

Available online at www.sciencedirect.com



C. R. Physique 6 (2005) 549-565



http://france.elsevier.com/direct/COMREN/

Aircraft trailing vortices/Tourbillons de sillages d'avions

# Formation, properties and climatic effects of contrails

Ulrich Schumann

Institut für Physik der Atmosphäre, Deutsches Zentrum für Luft- und Raumfahrt (DLR), Oberpfaffenhofen, 82230 Wessling, Germany

Available online 5 July 2005

# Abstract

Condensation trails (contrails) are aircraft induced cirrus clouds, which may persist and grow to large cirrus cover in icesupersaturated air, and may cause a warming of the atmosphere. This paper describes the formation, occurrence, properties and climatic effects of contrails. The global cover by lined-shaped contrails and the radiative impact of line-shaped contrails is smaller than that assessed in an international assessment in 1999. Contrails trigger contrail cirrus with far larger coverage than observed for line-shaped contrails, but still unknown radiative properties. Some model simulations indicate an impact of particles and particle precursors emitted from aircraft engines on cirrus clouds properties. However, the magnitude of this effect cannot yet be assessed. Contrail formation can be avoided only by flying in sufficiently warm and dry air. The formation of contrail cirrus can be reduced by avoiding flights in ice-supersaturated regions of the atmosphere, e.g., by raising the flight level into the lower-most stratosphere. *To cite this article: U. Schumann, C. R. Physique 6 (2005).* © 2005 Académie des sciences. Published by Elsevier SAS. All rights reserved.

## Résumé

**Formation, propriétés et conséquences climatiques des traînées de condensation.** Les traînées de condensation sont des nuages de type cirrus génères par les avions qui peuvent persister et croître jusqu'à produire une couverture nuageuse importante si l'air est sursaturé en glace, et peuvent également conduire à un réchauffement de l'atmosphère. Cet article décrit les conditions d'apparition, la formation, les propriétés et les conséquences climatiques des traînées de condensation. La couverture nuageuse globale et l'impact radiatif des traînées rectilignes sont plus faibles que ce qui avait été évalué dans un rapport international en 1999. Les traînées de condensation déclenchent des cirrus qui ont une couverture beaucoup plus grande que celle observée pour les traînées rectilignes avec cependant des propriétés radiatives encore inconnues. Des simulations basées sur plusieurs modèles montrent l'existence d'un impact des particules et des précurseurs de particules émis par les moteurs d'avions sur les propriétés des cirrus générés par les traînées. Cependant, l'importance de cet impact ne peut pas encore être évaluée. La formation des traînées ne peut être évitée qu'en présence d'une atmosphère suffisamment chaude et humide. La formation des cirrus générés par les traînées peut être réduite uniquement en évitant de voler dans les régions de l'atmosphère sursaturées en glace, c'est-à-dire en augmentant l'altitude de vol jusqu'à atteindre les basses couches de la stratosphère. *Pour citer cet article : U. Schumann, C. R. Physique 6 (2005).* 

© 2005 Académie des sciences. Published by Elsevier SAS. All rights reserved.

Keywords: Emissions; Contrails; Aircraft; Cirrus; Particles; Climate; Mitigation

Mots-clés : Émissions ; Traînées de condensation ; Cirrus ; Particules ; Climat ; Réduction

1631-0705/\$ – see front matter @ 2005 Académie des sciences. Published by Elsevier SAS. All rights reserved. doi:10.1016/j.crhy.2005.05.002

*E-mail address:* ulrich.schumann@dlr.de (U. Schumann).

## 1. Introduction

Contrails are visible line clouds that form behind aircraft flying in sufficiently cold air due to water vapour emissions. Contrails occur both behind propeller and jet driven aircraft and were first observed in 1915. The thermodynamic formation of contrails was first explained in 1941 [1]. Contrails evaporate quickly if the ambient air is dry; they persist, evolve into more extended cirrus clouds and grow in particle size by deposition of ambient water vapour on the ice particles in the contrails if the ambient air is humid enough. Contrails are of importance for aviation because of their visibility, and because of their potential climate impact [2,3]. Several recent reviews discuss the properties of contrails [4–9].

The most essential properties for climate are the changes in regional and global cloud cover, the optical depth, the resultant radiative forcing, and possible effects on air composition and the hydrological cycle. These issues are still not settled. This paper gives an overview on the present state of knowledge.

#### 2. Thermodynamics of contrail formation

According to the Schmidt–Appleman criterion [1,10], contrail formation is expected thermodynamically due to the increase in relative humidity (RH) that occurs in the engine plume as a result of mixing of heat and water vapour between the warm and moist exhaust and the cool ambient air, and as a consequence of the non-linear increase of saturation humidity with temperature, see Fig. 1. When the ambient atmosphere is cold enough, with temperature below a threshold temperature, humidity may reach liquid saturation in the young plume behind the aircraft. Then liquid water droplets form, by condensation of water vapour mainly on soot and volatile particles in the exhaust plume. Saturation with respect to ice is not sufficient for contrail formation [10–15]. Many of the liquid droplets freeze soon thereafter and form ice particles.

Engine exhaust contains water vapour due to the combustion of hydrogen containing fuels with air. Because of high temperature, the relative humidity is low initially. Therefore contrails, like fog forming from breathing people in outside winter air, form only in cold ambient air.

In very humid air, contrails are sometimes observed to form also from wing tip vortices and edges of the high-lift devices at the wings because of local pressure reduction by the strong curvature of air flow around such tips and edges. However, such contrails are observed only rarely, and are of little practical importance.

The threshold temperature for contrail formation from engine exhaust depends on ambient RH and on the ratio G between changes in water vapour pressure and temperature during mixing,

$$G = EI_{\text{H2O}} pc_p / \left[ \varepsilon Q(1 - \eta) \right]$$

and hence on the fuel combustion properties in terms of the emission index  $EI_{\text{H2O}}$  of water vapour and the combustion heat Q, on ambient pressure p at flight level, on the specific heat capacity of air  $c_p$  (ca. 1004 J kg<sup>-1</sup> K<sup>-1</sup>), the ratio  $\varepsilon = 0.622$  of molar masses of water vapour and air, and on the overall efficiency  $\eta$  of propulsion of the aircraft/engine system at cruise [1,11,12]. A Fortran subroutine to evaluate the threshold temperature is available from http://www.dlr.de/ipa/Schumann/Index.



Fig. 1. Mixing lines (dashed) and saturation curve over liquid water (full) in a diagram of partial water vapour pressure e versus temperature T. The mixing lines are plotted for environmental conditions with temperature  $T_e$  in the environment below (point E) and at (point C) the threshold temperature. The gradient of the mixing lines is given by the parameter G (see text). At point M the relative humidity during mixing under threshold conditions is a maximum.



Fig. 2. Threshold temperatures for 0, 40%, 60%, and 100% relative humidity relative to liquid saturation (*RH*) for overall propulsion efficiency  $\eta = 0.3$ , a fuel with water vapour emission index  $EI_{H2O} = 1.223$ , and a combustion heat Q = 43.2 MJ kg<sup>-1</sup>. At temperatures below the line for zero relative humidity, contrails always form, at temperatures above the line for 100% relative humidity, contrails never form. Also shown is the temperature profile of the standard atmosphere versus altitude z.

The overall efficiency of propulsion is the ratio

$$\eta = FV/(m_f Q)$$

between the work rate FV performed by the thrust F of the engine at true air speed of the aircraft V relative to the amount of chemical energy  $m_f Q$  provided by the fuel with specific combustion heat Q at flow rate  $m_f$ . The overall propulsion efficiency depends on speed V and on the state of aircraft operation. The specific fuel consumption SFC =  $m_f/F$ , is often published for cruise conditions by engine manufacturers or can be computed with an engine cycle model so that  $\eta$  can be determined. The average overall efficiency  $\eta$  at cruise was close to 0.2 in the 1950s, near 0.3 on average for the subsonic airliner fleet in 1992, and may reach 0.5 for new engines to be built after 2010 [12].

Fig. 2 shows the threshold temperature of the atmosphere below which contrails form versus altitude for kerosene fuel, for given relative humidity *RH*, and for given overall propulsion efficiency  $\eta$  of 0.3. If the ambient temperature corresponds to the standard atmosphere (-15 °C at surface, lapse rate of  $6.5^{\circ}$ /km up to a tropopause at 11 km, -56.5 °C constant temperature in the stratosphere), aircraft cause contrails within the altitude range 8.2–19 km for liquid saturation (*RH* = 100%, which does not normally occur in this altitude range below  $-40^{\circ}$ C), and 10.2–14 km for zero humidity.

The validity of the Schmidt–Appleman criterion for contrail formation has been verified experimentally in several cases based on visual observation indicating whether a contrail formed or not, together with measured ambient pressure, temperature and humidity and known engine and fuel properties [11–17]. The experiments confirmed that liquid saturation is necessary for contrail formation, and that more fuel-efficient engines with higher  $\eta$  cause contrails to form at higher ambient temperatures [12,16]. Changes in soot particles and fuel sulphur content (FSC) have little impact on the threshold temperature for contrail formation (less than 0.4 K) [17].

### 3. Formation of contrail ice particles

Aircraft engines emit hot exhaust gases containing mainly water vapour and carbon dioxide, and small amounts of nitrogen oxides, hydrocarbons, carbon monoxide, sulphuric oxides (oxidation products of sulphur containing molecules included in aviation fuels), some organic material (partially condensable), and chemi-ions (charged molecular clusters formed during combustion in the engine), soot (organic and black carbon) and possibly small metal particles (from mechanical erosion) [4].

