

**Please cite the Published Version**

De Leon, Rubén Rodríguez and Lee, David S (2023) Contrail radiative dependence on ice particle number concentration. *Environmental Research: Climate*, 2 (3). 035012-035012. ISSN 2752-5295

**DOI:** <https://doi.org/10.1088/2752-5295/ace6c6>

**Publisher:** IOP Publishing

**Version:** Published Version

**Downloaded from:** <https://e-space.mmu.ac.uk/632334/>

**Usage rights:**  [Creative Commons: Attribution 4.0](https://creativecommons.org/licenses/by/4.0/)

**Additional Information:** This is an Open Access article published in *Environmental Research: Climate*, by IOP Publishing.

**Data Access Statement:** The data cannot be made publicly available upon publication because they contain commercially sensitive information. The data that support the findings of this study are available upon reasonable request from the authors.

**Enquiries:**

If you have questions about this document, contact [openresearch@mmu.ac.uk](mailto:openresearch@mmu.ac.uk). Please include the URL of the record in e-space. If you believe that your, or a third party's rights have been compromised through this document please see our Take Down policy (available from <https://www.mmu.ac.uk/library/using-the-library/policies-and-guidelines>)

PAPER • OPEN ACCESS

## Contrail radiative dependence on ice particle number concentration

To cite this article: Rubén Rodríguez De León and David S Lee 2023 *Environ. Res.: Climate* **2** 035012

View the [article online](#) for updates and enhancements.

### You may also like

- [On the Solved Turbulent Scales in Turbulent Plume Fires](#)  
F.S. Ciani, P. Bonfiglio and S. Piva
- [Long-term fuel retention in JET ITER-like wall](#)  
K Heinola, A Widdowson, J Likonen et al.
- [Divergent trends in irrigation-water withdrawal and consumption over mainland China](#)  
Ling Zhang, Donghai Zheng, Kun Zhang et al.

# ENVIRONMENTAL RESEARCH CLIMATE



## PAPER

# Contrail radiative dependence on ice particle number concentration

### OPEN ACCESS

RECEIVED  
3 February 2023

REVISED  
10 July 2023

ACCEPTED FOR PUBLICATION  
12 July 2023

PUBLISHED  
1 August 2023

Rubén Rodríguez De León\*  and David S Lee 

Department of Natural Sciences, Manchester Metropolitan University, Manchester M1 5GD, United Kingdom

\* Author to whom any correspondence should be addressed.

E-mail: [r.deleon@mmu.ac.uk](mailto:r.deleon@mmu.ac.uk)

**Keywords:** radiative forcing, ice particle size distribution, contrails, ice water content, particle effective size, ice crystal number

Original content from this work may be used under the terms of the [Creative Commons Attribution 4.0 licence](https://creativecommons.org/licenses/by/4.0/).

Any further distribution of this work must maintain attribution to the author(s) and the title of the work, journal citation and DOI.



## Abstract

Recent studies on low aromatic fuels have shown that lower soot number emissions may reduce contrail ice particle number concentrations ( $N_{ice}$ ). Here we implemented, in a sophisticated radiative transfer model, two ice particle size distribution schemes in order to estimate the contrail radiative forcing's (RFs) dependence on these prospective  $N_{ice}$  reductions resulting from the introduction of sustainable aviation fuels. The results show that an 85% contrail  $N_{ice}$  reduction produces a 35% smaller contrail RF, while neglecting all non-radiative effects. This estimate of an RF reduction only considers the effects of the  $N_{ice}$  change assumed here, and neglects other potentially important microphysical mechanisms that may change the relationship between soot number emissions and  $N_{ice}$ . A comparison of our results with previous published estimates from full climate model simulations, shows similar RF reductions to those which also take into account non-radiative mechanisms, evidencing the need for more studies in order to allocate the contribution from radiative and non-radiative changes, as this would guide possible mitigation implementations. Despite these modeled contrail RF reductions being largely independent of the assumed ice water content (IWC), it is only through simultaneous improvement of the IWC and  $N_{ice}$  representation in models that contrail RF estimates can be better constrained. This is because our calculated RF varied by a factor of 3 when assuming a  $\pm 30\%$  IWC range; and by a factor of 5 if a, still conservative,  $\pm 60\%$  IWC range was prescribed, suggesting that the differences in the prescribed IWC and  $N_{ice}$  values in different models may explain the large discrepancies amongst published RF estimates. Recent estimates of higher  $N_{ice}$  values, and lower IWCs found in contrails even after several hours, compared to surrounding cirrus under similar atmospheric conditions, were assessed to conclude that it is mainly the differences in IWC that make young contrails have a smaller RF, and to reduce our previous estimate for linear contrail RF for year 2006 by 65%.

## 1. Introduction

The ice particle number concentration ( $N_{ice}$ ) of contrails depends upon the initial soot aerosol number concentration emissions [1, 2] for kerosene-based fuel. Given that, in turn, contrail radiative forcing (RF) is dependent on  $N_{ice}$ , understanding the dependency of contrails' RF on  $N_{ice}$  becomes relevant in the ongoing discussion around the usage of lower-carbon footprint fuels (often referred to as 'Sustainable Aviation Fuels', or 'SAF'). SAF have low to zero aromatic content, and many measurements at ground and at altitude show that the soot number emissions depend upon this (Moore *et al.* [3] and references therein). Understanding how SAF might impact the optical properties and otherwise of contrails [4, 5] is of critical importance to better-quantifying the potential climate co-benefits of low-carbon footprint fuels in reducing contrails' RF.

