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Visible contrail formation from fuels with different sulfur contents

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Abstract. As a test for postulated influences of sulfur emissions on nucleation, the contrail formation from a two-engine jet aircraft was investigated using fuels with different sulfur contents for the two engines during the same flight. The sulfur mass fractions in the fuels were about 2 and 250 ppm, respectively, typical for aviation fuels. Other engine and fuel parameters were about the same for both engines. Contrail formation was observed visually from distances as close as 100 m and documented by video and photos. The flight took place at 302 hPa (9 km altitude), at ambient temperatures of about -50°C, and relative humidity for liquid water of about 34 %. Short contrails formed about 30 m after the engines. No visible differences were detected in the contrails forming from the two engines. The observed conditions for contrail formation are close to those predicted by *Appleman* [1953] if the propulsion efficiency of the aircraft/engine combination during flight is taken into account.

Introduction

Contrails from engine exhaust of high-flying aircraft may influence the climatological and chemical state of the atmosphere [*Schumann*, 1994; *WMO*, 1995]. Small changes in the threshold conditions for contrail formation cause large changes in the regions where contrails may be found [*Miake-Lye et al.*, 1993]. Several recent studies suggest that emissions of sulfur oxides from engine fuel burning may induce sulfuric acid formation, binary nucleation of sulfuric acid with water, and subsequent heteromolecular condensation on soot particles or ions in the exhaust, possibly causing contrail formation at higher ambient temperature than without sulfur emissions [e.g., *Reiner and Arnold*, 1993; *Miake-Lye et al.*, 1994]. This hypothesis is tested by visual observations of contrail formation from a jet aircraft operating under cruise conditions with different fuels for the two engines, one with low and the other with hundred times larger sulfur content.

Jet fuel specifications [*ASTM*, 1994] allow for up to 3 g of sulfur per kg of fuel (3000 ppm mass fraction). Depending on the source of the crude petroleum and type of refinery processing, sulfur contents vary from values below 100 ppm to levels around 2000 ppm, with averages around 500 ppm [see *Schumann*, 1994].

According to *Appleman* [1953], contrails form when isobaric mixing between the hot and humid exhaust plume and cold ambient air leads to a mixture reaching saturation with respect to water so that cloud droplets form which then might freeze to form ice particles. The threshold temperature for contrail formation depends on the relative humidity of the ambient air, but contrails form even in perfectly dry air when the temperature is low enough, typically less than -50°C at altitudes above 300 hPa. Because of limited visibility, in particular for large ice particles, large viewing angles, and large observation distances, contrails might be observable only when the air is a few degrees cooler than the threshold temperature [*Pilié and Jiusto,* 1958; *Knollenberg,* 1972].

The particle formation in the exhaust plume may get enhanced by sulfur oxides (SO_x) emissions. *Hofmann and Rosen* [1978] observed large concentrations of volatile aerosols in a layer of stratospheric air which they related to the exhaust plume of a stratospheric aircraft. They concluded that the aerosol consists of sulfuric acid droplets formed in the aircraft plume which may act as cloud condensation nuclei. Such nuclei were also measured in jet aircraft plumes of cruising subsonic aircraft [*Pitchford et al.*, 1991; *Hagen et al.*, 1992]. *Reiner and Arnold* [1993] and *Frenzel and Arnold* [1994] measured sulfuric acid gas formed from the oxidation of SO_x in laboratory and in young jet exhaust plumes at ground levels at time scales of less than 0.1 s. The H_2SO_4 vapor pressure quickly exceeds saturation, with high embryo formation rates due to binary condensation of H_2SO_4 with H_2O [*Miake-Lye et al.*, 1994].

However, from a nucleation model, *Kärcher et al.* [1995] found that the sulfuric acid particles do not become large enough to be visible under threshold conditions. Also other factors may be responsible for early contrail formation. *Miake-Lye et al.* [1993] identified the influence of adiabatic cooling of jet plumes which enter the cores of strongly rotating wake vortices. *Kärcher* [1994] noted the importance of radial mixing. *Peters* [1993] observed that contrails behind modern high-bypass engines form earlier than behind older low-bypass engines. This was explained with different ratios of moisture to heat release from the various engines. A possible impact of engine efficiency was noted by *Knollenberg* [1972] who observed a contrail at a temperature several degrees above the Appleman threshold. *W. A. Cooper and L. D. Nelson* (personal communication, "Threshold conditions for the formation of contrails," NCAR, Boulder, CO 80307-3000, to be published, 1994) observed contrails at temperatures about 1.5 K above the Appleman threshold temperature; they suggested that this might result from the freezing of solution droplets formed from deliquescence of any soluble constituents of the particles in the exhaust. Hence, the question of the impact of sulfur on contrail formation is open. This led us to perform the explorative test described in this paper.

