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Nucleation of Supercooled Water by Neutrons: Latitude Dependence and Implications for Cloud Modelling

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Abstract

It has recently been shown that incident particles, neutrons, can initiate the freezing in a supercooled water volume. This new finding may have ramifications for the interpretation of both experimental data on the nucleation of laboratory samples of supercooled water and perhaps more importantly on the interpretation of ice nucleation involved in cloud physics. For example, if some fraction of the cloud nucleation previously attributed to dust, soot, or aerosols has been caused by cosmogenic neutrons, fresh consideration is required in the context of climate models. Moreover, as cosmogenic neutrons, most being muon-induced, have much greater flux at high latitudes, estimates of ice nucleates in these regions may be larger than required to accurately model cloud and condensation properties. This discrepancy has been pointed out in IPCC reports. Our paper discusses the connection between the new concept of neutrons nucleating supercooled water and the need for a new source of nucleation in high latitude clouds, ideally causing others to review current data, or to analyse future data with this idea in mind.

Keywords

Climate Models, Ice Nucleation, Neutrons, Supercooling

1. Introduction

The reports by Szydagis *et al.* [1] [2] that neutrons can cause the nucleation, and so the subsequent freezing, of supercooled water have widespread ramifications. Existing laboratory studies of deeply supercooled water could, in some cases, be re-evaluated and anomalous results perhaps be more easily explained. As an example, we consider the relatively large spread of values historically reported as the temperature threshold for homogeneous nucleation of pure water. These vary from warmer than −35˚C [3] to colder than −41˚C. The only way to reach these levels of deep supercooling is by having no active nucleation sites present, either in the water or at the containment walls. However, if neutrons are present in that laboratory, at that latitude and elevation, from cosmogenic, or other sources, they may be causing the nucleation in the water, making it appear as homogeneous nucleation, as they appear to nucleate both in the bulk and at the water's surface.

Cirrus clouds play a significant role in terms of regulating radiative fluxes at the top of the troposphere; however, we understand rather poorly the microphysics of heterogeneous freezing of water within clouds [4] [5]. Precipitation in mid-latitude clouds is predominantly initiated via the Wegener-Bergeron-Findeisen process, involving heterogeneous freezing of supercooled water droplets by ice-active aerosol particles [6].

We suggest now that this may, at least in part, be because of neutrons causing ice nucleation, when no known active nucleation sites are present, or are at least present in significantly lower numbers than required to explain the observed effects. Murray *et al.* [7] comment upon the poor understanding of ice nucleation in clouds, where, not only are the uncertainties surrounding the effects of aerosols large, but the effects of ice nucleation are not deeply understood. The Intergovernmental Panel on Climate Change (IPCC), in its 2014 report [8] was unable to estimate the *radiative forcing* of aerosols on clouds through ice nucleation, that report stating that "the atmospheric concentrations of ice nucleators (IN) are very uncertain because of the aforementioned uncertainties in freezing mechanisms and the difficulty of measuring IN in the upper troposphere." *Radiative forcing*, also known as climate forcing, refers to the change in energy flux in the atmosphere caused by natural or anthropogenic factors of climate change. This incomplete understanding of numbers of IN is well described by an extensive analysis of both experimental data and modelling from Curry and Khvorostyanov [9], who show clearly the lack of known INs required to explain observed effects in clouds.

The authors are very cognisant of the fact that what follows is not a review article, nor does it present any new data. What it does do, is tie together ideas from several disparate disciplines, ideas which may have significance to cloud microphysics and so to climate models going forward.