Burkhardt *et al* [6] estimated, from global climate model (GCM) simulations, that an 80% reduction in  $N_{ice}$  resulted in a 50% reduction in contrail cirrus RF, and a 50% reduction would lead to a significant decrease in contrail cirrus optical depth and coverage, leading to a decrease in RF by approximately 20%. Similarly, Bock and Burkhardt [7] found a 14% RF reduction from an  $N_{ice}$  reduction of 50%. Both studies

account for changes in contrails coverage, lifetime and optical depth, while the aim of the present study is to estimate the sensitivity of the radiative-only properties of contrails with respect to differences in  $N_{ice}$  in order to isolate their dependence on the prospective introduction of SAF, assuming a relationship between soot number and  $N_{ice}$ .

Contrails are ice clouds formed by aircraft under sufficiently cold and humid atmospheric conditions [8]. They may persist for several hours in ice supersaturated regions, eventually losing their linear shape, and becoming contrail cirrus as they increase their horizontal and vertical extent [9]. The confidence in the understanding of the RF of linear contrails remains constrained by the difficulties in the measurement, both *in situ* and from remote sensing, of their complex microphysical and optical properties, as well as by the complexity involved in parameterizing their broad variability [10] in large-scale models.

The present study is not only aimed at assessing the differences in the radiative properties of contrails linked to the introduction of SAF, but also at estimating the radiative differences between natural and contrail cirrus, as well as the sensitivity of their calculated RF to differences in the  $N_{ice}$  and ice water content (IWC) amongst different models.

Most ice radiative contrail and cirrus parameterizations make use of two variables, commonly an effective size, like the equivalent diameter ( $D_e$ ), and IWC.  $D_e$  is defined as the ratio of the surface area to the volume of the ice particle size distribution (PSD), while the IWC is simply the mass concentration for a given volume. The fact that the smallest ice crystals largely determine  $N_{ice}$  without significantly influencing  $D_e$ , nor IWC, implies that with this approach two PSDs may show similar  $D_e$  and IWC values despite large  $N_{ice}$  differences, which may significantly influence the calculated radiative properties. This has already been shown [11–13] for an atmospheric single column, as well as for ice single scattering properties, but not for global coverage RF calculations, prompting the need for the present study. Equally important is the use of a sophisticated efficient radiative transfer model that allows a realistic representation of the particle size dependence on IWC and temperature. This has not always been the case in previous contrail sensitivity studies (e.g. Markowicz and Witek [14]) in which the contrails' optical depth, or altitude, are prescribed using fixed values. This approach neglects the dependence that contrail IWC and  $D_e$  have on temperature and therefore on altitude. Furthermore, these simplifications neglect the fact that the results may be different if a different fixed value is assumed.

Given the lack of climatological contrail data, some linear contrail studies have prescribed natural cirrus properties (e.g. Ponater *et al* [15] and Rodríguez De León *et al* [16]), concluding that, like in the case of natural cirrus, the calculated RF of persistent contrails is mainly driven by their IWC, while being marginally dependent upon particle size, but evidence has grown about contrails showing higher  $N_{ice}$  than natural cirrus, as reported in several studies [17–20], suggesting the need to reassess this conclusion.

Despite the currently limited information about contrail  $N_{ice}$  and IWC properties and their temporal evolution, young contrails have been shown to have higher  $N_{ice}$  values than natural cirrus by an order of magnitude [19, 20], with enhanced  $N_{ice}$  values being found even after several hours compared to surrounding cirrus under similar atmospheric conditions [18]. A more recent study [21] found more modest  $N_{ice}$  differences of around 50% between natural and contrail cirrus. Based on these findings here we also here estimate the calculated contrail net RF sensitivity to  $N_{ice}$  and IWC differences.

## 2. Model setup

The model setup is described in the supplementary information section, and is similar to that of Rodríguez De León *et al* [16] (RL18, hereafter), from which the reference case is here included for comparison purposes. The calculated linear contrail cover is prescribed as monthly mean 6-hourly vertical profiles, based on the REACT4C [22] air traffic inventory for year 2006. Given that the vertical layers are variable and predetermined in the model, their IWC was reduced in order to approximate a thickness of approximately 500 m. Also, given that only one ice cloud type can be prescribed in the model, we run the simulations under clear sky conditions.

In order to assess the sensitivity to  $N_{ice}$ , two radiative parameterizations were here used to estimate the global linear contrail instantaneous RF. These parameterizations are based on two PSD schemes with different  $N_{ice}$  values. The PSDs are defined as functions of ambient temperature and IWC.

