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CONTRAIL MICROPHYSICS

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With our present state of knowledge, we cannot distinguish contrail cirrus from natural cirrus on the basis of their microphysical properties, making the effects of contrails on climate uncertain.

Contrails, which are products of aircraft emissions in the upper troposphere at temperatures of about −40°C and below, are among the most visible human influences on the Earth's climate. Initially, the microphysical properties of contrails differ from those of natural cirrus, but with age contrails may lose their shape and spread, becoming virtually indistinguishable from natural cirrus, both visually and perhaps also in their microphysical properties. Whether contrails disappear or develop into contrail cirrus clouds depends upon the environmental relative humidity with respect to ice. Contrails will persist in an ice-saturated atmosphere. With supersaturation, ice crystals develop and extract excess environmental water vapor. The transition of line-shaped contrails into cirrus-like clouds, however, is neither well understood nor well represented in climate models.

Contrail formation is described by the Schmidt–Appleman Criterion (SAC)[1], a simple equation that is a function of atmospheric temperature and pressure, the fuel energy content, the amount of water vapor exhausted, and the aircraft's overall propulsion efficiency. The reliability of the SAC to predict visible contrail-forming conditions has been confirmed by observations from various research flights, summarized in IPCC (1999). The same observations indicate that thermodynamics, not the physicochemistry of the exhaust soot from the aircraft, is the primary controlling factor for contrail formation. The SAC states that a contrail forms during the plume expansion process if the mixture of exhaust gases and ambient air briefly reaches or surpasses saturation with respect to liquid water. The fact that the mixture must reach water saturation to form a visible contrail is the only empirical component of the thermodynamic approach, and it is the only component that is related to the ice-forming properties of emitted Aitken-mode soot particles and the ultrafine liquid particles that form prior to the formation of contrails. That is, these plume particles act primarily as cloud condensation nuclei and are poor ice-forming nuclei, at least prior to water activation. Small differences in the assumed ice nucleation behavior of the soot do not significantly affect the SAC for visible contrail formation (Kärcher et al. 1996).

An aircraft's contribution of water vapor to the total vapor content of the jet plume is appreciable. Initially, the microphysics is determined by processes

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[1] This is formulated in a convenient format by Schumann et al. (1996) based on earlier work by Schmidt (1941) and Appleman (1953).

occurring within approximately one wingspan behind the aircraft—chemical and water activation of soot aerosols and the subsequent formation of ice mostly on these particles (Kärcher et al. 1996). Contrail crystals are then captured within the downward-traveling vortex pair generated by the aircraft, descending a few hundred meters with an average speed of about 2 m s^{-1}, which induces adiabatic compression, heating, and ice crystal sublimation (Lewellen and Lewellen 2000; Unterstraßer et al. 2008). This phase reduces the ice particle concentration and may modify the mean ice particle size. Subsequent growth of the contrail crystals depends on the ambient relative humidity, the vertical wind velocities, wind shear, and turbulence.

The objective of this paper, as part of the goal of the Aviation Climate Change Research Initiative (ACCRI) to provide scientific input to inform mitigation policies, is threefold: First, we review the current state of understanding of the science of contrails: how they are formed; their microphysical properties as they evolve and develop into contrail cirrus, and whether their microphysical properties can be distinguished from natural cirrus; and the ice-nucleating properties of soot aerosols, and whether these aerosols can nucleate cirrus crystals. Second, we identify key uncertainties and gaps in the present state of knowledge; and third, we make recommendations for future research to address the impacts of contrails and contrail cirrus on climate change. For other research within ACCRI, see the related articles in this issue. [More detailed information may be found in the subject-specific white paper by Heymsfield et al. (2008).]

CURRENT UNDERSTANDING OF THE MICROPHYSICAL PROPERTIES. Contrail microphysical properties—the ice water content (IWC), the total ice particle number concentration (N_t), ice particle size distributions (PSD), ice particle effective radii, and ice particle shapes—have been reported in a number of published articles over the past three decades (Table 1). Contrail properties just after inception (Table 1, entries T3, T4, and T8) through their development into contrail cirrus an hour or more later (T1, T5, T6, T7, T8, and T11) are represented. Observations include temperatures from –78° (T9) to –30°C (T2). Contrail ice crystals have been captured, preserved, and analyzed (T3) and imaged with high-resolution digital cameras (T5). Most measurements of microphysical properties to date have been made with optical 1D light scattering [e.g., forward scattering spectrometer probe (FSSP)] or 2D imaging probes [e.g., 2D optical array probe (2D-C)].

