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Key Points:
- Avoiding contrail cirrus formation mitigates aviation climate impact
- Altering contrail formation stage has large but unexplored mitigation potential
- Improved process understanding enables more reliable estimations of contrail cirrus climate impact

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The importance of contrail ice formation for mitigating the climate impact of aviation

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Abstract Aircraft contrails and the cirrus clouds arising from them contribute substantially to aviation-induced climate forcing. The share of aviation in anthropogenic climate change can be reduced by avoiding contrail cirrus formation. The mitigation potential of altering the contrail formation stage is explored using a microphysical model to show how reductions in soot particle number emissions from jet engines, reductions in mean soot particle size, and a decrease in the supersaturation of aircraft exhaust plumes substantially lowers the optical depth of young contrails thereby decreasing the occurrence, lifetime, and radiative impact of contrail cirrus. The improved scientific understanding of initial ice formation processes allows atmospheric effects of mitigation options related to contrail cirrus to be investigated in unprecedented detail, especially those associated with the use of alternative aviation fuels. This study will enable a leap forward toward more reliable simulations addressing global climatic effects of contrail-induced cloudiness.

1. Background

Due to the long-term impact of carbon dioxide (CO_2) on climate [*Solomon et al.*, 2009], reducing CO_2 emissions from aviation is of central importance. However, spreading contrails and the cirrus clouds that evolve from them—collectively known as contrail cirrus—have a greater radiative forcing (RF) today than all aviation CO_2 emissions since the first powered airplane flight [*Burkhardt and Kärcher*, 2011; *Boucher*, 2011]. The RF due to accumulated CO_2 emissions from aviation amounts to 28 mW m^{-2} corresponding to 1.6% of the total anthropogenic RF [*Lee et al.*, 2009]. Global contrail cirrus RF is assessed to be 50 mW m^{-2} [*Boucher et al.*, 2013]. These RF estimates are associated with several reference years since 2005 and therefore with different emission levels. Noting that aviation emissions have been increasing, these estimates can be combined to conclude that present-day RF from aviation CO_2 emissions and contrail cirrus alone amounts to about 5% of current anthropogenic forcing. In addition, the global RF from aircraft emissions of nitrogen oxides has been assessed to be 4.5 mW m^{-2} resulting from a combination of long-lived responses of atmospheric ozone and methane and a short-lived ozone response [*Holmes et al.*, 2011]; the direct RF due to aviation particulate (soot and sulfate) and water vapor emissions was −1.3 mW m^{-2} [*Lee et al.*, 2009] and 0.9 mW m^{-2} [*Wilcox et al.*, 2012], respectively. The climate response of all aircraft exhaust components together has been evaluated within a single model framework by *Jacobson et al.* [2013].

Future RF due to aviation may triple by 2050 [*Lee et al.*, 2009]; therefore, action on mitigation is important. Contrail-induced cloudiness is currently the largest short-lived atmospheric perturbation due to aviation. Therefore, contrail cirrus clouds are a key object for mitigation, and reducing their occurrence and atmospheric impact would make an immediate difference to the Earth's radiation budget. However, contrail cirrus and associated perturbations of natural cloudiness need to be well understood scientifically at the process level before mitigation action can be taken.

Reducing the contrail cirrus climate impact may target the contrail formation stage [*Kärcher et al.*, 2015] or the spreading stage [*Haywood et al.*, 2009]. Both stages are not independent inasmuch as spreading contrails inherit signatures of their formation for much of their lifetime [*Uthe et al.*, 1998]. The spreading stage is difficult to address since the meteorological processes creating ice supersaturation and interrelated microphysical processes affecting growth and sedimentation of ice particles [*Burkhardt and Kärcher*, 2009; *Laken et al.*, 2012] can no longer be influenced after contrail formation. Moreover, air traffic management options may not efficiently reduce contrail cirrus RF due to the prevalence and extent of ice-supersaturated areas in the upper troposphere [*Gettelman et al.*, 2006; *Lamquin et al.*, 2012] and the broad spectrum of contrail cirrus lifetimes

[*Newinger and Burkhardt*, 2012; *Irvine et al.*, 2014a] and may lead to increased fuel consumption hence CO_2 emissions [*Irvine et al.*, 2014b]. Flying entirely in the stratosphere would suppress the formation of long-lived contrails; however, the projected increase of air travel volume is stronger at low latitudes where it is difficult to reach even tropopause altitudes. Here I examine whether altering the microphysical properties of ice particles at formation potentially mitigates the aviation climate impact without the need to address air traffic routing [*Grewe et al.*, 2014] or rescheduling options [*Stuber et al.*, 2006].

Particulates present in aircraft exhaust plumes, especially black carbon (soot) particles formed during combustion of fossil fuels and emitted from aircraft jet engines, are crucial for the formation of ice in contrails [*Heymsfield et al.*, 2010]. The plume relative humidity must surpass liquid water saturation in order to form visible linear contrails [*Fahey and Schumann*, 1999]. At the high levels of supersaturation over liquid water encountered in contrail jet plumes — much above values commonly observed in the atmosphere — soot particles serve as water vapor condensation nuclei that activate into water droplets which subsequently freeze homogeneously into ice particles. The condensation nuclei activity of the largely insoluble soot particles is mainly determined by their size. Therefore, contrail ice particle properties and optical depth can be altered in significant ways by reducing the total number and mean size of emitted soot particles and by generating less supersaturated plumes. The optical depth depends on the surface area of optically active particles and contrail RF is roughly proportional to contrail optical depth [*Kärcher et al.*, 2010].

