surface oxidation of starch molecules in the presence of ozone. Longer exposure of O<sub>3</sub> (more than 45 min) to the sample solutions has practically no effect on the ice nucleation efficiency. As O<sub>3</sub> was produced from O<sub>2</sub>, there is a possibility that some amount of O<sub>2</sub> may also present along with O<sub>3</sub> during O<sub>3</sub> generation. To eliminate the effect of O<sub>2</sub>, it was bubbled through all the sample solutions for 30 min. We have not observed any significant change in the ice nucleation of the starchy sample solutions after O<sub>2</sub> bubbling, under our experimental conditions.

**Effect of NO<sub>x</sub> Exposure.** Acidic materials are known to change the surface of the particles resulting in the alteration of ice nucleation of the particles.<sup>70</sup> Anthropogenic activities that generate NO<sub>x</sub> frequently acidify the cloud.<sup>67</sup> To explore the effect of NO<sub>x</sub>, NO<sub>2</sub> was bubbled through the sample solutions in an open container at 5 kPa flow for about 15, 30, and 45 min. After NO<sub>x</sub> treatment, a decrease in the INE of the starchy sample solutions (BA and PO) with an increase in flow time was observed (Table S3). The corresponding ice nucleation spectra are shown in Figure 4D. After continuous NO<sub>2</sub> flow, the solutions become completely acidic. Lowering the pH means that solutions contain plenty of H<sup>+</sup> ions. We have already discussed in the previous section that in the presence of acid starch undergoes hydrolysis. Hydrophobicity, surface roughness, and reduction of crystallinity due to acid hydrolysis are the possible reasons for the reduction in the ice nucleation efficiency.

**Ice Nucleation Studies of Barley and Potato along with Their Skin.** BA and PO show a significantly higher ice nucleation temperature of all the starchy materials used for ice nucleation studies. The starch in these food materials was only used for the ice nucleation experiment during our experiments. However, we focused on something other than potato peel or barley bran, which is also essential food waste.<sup>71–73</sup> As PO and BA show significantly higher ice nucleation, we were interested in the combined effect of PO peel + PO starch and BA bran + BA starch on the ice nucleation ability. Freshly cut potato peel with potato starch was sonicated for 30 min to prepare the sample solution. Similarly, barley grain containing starch and bran was sonicated for 30 min. The difference in weight of barley gran before and after the sample preparation was calculated.

The presence of potato peel (powdered) and barley bran increases the  $T_{50}$  values of PO and BA significantly from  $-7.0$  and  $-7.2$  to  $-5.6$  and  $-6.5$  °C, respectively (Figure S15). Unlike PO (potato starch), the chemical composition of potato peel is entirely different from potato starch. Potato peel



**Figure 5.** (A) Ice nucleation plots for PO and BA three different dilutions and neutral pH. (B) Comparison of  $T_{50}$  values of PO and BA at three different concentrations.

contains proteins, lipids, dietary fiber, flavonoids, and phenolics.<sup>72</sup> Starch contents are less in the potato peel. Hence, some proteins, lipids, or other compounds in potato peel are responsible for incrementing the ice nucleation efficiency. However, we are still determining the exact material or combination of materials responsible for the enhancement. Similarly, barley bran also contains a large amount of proteins, lipids, ash, and  $\beta$ -glucan<sup>33</sup> along with starch that may be responsible for the shifting of ice nucleation toward higher temperatures.

In the previous section, it was observed that few cocontaminants increase the ice nucleation of the starchy materials. To check whether the presence of a cocontaminant can increase the ice nucleation temperature or not, (a) ZnO, (b) kaolin, (c) phenanthrene, and (d) nano/microplastics were added. The addition of ZnO, kaolin, and phenanthrene to both the potato peel (with PO) and barley bran (with BA) slightly shifts the ice nucleation toward warmer temperature, whereas micro/nanoplastics shift toward cooler temperature (Figure S16).

**Relevance to Environmental Conditions.** Starch-based aerosols are the most neglected particles that may have a lot of significance for animal and human health and the environment. Supplementary data files S2 and S3 show the United Nations Food and Agricultural Organization (UN FAO) data for potato and barley loss worldwide, respectively, during the period 2000–2022.<sup>74</sup> As the data suggest, a significant amount of barley and potato loss is observed worldwide during this period. This loss is attributed to the potato and barley cultivation, harvesting, and postharvest storage. These starchy waste materials can coat moisture, organic matter, dust, and microorganisms and transform aerosols in the atmosphere, which can act as nuclei under specific conditions.

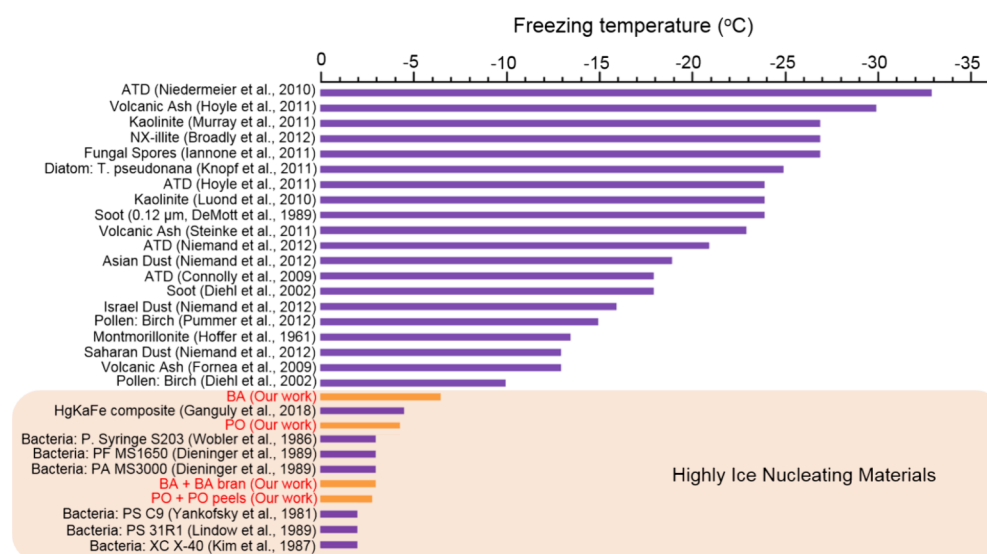
Our studies have shown that these particles significantly influence ice nucleation, which may seriously impact atmospheric clouds and precipitation, affecting the global hydrological cycle and climate change. It is noteworthy that in some instances, food waste is disposed of through burning in the fields, resulting in the release of smoke into the environment indirectly through agricultural biomass burning. This process introduces starchy particulate matter from food waste into the atmosphere, often categorized as “black/brown carbon”, which can significantly impact climatic conditions in various regions<sup>75</sup> and affect human and wildlife health.<sup>76–78</sup>