Ice particle size distributions in young through aged line-shaped contrails have been reported (Table 1), and modeling studies built upon them have elucidated the important microphysical processes. The number concentration (~10^4–10^5 cm^{-3}) and size (fractions of a micrometer) of contrail ice particles at formation are determined by the water activation and ice nucleation properties of plume particles and very high plume cooling rates (Kärcher et al. 1996).

At present, we must rely on models and laboratory studies to account for the initial microphysical and thermodynamic properties of contrails. A large fraction of emitted soot particles, based on the soot emission indices of current aircraft jet engines (Petzold et al. 1999), triggers ice formation in cold conditions or for sulfur-rich jet fuel, although a greater number of liquid plume particles contribute to ice formation in contrails (Schumann et al. 1996). Some further turbulent mixing with ambient air and depositional growth of contrail ice particles then occurs. At this point, the majority of the ice crystals are still small (diameters < 1 μm) and concentrations have decreased to ~10^3–10^4 cm^{-3}. Individual contrail plumes are then captured in the aircraft vortices, and this process suppresses the mixing. Numerical studies (Sussmann and Gierens 1999; Unterstraßer et al. 2008) show that a variable fraction of the initial ice crystals sublimates during the vortex phase, depending on the ambient humidity, temperature, stability, and turbulence.

Observations of the ice crystal shapes during the early phase of contrail formation and beyond are sparse. Observations of contrail crystal growth over a 1-h period have shown that the mean crystal diameters reach about 8 μm, and concentrations resulting from plume mixing are reduced to 10–15 cm^{-3} (T8). Heymsfield et al. (1998) report on the microphysical

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properties of a contrail they sampled for over an hour, from the time of its generation at a temperature of −52°C and with relative humidity with respect to ice (RH_i) reaching 160%. The total concentrations of ice crystals were 10–100 cm^{-3} above 2 μm in maximum dimension and ranged from 10 to 100 L^{-1} above 50 μm in size.

The IWC is an important bulk property of particle ensembles in the upper troposphere and often is used to parameterize cloud optical properties in models. Contrail IWCs have been estimated from the PSDs with approximately a factor of 2 uncertainty. Schumann (2002) summarized most IWC estimates in contrails and fit the temperature (T)-dependent relationship to them as follows:

$$\mathrm{IWC}\ (\mathrm{g\,m^{-3}}) = \exp(6.97 + 0.103T[^\circ\mathrm{C}]) \times 10^{-3}. \quad (1)$$

This curve (see Fig. 1) is a fit to data from −30° to −67°C and to IWCs from 0.001 to 0.07 $g\,m^{-3}$. An alternative (estimate) for the IWC—the potential IWC—relates the IWC to RH_i as

Table 1. Key contrail microphysical measurements. 1D-C: one-dimensional optical array probe; 2D-C; MASP: Multi-Angle Aerosol Spectrometer Probe; MODIS: Moderate Resolution Imaging Spectroradiometer; PI: particle imaging nephelometer; Replicator: continuous impactor-type probe producing ice crystal replicas; TDL: tunable laser diode hygrometer; VIPS: video ice particle sampler, continuous impactor-type probe producing videos of ice crystals; Wire impactor: impactor-type ice crystal replication technique.