Compared to thermodynamics, particle emissions play a secondary role in contrail formation. If the atmosphere is cold enough, a contrail will form even for zero particle emission from the aircraft engines because of condensation nuclei entrained into the exhaust plume from the ambient air. The upper troposphere always contains high concentrations (typically  $10^2$  to  $10^4$  cm<sup>-3</sup>) of condensation nuclei [18–20]. However, the engine exhaust usually contains combustion particles (mainly soot and small volatile particles) at far higher concentrations (up to  $10^9$  cm<sup>-3</sup> at the engine exit) [21], which may act as condensation nuclei in addition to particles mixed in from ambient air.

Soot particles emitted from aircraft engines contain fuel impurities (including sulfur compounds) and have experienced strong oxidation before leaving the combustion chamber and hence are mostly hydrophilic in contrast to pure black carbon particles [22–24]. By coating with sulphuric acid, soot particles may become even better suited as condensation nuclei [22]. The number and size of small liquid particles acting as condensation nuclei depends also on the presence of chemi-ions which support coagulation of the initially forming very small particles [4,25]. The amount, size and composition of emitted and entrained particles influence the number of ice particles in the contrail.

The ice particles form either by homogeneous freezing in the bulk of the liquid particles (the freezing probability increases linearly with the volume of the droplets and exponentially with decreasing temperature) or by heterogeneous freezing due to surface contact with solid particles. Because of condensing sulphuric acid, the number of liquid particles and the coating of soot particles increases with the FSC, but ice formation seems to occur at uncoated soot particles better than at coated soot particles [26,27]. Aircraft also emit condensable organic material which contributes to volatile particles in the exhaust plume, but such particles seem to be less suited for homogeneous ice formation than sulfuric acid [28]. Although measurements performed so far were limited in accuracy because measurement of ice particles at such high concentrations is difficult, it appears that the number of ice particles in young contrails is close to the number of soot particles and is only weakly dependent on FSC, see Fig. 3 [17]. This is consistent with model studies by Kärcher et al. [14], which also suggest that the number of ice particles increases more strongly with FSC at lower ambient temperatures.

Knowledge about contrail particle properties was summarised in [29,9]. An extensive in situ study of young and aged contrails in transition to cirrus has been presented by Schröder et al. [30] who measured the concentration of ice particles in



Fig. 3. Number of soot particles and ice particles per kg of fuel in contrails versus fuel sulfur content (FSC), behind various aircraft: ATTAS (squares), B737 (circles), and A310 (diamonds). The full symbols with dashed lines approximate the mean soot particle emission indices measured for the three aircraft in non-contrail plumes. The grey symbols with error bars denote the number of ice particles formed per kg of fuel burned in contrails for the B737 and the ATTAS (adapted from [17]).



Fig. 4. Ice crystal concentrations in contrails per fuel mass burned versus contrail age as derived from observations behind 12 cruising aircraft of different type by Schröder et al. [30]. The normalised concentration is computed from  $EI = cN(t)/\rho$ ; with  $N(t) = 7000(t/t_0)^{0.8}$  [31], for measured concentration *c* and plume age *t*. For cases without know pressure and temperature at flight level, the density  $\rho$  is estimated to be 0.4 kg m<sup>-3</sup>. In the sequence of contrail ages, the aircraft types were: VFW614, A310, B757, A310, B737, DC9, A300, unknown, B737, 3 unknowns.

contrails at various distances behind cruising aircraft. For typical dilution [31], the measured concentrations can be normalised by the amount of fuel consumption to provide quasi an emission index of ice particles for persistent contrails as a function of age, see Fig. 4. In persistent contrails the number of particles formed per unit mass of fuel is of the order  $10^{14}$  to  $10^{15}$  per kg of burned fuel. The data show no significant trend with age.

#### 4. Persistent contrails

Contrails are short-lived when the ambient air is dry, with relative humidity below saturation over an ice surface so that the ice particles forming in the contrail evaporate. Often, contrails disappear after seconds to minutes, between one and several ten kilometres behind the aircraft.

Contrails persist if the ambient humidity is larger than saturation humidity over ice surfaces (relative humidity over ice RHi larger than 100%). In such ice-supersaturated air masses, the ice particles within the contrails grow by deposition of water vapour molecules from the ambient air. Contrails may persist as long as the ambient air in which the contrail forms stays ice-supersaturated [32].

Although the first detection of ice-supersaturation in the upper troposphere dates back at least to the 1940s [33–35], it was generally believed that ice-supersaturation occurs only exceptionally, and that clouds of ice particles form soon after the humidity exceeds saturation. Most weather and climate models still assume that cirrus clouds form immediately when the humidity reaches ice saturation. Examples are the operational weather prediction model of the European Centre for Medium-Range Weather Forecasts and climate models derived from it [36].

However, evidence for ice supersaturation occurring in the free atmosphere is available to observers in terms of cirrus fallstreaks that grow while falling through supersaturated air layers [37] and, in fact, by persistent contrails [35]. Although contrails are often observed to form in or near cirrus clouds [38,30], contrails can form and persist even when no cirrus clouds are around [9].

Contrails form and persist also in weakly ice-supersaturated air. However, the formation of cirrus requires higher relative humidity than for contrail persistence: Ice particles form from the abundant small droplets by homogeneous freezing only at high relative humidity with respect to ice-saturation, at RHi of the order 145 to 165% or higher [39]. Also the formation of ice particles by heterogeneous nucleation often requires RHi of 110% or more [27] (with few exceptions such as desert dust particles). Often, RHi is large enough to let contrails persist and develop into cirrus but not large enough to let cirrus clouds form naturally. Since contrail persistence requires at least ice saturation, a sky full of contrails but without natural cirrus shows that cases occur with humidity above ice-saturation but below the threshold for cirrus formation.

The existence of ice-supersaturated air masses in the free atmosphere has been confirmed by several airborne measurements in the last decade with various types of hygrometers on aircraft [40–46], with carefully calibrated and corrected hygrometers on radiosondes [47], and with satellite data [48–50].



Fig. 5. Relative humidity with respect to liquid water versus ambient temperature. The thin full curve denotes the relative humidity for ice saturation. The dashed line denotes liquid saturation. The symbols denote measured data as derived from a frostpoint hygrometer (Ovarlez et al. [43]) and temperature sensors (extended from Schumann et al. [44]). The pair of curves with various line notations represent the relative humidity for constant water vapour content at various temperatures.

As shown by Fig. 5, more than 40% of all data collected during a measurement campaign over the North Atlantic [43, 44] were taken in ice-supersaturated regions. Measurements on modern airliners [51] indicated that such aircraft fly in ice-supersaturated air masses about 15% of the flight time [42].

Ice-supersaturated air masses form in regions with rising air motion and are partially filled with cirrus clouds [52]. Regions with ice supersaturation have been found with horizontal extensions of the order 150 km [53] and vertical extensions of about 500 m in the mean [47]. Supersaturation is also observed at least occasionally in the lower stratosphere up to the hygropause in the polar winter and also, rarely [47,54], in the lower stratosphere at mid-latitudes and in the tropics up to a about one kilometre above the local tropopause [55,56].

The area size of the regions with ice-supersaturated air masses defines the potential contrail cover which would appear if aircraft were to fly everywhere and at all times. Global distribution maps of ice-supersaturated regions have been produced from satellite data [48] and from analysis of meteorological data [57]. Such analysis suggests that the global average contrail cover (partially overlapping with cirrus clouds) could approach 16% [57]. Over Europe, the potential contrail cover reaches 12%, which is consistent with estimates derived from satellite observations of the size of regions with clusters of persistent contrails [58] and by in situ humidity measurements [42,53]. If these numbers were realised it would mean a very large change in high cloudiness. For comparison, the mean high cloud (cirrus) cover at northern mid-latitudes is about 20–30% according to different observations [59,60]. Subvisible clouds with optical depths below about 0.03 are even more abundant [61].

#### 5. Contrail dynamics

The initial development of contrails is driven by the exhaust gases, ambient temperature and humidity, and by the jet and vortex dynamics generated in the wake of an aircraft [62,63]. The structure of contrails depends on the air flow behind an aircraft depending on the size, weight, speed, number and position of engines, and geometry of the aircraft. Jet turbulence and the early wake dynamics can have a strong influence on the properties of persistent contrails even at late times [64–67]. Because of thermodynamics, contrails form first at the outer boundaries of the hot exhaust jets mixing with the cool ambient air. The total ice crystal number can be significantly reduced due to adiabatic compression resulting from the downward motion of the vortex system [65,66]. Small and large transport aircraft may produce persistent contrails of similar size, even though the fuel consumption may differ by a factor of five [66]. Under subsaturated conditions, contrails of 2-engined aircraft evaporate mostly already during the jet phase (< 20 s), contrails of 4-engined aircraft often survive until the end of the vortex phase (ca. 2 min) [68]. After decay of the wake vortex, the further development depends on ambient humidity, wind shear, turbulence, and stratification, and possibly radiative cooling [69]. Turbulent mixing causes an increase of the contrail diameter mainly horizontally because of anisotropic turbulence in a stably stratified atmosphere with weaker vertical than horizontal turbulent motions [70–72].

At present, only a few exploratory studies have dealt with the later stage of the persistent contrail dynamics which depends on the mesoscale atmospheric flows with rising or sinking motions of turbulent or wavy character and on shear, radiation and ice particle sedimentation. A vertical shear in the wind perpendicular to the contrail causes a contrail spread which may reach several kilometres within hours [73–76].