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Experimental

A twin-engine jet aircraft of type VFW 614, known as the ATTAS (Advanced Technology Testing Aircraft System) of DLR Braunschweig, Germany, is used for the contrail formation study. The aircraft has a wing span of 21.50 m, length overall of 20.60 m, and maximum speed of Mach 0.65 [*JANE,* 1976; p. 98]. It is equipped with two Rolls-Royce/SNECMA M45H Mk501 turbofan engines [*JANE,* 1976; p. 704], with a bypass ratio of 3:1, and a take-off thrust of 32.4 kN. The engines are mounted on overwing pylons aft of the wing rear spar. The integral wing tanks have a capacity of 3115 litres, each. Normally the fuel tanks in the wings are connected. For this test, the connecting valve was closed so that each engine is supplied with fuel from the respective tank separately. The two tanks were filled with different Jet A-1 fuels.

The properties and testing methods for Jet A-1 fuel, a standard kerosene used in civil aviation, are described in *ASTM* [1994]. A survey of data from nine German fuel suppliers revealed sulfur contents between 2 ppm and 1040 ppm, a mean value of 375 ppm, with 4 samples below 100 ppm and 2 above 900 ppm. Because of the availability of the low sulfur fuel from a refinery near Hamburg, the aircraft tanks were filled at Hamburg airport (HAM) with a low sulfur fuel for the left engine (after twice flushing the tank to eliminate residuals from earlier tank fillings) and with the standard fuel-mix available at that airport for the right engine. Refinery certificates are available for the special supply and for the three last fillings of the storage tanks at that airport. As required for low sulfur fuels with this engine, 20 ppm of a lubricity improver (Nalco 5403 of Deutsche Nalco-Chemie, Frankfurt; a liquid organic film former; free of nitrogen, phosphorus and sulfur), was added to both fuel loadings during filling of the wing tanks. Thereafter, samples were taken from the wing tanks before and after the flight and analyzed (with standard method as identified) to determine the total sulfur content (Wickbold oxy-hydrogen method, IP 243/EN 41), the aromatics (ASTM D 1319) and hydrogen (ASTM D 3334) contents, and the specific combustion energies (ASTM D 1405). These data together with (mean) data from the refinery certificates are listed in Table I.

The test strategy was to find conditions for the onset of contrail formation, which is most sensitive to details of the nucleating process. The observations took place along a route from Schleswig (54°32'N, 9°33'E), where a radiosounding was started at 1045 UTC, in southerly direction (206°) towards Bremen at times in between 1011 and 1030 UTC 13 December 1994. After initial climb to FL 305 (flight level, in hft) (294 hPa), where a strong contrail formed, the aircraft descended until the level of contrail onset was found at FL 299 (302 hPa, about 9 km). At 1029 UTC its instruments indicated a fuel flow rate, exhaust gas temperature, and percentage speed of the low pressure compressor of $m_f = 125$ g s^{-1}, 420°C, and 84 %, respectively, for the left engine, and 126 g s^{-1}, 416°C, and 83 %, for the right engine. The true air speed, $V = 115$ m s^{-1}, was slower than normal to allow an observing aircraft to follow behind and below at variable distance and displacement approaching the ATTAS up to about 100 m. The observations were documented with a video camera and with black and white photographs.

Table I. Fuel parameters for left and right engines

Parameter	Unit	Left	Right
Sulfur cont. before flight	ppm	1.6	245
Sulfur content after flight	ppm	2.7	267
Hydrogen content	mass %	13.50	13.60
Water vapor emission	kg/kg	1.206	1.215
Density at 15°C	kg m^{-3}	800.9	801.2
Anilin point	°C	57.8	58.2
Spec. heat of combustion	MJ/kg	43.238	43.236
Aromatics content	vol. %	18.8	18.0
Olefins content	vol. %	0.5	1.0
Naphthalens content	vol. %	0.1	1.2
Initial boiling point	°C	156	154
Boiling end point	°C	236	243
Smoke point	mm	25	24