2. Methodology

This study is based on a theoretical model [*Kärcher et al.*, 2015] — vetted by numerous observational and computational studies — predicting properties of water droplets and ice particles in jet contrails in agreement with airborne measurements and laboratory studies. The model provides analytical relationships quantifying and connecting the fundamental processes that lead to the formation of ice in jet exhaust plumes within one wingspan behind cruising aircraft. It provides deeper insights into underlying physical mechanisms and might form the basis of a process-oriented parameterization for application in models with coarse resolution. Results reported here were obtained using parameters for a representative large airliner with kerosene-based propulsion technology cruising in supersaturated upper tropospheric air. Supersaturation over ice is a prerequisite for contrail persistence.

The key dynamical, thermodynamical, and microphysical elements of contrail particle formation are summarized as follows. Turbulent mixing of jet plumes with environmental air constrains plume dilution and cooling rates. Thermodynamics and mixing constrains the evolution and magnitude of plume supersaturation depending on the slope of the bulk plume mixing line, which together with the atmospheric relative humidity determines contrail formation threshold temperatures. The mixing line describes the dependence of water vapor partial pressure versus temperature in the cooling exhaust plume in a bulk thermodynamic approach [*Schumann*, 1996]. Changes in its slope are brought about by changes in air pressure and the ratio of water vapor emission index and fraction of fuel combustion energy converted into exhaust heat. While air pressure and ambient relative humidity over ice have only a small impact on our results, since the ice formation process is dominated by the high levels of emitted water vapor, the choices 250 hPa and 115%, representative for most flight routes, affect the slope of the plume mixing line, G, and the thermodynamic contrail formation threshold temperature, $\Theta = 225.2$ K (for a baseline value $G = 1.64$ Pa K^{-1}). Atmospheric temperature, T, is varied from 225 K to 210 K covering the range of mean values prevalent in the northern extratropical midlatitudes at cruise altitudes.

Figure 1 shows the maximum saturation ratio over liquid water, S_{max} (solid curves), attained in a diluting jet plume without condensation and the temperature offsets, $\Delta T = \Theta - T$ (dashed curves), as a function of G for three different ambient temperatures. Contrails form in supersaturated plumes if $S_{max} > 1$, or equivalently, if the air is colder than the threshold, $\Delta T > 0$. The peak supersaturation is a measure of the potential to activate plume particles into water droplets. A minimum slope, determined by $S_{max} = 1$ or $\Delta T = 0$, is necessary to form contrails for given ambient conditions. As G increases or T decreases, peak supersaturation rises enhancing water droplet and ultimately ice formation. The temperature offsets allow to distinguish between near-threshold cases ($\Delta T < 1-2$ K) and below-threshold cases ($\Delta T > 1-2$ K). The separation between these regimes depends on the total number, size distribution, and hygroscopicity of the plume particles. Near-threshold contrails are optically thinnest since water activation diminishes as $S_{max} \rightarrow 1$.

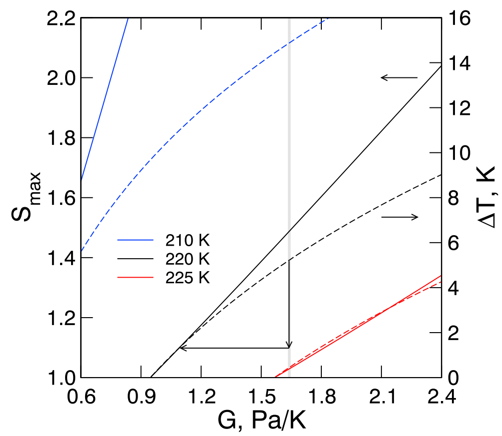

Figure 1. Peak saturation ratio over liquid water (fractional relative humidity) in jet exhaust plumes without condensation (solid curves) and offsets between the thermodynamic formation threshold temperature and the ambient temperature (dashed) as a function of the slope of the plume mixing line. Calculations assume a pressure of 250 hPa and an ambient relative humidity over ice of 115%. Three sets of curves are shown for cold (blue), close to average (black), and warm (red) ambient temperatures. The grey line marks the value of the slope, 1.64 Pa K^{-1}, used as a baseline. Long arrows indicate a decrease in the threshold temperature by about 4 K (at 220 K) brought about by lowering *G* by a factor 2/3.

Two particle types with mean dry sizes of some 10 nm across act as water condensation nuclei: emitted soot particles and entrained atmospheric aerosol particles. Nonvolatile soot particles are rather hydrophobic; however, they can be water activated forming early visible contrails even when using fuel with a sulfur content two orders of magnitude below average values [*Busen and Schumann*, 1995]. Entrained particles are more hygroscopic than soot and therefore easier to activate but are much less abundant by number. At the onset of contrail formation plume supersaturation increases by cooling, while soot numbers decrease due to dilution and the number of atmospheric particles increases due to entrainment of ambient air.

The number of plume particles activated into water droplets at any given plume supersaturation and rate of plume cooling is calculated dependent on particle size and chemical composition. In the subsaturated plume, the water activity of soot particles is largely determined by scavenging of smaller liquid plume particles and uptake of sulfuric acid molecules derived from aircraft sulfur dioxide emissions [*Kärcher*, 1998]. Size distribution and chemical composition of combustion aerosol particles in the exhaust plume are affected by van-der-Waals forces and fractal geometry enhancing coagulation rates [*Hagen et al.*, 1991; *Jacobson and Seinfeld*, 2004]. The underlying heterocoagulation and condensation processes are not explicitly modeled, but their effect on droplet formation is taken into account by constraining a solubility model [*Petters and Kreidenweis*, 2007]. Self-coagulation among soot particles, water droplets, or ice particles is not capable of affecting ice particle numbers significantly in contrails at the prevailing particle number concentrations due to the short time available for contrail particle formation and, in the case of ice particles, additionally due to the presumably very low sticking efficiency [*Connolly et al.*, 2012].

