Cirrus cloud formation and the role of heterogeneous ice nuclei

Cirrus cloud formation and the role of heterogeneous ice nuclei

Karl D. Froyd, Daniel J. Cziczo, Corinna Hoose, Eric J. Jensen, Minghui Diao, Mark A. Zondlo, Jessica B. Smith, Cynthia H. Twohy, and Daniel M. Murphy

Citation: AIP Conference Proceedings **1527**, 976 (2013); doi: 10.1063/1.4803436
View online: http://dx.doi.org/10.1063/1.4803436
View Table of Contents: http://scitation.aip.org/content/aip/proceeding/aipcp/1527?ver=pdfcov
Published by the AIP Publishing

Cirrus Cloud Formation and the Role of Heterogeneous Ice Nuclei

Karl D. Froyd[a,b], Daniel J. Cziczo[c], Corinna Hoose[d], Eric J. Jensen[e], Minghui Diao[f], Mark A. Zondlo[f], Jessica B. Smith[g], Cynthia H. Twohy[h], and Daniel M. Murphy[a]

[a] NOAA Earth System Research Laboratory, Chemical Sciences Division, Boulder, CO 80305 USA.
[b] Cooperative Institute for Research in Environmental Science, University of Colorado, Boulder, CO 80309 USA.
[c] Department of Earth, Atmospheric and Planetary Sciences, Massachusetts Institute of Technology, 77 Massachusetts Ave., Cambridge, MA, 02139, USA.
[d] Institute for Meteorology and Climate Research – Atmospheric Aerosol Research, Karlsruhe Institute of Technology, 76021 Karlsruhe, Germany.
[e] NASA Ames Research Center, Moffett Field, CA 94035, USA.
[f] Department of Civil and Environmental Engineering, Princeton University, Princeton, NJ 08544, USA.
[g] School of Engineering and Applied Sciences, Harvard University, Cambridge, MA, 02138, USA.
[h] College of Earth, Ocean and Atmospheric Sciences, Oregon State University, Corvallis, OR 97331, USA.

Abstract. Composition, size, and phase are key properties that define the ability of an aerosol particle to initiate ice in cirrus clouds. Properties of cirrus ice nuclei (IN) have not been well constrained due to a lack of systematic measurements in the upper troposphere. We have analyzed the size and composition of sublimated cirrus particles sampled from a high altitude research aircraft using both in situ and offline techniques. Mineral dust and metallic particles are the most enhanced residue types relative to background aerosol. Using a combination of cirrus residue composition, relative humidity, and cirrus particle concentration measurements, we infer that heterogeneous nucleation is a dominant cirrus formation mechanism for the mid-latitude, subtropical, and tropical regions under study. Other proposed heterogeneous IN including biomass burning particles, elemental carbon, and biological material were not abundant in cirrus residuals.

Keywords: Cirrus. Ice nucleation. Mineral dust.
PACS: 92.60.Mt, 64.60.Q-, 92.70.Kb, 92.60.Qx

CIRRUS CLOUDS FORMATION

The effect of clouds on the climate system is more uncertain than the influence of heat-trapping greenhouse gases. High altitude cirrus clouds have extensive global coverage and are the first clouds to attenuate the sun's energy and the last which can trap the Earth's heat. Recent satellite observations indicate a slight increase in cirrus coverage. Humans can influence cirrus clouds and radiative impact by altering emissions of aerosol particles that either promote or inhibit ice formation. Projections

Nucleation and Atmospheric Aerosols
AIP Conf. Proc. 1527, 976-978 (2013); doi: 10.1063/1.4803436

for the following decades indicate increasing emissions for several aerosol types known to nucleate ice in laboratory studies.

Ice initiation in cirrus clouds can occur as heterogeneous process whereby water molecules nucleate at a solid surface or by homogeneous freezing of a liquid aerosol. The ice initiation process helps define cirrus microphysical and radiative properties. In homogeneous freezing a substantial fraction of the ambient aerosol population can form ice, whereas heterogeneous ice nuclei (IN) typically number $<<1\%$ of atmospheric aerosol particles and generate correspondingly fewer cloud particles. Different aerosol types have characteristic ice onset temperatures and relative humidities and promote different freezing mechanisms.

We have determined the chemical and physical properties of the particles on which cirrus ice crystals formed from measurements acquired aboard a high altitude NASA research aircraft. These data reveal which particle types are relevant for cirrus formation, and we can also infer the mechanism by which these clouds formed.

MEASUREMENTS IN THE UPPER TROPOSPHERE

Several aircraft measurement campaigns were conducted between 2002 and 2011 over North and Central America and nearby ocean. Ice crystals were sampled into a flow stream where the water was removed. The remaining material was analyzed in situ using the NOAA Particle Analysis by Laser Mass Spectrometry (PALMS) single-particle mass spectrometer. The in situ measurements were complemented by ice residual collection for off-line electron microscopy compositional analysis. We term the material within an ice crystal an ice residual (IR) and not an IN because particles and gases may be scavenged after ice nucleation.

