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# Impact of biofuels on contrail warming

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## LETTER

### Impact of biofuels on contrail warming

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#### Abstract

Contrails and contrail-cirrus may be the largest source of radiative forcing (RF) attributable to aviation. Biomass-derived alternative jet fuels are a potentially major way to mitigate the climate impacts of aviation by reducing lifecycle CO<sub>2</sub> emissions. Given the up to 90% reduction in soot emissions from paraffinic biofuels, the potential for a significant impact on contrail RF due to the reduction in contrail-forming ice nuclei (IN) remains an open question. We simulate contrail formation and evolution to quantify RF over the United States under different emissions scenarios. Replacing conventional jet fuels with paraffinic biofuels generates two competing effects. First, the higher water emissions index results in an increase in contrail occurrence ( $\sim +8\%$ ). On the other hand, these contrails are composed of larger diameter crystals ( $\sim +58\%$ ) at lower number concentrations ( $\sim -75\%$ ), reducing both contrail optical depth ( $\sim -29\%$ ) and albedo ( $\sim -32\%$ ). The net changes in contrail RF induced by switching to biofuels range from  $-4\%$  to  $+18\%$  among a range of assumed ice crystal habits (shapes). In comparison, cleaner burning engines (with no increase in water emissions index) result in changes to net contrail RF ranging between  $-13\%$  and  $+5\%$  depending on habit. Thus, we find that even 67% to 75% reductions in aircraft soot emissions are insufficient to substantially reduce warming from contrails, and that the use of biofuels may either increase or decrease contrail warming—contrary to previous expectations of a significant decrease in warming.

## 1. Introduction

Condensation trails form in the wake of aircraft under certain meteorological conditions of temperature and humidity (Schumann 1996), predominantly through the formation of water droplets around particles emitted by aircraft, which serve as ice nuclei (IN) that lead to ice crystals predominantly through heterogeneous freezing. Aircraft-emitted IN are mainly nonvolatile particles, in particular soot (Kärcher and Yu 2009, Schumann 2012), although the role of volatile particles is uncertain especially in low soot conditions (Kärcher and Yu 2009), and homogeneous nucleation is potentially significant as well. In an ice-supersaturated atmosphere, contrails can last several hours, evolving into contrail cirrus indistinguishable from natural cirrus (Burkhardt and Kärcher 2011, Haywood *et al* 2009). Acting like high, thin ice clouds, contrails are

more effective at trapping outgoing longwave radiation than at reflecting incoming shortwave radiation back to space (Burkhardt and Kärcher 2011, Hartmann *et al* 1992, IPCC 2013, Schumann and Graf 2013). As a result, contrails are estimated to be the largest radiative forcing (RF) component attributable to aviation (Burkhardt and Kärcher 2011, IPCC 2013). Biofuels have been identified as an opportunity to mitigate aviation's climate impact by reducing fossil CO<sub>2</sub> emissions. Based on measurements of reduced soot emissions at cruise (Moore *et al* 2017) and model results showing reductions in contrail optical depth (Kärcher 2016), the potential of biofuels to reduce the climate impact of contrails has become an important question. Here, we conduct the first assessment of the effect of biofuels on contrail radiative forcing, as well as the related impact of reductions in soot emissions associated with improvements in combustor technology.

**Table 1.** Fuel properties and total, daytime and nighttime RF for all fuel/emissions configurations simulated by CERM over the US (57° W–140° W longitude and 16° N–58° N latitude). The range of RF values represent uncertainty dependent on the assumption of ice crystal habit. Percentages are reported for pairwise comparisons.

	Fuel properties			Radiative impact of contrails/contrail cirrus over the US domain		
	El <sub>H<sub>2</sub>O</sub> [kg kg <sub>f</sub> <sup>−1</sup> ]	LHV [MJ kg <sub>f</sub> <sup>−1</sup> ]	El <sub>IN</sub> [10 <sup>15</sup> kg <sub>f</sub> <sup>−1</sup> ]	RF [mW m <sup>−2</sup> ]	RF <sub>day</sub> [mW m <sup>−2</sup> ]	RF <sub>night</sub> [mW m <sup>−2</sup> ]
Conventional Fuel (Baseline)	1.23	43.13	2.63	22 to 68	−29 to 34	53 to 108
Paraffinic Biofuel	1.37	44.08	0.66	23 to 75 (−4% to 18%)	−10 to 53 (56% to 550%)	44 to 96 (−31% to −6%)
Clean Burn	1.23	43.13	0.90	21 to 67 (−13% to 5%)	−12 to 45 (24% to 425%)	42 to 89 (−31% to −14%)

El<sub>H<sub>2</sub>O</sub> and LHV indicate the emission index of water vapor and the lower heating value of the two fuel types used, respectively. For all fuel cases, El<sub>H<sub>2</sub>O</sub> is computed from stoichiometry, assuming complete combustion.