Droplet formation is terminated once the rise of supersaturation over liquid water is halted by water vapor condensation on activated nuclei. Due to large cooling rates, all water droplets freeze homogeneously at plume temperatures 229–232 K, i.e., lower than in natural liquid-phase clouds [*Kärcher and Seifert*, 2016] due to the small size of water droplets (several 100 nm) and the short freezing time scale (a few 0.1 s) in contrails. Ice formation occurs in a burst because ice particle growth is insufficient to deplete the supersaturation sustaining the water droplets during freezing. Consistent with observed contrail formation temperatures, exhaust soot particles act in the homogeneous ice nucleation mode as passive inclusions in the water droplets without the requirement to serve as heterogeneous ice nuclei. The model allows water uptake on contrail particles to become kinetically hindered in the case of low particle numbers ($<10^{14}$/kg fuel).

Nanometer-sized (ultrafine) particles are another source of condensation nuclei in young jet exhaust plumes [*Brock et al.*, 2000]. They are generated by ion-mediated nucleation and uptake of sulfuric acid and low-volatile organic vapors prior to contrail droplet and ice formation [*Yu and Turco*, 1997]. Their mean dry size is determined by the amount of oxidized sulfur and condensable hydrocarbons present in fresh exhaust and can therefore be reduced by desulfurization of the jet fuel. They do not readily serve as droplet nuclei in the presence of much larger emitted and entrained particles acting as condensation sinks. If soot numbers are reduced below the level of entrained particles and atmospheric temperatures are low, some 10^{17} ice particles per kilogram of fuel burnt can form on ultrafine particles [*Kärcher and Yu*, 2009]. The sulfur content is assumed to be sufficiently low and the particulate organic matter in the exhaust to be hydrophobic enough inhibiting ultrafine particles to serve as contrail droplet nuclei. Aircraft jet engines may also emit metal particles and lubricant aerosol particles depending on maintenance and power setting, but those particles are too few by number and occur too intermittently to explain contrail formation.

Soot number emission indices, EI_s, vary depending on fuel combustion characteristics and in-flight jet engine performance. Current values lie in the range 10^{14}–10^{15}/kg fuel [*Anderson et al.*, 2011] within the soot-rich regime ($EI_s > 10^{13}$/kg fuel); the exhaust is considered to be soot poor for $EI_s < 10^{12}$/kg fuel. The boundary between both regimes is determined by the number of entrained particles: 600 particles per cubic centimeter of ambient air. Guided by measurements [*Petzold et al.*, 2005], we assume exhaust soot particles to be lognormally distributed with a mode radius of 12.5 nm and a geometric standard deviation of 1.6 and to be internally mixed with a 1% volume fraction of water-soluble sulfate and organic exhaust compounds.

Mean ice particle radii and contrail optical depths are reported at a plume age of 1 s, corresponding to a distance of several wingspans behind the contrail-producing aircraft, i.e., shortly after ice formation is completed. Optical depth of contrail ice particles due to visible light scattering is computed based on Mie theory in the absence of more accurate optical models for micrometer-sized ice particles, neglecting absorption of soot cores due to small soot-to-ice volume ratios. Optical depth values calculated for a given ice particle size have been integrated over a lognormal size distribution with a geometric standard deviation of 1.4 to account for turbulent mixing of air parcels with different microphysical histories. Taken after the initial growth phase depleting plume supersaturation over ice in the case of high particle numbers, peak optical depth values predicted here for current soot emissions (0.5–2) partly overlap with measurements (0.1–1) in older contrails after further plume dilution [*Kärcher et al.*, 2009] or sublimation [*Voigt et al.*, 2011].

Emission indices are not affected by plume dilution; the apparent number emission index of ice particles, AEI, is directly comparable to EI_s. The term "apparent" indicates that contrail ice particles do not belong to emissions. Plume-averaged susceptibilities of ice variable x, $\sigma_x = \partial \ln(x)/\partial \ln(EI_s)$, are calculated allowing variables besides x to vary upon changing EI_s in order to capture correlations between microphysical processes. For a given small relative change in EI_s, σ_x quantifies the dependence on (or relative change in) x. Ice number, radius, and optical depth dependencies ($x = \mathrm{AEI}, r, \tau$) are discussed, which may be thought of as the effect of changes in soot emissions on contrail persistence in ice-supersaturated areas and on the RF of young contrails, respectively.

3. Discussion of Results

The results suggest significant dependencies of apparent ice particle number emission index, mean radius, and optical depth on the soot particle number emission index, and, close to the contrail formation threshold, on atmospheric temperature (Figures 2a–2c). Ice number and size affect, via growth and sedimentation, radiative response and life span of the evolving contrail cirrus.

Ice numbers decrease about 100-fold from soot-rich to soot-poor conditions in the baseline below-threshold cases ($T = 210$ K, 220 K) approaching constant low values determined by the number of entrained atmospheric particles for $EI_s < 10^{12}$/kg fuel. In a colder atmosphere, contrail particle formation takes place at earlier plume ages where increasingly fewer particles acting as contrail nuclei are entrained into the plume. Ice numbers rise markedly as the fuel becomes soot rich ($EI_s > 10^{13}$/kg fuel), because ice particle formation is not limited by the plume cooling rate; the more soot particles are present in the exhaust, the more can be water activated and freeze. While AEI approaches EI_s in below-threshold contrails, ice numbers decline strongly in the near-threshold case ($T = 225$ K), since water activation and freezing are impeded by much smaller plume supersaturation rendering soot particle reductions less effective in reducing ice numbers.