Day-to-day increase in biomass burning<sup>79,80</sup> is expected to increase the global temperature and will lead to a drastic change in the global hydrological cycle.<sup>81,82</sup> Smoke from burning vegetation not only helps in cloud formation by acting as a nucleating site for water droplets but also changing the clouds’ coverage and behavior.<sup>83</sup> Agricultural burning of vegetation changes the land surface characteristics that lead to a reduction of the convective cloud formation by limiting the vertical development of the boundary layer.<sup>84</sup> Additionally, aerosols associated with agricultural burning, such as those from biomass burning, can influence cloud behavior. More aerosols lead to smaller ice crystals in towering cumulus clouds and more water vapor entering the stratosphere. High concentrations of cloud condensation nuclei (CCN) from anthropogenic sources, including sugar cane burning, can increase cloud droplet number concentration (CDNC), potentially reducing precipitation efficiency.<sup>85</sup> Therefore, investigation of the ice nucleation mechanism of food waste materials is essential in understanding cloud microphysics.

Experiments performed in the laboratory-use instrumental techniques equipped with critical technology to understand the physicochemical processes. Even now, these do not have the sensitivity or resolution to analyze the sample of trace concentrations relevant to the environment. As a result, many studies of ice nucleation were performed at much higher concentrations than those present in the atmosphere. Although no reports exist on the concentrations of starch present in the atmosphere or its precarious effects on climate, we have used extremely low concentrations of starch for ice nucleation studies that could be of environmental relevance. For most of our studies, we have employed 100  $\mu\text{g}/1.5$  mL of the sample solution. For concentration-dependent studies, the above solution was further diluted to three different orders of magnitude (1000 $\times$ , 10,000 $\times$ , and 100,000 $\times$ ). BA and PO sample solutions were used for these experiments as they have higher ice nucleation efficiency. Figure 5A displays BA and PO concentration-dependent ice nucleation studies performed at neutral pH. The  $T_{50}$  reference value (milli-Q water) was  $-21.2 \pm 0.1$  °C (Figure S1).

The ice nucleation studies of BA and PO showed that the  $T_{50}$  values of BA or PO do not change much with dilution ( $\Delta T_{50} \leq 3$  °C). Dilution often changes properties such as coagulation, aggregation, and splitting that may affect the physicochemical processes such as the freezing temperature.





**Figure 6.** Comparison of ice nucleating efficiency of some heterogeneous ice nuclei with our materials.

Yet, a slight change in  $T_{50}$  upon a wide range of dilution (Figure 5B) suggests that the starch present in BA or PO has more or less the mean freezing temperature around  $-7.0$ , and they can act as catalysts for ice nucleation in environmental matrices.

**Comparison of Ice Nucleation Efficiency of Different Heterogeneous Ice Nuclei.** A comparison of the freezing temperatures of our starchy materials along with other materials such as soot,<sup>86,87</sup> mineral dust,<sup>88–95</sup> volcanic ash,<sup>95–97</sup> pollen grains,<sup>98,99</sup> bacteria,<sup>100–104</sup> fungal spores,<sup>105</sup> algae,<sup>106</sup> and inorganic composite materials<sup>66</sup> was done, which are widely known to affect the ice nucleation properties (Figure 6).

Our comparison is based on the values reported in the literature. The freezing temperature used for comparison corresponds to the warmest temperature at which the droplet freezing occurs. Soot particles, a major ice nucleating particle, can contribute to cirrus cloud formation that has an important influence on the climate. DeMott<sup>86</sup> measured the ice nucleation ability of soot produced using oxygen deficient acetylene burner and showed that ice freezes below  $-24$  °C (freezing range  $-24$  to  $-34$  °C). Diehl and Mitra<sup>87</sup> used aviation kerosene to generate soot and tested the freezing temperature of the droplets that came out as high as  $-18$  °C (freezing range  $-18$  to  $-28$  °C).

Mineral dust (kaolinite, illite, montmorillonite, etc.,) is also known to be an important source of ice nucleation in the atmosphere. Murray et al.<sup>88</sup> showed kaolinite freezes at a much lower temperature at  $-27$  °C (freezing range  $-27$  to  $-38$  °C) whereas Broadly et al.<sup>91</sup> and Hoffer<sup>90</sup> reported a freezing temperature of  $-27$  °C (freezing range  $-27$  to  $-38$  °C) and  $-13.5$  °C (freezing range  $-13.5$  to  $-35.5$  °C) for NX-illite and montmorillonite, respectively. Niemand et al.<sup>94</sup> also measured the ice nucleation capacities of a variety of dust particles including Arizona test dust (ATD), Asian dust, Sharan dust, and Israel dust most of which freezes at lower temperatures, that is, at  $-18$ ,  $-19$ ,  $-13$ , and  $-16$  °C, respectively. Several reports also exist on the ice nucleation properties of volcanic ash<sup>95,96</sup> and pollens<sup>98,99</sup> but their ice nucleation temperature is low and not considered as effective ice nucleating agents.