To this end, we developed the Contrail Evolution and Radiation Model (CERM) to simulate the main dynamical and microphysical processes occurring throughout a contrail's lifetime to compute contrail RF.

CERM simulations of contrails and contrail cirrus are run for one year over the United States under a set of three scenarios with varying fuel types and emission indices of suitable ice nuclei (El<sub>IN</sub>). In the first (baseline) scenario, the US aviation fleet utilizes conventional jet fuel with El<sub>IN</sub> = 2.63 × 10<sup>15</sup> kg<sub>f</sub><sup>−1</sup>. This is consistent with recent estimates (Stettler *et al* 2013), assuming most of the ice nucleation at contrail formation occurs around aviation-emitted black carbon (soot) (Kärcher and Yu 2009). We present our results in terms of IN emissions to separate our analysis from uncertainty associated with which specific particles constitute IN. In the second scenario, paraffinic biofuels are assumed to be used by the entire US fleet (and all over-flights). This scenario uses paraffinic biofuels (e.g. biomass-derived Fischer-Tropsch (FT) or hydroprocessed esters and fatty acids (HEFA) fuels) with El<sub>IN</sub> = 0.66 × 10<sup>15</sup> kg<sub>f</sub><sup>−1</sup>, a reduction of 75% (Speth *et al* 2015) with respect to the baseline, and the (higher) stoichiometric water vapor emissions. This reduction is consistent with the results of Moore *et al* (2017) who found 26%–48% reductions in the particle number emissions index for a 50% HEFA blend at cruise conditions. (We note that results from our paraffinic biofuel scenario are also applicable to other chemically similar fuels, e.g. coal-derived FT alternative jet fuel.) In the third and final scenario, we assess the potential impact of cleaner burning engines using conventional jet fuel, with El<sub>IN</sub> = 0.90 × 10<sup>15</sup> kg<sub>f</sub><sup>−1</sup> (Wilkerson *et al* 2010), which is intended to demonstrate the effect of advances in combustor technology reducing soot emissions.

## 2. Methods

### 2.1. Model structure and input data

The Contrail Evolution and Radiation Model (CERM) developed for this study evaluates contrails and contrail cirrus characteristics and radiative impact over a

full year, using a 1 hour time resolution and a 13.5 km horizontal grid resolution covering the contiguous United States (approximately 57° W–140° W longitude and 16° N–58° N latitude), with 10 equally spaced vertical pressure levels ranging from 400 to 150 hPa. CERM uses aircraft tracking and fuel burn data for 2006 taken from the Aviation Environmental Design Tool (AEDT) (Wilkerson *et al* 2010). Meteorological fields are taken from the National Oceanic and Atmospheric Administration (NOAA) Rapid Refresh (RAP) dataset (National Oceanic and Atmospheric Administration Earth System Research Laboratory 2015). Data include temperature, three-dimensional wind speed fields and relative humidity with respect to liquid water. Contrail simulation results are highly dependent on the accuracy of the meteorological fields at flight altitudes. We find that the RAP dataset is locally ice-supersaturated approximately 12% of the time over an annual cycle at altitudes between 7 km and 13 km. This is lower than some other estimates in the literature with values closer to 15% (Gierens *et al* 2012, Irvine and Shine 2015). However, the RAP dataset is specifically for North America while other results are global.