2. Stochastic Nature of Ice Nucleation

For any given volume of liquid water cooled below its melting point, there exists an inherent spread of nucleation temperatures due to the stochastic nature of the phase transition, even when one, efficient, nucleation site is present [10]. This spread of temperatures is found to be present in any heterogeneous nucleation process, *i.e.* added mineral, biological or physical processes, e.g. sonication, as well as for so-called homogeneous nucleation, when no obvious/known nucleators are present. Generally, in laboratory measurements of a given sample, spreads of a few ˚C are reported [11] but when the same sample is used and sufficient repeat measurements are made, or multiple (nearly) identical samples are used and multiple measurements are made, the spread of nucleation temperatures converges to approximately 0.7˚C or even lower [12] [13]. Spreads that are greater than this value are hypothesised to be due to multiple nucleation sites, too few samples for statistical validity or a nucleation site which is changing with time [14]. By spread, we mean the 10% - 90% width of a "survival curve" plot, or probability curve. Incoming particles causing nucleation could naturally lead to multiple sites, as demonstrated by Szdagis *et al.* [2].

3. Nucleation in Clouds

Ice nucleation in clouds also occurs via either a homogeneous freezing of liquid particles or heterogeneous freezing, triggered from INs which possess surface properties favourable to lowering the energy barrier to crystallisation [15]. The radiative properties of mixed-phase clouds are thought to be dominated by the supercooled liquid phase [16], and therefore understanding the ice formation process is essential for the modelling of climatic radiative impact. Better knowledge of the effects of IN is required for improving climate models and predicting precipitation such as rain, snow, and hail in weather forecast models. Currently, there is significant research activity because field measurements have shown that IN seem to be of natural origin but are somewhat poorly understood [16] [17] [18] [19].

If neutrons, or other radiation, can cause deeply supercooled water droplets in clouds to nucleate and freeze, there exists the potential for re-evaluation of existing data globally on nucleation when no obvious INs were present, or at least were thought to be present in insufficient numbers to explain the observed cloud freezing. It is known the cosmogenic neutron flux toward Earth is larger at the North Pole than at the equator [20] [21], although likely due to the neutrons being secondary particles created by muons and other charged particles, which would be more prevalent at the poles, since they are affected by the magnetic field [22] [23] [24]. However, a connection between atmospheric dynamics in the troposphere and the flux of high-energy cosmic rays has been sought for many years [25] [26]. Scenarios that have been proposed whereby cosmic rays affect the atmosphere often include some mechanism for enhanced ice nucleation within clouds [26], but contradictory results are present in the existent literature as to whether any type of particle or radiation can actually cause supercooled water to freeze, as we discuss next. It must be noted that there exist much less data for the South Pole and high Southern latitudes, although there appears to be no reason for a different flux there compared to Northern latitudes.

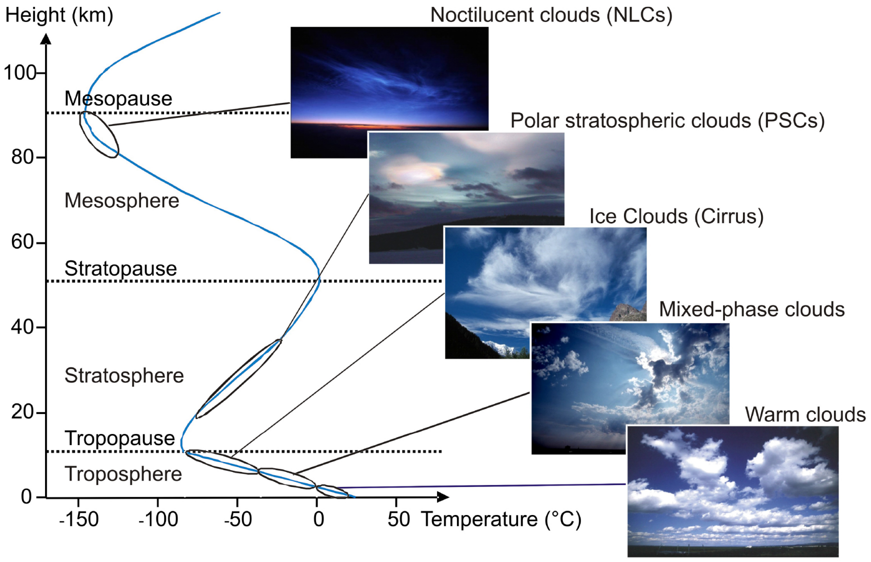

Figure 1. Ice clouds and ice-containing clouds are found from about 2 km altitude through 10 km and form a significant part of the climate-dependent system. (Figure courtesy of Karlsruhe Institute of Metereology and Climate Research).