Study	Contrail	Key findings	Particle probe
Knollenberg (1972) (T1)	Contrail→cirrus uncinus	Unexpectedly large IWC, contrail evolved into natural cirrus, some crystals developed to > 0.5 mm	1D-C (75 μm–2.175 mm)
Gayet et al. (1996) (T2)	Contrail→cirrus	N_t (> 50 μm) up to 0.175 cm^{-3}, larger than natural cirrus	2D-C (25–800 μm)
Baumgardner and Gandrud 1998 (T3)	Contrail, 210 K	30 s after generation: 200 cm^{-3}, bimodal size distribution; peaks at 0.7 and 2 μm	MASP (0.3–20 μm)
Goodman et al. (1998) (T4)	Contrail	1 mi after generation: N_t~5–10 cm^{-3}, $\bar{D}_v$ = 4–5 μm; crystal habits are predominately plates; shapes formed when crystal D > 0.5 μm	Wire impactor (> 0.5 μm)
Heymsfield et al. (1998) (T5)	Contrail→cirrus	N_t = 10–100 cm^{-3}, $\bar{D}$ = 1–10 μm, contrail visible for > 6 h	MASP (0.3–20 μm), VIPS (20–200 μm), PI (50–1000 μm), 2D-C (50–1600 μm)
Lawson et al. (1998) (T6)	Contrail→cirrus	Crystal habits: columns and bullet rosettes; when time > 40 min, 1–20-μm crystals in contrail core with N_t ~1 cm^{-3}	MASP (0.3–20 μm), PI (50–1000 μm)
Poellot et al. (1999) (T7)	21 contrail clouds	N_t > 10 cm^{-3}, $\bar{D}$ ~10 μm	FSSP-100 (2–47 μm), 1D-C (20–600 μm), 2D-C (33–1056 μm)
Schröder et al. (2000) (T8)	12 contrail flights (sampled up to 30 min from generation)	N_t > 100 cm^{-3}, $\bar{D}$ = 1–10 μm, ice particles spherical	FSSP-300 (0.3–20 μm), FSSP-100 (6–98 μm), 2D-C (20–650 μm), replicator (>2 μm)
Schumann (2002) (T9)	Contrail→cirrus	Compilation of contrail IWC estimates	Many instruments
Gao et al. (2004) (T10)	Cold contrail	Presence of new class of HNO_3 containing ice crystals at T < 202 K	MASP (TDL)
Atlas et al. (2006) (T11)	Contrail→cirrus	Tracked contrails with satellite and lidar	Ground-based lidar MODIS

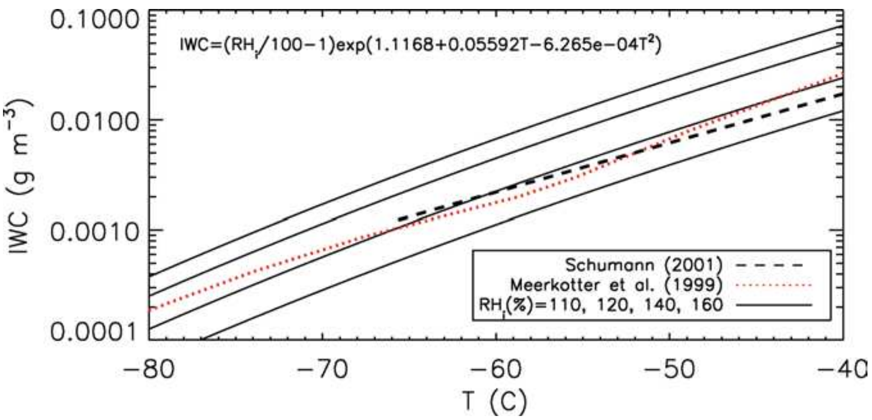

Fig. 1. Contrail ice water content as a function of temperature fitted to observations by Schumann (2002), from the model of Meerkötter et al. (1999) and as given by Eq. (1).

$$\text{IWC (g m}^{-3}) = \rho_a[X_i(\text{RH}_i/100 - 1)], \qquad (2)$$

where ρ_a is the density of air, X_i is the saturation mixing ratio with respect to ice at the ambient temperature, and RH_i is in percent. For a standard atmosphere this result is well described by the relationship

$$\text{IWC} = (\text{RH}_i/100 - 1)^*\exp(a_o + a_1T + a_2T^2), \qquad (3)$$

where $a_0 = -3.4889$, $a_1 = 0.05588$, and $a_2 = 6.268 \times 10^{-4}$, and T is in degrees Celcius. Equations (1) and (3) are obvious approximations because contrails are expected to show substantial variability.