The first scheme was developed by Field *et al* [23] (F07 hereafter) and the second one, which was previously implemented in RL18, was developed by McFarquhar and Heymsfield [24] (MH97 hereafter). The MH97 scheme is based on PSD measurements of deep convection cirrus from the Central Equatorial Pacific Experiment (CEPEX), while the F07 parameterization is based on mid-latitude ice clouds. The MH97's PSD parameterization provides  $N_{ice}$  and its partition into the small and large ice particle PSD modes for any specified IWC, which is here prescribed, together with its dependence on temperature, from the IWC climatology developed by Schiller *et al* [25] (S08, hereafter). The contrail radiative properties are then

calculated by integrating their single scattering properties over the MH97 PSDs in order to produce a parameterization in terms of the PSD's IWC and  $D_e$ . For the F07 scheme, the radiative parameterization described in Baran *et al* [26] was applied, which is based on an ensemble of ice crystal shapes, described in Vidot *et al* [27]. This radiative parameterization does not include the dependence on the effective ice particle size, because the particle shape ensemble implicitly incorporates the size dependence from the combined IWC and ambient temperature values.

It was shown in RL18 that  $N_{ice}$  values from the MH97 scheme were slightly larger than the climatological values reported in Krämer *et al* [28]. In contrast, the F07 scheme values are smaller than those from the MH97 parameterization by around 85% at representative persistent contrail temperatures, and for this reason these schemes provide a wide range of modeled  $N_{ice}$  values to analyze their impact on the calculated RFs.

For the MH97 PSDs, we assumed a hexagonal cylinder particle habit with a variable aspect ratio, which reproduced the observed mass and area contributions from small and large particles reported by MH97. The  $D_e$  values prescribed in the simulations are also obtained from the MH97 scheme, ensuring that a realistic dependence of  $D_e$  and of the IWC on the local ambient temperature was always assumed in the sensitivity cases. As mentioned in the introduction section, the accuracy of the results from sensitivity studies relies on prescribing physically realistic values, which take into account their interdependencies.

The IWC was prescribed as a function of local ambient temperature in all the cases, but in order to assess the RF dependence on the IWC, we scaled it by 30% and 60% increments/decrements with respect to the mean IWC compiled by S08 for middle latitudes, assumed in the base case. Prescribing  $\pm 60\%$  IWC differences results in an IWC range covering a factor of 4. This can be contrasted to the ice water path differences between GCMs used in the IPCC's 4th Assessment Report, which showed a factor of 20 difference between the largest and smallest values, and a factor of 6 after removing outliers [29], while reporting regional differences of nearly 2 orders of magnitude. Given that the ice water path would also include an uncertainty linked to the physical thickness of the cloud layer, it seems reasonable to assume that the  $\pm 60\%$  IWC uncertainty implemented here is similar to that between GCMs. Also, contrail cirrus are reported to have a 60% smaller IWC compared to cirrus under the same atmospheric conditions [21].

### 3. Results

The RF results, shown in table 1, comprise longwave (LW), shortwave (SW) and net (LW + SW) annual mean global instantaneous contrail RF values, as well as their percentage differences with respect to the MH97's base case. The cases combine scaled IWC values with the two described PSD schemes, amongst which  $N_{ice}$  values differ by around 85% at representative persistent contrail temperatures, emulating plausible differences between contrails and natural cirrus [30], as well as possible soot reductions linked to a prospective introduction of SAF.

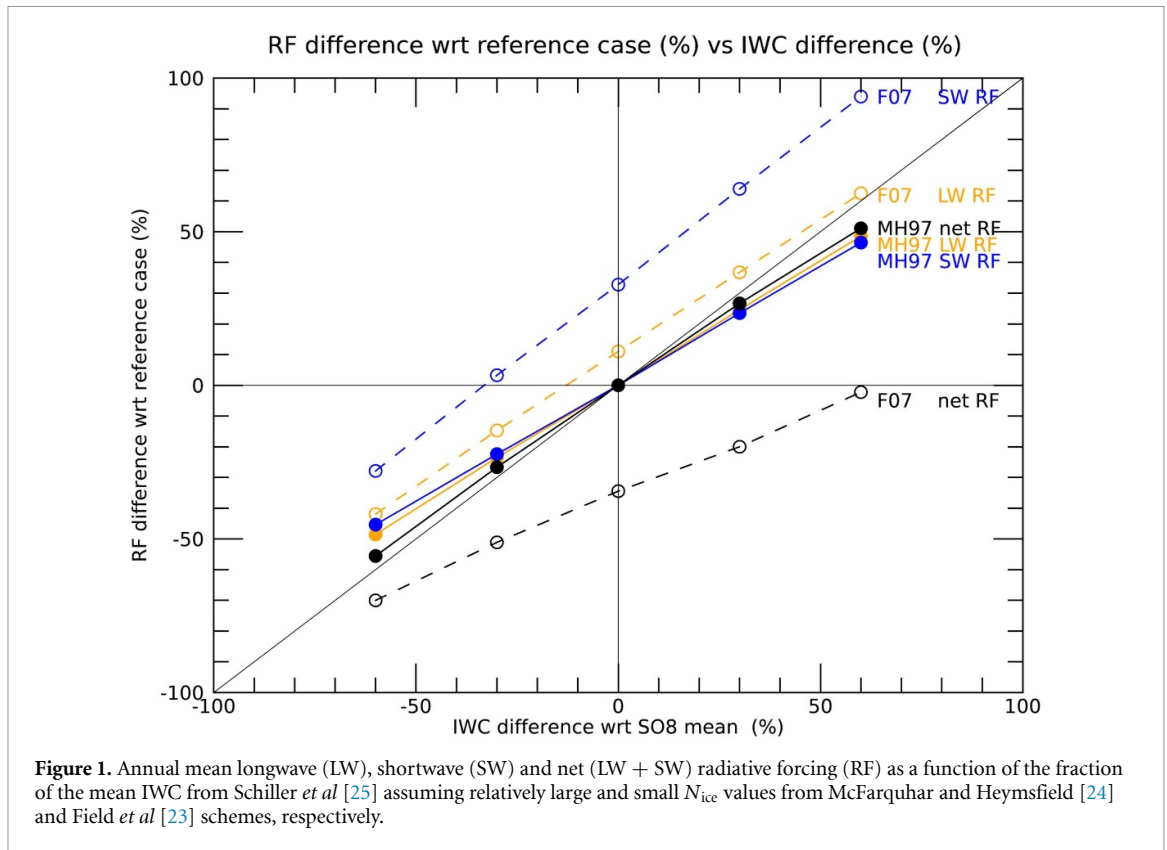
Figure 1 shows the percentage differences with respect to the MH97 base case, from which it can be seen that the  $N_{ice}$  differences resulted in the F07 net RFs being around 35% smaller than those calculated with the MH97 scheme. This reduction resulted from F07 SW RF magnitudes being around 33% larger than those from MH97, partially counterbalanced by a comparatively modest strengthening of around 11% in the LW. As a result, all F07 net RFs showed smaller values than the MH97 base case, even for case 5 which had a 60% larger IWC prescribed value.