Mean ice particle radii approach constant high values in soot-poor exhaust due to the control of ice formation by the low number of entrained particles. They also increase with lower temperatures due to higher plume supersaturation. Changes in total ice number and mean size are anticorrelated in soot-rich conditions where water vapor uptake on contrail particles is fast and the same amount of vapor is distributed among

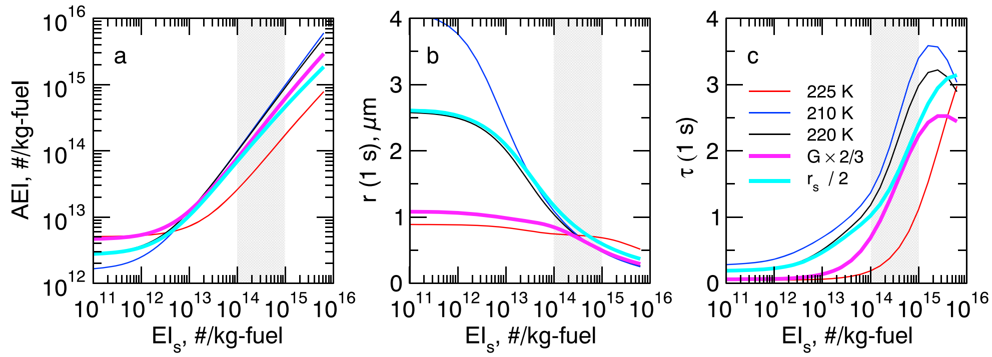

Figure 2. Changes in ice particle properties of young contrails imposed by changes in aircraft soot emissions and exhaust plume supersaturation. (a) Apparent total number emission index, (b) mean radius, and (c) visible optical depth of contrail ice particles at 1 s plume age versus soot number emission index. Baseline cases (thin curves) have been calculated for different offsets between the thermodynamic formation threshold temperature and the ambient temperature: ΔT = 14.9 K for T = 210 K (blue); 5.2 K for 220 K (black); 0.45 K for 225 K (red), for a mixing line slope G = 1.64 Pa K^{-1}, and mean soot particle radius r_s=12.5 nm. Thick curves are results from sensitivity studies of the case 220 K, either brought about by reducing G by a factor 2/3 (magenta) or calculated with halved r_s (cyan). Grey areas indicate the approximate range of soot number emission indices for the current fleet.

more particles. This correlation weakens due to kinetic limitations of water vapor condensation as conditions become soot poor. Contrail optical depth declines at the highest EI_s values. Optical depth attains small constant values in soot-poor exhaust and diminishes strongly near the formation threshold consistent with decreased ice particle surface area. For $EI_s > 3 \times 10^{15}$/kg fuel, the decrease in size reduces the light scattering efficiency per ice particle outweighing the increase in number in determining optical depth.

With the help of Figures 3a–3c, the conditions in which contrail ice properties are particularly sensitive to changes in soot emissions are identified. Number susceptibility is large (>0.9) in soot-rich conditions for below-threshold cases and declines rapidly as EI_s decreases. Ice number becomes nearly unaffected by soot reductions once soot-poor conditions are approached. Due to the number-size anticorrelation, radius susceptibility exhibits a reverse but otherwise similar pattern. Optical depth susceptibility is moderate (≈ 0.4) in the below-threshold cases but may nonetheless be effectively reduced by lowering EI_s over a wide range of soot emissions (10^{12}–10^{15}/kg fuel). Optical depth susceptibility is high (≈ 0.8) at current emission levels near the formation threshold. Near the threshold, susceptibilities shift to larger EI_s values and ice number susceptibility is slightly reduced.

Lowering the slope of the thermodynamic plume mixing line, G, reduces the plume supersaturation and thereby the number of freezing droplets. Moreover, smaller G values reduce the formation threshold temperature and thereby lower the frequency of contrail occurrence. In a sensitivity study, G is reduced by a factor of 2/3 lowering the contrail formation threshold by about 4 K, leading to ΔT = 1.3 K (Θ = 221.3 K) and thereby turning the baseline below-threshold case at T = 220 K into a near-threshold case (arrows in Figure 1).

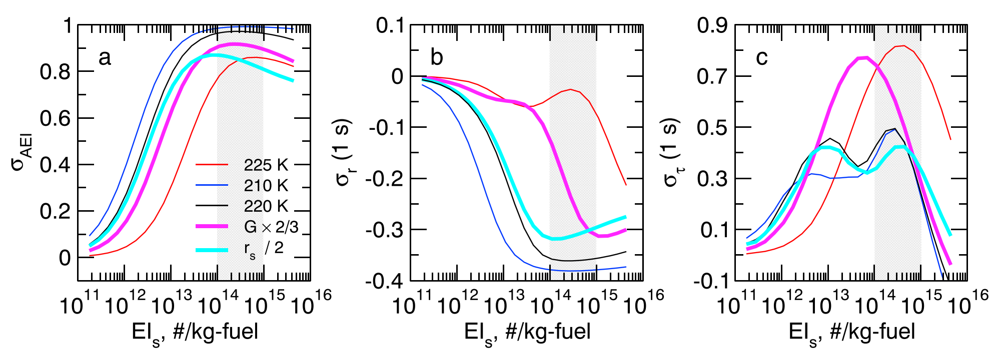

Figure 3. Dependence of contrail ice particle (a) number, (b) radius, and (c) optical depth on the soot number emission index. Conditions as in Figure 2.

