

Article

Addressing Confidence in Modeling of Contrail Formation from E-Fuels in Aviation Using Large Eddy Simulation Parametrization

Eduardo Cabrera ¹ and João M. Melo de Sousa ^{2,*} ¹ Instituto Superior Técnico, Av. Rovisco Pais, 1049-001 Lisboa, Portugal; eduardo.cabrera@tecnico.ulisboa.pt² IDMEC, Instituto Superior Técnico, Universidade de Lisboa, Av. Rovisco Pais, 1049-001 Lisboa, Portugal

* Correspondence: msousa@tecnico.ulisboa.pt

Abstract: As it becomes increasingly necessary to reduce aviation-related emissions, condensation trails present an additional challenge. These are arguably responsible for the largest contribution to radiative forcing in the sector, but the phenomenon is still not as well understood as those involving other agents. The present study employs a large eddy simulation (LES) parametrization to validate a previously developed contrail model in order to assess the feasibility of a multi-model approach to increase confidence in simulations of contrail cirrus formation. Subsequently, the computational model was used to analyze the impact of e-fuels in contrail dynamics, resulting in reductions of over 7% and 14%, respectively, in average contrail lifetime and optical depth, with such improvements increasing if higher blending limits are utilized. This confirmed the potential for e-fuels as the most viable option for near-future large-scale implementations among all sustainable aviation fuel alternatives.

Keywords: contrails; large eddy simulation parametrization; e-fuels; sustainable aviation fuel



Citation: Cabrera, E.; de Sousa, J.M.M. Addressing Confidence in Modeling of Contrail Formation from E-Fuels in Aviation Using Large Eddy Simulation Parametrization. *Energies* **2024**, *17*, 1442. <https://doi.org/10.3390/en17061442>

Academic Editor: Paul Stewart

Received: 6 January 2024

Revised: 6 March 2024

Accepted: 14 March 2024

Published: 17 March 2024



Copyright: © 2024 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (<https://creativecommons.org/licenses/by/4.0/>).

1. Introduction

As with all other transport industries, the aviation sector is faced with the both current and future problem of reducing pollutant emissions in mankind's struggle to combat climate change. Furthermore, amid the disruptions associated with the COVID-19 pandemic years also came confirmation of the scale of the sector's carbon footprint, as studies carried out during the most intense lockdown periods showed an immediate reduction in the emissions of greenhouse gases (GHG) and improvement in air quality in areas of otherwise usually high air traffic density [1]. Nevertheless, the steady, albeit slow, return to normality highlighted the need to resume and accelerate efforts in research and development of technologies that may facilitate the transition to carbon-neutral aviation, namely, sustainable aviation fuels (SAF). The latter will play the most important role towards achieving the established near- and medium-term goals, as evidenced by the increasing number of test flights conducted with 100% blends of SAF [2,3].

The most visible and recognizable pollutant emissions associated with the aviation sector are condensation trails (or contrails). Contrails are aircraft-induced cirrus clouds, which may persist and grow to large cirrus cover in ice-supersaturated air. These are triggered from the mixing of the hot and humid engine exhaust gases with ambient cold and dry air. Their optical properties depend not only on the ambient atmospheric conditions but also on both the particles emitted by the aircraft (which, in turn, are determined by the fuel properties and engine control settings) and the particles formed in the aircraft plume [4].

From a physicochemical point of view, contrails originate from the water vapor emitted by aircraft engines that condensates on particle nuclei and subsequently freezes. In order to meet the necessary formation conditions, the ambient air must reach water saturation, and for contrails to persist, the air needs to maintain supersaturation with respect to ice. It is

worth pointing out that, due to the fact that the bonds established between molecules in ice are much stronger than the ones in the case of liquid water, there will be a lower flux out of the ice into the vapor space, which means that saturation with respect to ice is achieved with a lower concentration of water molecules than saturation with respect to liquid water. In turn, this means that water saturation will always imply ice supersaturation [5].