The combined  $N_{ice}$  and IWC sensitivity in table 1 covers a range of net RF values that spans a factor of 3 when prescribing IWC differences of  $\pm 30\%$ , and a factor of 5 when IWC is varied by  $\pm 60\%$ , compared to a more modest factor of 3 for the  $\pm 60\%$  IWC scaling if only one PSD scheme is prescribed. This shows that despite the fact that the results are more sensitive to the prescribed IWC differences compared to those from  $N_{ice}$  when taken separately, their contribution to the combined RF sensitivity is not largely dissimilar, and therefore it is only by simultaneously improving their model representation and validation that the uncertainty of linear contrail and contrail cirrus RF estimates can be constrained.

### 4. Discussion

The impact that differences in  $N_{ice}$  play on the radiative properties of contrails acquires particular relevance when considering theoretical calculations that have shown a dependence of  $N_{ice}$  on soot number emissions [2], as well as limited *in situ* measurements [31] reporting soot and  $N_{ice}$  reductions between 50 and 70%. This discussion becomes especially pertinent to current efforts to produce low-carbon footprint fuels (bio, synthetic), since they will have low to zero aromatic content, and much lower soot emissions [3].

Contrasting the 50% reduction in contrail cirrus RF reported by Burkhardt *et al* [6] from an 80%  $N_{ice}$  reduction, which took into account changes both in contrail cirrus optical depth and coverage, with the 35%



**Table 1.** Instantaneous radiative forcing (RF) [ $mW m^{-2}$ ] for prescribed IWC and  $N_{ice}$  cases and their percentage differences with respect to the MH97 base case. Longwave (LW), shortwave (SW) radiative forcing (RF), net (LW + SW) and ratio ( $-SW/LW$ ). The mean IWC from Schiller *et al* is scaled and the radiative calculations are based on PSD schemes from McFarquhar and Heymsfield [24] (MH97) and Field *et al* [23] (F07), which have relatively larger and smaller  $N_{ice}$  values, respectively.

	IWC	0.4*S08	0.7*S08	S08	1.3*S08	1.6*S08
RF	cases:	1	2	3	4	5
Net	MH97	4.0	6.6	9.0 base case	11.4	13.6
		-56%	-27%	0%	26%	52%
Net	F07	2.7	4.4	5.9	7.2	8.8
		-70%	-52%	-34%	-20%	-2%
LW	MH97	14.0	20.8	27.2	33.9	40.4
		-49%	-24%	0%	25%	49%
LW	F07	15.8	23.2	30.2	37.2	44.2
		-42%	-15%	11%	37%	63%
SW	MH97	-10.0	-14.2	-18.3	-22.6	-26.8
		-46%	-22%	0%	24%	47%
SW	F07	-13.2	-18.9	-24.3	-30.0	-35.5
		-28%	3%	33%	64%	94%
-SW/LW	MH97	0.71	0.68	0.67	0.67	0.66
	F07	0.83	0.81	0.80	0.81	0.80

RF reduction found in the present study for a similar reduction (85%) in  $N_{ice}$ , we conclude that, despite the fact that our model only considers the change in radiative properties linked to  $N_{ice}$ , both results show an encouraging similarity. It becomes clear that, given the relevance of the topic, more studies from different models and approaches will be needed in order to understand the implications of introducing SAF.

It should be stressed here that although a reduction in RF is found with lower  $N_{ice}$ , that in the context of low-aromatic SAF such a response is not assumed. Theoretical modeling has shown that at low soot number

emissions, enhanced activation of ultrafine aqueous particles may occur at very cold temperatures, resulting in an increase in  $N_{ice}$  for low soot numbers, past a critical threshold of  $10^{14}$  soot particles  $kg^{-1}$  fuel.