The results (thick magenta curves) demonstrate that ice number and radius dependencies are still high for current emissions. Relative to the baseline-*G* case, the mean ice particle radius is smaller below about $EI_s = 10^{14}$/kg fuel. Optical depth can be significantly reduced below $EI_s = 10^{15}$/kg fuel.

Smaller aerosol particles activate into water droplets at higher supersaturation due to the curvature (Kelvin) effect. Lowering the mean soot particle size therefore decreases contrail particle numbers. In a second sensitivity study of case $T = 220$ K, retaining the baseline value for *G*, the mean soot particle radius is halved. The results (thick cyan curves) show that ice number and optical depth are markedly reduced for current soot emission levels, while the mean ice particle radius is hardly affected due to the large amount of condensable water vapor provided by the emissions. Susceptibility parameters are only slightly altered.

4. Implications for Mitigation

The findings discussed above indicate how the climate impact of contrail cirrus might be reduced. The largest possible reduction in ice numbers is achieved by about a 100-fold decrease in current soot number emissions, in combination with a reduction of the thermodynamic mixing line slope by one third or with halving the mean soot particle size. These changes decrease the optical depth of young contrails by more than an order of magnitude. The associated fourfold to fivefold increase in radius from soot-rich to soot-poor conditions (for $T = 220$ K and baseline value of *G*) increases initial ice particle terminal fall speeds by about a factor of 20, thereby reducing contrail cirrus lifetimes due to enhanced sedimentation. Reducing soot numbers less than 100-fold is a less effective means to reduce contrail cirrus formation. Reductions of EI_s in soot-poor exhaust do not lead to further changes in contrail properties as ice formation in contrails is then controlled by the availability of atmospheric aerosol particles.

The number of contrail ice particles can be reduced by cutting the number of emitted soot particles and also by reducing the mean soot particle mass or size. Particulate mass alone is not an appropriate indicator for ice formation. Validated number-size relationships for exhaust soot particles in cruising conditions are not available. A novel practice to quantify soot particle sizes and number emission indices at cruise should replace the current methodology of quantifying smoke number at ground—a poor surrogate for the mass of particulate matter in aircraft exhaust [*Stettler et al.*, 2013; *Lobo et al.*, 2015]. While accurate predictions of soot particle number and size varies with combustor design and engine operation and are therefore difficult to obtain, they are needed in order to provide aircraft emission inventories with data enabling proper initialization of global model simulations. The availability of black carbon emission measurements from gas turbines heightens the prospect of obtaining predictive relationships of soot number emission reductions from (blended) alternative fuels [*Speth et al.*, 2015].

Reducing the plume mixing line slope poses a technological challenge since modern jet engines—operating at high pressure ratios and high combustion temperatures in order to reduce fuel burn and associated CO_2 emissions—tend to produce cooler exhaust which, in turn, increases *G*. Steeper plume mixing line gradients causes contrails to form at higher atmospheric temperatures [*Schumann et al.*, 2000] thereby increasing the frequency of contrail occurrence. The amount of soot emitted by jet engines results from two opposing effects: formation (nucleation and growth) and burnoff (oxidation) of polycyclic aromatic hydrocarbons [*Richter and Howard*, 2000]. Higher combustion pressures favor soot formation. The lean combustor technique—designed to minimize nitrogen oxide emissions—also leads to reduced soot number emissions already quantified in ground measurements [*European Aviation Safety Agency*, 2013]. Other options to reduce *G* have been proposed [*Noppel and Singh*, 2007].

In the light of these challenges, the use of alternative aviation fuels might offer a clue for mitigation. Ground-based measurements indicate up to tenfold reduction in soot number emissions when using pure synthetic jet fuels [*Moore et al.*, 2015] and also significant reductions in mean soot particle sizes [*Beyersdorf et al.*, 2014]. The use of fuel blends in order to meet technical standards for aviation turbine fuels leads to smaller reductions; it remains to be shown whether those or even larger reductions can be achieved in cruise conditions. Studies describing the outcomes of related measurements at cruise and within contrails are under way [*National Aeronautics and Space Administration*, 2015]; [*Deutsches Zentrum für Luft- und Raumfahrt*, 2015]. Besides causing fewer pollutant emissions, alternative fuels may have different hydrogen content (i.e., water vapor emissions) affecting *G* and thereby contrail occurrence and plume supersaturation.

Combining soot emission reductions leading to soot-poor exhaust with desulfurization of aviation fuel renders mitigation more effective as this would eliminate the generation of cold and—especially in high latitudes—potentially long-lived contrail cirrus due to the inhibition of ultrafine plume particle activation. It would also make exhaust soot particles poorer droplet nuclei therefore enhancing the number of shorter-lived near-threshold contrails; however, no significant effect on ice numbers is found upon lowering the soluble soot mass fraction, since exhaust soot particles are already treated as largely insoluble particles to make calculated contrail optical depth values consistent with observations. As an added benefit, reducing soot particle emissions would alleviate hypothesized heterogeneous ice nucleation effects on large-scale clouds [*Zhou and Penner*, 2014]. Economic and environmental benefits of the use of desulfurized aviation fuel have recently been assessed [*Barrett et al.*, 2012]. Moreover, desulfurization of aviation fuel would alleviate aerosol effects on low-level clouds attributed to particle emissions from aircraft [*Unger*, 2011; *Righi et al.*, 2013; *Gettelman and Chen*, 2013].