The enhanced freezing of liquid cloud droplets by nucleation of ice crystals in the ionisation tracks produced by the flux of cosmic rays was suggested by Varshneya [27]. Varshneya [28] subsequently developed a theory whereby the energy barrier to ice nucleation is reduced via confinement of an ionisation charge to a forming ice nucleus, and showed data using gammas. However, more recently Detwiler and Vonnegut [29] tested Varshneya's results when they exposed 10 μm diameter cloud droplets at −15˚C (see **Figure 1**) to a source of 5.3 MeV alpha particles. They found that fewer than 1 in 2.5×10^6 collisions between alpha particles and cloud droplet atoms resulted in a nucleation event, suggesting that ionising radiation is ineffective as a direct source of enhanced ice nucleation in clouds above −15˚C. Seeley *et al.* [19] subsequently also concluded that ionisation tracks produced by 5 MeV alpha particles do not have an observable effect on the nucleation of ice from strongly supercooled liquid water. These two works however did not exclude the possibility that another type, or energy, or track range, of cosmic-ray particles could lead to nucleation. We are suggesting that neutrons are that source.

Gammas are also a potential source. It is worth noting that alpha particles do have relatively high energy but may be below the stopping power threshold in supercooled water. This would be the case if supercooled water acts as superheated water does, and any other superheated liquid, as in bubble-chamber based dark matter experiments such as COUPP, PICO, and PICASSO (see for example [30]). It is worth noting these experiments also have a high level of blindness to gamma-rays, suggesting neutrons are a more likely source of ice nucleation than gammas.

The latitude dependence, *i.e.* the flux at 90˚ latitude compared with equatorial flux, of neutrons has been measured to be between 2.5 and 8 times [21] [31]. The flux is a factor of 10 higher at 5 km altitude than the generally agreed-upon value of $1.0*10^{-5}$ $cm^{-2} \cdot s^{-1}$ measured at sea level [31] [32]. At sea level, cosmic rays are thought to consist of ~10% neutrons [30].

Since the 80s it is known that Terrestrial Cosmic Rays, mainly reported as Atmospheric Neutrons, can penetrate the natural shielding of buildings and equipment and produce errors in integrated circuits. The high-energy neutron fluxes range between 10 particles $cm^{-1} \cdot h^{-1}$ at sea level and some 10^3 particles $cm^{-1} \cdot h^{-1}$ at 10 km altitude [33]. Atmospheric neutrons are end-products of the interactions of cosmic rays with our atmosphere. The cosmic rays interact with air nuclei to generate particles comprising protons, neutrons, etc. The radiation intensity is maximum at 20 km and then drops off to sea level at which the neutron flux remains significant.

A brief look at latitude dependence on IN effects includes the work of Murray *et al.* [34], who report on the measured fraction of mid-level stratus clouds containing ice at four locations and they found striking differences. They described, for example, lower than expected INs at Cape Verde and at Morocco [35] [36]. We suspect that this finding is due to lower neutron counts near the equator, despite the area being rich in Saharan dust [37], although any true trend with latitude change is likely to be obscured by systematics stemming from markedly different experimental setups including different water volumes and amounts of (unintentional) shielding from radiation from the lab environment, space, or multiple sources. We are thus only speculating here and inviting others to re-evaluate their (existing) data sets, in light of our hypothesis.