The European Meteorological Bulletin of the German Weather Service for 1200 UTC that day exhibits strong and uniform flow from north-west (300°) in between 250 and 400 hPa, with a maximum wind speed of 64 m s^{-1} over Schleswig at 286 hPa. For 300 hPa, the radiosoundings of Schleswig and Hannover (52°28'N, 9°42'E) report temperatures -50°C and -48°C, and dewpoint spreads of 9 K and 6 K, respectively, corresponding to 34 and 50 % relative humidity with respect to water (55 and 80 % ice saturation). Along the route at FL 299, the air temperature measured on board the ATTAS varied by at most 1K, so that the Schleswig soundings should be representative. No humidity measurements were available from the aircraft, but a slowly increasing tendency for contrail formation along the route and the higher humidity at Hannover suggest that humidity was increasing slightly, perhaps 10 %, on the way south. The visibility was at least 80 km, without persistent contrails, indicating humidity below ice saturation (62 %). Low clouds formed below 1700 m, and a few thin cirrus clouds with persistent contrails were visible at higher levels. The soundings revealed a Brunt-Väisälä frequency of about 0.01 s^{-1} and a Richardson number of about 2 to 4. No turbulence was noted during the flight.

Results

At the top level (FL 305), the resulting contrail was more than 1 km long. At FL 299, the contrails were occasionally disappearing, usually shorter than 1000 m, and the two individual jet contrails were clearly discernible. No contrails were visible at lower flight levels. The observations showed no significant visible difference in contrail formation from the two engines. Figure 1 documents the view from the observing aircraft at 1024 UTC at a distance of about 150 m. The two jet contrails have essentially equal shape and equal start points of contrail formation. The sun is about 40° to the left, giving good visibility but different contrasts for the two contrails. Their contrasts are equal if viewed from positions further right, as documented in other photos. Sideview photos show that the contrail formed about 25 - 35 m behind engine exits. The distances where contrails formed behind the engines were the same for both jets, at least up to the accuracy of observations of about 2 m. Figure 2 shows the temperature profile of the radiosonde of Schleswig versus altitude in terms of pressure. The symbol indicates the situation of contrail formation at – 49.7 ± 0.5 °C, 302.3 ± 0.7 hPa.

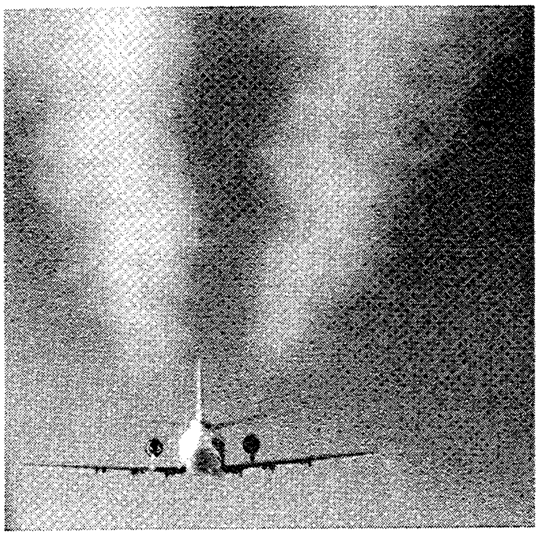

Figure 1. The contrail forming aircraft at FL 299, 1024 UTC (digitized photo). The sun is about 40° to the left, giving higher contrast for the left than for the right contrail.

Discussion

The fuel provided for the right engine contains a sulfur content of about 250 ppm, slightly less than average, but well within the wide range typical for aviation fuels. It is hundred times larger than for the left engine. Hence, visible differences were to be expected if such sulfur levels are important for contrail formation.

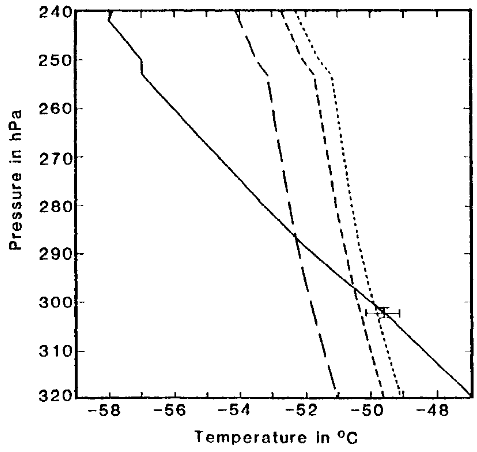

Figure 2. Pressure versus temperature from Schleswig radiosounding (full curve), Appleman criterion for the humidity of that sounding, for a propulsion efficiency $\eta = 0$ (long dashed) and $\eta = 0.14$ (short dashed), and for $\eta = 0.14$ with 10 % additional relative humidity (dotted). The symbol indicates observed contrail formation conditions with estimated error margins.