The effect of  $N_{ice}$  differences had already been explored by Zhang *et al* [11] for single column simulations for spherical ice crystals hexagonal columns and polycrystals, but not for global simulations nor for ice particle shape ensembles. The results presented here showed that the effect of an 85%  $N_{ice}$  reduction is a reduction of around 35% in the calculated net RF, irrespective of the assumed IWC.

IWC differences between contrails and natural cirrus have been also measured. Li *et al* [21] reported that contrail cirrus present smaller IWCs by around 60%, while Chauvigné *et al* [19] reported smaller contrail and contrail cirrus IWC values in all stages of contrails compared to natural cirrus, and larger  $N_{ice}$  for young contrails suggesting the MH97 scheme better represents linear contrail  $N_{ice}$  values. Based on these results, we reduced our previous estimate from  $9 \text{ mW m}^{-2}$ , reported in RL18 (represented here as case 3 for MH97), to  $4 \text{ mW m}^{-2}$ , based on case 1 of the MH97 scheme, which assumes an IWC reduction of 60% as reported by Li *et al* [21]. This significant reduction evidences the importance that contrail measurement campaigns have on reducing global RF uncertainties by improving combined IWC and  $N_{ice}$  climatological data compared to surrounding natural cirrus.

Important discrepancies amongst published studies on the linear contrail net RF's dependence on the prescribed particle size include RL18 reporting a RF sensitivity of  $-9\%$  when implementing the MH97 parameterization used here, while Yi *et al* [32] reported differences of  $+46\%$ . In both studies the differences emulated a 50% increment in the effective particle size, suggesting that, as pointed out by Zhang *et al* [11] and Rodríguez De León and Haigh [12], the commonly adopted approach of parameterizing the radiative properties of ice crystals using their IWC and an effective size produces RFs which are not univocally estimated unless some information about the PSD's shape is also included.

Here we suggest that information about the  $N_{ice}$  values of the PSDs used to parameterize the radiative properties of persistent contrails can provide useful information to explain differences amongst RF estimates. Similarly, the sensitivity to particle size obtained by Yi *et al* [32] would also be affected by the fact that in their simulations they assumed fixed contrail altitude and optical depth values. As a result of this, the RF differences would be biased in one direction compared to a setup in which ambient conditions would more realistically determine contrail properties. It is therefore suggested, that despite the advantages of implementing simplified setup cases in sensitivity and inter-comparison studies, these simplifications often produce results not only linked to the assessed variable.

The radiative differences from the PSD schemes used here are also related to the fact that they assume two different particle shapes, hexagonal cylinders and an ice crystal ensemble. Similarly to the present study, Yi *et al* [32] also compared schemes based on a hexagonal cylinder habit and on an ice habit mixture. They obtained a 11% larger net RF from hexagonal cylinders, which they attributed to the fact that roughened ice particles were assumed in their multi-habit mixture, which would present smaller SW asymmetry factors compared to smooth hexagonal cylinders, and therefore would reflect more SW radiation.

This, in turn, would result in a reduction of the net RF for the multiple habit mixture. The discrepancy between the 11% difference obtained by Yi *et al* [32] and the 35% obtained in the present study may be explained by the use of fixed optical properties and contrail altitude assumed by Yi *et al* [32], which would exclude the  $N_{ice}$  variability expected at different temperatures if the altitude and the optical depth were allowed to vary. Again, these results suggest that simplified setups may render their sensitivity estimates unrealistic when fixing properties.

The results in table 1 show  $-SW/LW$  values of consistently around 67%, and 83% for the MH97 and the F07 scheme, respectively, showing that, with all other variables being fixed, this ratio shows little variability for each PSD scheme. On the other hand, this ratio is expected to be linked to the contrails' altitude, as De León *et al* [33] reported  $-SW/LW$  percentages between 64% to 81% when fixing contrail altitudes between 30 000 and 38 000 feet, while allowing IWC and the effective size to vary. Schumann and Graf [34] compiled  $-SW/LW$  values from various published studies, and reported a similar range size, from 32% to 52%, not only linked to  $N_{ice}$  differences, but in that case comprising differences in the contrail fields, parameterizations, radiative transfer schemes, and the background meteorology differences amongst models.

The results in table 1 and figure 1 show that the F07 scheme, which comprises smaller  $N_{ice}$  values by 85% compared to the MH97 scheme, produces net RF values that are always smaller than the reference case; even cancelling out the effect of an IWC 60% increase. In contrast, by excluding the IWC variability, the net RF differences linked to the  $N_{ice}$  differences produce a net RF reduction of around 35%.

A model intercomparison study by Myhre *et al* [35], reported contrail RF differences amongst models within a factor of 2 when prescribing a homogeneous 1% global contrail cover at a fixed altitude. They also reported global net RF values between  $9.3$  and  $15 \text{ mW m}^{-2}$  when prescribing a realistic 3D linear contrail cover. Their contrail setup consisted of a fixed PSD with fixed optical properties, which is equivalent to fixing

$N_{ice}$ , meaning that their setup excluded the  $N_{ice}$  sensitivity analyzed here, instead quantifying RF differences linked to the radiative schemes and background meteorologies amongst the compared models. Their 3D results implied a relatively modest (22%) standard uncertainty compared to 60% differences from the combined  $N_{ice}$  and IWC obtained in the present study if we assume a 30% IWC uncertainty, and 80% differences if a 60% IWC uncertainty is assumed, suggesting that the discrepancies between published contrail RF estimates are more likely explained by the uncertainties in the ice properties fed into the radiative schemes than by other differences in the radiative treatment in large scale models, including the background meteorology.