### 2.2. Contrail formation and initialization

Meteorology and flight data are used to compute contrail formation using the Schmidt–Appleman Criterion (SAC) (Schumann 1996), which is formulated in CERM following the method described in Ponater *et al* (2002). Overall engine efficiency is set to 0.45, a value consistent with modern jet engine performance parameters (Cumpsty 2003), though this may be higher than the fleet average for 2006. Only persistent contrails are simulated in CERM; therefore the SAC is integrated with a persistence condition of ice-supersaturated background atmosphere, with ambient relative humidity with respect to ice computed from input meteorological data of temperature and relative humidity with respect to liquid water (Alduchov and Eskridge 1996, Murphy and Koop 2005). The initial number of ice crystals within each contrail is obtained from aircraft fuel burn data by applying the IN emission index corresponding to the specific fuel/emission scenario simulated (see table 1). Aircraft-emitted ice

nuclei responsible for contrail formation are treated in CERM as monodisperse with diameter of 40 nm (Petzold *et al* 2003), and bulk density of  $1000 \text{ kg m}^{-3}$  (Durdina *et al* 2014). These characteristics are typical of aircraft-emitted soot particles, which are assumed to be contrail formation precursors following several models available in the literature (Burkhardt and Kärcher 2011, Kärcher *et al* 1998, Schumann 2012).

### 2.3. Contrail vortex phase modeling

After contrail formation is simulated, CERM models processes occurring at the early stages of contrail lifetime. Wake vortex downwash determines adiabatic heating of contrails, and a consequent ice crystal sublimation (Holzäpfel and Gerz 1999), which is modeled through a parameterization by Schumann (Schumann 2012). The parameterization utilizes a constant vortex sinking distance  $\Delta z = 100$  meters, a value suggested both by empirical (Sussmann and Gierens 1999) and computational (Lewellen and Lewellen 2001) evidence. Contrail dilution and spreading in the vortex phase are modeled as described by Schumann (2012). CERM also accounts for entrainment of potential ice nuclei from the ambient atmosphere, a phenomenon consistent with cirrus ice residuals sampling campaigns (Cziczo and Froyd 2014, Twohy and Gandrud 1998). Given the high uncertainties in the type of particles serving as ice nuclei (Cziczo and Froyd 2014, Twohy and Gandrud 1998), no characterization is given about the nature of ice nuclei in the background atmosphere, and the concentrations are taken as uniformly distributed IN across the domain (well-mixed upper troposphere) (Hendricks *et al* 2004). Background ice nuclei in this study are assumed at the concentration of  $2/L$ , following results from measurement campaigns (DeMott *et al* 2010, DeMott *et al* 2011). Entrainment of ice nuclei from the background atmosphere starts in the vortex phase and occurs at all stages of contrail lifetime, and affects the total number of ice crystals within aging contrails, the processes of ice crystal growth and of ambient water vapor uptake.

### 2.4. Dynamics, turbulent diffusion and wind shear

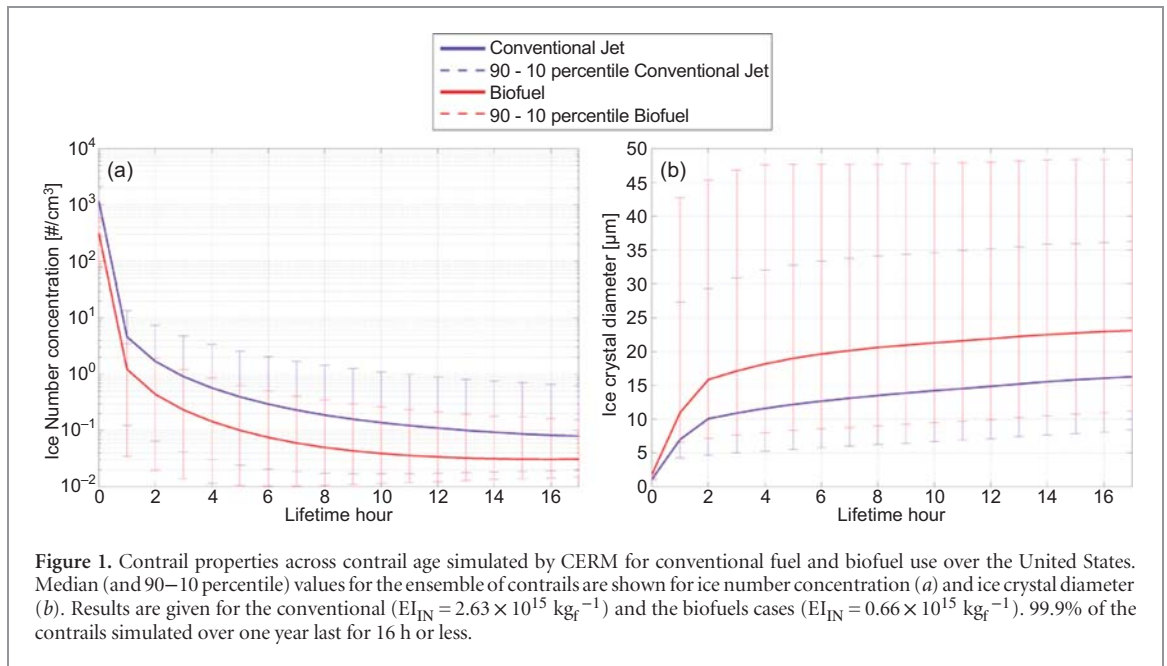
CERM simulates the advection and gravitational settling of contrails through a Lagrangian dynamics module. Contrail dynamical processes are assumed to occur simultaneously and uniformly across all the ice crystals (assumed spherical) (Li *et al* 2013), and are driven by the wind velocity fields throughout the domain (Schumann 2012) and by the gravitational fall speed of the ice crystals within the contrails (Seinfeld and Pandis 2016). Throughout their lifetime, contrails are modeled as Gaussian plumes with elliptical cross-sections by a parameterization of dilution, turbulent diffusion and wind shear (Schumann 2012). Atmospheric turbulence levels are assumed following Schumann (2012). At the hourly time resolution of the model, contrails evolve in the atmosphere and persist as long as they remain in ice-supersaturated regions.