In dry air environments, contrails are typically short-lived as lack of sufficient relative humidity levels leads to the evaporation of the ice particles that form in the contrail. In these cases, contrails can disappear only seconds or a few minutes after formation and, therefore, do not achieve extensive lengths. For a contrail to persist, the ambient humidity usually must be larger than the saturation humidity over ice surfaces; relative humidity over ice $RH_i > 100\%$. In such conditions, the ice particles within the contrails grow by deposition of water vapor molecules from the ambient air, and the contrails will persist while the ambient air remains in a state of ice-supersaturation. Contrail cirrus, however, can also form and persist in weakly ice-supersaturated air. Aircraft engines release not only water vapor, which promotes water saturation, but also heat, which counteracts the former. Hence, if the water vapor concentration increase due to the emitted water vapor is higher than the total increase in the local liquid water saturation point due to the emitted heat, cirrus cloud formation conditions can be met with only an ice saturation environment instead of typical high ice-supersaturation (of at least 145 to 165%). As a consequence, the latter contrail cirrus clouds can maintain their shape for up to hours, until they evaporate or lose their shape, thus becoming indistinguishable from natural cirrus clouds [6,7].

The impact of contrails and aviation-induced cloudiness on the global climate is still under debate, as this problem only started to be properly addressed in the last decades of the twentieth century, especially following the detection of the ozone hole. Despite having a much shorter lifetime than long-lived GHG, studies have estimated a net radiative forcing (RF) associated with contrail cirrus of 37.5 mW m^{-2} , making it the largest contribution in the aviation sector [7]. There are, however, high uncertainties associated with this estimate, namely, when compared to the uncertainties corresponding to the RF calculations made for major pollutants, such as CO_2 and NO_x , as portrayed in Figure 1 (only the impact of aerosol-cloud interactions presents a lower confidence level, due to lack of studies and very poor understanding of the processes [8]).

Notably, and despite the fact that the sulfur content in commercial aviation fuel (Jet A/A-1) currently averages 500 ppm worldwide (well below the specifications limit), consensus is still lacking concerning the role of sulfur in the formation of contrails [9–14]. However, it must also be noted that a comprehensive review of the properties of individual contrails [10] has reported changes in fuel sulfur content to have minimal impact on contrail onset. Regardless, some of the aforementioned studies have introduced modeling of activation of hydrophobic soot particles, via surface adsorption of sulfur compounds resulting from jet fuel combustion, in order to make those hydrophilic and enable the formation of liquid droplets or ice nuclei. Nevertheless, recent experiments [15] have demonstrated that fuel sulfur up to 700 ppm does not affect soot particle hygroscopicity, thus supporting the view of a weak impact of sulfur compounds on contrail formation.

While plume ice cloud models with various degrees of complexity for mixing and particle microphysics have been developed, most models do not account for the whole contrail life-cycle from contrail formation until dissipation [9]. Furthermore, other more elaborate models, due to their nonlinear nature and strong dependency on meteorological parameters related to contrail formation, suffer from being too computationally demanding, and thus are not suitable for large-scale simulations. The implications of the foregoing scenario may be that the full climatic impact of contrails is still being overlooked. This, nevertheless, also offers an opportunity for the study of effective mitigation measures, due to the fact that any effects would quickly become apparent.

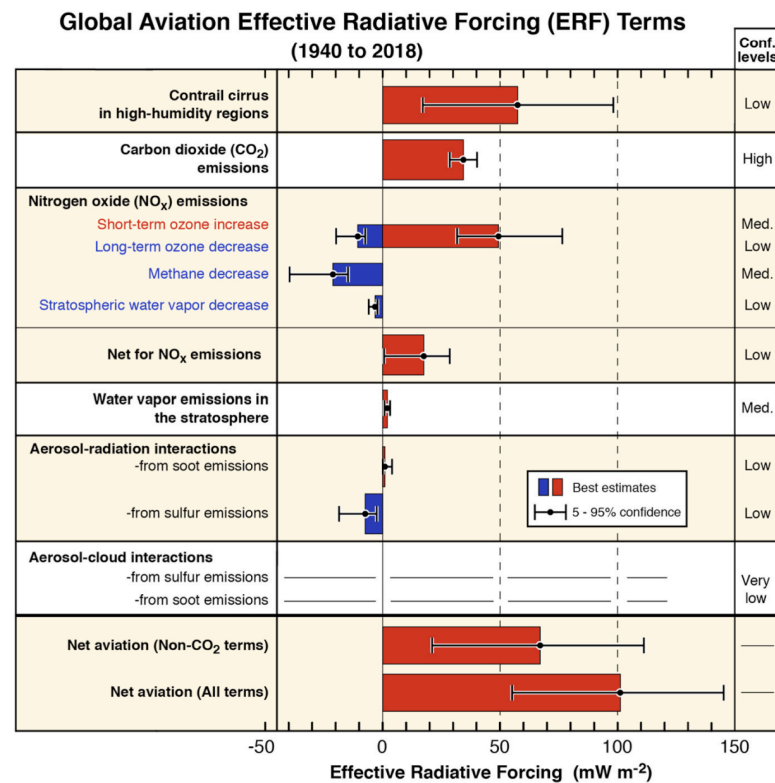


Figure 1. Best-estimates for climate forcing terms from global aviation from 1940 to 2018. Confidence levels were defined based on available evidence and agreement. Adapted from Lee et al. [8].