#### 6. Line-shaped contrail occurrence

It is often difficult to decide whether a certain cirrus has formed naturally or resulted from aged contrails [77–79]. Contrail clouds have been identified and discriminated from natural cirrus clouds in satellite images based on the linear appearance of these contrails, see Fig. 6, for example. The fractional cloud cover of persistent contrails was estimated first by Bakan et al. [80] by manually investigating satellite images from an Eastern North Atlantic and Western European region  $(30^{\circ}W-30^{\circ}E, 35^{\circ}N-75^{\circ}N)$ . They found an average cloud cover from contrails in this region of 0.5% during daytime (about 0.37% in the daily average), possibly overestimating the real cover. Later analysis for Central Europe (were traffic density is higher) partly overlapping with the region considered by Bakan et al. [80] gave smaller cover values [58,81].

Mannstein et al. [58] developed a fully automated method for line-shaped contrail detection. Much care was taken to avoid false identification of contrails or underestimation of contrail cover over surfaces with strongly inhomogeneous surface temperatures [81–83]. Such methods have now been used to analyse the contrail cover for several geographical areas: Central Europe [81,84], Eastern North Atlantic [80], Southern and Eastern Asia [85], North America [83], and the Eastern North Pacific [82].

The day-time mean contrail cloud cover over Central Europe  $(8^{\circ}W-23^{\circ}E, 42^{\circ}N-56^{\circ}N)$  during the years 1995–2000 is 0.75%, see Fig. 7 [84]. This six year analysis confirms results of Meyer et al. [81] from only 2 years of similar data. The night-time contrail coverage in that region was found approximately one-third compared to that of daytime cover, in rough agreement with traffic variations, so that the daily average is slightly larger than 0.5%.



Fig. 6. Contrails over the Atlantic west of Gibraltar in a scene of METEOSAT Second Generation (METEOSAT 8; data provided by EUMET-SAT) at 14:30 UTC March 3, 2004, processed by W. Krebs, L. Bugliaro and H. Mannstein, DLR (personal communication, 2005).

For extrapolations to the globe and for future traffic scenarios, the mean global cover by linear contrails has been computed using temperature and humidity data from numerical weather analysis, and aircraft fuel (or flight distance) inventories estimating the amount of air traffic in a global model grid box [57,86]. Since the meteorological analyses available restrict relative humidity over ice (RHi) to values below 100% (viewed as values that are representative for a larger spatial region such as a global model grid box), and since the local values may be higher than the mean grid box value, the contrail analysis assumes that persistent contrails form with growing cover amounts already between about 60% and 100% RHi in regions where the Schmidt–Appleman criterion predicts contrail formation. This kind of analysis provides also an estimate of the regions covered with ice-supersaturated air masses (the potential contrail cover). In connection with the air traffic data and calibration to satellite observations it provides estimates for the real contrail cover.

The first such analysis by Sausen et al. [57] was calibrated to match preliminary contrail cover estimates in the 'Bakan' region west of Europe [80]. With this normalisation and for fuel consumption as in 1991/1992, the calculated global averaged contrail cover amounted to 0.087%. Taking more recent satellite observations and accounting for the smaller cover during night, the global line-shaped contrail cover averaged over day and night amounts to about 0.07% [87].

Gierens et al. [86] computed that the mean contrail cover could reach 0.25% in 2015 and between 0.26% and 0.75% in 2050, depending on the scenario for the increase in air traffic. The increase in contrail cover is stronger than for total fuel use alone, partly due to more efficient engines ( $\eta$  up to 0.5). While these estimates [86] were performed for a static climate, Marquart et al. [87] considered a changing climate when estimating the fractional contrail cover. They found smaller values than in [86], mainly due to fewer contrails in a warmer climate in the future caused by the increase in greenhouse gas concentrations.

# 7. Cirrus induced by aviation

The cloud change induced by aircraft is larger than that indicated by line-shaped contrails for two reasons:

(i) water vapour and particles emitted from aircraft engines induce contrails which grow to larger-scale clouds (*contrail-cirrus*) if the ambient atmosphere is so humid that the humidity exceeds ice-saturation;



Fig. 7. Annual mean cover by contrails over central Europe  $(10^{\circ} W \text{ to } 30^{\circ} \text{E and } 38^{\circ} \text{N} \text{ to } 62^{\circ} \text{N})$  at noon time averaged over the six years 1995 to 2000, together with typical upper air traffic routes from EuroControl, as derived by Meyer et al. [84]. The daily mean contrail cover is 0.55%.

 (ii) particles emitted from the aircraft change the concentration of particles in the atmospheric *aerosol* that influence cirrus formation over the whole life-time of the particles formed.

The formation of *contrail-cirrus* is clearly visible to ground observers and observations by satellites from space [77–79]. However, modelling and prediction of contrail cirrus for observable cases is still in its beginning [88]. Proper models and validation data for such studies, including the state of the atmosphere, at scales comparable to the size if supersaturated regions are still to be provided.

No conclusive observational evidence exists for an impact of aviation *aerosol* on cirrus properties. It is to be expected that aviation aerosol and aerosol precursor emissions may impact the upper tropospheric aerosol over their entire life cycle, which may last over a time scale of up to a few weeks depending on season and altitude. It is also to be expected that aviation aerosol acts as ice nuclei both by homogeneous and heterogeneous processes. In particular, soot particles originating from aircraft exhaust may act as efficient heterogeneous ice nuclei [22,57]. Aviation aerosols may trigger the formation of clouds long after the emission, when the background atmosphere has changed to a state allowing cloud formation (supersaturation). Aircraft-induced aerosols can modify the micro-physical properties of clouds, change cloud particle sizes and forms, and the number of cloud particles [89,90]. The result of such modification may include a change in the precipitation rate, in cloud life time, and in cloud radiative properties. A quantification of the impact of aviation aerosol on cirrus properties is subject of ongoing research.

A correlation between aviation soot and cirrus particle concentrations has been observed in cirrus only in one case study, apparently in young persistent contrails [91]. The potential for a connection between aerosols and cirrus has been found in experiments which have shown differences in aerosol and cirrus particle concentration in clean and polluted air masses [20,92–95]. However, the contribution of aviation emissions to cirrus formation in the atmospheric aerosol has not yet been observed at ages beyond about one hour, nor has the formation of cirrus been documented which forms from aviation aerosol without

presence of a contrail. The potential for an impact of aviation aerosol on cirrus has been shown in still tentative numerical simulations of soot concentrations and ice particle formation [96,97].

The few simulation studies performed so far show only very small impact of aviation emissions on the soot mass concentration in the free atmosphere but show significant changes in the number density of soot particles in the atmosphere [97]. For cirrus formation, the number of soot particles may be more important than their mass. The ice formation processes are very complex and not yet finally understood [5,6,92,98,99]. The changes in concentrations of ice nuclei (such as aircraft soot) may cause an increased cirrus cover but may also cause a reduced cirrus cover, so even the sign of this effect is presently uncertain [100].

Some long-term ground-based and satellite-based observations suggest increases in cloud cover correlated with increased air traffic [101–104,50]. Boucher [101] used routine reports of meteorological observers on the ground (on ships or on land) and found an increase of cirrus occurrence (not of cirrus cover) which correlates with the fuel consumption by aviation. The studies by Minnis et al. [102], Zerefos et al. [103] and Stordal et al. [104] were based on 16 years of cloud data from the International Satellite Cloud Climatology Project (ISCCP) [59], using different statistical approaches (and partially additional data). The observed trends over Europe, possibly due to aircraft, amount to between 1.3 and 2% /decade. Stubenrauch and Schumann [50] used 9 years of objective data from the TOVS sensor, a multispectral infrared and microwave sensor which provides vertical profiles of humidity and the cloud amount. Trends of seasonal mean effective high cloud amount are analysed in regions with high and low air traffic density. For a more direct attribution of aviation impact, they use TOVS derived humidity and temperature to identify situations with sufficiently cold and humid air masses favourable for contrails. In regions with especially high air traffic density, a significantly stronger increase of effective high cloud amount is found for situations favourable for contrails than for all situations. In situations favourable for contrails, which occur in about 5 to 10% of all situations, the cloud amount increased by about 5% per decade over Europe and the North Atlantic flight corridor. When averaged over all situations regardless of air mass humidity, the increase amounts to 0.2% per decade in regions with high air traffic densities (Europe and North Atlantic flight corridors). Hence, compared to earlier studies, they find smaller trends but their results may underestimate contrail cirrus because of the low vertical resolution.

From all these studies, the total cirrus cover increase over a, say, two decade growth period may amount to 0.4 to 4%, which is 1 to 8 times larger than the line-shaped contrail cover [81]. These observations suggest that air traffic is responsible for observed cirrus changes, and some of these studies may provide more direct evidence for this attribution than others, but none of them can prove this attribution. Moreover these observations do not allow us to decide on the relative importance of the contrail-cirrus and the aerosol effects. The satellite measurements are unable to detect optically thin cirrus (optical depths below about 0.1), but these thin ice clouds are expected to be particularly susceptible to changes in the concentrations of heterogeneous ice nuclei including aircraft exhaust [100].

A more direct identification of cirrus induced by air traffic has been provided by Mannstein and Schumann [105]. They correlate satellite observations of cirrus with simultaneous data on air traffic movements. The analysis shows that cirrus cover is systematically larger in regions with high air traffic than in regions with low air traffic. The correlation between cirrus cover and traffic data increases when one compares air traffic with cirrus about one hour later. This is a strong indication that the additional cirrus cover is caused by spreading persistent contrails. The study finds that the contrail-cirrus cover over Europe is about ten times larger than the line-shaped contrail cover. The study also shows a direct correlation between air traffic density and reduced infrared radiation from Earth to Space.