To date, only microorganisms such as bacteria (*P. syringae*) are reported to function as high-temperature ice nucleating particles and thereby effectively contribute to cloud seeding and trigger glaciation and precipitation. *P. syringae* shows ice nucleation at  $-2$  °C.<sup>27,100,102</sup> We have shown that starchy foods, especially, PO and BA, show ice nucleation at a much warmer temperature around  $-4.3$  and  $-6.5$  °C, whereas PO + PO peels and BA + BA bran show at  $-2.8$  and  $-3.0$  °C, respectively. These values are close to that reported for bacteria and the second-best ice nucleating species reported so far. Although these starch-based aerosols end up in the environment through pollens and agricultural waste and have the potential to seed cloud at much warmer temperatures, they are often neglected owing to their biodegradable properties. More studies and understanding are required in this area to know its potential impact on cloud condensation and climate change.

The ice nucleation ability of various starch-containing food materials relevant to the atmosphere and atmospheric processes was investigated. Among tested starchy foods, PO and BA, PO peels, and BA bran were highly efficient ice nuclei. The responses of atmospherically significant physicochemical parameters and common contaminants on the highly efficient ice nucleating particles were established. Alterations in size, shape, surface area, hydrophobicity, and crystallinity cause the change in the INE. We have investigated the INE of these materials at trace concentrations with environmental relevance and found that nucleation is not much affected by dilution. It suggests that they act as catalysts for ice nucleation in environmental matrices. Some starchy food materials studied in this work are very close to the INE of the most efficient biological particles reported. Further, theoretical, experimental, and modeling research is required to provide more insight into the effect of these aerosols on climate change.

## ■ ASSOCIATED CONTENT

### Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acsfoodscitech.3c00561>.

Additional information includes a detailed description of instrumentation, spectroscopic data, and other results (PDF)

United Nations Food and Agricultural Organization (UN FAO) data for potato (XLSX)

United Nations Food and Agricultural Organization (UN FAO) data for barley (XLSX)

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### Author Contributions

S.B. has performed most of the experiments, analysis, and drafting of the first version of the manuscript. D.P. helped in SAXS and XRD analysis, revising the manuscript, and also in the experimental setup related to ozone and NO<sub>x</sub> experiments. P.A.A., the principal investigator, proposed the project, supervised all coauthors, and completely revised the manuscript.

### Notes

The authors declare no competing financial interest.

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## REFERENCES

- (1) Knopf, D. A.; Alpert, P. A.; Wang, B. The Role of Organic Aerosol in Atmospheric Ice Nucleation: A Review. *ACS Earth Sp. Chem.* **2018**, *2* (3), 168–202.
- (2) Lau, K. M.; Wu, H. T. Warm Rain Processes over Tropical Oceans and Climate Implications. *Geophys. Res. Lett.* **2003**, *30* (24), CLM7-1.
- (3) Bartels-Rausch, T. Things We Need to Know about Ice and Snow. *Nature* **2013**, *494* (7435), 27–29.
- (4) Mazur, P. Cryobiology: The Freezing of Biological Systems. *Science* (80-) **1970**, *168* (3934), 939–949.
- (5) McNeill, V. F.; Grannas, A. M.; Abbott, J. P. D.; Ammann, M.; Ariya, P.; Bartels-Rausch, T.; Domine, F.; Donaldson, D. J.; Guzman,

M. I.; Heger, D.; Kahan, T. F.; Klán, P.; Masclin, S.; Toubin, C.; Voisin, D. Organics in Environmental Ices: Sources, Chemistry, and Impacts. *Atmos. Chem. Phys.* **2012**, *12* (20), 9653–9678.

(6) Harrison, R. G. Cloud Formation and the Possible Significance of Charge for Atmospheric Condensation and Ice Nuclei. *Space Sci. Rev.* **2000**, *94* (1), 381–396.

(7) Vali, G.; DeMott, P. J.; Möhler, O.; Whale, T. F. Technical Note: A Proposal for Ice Nucleation Terminology. *Atmos. Chem. Phys.* **2015**, *15* (18), 10263–10270.

(8) Baker, H. G.; Baker, I. Starch in Angiosperm Pollen Grains and Its Evolutionary Significance. *Am. J. Bot.* **1979**, *66* (5), 591–600.

(9) Laurence, A. R.; Thoms, A. V.; Bryant, V. M.; McDonough, C. Airborne Starch Granules as a Potential Contamination Source at Archaeological Sites. *J. Ethnobiol.* **2011**, *31* (2), 213–232.

(10) Newson, R.; Strachan, D.; Archibald, E.; Emberlin, J.; Hardaker, P.; Collier, C. Effect of Thunderstorms and Airborne Grass Pollen on the Incidence of Acute Asthma in England, 1990–94. *Thorax* **1997**, *52* (8), 680–685.

(11) Harun, N.-S.; Lachapelle, P.; Douglass, J. Thunderstorm-Triggered Asthma: What We Know so Far. *J. Asthma Allergy* **2019**, *12*, 101–108.

(12) McIlouth, M. E.; Paulus, H. J. Air Pollution from the Grain Industry. *J. Air Pollut. Control Assoc.* **1961**, *11* (7), 313–317.