5. Future Developments

Attempts to include contrail-induced cloudiness in mitigation have long been frustrated by a poor scientific understanding of atmospheric effects [*Green*, 2009]. The improved scientific understanding of contrail ice formation should be transferred into cloud schemes of climate models; technological research and development efforts should be enhanced in order to realize the proposed changes to emissions and plume conditions. Evaluating the benefits of—and trade-offs between—mitigation options requires more scientific scrutiny, including studies capable of addressing this topic in the context of anthropogenic climate change [*Karnauskas et al.*, 2015; *Williams*, 2016]. The present study supports such efforts by laying the groundwork for subsequent global climate studies addressing mitigation effects due to alteration of the contrail formation stage with its large but unexplored potential for limiting the environmental impact of future air transportation.

In future atmospheric research, the atmospheric response to reductions in initial contrail ice number should be explored using global climate models with an interactive parameterization scheme for contrail ice formation depending on variable soot particle number emissions and atmospheric conditions. This is necessary in order to evaluate the integrated climate impact of an altered formation stage. It would be interesting to study how upper tropospheric aerosol particles affect contrail cirrus through their effect on ice number in soot-poor conditions, because the total number of atmospheric particles is geographically and seasonally variable. Since air temperatures at high flight levels at tropical and subtropical latitudes are warmer than at higher latitudes and therefore more often include formation thresholds, studies contrasting tropical versus extratropical responses to contrail cirrus perturbations could unravel effects of enhanced occurrence of near-threshold contrails on the overall climate impact. The results will depend on the probability distribution of air temperatures at cruise relative to local contrail formation threshold temperatures. Liquid cryogenic fuels with low to zero carbon content are particularly interesting to study owing to their opposing effects on contrail longevity and frequency of occurrence. On the one hand, those fuels have no particulate emissions minimizing ice particle numbers and maximizing mean sizes thereby enhancing sedimentation loss rates. On the other hand, they increase G thereby forming contrails over a larger range of cruise altitudes. Overseeding the exhaust with excess contrail nuclei may be problematic since the resulting increased lifetime of contrail cirrus due to reduced initial ice particle sizes might outweigh the initial reduction in optical depth in affecting RF.

In the case of low soot emissions, organic matter in ultrafine plume particles should be identified and its propensity for water vapor condensation specified. Effects of enhanced coagulation between combustion particles on contrail droplet and ice formation could be studied dependent on the sulfur content of the jet fuel. Microphysical models might be tested with observations of ultrafine particle and contrail ice formation from soot-poor exhaust. Moreover, high-resolution simulations of dissipating aircraft wakes could be used to study how changes in formation conditions affect the subsequent processing of newly formed contrail ice particles in wake vortices. The representation of contrail cirrus within large-scale models improved significantly. Many global climate models include a number and mass-based representation of cloud hydrometeors and should be coupled with a new generation of soot emission inventories and parameterizations of initial contrail ice properties. A physically based description of the evolution of contrail ice particles in decaying aircraft wakes should be linked to ice nucleation in order to arrive at a self-consistent treatment of all subgrid-scale processes including their couplings and feedbacks in order to be useful for adaption and integration into the cloud schemes of global models.

Along with these efforts, engineering science research into aviation fuel design and propulsion technology needs to investigate which of the proposed emission and plume changes might be technically and economically feasible and to which degree and on which time horizon the most promising mitigation solutions can be realized. The development of alternative fuels for commercial aviation is motivated in part by the potential increase in environmental performance they offer. Reduced emissions of soot, sulfate, and water vapor from alternative fuels tailored for future midterm jet aircraft and engines offer the promise of a substantial mitigation of the aviation climate impact, if life cycle environmental questions associated with the manufacturing of those fuels — most notably regarding CO_2 emissions — can be addressed. Accompanying measures unrelated to fuel and its combustion might include changes in jet engine architecture and airframe concepts affecting plume and wake dynamics and hence the transition of contrails into cirrus clouds.

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References

Anderson, B. E., et al. (2011), *Alternative Aviation Fuel Experiment (AAFEX), Technical Memorandum NASA/TM-2011-217059*, National Aeronautics and Space Administration, Langley Research Center, Hampton, VA., Washington, D. C.

Barrett, S. R. H., et al. (2012), Public health, climate, and economic impacts of desulfurizing jet fuel, *Environ. Sci. Technol.*, *46*, 4275–4282.

Beyersdorf, A. J., et al. (2014), Reductions in aircraft particulate emissions due to the use of Fischer-Tropsch fuels, *Atmos. Chem. Phys.*, *14*, 11–23.

Boucher, O. (2011), Seeing through contrails, *Nat. Clim. Change*, *1*, 24–25.

Boucher, O., et al. (2013), Clouds and Aerosols, in *Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change*, edited by T. F. Stocker et al., pp. 571–658, Cambridge Univ. Press, Cambridge, U. K., and New York, doi:10.1017/CBO9781107415324.016.

Brock, C. A., F. Schröder, B. Kärcher, A. Petzold, R. Busen, and M. Fiebig (2000), Ultrafine particle size distributions measured in aircraft exhaust plumes, *J. Geophys. Res.*, *105*, 26,555–26,568.

Burkhardt, U., and B. Kärcher (2009), Process-based simulation of contrail cirrus in a global climate model, *J. Geophys. Res.*, *114*, D16201, doi:10.1029/2008JD011491.

Burkhardt, U., and B. Kärcher (2011), Global radiative forcing from contrail cirrus, *Nat. Clim. Change*, *1*, 54–58, doi:10.1038/NCLIMATE1068.

