atmosphere

MDPI

Review

Anthropogenic Aerosols Effects on Ice Clouds: A Review

Yang Yang [1] and Run Liu [1,2,*]

[1] Institute for Environmental and Climate Research, Jinan University, Guangzhou 511443, China; yangyang@stu2019.jnu.edu.cn
[2] Guangdong-Hong Kong-Macau Joint Laboratory of Collaborative Innovation for Environmental Quality, Institute for Environmental and Climate Research, Jinan University, Guangzhou 511443, China
* Correspondence: liurun@jnu.edu.cn; Tel.: +86-20-37336618

Abstract: Since the ability of anthropogenic aerosols to act as ice nucleation particles has been recognized, the effect of anthropogenic aerosols on ice clouds has attracted increasing attentions. In recent years, some progress has been made in investigating the effects of anthropogenic aerosols on ice clouds. In this paper, we briefly review the study on the impact of anthropogenic aerosols on ice nuclei, properties and radiative forcing of ice clouds. Anthropogenic aerosols can form ice nuclei through homogeneous nucleation and heterogeneous nucleation. Convective strength can modulate the response of ice clouds to anthropogenic aerosols by affecting the nucleation activities. There have been large uncertainties in calculating the radiative forcing of anthropogenic aerosols on ice clouds in climate models. Further studies on the impact of anthropogenic aerosols on ice clouds are imperative to provide better parameterization schemes and reduce the uncertainties of aerosol indirect effects.

Keywords: anthropogenic aerosols; ice crystal; ice cloud

check for updates

Citation: Yang, Y.; Liu, R. Anthropogenic Aerosols Effects on Ice Clouds: A Review. *Atmosphere* **2022**, *13*, 910. https://doi.org/10.3390/atmos13060910

Academic Editors: Ling Zou and Alberto Cazorla

Received: 15 April 2022
Accepted: 31 May 2022
Published: 3 June 2022

Publisher's Note: MDPI stays neutral with regard to jurisdictional claims in published maps and institutional affiliations.

Copyright: © 2022 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https://creativecommons.org/licenses/by/4.0/).

1. Introduction

Global aerosol concentrations have increased substantially from pre-industrial time to the present [1–6]. Aerosols can alter the Earth's energy balance, thereby affecting the climate system [7,8]. The direct aerosol effect accounts for the ability of aerosol particles to absorb and scatter solar radiation [9]. The semi-direct aerosol effect accounts for the burning of clouds, due to the warming of the atmosphere by absorbing aerosols [10]. The indirect aerosol effect accounts for the ability of aerosols to act as cloud condensation nuclei (CCN) or ice nuclei (IN) [11]. Changes in aerosol concentration and chemical composition can affect cloud microphysical and optical properties [12,13], such as cloud albedo and cloud lifetime. The indirect aerosol effect is one of the largest uncertainties of global radiative forcing.

Ice clouds, which are composed entirely, or almost entirely, of ice crystals, cover over 30% of the Earth's surface, and they are mainly in the form of cirrus clouds [14,15]. Ice clouds play an important role in regulating the global radiation budget [16]. Two major types of ice clouds are characterized by distinct formation mechanisms: ice clouds formed by deep convections and those formed in situ, due to updrafts caused by frontal systems, gravity waves, or orographic waves [17–19]. Due to the immaturity of the ice crystal observations and identification techniques, the research related to ice clouds is limited to some extent. So far, there are extensive studies on the aerosol impact on liquid and mixed-phase clouds [20–22]. By contrast, less attention has been paid on the connections between aerosols and ice clouds. With the development of satellite observation and numerical simulation technology, the interaction between ice clouds and aerosols, especially the influence of anthropogenic aerosols on ice clouds, has attracted more and more attention [23–27].

Compared with the number of particles that can serve as CCN, the proportion of particles that can serve as IN is very low. Only a fraction of 10^{-3} to 10^{-5} of the background aerosols can serve as IN [28], and the fraction is even several orders of magnitude lower

in marine regions [29]. IN can significantly influence the microphysical properties of ice clouds, such as ice particle number and size [30,31].

In addition, it is still challenging to study the impact of anthropogenic aerosols on microphysical characteristics, such as the cloud cover of ice clouds. This review covers the mechanism and process of the impact of anthropogenic aerosols on ice clouds to help further the understanding of aerosol-cloud-climate interaction and reduce the uncertainty of aerosol indirect effect.

2. Anthropogenic Aerosols as Effective Ice Nucleating Particles

Mineral dust, volcanic ash, and bio-aerosols (such as bacteria, fungal spores, and pollen) are known as effective ice nucleating particles (INPs) that can serve as IN [32,33]. Recently, Sato and Inoue [34] suggested that marine aerosols can act as INPs for ice-cloud formation over near-coastal Antarctic ice-covered areas. However, some aerosols related to anthropogenic activities have also been proven to play a similar role in nucleating ice.

Two studies conducted in Beijing, China compared INP activities during events of heavy pollution haze and dust storms. They found that average INP concentrations, in heavy pollution haze cases, were two times higher than those in dust storms [35]. In 1995, similar observations were conducted for the same season and location, and the severity of storm was even more serious. It was found that the air pollution case still had INP activity comparable to that of the heavy dust counterpart [36]. Yang et al. [37] observed INP concentrations in Nanjing, China, distinguished air masses originating in different regions, including deserts and anthropogenic pollution regions, and showed that INP concentrations in air masses originating in pollution regions were usually larger than those originating from deserts. Yin et al. [38] found that INP concentrations in China increased by a factor of five to ten from 1980 to 2000, which could not be explained by natural processes alone, confirming that anthropogenic aerosols were the main reason for the increase in INP concentrations.