Against this background, the present investigation seeks to expand on the uncertainties associated with most current contrail prediction modeling approaches. Aiming to achieve this goal, the contrail model developed by Narciso and Sousa [16], that was assembled together with the 0-D aircraft engine model proposed earlier by Gaspar and Sousa [17], will be utilized. In previous studies the intermediate calculations in the modeling resulted in larger than initially expected errors, which were attributed to the simplifications and assumptions made due to the high uncertainties and complexity associated with contrail formation, and thus require further validation. In that respect, an LES-based contrail parametrization developed by Unterstrasser [18] was selected to be implemented, and a comparative analysis between its results and those provided by the original modeling for young contrails was conducted with the aim of improving the fidelity of contrail formation predictions. Furthermore, the present research is part of a larger effort to continue the work initiated by Cabrera and Sousa [1], who analyzed in detail the current state of SAF development and production to conclude that synthetic or e-fuels show the most promise to be incorporated in future large-scale implementations. Hence, the aforementioned engine model was also utilized to enable computational simulations regarding the impact of e-fuel usage in different blending ratios.

2. Original Contrail Model

2.1. Contrail Formation

Contrail formation can be explained through the Schmidt–Appleman criterion (SAC), which is used to decide whether an atmosphere is cool and humid enough to allow for the formation of contrails. When the SAC is satisfied, it means that the ambient temperature is below a critical temperature T_C , or, alternatively, the ambient relative humidity is above the critical value U_C , and the plume mix of exhaust gases and ambient air achieves liquid saturation.

The amount of water vapor has an extreme importance in determining if the SAC is satisfied. The water vapor emission index EI_{H_2O} can be calculated from Equation (1), where m_H is the hydrogen mass ratio in the fuel, and M_{H_2O} and M_H are the molar masses of water and hydrogen, respectively, as follows:

$$EI_{H_2O} = \frac{m_H M_{H_2O}}{2M_H}. \quad (1)$$

From here, with the use of gas and ambient data, such as the individual gas constants of water vapor and air, respectively, R_{H_2O} and R_{air} , the pressure P , and the specific heat of air at constant pressure c_p , together with engine outputs, such as the overall propulsion efficiency η , and fuel properties, such as the lower heating value Q , the mixing line gradient G , relating the variation in the water vapor partial pressure with temperature, can be computed following a series of steps that conclude with Equation (2), as given by:

$$G = \frac{R_{H_2O}}{R_{air}} \frac{c_p EI_{H_2O} P}{Q(1 - \eta)}. \quad (2)$$

Using parameter G , the critical temperature can be calculated by solving Equation (3), where e_{sat} stands for the partial water pressure at saturation, and T_M refers to the temperature at the point where the saturation curve and the mixing line meet. The temperature T_C is thereby obtained by equating the gradients, as follows:

$$T_C = T_M - \frac{1}{G} (e_{sat}(T_M) - U_{amb} e_{sat}(T_C)). \quad (3)$$

2.2. Early Stage Contrails

Early stage contrails interact with the engine jet, the aircraft wake vortex, as well as with the ambient turbulence, stratification, and wind shear S . Modeling all these interactions in detail would be difficult due to the physical complexity associated with wake dynamics. Following Schumann [9], the present model estimates the contrail properties at the end of this phase from aircraft and atmospheric parameters instead, as illustrated in Figure 2. The baseline aircraft dimensions were modeled after the BAe 146, as it uses the Lycoming ALF 502 engines that were considered in the development of the 0-D engine model by Gaspar and Sousa [17].