## 5. Conclusions

The relevance of the dependence of the contrails' RF on their assumed  $N_{ice}$  is manifold. The results obtained here showed that smaller  $N_{ice}$  values produce a SW RF enhancement that is 3 times that of the LW, resulting in a NET RF reduction. Given that our calculations only cover the radiative aspects of an  $N_{ice}$  reduction, our results of a contrail net RF reduction potentially linked to an  $N_{ice}$  reduction by introducing SAF can be expected from the corresponding change in optical properties alone, while a similar RF reduction has been reported from GCM calculations in the literature. This is an encouraging result, but until further measurements are made, a simple relationship between lower soot numbers from SAF and  $N_{ice}$  cannot be assumed since theoretical modeling shows that at very low soot numbers ( $<10^{14}$  soot  $\text{kg}^{-1}$  fuel) and cold conditions ( $\geq 12$  K below contrail formation threshold temperatures), ultrafine aqueous particles may act to increase  $N_{ice}$  again [2]. Nonetheless, reducing soot, and therefore  $N_{ice}$ , at temperatures above contrail formation values minus 12 K, to this threshold point of  $10^{14}$  soot  $\text{kg}^{-1}$  fuel, could represent a mitigation option for contrails by reducing  $N_{ice}$  for fossil-based fuel, with no added potential  $\text{CO}_2$  penalties, as opposed to other strategies like contrail avoidance by flight rerouting.

Based on the differences in  $N_{ice}$  and IWC between natural cirrus and contrails reported in the literature, our results show that a 60% IWC reduction, similar to the differences found by Li *et al* [21], produces a larger net RF reduction ( $\sim 55\%$ ), compared to that ( $\sim 35\%$ ) from the reported larger  $N_{ice}$  values also reported for contrails. It is nevertheless concluded that the accuracy and representation of these variables combined needs to be improved in order to better estimate contrail RF and its dependence on the introduction of SAF.

## Data availability statement

The data cannot be made publicly available upon publication because they contain commercially sensitive information. The data that support the findings of this study are available upon reasonable request from the authors.

## Acknowledgments

This work was carried out under the EU funded project ACACIA (Grant Agreement No. 875036) ([www.acacia-project.eu](http://www.acacia-project.eu)), and the UK Department of Transport under contract TISEA00001 (Aviation Atmospheric, Environmental Technical Support). The traffic data used in this study are available through the REACT4C Project to non-commercial users after signing a data agreement. The authors thank Paul Field, Anthony Baran, and David Mitchell for very useful discussions and the thorough suggestions from two anonymous reviewers which greatly improved the model setup and the interpretation of the results. The authors declare no conflict of interest with regards to the results presented in this study.

## Appendix

The contrail cover distribution used in the simulations was generated using the Contrail Modeling and Analysis tool (COMA), which is based on the ECHAM4 contrail parameterization and the offline methodology described in Sausen *et al* [36]. The calculations used aircraft movements from the International Civil Aviation Organization (ICAO) Committee on Aviation Environmental Protection (CAEP) Modeling and Database Group (MDG), which were incorporated into the FAST emissions model to calculate the distance travelled for the year 2006 (ICAO) [37].

The global distance travelled, from ground level up to 44 000 ft on 2000 ft intervals, was approximately 38.9 billion km, with 87% occurring above 500 hPa. The diurnal distance travelled over 4 time bands (00, 06, 12 and 18 UTC) was coupled with the 2006 European Centre for Medium-Range Weather Forecasts (ECMWF) ERA-Interim meteorological datasets (regular grid,  $2.5^\circ \times 2.5^\circ$ ). We plan to update this and the background climatology in our models to use the more recent ERA5 dataset in the near future. The cloud



**Table A2.** Input for the SW and LW parameterizations based on 6 temperature values covering a representative range of contrail temperatures, and minimum, mean, and maximum climatological IWC fits from Schiller *et al* [25] (SO8). These pairs of 6 temperature and 3 IWC values are used as input to produce 18 PSDs following the scheme from McFarquhar and Heymsfield [24] (MH97). The radiative runs use IWC and  $D_{ge}$  calculated for each cell, also based on SO8 and MH97.

Parameterizations Input	SO8	MH97	Radiative Parameterizations	
Temperature	IWC→	$D_{ge}$ →	Fu (1996), Fu (1998)	
6 sample Temperatures	Minimum	→		
	Mean	→	→	SW LW RF
	Maximum	→		
Runs→ Input	Temperature→ ECMWF	IWC + $D_{ge}$ → SO8 + MH97	↑	

cover from ERA-Interim was used by COMA to calculate the critical relative humidity for cloud formation. The specific humidity and temperature were then used to determine the potential contrail coverage. This coverage was then multiplied by the travelled distance in each cell to produce the actual contrail coverage, assuming an aircraft fleet propulsion efficiency of 0.36. This resulted in an annual global contrail cover of 0.10%, based on traffic for the year 2006, which incorporated a representative diurnal variability.