Investigations outside China have similar findings. Knopf et al. [39] observed the heavily polluted atmosphere in Mexico City and found that anthropogenic organic particles had potentially contributed to the formation of cirrus clouds. Cziczo et al. [40] suggested that the residual components of cirrus cloud collected in the field were mainly metallic particles, followed by mineral dust. In addition, fire aerosols have significant impacts on ice clouds over southern Mexico and the central United States during fire events [41].

While field measurements were only conducted at several discrete sites and at the surface, satellite observations can provide a much larger coverage. Using aerosol optical depth (AOD) data, obtained from the Moderate Resolution Imaging Spectroradiometer (MODIS), and aerosol classification information, available from the Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) space-borne instrument [42], Rosenfeld et al. [43] compared the glaciation temperature of growing convective clouds contaminated by different aerosol subtypes over East Asia and showed that desert dust and heavy air pollution over East Asia have a similar ability to glaciate the tops of growing clouds. Recently, using MODIS and CALIOP satellite observations, Zhao et al. [25] found a similar observed effective radius of ice crystals' (R_{ei})-AOD relationships between dusts and polluted continental aerosols, verifying that a portion of polluted continental aerosols had a certain ice nucleating ability.

Aside from field and satellite observations, the ability of anthropogenic aerosols, serving as INPs, is also validated by cloud-resolving sensitivity simulations [25]. Zhao et al. [25] found that, only if a portion of polluted continental aerosols can serve as INPs, the observed relationship between R_{ei} and polluted continental AOD can be produced by the model simulations.

The abovementioned studies fully demonstrate that anthropogenic aerosols have the ability to serve as INPs (Table 1). It should be noted that, although anthropogenic aerosols can serve as INPs, compared to the aerosols from the natural sources, the portion of ice nuclei from anthropogenic aerosols are orders of magnitude smaller. However, it is still of great importance to investigate the effects of anthropogenic aerosols on ice clouds.

Table 1. Summary of investigations on the ability of anthropogenic aerosols serving as INPs.

Location/Period	Method	Remarks	Reference
Beijing, China/spring, 1963	Field measurements	INP concentrations in heavy pollution haze cases were two times higher than those in dust storms	[35]
Beijing, China/spring, 1995	Field measurements	The INP activity of air pollution is comparable to that of heavy dust	[36]
Nanjing, China/winter, 2011	Field measurements	INP concentrations originating from pollution regions were larger than those originating from desserts	[37]
China/1980–2010	Survey of the existing literature on field measurements	Natural processes alone could not explain the increase in INP concentrations	[38]
Mexico City, Mexico/March, 2006	Field measurements	potential contribution of anthropogenicIn highly polluted environment, anthropogenic organic particles had potential contribution to the formation of cirrus clouds	[39]
North and Central America and nearby ocean/2002–2011	Aircraft measurements	Dominant component of ice residuals of cirrus clouds was metallic particle	[40]
East Asia	Satellite observations	Heavy air pollution and desert dust have similar ability to glaciate the tops of growing clouds	[43]
East Asia	Satellite observations	Similar R_{ei}-AOD relationships between dust and polluted continental aerosols was observed	[25]
East Asia	Cloud-resolving model	A portion of polluted continental aerosols can serve as INPs.	[25]

3. Effect of Anthropogenic Aerosols on Ice Cloud Properties

The mechanism and influencing factors of interactions between aerosols and ice clouds are complicated, and they have been recognized as one of the substantial uncertainties in climate change assessment [22]. Limited studies of the effects of anthropogenic aerosols on ice cloud properties have shown contradicting results [44,45].

Ice nucleation occurs following two primary pathways: homogenous freezing of supercooled cloud droplets, or solution particles below about −37 °C [46], and heterogeneous nucleation triggered by INPs [31,47]. Heterogeneous nucleation is the dominant formation mechanism of cirrus clouds [40,48]. With the increases in INPs, the contribution of ice crystals from heterogeneous nucleation is significantly increased. More heterogeneous nucleation could inhibit the production of ice crystals from homogeneous nucleation [48]. The portion of heterogeneous nucleation can significantly alter the subsequent properties of ice clouds. Meanwhile, the chemical composition of anthropogenic aerosols is complex, and it easily changes over time and varies from region to region, due to photochemical processing, mixing state variations, phase state change, and cloud recycling. Organic aerosols [47], lead-bearing aerosols [49–51], black carbon [32,52–54], solid ammonium sulfate [55], and metallic compounds (other than lead) [56] have been discussed as anthropogenic aerosol constituents serving as INPs. Therefore, different studies showed contradictory ice cloud formation activities [47,52,56,57].

3.1. Effect of Anthropogenic Aerosols on Effective Radius of Ice Crystals

R_{ei} is the most important factor to determine the net radiative effect of ice clouds. According to the rule of the Twomey effect, when the cloud water path remains constant, cloud droplet effective radius decreases with the increase in aerosol concentrations [12]. For R_{ei}, an increase in aerosol concentrations lead to the increase in INPs, which promotes the occurrence of heterogeneous nucleation. Since heterogeneous nucleation leads to the formation of less and larger ice crystals compared to homogeneous nucleation [31,58], theoretically, an increase in anthropogenic aerosol concentrations leads to an increase in R_{ei}.