Properties of sampled cirrus IR are shown in Figure 1. The predominant particle category on which freezing took place was mineral dust/metallic particles. These particle are easily distinguished from other types based on their single-particle mass spectra (Fig 1 A & B) and their morphology combined with energy dispersive X-ray microanalysis (Fig 1 D-G). It should be noted that ice crystal impaction onto aircraft surfaces and sampling inlets generates artifact particles that can be mistaken for cirrus IR (Fig 1 C & H). These artifacts were often more abundant than cirrus IR, yet the composition techniques employed allow for reliable removal from the analysis.

The mode of cirrus ice formation is inferred from the composition: when the IR were predominantly sulfate and organic and similar to the near-cloud particles, homogeneous freezing is inferred, whereas dissimilar IR and near-cloud particles are categorized as heterogeneous. Based on these criteria, heterogeneous nucleation was the freezing mechanism in the large majority of cloud encounters. The IR composition data that infer a predominant heterogeneous mechanism are consistent with clear sky relative humidity measurements that rarely exceed 140%.

A global simulation of upper tropospheric aerosol particle concentrations combined with laboratory measurements of IN activity was used to determine cirrus-forming properties for different aerosol types. In agreement with, but independent of the IR composition measurements, this analysis asserts the dominance of mineral dust among cirrus IR.

The combination of in situ determination of IR composition, relative humidity measurements and ice crystal number densities from multiple field campaigns in different regions present a compelling case to consider heterogeneous freezing the dominant mechanism of cirrus formation throughout the study regions. These data stand in contrast to recent model studies which suggest the opposite.

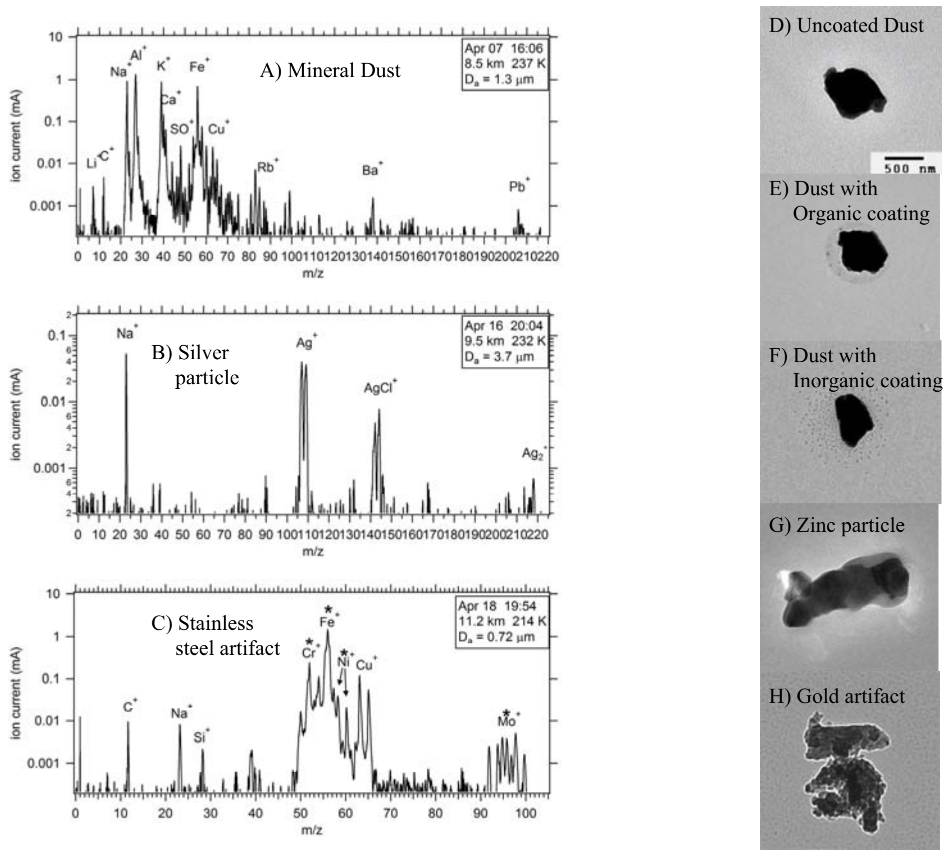

FIGURE 1. The composition of the most abundant types of cirrus ice residuals. Representative PALMS spectra show mineral dust (A) and metallic particles (B). EM images and elemental analysis confirm mineral dust with no coating (D), a thick organic coating (E), an inorganic coating (F), and also a metallic particle (G). Artifacts from ice crystal impaction on aircraft and inlet surfaces are evident as stainless steel particles (C) and gold particles from electroplated surfaces (H).