### 2.5. Ice crystal growth

While they are advected and spread in the atmosphere, contrails entrain background air and incorporate new available ice nuclei (DeMott *et al* 2010, DeMott *et al* 2011). These fresh IN generate new ice crystals within the contrails, and induce further entrainment of ambient water offering a total larger surface for condensation of ice-supersaturated water vapor. Throughout their lifetime, ice crystals are simulated to grow in size by depositional uptake of background water vapor. This is modeled from Fick's first and second laws, which regulate the diffusion of aerosol particles and predict a larger water uptake for a larger atmospheric supersaturation with respect to ice (Pruppacher and Klett 2010). The number of ice crystals within contrails changes as contrails age due to entrainment of ambient ice nuclei and to aggregation related to the mixing of contrail with ambient air and plume-internal turbulence (Schumann 2012).

### 2.6. Optical depth and radiative forcing

Optical depth at 550 nm at every stage of contrail lifetime is computed utilizing an approach developed by Schumann (2012), using Mie theory and taking the refractive index for ice to be 1.31. At each time step, ice crystal size, ice crystal number concentration, horizontal area cover, depth, ice water content and optical depth are computed for each of the simulated contrails. A parameterized radiative forcing model (Schumann *et al* 2012) is used to compute the radiative forcing from the modeled contrails and contrail cirrus over the United States of America. Planetary albedo, radiative fluxes and background cloud cover data necessary for the RF computations are obtained from the NASA CERES satellite inventory (NASA Langley Research Center Atmospheric Science Data Center 2015). The solar zenith angle is computed at each location according to the local time of day, as described in the supplementary material available at [stacks.iop.org/ERL/12/114013/mmedia](http://stacks.iop.org/ERL/12/114013/mmedia). Shortwave and longwave radiative forcing are therefore calculated with five ice crystal habit assumptions considered (spherical, solid hexagonal columns, hollow hexagonal columns, plates and droxtals) as modeled by Schumann *et al* (2012) that correspond to the range of RF values presented in this paper. Comparisons between cases are made pairwise assuming the same ice crystal habit in each case. The percentage changes in RF presented in table 1 follow this paired comparison approach, and further details broken down by ice crystal habit are shown in the supplementary material. For the purpose of presenting results, the daytime and nighttime RF components are separated by computing local sunrise and sunset throughout the computational domain (Kalogirou 2014) and average values are computed over these time periods. A detailed description of CERM's structure and dynamics, microphysics, and radiation modules, together with comparisons of the results with existing literature is available in the supplementary material.



### 3. Results

Considering one-year contrail simulations in the conventional and paraffinic biofuel cases, we find that utilizing biofuels results in 8% more contrails over the United States. This is due to the 11% higher water emissions index of biofuels with respect to conventional jet fuel (see table 1). Higher water vapor emissions increase humidity within plumes, resulting in higher threshold temperatures below which contrails form after the passage of an aircraft (Schumann 1996). This enhances the likelihood of contrail occurrence. Contrails forming in the biofuel case are characterized by ice crystal number concentrations on average 75% lower than their counterparts in the conventional fuel case (see figure 1(a)). This reduction is due to the lower number of IN available for the formation of crystals in the early stages of contrail lifetime. The lower number of ice crystals reduces the competition for uptake of ambient water vapor above ice-saturation throughout contrail lifetime, yielding larger crystals ( $\sim +58\%$  in diameter with respect to the conventional fuel case; see figure 1(b)).