However, in the real atmosphere, the effect of aerosols on R_{ei} is complicated, and studies have reported contrary results.

Jiang et al. [44,59] found that, compared with ice clouds in clean regions, the R_{ei} of ice clouds in polluted regions was smaller, which was in agreement with the classical Twomey effect for liquid clouds. Chylek et al. [58] reported that R_{ei} was larger when pollution was more serious. Zhao et al. [17] further confirmed that the response of the R_{ei} of cirrus clouds to aerosol loading could be either positive (anti-Twomey effect) or negative (Twomey effect), depending on meteorological conditions.

The underlying possible mechanism is revealed in Zhao et al. [25] by conducting cloud-resolving simulations. Generally, in a rising air parcel, because the requirements of the temperature or supersaturation of heterogeneous nucleation are lower, heterogeneous nucleation occurs prior at homogeneous nucleation. For strong convective systems, plenty of water vapor is transported to the upper troposphere, and the ice formation is dominated by the homogeneous freezing of cloud droplets; therefore, R_{ei} decreases with the increasing loading of anthropogenic aerosols (Twomey effect). By contrast, for moderate convective systems, water vapor brought to the upper troposphere by convection is limited. The heterogeneous nucleation can efficiently compete with, and even prevent, homogeneous nucleation. The heterogeneous nucleation could produce less and larger ice crystals than its homogeneous counterpart. Therefore, R_{ei} increases with the increasing loading of anthropogenic aerosols (anti-Twomey effect) [25]. The convection strength does not only change the content of water vapor in the upper troposphere, but it also alters the updraft velocity. A stronger convection with higher updraft velocity is favorable for homogeneous freezing by reducing the growth time of heterogeneously formed ice crystals [25].

3.2. Effect of Anthropogenic Aerosols on Ice Cloud Cover

The response of ice cloud cover to anthropogenic aerosols is complicated. One study using Himawari-8 data found that convective cloud cover first increases then decreases with anthropogenic aerosol loading, and this phenomena is consistent with the combined action of the aerosol microphysical, and radiative effects [60]. There seems to be a turning point of the response of cloud cover to anthropogenic aerosol loadings. The aerosol absorption effect plays an important role only at large aerosol loading [61]; therefore, when aerosol loading is relatively small, cloud cover increases with aerosol concentration, while it decreases with aerosols when aerosol loading is large [26,62,63].

3.3. Effect of Anthropogenic Aerosols on Other Properties of Ice Clouds

The response of the altitude of convective clouds to anthropogenic aerosols is not monotonic [26]. For small anthropogenic loading (AOD ~0.2–0.3), the altitude of convective cloud gets higher with increasing AOD [26], which is in line with the aerosol invigoration hypothesis (that is, through the latent heat release, due to the freezing of a larger amount of water droplets, deep convection can be energized by an increase in CCN) [11]. For heavy aerosol loading conditions (AOD > 0.3), the altitude of convective cloud decreases with AOD, which could be explained by the reduction in sunlight reaching the surface due to the substantial pollution, and in turn, it could weaken the convective intensity. It can be seen that the non-monotonic relationship between the altitude of convective cloud and anthropogenic aerosol concentration is caused by the competition between microphysical and radiative effects of aerosols [26,61,64,65]. The feedback from environmental condition changes might be another contributor to the non-monotonic relationship as well, since this feedback could also be non-monotonic [66,67].

Responses of cloud optical thickness (COT), for different types of ice clouds, to changes in anthropogenic aerosols are different. For convection-generated ice clouds, COT increases with small-to-moderate AODs of <0.3. At larger AODs, COT decreases when AOD increases [24]. A similar "boomerang shape" relationship has also been found for warm clouds [61]. For in situ ice clouds, COT increases rapidly with layer AOD [24]. This could be explained by the aerosols' microphysical effect, which increases COT, far exceeding the

effect of absorptive heating that reduces COT. Compared to other types of aerosols, the fractional increases in COT with anthropogenic AOD are the largest, which is probably because the number concentration of anthropogenic pollution aerosols is larger than other aerosol type for a given AOD [24].

4. Effect of Anthropogenic Aerosols on Radiative Forcing through Ice Clouds

There are considerable uncertainties in calculating the radiative forcing of ice clouds using global climate models, in terms of both its magnitude and its sign [68]. The radiative effects of anthropogenic aerosols on ice clouds are influenced by competition between homogeneous and heterogeneous ice nucleation processes [27]. Parameterization schemes have been developed to deal with both homogeneous and heterogeneous nucleation in global climate models [69–71]. Global numerical simulation experiments, using different ice nucleation parameterization and updraft treatments, result in completely different results. Since it is hard to clearly separate the anthropogenic aerosol forcing through ice clouds from other aerosol forcing (i.e., anthropogenic aerosol forcing through warm clouds and mixed-phase clouds) [48], very few published papers present the global forcing of anthropogenic aerosols through cirrus clouds.