The instantaneous RF, defined as the difference at the top of the atmosphere of the radiative fluxes with and without contrails, was calculated using the UK's Met Office's GCM's two-stream radiative transfer scheme (SOCRATES) [38] with a 144 (longitude)  $\times$  72 (latitude)  $\times$  23 (altitude) resolution. Solar zenith angles and day lengths at the middle of the month were used to represent the monthly mean SW fluxes with a five points Gaussian integration. The contrail radiative properties were represented offline in SOCRATES as functions of two prognostic variables, IWC and ambient temperature. The latter is prescribed using ERA40 [39] ECMWF reanalysis data, from which all background meteorology is prescribed as monthly climatological averages for water vapor, temperature, and surface albedo.

The parameterizations used in the simulations are schematically represented in table A2. The contrail's IWC is determined using its climatological dependence on ambient temperature developed by Schiller *et al* [25] and used as input in McFarquhar and Heymsfield's [24] (MH97 hereafter) PSD scheme by choosing six contrail representative temperature values, covering a range from 195 to 245 K. Three fits from Schiller *et al* [25] for minimum, mean and maximum IWCs for each temperature are then used to produce 18 PSDs. These PSDs are then used to integrate the ice crystal single scattering properties (extinction coefficient, single scattering albedo, and asymmetry parameter) of randomly oriented hexagonal cylinders to parameterize these properties in terms of the PSD's IWC and generalized diameter ( $D_{ge}$ ) [40] which ranged from 11 to 75  $\mu\text{m}$ .

The methodology presented in Fu *et al* [40] is used to integrate the LW optical properties calculated by Baran *et al* [41]. In the SW, the parameterization described by Fu [42] and the single scattering properties calculated by Yang *et al* [43] are applied, integrated over 6 and 9 bands in the SW and in the LW spectral regions, respectively. The aspect ratio (ratio of the ice crystal length and width) of the prescribed hexagonal cylinders varies with particle size in a way that matches both the total volume and projected areas retrieved by MH97, making the micro-physical and the optical properties in the model consistent.

## ORCID iDs

Rubén Rodríguez De León  <https://orcid.org/0000-0002-6640-3211>

David S Lee  <https://orcid.org/0000-0002-5984-8861>

## References

- [1] Kärcher B and Yu F 2009 Role of aircraft soot emissions in contrail formation *Geophys. Res. Lett.* **36** 1–5
- [2] Kärcher B 2018 Formation and radiative forcing of contrail cirrus *Nat. Commun.* **9** 1–17
- [3] Moore R *et al* 2017 Biofuel blending reduces particle emissions from aircraft engines at cruise conditions *Nature* **543** 411–5
- [4] Kärcher B 2016 The importance of contrail ice formation for mitigating the climate impact of aviation *J. Geophys. Res.* **121** 3497–505
- [5] Kärcher B and Voigt C 2017 Susceptibility of contrail ice crystal numbers to aircraft soot particle emissions *Geophys. Res. Lett.* **44** 8037–46
- [6] Burkhardt U, Bock L and Bier A 2018 Mitigating the contrail cirrus climate impact by reducing aircraft soot number emissions *npj Clim. Atmos. Sci.* **1** 1–7
- [7] Bock L and Burkhardt U 2019 Contrail cirrus radiative forcing for future air traffic *Atmos. Chem. Phys.* **19** 8163–74
- [8] Schumann U 1996 On conditions for contrail formation from aircraft exhausts *Meteorol. Z.* **5** 4–23
- [9] Schumann U and Heymsfield A J 2017 On the life cycle of individual contrails and contrail cirrus *Meteorol. Monogr.* **58** 3–1