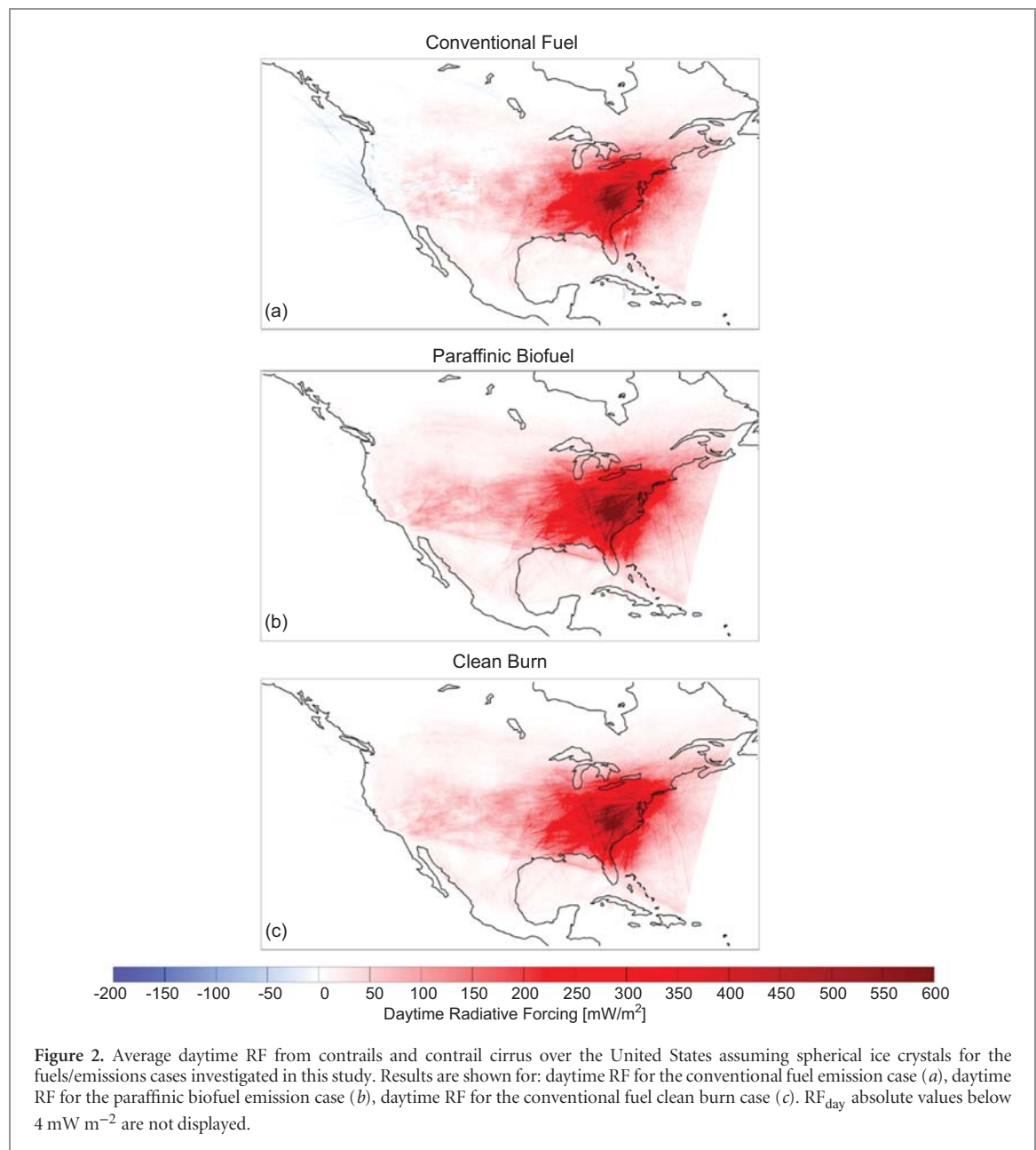
Changes in crystal concentration and size affect contrail optical properties (Schumann *et al* 2012), resulting in a lower average contrail optical depth (within regions where contrails are present) at 550 nm for contrails formed by biofuels, decreasing to  $\sim 0.020$  from the average  $\sim 0.028$  computed in the conventional fuel case ( $-29\%$ ). A reduction in contrail optical depth is expected to decrease the warming longwave RF to a larger extent than the cooling shortwave RF, thus generally yielding a lower net radiative forcing (Schumann *et al* 2012). Nevertheless, the cooling effect due to the reduction in optical depth for contrails formed in the biofuel case is counterbalanced by a lower contrail albedo, i.e. the extent to which these