The contrail formed rather shortly behind this medium-sized aircraft, at plume ages of about 0.3 s. Visible contrails are often seen close behind jet airliners, but models predict larger distances (> 100 m) for B747 and B707 aircraft [*Kärcher,* 1994; *Miake-Lye et al.*, 1994]. Later onset of contrail formation may allow for stronger sulfur impact.

The experimental procedure avoids differences in the formation of the two jet contrails due to wake vortex dynamics, engine types, aircraft properties, and ambient air conditions, because these were equal in this test. The ratios of the specific heat of combustion and the emission index of water vapor of the two fuels, see Table I, differ by less than 0.7 %, implying a 0.07 K higher threshold temperature for the right engine. Fuel properties important for soot formation [*ASTM,* 1994] are the aromatics content and smoke point as given in Table I. We do not see any essential difference that could have masked the sulfur impact.

Figure 2 shows also the threshold temperature versus pressure separating regions where contrails are expected (to the left) from regions without contrails. The curves are computed as in *Appleman* [1953]. (The approximative eq. (52) of *Iribarne and Godson* [1981, p. 134] gives about 1 K higher threshold temperatures.) We use the water vapor emission index and the specific combustion heat (Q) values of the given fuels, and the relative humidity values reported from the Schleswig radiosounding. Different from the standard Appleman criterion, we account for the fact that only the fraction $(1-\eta)$ of the specific combustion heat Q is given directly to the exhaust jet plume, where η measures the engine/aircraft efficiency, i.e., the amount of propulsion work per combustion heat. The propulsion work is converted into heating of the entire wake after dissipation of the wake vortex systems, but this energy is not available for immediate heating of the young jet plume. According to flight performance calculations of DLR for the engine/aircraft combination for the given flight conditions at FL 299 at 1029 UTC (actual aircraft weight 17650 kg), each engine produces a thrust of $F = 6.4 \pm 0.1$ kN. Hence, $\eta = FV/(m_f Q) \cong 0.14$. A change in η from 0 to 0.14 causes a 1.4 K higher threshold temperature for contrail formation at 300 hPa. Modern aircraft/engine combinations (e.g., a B747 with CF6-80 engines) [*Deidewig and Lecht,* 1994] have a larger efficiency (about 0.3) with 3.4 K higher contrail threshold temperature for cruise at 300 hPa. From Figure 2, we see that the contrails formed about 2 K above the Appleman criterion for $\eta = 0$, while the observations are more consistent with $\eta = 0.14$. Of course, the nearby radiosonde represents the flight conditions only approximately. For 10 % additional relative humidity, the threshold temperature would have been 0.4 K higher and closer to the observed value. Other plume properties may be relevant also, but the observed contrail formation is roughly consistent with the Appleman criterion if the propulsion efficiency is taken into account.

This criterion assumes condensation at small supersaturation. The necessary nuclei may form from the fuel sulfur even at the lower level of 2 ppm. The engines burn at an air/fuel mass ratio of about 70 (DLR analysis). Hence, 2 ppm fuel sulfur causes 0.028 ppm of sulfur mass concentration in the exhaust at the jet exit, which is much larger than background values (order 0.001 ppm) [*WMO,* 1995]. On the other hand, for 302.3 hPa, 34 % humidity, and $\eta = 0.14$, the Appleman theory predicts plume saturation when the exhaust gases have been mixed with ambient air at an air/fuel mass ratio of 5400 (plume 8 K warmer than environment). At this stage, the fuel sulfur mass contribution is small compared to ambient values. It may still be large enough to cause considerable generation of sulfuric acid gas in the plume between the engine and the contrails. Hence, it remains open whether 2 ppm sulfur content in the fuel is to be considered very small.