#### 8. Radiative forcing by contrails

Contrails, similar to thin cirrus, reduce the amount of short wave radiation reaching the Earth (albedo effect) and reduce also the long-wave radiation leaving from Earth to space (greenhouse effect) [4,29]. Contrail cirrus impacts solar radiation mainly as a function of the optical depth and impacts infrared radiation mainly as a function of emissivity and temperature. The emissivity of cirrus is close to about half the optical depth in the solar wavelength range. The optical depth depends on particle properties and ice water content.

The value of the global mean contrail optical depth  $\tau$  (in the solar range near 550 nm wavelength) of persistent line-shaped contrails is still uncertain, possibly between 0.15 and 0.25 as suggested by Minnis et al. [102]. From lidar observations [38,106, 107],  $\tau$  was estimated between 0.05 and 0.5 with best estimate 0.3 [29,108,4]. More recent satellite data analyses and model studies reveal  $\tau$  near 0.1 over Europe [81,109]. Larger  $\tau$  values of 0.2 to 0.26 were found over the USA [102]. Ponater et al. [109] explain the different findings on optical depth in a model study by the different climatological conditions in various regions of the Earth. They compute cover and optical properties of contrails using a parameterisation similar to those for cirrus clouds within a global circulation model, including the Schmidt–Appleman criterion. With respect to cover computations, the method is the same as that used in Sausen et al. [57], but uses the meteorological fields computed by a global climate circulation model, and also computes the ice water content and radiative properties of contrails. The compute globally mean optical depth

of contrails is 0.15. The optical depth is variable and smaller over Europe than over the USA because of less water vapour for condensation, presumably because of lower temperature of the European atmosphere.

So far, the radiative forcing (RF) [3] by contrails was computed assuming the contrails to be geometrically and optically thin plane parallel homogeneous cirrus layers in a static atmosphere [110,29,108]. The computations reveal a positive RF at the top of the atmosphere during night and day at least in many cases with thin cirrus [29]. Negative RF values occur for thick cirrus with optical depth above 10 or ice water contents above 10 g m<sup>-2</sup> [111,112]. Such large values are unlikely to occur in persistent contrails, in which the optical depth is usually below 1 and the ice water content below 5 g m<sup>-2</sup>. (This may be different for contrail cirrus.) Negative RF values occur, even for thin cirrus, over dark and cold surfaces such as over the North Atlantic in winter [29,81].

Mainly as a consequence of the different values for contrail cover and optical depth, global mean RF estimates have been reported which differ by a factor five: For 1992 air traffic conditions, Marquart et al. [87], Myhre and Stordal [113], and Minnis et al. [108] have yielded values of  $3.5 \text{ mW m}^{-2}$ ,  $9 \text{ mW m}^{-2}$ , and  $17 \text{ mW m}^{-2}$ , respectively. The latter was used for IPCC [3]. Minnis et al. [108] assume a global contrail cover of 0.1% for 1992, an optical thickness of 0.3, contrails at 200 hPa altitude, and hexagonal ice particles. Myhre and Stordal [113] use the same optical depth and global contrail cover but find smaller RF values because of different approaches for the daily traffic cycle, the scattering properties of ice particles, and contrail altitude. Marquart et al. [87] normalise the computed contrail cover by reference to more recent satellite observations [81] implying a smaller global contrail cover (0.05 to 0.07% for 1992); they compute smaller optical thickness values [109], including the daily cycle, more proper altitude distributions of the contrails, and improved radiative transport algorithms [114]. Their RF result is five times smaller than the value used in the IPCC assessment. The global distribution of radiative forcing as computed by Marquart et al. [87] is shown in Fig. 8 [115]. It should be noted that the net RF is the difference between two large values:



Fig. 8. Contrail radiative forcing for 1992 as computed by Marquart et al. [115] using an air-traffic scenario, contrail cover deduced from traffic and climate model simulations, and radiative transfer calculations. From top to bottom: longwave, shortwave and net radiative forcing (mean values:  $4.9 \text{ mW/m}^2$ ,  $-1.4 \text{ mW/m}^2$ ,  $3.5 \text{ mW/m}^2$ ); left: January; right: July.

the large, mostly negative RF in the solar range and the large positive terrestrial RF. Hence, any small error in either of them has large impact on the computed net effect. Contrail longwave RF derived from satellite data appears to be much larger than computed theoretically [82]. Moreover, the RF values are computed for 1992. Air traffic has increased considerably since then and is expected to increase further.

For contrail cirrus, no reliable estimate of the optical properties and of the radiative forcing exists. The IPCC estimate of an upper bound of radiative forcing of 40 mW m<sup>-2</sup> by aviation induced cirrus changes is based on the assumptions of 0.2% global additional cirrus coverage with an optical thickness of 0.3 (same as for line-shaped persistent contrails) [4]. Both assumptions are very uncertain [3,4,104]. Mannstein and Schumann [105] find that the cover by contrail cirrus is about ten times larger than the cover by line-shaped contrails. For the same optical properties, this would imply a radiative forcing ten times larger the RF value from line-shaped contrails. However, the optical properties of the contrail cirrus are likely different from that of line-shaped contrails. The radiative forcing depends nonlinearly on the optical depth. It increases about linearly for small optical depth values, reaches a maximum in between 2 and 5 and may be negative for optical depth values larger than 10 [29]. It is conceivable that contrails within cirrus enhance the optical depth of existing cirrus to values for which the RF becomes negative. Hence, a reliable estimate of the RF by contrail cirrus cannot be given without knowing its optical depth. We plan to use analysis of the change in visible and infrared radiation related to air traffic density and contrail cirrus in the future to close this gap [105].

### 9. Climate impact of contrails

The climatic impact of contrail cirrus is not known. Besides radiative forcing by contrails, contrail cirrus, and changed cirrus, climate may also be impacted by changes in air composition due to reactions on the surface or inside of the induced cloud particles and by changes in the hydrological cycle, e.g., by particle sedimentation of water in the upper troposphere or by changes in precipitation. Only preliminary estimates of some aspects are available so far, mainly for radiative forcing by line-shaped contrails [4].

For 1% additional cirrus cloud cover regionally (optical depth 0.28), a regional surface temperature increase of the order 0.1 K was expected from a regional study by Strauss et al. [110]. For 1% additional cirrus cloud cover globally (optical depth 0.33) a global circulation model coupled to a mixed layer ocean model computed 0.43 K global warming (Rind et al. [116]). Ponater et al. [117] find a smaller climate feedback from contrails than for CO<sub>2</sub> increases in their climate model: the equilibrium response of surface temperature to radiative forcing from contrails is 0.43 K/(W m<sup>-2</sup>) while 0.73 K/(W m<sup>-2</sup>) for CO<sub>2</sub> [117]. For a global contrail cover of about 0.06% and 0.15%, with mean RF of 3.5 mW/m<sup>2</sup> and 9.8 mW/m<sup>2</sup>, in 1992 and 2015, respectively (optical depth between 0.05 and 0.2 depending on region and season) [85], the computed transient global mean surface temperature increase until 2000 amounts to about 0.0005 K in this model [117].

A far larger climate impact has been deduced by Minnis et al. [102], who have analysed a trend in cirrus clouds of about 1%/decade over the continental USA between 1971 and 1995, which they attribute almost exclusively due to air traffic increase during the period. Assuming an optical depth of 0.25 this increase of high clouds was calculated to induce a global mean RF of 25 mW/m<sup>2</sup> at maximum and a tropospheric temperature response of 0.2 to 0.3 K/decade in the region of the forcing, which would explain practically all observed warming over the respective area between 1975 and 1994. However, Shine et al. [118] and Ponater et al. [117] point out several simplifications in this study and find about two orders of magnitude smaller temperature changes.

Travis et al. [119] claim observable increases in the daily temperature range (DTR) due to reduced contrails in the three days period of 11–14 September 2001, when air traffic over parts of the USA was reduced. They report that the DTR was 1 K above the 30-year average for the three days grounding period, which was interpreted as evidence that jet air-craft do have an impact on the radiation budget over the USA. However the DTR in 1982 was also nearly 1 K above the average [120], certainly for other reasons. Hence, the statistical significance of the data is too weak for strong conclusions. Travis et al. [121] support their hypothesis by analysis of the spatial variations of the DTR and of minimum and maximum temperature, and compute an estimate of the contrail cover that would have occurred under normal traffic conditions. The potential contrail cover appears to be related to the observed variation in DTR. However, a quantitative model which relates the DTR to the change in contrail cloud cover is not provided, and other reasons may be responsible for the observed DTR variations. Kalkstein and Balling [122] analyse the air-mass and weather conditions over the USA following the period after 9 September 2001, in relation to the observed patterns in the temperature range. They also find a higher-than-average DTR shortly after this period, but unusually clear weather in that region could also explain the observed DTR.

## 10. Mitigation options

The main contrail induced climate problem identified so far is the greenhouse effect of contrail-cirrus. Short-lived contrails are less a problem because of very small global area coverage [29]. If short contrails are to be avoided for other reasons one has to fly in sufficiently warm and dry atmospheres, as indicated by the Schmidt–Appleman threshold temperature. The introduction of more efficient aircraft engines does not solve the problem of contrail formation; just the opposite is true, because the threshold temperature increases with increased overall efficiency  $\eta$  of the engines [12]. An increased efficiency  $\eta$  is desirable of course for economic and climate reasons by reducing the fuel consumption and reducing emissions of carbon dioxide (CO<sub>2</sub>).