clouds scatter incoming solar radiation back to space. The annually-averaged planetary albedo change induced by contrails decreases from  $1.18 \times 10^{-4}$  in the conventional fuel case to  $8.03 \times 10^{-5}$  in the biofuel case ( $-32\%$ ). This albedo reduction is due to the larger ice crystals and lower ice number concentrations, i.e. a (reverse) Twomey effect (Twomey 1974). The Twomey effect is an increase in albedo for clouds ‘polluted’ by anthropogenic emissions, and thus composed of droplets in larger number and smaller sizes. The effect also occurs in contrails, since the solar albedo of ice crystals increases with crystal size reduction faster than the infrared emittance (Zhang *et al* 1999). Overall, the increase in contrail occurrence and the lower albedo of contrails forming in the biofuel case outweigh the cooling effect of optical depth reduction, caused by its prevailing effect on longwave versus shortwave radiation. This leads to a higher average net radiative forcing of between 0% and  $+18\%$  for all cases except plate crystals, which show a 4% reduction in net RF. This is contrary to expectations of a substantial RF benefit associated with the decrease in optical depth (e.g. Kärcher (2016)).

The contrast between the cooling effect brought by contrail optical depth reduction and the warming effect due to contrail albedo reduction is shown in figures 2(a) and (b) and figures 3(a) and (b) assuming spherical ice crystals, displaying daytime ( $RF_{\text{day}}$ ) and nighttime ( $RF_{\text{night}}$ ) components of radiative forcing for the conventional and biofuel cases. Compared to conventional fuel, use of paraffinic biofuels increases daytime radiative forcing between 56% and 550% (figure 2(b)) and decreases nighttime RF between 6% and 31% (figure 3(b)).

Comparing the clean burn case ( $EI_{IN} = 0.90 \times 10^{15} \text{ kg}_f^{-1}$ ) with the baseline emission case ( $EI_{IN} = 2.63 \times 10^{15} \text{ kg}_f^{-1}$ ), the reduction of suitable IN