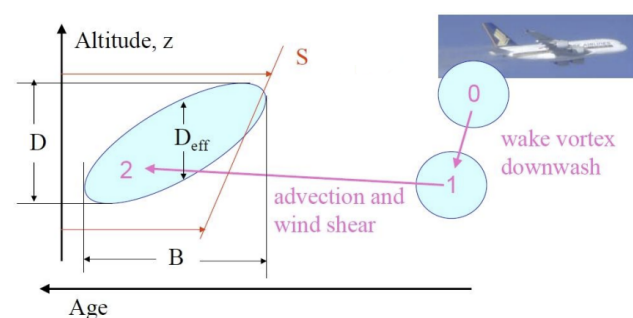


Figure 2. Schematic of contrail size and dynamics with respect to altitude and age, subjected to wind shear S . Stage 0 represents the contrail at the time of formation, $t = 0$; stage 1 represents the contrail after sinking due to the wake vortex downwash at $t = t_0$; stage 2 represents the cross-section of an aged contrail. Adapted from Schumann [9].

The subsequent modeling is formulated in normalized form, with the characteristic scales based on the initial vortex separation b_0 , and the initial vortex circulation Γ_0 , respectively, obtained from Equations (4) and (5). In the following expressions, s_a , M_a , and TAS denote the wingspan, the mass, and the true airspeed of the aircraft, respectively, ρ is the air density, and g stands for the gravitational acceleration, as follows:

$$b_0 = \frac{\pi}{4} s_a, \quad (4)$$

$$\Gamma_0 = \frac{4M_a g}{\pi s_a \rho T A S}. \quad (5)$$

From here, the timescale t_0 is obtained using Equation (6). The initial descent speed ω_0 and the normalized eddy dissipation rate ϵ^* (from its dimensional value ϵ) are also calculated using Equations (7) and (8), given by:

$$t_0 = \frac{2\pi b_0^2}{\Gamma_0}, \quad (6)$$

$$\omega_0 = \frac{\Gamma_0}{2\pi b_0}, \quad (7)$$

$$\epsilon^* = \frac{(\epsilon b_0)^{\frac{1}{3}}}{\omega_0}. \quad (8)$$

For distinction between weak and strong stable stratification, the Brunt-Väisälä frequency is normalized in Equation (9) as N^* from its dimensional value N_{BV} computed in Equation (10). In Equation (11), θ represents the potential temperature that a parcel of dry air at temperature T_E and pressure P_E would have if isentropically brought to the reference pressure $P_{ref} = 1000$ mbar, as follows:

$$N^* = N_{BV} t_0, \quad (9)$$

$$N_{BV} = \sqrt{\frac{g}{\theta} \frac{d\theta}{dz}}, \quad (10)$$

$$\theta = T_E \left(\frac{P_{ref}}{P_E} \right)^{R_{air}/c_p}. \quad (11)$$

Depending on the value of N^* , the maximum descent value of the contrail Δz_w will be calculated according to the brackets defined in Equation (12). At this point, it is now possible to empirically calculate both the contrail depth D , using Equation (13), and the contrail width (or breadth) B , using Equation (14), at the stage where the initial sinking ends (previously defined in Figure 2 as stage 1 and denoted by the latter index in subscript). Based on the foregoing parameters, an effective vertical depth D_{eff} can also be calculated based on the elliptic (Gaussian) cross-section shape assumed in Figure 2 for the purpose of optical depth computations of aged contrails. In the following equations, m_f denotes the mass flow of fuel per flight distance and C_{D0} is set to 0.5, but the numerical value of the other empirical constants has already been inserted, as shown below:

$$\begin{cases} \Delta z_w = 1.49 \frac{\omega_0}{N_{BV}}, & N^* \geq 0.8 \\ \Delta z_w = b_0 (7.68(1 - 4.07\epsilon^* + 5.67\epsilon^{*2})(0.79 - N^*) + 1.88), & N^* < 0.8 \wedge \epsilon^* \leq 0.36 \end{cases} \quad (12)$$

$$D_1 = C_{D0} \Delta z_w, \quad (13)$$

$$B_1 = \frac{7000 t_0^{0.8} m_f}{\frac{\pi}{4} \rho D_1}. \quad (14)$$

2.3. Ice Properties

Contrail ice properties at different stages are defined by the following quantities: the mass mixing ratio of ice I and the total number concentration of contrail ice particles per contrail length N_i . When in soot-rich conditions (consistent with present-day emissions from aero-engines), the homogeneous freezing of water-activated soot is the primary ice formation mechanism; therefore, soot emissions dominate ice particle formation [13]. Hence, the initial number of ice particles N_{form} is calculated using Equation (15), as follows:

$$N_{form} = \frac{EI_N m_f}{TAS}. \quad (15)$$

For the computation of I , thermal equilibrium between the contrails and the ambient air after contrail formation is assumed, which allows the use of thermodynamic relations. The initial mass mixing ratio of ice I_0 is calculated using Equation (16). This quantity is a function of two ambient properties, namely, the ambient humidity q_0 and the saturation humidity q_{sat} , which, in turn, depends on the local pressure P_0 and temperature T_0 , as given by:

$$I_0 = \frac{EI_{H_2O} m_f}{\frac{\pi}{4} \rho D_1 B_1} + q_0 - q_{sat}(P_0, T_0). \quad (16)$$