The usage of cryogen fuels instead of kerosene also increases the frequency of contrail formation for constant air traffic because the threshold temperature increases with the hydrogen content in the fuel and liquid hydrogen driven engines emit 2.5 more water vapour mass than engines using conventional fuel for the same energy content. Hence, such aircraft cause contrails over a larger range of altitudes than kerosene driven aircraft [11]. The potential contrail cover would be about 30% larger if all aircraft were driven by liquid hydrogen instead of kerosene [123]. Still, the climate impact may not be much different: Because of a cleaner exhaust with fewer particles available to form ice particles, the contrails would be optically thinner if cryogenic fuels were to be used instead of kerosene [11,123–125]. This effect was not taken into account in an earlier study [126].

The formation of contrail cirrus induced from persistent contrails can be reduced by reducing flights in ice-supersaturated air masses. Some authors [127,128] have suggested flying generally lower to avoid contrail formation. However, drastic flight altitude reductions might be necessary if contrails are to be avoided to a large percentage. The global annual mean contrail coverage may be reduced by 45% for a 6000 ft (1830 m) lower cruise altitude [129]. However, this would imply an increase in fuel consumption (and emissions of  $CO_2$ ), nitrogen oxides emissions [130], longer flight times, and more work load for air traffic control [127,131].

Since atmospheric regions with ice supersaturation are not very thick (typically 500 m with large variance) [47], it often would suffice to fly a few hundred meters higher or lower to avoid such regions [132]. However, in order to do so, one needs information on the occurrence of such regions ahead of flight. This forecast problem (using weather predictions and traffic predictions) is not yet solved and requires meteorological models with accurate representation of ice supersaturation and suitable observational data (e.g., water vapour and contrail observations from airliners).

Contrail-cirrus could generally be avoided if flying slightly higher in the extratropical lowermost stratosphere. Flying higher generally would cause more contrails at lower latitudes and in the tropics [57,129]. However, for long range traffic in the extratropics, flying a little higher than today, in the lower-most stratosphere just above the tropopause, may be recommendable both under climate and aviation aspects. This option should be considered if contrail cirrus dominates the climate impact by aviation and if the side-effects such as ozone changes are sufficiently small. The stratosphere is usually dry so that formation of contrail cirrus in this region is avoided. Aviation can be performed efficiently at such altitudes with the potential for reduced fuel consumption and nitrogen oxides emission. The mid-latitude lower-most stratosphere appears to be only weakly sensitive to ozone changes from nitrogen oxides emitted by aircraft [3,130]. Some authors point to the potential of ozone loss by chlorine activation at contrail and cirrus ice particles in the lowermost stratosphere by heterogeneous chemistry [4,6,133,134]. However, high humidity in the lower-most stratosphere results from transport of humid tropospheric air masses into the lower stratosphere [135] containing low amounts of inorganic chlorine [54]. Three-dimensional model studies suggest ozone to be reduced by not more than a few percent from heterogeneous reactions on subvisible cirrus from all sources in the tropopause region and this decrease may be of comparable magnitude as ozone formation by nitrogen oxides emissions from commercial air traffic [133]. Since aviation causes only a small fraction of all cirrus, the contrail cirrus impact on ozone may also be small.

Reduction of particle and particle precursors from aircraft engines might provide a further mitigation option. Aircraft emissions contribute to condensation and to ice nuclei which change the formation and optical properties of contrail cirrus and of cirrus in general. There is now sufficient evidence (see Fig. 3) to expect that a decrease in soot particle emissions does decrease the number of ice particles forming in contrails [17]. The experimental evidence is weak but models imply that the number of ice particles formed increases with the sulphur content of aviation fuels [14]. As a consequence, the optical depth of line-shaped contrails decreases with the reduction in the amount of particle emissions. The climatic impact of such a change has not yet been fully assessed, but from the studies on contrails with different fuels, a reduced RF is to be expected if the number of ice particles in contrails gets reduced [98,123].

#### 11. Conclusions

Many aspects of contrail formation are well understood. Contrails form for thermodynamic reasons when the ambient air is cold enough. Persistent contrails form in ice-supersaturated air masses. In such cases contrail cirrus often form where no cirrus would form otherwise because ice supersaturation is often too low for natural cirrus particle nucleation. Airliners fly on average about 15% of their time in ice-supersaturated air masses. Because of rather small vertical extent, the fraction of flights

that occurs within ice supersaturated regions could be reduced considerably by slight variations of flight altitude if the actual weather conditions would be known precisely enough. Contrail cirrus forms very rarely for flights in the lower stratosphere above the tropopause. The region just above the tropopause may be also only weakly sensitive in terms of ozone changes to nitrogen oxides emissions, but this needs further studies. The radiative forcing by line-shaped contrails appears to be about five times smaller than assessed in IPCC [3] mainly because of smaller contrail cover and smaller optical depth. The uncertainty in this result is still large. For example, the time lag between contrail cover and traffic is not yet taken into account in these studies. Recent findings suggest that the cover by contrail cirrus is about ten times larger than the cover by line-shaped contrails but cannot be determined yet because of unknown optical properties of the changed cirrus. The impact of aviation induced aerosol (mainly soot) on cloud formation is still not quantified. The number of ice particles formed in contrails and their climate impact may be reduced by lowering soot emissions and sulphur content of aviation fuels, but the efficiency of such a measure has not yet been quantified. Contrails can have caused only a small part of the observed changes in surface temperature. Findings of higher-than-average daily temperature range during short periods without air traffic cannot be attributed to aviation impact without doubt.

Better insight into the climate impact of contrail cirrus requires measurements and modelling of the radiative properties of observed contrail cirrus. Meteorological models which are able to predict realistic ice-supersaturation in the atmosphere and contrail cirrus as a function of traffic in detail at mesoscale and at global scale have still to be developed and verified. At present also a complete set of validation data for model studies of contrail-cirrus is missing. Better insight into the aerosol impact of aviation on cirrus can hardly be determined from observations alone. Here models have to be developed which account for the changes in aerosol distribution due to aircraft emissions and the impact of the aerosol on cirrus formation. Some work into these directions is under progress.

#### Acknowledgements

I thank Luca Bugliaro, Francois Garnier, Bernd Kärcher, Waldemar Krebs, Michael Ponater, Hermann Mannstein, Susanne Marquart, Roberto Paoli, and Robert Sausen for provision of figures and for helpful suggestions.

#### References

- E. Schmidt, Die Entstehung von Eisnebel aus den Auspuffgasen von Flugmotoren, in: Schriften der Deutschen Akademie der Luftfahrtforschung, vol. 44, Verlag R. Oldenbourg, München/Berlin, 1941, pp. 1–15.
- [2] W.B. Murcray, On the possibility of weather modification by aircraft contrails, Mon. Wea. Rev. 98 (1970) 745–748.
- [3] J.E. Penner, D.H. Lister, D.J. Griggs, D.J. Dokken, M. McFarland (Eds.), IPCC, Aviation and the Global Atmosphere. A Special Report of IPCC (Intergovernmental Panel on Climate Change), Cambridge Univ. Press, Cambridge, UK, 1999, pp. 65–120.
- [4] D.W. Fahey, U. Schumann, S. Ackerman, P. Artaxo, O. Boucher, M.Y. Danilin, B. Kärcher, P. Minnis, T. Nakajima, O.B. Toon, Aviationproduced aerosols and cloudiness, in: J.E. Penner, D.H. Lister, D.J. Griggs, D.J. Dokken, M. McFarland (Eds.), Aviation and the Global Atmosphere, A Special Report of IPCC (Intergovernmental Panel on Climate Change), Cambridge Univ. Press, Cambridge, UK, 1999, pp. 65–120.
- [5] B. Kärcher, Aviation-produced aerosols and contrails, Surveys Geophys. 20 (1999) 113–167.
- [6] B. Kärcher, Contrails: Observations, formation mechanisms, atmospheric impacts, uncertainties, DLR-Mitt. 2000-01, DLR Oberpfaffenhofen, 2000.
- [7] D.S. Lee, P.E. Clare, J. Haywood, B. Kärcher, R.W. Lunnon, I. Pilling, A. Slingo, J.R. Tilston, Identifying the uncertainties in radiative forcing of climate from aviation contrails and aviation-induced cirrus, Report DERA/AS/PT/CR000102, DERA, Pyestock, UK, 2000.
- [8] U. Schumann, J. Ström, F. Arnold, T.K. Berntsen, P.M. de F. Forster, D. Hauglustaine, Aviation impact on atmospheric composition and climate, in: European Research in the Stratosphere 1996–2000, EUR 19867, European Commission, Brussels, 2001, pp. 257–307, Chapter 7.
- [9] U. Schumann, Contrail Cirrus, in: D.K. Lynch, K. Sassen, D.O'C. Starr, G. Stephens (Eds.), Cirrus, Oxford Univ. Press, London, 2002, pp. 231–255.
- [10] H. Appleman, The formation of exhaust contrails by jet aircraft, Bull. Amer. Meteor. Soc. 34 (1953) 14-20.
- [11] U. Schumann, On conditions for contrail formation from aircraft exhausts, Meteor. Z. 5 (1996) 4–23.
- [12] U. Schumann, Influence of propulsion efficiency on contrail formation, Aerosp. Sci. Tech. 4 (2000) 391-401.
- [13] R. Busen, U. Schumann, Visible contrail formation from fuels with different sulfur contents, Geophys. Res. Lett. 22 (1995) 1357–1360.
- [14] B. Kärcher, B. Busen, A. Petzold, F.P. Schröder, U. Schumann, E.J. Jensen, Physicochemistry of aircraft-generated liquid aerosols, soot, and ice particles, 2, Comparison with observations and sensitivity studies, J. Geophys. Res. 103 (1998) 17129–17148.
- [15] E.J. Jensen, O.B. Toon, S. Kinne, G.W. Sachse, B.E. Anderson, K.R. Chan, C.H. Twohy, B. Gandrud, A. Heymsfield, R.C. Miake-Lye, Environmental conditions required for contrail formation and persistence, J. Geophys. Res. 103 (1998) 3929–3936.