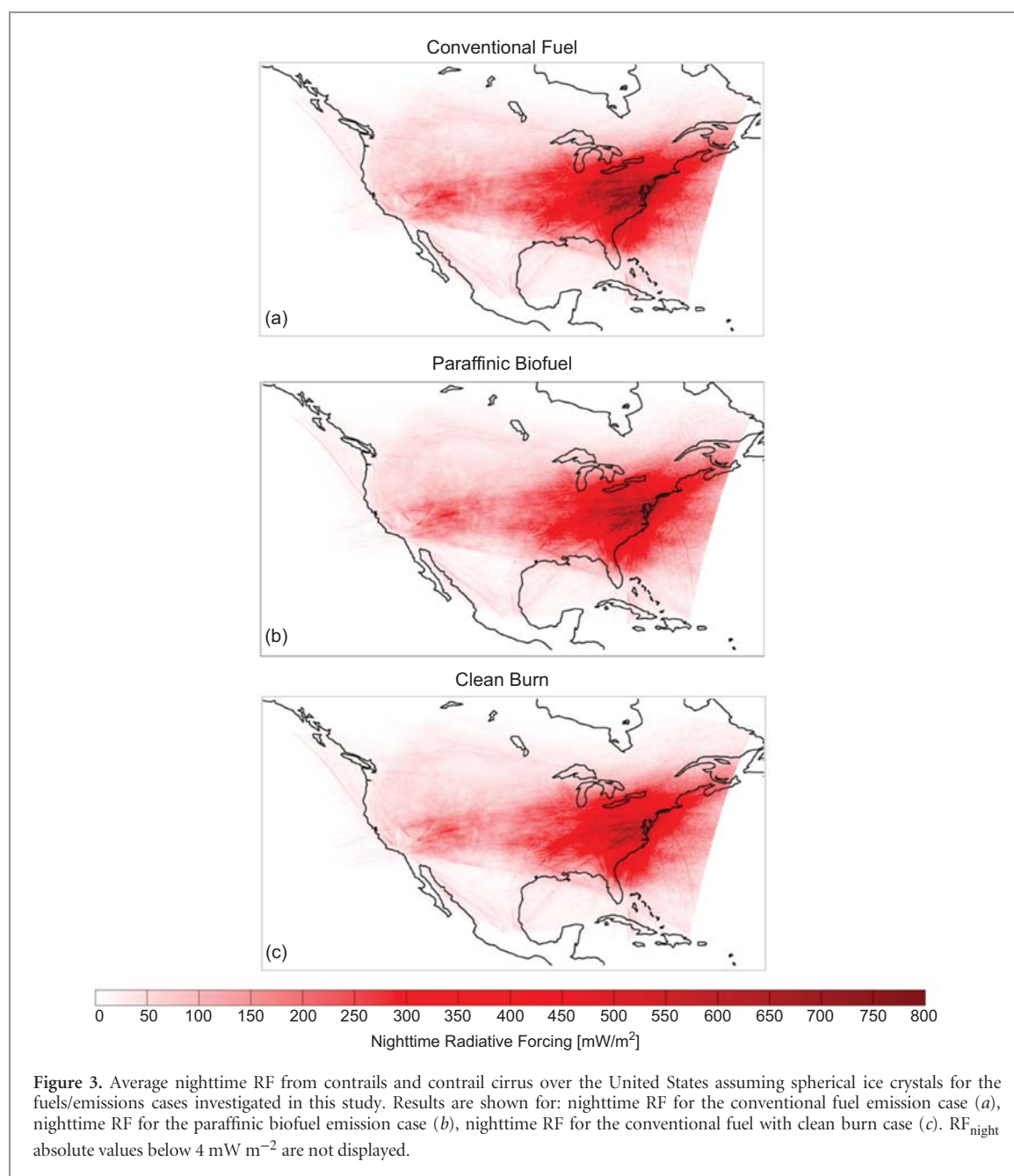




emissions results in changes in contrail properties similar to the ones produced by alternative fuels. On average, contrails in the clean burn case are characterized by larger crystals (+43%) at smaller number concentration (−67%) with respect to the baseline case. The resulting contrail optical depth is therefore decreased (−27%), but this cooling effect is again counterbalanced by the lower planetary albedo change induced by contrails (−33%). The effect on net radiative forcing (figure 4(c)) is less clear than the biofuel case, ranging between 13% lower and 5% higher than the conventional fuel case. This result shows that, since the decreased albedo (reverse Twomey effect) offsets most of the benefits of reducing long-wave RF, even substantial reductions in IN emissions offer, at best, limited potential for reducing contrail warming.