The ice mass variation between stage 0 and stage 1, denoted by ΔI_{ad} , is obtained using Equations (17) and (18), where ΔT_{ad} is the adiabatic temperature variation and the subscript *ice* stands for the ice saturation, as follows:

$$\Delta I_{ad} = \frac{P_{ice}(T_0 + \Delta T_{ad})}{P_1} - \frac{P_{ice}(T_0)}{P_0}, \quad (17)$$

$$\Delta T_{ad} = T_0 \frac{R_{air}}{c_p} \frac{P_1 - P_0}{P_0}. \quad (18)$$

From the ice mass variation, the number of ice particles per length at stage 1, denoted by N_1 , is provided using the survival factor $f_{surv} = \frac{I_1}{I_0}$, as in Equation (19) given by:

$$N_1 = f_{surv} N_{form}. \quad (19)$$

2.4. Contrail Cirrus

After the initial contrail phase, the model enters a loop, thus allowing calculation of the contrail properties along its remaining life-span in order to determine the potential environmental impact. The procedure followed to simulate the life and properties of the contrail cirrus in the original modeling is based on the CoCiP model proposed by Schumann [9], and additional details of the present approach are given by Narciso and Sousa [16].

Briefly, for each calculation point, the contrail trajectory is first updated to account for advection effects using a second-order two-step Runge–Kutta scheme, with the latitude and longitude changing due to the northward and eastward winds, while the vertical trajectory is represented by the ambient pressure variation. For ice crystal particle loss, the present model accounts for sublimation of smaller particles during the turbulent mixing, as well as sedimentation-induced aggregation. Due to this, the contrail is taken as being displaced downward according to the ice particle terminal fall velocity. Contrail dimensions, ice mass, ice particle number, and the optical depth τ are thereby obtained at each trajectory point. The simulation is terminated when the contrail lifetime is assumed to end, which can be defined when there is no more mass content, or when the optical depth reaches a small enough value. In the present case, the calculation loop ends when $\tau < 10^{-4}$.

3. LES-Based Contrail Parametrization

As mentioned in Section 2, simplifying assumptions were made in the original modeling due to the complexities and high uncertainties associated with contrail formation. In the early stage model, calculations were simplified to account for the lack of suitable models that can accurately simulate the highly complex wake vortex dynamics without incurring a high computational cost. However, this may imply that the uncertainties associated with that calculation phase, while initially accepted as within reasonable margins, could affect the reliability of the results generated by the subsequent contrail cirrus loop, and ultimately lead to undesirable consequences for the validity of the final results.

To assess uncertainties associated with the original contrail model, the parametrization proposed by Unterstrasser [18] was implemented, thus providing an alternative approach

for calculation of the early evolution stages of contrails. Since this parametrization is based on LES modeling and, thus, incorporates fine details of the wake vortex dynamics as well as Lagrangian ice microphysics, aiming to explore how vortex processes affect later contrail-cirrus properties [19], a higher confidence level of the corresponding results may be expected. Furthermore, as the parametrization condenses the LES data in a simpler analytical form, its implementation can be performed without severely compromising the run time of the original model, which made its choice suitable for the ensuing validation study. More information about the LES approach can be found in Appendix B.

3.1. Compatibilization of Inputs

As mentioned earlier, the implemented parametrization is based on fully three-dimensional (3D) LES simulations of the contrail vortex phase. These simulations begin at an initial state of seconds after the start of the wake vortex and proceed to obtain the contrail depth and the number of ice particles per length (via computation of the survival factor f_{surv}) for 5 min old contrails. A compatibilization of the inputs with the original model was required first, and these are summarized below.