Favorable conditions for contrails to develop cirrus-like properties (Fig. 2) include a largely cirrus-free environment, a sustained growth period for crystals in high supersaturated conditions (see Gierens et al. 1999), and sustained upward vertical motions leading to a deep layer of high ice supersaturation. The development of virga falling from contrails is a manifestation of a favorable environment for growth and the development of contrail cirrus (T1, T4, and T10). With a sustained ice-supersaturated environment, some contrail microphysical properties become similar to those of natural cirrus, for example, concentrations of ice crystals larger than 100 μm in diameter are of the order of 10–100 l^{-1}, and the habits are bullet rosettes (T5).

Soot almost certainly triggers contrail formation given the current soot emission indices. In contrast, the role of soot in forming cirrus ice crystals downstream, without contrail formation or after dissolution of short-lived contrails, is highly uncertain. The main problem is to demonstrate unambiguously that ice nucleates on soot particles from aircraft and not on other atmospheric ice nuclei (IN), including soot from other sources and mineral dust. A prioritized list of uncertainties associated with this problem has been provided and discussed by Kärcher et al. (2007).

Ice formation by black carbon particles, in general, remains poorly understood. It is apparent that some black carbon particles act as IN, because carbonaceous particles have been found to be one of the major types of apparent nuclei of ice crystals formed on aerosols sampled from the background upper troposphere and cold regions of the lower troposphere (Chen et al. 1998; Rogers et al. 2001). The efficiency of black carbon particles acting as IN depends very sensitively, but in unclear ways, on temperature and supersaturation, soot size and surface oxidation characteristics, and the mechanism of ice formation (DeMott 1990; Gorbunov et al. 2001; Diehl and Mitra, 1998; Möhler et al. 2005a,b; Dymarska et al. 2006). Most studies have focused on temperatures warmer than 235 K; some laboratory studies have addressed the ice nucleation properties of soot particles at lower temperatures (DeMott et al. 1999; Möhler et al. 2005a,b), and only a few have carefully quantified the freezing fraction of soot particles on a single particle basis. Unfortunately, the studies most relevant to cirrus formation have used idealized soot particles of unknown relevance to aircraft exhaust soot.

MOVING FORWARD. *Contrail formation.* Current knowledge of contrail formation conditions is sufficient for forecasting contrail occurrence. Crucial parameters that must be known with high accuracy include the ambient relative humidity, the overall aircraft propulsion efficiency, and the number emission index of aircraft soot particles.

Several contrail microphysical properties and processes warrant new measurements and a more complete understanding, in part because of their potential implications for persistent contrail development and subsequent influence on contrail cirrus formation. Most crucial unknowns stem from the extreme difficulty in making the necessary measurements.

Measuring the ice concentration at the onset of contrail formation is one such measurement problem, yet it is important for assessing how well the models represent the physics of the initial formation of contrails. The fraction of ice number and ice mass that survives the vortex phase and the aircraft-specific effects on the vortex dynamics influence subsequent growth of contrail crystals and contrail spread, but these processes are extremely difficult to measure in situ.

Microphysical measurement issues. Recent studies (McFarquhar et al. 2007; Heymsfield 2007) have shown that measurements of small ice crystals can be artificially enhanced as a result of several hundred micron or larger particles either shattering on the inlets or arms of optical spectrometers or bouncing or shattering on upstream surfaces of the measurement aircraft. Because of shattering, previous measurements of the concentrations and sizes of particles in contrails and contrail cirrus are likely unreliable, as are measurements for the natural cirrus itself. Measurement contamination from shattered ice fragments confounds our ability to differentiate contrail cirrus from natural cirrus. New measurements using probes that have been designed to either reduce or eliminate shattering are vital if we are to adequately model contrail crystal formation and growth and the microphysical processes for those contrails that evolve into contrail cirrus.