Busen, R., and U. Schumann (1995), Visible contrail formation from fuels with different sulfur contents, *Geophys. Res. Lett.*, *22*, 1357–1360.

Connolly, P. J., C. Emersic, and P. R. Field (2012), A laboratory investigation into the aggregation efficiency of small ice crystals, *Atmos. Chem. Phys.*, *12*, 2055–2076.

Deutsches Zentrum für Luft- und Raumfahrt (2015), *Deutsches Zentrum für Luft- und Raumfahrt e.V.*, German Aerospace Center, Corporate Communications, Cologne, Germany. [Available at http://www.dlr.de/dlr/en/desktopdefault.aspx/tabid-10261/371_read-10228/#/gallery/14652.]

European Aviation Safety Agency (2013), *Table 1 in: Report EASA.2010.FC10-SC03*, European Aviation Safety Agency, Cologne, Germany. [Available at https://easa.europa.eu/document-library/research-projects.]

Fahey, D. W., and U. Schumann (1999), Aviation-Produced Aerosols and Cloudiness, in *Aviation and the Global Atmosphere. A Special Report of IPCC Working Groups I and III. Intergovernmental Panel on Climate Change*, edited by J. E. Penner, Cambridge Univ. Press, Cambridge, U. K.

Gettelman, A., E. J. Fetzer, A. Eldering, and F. W. Irion (2006), The global distribution of supersaturation in the upper troposphere from the Atmospheric Infrared Sounder, *J. Climate*, *19*, 6089–6103.

Gettelman, A., and C. Chen (2013), The climate impact of aviation aerosols, *Geophys. Res. Lett.*, *40*, 2785–2789.

Green, J. E. (2009), The potential for reducing the impact of aviation on climate, *Technol. Anal. Strat. Manag.*, *21*, 39–59.

Grewe, V., et al. (2014), Aircraft routing with minimal climate impact: The REACT4C climate cost function modelling approach (V1.0), *Geosci. Model Dev.*, *7*, 175–201.

Hagen, D. E., M. B. Trueblood, and J. Podzimek (1991), Combustion aerosol scavenging, *Atmos. Environ.*, *25A*, 2581–2586.

Haywood, J. M., R. P. Allan, J. Bornemann, P. M. Forster, P. N. Francis, S. Milton, G. Rädel, A. Rap, K. P. Shine, and R. Thorpe (2009), A case study of the radiative forcing of persistent contrails evolving into contrail-induced cirrus, *J. Geophys. Res.*, *114*, D24201, doi:10.1029/2009JD012650.

Heymsfield, A., D. Baumgardner, P. DeMott, P. Forster, K. Gierens, and B. Kärcher (2010), Contrail microphysics, *Bull. Am. Meteorol. Soc.*, *91*, 465–472.

Holmes, C. D., Q. Tang, and M. J. Prather (2011), Uncertainties in climate assessment for the case of aviation NO, *Proc. Natl. Acad. Sci. U.S.A.*, 10,997–11,002.

Jacobson, M. Z., and J. H. Seinfeld (2004), Evolution of nanoparticle size and mixing state near the point of emission, *Atmos. Environ.*, *38*, 1839–1850.

Jacobson, M. Z., J. T. Wilkerson, A. D. Naiman, and S. K. Lele (2013), The effects of aircraft on climate and pollution. Part II: 20-year impacts of exhaust from all commercial aircraft worldwide treated individually at the subgrid scale, *Faraday Disc.*, *165*, 369–382.

Irvine, E. A., B. J. Hoskins, and K. P. Shine (2014a), A Lagrangian analysis of ice-supersaturated air over the North Atlantic, *J. Geophys. Res.*, *119*, 90–100, doi:10.1002/2013JD020251.

Irvine, E. A., B. J. Hoskins, and K. P. Shine (2014b), A simple framework for assessing the trade-off between the climate impact of aviation carbon dioxide emissions and contrails for a single flight, *Environ. Res. Lett.*, *9*, 064021.

Kärcher, B. (1998), On the potential importance of sulfur-induced activation of soot particles in nascent jet aircraft exhaust plumes, *Atmos. Res.*, *46*, 293–305.

Kärcher, B., and A. Seifert (2016), On homogeneous ice formation in liquid clouds, *Q. J. R. Meteorol. Soc.*, *142*, doi:10.1002/qj.2735.

Kärcher, B., and F. Yu (2009), Role of aircraft soot emissions in contrail formation, *Geophys. Res. Lett.*, *36*, L01804, doi:10.1029/2008GL036649.

Kärcher, B., U. Burkhardt, S. Unterstrasser, and P. Minnis (2009), Factors controlling contrail cirrus optical depth, *Atmos. Chem. Phys.*, *9*, 6229–6254.

Kärcher, B., U. Burkhardt, M. Ponater, and C. Frömming (2010), Importance of representing optical depth variability for estimates of global line-shaped contrail radiative forcing, *Proc. Natl. Acad. Sci. U.S.A.*, *107*, 19,181–19,184.

Kärcher, B., U. Burkhardt, A. Bier, L. Bock, and I. J. Ford (2015), The microphysical pathway to contrail formation, *J. Geophys. Res.*, *120*, doi:10.1002/2015JD023491.

Karnauskas, K. B., J. P. Donnelly, H. C. Barkley, and J. E. Martin (2015), Coupling between air travel and climate, *Nat. Clim. Change*, *5*, doi:10.1038/NCLIMATE2715.

Laken, B. A., E. Pallé, D. R. Kniveton, C. J. R. Williams, and D. A. Kilham (2012), Contrails developed under frontal influences of the North Atlantic, *J. Geophys. Res.*, *117*, D11201, doi:10.1029/2011JD017019.