- Ambient temperature T , from the atmospheric data model
- Supersaturation s_i , which is related to the ambient relative humidity $s_i = U_{amb} - 1$
- The dimensional Brunt-Väisälä frequency N_{BV}
- The ice crystal emission index EI_{ice} , which, again, is assumed to be equal to the soot emission index for soot-rich regimes
- The wingspan s_a and the mass M_a of the aircraft
- The mass flow of fuel per flight distance m_f

The parametrization was first implemented on a large dataset, with certain parameters having a defined range of values that are within the desired simulation conditions. For example, the temperature ranges between 209 and 225 K, while RH_i is set between 100 and 140%; both these limits are set so as to represent conditions of high likelihood of contrail formation and persistence. Furthermore, the Brunt-Väisälä frequency ranges between 0.005 and 0.0115 s⁻¹ to model typical upper tropospheric stable conditions, and the ice crystal emission index ranges between 1.4×10^{13} and 7×10^{15} kg⁻¹ to allow for both present engine soot emission values as well as the expected future decrease due to technological improvements. Likewise, though it will not be addressed in this paper, other aircraft-dependent parameters can be changed to reflect different types of aircraft. More information about the validity of the parametrization for different sets of values was given by Unterstrasser [18].

Similarly to the simulation model described in Section 2, the characteristic quantities b_0 and Γ_0 , as well as the timescale t_0 and the descent speed ω_0 , can be calculated using Equations (4) through (7).

3.2. Length Scales

Three length scales are introduced to characterize the effects of different phenomena in the evolution of early downward-sinking contrails, namely, z_{desc} , which measures the final vertical displacement of the wake vortex, whereas z_{atm} and z_{emit} quantify the impact of supersaturation and water vapor emission in the evolution of ice crystal mass, respectively. Regarding z_{desc} , an analytical approximation will be represented by $\hat{z}_{desc}$. After the circulation Γ_0 has been obtained, $\hat{z}_{desc}$ can be calculated using Equation (20) as follows:

$$\hat{z}_{desc} = \sqrt{\frac{8\Gamma_0}{\pi N_{BV}}}. \quad (20)$$

The water vapor emitted from the aircraft engine exhaust increases the water vapor concentration ρ until the plume is supersaturated. Assuming a uniform distribution, the added water vapor concentration ρ_{emit} is obtained using Equation (21), given by:

$$\rho_{emit} = \frac{\nu}{A_p}. \quad (21)$$

In turn, the water vapor emission ν and the plume area A_p appearing in Equation (21) are calculated for a plume of radius r_p from Equations (22)–(24), as follows:

$$\nu = \frac{\dot{m}_f}{TAS} EI_{H_2O}, \quad (22)$$

$$r_p = 1.5 + 0.314s_a, \quad (23)$$

$$A_p = 4\pi r_p^2. \quad (24)$$

Next, the length scales z_{atm} and z_{emit} can be obtained by solving Equations (25) and (26) employing a root-finding bisection method. The dry adiabatic lapse rate Γ_d and the gas constant for water vapor R_{H_2O} are set to 9.8 K km^{-1} and $461 \text{ J kg}^{-1} \text{ K}^{-1}$, respectively, in the aforementioned equations written here as:

$$(1 + s_i) \frac{e_{sat}(T)}{T} = \frac{e_{sat}(T + \Gamma_d z_{atm})}{(T + \Gamma_d z_{atm})}, \quad (25)$$

$$\frac{e_{sat}(T)}{R_{H_2O}T} + \rho_{emit} = \frac{e_{sat}(T + \Gamma_d z_{emit})}{R_{H_2O}(T + \Gamma_d z_{emit})}. \quad (26)$$

Finally, the three length scales previously calculated are combined in Equation (27). The parametrization coefficients α_{atm} and α_{emit} are defined by Equations (28) and (29), assuming a reference ice crystal emission index $EI_{ice,ref} = 2.8 \times 10^{14}$. Whereas α_{desc} remains fixed at 0.6, the values of γ_{atm} and γ_{emit} are set depending upon the intended output (see next subsection) from the following expressions:

$$z_{\Delta} = \alpha_{atm}z_{atm} + \alpha_{emit}z_{emit} - \alpha_{desc}\hat{z}_{desc}, \quad (27)$$

$$\alpha_{atm} = 1.7 \left(\frac{EI_{ice}}{EI_{ice,ref}} \right)^{-\gamma_{atm}}, \quad (28)$$

$$\alpha_{emit} = 1.15 \left(\frac{EI_{ice}}{EI_{ice,ref}} \right)^{-\gamma_{emit}}. \quad (29)$$

3.3. Ice Crystal Loss

In the determination of the surviving ice crystals, γ_{atm} and γ_{emit} are both set to 0.18 when using Equation (27), aiming to account for the impact of excess moisture in ice crystal loss. The approximated survival factor $\hat{f}_{surv}$ is then obtained from Equations (30) and (31), with $\beta_0 = 0.45$, $\beta_1 = 1.19$ and $\alpha_0 = -1.35$, as follows:

$$\hat{f}_{surv} = \hat{a}(z_{\Delta}), \quad (30)$$

$$\hat{a}(x) = \beta_0 + \frac{\beta_1}{\pi} \arctan(\alpha_0 + \frac{x}{100}). \quad (31)$$

Once the survival factor has been determined, the number of ice crystals in the young contrails of age $t = 5 \text{ min}$ can be obtained again from Equations (15) and (19), as previously expressed for the original modeling.