Conclusions

The observations of contrails behind a medium-sized jet aircraft with two engines using fuels with different sulfur contents, in the range typical for aviation fuels, revealed no visible differences in the contrails forming about 30 m behind the engines or in the properties of the contrails for a length up to about 1 km. Fuel, engine and aircraft parameters were sufficiently symmetric so that any masking of the impact of sulfur on contrail formation by other differences is unlikely. Hence, we conclude that variations in sulfur content between 2 and 250 ppm have no visible impact on the onset of contrail formation. This fact should be helpful to validate particle formation and jet mixing models. The simple observations do not exclude sulfur impact on the early formation of subvisible particles and on their evolution at later stages of the contrail. Tests with higher and lower sulfur contents together with quantitative measurements of particle properties within the plume remain to be performed.

The threshold of contrail formation is, however, sensitive to the engine/aircraft efficiency. Modern aircraft/engine combinations flying with higher propulsion efficiency cause contrails at higher temperatures than older engines.

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References

Appleman, H., The formation of exhaust condensation trails by jet aircraft, *Bull. Amer. Meteorol. Soc.*, *34*, 14-20, 1953.

ASTM, Standard specification for aviation fuels, D 1655-93a, *Annual Book of ASTM Standards, 05.01*, American Society for Testing and Materials, Philadelphia, PA 19103-1187, p. 555-563, 1994.

Deidewig, F., and M. Lecht, NO_x emissions from aircraft/engine combinations in flight, in DLR-Mitt. 94-06, DLR, D-51140 Köln, p. 44-49, 1994.

Frenzel, A., and F. Arnold, Sulfuric acid cluster ion formation by jet engines: implications for sulfuric acid formation and nucleation, in DLR-Mitt. 94-06, DLR, D-51140 Köln, p. 106-112, 1994.

Hagen, D. E., M. B. Trueblood, and P. D. Whitefield, A field sampling of jet exhaust aerosols, *Particulate Sci. & Techn.*, *10*, 53-63, 1992.

Hofmann, D. J., and J. M. Rosen, Balloon observations of a particle layer injected by a stratospheric aircraft at 23 km, *Geophys. Res. Lett.*, *5*, 511-514, 1978.

Iribarne, J. V., and W. L. Godson, *Atmospheric Thermodynamics*, Reidel, Dordrecht, 1981.

JANE, *Jane's all the World's Aircraft 1975-1976*, edited by J.W.R. Taylor, Jane's Yearbooks, Paulton House, London, England, 1976.

Kärcher, B., Transport of exhaust products in the near trail of a jet engine under atmospheric conditions, *J. Geophys. Res.*, *99*, 14509-14517, 1994.

Kärcher, B., Th. Peter, and R. Ottmann, Contrail formation: Homogeneous nucleation of H_2SO_4/H_2O droplets, *Geophys. Res. Lett.*, in press, 1995.

Knollenberg, R. G., Measurements of the growth of the ice budget in a persisting contrail, *J. Atmos. Sci.*, *29*, 1367-1374, 1972.

Miake-Lye, R. C., M. Martinez-Sanchez, R. C. Brown, and C. E. Kolb, Plume and wake dynamics, mixing, and chemistry behind a high speed civil transport aircraft, *J. Aircraft*, *30*, 467-479, 1993.

Miake-Lye, R. C., R. C. Brown, M. R. Anderson, and C. E. Kolb, Calculations of condensation and chemistry in an aircraft contrail, in DLR-Mitt. 94-06, DLR, D-51140 Köln, p. 274-279, 1994.

Peters, J. L., New techniques for contrail forecasting, Air Weather Service, Scott Air Force Base, Illinois 62225-5206, AWS/TR-93/001, AD-A269 686, 1993.

Pilié, R. J., and J. E. Jiusto, A laboratory study of contrails, *J. Meteorol.*, *15*, 149-154, 1958.

Pitchford, M., J. G. Hudson, and J. Hallet, Size and critical supersaturation for condensation of jet engine exhaust particles, *J. Geophys. Res.*, *96*, 20787-20793, 1991.

Reiner, T., and F. Arnold, Laboratory flow reactor measurements of the reaction $SO_3+H_2O+M \rightarrow H_2SO_4+M$: Implications for gaseous H_2SO_4 and aerosol formation in the plumes of jet aircraft, *Geophys. Res. Lett.*, *20*, 2659-2662, 1993.

Schumann, U., On the effect of emissions from aircraft engines on the state of the atmosphere, *Ann. Geophys.*, *12*, 365-384, 1994.

WMO, *Scientific Assessment of Ozone Depletion: 1994*, Global Ozone Research and Monitoring Project - Report No. 37, WMO, C. P. No. 5, CH-1211 Geneva 20, Switzerland, 1995.

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