- [10] Schumann U *et al* 2017 Properties of individual contrails: a compilation of observations and some comparisons *Atmos. Chem. Phys.* **17** 403–38
- [11] Zhang Y, Macke A and Albers F 1999 Effect of crystal size spectrum and crystal shape on stratiform cirrus radiative forcing *Atmos. Res.* **52** 59–75
- [12] Rodríguez De León R and Haigh J D 2007 Infrared properties of cirrus clouds in climate models *Q. J. R. Meteorol. Soc.* **133** 273–82
- [13] Mitchell D, Lawson R and Baker B 2011 Understanding effective diameter and its application to terrestrial radiation in ice clouds *Atmos. Chem. Phys.* **11** 3417
- [14] Markowicz K M and Witek M 2011 Sensitivity study of global contrail radiative forcing due to particle shape *J. Geophys. Res.* **116** 1–15
- [15] Ponater M, Marquart S and Sausen R 2002 Contrails in a comprehensive global climate model: parameterisation and radiative forcing results *J. Geophys. Res.* **107** 4164
- [16] Rodríguez De León R, Lim L L, Lee D S, Bennett M and Krämer M 2018 Simple versus complex physical representation of the radiative forcing from linear contrails: a sensitivity analysis *J. Geophys. Res.* **123** 2831–40
- [17] Schumann U, Schlager H, Arnold F, Baumann R, Haschberger P and Klemm O 1998 NO and NO<sub>y</sub> in the upper troposphere: nine years of CARIBIC measurements onboard a passenger aircraft *Atmos. Environ.* **32** 3097–103
- [18] Voigt C *et al* 2017 ML-CIRRUS: the airborne experiment on natural cirrus and contrail cirrus with the high-altitude long-range research aircraft HALO *Bull. Am. Meteorol. Soc.* **2** 271–88
- [19] Chauvigné A *et al* 2018 Statistical analysis of contrail to cirrus evolution during the contrail and cirrus experiment (CONCERT) *Atmos. Chem. Phys.* **18** 9803–22
- [20] Jeßberger P, Voigt C, Schumann U, Sölch I, Schlager H, Kaufmann S, Petzold A, Schäuble D and Gayet J F 2013 Aircraft type influence on contrail properties *Atmos. Chem. Phys.* **13** 11965–84
- [21] Li Y *et al* 2023 Upper-tropospheric slightly ice-subsaturated regions: frequency of occurrence and statistical evidence for the appearance of contrail cirrus *Atmos. Chem. Phys.* **23** 2251–71
- [22] Søvdø O *et al* 2014 Aircraft emission mitigation by changing route altitude: a multi-model estimate of aircraft NO<sub>x</sub> emission impact on O<sub>3</sub> photochemistry *Atmos. Environ.* **95** 468–79
- [23] Field P R, Heymsfield A J and Bansemir A 2007 Snow size distribution parameterization for midlatitude and tropical ice clouds *J. Atmos. Sci.* **64** 4346–65
- [24] McFarquhar G M and Heymsfield A J 1997 Parameterization of tropical cirrus ice crystal size distributions and implications for radiative transfer: results from CEPEX *J. Atmos. Sci.* **54** 2187–200
- [25] Schiller C, Krämer M, Afchine N, Spelten A A and Sitnikov N 2008 Ice water content of Arctic, midlatitude, and tropical cirrus *J. Geophys. Res.* **113** 1–12
- [26] Baran A J, Hill P, Walters D, Hardiman S C, Furtado K, Field P R and Manners J 2016 The impact of two coupled cirrus microphysics–radiation parameterizations on the temperature and specific humidity biases in the tropical tropopause layer in a climate model *J. Clim.* **29** 5299–316
- [27] Vidot J, Baran A J and Brunel P 2015 A new ice cloud parameterization for infrared radiative transfer simulation of cloudy radiances: evaluation and optimization with IIR observations and ice cloud profile retrieval products *J. Geophys. Res. Atmos.* **120** 6937–51
- [28] Krämer M *et al* 2009 Ice supersaturations and cirrus cloud crystal numbers *Atmos. Chem. Phys.* **9** 3505–22
- [29] Waliser D E *et al* 2009 Cloud ice: a climate model challenge with signs and expectations of progress *J. Geophys. Res.* **114** 1–27
- [30] Teoh R, Schumann U, Voigt C, Schripp T, Shapiro M, Engberg Z, Molloy J, Koudis G and Stettler M E 2022 Targeted use of sustainable aviation fuel to maximize climate benefits *Environ. Sci. Technol.* **56** 17246–55
- [31] Voigt C *et al* 2021 Cleaner burning aviation fuels can reduce contrail cloudiness *Commun. Earth Environ.* **2** 114
- [32] Yi B, Yang P, Liou K N, Minnis P and Penner J E 2012 Simulation of the global contrail radiative forcing: a sensitivity analysis *Geophys. Res. Lett.* **39** 1–5
- [33] De León R R, Krämer M, Lee D and Thelen J 2012 Sensitivity of radiative properties of persistent contrails to the ice water path *Atmos. Chem. Phys.* **12** 7893–901
- [34] Schumann U and Graf K 2013 Aviation-induced cirrus and radiation changes at diurnal timescales *J. Geophys. Res.* **118** 2404–21
- [35] Myhre G *et al* 2009 Intercomparison of radiative forcing calculations of stratospheric water vapour and contrails *Meteorol. Z.* **18** 585–96
- [36] Sausen R, Gierens K and Ponater M 1998 U. S. A diagnostic study of the global distribution of contrails part I: present day climate *Theor. Appl. Climatol.* **61** 121–41
- [37] ICAO, International Civil Aviation Organization (ICAO)/Committee on Aviation Environmental Protection (CAEP), agenda item 4: modeling and databases task force (MODTF) goals assessment results *Proc. ICAO/CAEP Working Paper, Steering Group Meeting (Salvador, Brazil, 22–26 June 2009)*
- [38] Edwards J and Slingo A 1996 Studies with a flexible new radiation code I: choosing a configuration for a large-scale model *Q. J. R. Meteorol. Soc.* **122** 689–719
- [39] Simmons A and Gibson J 2000 The ERA-40 project plan *ERA-40 Proj. Rep. Ser 1*
- [40] Fu Q, Yang P and Sun W B 1998 An accurate parameterization of the infrared radiative properties of cirrus clouds for climate models *J. Clim.* **11** 2223–37
- [41] Baran A J, Havemann S and Mackowski D 2001 A database of hexagonal column optical properties for wavelengths ranging between 0.2  $\mu\text{m}$ s to 30  $\mu\text{m}$ s produced for ANNEX 7 (Department of the Environment, Food and Rural Affairs)
- [42] Fu Q 1996 An accurate parameterization of the solar radiative properties of cirrus clouds for climate models *J. Clim.* **9** 2058–82
- [43] Yang P, Liou K, Wyser K and Mitchell D 2000 Parameterization of the scattering and absorption properties of individual ice crystals *J. Geophys. Res.* **105** 4699–718