3.4. Contrail Depth and Width

In contradistinction, as the contrail depth depends mostly on the impact of the wake vortex, and only slightly on crystal loss, the quantities z_{Δ} and $\hat{f}_H$ are computed with γ_{atm} and γ_{emit} set to 0; here, it must be noted that $\hat{f}_H$ is calculated in the same manner as $\hat{f}_{surv}$ using Equation (30).

The parametrized contrail depth $\hat{H}$ can be obtained through Equations (32) and (33), with $\eta_1 = 6$, $\eta_2 = 0.15$ and $x_s = 0.2$, given by:

$$\hat{H} = \hat{z}_{desc} \times \hat{b}(\hat{f}_H), \quad (32)$$

$$\hat{b}(x) = \begin{cases} \eta_1 x, & x \in [0, x_s] \\ \eta_2 x + (\eta_1 - \eta_2)x_s, & x \in [x_s, 1] \end{cases}. \quad (33)$$

The LES parametrization does not enable a direct computation of the contrail width. In fact, this parameter will not be as relevant as the contrail depth or the ice crystal number for the remainder of the contrail evolution. Nevertheless, an estimation of the horizontal spreading rate of a contrail can be obtained from the simple expression given by Equation (34), which is based on the evaluation of transverse profiles of the ice crystal mass after the vortex phase, as follows:

$$\dot{W} = \hat{H} \times S. \quad (34)$$

It is noteworthy that for typical values of the parametrized contrail depth $\hat{H} = 400$ m and vertical wind shear $S = 0.005 \text{ s}^{-1}$ (Cf. Figure 2), the parametrized contrail width $\hat{W}$ increases at a rate of $1/8 \text{ km min}^{-1}$, which, in practice, means that any errors eventually introduced by the initial width estimation would become less relevant in contrail-to-cirrus evolutions of several hours.

When the values of parameters $\hat{N}_{surv}$ (knowing the parametrized survival factor $\hat{f}_{surv}$), $\hat{H}$, and $\hat{W}$ have been calculated, the mean ice crystal number concentration n_{mean} can be estimated from Equation (35), provided that it is assumed that the ice crystals are spread over a rectangular cross section, as follows:

$$\hat{n}_{mean} = \frac{\hat{N}_{surv}}{\hat{H}\hat{W}}. \quad (35)$$

3.5. Implementation, Verification, and Validation

Aiming at a validation of the computational implementation of the parametrization modeling, Figure 3 shows a comparison between the simulated and parametrized survival factors. The selected simulation values were extracted from data available in reference [18], and were obtained from a series of 3D LES simulations of the contrail vortex phase with the use of an EULAG-LCM model system developed at the German Aerospace Center (DLR).

These simulations [19,20] study different conditions, with variations in temperature, relative humidity, ice crystal emission index, and fuel flow, in order to assess how these quantities impact contrail formation for an aircraft of the type of a Boeing 777. Additional data from similar simulations made for other aircraft types are also available in reference [18]. Altogether, these simulations indicate that the contrail depth depends most strongly on the supersaturation level and aircraft wingspan, and to a lesser extent on the temperature and the Brunt-Väisälä frequency, whilst the survival factor is mainly dependent on the supersaturation and ice crystal emission. As the supersaturation level and, consequently, the relative humidity, affects both the contrail depth and the survival factor, differences in its value for the different cases have been emphasized using a color system in Figure 3. The results show an average relative error of 12.4% for the cases analyzed, which implies a good level of confidence in the approximations.

A similar analysis for the contrail depth is presented in Figure 4, in which the average relative error was approximately 9.6%, which can be deemed acceptable, as even a difference of 20 to 30 m will not have a major impact in subsequent contrail evolution calculations. As the contrail depth is affected by the wingspan of the aircraft, most values for the cases shown in Figure 4 are within the 350 to 450 m range. Moreover, the calculated parametrized values show differences of less than 1% with respect to counterparts published by Unterstrasser [18], which proved that the computational implementation of the LES parametrization was successfully verified.