At −33˚C, the reported nucleation rates [7] for water vary by up to six orders of magnitude, depending upon experimental arrangement. This variation is unable to be explained by stochasticity, nor the difference between volume and surface nucleation. Generally, theory and experimental data are not good fits, for cold temperatures [38] [39]. Recently, it was noted that there is considerable divergence in parameterisations of homogeneous nucleation, producing significant differences within cloud models [3]. Koop and Murray [3] discuss a three orders of magnitude difference in nucleation rate for temperatures reaching −36˚C and lower.

In the Northern hemisphere, at high latitude (51˚ N), almost all clouds are ice-filled, and so nucleation has occurred by −20˚C. Conversely, at low latitude (16˚N), even by −30˚C, only 70% of clouds are ice-filled [40].

Atmospheric dust volumes vary by factors of up to 10^3 between the equator and poles [36], and soot levels by 10^4, and, although lower in number, bacteria/spores vary by 10^4 [36]. Murray *et al.* [3] report a six order of magnitude variation for IN/cm^3 at −30˚C. For −20˚C to −40˚C concentrations have been estimated to range over three orders of magnitude and, significantly, the spread of measurements at −30˚C for experimental data taken at Colorado is 3× that taken at Alaska [41].

In **Figure 2**, we demonstrate some data adapted from Kanitz *et al.* [40] where, at high latitude (51 degrees N) almost all clouds are ice-filled, *i.e.* nucleation has

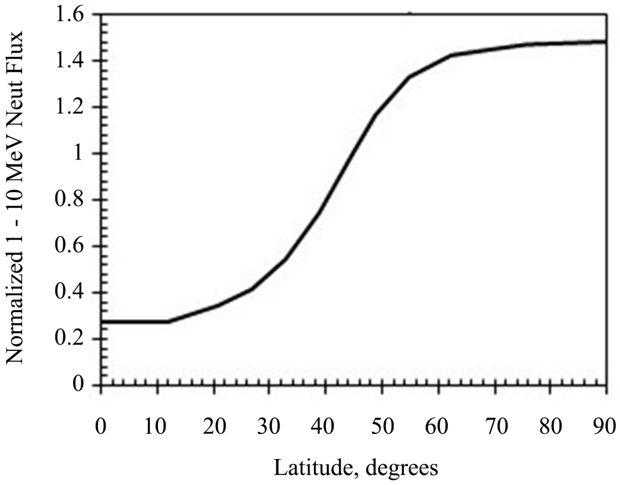

Figure 2. Neutron flux versus latitude (source [31]).

occurred by −20˚C, whereas at low latitude (16˚N) at −30˚C only 70% of clouds are ice-filled (in Northern Hemisphere).

We know that if nucleation is from the same source, or from very similar sources, then the inherent width (10% - 90%) of the nucleation curve narrows [12]. Thus, it seems possible that the measured nucleation at Alaska (51˚N) was due, at least partially, to neutrons causing the less narrow spread. Moreover, Hoose *et al.* [39] state that the "nature and origin of other particles which make up 1% - 34% of the measured IN per ice crystal residue remain to be resolved".

Marine bacteria are also thought to be a source of ice nucleation, with concentrations difficult to enumerate but thought to be on the order of 10 L^{-1} in air [42] [43] [44]. A conservative lower limit on airborne diatom fragment concentrations is thought to be 0.1 L^{-1} in air. Alpert *et al.* [42] point out that ice crystal concentrations in cirrus clouds impacted by heterogeneous INs are "ill-defined and remain largely uncertain". DeMott and Prenni [45] argue the most important carbonaceous particles capable of serving as ice nuclei in clouds warmer than −15˚C may be from biological sources.

Knowledge of cloud processes remains incomplete, yet global precipitation is known to be predominantly produced by clouds with ice. De Mott and Rogers [46] and Liu *et al.*, [47] have combined observations from field studies spanning more than a decade, from a variety of locations around the globe and describe a new relationship which does seem to reduce unexplained variability in IN concentrations at a given temperature from ~10^3 to less than a factor of 10. They describe the remaining variability as "apparently due to variations in aerosol chemical composition or other factors".