- [16] U. Schumann, B. Busen, M. Plohr, Experimental test of the influence of propulsion efficiency on contrail formation, J. Aircraft 37 (2000) 1083–1087.
- [17] U. Schumann, F. Arnold, B. Busen, J. Curtius, B. Kärcher, A. Kiendler, A. Petzold, H. Schlager, F. Schröder, K.-H. Wohlfrom, Influence of fuel sulfur on the composition of aircraft exhaust plumes: The experiments SULFUR 1–7, J. Geophys. Res. 107 (2002) AAC 2-1–AAC 2-27, 10.1029/2001JD000813.
- [18] F. Schröder, J. Ström, Aircraft measurements of sub micrometer aerosol particles in the midlatitude free troposphere and tropopause region, Atmos. Res. 44 (1997) 333–356.
- [19] F. Schröder, B. Kärcher, M. Fiebig, A. Petzold, Aerosol states in the free troposphere at northern midlatitudes, J. Geophys. Res. 107 (D21) (2002) LAC 8-1–LAC 8-8, 10.1029/2000JD000194.
- [20] A. Minikin, A. Petzold, J. Ström, R. Krejci, M. Seifert, P. van Velthoven, H. Schlager, U. Schumann, Aircraft observations of the upper tropospheric fine particle aerosol in the Northern and Southern Hemispheres at midlatitudes, Geophys. Res. Lett. 30 (2003), 10.1029/2002GL016458.
- [21] F.P. Schröder, B. Kärcher, A. Petzold, R. Baumann, B. Busen, C. Hoell, U. Schumann, Ultrafine aerosol particles in aircraft plumes: In situ observations, Geophys. Res. Lett. 25 (1998) 2789–2792.
- [22] B. Kärcher, T. Peter, U.M. Biermann, U. Schumann, The initial composition of jet condensation trails, J. Atmos. Sci. 53 (1996) 3066– 3083.
- [23] A. Petzold, C. Stein, S. Nyeki, M. Gysel, E. Weingartner, U. Baltensperger, H. Giebl, R. Hitzenberger, A. Döpelheuer, S. Vrchoticky, H. Puxbaum, M. Johnson, C.D. Hurley, R. Marsh, C.W. Wilson, Properties of jet engine combustion particles during the PartEmis experiment: Microphysics and chemistry, Geophys. Res. Lett. 30 (2003), 10.1029/2003GL017283, 52-1–52-4.
- [24] O.B. Popovicheva, N.M. Persiantseva, E.E. Lukhovitskaya, N.K. Shonija, N.A. Zubareva, B. Demirdjian, D. Ferry, J. Suzanne, Aircraft engine soot as contrail nuclei, Geophys. Res. Lett. 31 (2004) L11104, 10.1029/2003GL018888.
- [25] B. Kärcher, R.P. Turco, F. Yu, M.Y. Danilin, D.K. Weisenstein, R.C. Miake-Lye, B. Busen, On the unification of aircraft ultrafine particle emission data, J. Geophys. Res. 105 (2000) 29379–29386.
- [26] O. Möhler, O. Stetzer, S. Schaefers, C. Linke, M. Schnaiter, R. Tiede, H. Saathoff, M. Krämer, A. Mangold, P. Budz, P. Zink, J. Schreiner, K. Mauersberger, W. Haag, B. Kärcher, U. Schurath, Experimental investigations of homogeneous freezing of sulphuric acid particles in the aerosol chamber AIDA, Atmos. Chem. Phys. 2 (2003) 211–223.
- [27] O. Möhler, S. Büttner, C. Linke, M. Schnaiter, H. Saathoff, O. Stetzer, R. Wagner, M. Krämer, A. Mangold, U. Ebert, U. Schurath, Effect of sulfuric acid coating on heterogeneous ice nucleation by soot aerosol particles, J. Geophys. Res. (2005), in press.
- [28] B. Kärcher, T. Koop, The role of organic aerosols in homogeneous ice formation, Atmos. Chem. Phys. 5 (2005) 703-714.
- [29] R. Meerkötter, U. Schumann, P. Minnis, D.R. Doelling, T. Nakajima, Y. Tsushima, Radiative forcing by contrails, Ann. Geophys. 17 (1999) 1080–1094.
- [30] F. Schröder, B. Kärcher, C. Duroure, J. Strom, A. Petzold, J.-F. Gayet, B. Strauss, P. Wendling, A. Thomas, On the transition of contrails into cirrus clouds, J. Atmos. Sci. 57 (2000) 464–480.
- [31] U. Schumann, H. Schlager, F. Arnold, R. Baumann, P. Haschberger, O. Klemm, Dilution of aircraft exhaust plumes at cruise altitudes, Atmospheric Environment 32 (1998) 3097–3103.
- [32] A. Detwiler, R. Pratt, Clear-air seeding: Opportunities and strategies, J. Wea. Mod. 16 (1984) 46-60.
- [33] E. Glückauf, Notes on upper air hygrometry II: On the humidity in the stratosphere, Q. J. Roy. Meteor. Soc. 71 (1945) 110-112.
- [34] H. Weickmann, Formen und Bildung atmosphärischer Eiskristalle, Beitr. Physik der freien Atmosphäre 28 (1945) 12–52.
- [35] A. Brewer, Condensation trails, Weather 1 (1946) 34-41.
- [36] K. Gierens, U. Schumann, Ice-supersaturation in the upper troposphere, GEWEX News 14 (1) (2004) 6-7.
- [37] F.H. Ludlam, Clouds and Storms, The Pennsylvania State Univ. Press, University Park, PA, 1980.
- [38] K. Sassen, Contrail-cirrus and their potential for regional climate change, Bull. Amer. Meteor. Soc. 78 (1997) 1885–1903.
- [39] T. Koop, B. Luo, A. Tsias, T. Peter, Water activity as the determinant for homogeneous ice nucleation in aqueous solutions, Nature 406 (2000) 611–614.
- [40] D.M. Murphy, K.K. Kelly, A.F. Tuck, M.H. Proffitt, S. Kinne, Ice saturation at the tropopause observed from the ER-2 aircraft, Geophys. Res. Lett. 17 (1990) 353–356.
- [41] A.J. Heymsfield, L.M. Miloshevich, Relative humidity and temperature influences on cirrus formation and evolution: Observations from wave clouds and FIRE II, J. Atmos. Sci. 52 (1995) 4302–4326.
- [42] K. Gierens, U. Schumann, M. Helten, H. Smit, A. Marenco, A distribution law for relative humidity in the upper troposphere and lower stratosphere derived from three years of MOZAIC measurements, Ann. Geophys. 17 (1999) 1218–1226.
- [43] J. Ovarlez, P. van Velthoven, G. Sachse, S. Vay, H. Schlager, H. Ovarlez, Comparison of water vapor measurements from POLINAT 2 with ECMWF analyses in high-humidity conditions, J. Geophys. Res. 105 (2000) 3737–3744.
- [44] U. Schumann, H. Schlager, F. Arnold, J. Ovarlez, H. Kelder, Ø. Hov, G. Hayman, I.S.A. Isaksen, J. Staehelin, P.D. Whitefield, Pollution from aircraft emissions in the North Atlantic flight corridor: Overview on the POLINAT projects, J. Geophys. Res. 105 (2000) 3605–3631.
- [45] J. Ovarlez, J.-F. Gayet, K. Gierens, J. Ström, H. Ovarlez, F. Auriol, B. Busen, U. Schumann, Water vapour measurements inside cirrus clouds in Northern and Southern hemispheres during INCA, Geophys. Res. Lett. 29 (16) (2002), 10.1029/2001GL014440, 60-1–60-4.
- [46] E.J. Jensen, O.B. Toon, S.A. Vay, J. Ovarlez, R. May, P. Bui, C.H. Twohy, B. Gandrud, R.F. Pueschel, U. Schumann, Prevalence of ice supersaturated regions in the upper troposphere: Implications for optically thin ice cloud formation, J. Geophys. Res. 106 (2001) 17253–17266.
- [47] P. Spichtinger, K. Gierens, U. Leiterer, H. Dier, Ice supersaturation in the tropopause region over Lindenberg, Germany, Meteor. Z. 12 (2003) 143–156.