Barahona and Nenes [71] obtained the parameterization scheme (BN Scheme) from the analytical solution of the cloud parcel equation. Liu and Penner [69] obtained the parameterization scheme (LP Scheme) by the simulation of an adiabatic ascending cloud parcel, however, LP Scheme could only deal with the case where the velocity of the updraft was positive, ignoring the evaporation of drops in the downdraft. The parameterization of Kärcher et al. [70] (KL Scheme) explicitly calculates the evolution of ice supersaturation in a rising cloud parcel. KL Scheme is mostly used in studies on aerosol effects on cirrus clouds because it explicitly expresses relevant physical processes [72,73]. However, the competition of aerosol particles, with different size bins for water deposition, is not considered in the KL Scheme. Therefore, homogeneous nucleation of particles with small and medium sizes in the KL Scheme was underestimated [74]. Shi and Liu [48] conducted a sensitivity study of anthropogenic aerosol indirect forcing through cirrus clouds with three ice nucleation parameterizations (i.e., BN Scheme, LP Scheme, and KL Scheme) and found that the change in net cirrus cloud forcing ranged from -0.12 to 0.05 W m^{-2}, reconfirming that different ice nucleation parameterizations are of great importance to the uncertainty of anthropogenic ice aerosol indirect forcing.

To make use of the advantages of various parameterization schemes, different parameterization schemes are often combined. Penner et al. [72] estimated the radiative forcing of aerosols through cirrus clouds ranging from -0.53 to -0.67 W m^{-2} by applying KL and LP schemes on an offline ice nucleation and the radiative transfer model. Gettelman et al. [75] used the LP and BN schemes to calculate the radiative forcing, associated with aerosol effects through cirrus clouds, as 0.27 ± 0.10 W m^{-2}. Zhu and Penner [27] developed a hybrid parameterization scheme, combing the advantages of LP and KL schemes, and assessed the radiative forcing of anthropogenic aerosols through cirrus clouds as -0.20 ± 0.05 W m^{-2}. Since secondary organic aerosols (SOA) can also act as INPs [40,76–79], the radiative forcing turns to -0.04 ± 0.08 W m^{-2} when the influence of SOA is taken into consideration.

5. Summary

The interaction between aerosols and ice clouds has always been a complex and difficult problem to solve. Since the ability of anthropogenic aerosols to act as INPs has been recognized, several investigations on the influence of anthropogenic aerosols on ice clouds have been carried out, mainly focusing on the formation of ice nuclei, properties of ice clouds, radiative forcing of ice clouds, etc., but the understanding of this relationship is not clear.

One of the factors limiting the study of anthropogenic aerosol effects on ice clouds is the lack of observed data. In practice, only satellite observations can provide a global scale data. CALIPSO and CloudSat satellites provide information of different types of ice

clouds and aerosols, and they promote the study on the interaction between anthropogenic aerosols and ice clouds. Although satellite observations have irreplaceable advantages in space coverage, polar-orbiting satellites can only provide fixed time data, which can be used to study the transient relationship between anthropogenic aerosols and ice clouds, rather than the entire life cycle of an ice cloud. In addition, the uncertainty of satellite observation data is an important defect of its application. Ground-based observations can make up for the deficiency of satellite observations. Ground-based observation has the problem of insufficient space coverage; however, it has great advantages in accuracy and personalized observations. Meanwhile, the vertical distribution characteristics of aerosols and ice clouds can be obtained in alpine areas or by aircraft observations, which is convenient for the study of the influence of anthropogenic aerosols on ice clouds.

Multiple observations can be merged to reduce uncertainties. The challenge of using numerical models to study ice clouds lies in how to update laboratory, ground-based measurements and satellite observations to develop parameterized schemes that can accurately quantify the interaction between anthropogenic aerosols and ice clouds, as well as build more sophisticated numerical models to further study the mechanism of anthropogenic aerosols on ice clouds and climate.

Author Contributions: Writing—Original draft preparation, Y.Y.; writing—review and editing, Y.Y., R.L.; supervision, R.L.; project administration, R.L.; funding acquisition, R.L. All authors have read and agreed to the published version of the manuscript.

Funding: This research was funded by the National Natural Science Foundation of China (41805115), Guangzhou Municipal Science and Technology Project, China (202002020065), and Special Fund Project for Science and Technology Innovation Strategy of Guangdong Province (2019B121205004).

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Acknowledgments: We are grateful to the four anonymous reviewers for their thoughtful reviews and advice, which led to an improved revised manuscript.

Conflicts of Interest: The authors declare no conflict of interest.