#### 4. Discussion and conclusion

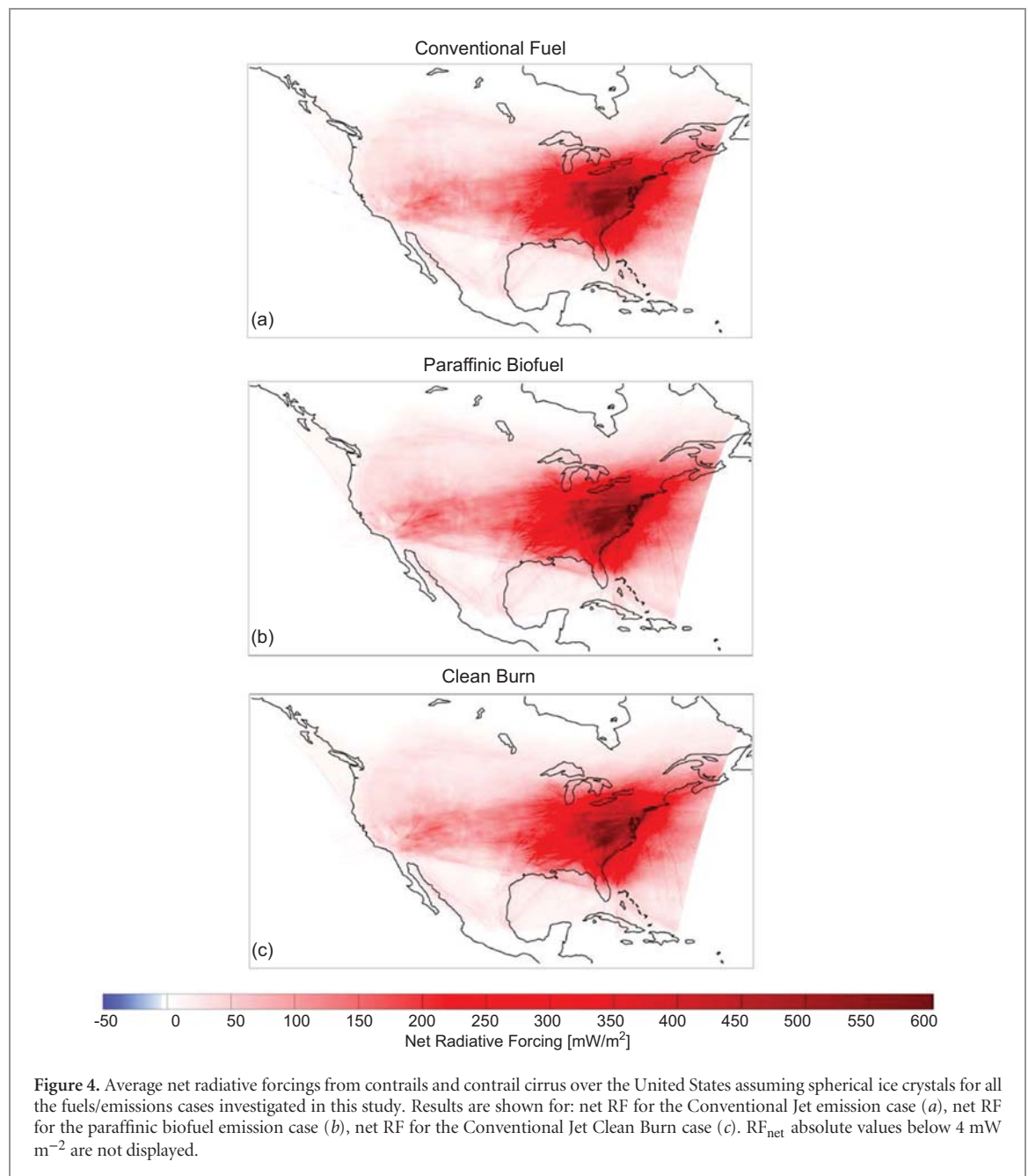
The Contrail Evolution and Radiation Model (CERM) has been used to evaluate the effects of changes in aircraft fuels and emissions on contrail warming using scenarios which consider reductions in ice nuclei emissions either from the use of paraffinic biofuels (or other paraffinic alternative fuels) or through improvements in combustor technology which decrease soot emissions. In the case of biofuels, contrails are found to form more frequently due to the higher water emissions index of paraffinic fuels, and this leads to a change in net RF of −4 to +18% compared to conventional fuels (figures 4(a) and 4(b)). This effect is composed of an increase in daytime RF (+10 to +22  $\text{mW m}^{-2}$ ) and a decrease in nighttime RF (−6 to −21  $\text{mW m}^{-2}$ ), so by selectively using biofuels



at night, a reduction in contrail RF could be achieved. In contrast, for cleaner burning engines, which would operate at all times of day, the increase in daytime RF ( $+8$  to  $+17 \text{ mW m}^{-2}$ ) and a decrease in nighttime RF ( $-11$  to  $-21 \text{ mW m}^{-2}$ ) nearly cancel out, leaving a net change in contrail RF of  $-5$  to  $+4 \text{ mW m}^{-2}$ . This suggests that advances in combustor technology which reduce soot emissions may not contribute to a significant reduction in contrail RF.

Modeling contrails requires capturing several complex phenomena from plume-scale growth to the microphysical aspects of contrails and their interaction with light. Assumptions are made and parameter values used throughout CERM that can alter the results of individual simulations, so there is uncertainty in the results presented in this paper. A major

source of uncertainty is the choice of ice crystal habit (Schumann *et al* 2012) and the range of RF values presented in this paper portray the effect of varying this assumption. Paired comparisons among simulations using the same ice crystal habit show that paraffinic (such as HEFA and FT) biofuels have a net warming effect in all cases except for plate crystals, while the effect of a clean burning engine is more varied. The sensitivity of the results shown in this paper to assumptions in the microphysical modeling suggest that further development of these models is warranted in order to understand and reduce the uncertainties associated with contrail modeling. Such model improvements are especially important at a time when the characteristics of aircraft emissions are changing through the adoption of biofuels and advancements in engine technology.



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