- [48] P. Spichtinger, K. Gierens, W. Read, The global distribution of ice-supersaturated regions as seen by the microwave limb sounder, Q. J. Roy. Meteor. Soc. 129 (2003) 3391–3410.
- [49] K. Gierens, R. Kohlhepp, P. Spichtinger, M. Schroedter-Homscheidt, Ice supersaturation as seen from TOVS, Atmos. Chem. Phys. 4 (2004) 539–547.
- [50] C.J. Stubenrauch, U. Schumann, Impact of air traffic on cirrus coverage, Geophys. Res. Lett. (2005), in press.
- [51] A. Marenco, V. Thouret, P. Nédélec, H. Smit, M. Helten, D. Kley, F. Karcher, P. Simon, K. Law, J. Pyle, G. Poschmann, R. von Wrede, C. Hume, T. Cook, Measurement of ozone and water vapor by Airbus in-service aircraft: The MOZAIC airborne program, an overview, J. Geophys. Res. 103 (1998) 25631–25642.
- [52] P. Spichtinger, K. Gierens, H. Wernli, A case study on the formation and evolution of ice supersaturation in the vicinity of a warm conveyor belt's outflow region, Atmos. Phys. Chem. 5 (2005) 973–987.
- [53] K. Gierens, P. Spichtinger, On the size distribution of ice-supersaturated regions in the upper troposphere and lowermost stratosphere, Ann. Geophys. 18 (2000) 499–504.
- [54] J.B. Smith, E.J. Hintsa, N.T. Allen, R.M. Stimpfle, J.G. Anderson, Mechanisms for midlatitude ozone loss: Heterogeneous chemistry in the lowermost stratosphere?, J. Geophys. Res. 106 (2001) 1297–1309.
- [55] B. Kärcher, S. Solomon, On the composition and optical extinction of particles in the tropopause region, J. Geophys. Res. 104 (1999) 27441–27459.
- [56] L. Goldfarb, P. Keckhut, M.-L. Chanin, A. Houchecorn, Cirrus climatological results from LIDAR measurements at OHP (44°N, 6°E), Geophys. Res. Lett. 28 (2001) 1687–1690.
- [57] R. Sausen, K. Gierens, M. Ponater, U. Schumann, A diagnostic study of the global distribution of contrails: Part I: Present day climate, Theor. Appl. Climatol. 61 (1998) 127–141.
- [58] H. Mannstein, R. Meyer, P. Wendling, Operational detection of contrails from NOAA-AVHRR data, Int. J. Remote Sensing 20 (1999) 1641–1660.
- [59] W.B. Rossow, R.A. Schiffer, Advances in understanding clouds from ISCCP, Bull. Amer. Meteor. Soc. 80 (1999) 2261–2287.
- [60] D.P. Wylie, W.P. Menzel, Eight years of high cloud statistics using HIRS, J. Climate 12 (1999) 170-184.
- [61] P.-H. Wang, P. Minnis, M.P. McCormick, G.S. Kent, K.M. Skeens, A 6-year climatology of cloud occurrence frequency from Stratospheric Aerosol and Gas Experiment II observations (1985–1990), J. Geophys. Res. 101 (1996) 29407–29429.
- [62] D.C. Lewellen, W.S. Lewellen, Large eddy simulations of the vortex-pair breakup in aircraft wakes, AIAA Journal 34 (1996) 2337–2345.
- [63] F. Holzäpfel, T. Gerz, R. Baumann, The turbulent decay of trailing vortex pairs in stably stratified environments, Aerosp. Sci. Techn. 5 (2001) 95–108.
- [64] U. Schumann, A. Dörnbrack, T. Dürbeck, T. Gerz, Large-Eddy Simulation of Turbulence in the Free Atmosphere and behind Aircraft, Fluid Dynam. Res. 20 (1997) 1–10.
- [65] D.C. Lewellen, W.S. Lewellen, The effects of aircraft wake dynamics on contrail formation, J. Atmos. Sci. 58 (2001) 390-406.
- [66] K. Gierens, E. Jensen, A numerical study of the contrail-to-cirrus transition, Geophys. Res. Lett. 25 (1998) 4341–4344.
- [67] R. Paoli, J. Hélie, T. Poinsot, Contrail formation in aircraft wakes, J. Fluid Mech. 502 (2004) 361–373.
- [68] R. Sussmann, K. Gierens, Differences in early contrail evolution of 2-engined versus 4-engined aircraft. Lidar measurements and numerical simulations, J. Geophys. Res. 106 (2001) 4899–4911.
- [69] E.J. Jensen, A.S. Ackerman, D.E. Stevens, O.B. Toon, P. Minnis, Spreading and growth of contrails in a sheared environment, J. Geophys. Res. 103 (1998) 31557–31568.
- [70] U. Schumann, P. Konopka, R. Baumann, B. Busen, T. Gerz, H. Schlager, P. Schulte, H. Volkert, Estimate of diffusion parameters of aircraft exhaust plumes near the tropopause from nitric oxide and turbulence measurements, J. Geophys. Res. 100 (1995) 14147–14162.
- [71] T. Dürbeck, T. Gerz, Dispersion of aircraft exhausts in the free atmosphere, J. Geophys. Res. 101 (1996) 26007–26015.
- [72] T. Gerz, T. Dürbeck, P. Konopka, Transport and effective diffusion of aircraft emissions, J. Geophys. Res. 103 (1998) 25905–25913.
- [73] V. Freudenthaler, F. Homburg, H. Jäger, Contrail observations by ground-based scanning lidar: Cross-sectional growth, Geophys. Res. Lett. 22 (1995) 3501–3504.
- [74] V. Freudenthaler, Lidarmessungen der räumlichen Ausbreitung sowie mikrophysikalischer und optischer Parameter von Flugzeugkondensstreifen. Schriftenreihe des Fraunhofer Institut Atmosphärische Umweltforschung, Garmisch, Band 63-2000, 2000.
- [75] R. Sussmann, Vertical dispersion of an aircraft wake: Aerosol-lidar analysis of entrainment and detrainment in the vortex regime, J. Geophys. Res. 104 (1999) 2117–2129.
- [76] R. Sussmann, K. Gierens, Lidar and numerical studies on the different evolution of vortex pair and secondary wake in young contrails, J. Geophys. Res. 104 (1999) 2131–2142.
- [77] U. Schumann, P. Wendling, Determination of contrails from satellite data and observational results, in: Proc. of a DLR Intern. Coll., Bonn, Nov. 15/16, 1990, in: Lecture Notes in Engrg., vol. 60, Springer-Verlag, Berlin, 1990, pp. 138–153.
- [78] P. Minnis, D.F. Young, D.P. Garber, L. Nguyen, W.L. Smith Jr., R. Palikonda, Transformation of contrails into cirrus during SUCCESS, Geophys. Res. Lett. 25 (1998) 1157–1160.
- [79] J.A. Screen, A.R. MacKenzie, Aircraft condensation trails and cirrus, Weather 59 (2004) 116–121.
- [80] S. Bakan, M. Betancor, V. Gayler, H. Grassl, Contrail frequency over Europe from NOAA-satellite images, Ann. Geophys. 12 (1994) 962–968.
- [81] R. Meyer, H. Mannstein, R. Meerkötter, U. Schumann, P. Wendling, Regional radiative forcing by line-shaped contrails derived from satellite data, J. Geophys. Res. 107 (2002) 4104, 10.1029/2001JD000426.
- [82] P. Minnis, R. Palikonda, B.J. Walter, J.K. Ayers, H. Mannstein, Contrail coverage over the North Pacific from AVHRR data, Meteor. Z. (2005), in press.