References

1. Hoyle, C.R.; Myhre, G.; Berntsen, T.K.; Isaksen, I.S.A. Anthropogenic influence on SOA and the resulting radiative forcing. *Atmos. Chem. Phys.* **2009**, *9*, 2715–2728. [CrossRef]
2. Skeie, R.B.; Berntsen, T.K.; Myhre, G.; Tanaka, K.; Kvalevåg, M.M.; Hoyle, C.R. Anthropogenic radiative forcing time series from pre-industrial times until 2010. *Atmos. Chem. Phys.* **2011**, *11*, 11827–11857. [CrossRef]
3. Skeie, R.B.; Berntsen, T.; Myhre, G.; Pedersen, C.A.; Ström, J.; Gerland, S.; Ogren, J.A. Black carbon in the atmosphere and snow, from pre-industrial times until present. *Atmos. Chem. Phys.* **2011**, *11*, 6809–6836. [CrossRef]
4. Houghton, J.T.; Meira Filho, L.G.; Callander, B.A.; Harris, N.; Kattenberg, A.; Maskell, K. *Climate Change 1995: The Science of Climate Change*; Contribution of WGI to the Second Assessment Report of the Intergovernmental Panel on Climate Change; Cambridge University Press: Cambridge, UK, 1996; 572p.
5. Shen, X.; Liu, B.; Li, G.; Wu, Z.; Jin, Y.; Yu, P.; Zhou, D. Spatiotemporal change of diurnal temperature range and its relationship with sunshine duration and precipitation in China. *J. Geophys. Res. Atmos.* **2014**, *119*, 13163–13179. [CrossRef]
6. Williams, A.S.; Igel, A.L. Cloud top radiative cooling rate drives non-precipitating stratiform cloud responses to aerosol concentration. *Geophys. Res. Lett.* **2021**, *48*, e2021GL094740. [CrossRef]
7. Haywood, J.; Boucher, O. Estimates of the direct and indirect radiative forcing due to tropospheric aerosols: A review. *Rev. Geophys.* **2000**, *38*, 513–543. [CrossRef]
8. Kaufman, Y.J.; Tanré, D.; Boucher, O. A satellite view of aerosols in the climate system. *Nature* **2002**, *419*, 215–223. [CrossRef]
9. Hansen, J.; Sato, M.; Ruedy, R. Radiative forcing and climate response. *J. Geophys. Res. Atmos.* **1997**, *102*, 6831–6864. [CrossRef]
10. Charlson, R.J.; Pilat, M.J. Climate: The influence of aerosols. *J. Appl. Meteorol. Climatol.* **1969**, *8*, 1001–1002. [CrossRef]
11. Rosenfeld, D.; Lohmann, U.; Raga, G.B.; O'Dowd, C.D.; Kulmala, M.; Fuzzi, S.; Reissell, A.; Andreae, M.O. Flood or drought: How do aerosols affect precipitation? *Science* **2008**, *312*, 1309–1313. [CrossRef]
12. Twomey, S. The influence of pollution on the shortwave albedo of clouds. *J. Atmos. Sci.* **1977**, *34*, 1149–1152. [CrossRef]
13. Albrecht, B. Aerosols, cloud microphysics, and fractional cloudiness. *Science* **1989**, *245*, 1227–1230. [CrossRef] [PubMed]
14. Wylie, D.P.; Jackson, D.L.; Menzel, W.P.; Bates, J.J. Trends in global cloud cover in two decades of HIRS observations. *J. Clim.* **2005**, *18*, 3021–3031. [CrossRef]