(13) Stobnicka, A.; Górny, R. L. Exposure to Flour Dust in the Occupational Environment. *Int. J. Occup. Saf. Ergon.* **2015**, *21* (3), 241–249.

(14) Šikoparija, B.; Matavulj, P.; Mimić, G.; Smith, M.; Grewling, Ł.; Podražčanin, Z. Real-Time Automatic Detection of Starch Particles in Ambient Air. *Agric. For. Meteorol.* **2022**, *323*, No. 109034.

(15) Boac, J. M.; Maghirang, R. G.; Casada, M. E.; Wilson, J. D.; Jung, Y. S. Size Distribution and Rate of Dust Generated During Grain Elevator Handling. *Appl. Eng. Agric.* **2009**, *25* (4), 533–542.

(16) Efig. AP-42 9.9.1 Grain Elevators And Processes.

(17) Grunewald, J.; Eklund, A.; Katchar, K.; Moshfegh, A.; Lidén, C.; Lundgren, L.; Skare, L.; Tornling, G. Lung Accumulations of Eosinophil Granulocytes after Exposure to Cornstarch Glove Powder. *Eur. Respir. J.* **2003**, *21* (4), 646–651.

(18) Skjold, T.; Dahl, R.; Juhl, B.; Sigsgaard, T. The Incidence of Respiratory Symptoms and Sensitisation in Baker Apprentices. *Eur. Respir. J.* **2008**, *32* (2), 452–459.

(19) Page, E. H.; Dowell, C. H.; Mueller, C. A.; Biagini, R. E.; Heederik, D. Exposure to Flour Dust and Sensitization among Bakery Employees. *Am. J. Ind. Med.* **2010**, *53* (12), 1225–1232.

(20) FAO. *The State of Food and Agriculture. Moving Forward on Food Loss and Reduction*; Food and Agricultural Organization (FAO): Rome, Italy, 2019; pp 1–182. <https://www.fao.org/3/ca6030en/ca6030en.pdf>.

(21) Balasubramanian, S.; Domingo, N. G. G.; Hunt, N. D.; Gittlin, M.; Colgan, K. K.; Marshall, J. D.; Robinson, A. L.; Azevedo, I. M. L.; Thakrar, S. K.; Clark, M. A.; Tessum, C. W.; Adams, P. J.; Pandis, S. N.; Hill, J. D. The Food We Eat, the Air We Breathe: A Review of the Fine Particulate Matter-Induced Air Quality Health Impacts of the Global Food System. *Environ. Res. Lett.* **2021**, *16* (10), 103004.

(22) Domingo, N. G. G.; Balasubramanian, S.; Thakrar, S. K.; Clark, M. A.; Adams, P. J.; Marshall, J. D.; Muller, N. Z.; Pandis, S. N.; Polasky, S.; Robinson, A. L.; Tessum, C. W.; Tilman, D.; Tschofen, P.; Hill, J. D. Air Quality-Related Health Damages of Food. *Proc. Natl. Acad. Sci. U. S. A.* **2021**, *118* (20), No. e2013637118.

(23) Wang, X. Managing Land Carrying Capacity: Key to Achieving Sustainable Production Systems for Food Security. *Land* **2022**, *11*, 484.

(24) Ciuraru, R.; Kammer, J.; Decuq, C.; Vojkovic, M.; Haider, K.; Carpentier, Y.; Lafouge, F.; Berger, C.; Bourdat-Deschamps, M.; Ortega, I. K.; Levavasseur, F.; Houot, S.; Loubet, B.; Petitprez, D.; Focsa, C. New Particle Formation from Agricultural Recycling of Organic Waste Products. *npj Clim. Atmos. Sci.* **2021**, *4* (1), 5.

(25) Zihan, S.; Lu, L.; Tao, W.; Bolin, Z.; Hongfei, Z. Starch Nanoparticles as a New Ice Crystal Nucleator in Lactobacillus