- [83] R. Palikonda, P. Minnis, D.P. Duda, H. Mannstein, Contrail coverage derived from 2001 AVHRR data over the continental United States of America and surrounding areas, Meteor. Z. (2005), in press.
- [84] R. Meyer, H. Mannstein, R. Meerkötter, P. Wendling, Contrail and cirrus observations over Europe from 6 years of NOAA-AVHRR data, in: Proc. of the 2002 EUMETSAT Meteorological Satellite Conference, Dublin, Ireland, 2002 September 2–6, 2002, pp. 728–735.
- [85] R. Meyer, R. Buell, C. Leiter, H. Mannstein, S. Marquart, T. Oki, P. Wendling, Contrail observations over Southern and Eastern Asia in NOAA/AVHRR data and comparisons to contrail simulations in a GCM, Int. J. Remote Sensing (2005), in press.
- [86] K. Gierens, R. Sausen, U. Schumann, A diagnostic study of the global coverage by contrails. Part II: Future air traffic scenarios, Theor. Appl. Climatol. 63 (1999) 1–9.
- [87] S. Marquart, M. Ponater, F. Mager, R. Sausen, Future development of contrail cover, optical depth, and radiative forcing: impacts of increasing air traffic and climate change, J. Clim. 16 (2003) 2890–2904.
- [88] D.P. Duda, P. Minnis, D.P. Garber, R. Palikonda, CONUS contrail frequency and coverage estimated from RUC and flight track data, Meteor. Z. (2005), in press.
- [89] A. Kristensson, J.-F. Gayet, J. Ström, F. Auriol, In situ observations of a reduction in effective crystal diameter in cirrus clouds near a flight corridor, Geophys. Res. Lett. 27 (2000) 681–684.
- [90] K. Wyser, J. Ström, A possible change in cloud radiative forcing due to aircraft exhaust, Geophys. Res. Lett. 25 (1998) 1673–1676.
- [91] J. Ström, S. Ohlsson, In situ measurements of enhanced crystal number densities in cirrus clouds caused by aircraft exhaust, J. Geophys. Res. 103 (1998) 11355–11361.
- [92] B. Kärcher, U. Lohmann, A parameterization of cirrus cloud formation: Homogeneous freezing including effects of aerosol size, J. Geophys. Res. 107 (2002) 4698, 10.1029/2001JD001429.
- [93] J. Ström, M. Seifert, J. Ovarlez, A. Minikin, J.-F. Gayet, R. Krejci, A. Petzold, F. Auriol, B. Busen, U. Schumann, B. Kärcher, W. Haag, H.-C. Hansson, Cirrus cloud occurrence as function of ambient relative humidity: a comparison of observations obtained during the INCA experiment, Atmos. Chem. Phys. 3 (2003) 1807–1816.
- [94] B. Kärcher, J. Ström, The roles of dynamical variability and aerosols in cirrus cloud formation, Atmos. Chem. Phys. 3 (2003) 823-838.
- [95] J.-F. Gayet, J. Ovarlez, V. Shcherbakov, J. Ström, U. Schumann, A. Minikin, F. Auriol, A. Petzold, M. Monier, Cirrus cloud microphysical and optical properties at southern and northern midlatitudes during the INCA experiment, J. Geophys. Res. 109 (2004) D20206, 10.1029/2004JD004803.
- [96] U. Lohmann, B. Kärcher, Hendricks J., Sensitivity studies of cirrus clouds formed by heterogeneous freezing in the ECHAM GCM, J. Geophys. Res. 109 (2004) D16204, 10.1029/2003JD004443.
- [97] J. Hendricks, B. Kärcher, A. Döpelheuer, J. Feichter, U. Lohmann, D. Baumgardner, Simulating the global atmospheric black carbon cycle: a revisit to the contribution of aircraft emissions, Atmos. Chem. Phys. 4 (2004) 2521–2541.
- [98] E.J. Jensen, O.B. Toon, The potential impact of soot particles from aircraft exhaust on cirrus clouds, Geophys. Res. Lett. 24 (1997) 249–252.
- [99] W. Haag, B. Kärcher, J. Ström, A. Minikin, U. Lohmann, J. Ovarlez, A. Stohl, Freezing thresholds and cirrus cloud formation mechanisms inferred from in situ measurements of relative humidity, Atmos. Chem. Phys. 3 (2003) 1781–1806.
- [100] W. Haag, B. Kärcher, The impact of aerosols and gravity waves on cirrus clouds at midlatitudes, J. Geophys. Res. 109 (2004) D12202, 10.1029/2004JD004579.
- [101] O. Boucher, Influence of air traffic on cirrus occurrence, Nature 397 (1999) 30–31.
- [102] P. Minnis, J.K. Ayers, R. Palikonda, D. Phan, Contrails, cirrus trends, and climate, J. Clim. 17 (2004) 1671–1685.
- [103] C. Zerefos, K. Eleftheratos, D. Balis, P. Zanis, G. Tselioudis, C. Meleti, Evidence of impact of aviation on cirrus cloud formation, Atmos. Chem. Phys. 3 (2003) 1633–1644.
- [104] F. Stordal, G. Myhre, W. Arlander, T. Svendby, E.J.G. Stordal, W.B. Rossow, D.S. Lee, Is there a trend in cirrus cloud cover due to aircraft traffic?, Atmos. Chem. Phys. Discuss. 4 (2004) 6473–6501.
- [105] H. Mannstein, U. Schumann, Aircraft induced contrail cirrus over Europe, Meteor. Z. (2005), in press.
- [106] V. Freudenthaler, F. Homburg, H. Jäger, Optical parameters of contrails from lidar measurements: Linear depolarization, Geophys. Res. Lett. 23 (1996) 3715–3718.
- [107] H. Jäger, V. Freudenthaler, F. Homburg, Remote sensing of optical depth of aerosols and cloud cover related to air traffic, Atmos. Environ. 32 (1998) 3123–3127.
- [108] P. Minnis, U. Schumann, D.R. Doelling, K.M. Gierens, D.W. Fahey, Global distribution of contrail radiative forcing, Geophys. Res. Lett. 26 (1999) 1853–1856.
- [109] M. Ponater, S. Marquart, R. Sausen, Contrails in a comprehensive climate model: parameterisation and radiative forcing results, J. Geophys. Res. 107 (2002) 4164, 10.1029/2001JD000429.
- [110] B. Strauss, R. Meerkötter, B. Wissinger, P. Wendling, M. Hess, On the regional climatic impact of contrails: Microphysical and radiative properties of contrails and natural cirrus clouds, Ann. Geophys. 15 (1997) 1457–1467.
- [111] J.P.F. Fortuin, R. van Dorland, W.M.F. Wauben, H. Kelder, Greenhouse effects of aircraft emissions as calculated by a radiative transfer model, Ann. Geophys. 13 (1995) 413–418.
- [112] Y. Zhang, A. Macke, F. Albers, Effect of crystal size spectrum and crystal shape on stratiform cirrus radiative forcing, Atmosph. Res. 52 (1999) 59–75.
- [113] G. Myhre, F. Stordal, On the tradeoff of the solar and the thermal infrared radiative impact of contrails, Geophys. Res. Lett. 28 (2001) 3119–3122.
- [114] S. Marquart, B. Mayer, Towards a reliable GCM estimation of contrail forcing, Geophys. Res. Lett. 29 (2002) 1179, 10.1029/2001GL014075.

- [115] S. Marquart, M. Ponater, F. Mager, R. Sausen, Future development of contrails: Impacts of increasing air traffic and climate change, in: Proc. AAC-Conference, June 30 – July 3, 2003, Friedrichshafen, Germany, EUR 21051, 2004, pp. 255–260.
- [116] D. Rind, P. Lonergan, K. Shah, Modeled impact of cirrus cloud increases along aircraft flight paths, J. Geophys. Res. 105 (2000) 19927– 19940.
- [117] M. Ponater, S. Marquart, R. Sausen, U. Schumann, On contrail climate sensitivity, Geophys. Res. Lett. (2005) 32, L10706, 10.1029/2005GL022580.
- [118] K.P. Shine, Comments on Contrails, cirrus trends, and climate, J. Clim. 68 (2005), in press.
- [119] D.J. Travis, A.M. Carleton, R.G. Lauritsen, Jet aircraft contrails: Surface temperature variations during the aircraft groundings of September 11–13, 2001, in: 10th Conference on Aviation, Range & Aerospace Meteorology, 13–16 May 2002, Portland, Oregon, American Meteorological Society, 2002, paper J1.1.
- [120] D.J. Travis, A.M. Carleton, R.G. Lauritsen, Contrails reduce daily temperature range, brief communications, Nature 418 (2002) 601.
- [121] D.J. Travis, A.M. Carleton, R.G. Lauritsen, Regional variations in U.S. diurnal temperature range for the 11–14 September 2001 aircraft groundings: Evidence of jet contrail influence on climate, J. Clim. 17 (2004) 1123–1134.
- [122] A.J. Kalkstein, R.C. Balling Jr, Impact of unusually clear weather on United States daily temperature range following 9/11/2001, Climate Res. 26 (2004) 1–4.
- [123] S. Marquart, M. Ponater, L. Ström, K. Gierens, An upgrade estimate of the radiative forcing by cryoplane contrails, Meteor. Z. (2005), in press.
- [124] L. Ström, K. Gierens, First simulations of cryoplane contrails, J. Geophys. Res. 107 (2002) 4346, 10.1029/2001JD000838.
- [125] F. Svensson, A. Hasselrot, J. Moldanova, Reduced environmental impact by lowered cruise altitude for liquid-hydrogen fuelled aircraft, Aerosp. Sci. Technol. 8 (2004) 307–320.
- [126] S. Marquart, R. Sausen, M. Ponater, V. Grewe, Estimate of the climate impact of cryoplanes, Aerosp. Sci. Technol. 5 (2001) 73-84.
- [127] V. William, R.B. Noland, R. Toumi, Reducing the climate change impacts of aviation by restricting cruise altitudes, Transportation Res. Part D 7 (2002) 451–464.
- [128] V. William, R.B. Noland, R. Toumi, Air transport cruise altitude restrictions to minimize contrail formation, Climate Policy 3 (2003) 207–219, S1469-3062(03)00054-8.
- [129] C. Fichter, S. Marquart, R. Sausen, D.S. Lee, The impact of cruise altitude on contrails and related radiative forcing, Meteor. Z. (2005), in press.
- [130] V. Grewe, M. Dameris, C. Fichter, R. Sausen, Impact of Aircraft NOx Emissions. Part 1: Interactively coupled climate-chemistry simulations and sensitivities to climate-chemistry feedback, lightning and model resolution, Meteor. Z. 11 (2002) 177–186.
- [131] M.J. Garrison, D.P. DuBois, S.L. Baughcum, The Effect of Constrained Cruise Altitudes on Fuel Usage, NOx Production, and Flight Time for Commercial Airplanes, NASA/CR, 213305, 2004.
- [132] H. Mannstein, P. Spichtinger, K. Gierens, How to avoid contrail cirrus, Transportation Research Part D, Transport and Environment (2005), in press.
- [133] B. Bregman, P.-H. Wang, J. Lelieveld, Chemical ozone loss in the tropopause region on subvisible ice clouds, calculated with a chemistrytransport model, J. Geophys. Res. 107 (2002) 4032, 10.1029/2001JD000761, ACH 5-1 – 5-15.
- [134] S.K. Meilinger, B. Kärcher, T. Peter, Microphysics and heterogeneous chemistry in aircraft plumes—high sensitivity on local meteorology and atmospheric composition, Atmos. Chem. Phys. 5 (2005) 533–545.
- [135] A. Zahn, C.A.M. Brenninkmeijer, M. Maiss, D.H. Scharffe, P.J. Crutzen, M. Hermann, J. Heintzenberg, A. Wiedensohler, H. Güsten, G. Heinrich, H. Fischer, J.W.M. Cuijpers, P.J.F. van Velthoven, Identification of extratropical two-way troposphere-stratosphere mixing based on CARIBIC measurements of O<sub>3</sub>, CO and ultrafine particles, J. Geophys. Res. 105 (2000) 1527–1535.