15. Wylie, D.P.; Menzel, W.P.; Woolf, H.M.; Strabala, K.I. Four years of global cirrus cloud statistics using HIRS. *J. Clim.* **1994**, *7*, 1972–1986. [CrossRef]
16. Liou, K.N. Influence of cirrus clouds on weather and climate processes: A global perspective. *Mon. Weather Rev.* **1986**, *114*, 1167–1199. [CrossRef]
17. Mace, G.G.; Clothiaux, E.E.; Ackerman, T.P. The composite characteristics of cirrus clouds: Bulk properties revealed by one year of continuous cloud radar data. *J. Clim.* **2001**, *14*, 2185–2203. [CrossRef]
18. Mace, G.G.; Benson, S.; Vernon, E. Cirrus clouds and the large-scale atmospheric state: Relationships revealed by six years of ground-based data. *J. Clim.* **2006**, *19*, 3257–3278. [CrossRef]
19. Krämer, M.; Rolf, C.; Luebke, A.; Afchine, A.; Spelten, N.; Costa, A.; Meyer, J.; Zöger, M.; Smith, J.; Herman, R.L.; et al. A microphysics guide to cirrus clouds-Part I: Cirrus types. *Atmos. Chem. Phys.* **2016**, *16*, 3463–3483. [CrossRef]
20. Zhao, B.; Jiang, J.H.; Gu, Y.; Diner, D.; Worden, J.; Liou, K.N.; Su, H.; Xing, J.; Garay, M.; Huang, L. Decadal-scale trends in regional aerosol particle properties and their linkage to emission changes. *Environ. Res. Lett.* **2017**, *12*, 054021. [CrossRef]
21. Rosenfeld, D.; Andreae, M.O.; Asmi, A.; Chin, M.; de Leeuw, G.; Donovan, D.P.; Kahn, R.; Kinne, S.; Kivekäs, N.; Kulmala, M.; et al. Global observations of aerosol-cloud-precipitation-climate interactions. *Rev. Geophys.* **2014**, *52*, 750–808. [CrossRef]
22. IPCC. *The Physical Science Basis*; Contribution of Working Group I to the Sixth Assessment Report of the Intergovernmental Panel on Climate Change; Cambridge University Press: Cambridge, UK, 2021.
23. Zhao, B.; Liou, K.N.; Gu, Y.; Jiang, J.H.; Li, Q.; Fu, R.; Huang, L.; Liu, X.; Shi, X.; Su, H.; et al. Impact of aerosols on ice crystal size. *Atmos. Chem. Phys.* **2018**, *18*, 1065–1078. [CrossRef] [PubMed]
24. Zhao, B.; Gu, Y.; Liou, K.N.; Wang, Y.; Liu, X.; Huang, L.; Jiang, J.H.; Su, H. Type-dependent responses of ice cloud properties to aerosols from satellite retrievals. *Geophys. Res. Lett.* **2018**, *45*, 3297–3306. [CrossRef]
25. Zhao, B.; Wang, Y.; Gu, Y.; Liou, K.N.; Jiang, J.H.; Fan, J.; Liu, X.; Huang, L.; Yung, Y.L. Ice nucleation by aerosols from anthropogenic pollution. *Nat. Geosci.* **2019**, *12*, 602–607. [CrossRef]
26. Jiang, J.H.; Su, H.; Huang, L.; Wang, Y.; Massie, S.; Zhao, B.; Omar, A.; Wang, Z. Contrasting effects on deep convective clouds by different types of aerosols. *Nat. Commu.* **2018**, *9*, 3874. [CrossRef] [PubMed]
27. Zhu, J.; Penner, J.E. Radiative forcing of anthropogenic aerosols on cirrus clouds using a hybrid ice nucleation scheme. *Atmos. Chem. Phys.* **2020**, *20*, 7801–7827. [CrossRef]
28. Rogers, D.C.; DeMott, P.J.; Kreidenweis, S.M.; Chen, Y. Measurements of ice nucleating aerosols during SUCCESS. *Geophys. Res. Lett.* **1998**, *25*, 1383–1386. [CrossRef]
29. Rosinski, J.; Morgan, G.M. Ice-forming nuclei in transvaal, Republic of South Africa. *J. Aerosol Sci.* **1988**, *19*, 531–538. [CrossRef]
30. Seinfeld, J.H.; Bretherton, C.; Carslaw, K.S.; Coe, H.; DeMott, P.J.; Dunlea, E.J.; Feingold, G.; Ghan, S.; Guenther, A.B.; Kahn, R.; et al. Improving our fundamental understanding of the role of aerosol–cloud interactions in the climate system. *Proc. Natl. Acad. Sci. USA* **2016**, *113*, 5781–5790. [CrossRef]
31. DeMott, P.J.; Prenni, A.J.; Liu, X.; Kreidenweis, S.M.; Petters, M.D.; Twohy, C.H.; Richardson, M.S.; Eidhammer, T.; Rogers, D.C. Predicting global atmospheric ice nuclei distributions and their impacts on climate. *Proc. Natl. Acad. Sci. USA* **2010**, *107*, 11217–11222. [CrossRef]
32. Hoose, C.; Möhler, O. Heterogeneous ice nucleation on atmospheric aerosols: A review of results from laboratory experiments. *Atmos. Chem. Phys.* **2012**, *12*, 9817–9854. [CrossRef]
33. Murray, B.J.; O'Sullivan, D.; Atkinson, J.D.; Webb, M.E. Ice nucleation by particles immersed in supercooled cloud droplets. *Chem. Soc. Rev.* **2012**, *41*, 6519–6554. [CrossRef]
34. Sato, K.; Inoue, J. Seasonal change in satellite-retrieved lower-tropospheric ice-cloud fraction over the Southern Ocean. *Geophys. Res. Lett.* **2021**, *48*, e2021GL095295. [CrossRef]
35. You, L.; Shi, A. The measurement and analysis of ice-nucleuscon centration at Peking during the period from march 18th to April 30th in 1963. *Acta Meteorol. Sin.* **1964**, *34*, 548–554.
36. You, L.; Yang, S.; Wang, X.; Pi, J. Study of ice nuclei concentration at Beijing in spring of 1995 and 1996. *Acta Meteorol. Sin.* **2002**, *60*, 101–109.
37. Yang, L.; Yin, Y.; Yang, S.; Su, H.; Jiang, H. Characteristics of atmospheric ice nuclei and its relationship to aerosols in winter in Nanjing. *Chin. J. Atmos. Sci.* **2013**, *37*, 983–993.
38. Yin, J.; Wang, D.; Zhai, G. Long-term in situ measurements of the cloud-precipitation microphysical properties over East Asia. *Atmos. Res.* **2011**, *102*, 206–217. [CrossRef]
39. Knopf, D.A.; Wang, B.; Laskin, A.; Moffet, R.C.; Gilles, M.K. Heterogeneous nucleation of ice on anthropogenic organic particles collected in Mexico City. *Geophys. Res. Lett.* **2010**, *37*, L11803. [CrossRef]
40. Cziczo, D.J.; Froyd, K.D.; Hoose, C.; Jensen, E.J.; Diao, M.; Zondlo, M.A.; Smith, J.B.; Twohy, C.H.; Murphy, D.M. Clarifying the dominant sources and mechanisms of cirrus cloud formation. *Science* **2013**, *340*, 1320–1324. [CrossRef]
41. Liu, Y.; Zhang, K.; Qian, Y.; Wang, Y.; Zou, Y.; Song, Y.; Wan, H.; Liu, X.; Yang, X.-Q. Investigation of short-term effective radiative forcing of fire aerosols over North America using nudged hindcast ensembles. *Atmos. Chem. Phys.* **2018**, *18*, 31–47. [CrossRef]
42. Omar, A.H.; Winker, D.M.; Vaughan, M.A.; Hu, Y.; Trepte, C.R.; Ferrare, R.A.; Lee, K.P.; Hostetler, C.A.; Kittaka, C.; Rogers, R.R.; et al. The CALIPSO automated aerosol classification and lidar ratio selection algorithm. *J. Atmos. Ocean Technol.* **2009**, *26*, 1994–2014. [CrossRef]