- Bulgarius CICC 6097 Cryoprotection. *Int. J. Biol. Macromol.* **2023**, 251, No. 126395.
- (26) Chang, M.-W.; Lee, C.-R.; Hung, H.-F.; Teng, K.-S.; Huang, H.; Chuang, C.-Y. Bioaerosols from a Food Waste Composting Plant Affect Human Airway Epithelial Cell Remodeling Genes. *Int. J. Environ. Res. Public Health* **2014**, 11 (1), 337–354.
- (27) Mortazavi, R.; Hayes, C. T.; Ariya, P. A. Ice Nucleation Activity of Bacteria Isolated from Snow Compared with Organic and Inorganic Substrates. *Environ. Chem.* **2008**, 5 (6), 373–381.
- (28) Archuleta, C. M.; DeMott, P. J.; Kreidenweis, S. M. Ice Nucleation by Surrogates for Atmospheric Mineral Dust and Mineral Dust/Sulfate Particles at Cirrus Temperatures. *Atmos. Chem. Phys.* **2005**, 5 (10), 2617–2634.
- (29) Zolles, T.; Burkart, J.; Häusler, T.; Pummer, B.; Hitzemberger, R.; Grothe, H. Identification of Ice Nucleation Active Sites on Feldspar Dust Particles. *J. Phys. Chem. A* **2015**, 119 (11), 2692–2700.
- (30) Welti, A.; Kanji, Z. A.; Lüönd, F.; Stetzer, O.; Lohmann, U. Exploring the Mechanisms of Ice Nucleation on Kaolinite: From Deposition Nucleation to Condensation Freezing. *J. Atmos. Sci.* **2014**, 71 (1), 16–36.
- (31) Salam, A.; Lesins, G.; Lohmann, U. Laboratory Study of Heterogeneous Ice Nucleation in Deposition Mode of Montmorillonite Mineral Dust Particles Aged with Ammonia, Sulfur Dioxide, and Ozone at Polluted Atmospheric Concentrations. *Air Qual. Atmos. Heal.* **2008**, 1 (3), 135–142.
- (32) Belosi, F.; Piazza, M.; Nicosia, A.; Santachiara, G. Influence of Supersaturation on the Concentration of Ice Nucleating Particles. *Tellus B* **2018**, 70 (1), 1–10.
- (33) Sibakov, J.; Lehtinen, P.; Poutanen, K. Cereal Brans as Dietary Fibre Ingredients. In *Fibre-Rich and Wholegrain Foods* Woodhead Publishing Series in Food Science, Technology and Nutrition; Delcour, J. A.; Poutanen, K. Eds.; Elsevier, 2013; pp 170–192. DOI: 10.1533/9780857095787.2.170.
- (34) Pawar, R.; Jadhav, W.; Bhusare, S.; Borade, R.; Farber, S.; Itzkowitz, D.; Domb, A. Polysaccharides as Carriers of Bioactive Agents for Medical Applications. In *Natural-Based Polymers for Biomedical Applications*; 2008; pp 3–53. DOI: 10.1533/9781845694814.1.3.
- (35) Radlinski, A. P.; Mastalerz, M.; Hinde, A. L.; Hainbuchner, M.; Rauch, H.; Baron, M.; Lin, J. S.; Fan, L.; Thiagarajan, P. Application of SAXS and SANS in Evaluation of Porosity, Pore Size Distribution and Surface Area of Coal. *Int. J. Coal Geol.* **2004**, 59 (3), 245–271.
- (36) Grishaev, A. Sample Preparation, Data Collection, and Preliminary Data Analysis in Biomolecular Solution X-Ray Scattering. *Curr. Protoc. Protein Sci.* **2012**, 70, 17.14.1–17.14.18.
- (37) Hamley, I. W. Applications and Specifics of SAXS. *Small-Angle Scattering* **2021**, 137–195.
- (38) Willhammar, T.; Daicho, K.; Johnstone, D. N.; Kobayashi, K.; Liu, Y.; Midgley, P. A.; Bergström, L.; Saito, T. Local Crystallinity in Twisted Cellulose Nanofibers. *ACS Nano* **2021**, 15 (2), 2730–2737.
- (39) Kaur, J.; Ganguly, M.; Rangel-Alvarado, R.; Pal, D.; Hall, R.; Ariya, P. A. Ice Nucleation of Pharmaceutical and Synthetic Organic Emerging Contaminants: The Impact of Selected Environmental Conditions. *ACS Earth Sp. Chem.* **2022**, 6 (9), 2236–2249.
- (40) Auer, S.; Frenkel, D. Quantitative Prediction of Crystal-Nucleation Rates for Spherical Colloids: A Computational Approach. *Annu. Rev. Phys. Chem.* **2004**, 55, 333–361.
- (41) Fasolo, M.; Sollich, P. Equilibrium Phase Behavior of Polydisperse Hard Spheres. *Phys. Rev. Lett.* **2003**, 91 (6), 68301.
- (42) Cai, T.; Lin, H.; Liu, Z.; Chen, K.; Lin, Y.; Xi, Y.; Chhuond, K. Starch Wastewater Treatment Technology. *IOP Conf. Ser. Earth Environ. Sci.* **2019**, 358 (2), No. 022054.
- (43) Johnson, D. B.; Aguilera, A. The Microbiology of Extremely Acidic Environments. *Man. Environ. Microbiol.* **2016**, 4–3.
- (44) Grant, W. D.; Jones, B. E. Alkaline Environments. In *Encyclopaedia of microbiology*; Academic Press: New York, NY, USA, 2000; Vol. 1, pp 126–133.