Lamquin, N., C. J. Stubenrauch, K. Gierens, U. Burkhardt, and H. Smit (2012), A global climatology of upper-tropospheric ice supersaturation occurrence inferred from the Atmospheric Infrared Sounder calibrated by MOZAIC, *Atmos. Chem. Phys.*, *12*, 381–405.

Lee, D. S., D. W. Fahey, P. M. Forster, P. J. Newton, R. C. N. Wit, L. L. Lim, B. Owen, and R. Sausen (2009), Aviation and global climate change in the 21st century, *Atmos. Environ.*, *43*, 3520–3537.

Lobo, P., L. Durdina, C. J. Smallwood, T. Rindlisbacher, F. Siegerist, E. A. Black, Z. H. Yu, A. A. Mensah, D. F. Hagen, and R. C. Miake-Lye (2015), Measurements of aircraft engine non-volatile PM emissions: Results of the Aviation-Particle Regulatory Instrumentation Demonstration Experiment (A-PRIDE) 4 campaign, *Aerosol Sci. Technol.*, *49*, 472–484.

Moore, R. H., et al. (2015), Influence of jet fuel composition on aircraft engine emissions: A synthesis of aerosol emissions data from the NASA APEX, AAFEX, and ACCESS missions, *Energy Fuels*, *29*, 2591–2600.

National Aeronautics and Space Administration (2015), *National Aeronautics and Space Administration*, Washington, D. C. [Available at http://www.nasa.gov/aero/access-ii-confirms-jet-biofuel-burns-cleaner.]

Newinger, C., and U. Burkhardt (2012), Sensitivity of contrail cirrus radiative forcing to air traffic scheduling, *J. Geophys. Res.*, *117*, D10205, doi:10.1029/2011JD016736.

Noppel, F, and R. Singh (2007), Overview on contrail and cirrus cloud avoidance technology, *J. Airc.*, *44*, 1721–1726.

Petters, M. D., and S. M. Kreidenweis (2007), A single parameter representation of hygroscopic growth and cloud condensation nucleus activity, *Atmos. Chem. Phys.*, *7*, 1961–1971.

Petzold, A., et al. (2005), On the effects of organic matter and sulphur-containing compounds on the (CCN) activation of combustion particles, *Atmos. Chem. Phys.*, *5*, 3187–3203.

Richter, H., and J. B. Howard (2000), Formation of polycyclic aromatic hydrocarbons and their growth to soot — A review of chemical reaction pathways, *Prog. Energy Combust. Sci.*, *26*, 565–608.

Righi, M., J. Hendricks, and R. Sausen (2013), The global impact of the transport sectors on atmospheric aerosol: Simulations for year 2000 emissions, *Atmos. Chem. Phys.*, *13*, 9939–9970.

Schumann, U. (1996), On conditions for contrail formation from aircraft exhausts, *Meteorol. Z.*, *5*, 4–23.

Schumann, U., R. Busen, and M. Plohr (2000), Experimental test of the influence of propulsion efficiency on contrail formation, *J. Aircr.*, *37*, 1083–1087.

Solomon, S., G.-K. Plattner, R. Knutti, and P. Friedlingstein (2009), Irreversible climate change due to carbon dioxide emissions, *Proc. Natl. Acad. Sci. U.S.A.*, *106*, 1704–1709.

Speth, R. L., C. Rojo, R. Malina, and S. R. H. Barrett (2015), Black carbon emissions reductions from combustion of alternative jet fuel, *Atmos. Environ.*, *105*, 37–42.

Stettler, M. E. J., A. M. Boies, A. Petzold, and S. R. H. Barrett (2013), Global civil aviation black carbon emissions, *Environ. Sci. Technol.*, *47*, 10,397–10,404.

Stuber, N., P. Forster, G. Rädel, and K. Shine (2006), The importance of the diurnal and annual cycle of air traffic for contrail radiative forcing, *Nature*, *441*, 864–867.

Unger, N. (2011), Global climate impact of civil aviation for standard and desulfurized jet fuel, *Geophys. Res. Lett.*, *38*, L20803, doi:10.1029/2011GL049289.

Uthe, E. E., N. B. Nielsen, and T. E. Osberg (1998), Airborne scanning Lidar observations of aircraft contrails and cirrus clouds during SUCCESS, *Geophys. Res. Lett.*, *25*, 1339–1342.

Voigt, C., U. Schumann, P. Jessberger, T. Jurkat, A. Petzold, J.-F. Gayet, M. Krämer, T. Thornberry, and D. W. Fahey (2011), Extinction and optical depth of contrails, *Geophys. Res. Lett.*, *39*, L11806, doi:10.1029/2011GL047189.

Wilcox, L. J., K. P. Shine, and B. J. Hoskins (2012), Radiative forcing due to aviation water vapour emissions, *Atmos. Environ.*, *63*, 1–13.

Williams, P. D. (2016), Transatlantic flight times and climate change, *Environ. Res. Lett.*, *11*, 024008, doi:10.1088/1748-9326/11/2/024008.

Yu, F., and R. P. Turco (1997), The role of ions in the formation and generation of particles in aircraft plumes, *Geophys. Res. Lett.*, *24*, 1927–1930.

Zhou, C., and J. E. Penner (2014), Aircraft soot indirect effect on large-scale cirrus clouds: Is the indirect forcing by aircraft soot positive or negative?, *J. Geophys. Res.*, *119*, 11,303–11,320, doi:10.1002/2014JD021914.