43. Rosenfeld, D.; Yu, X.; Liu, G.; Xu, X.; Zhu, Y.; Yue, Z.; Dai, J.; Dong, Z.; Dong, Y.; Peng, Y. Glaciation temperatures of convective clouds ingesting desert dust, air pollution and smoke from forest fires. *Geophys. Res. Lett.* **2011**, *38*, L21804. [CrossRef]
44. Jiang, J.H.; Su, H.; Schoeberl, M.R.; Massie, S.T.; Colarco, P.; Platnick, S.; Livesey, N.J. Clean and polluted clouds: Relationships among pollution, ice clouds, and precipitation in South America. *Geophys. Res. Lett.* **2008**, *35*, L14804. [CrossRef]
45. Patnaude, R.; Diao, M. Aerosol indirect effects on cirrus clouds based on global aircraft observations. *Geophys. Res. Lett.* **2020**, *47*, e2019GL086550. [CrossRef]
46. Koop, T.; Luo, B.; Tsias, A.; Peter, T. Water activity as the determinant for homogeneous ice nucleation in aqueous solutions. *Nature* **2000**, *406*, 611–614. [CrossRef] [PubMed]
47. Knopf, D.A.; Alpert, P.A.; Wang, B. The role of organic aerosol in atmospheric ice nucleation: A review. *ACS Earth Space Chem.* **2018**, *2*, 168–202. [CrossRef]
48. Shi, X.; Liu, X. Sensitivity study of anthropogenic aerosol indirect forcing through cirrus clouds with CAM5 using three ice nucleation parameterizations. *J. Meteorol. Res.* **2018**, *32*, 693–706. [CrossRef]
49. Cziczo, D.J.; Stetzer, O.; Worringen, A.; Ebert, M.; Weinbruch, S.; Kamphus, M.; Gallavardin, S.J.; Curtius, J.; Borr-mann, S.; Froyd, K.D.; et al. Inadvertent climate modification due to anthropogenic lead. *Nat. Geosci.* **2009**, *2*, 333–336. [CrossRef]
50. Ebert, M.; Worringen, A.; Benker, N.; Mertes, S.; Weingartner, E.; Weinbruch, S. Chemical composition and mixing state of ice residuals sampled within mixed phase clouds. *Atmos. Chem. Phys.* **2011**, *11*, 2805–2816. [CrossRef]
51. Kamphus, M.; Ettner-Mahl, M.; Killmach, T.; Drewnick, F.; Keller, L.; Cziczo, D.J.; Mertes, S.; Borrmann, S.; Curtius, J. Chemical composition of ambient aerosol, ice residues, and lcoud droplet residues in mixed-phase clouds: Single particle analysis during the Cloud and Aerosol Characterization Experiment (CLACE 6). *Atmos. Chem. Phys.* **2010**, *10*, 8077–8095. [CrossRef]
52. Bond, T.C.; Doherty, S.J.; Fahey, D.W.; Forster, P.M.; Berntsen, T.; DeAngelo, B.J.; Flanner, M.G.; Ghan, S.; Kärcher, B.; Koch, D.; et al. Bounding the role of black carbon in the climate system: A scientific assessment. *J. Geophys. Res. Atmos.* **2013**, *118*, 5380–5552. [CrossRef]
53. Mahrt, F.; Marcolli, C.; David, R.O.; Grönquist, P.; Barthazy Meier, E.J.; Lohmann, U.; Kanji, Z.A. Ice nucleation abilities of soot particles determined with the Horizontal Ice Nucleation Chamber. *Atmos. Chem. Phys.* **2018**, *18*, 13363–13392. [CrossRef]
54. Charnawskas, J.C.; Alpert, P.A.; Lambe, A.T.; Berkemeier, T.; O'Brien, R.E.; Massoli, P.; Onasch, T.B.; Shiraiwa, M.; Moffet, R.C.; Gilles, M.K.; et al. Condensed-phase biogenic-anthropogenic interactions with implications for cold cloud formation. *Faraday Discuss.* **2017**, *200*, 165–194. [CrossRef] [PubMed]
55. Hung, H.M.; Malinowski, A.; Martin, S.T. Ice nucleation kinetics of aerosols containing aqueous and solid ammonium sulfate particles. *J. Phys. Chem. A* **2002**, *106*, 293–306. [CrossRef]
56. Kanji, Z.A.; Ladino, L.A.; Wex, H.; Boose, Y.; Burkert-Kohn, M.; Cziczo, D.J.; Krämer, M. Chapter 1: Overview of Ice Nucleating Particles. In *Ice Formation and Evolution in Clouds and Precipitation: Measurement and Modeling Challenges*; Baumgardner, D., McFarquhar, G.M., Heymsfield, A.J., Eds.; American Meteorological Society: Boston, MA, USA, 2017; Volume 58, pp. 1.1–1.33.
57. Cziczo, D.J.; Ladino, L.; Boose, Y.; Kanji, Z.A.; Kupiszewski, P.; Lance, S.; Mertes, S.; Wex, H. Chapter 8: Measurements of Ice Nucleating Particles and Ice Residuals. In *Ice Formation and Evolution in Clouds and Precipitation: Measurement and Modeling Challenges*; Baumgardner, D., McFarquhar, G.M., Heymsfield, A.J., Eds.; American Meteorological Society: Boston, MA, USA, 2017; Volume 58, pp. 8.1–8.13.
58. Chylek, P.; Dubey, M.K.; Lohmann, U.; Ramanathan, V.; Kaufman, Y.J.; Lesins, G.; Hudson, J.; Altmann, G.; Olsen, S. Aerosol indirect effect over the Indian Ocean. *Geophys. Res. Lett.* **2006**, *33*, L06806. [CrossRef]
59. Jiang, J.H.; Su, H.; Zhai, C.; Massie, S.T.; Schoeberl, M.R.; Colarco, P.R.; Platnick, S.; Gu, Y.; Liou, K.N. Influence of convection and aerosol pollution on ice cloud particle effective radius. *Atmos. Chem. Phys.* **2011**, *11*, 457–463. [CrossRef]
60. Chen, T.; Li, Z.; Kahn, R.A.; Zhao, C.; Rosenfeld, D.; Guo, J.; Han, W.; Chen, D. Potential impact of aerosols on convective clouds revealed by Himawari-8 observations over different terrain types in eastern China. *Atmos. Chem. Phys.* **2021**, *21*, 6199–6220. [CrossRef]
61. Koren, I.; Martins, J.V.; Remer, L.A.; Afargan, H. Smoke invigoration versus inhibition of clouds over the Amazon. *Science* **2008**, *321*, 946–949. [CrossRef] [PubMed]
62. Guo, J.; Su, T.; Li, Z.; Miao, Y.; Li, J.; Liu, H.; Xu, H.; Cribb, M.; Zhai, P. Declining frequency of summertime local-scale precipitation over eastern China from 1970 to 2010 and its potential link to aerosols. *Geophys. Res. Lett.* **2017**, *44*, 5700–5708. [CrossRef]
63. Wang, Q.; Li, Z.; Guo, J.; Zhao, C.; Cribb, M. The climate impact of aerosols on the lightning flash rate: Is it detectable from long-term measurements? *Atmos. Chem. Phys.* **2018**, *18*, 12797–12816. [CrossRef]
64. Tao, W.K.; Chen, J.P.; Li, Z.; Wang, C.; Zhai, C. Impact of aerosols on convective clouds and precipitation. *Rev. Geophys.* **2012**, *50*, RG2001. [CrossRef]
65. Fan, J.; Wang, Y.; Rosenfeld, D.; Liu, X. Review of aerosol-cloud interactions: Mechanisms, significance, and challenges. *J. Atmos. Sci.* **2016**, *73*, 4221–4252. [CrossRef]
66. Dagan, G.; Koren, I.; Altaratz, O.; Heiblum, R.H. Aerosol effect on the evolution of the thermodynamic properties of warm convective cloud fields. *Sci. Rep.* **2016**, *6*, 38769. [CrossRef]
67. Wall, C.; Zipser, E.; Liu, C. An investigation of the aerosol indirect effect on convective intensity using satellite observations. *J. Atmos. Sci.* **2014**, *71*, 430–447. [CrossRef]
68. Storelvmo, T. Aerosol effects on climate via mixed-phase and ice clouds. *Annu. Rev. Earth Planet Sci.* **2017**, *45*, 199–222. [CrossRef]
69. Liu, X.; Penner, J.E. Ice nucleation parameterization for global models. *Meteorol. Z.* **2005**, *14*, 499–514. [CrossRef]