- (45) Häusler, T.; Gebhardt, P.; Iglesias, D.; Rameshan, C.; Marchesan, S.; Eder, D.; Grothe, H. Ice Nucleation Activity of Graphene and Graphene Oxides. *J. Phys. Chem. C* **2018**, 122 (15), 8182–8190.
- (46) Yu, H.; Fang, Q.; Cao, Y.; Liu, Z. Effect of HCl on Starch Structure and Properties of Starch-Based Wood Adhesives. *Bio-Resources* **2015**, 11 (1), 1721–1728.
- (47) Biggs, C. I.; Packer, C.; Hindmarsh, S.; Walker, M.; Wilson, N. R.; Rourke, J. P.; Gibson, M. I. Impact of Sequential Surface-Modification of Graphene Oxide on Ice Nucleation. *Phys. Chem. Chem. Phys.* **2017**, 19 (33), 21929–21932.
- (48) Lupi, L.; Molinero, V. Does Hydrophilicity of Carbon Particles Improve Their Ice Nucleation Ability? *J. Phys. Chem. A* **2014**, 118 (35), 7330–7337.
- (49) Lupi, L.; Hudait, A.; Molinero, V. Heterogeneous Nucleation of Ice on Carbon Surfaces. *J. Am. Chem. Soc.* **2014**, 136 (8), 3156–3164.
- (50) Qin, Y.; Zhang, H.; Dai, Y.; Hou, H.; Dong, H. Effect of Alkali Treatment on Structure and Properties of High Amylose Corn Starch Film. *Materials* **2019**, 12 (10), 1705.
- (51) Reynolds, W. W.; Casterlin, M. E. The Role of Temperature in the Environmental Physiology of Fishes BT - *Environmental Physiology of Fishes*; Ali, M. A., Ed.; Springer US: Boston, MA, 1980; pp 497–518. DOI: 10.1007/978-1-4899-3659-2\_19.
- (52) Zhiguang, C.; Junrong, H.; Huayin, P.; Keipper, W. The Effects of Temperature on Starch Molecular Conformation and Hydrogen Bonding. *Starch/Stärke* **2022**, 74 (7–8), 1–9.
- (53) Liu, Q.; Donner, E.; Tarn, R.; Singh, J.; Chung, H.-J. Chapter 8 - *Advanced Analytical Techniques to Evaluate the Quality of Potato and Potato Starch*; Singh, J., Kaur, L. B. T.-A. in P. C. and T., Eds.; Academic Press: San Diego, 2009; pp 221–248. DOI: 10.1016/B978-0-12-374349-7.00008-8.
- (54) Zheng, Y.; Su, C.; Lu, J.; Loh, K. P. Room-Temperature Ice Growth on Graphite Seeded by Nano-Graphene Oxide. *Angew. Chem., Int. Ed. Engl.* **2013**, 52 (33), 8708–8712.
- (55) Li, Y.; Li, F.; Zhang, L.; Zhang, C.; Peng, H.; Lan, F.; Peng, S.; Liu, C.; Guo, J. Zinc Oxide Nanoparticles Induce Mitochondrial Biogenesis Impairment and Cardiac Dysfunction in Human iPSC-Derived Cardiomyocytes. *Int. J. Nanomedicine* **2020**, 15, 2669–2683.
- (56) Shen, Z.; Chen, Z.; Hou, Z.; Li, T.; Lu, X. Ecotoxicological Effect of Zinc Oxide Nanoparticles on Soil Microorganisms. *Front. Environ. Sci. Eng.* **2015**, 9 (5), 912–918.
- (57) Ganguly, M.; Ariya, P. A. Ice Nucleation of Model Nanoplastics and Microplastics: A Novel Synthetic Protocol and the Influence of Particle Capping at Diverse Atmospheric Environments. *ACS Earth Sp. Chem.* **2019**, 3 (9), 1729–1739.
- (58) Chong, E.; Marak, K. E.; Li, Y.; Freedman, M. A. Ice Nucleation Activity of Iron Oxides via Immersion Freezing and an Examination of the High Ice Nucleation Activity of FeO. *Phys. Chem. Chem. Phys.* **2021**, 23 (5), 3565–3573.
- (59) Wex, H.; Demott, P. J.; Tobo, Y.; Hartmann, S.; Rösch, M.; Clauss, T.; Tomsche, L.; Niedermeier, D.; Stratmann, F. Kaolinite Particles as Ice Nuclei: Learning from the Use of Different Kaolinite Samples and Different Coatings. *Atmos. Chem. Phys.* **2014**, 14 (11), 5529–5546.
- (60) Eeshwarasinghe, D.; Loganathan, P.; Kalaruban, M.; Sounthararajah, D. P.; Kandasamy, J.; Vigneswaran, S. Removing Polycyclic Aromatic Hydrocarbons from Water Using Granular Activated Carbon: Kinetic and Equilibrium Adsorption Studies. *Environ. Sci. Pollut. Res.* **2018**, 25 (14), 13511–13524.
- (61) Yan, J.; Wang, L.; Fu, P. P.; Yu, H. Photomutagenicity of 16 Polycyclic Aromatic Hydrocarbons from the US EPA Priority Pollutant List. *Mutat. Res.* **2004**, 557 (1), 99–108.
- (62) Brooks, S. D.; Suter, K.; Olivarez, L. Effects of Chemical Aging on the Ice Nucleation Activity of Soot and Polycyclic Aromatic Hydrocarbon Aerosols. *J. Phys. Chem. A* **2014**, 118 (43), 10036–10047.
- (63) Taniguchi, I.; Yoshida, S.; Hiraga, K.; Miyamoto, K.; Kimura, Y.; Oda, K. Biodegradation of PET: Current Status and Application Aspects. *ACS Catal.* **2019**, 9 (5), 4089–4105.
- (64) Ng, E.-L.; Huerta Lwanga, E.; Eldridge, S. M.; Johnston, P.; Hu, H.-W.; Geissen, V.; Chen, D. An Overview of Microplastic and