Exhaust soot particles as cirrus nuclei. Aircraft soot emissions provide a source of enhanced aerosol number concentration in cirrus-forming regions and may perturb cirrus properties and alter cirrus coverage (Hendricks et al. 2005); however, our knowledge of the properties of soot as ice nuclei is extremely limited. Fundamental laboratory studies are required to ascertain what makes certain soot particles more active than others, and what role (short lived) contrail and atmospheric processing might play in making exhaust soot more or less active as cirrus IN. New studies of the ice-nucleating properties of aircraft exhaust and other ambient IN measured in situ for conditions in the cirrus temperature and water vapor regimes are needed. Such investigation could be done independently, but it would be most useful within the context of a measurement effort to convincingly relate the ice activation properties of aerosols to the microphysical composition of cirrus clouds that

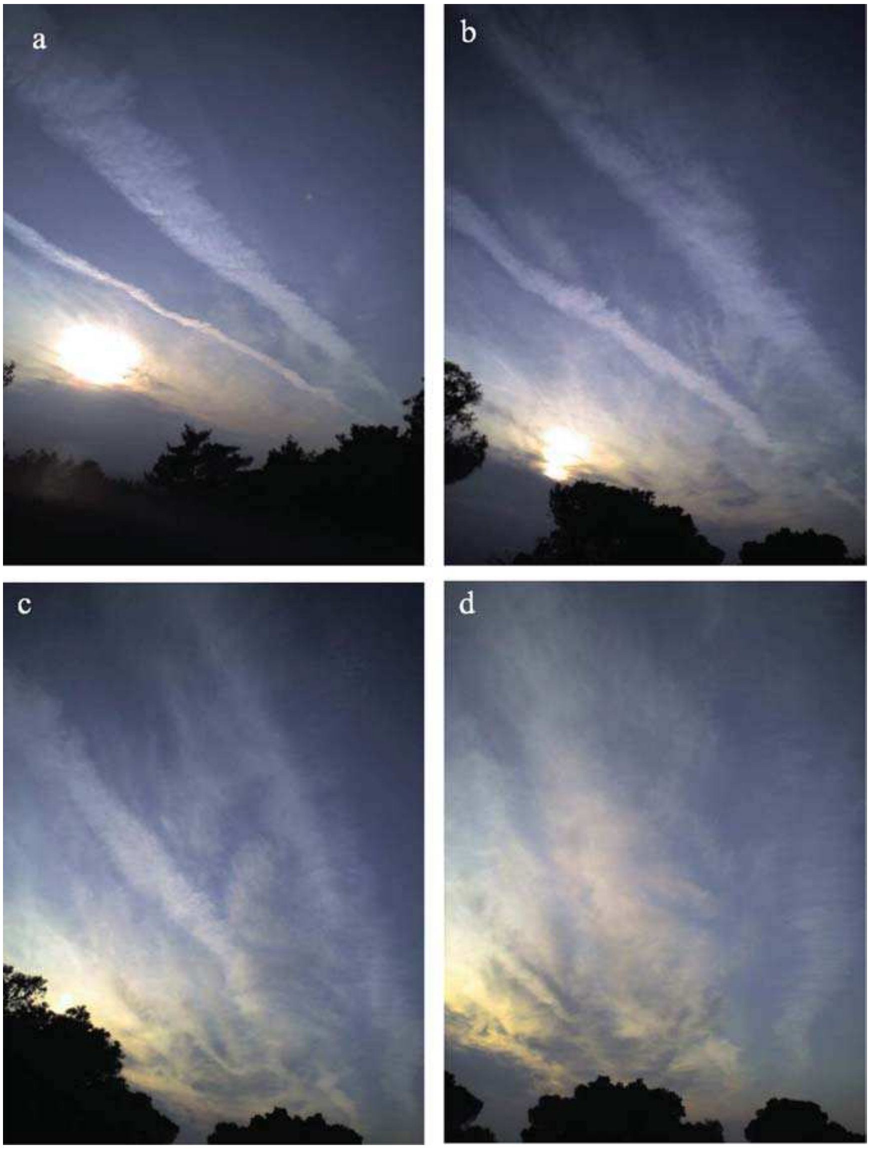

Fig. 2. Photographs of contrail spreading into cirrus taken from Athens, Greece, on 14 Apr 2007 at 1900, 1909, 1913, and 1920 local time (from top left to bottom right). Courtesy of Kostas Eleftheratos, University of Athens, Greece.

form on them. New instrumentation continues to be developed with ongoing improvement applicable to these challenges.