4. Discussion

Our recent study [1] held the water supercooled at about −18˚C to −19˚C, suggesting that at −30˚C supercooled water will be significantly more sensitive to the effects of incident neutrons, especially if higher in flux and energy, primarily the former, at higher altitudes [20] [22] [48]. We assume momentum transfer to a nucleus of H or O, pushing the molecule over the energy barrier, as described by classical nucleation theory. The barrier must be lower at −30˚C and it seems reasonable that incident high-energy neutrons will more efficiently force nucleation [48].

Phillips *et al.* [35] found that some samples of atmospheric dust had higher nucleating ability than expected and they postulated that there "may be chemical species of IN within the atmosphere not yet considered." They argue that future advances in observational technology may elucidate the apparent limited understanding of IN. We further argue here that this lack of agreement and agreed uncertainty is potentially due to *particles* not *particulates* being the cause of at least some fraction of nucleation events.

Neutron flux is about a factor seven higher at the poles than at the equator. The solar cycle variation is also much higher at the poles: around a few percent at the equator, up to around 30% at the poles. The Galactic Cosmic Ray (GCR) profile with altitude peaks around 15 km, as this is where the secondary production peaks, as a result of the increasing primary GCR flux with altitude and decreasing atmospheric cross section for collisions [49].

The history of cloud-GCR links suggests that effects on high clouds demonstrate some statistical evidence of possible relationships from surface observations. However, high cloud questions have never been fully resolved mechanistically, and certainly some of the apparently homogenous nucleation events could be caused by GCR interactions [5]. At altitudes of 5 to 10 km there are 5 to 10 supercooled water droplets per cm^3 and there are about 100 neutrons per cm^2 per second. Thus, if we assume 10 droplets per cm^3 and if we assume an average size of 10 μm [50] then we might expect a droplet to be struck by a neutron every 10 seconds, per 1 cm^3 volume of air.

A typical energy of high-altitude neutrons is O (1 MeV) [22] [51]. The total neutron cross section for a typical water molecule, summing across the cross sections for all the possible neutron reactions for two protons and one Oxygen-16 nucleus [52] [53], is ~10 barns, or 10^{-23} cm^2. This leads to a mean free path $\lambda = 1/(\sigma^{*}N) = 1/(10^{-23}\ cm^2 \times 3.345 \times 10^{22}\ cm^{-3}) - 3\ cm$, when assuming a mass density of ~1 g/cm^3 and molar mass of 18 g/mol. Thus, the naïve estimate of a neutron successfully interacting with a water droplet drops from 1 per 10 s to $1-e^{-0.001/3} - 3 \times 10^{-4}$ per second, per cm^3. Nevertheless, at an example temperature of −30˚C, this is several orders of magnitude higher than predictions for more traditional (non-neutron) sources of ice nucleation in the upper atmosphere [38].

5. Conclusions

Thus, even if our still-crude estimate remains too high due to not accounting for all possible factors such as the energy threshold, this is highly suggestive of radiation-induced nucleation being an important, but neglected, effect. The energy threshold question may be somewhat elucidated by the recent calculations of Szydagis *et al.* [2]. Ice growth following nucleation seeding further supercooled droplets should follow the same path and same parameters as occur when the original droplet is seeded with dust or biologics.

Regardless of the source of neutrons present within the atmosphere, at the heights relevant to clouds, they may be responsible for some ice nucleation. The effects of secondary nucleation must be incorporated into further models involving these particles and is beyond the scope of this discussion. It is the primary nucleation event considered here and further work is required within this area to determine the exact effect that neutrons have on the nucleation probability of supercooled water droplets. That work should then inform cloud models and allow for more accurate determination of cloud freezing and also for new studies both of weather and of climate change.

Data Availability Statement

Data analyzed in this study were a re-analysis of existing data, which are openly available at locations cited in the reference section.

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Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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