- Nanoplastic Pollution in Agroecosystems. *Sci. Total Environ.* **2018**, *627*, 1377–1388.
- (65) Revel, M.; Châtel, A.; Mouneyrac, C. Micro(Nano)Plastics: A Threat to Human Health? *Curr. Opin. Environ. Sci. Heal.* **2018**, *1*, 17–23.
- (66) Ganguly, M.; Dib, S.; Kurien, U.; Rangel-Alvarado, R. B.; Miyahara, Y.; Ariya, P. A. Influence of Environmentally Relevant Physicochemical Conditions on a Highly Efficient Inorganic Ice Nucleating Particle. *J. Phys. Chem. C* **2018**, *122* (32), 18690–18704.
- (67) Attard, E.; Yang, H.; Delort, A.-M.; Amato, P.; Pöschl, U.; Glaux, C.; Koop, T.; Morris, C. E. Effects of Atmospheric Conditions on Ice Nucleation Activity of *Pseudomonas*. *Atmos. Chem. Phys.* **2012**, *12* (22), 10667–10677.
- (68) Ryzhkov, A. B.; Ariya, P. A. A Theoretical Study of the Reactions of Carbonyl Oxide with Water in Atmosphere: The Role of Water Dimer. *Chem. Phys. Lett.* **2003**, *367* (3), 423–429.
- (69) Kanji, Z. A.; Welti, A.; Chou, C.; Stetzer, O.; Lohmann, U. Laboratory Studies of Immersion and Deposition Mode Ice Nucleation of Ozone Aged Mineral Dust Particles. *Atmos. Chem. Phys.* **2013**, *13* (17), 9097–9118.
- (70) Augustin-Bauditz, S.; Wex, H.; Kanter, S.; Ebert, M.; Niedermeier, D.; Stolz, F.; Prager, A.; Stratmann, F. The Immersion Mode Ice Nucleation Behavior of Mineral Dusts: A Comparison of Different Pure and Surface Modified Dusts. *Geophys. Res. Lett.* **2014**, *41* (20), 7375–7382.
- (71) Wu, D. Recycle Technology for Potato Peel Waste Processing: A Review. *Procedia Environ. Sci.* **2016**, *31*, 103–107.
- (72) Javed, A.; Ahmad, A.; Tahir, A.; Shabbir, U.; Nouman, M.; Hameed, A. Potato Peel Waste—Its Nutraceutical, Industrial and Biotechnological Applications. *AIMS Agric. Food* **2019**, *4* (3), 807–823.
- (73) Gebrechistos, H. Y.; Chen, W. Utilization of Potato Peel as Eco-Friendly Products: A Review. *Food Sci. Nutr.* **2018**, *6* (6), 1352–1356.
- (74) *Food loss and waste database: Food and Agriculture Organization of United Nations*. <https://www.fao.org/platform-food-loss-waste/flw-data/en/> 2011.
- (75) Bond, T. C.; Doherty, S. J.; Fahey, D. W.; Forster, P. M.; Bernsten, T.; DeAngelo, B. J.; Flanner, M. G.; Ghan, S.; Kärcher, B.; Koch, D.; Kinne, S.; Kondo, Y.; Quinn, P. K.; Sarofim, M. C.; Schultz, M. G.; Schulz, M.; Venkataraman, C.; Zhang, H.; Zhang, S.; Bellouin, N.; Guttikunda, S. K.; Hopke, P. K.; Jacobson, M. Z.; Kaiser, J. W.; Klimont, Z.; Lohmann, U.; Schwarz, J. P.; Shindell, D.; Storelvmo, T.; Warren, S. G.; Zender, C. S. Bounding the Role of Black Carbon in the Climate System: A Scientific Assessment. *J. Geophys. Res. Atmos.* **2013**, *118* (11), 5380–5552.
- (76) Jaffe, D. A.; O'Neill, S. M.; Larkin, N. K.; Holder, A. L.; Peterson, D. L.; Halofsky, J. E.; Rappold, A. G. Wildfire and Prescribed Burning Impacts on Air Quality in the United States. *J. Air Waste Manage. Assoc.* **2020**, *70* (6), 583–615.
- (77) Pardo, M.; Li, C.; He, Q.; Levin-Zaidman, S.; Tsoory, M.; Yu, Q.; Wang, X.; Rudich, Y. Mechanisms of Lung Toxicity Induced by Biomass Burning Aerosols. *Part. Fibre Toxicol.* **2020**, *17* (1), 4.
- (78) Reid, C. E.; Brauer, M.; Johnston, F. H.; Jerrett, M.; Balmes, J. R.; Elliott, C. T. Critical Review of Health Impacts of Wildfire Smoke Exposure. *Environ. Health Perspect.* **2016**, *124* (9), 1334–1343.
- (79) Hanes, C. C.; Wang, X.; Jain, P.; Parisien, M.-A.; Little, J. M.; Flannigan, M. D. Fire-Regime Changes in Canada over the Last Half Century. *Can. J. For. Res.* **2019**, *49* (3), 256–269.
- (80) Jain, P.; Castellanos-Acuna, D.; Coogan, S. C. P.; Abatzoglou, J. T.; Flannigan, M. D. Observed Increases in Extreme Fire Weather Driven by Atmospheric Humidity and Temperature. *Nat. Clim. Chang.* **2022**, *12* (1), 63–70.
- (81) Jones, M. W.; Smith, A.; Betts, R.; Canadell, J. G.; Prentice, I. C.; Le Quéré, C. Climate Change Increases the Risk of Wildfires. *ScienceBrief UEA* 2020. <https://tyndall.ac.uk/wp-content/uploads/2021/09/TWP-164.pdf>.
- (82) Abatzoglou, J. T.; Williams, A. P. Impact of Anthropogenic Climate Change on Wildfire across Western US Forests. *Proc. Natl. Acad. Sci. U. S. A.* **2016**, *113* (42), 11770–11775.
- (83) Day, C. Smoke From Burning Vegetation Changes the Coverage and Behavior of Clouds. *Phys. Today* **2004**, *57* (5), 24–26.
- (84) Lyons, T. J. Clouds Prefer Native Vegetation. *Meteorol. Atmos. Phys.* **2002**, *80* (1), 131–140.
- (85) Sherwood, S. A Microphysical Connection Among Biomass Burning, Cumulus Clouds, and Stratospheric Moisture. *Science* (80-) **2002**, *295* (5558), 1272–1275.
- (86) DeMott, P. J. An Exploratory Study of Ice Nucleation by Soot Aerosols. *J. Appl. Meteorol. Climatol.* **1990**, *29* (10), 1072–1079.
- (87) Diehl, K.; Mitra, S. K. A Laboratory Study of the Effects of a Kerosene-Burner Exhaust on Ice Nucleation and the Evaporation Rate of Ice Crystals. *Atmos. Environ.* **1998**, *32* (18), 3145–3151.
- (88) Murray, B. J.; Broadley, S. L.; Wilson, T. W.; Atkinson, J. D.; Wills, R. H. Heterogeneous Freezing of Water Droplets Containing Kaolinite Particles. *Atmos. Chem. Phys.* **2011**, *11* (9), 4191–4207.
- (89) Lüönd, F.; Stetzer, O.; Welti, A.; Lohmann, U. Experimental Study on the Ice Nucleation Ability of Size-Selected Kaolinite Particles in the Immersion Mode. *J. Geophys. Res.* **2010**, *115* (D14), 1–14.
- (90) Hoffer, T. E. A Laboratory Investigation of Droplet Freezing. *J. Atmos. Sci.* **1961**, *18* (6), 766–778.
- (91) Broadley, S. L.; Murray, B. J.; Herbert, R. J.; Atkinson, J. D.; Dobbie, S.; Malkin, T. L.; Condliffe, E.; Neve, L. Immersion Mode Heterogeneous Ice Nucleation by an Illite Rich Powder Representative of Atmospheric Mineral Dust. *Atmos. Chem. Phys.* **2012**, *12* (1), 287–307.
- (92) Niedermeier, D.; Hartmann, S.; Shaw, R. A.; Covert, D.; Mentel, T. F.; Schneider, J.; Poulain, L.; Reitz, P.; Spindler, C.; Clauss, T.; Kiselev, A.; Hallbauer, E.; Wex, H.; Mildenberger, K.; Stratmann, F. Heterogeneous Freezing of Droplets with Immersed Mineral Dust Particles – Measurements and Parameterization. *Atmos. Chem. Phys.* **2010**, *10* (8), 3601–3614.
- (93) Connolly, P. J.; Möhler, O.; Field, P. R.; Saathoff, H.; Burgess, R.; Choulaton, T.; Gallagher, M. Studies of Heterogeneous Freezing by Three Different Desert Dust Samples. *Atmos. Chem. Phys.* **2009**, *9* (8), 2805–2824.
- (94) Niemand, M.; Möhler, O.; Vogel, B.; Vogel, H.; Hoose, C.; Connolly, P.; Klein, H.; Bingemer, H.; Demott, P.; Skrotzki, J.; Leisner, T. A Particle-Surface-Area-Based Parameterization of Immersion Freezing on Desert Dust Particles. *J. Atmos. Sci.* **2012**, *69* (10), 3077–3092.
- (95) Hoyle, C. R.; Pinti, V.; Welti, A.; Zobrist, B.; Marcolli, C.; Luo, B.; Höskuldsson, A.; Mattsson, H. B.; Stetzer, O.; Thorsteinsson, T.; Larsen, G.; Peter, T. Ice Nucleation Properties of Volcanic Ash from Eyjafjallajökull. *Atmos. Chem. Phys.* **2011**, *11* (18), 9911–9926.
- (96) Steinke, I.; Möhler, O.; Kiselev, A.; Niemand, M.; Saathoff, H.; Schnaier, M.; Skrotzki, J.; Hoose, C.; Leisner, T. Ice Nucleation Properties of Fine Ash Particles from the Eyjafjallajökull Eruption in April 2010. *Atmos. Chem. Phys.* **2011**, *11* (24), 12945–12958.
- (97) Fornea, A. P.; Brooks, S. D.; Dooley, J. B.; Saha, A. Heterogeneous Freezing of Ice on Atmospheric Aerosols Containing Ash, Soot, and Soil. *J. Geophys. Res. Atmos.* **2009**, *114* (D13), D13201.
- (98) Diehl, K.; Matthias-Maser, S.; Jaenicke, R.; Mitra, S. K. The Ice Nucleating Ability of Pollen: Part II. Laboratory Studies in Immersion and Contact Freezing Modes. *Atmos. Res.* **2002**, *61* (2), 125–133.
- (99) Pummer, B. G.; Bauer, H.; Bernardi, J.; Bleicher, S.; Grothe, H. Suspendable Macromolecules Are Responsible for Ice Nucleation Activity of Birch and Conifer Pollen. *Atmos. Chem. Phys.* **2012**, *12* (5), 2541–2550.
- (100) Lindow, S. E.; Lauhe, E.; Govindarajan, A. G.; Panopoulos, N. J. D. G. Localization of Ice Nucleation Activity and the IceC Gene Product in *Pseudomonas Syringae* and *Escherichia Coli*. *Mol. Plant-Microbe Interact.* **1989**, *2* (5), 262–272.
- (101) Wolber, P. K.; Deininger, C. A.; Southworth, M. W.; Vandekerckhove, J.; van Montagu, M.; Warren, G. J. Identification