Field campaigns. Reliable models of contrail crystal formation and subsequent growth demand field campaigns that employ new instruments and technologies to measure the detailed microphysical and chemical structure of aircraft exhaust plumes and contrails during their initial development and subsequent evolution into mature systems that disperse and age. Without such improved measurements, it will be impossible to realistically model the development of contrail cirrus. "Closure" experiments are needed to evaluate the sensitivity of cirrus cloud formation and evolution resulting from soot particles emitted by aircraft.

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CLOUD TYPE AND CONTRAIL DEFINITION

In definitions of clouds according to morphology, *cirrus* denotes detached clouds in the form of white, delicate filaments, or white or mostly white patches or narrow bands. These clouds have a fibrous (hair-like) appearance, a silky sheen, or both (WMO 1975). Cirrus forms primarily in the upper troposphere, above ~8 km (25,000 ft), when temperatures are mostly below −35°C, and is entirely composed of ice crystals. Two further genera include cirrostratus, which totally or partly cover the sky, and cirrocumulus, which are small cloud elements in the form of separate or merged grains, ripples, etc. Cirrus clouds forming in situ develop in air that is supersaturated with respect to the ice phase. A wispy, layered cloud that forms at the top of a thunderstorm, termed an *anvil* because of its shape, is cirrus that consists initially of ice debris that spreads outward from the convective parts of the storm. Cirrus can have long lifetimes that are limited by sublimation resulting from subsidence or sedimentation of larger ice crystals.

A condensation trail, or *contrail*, is an artificial, linear cloud created by an aircraft. Jet aircraft cruising in the upper troposphere produce contrails, entirely composed of ice crystals, below about −40°C. Contrails are caused either by water condensation on and subsequent freezing of water-activated soot particles in engine exhaust plumes mixing with drier and cooler ambient air (exhaust contrail), or by water condensation and freezing of ambient particles resulting from the reduction in air pressure above the wing surface (aerodynamic contrail). The majority of the studies deals with exhaust contrails, because the formation processes of aerodynamic contrails have been investigated only recently.

Line-shaped persistent contrails transform into irregularly shaped ice clouds, or *contrail cirrus*, under the action of wind shear and turbulence in ice-supersaturated air. By definition, contrail cirrus is difficult, if at all possible, to distinguish visually from most natural cirrus. The time of transition of contrails into contrail cirrus, controlled by the latter factors, is not well defined. Neither are differences in microphysical and optical properties, hence, the radiative effects between contrail cirrus and natural cirrus. In heavily traveled areas, contrail cirrus merge and form long-lived, cirrus-like cloud decks occasionally covering large horizontal regions. Contrail cirrus may replace natural cirrus or alter natural cirrus properties by competition for the available water vapor.

In cases where no contrails form, soot particles produced by the combustion of kerosene in jet engines age in the decaying aircraft wakes that mix with ambient air and particles. The aging processes may transform exhaust soot particles into heterogeneous ice nuclei capable of nucleating ice crystals at lower supersaturations than the ambient aerosol particles. These soot particles may subsequently form cirrus or modify existing natural cirrus, *soot cirrus*, depending on the dynamic processes setting the stage for the nucleation of ice in supersaturated areas. The ice-nucleating ability of aging exhaust soot particles is not known, and experimental evidence for soot cirrus is lacking; however, soot cirrus may be potentially important. Changes of cirrus cloudiness caused by contrails, contrail cirrus, and soot cirrus together are denoted as *aircraft-induced cloudiness* (Solomon et al. 2007).

Cloud definitions	
Cirrus	Naturally occurring high cloud formed of ice crystals
Contrails	Line-shaped ice cloud created by aircraft
Contrail cirrus	Irregularly-shaped cirrus-like cloud developing from contrails
Soot cirrus	Artificial cirrus cloud forming from aircraft soot emissions in the absence of contrails
Aircraft-induced cloudiness	Changes of cirrus cloudiness caused by contrails, contrail cirrus, and soot cirrus

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