70. Kärcher, B.; Hendricks, J.; Lohmann, U. Physically based parameterization of cirrus cloud formation for use in global atmospheric models. *J. Geophys. Res. Atmos.* **2006**, *111*, D01205. [CrossRef]
71. Barahona, D.; Nenes, A. Parameterization of cirrus cloud formation in large-scale models: Homogeneous nucleation. *J. Geophys. Res. Atmos.* **2008**, *113*, D11211. [CrossRef]
72. Penner, J.E.; Chen, Y.; Wang, M.; Liu, X. Possible influence of anthropogenic aerosols on cirrus clouds and anthropogenic forcing. *Atmos. Chem. Phys.* **2009**, *9*, 879–896. [CrossRef]
73. Penner, J.E.; Zhou, C.; Garnier, A.; Mitchell, D.L. Anthropogenic aerosol indirect effects in cirrus clouds. *J. Geophys. Res. Atmos.* **2018**, *123*, 11652–11677. [CrossRef]
74. Liu, X.; Shi, X. Sensitivity of homogeneous ice nucleation to aerosol perturbations and its implications for aerosol indirect effects through cirrus clouds. *Geophys. Res. Lett.* **2018**, *45*, 1684–1691. [CrossRef]
75. Gettelman, A.; Liu, X.; Barahona, D.; Lohmann, U.; Chen, C. Climate impacts of ice nucleation. *J. Geophys. Res. Atmos.* **2012**, *117*, D20201. [CrossRef]
76. Ignatius, K.; Kristensen, T.B.; Järvinen, E.; Nichman, L.; Fuchs, C.; Gordon, H.; Herenz, P.; Hoyle, C.R.; Duplissy, J.; Garimella, S.; et al. Heterogeneous ice nucleation of viscous secondary organic aerosol produced from ozonolysis of α-pinene. *Atmos. Chem. Phys.* **2016**, *16*, 6495–6509. [CrossRef]
77. Wagner, R.; Höhler, K.; Huang, W.; Kiselev, A.; Möhler, O.; Mohr, C.; Pajunoja, A.; Saathoff, H.; Schiebel, T.; Shen, X.; et al. Heterogeneous ice nucleation of α-pinene SOA particles before and after ice cloud processing. *J. Geophys. Res. Atmos.* **2017**, *122*, 4924–4943. [CrossRef]
78. DeMott, P.J.; Cziczo, D.J.; Prenni, A.J.; Murphy, D.M.; Kreidenweis, S.M.; Thomson, D.S.; Borys, R.; Rogers, D.C. Measurements of the concentration and composition of nuclei for cirrus formation. *Proc. Natl. Acad. Sci. USA* **2003**, *100*, 14655–14660. [CrossRef] [PubMed]
79. Wilson, T.W.; Murray, B.J.; Wagner, R.; Möhler, O.; Saathoff, H.; Schnaiter, M.; Skrotzki, J.; Price, H.C.; Malkin, T.L.; Dobbie, S.; et al. Glassy aerosols with a range of compositions nucleate ice heterogeneously at cirrus temperatures. *Atmos. Chem. Phys.* **2012**, *12*, 8611–8632. [CrossRef]