and Purification of a Bacterial Ice-Nucleation Protein. *Proc. Natl. Acad. Sci. U. S. A.* **1986**, *83* (19), 7256–7260.

(102) Yankofsky, S. A.; Levin, Z.; Bertold, T.; Sandlerman, N. Some Basic Characteristics of Bacterial Freezing Nuclei. *J. Appl. Meteorol. Climatol.* **1981**, *20* (9), 1013–1019.

(103) Deininger, C. A.; Mueller, G. M.; Wolber, P. K. Immunological Characterization of Ice Nucleation Proteins from *Pseudomonas Syringae*, *Pseudomonas Fluorescens*, and *Erwinia Herbicola*. *J. Bacteriol.* **1988**, *170* (2), 669–675.

(104) Kim, H. K. *Xanthomonas Campestris* Pv. *Translucens* Strains Active in Ice Nucleation. *Plant Dis.* **1987**, *71* (11), 994–997.

(105) Iannone, R.; Chernoff, D. I.; Pringle, A.; Martin, S. T.; Bertram, A. K. The Ice Nucleation Ability of One of the Most Abundant Types of Fungal Spores Found in the Atmosphere. *Atmos. Chem. Phys.* **2011**, *11* (3), 1191–1201.

(106) Knopf, D. A.; Alpert, P. A.; Wang, B.; Aller, J. Y. Stimulation of Ice Nucleation by Marine Diatoms. *Nat. Geosci.* **2011**, *4* (2), 88–90.