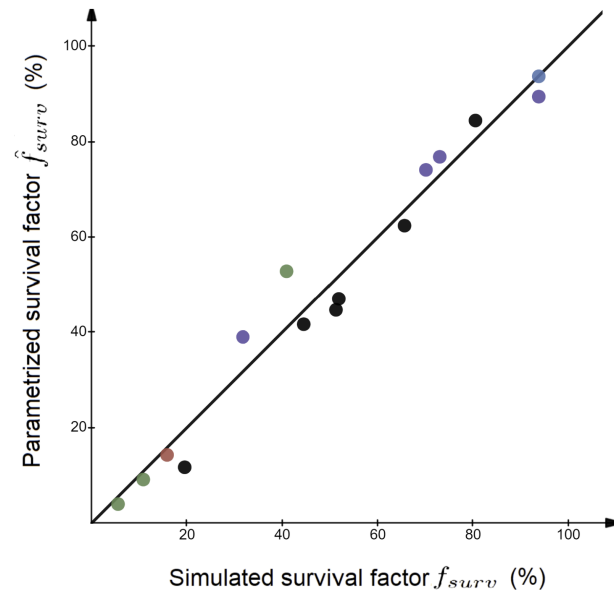


Figure 3. Comparison between simulated and parametrized survival factors. Red symbols represent simulations for saturated conditions, green for 10% supersaturation, black for 20%, purple for 30%, and blue for 40%.

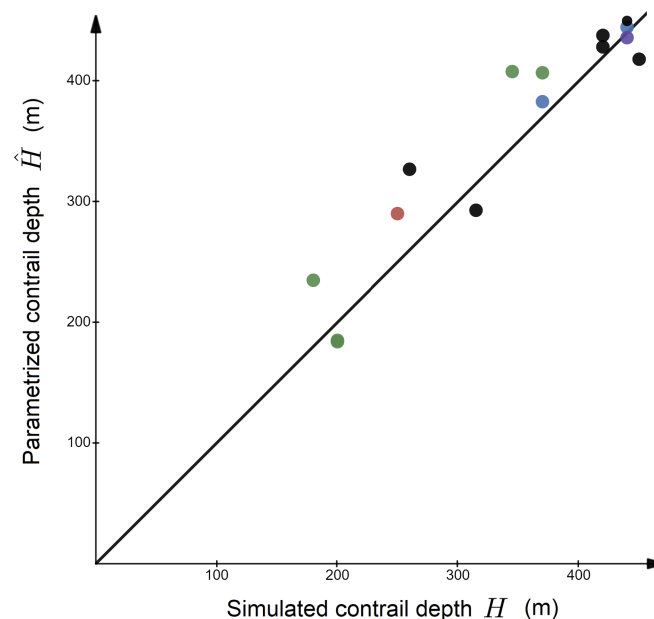


Figure 4. Comparison between simulated and parametrized contrail depths. Red dots represent simulations for saturated conditions, green for 10% supersaturation, black for 20%, purple for 30% and blue for 40%.

4. Simulation Results

4.1. Impact of E-Fuels

The computational tool based on the 0-D aircraft engine model mentioned in Section 2.2 requires a specific set of fuel properties to perform the subsequent operations. Hence, these properties need to be obtained from the existing literature or other available sources. As biofuel research and development have been carried out for decades, such properties are available through a selection of different sources.

Concerning e-fuels, since their development is considerably behind that of biofuels, and only more recently have efforts been made to accelerate it [1], detailed fuel properties are not readily available, as the existing literature is mainly focused on proof-of-concept

demonstrations. Nevertheless, since for the case of power-to-liquids (PtL) synthesized using the Fischer–Tropsch (FT) pathway the only difference in regard to other alternative fuels utilizing the FT-SPK process lies in the methods used for syngas production, one may, therefore, assume that the fuel properties remain relatively similar, and any conclusions regarding engine performance and contrail formation also apply. As such, the available data from FT fuels will be utilized in the present study, and the results will be averaged for comparison purposes.

The relevant fuels and respective fuel properties available in the database of the computational tool are presented in Table A1 of Appendix A. In addition, routes from the built-in database were selected so that not only the effect of different ambient conditions could be analyzed, but also so that these would accurately represent real flight routes routinely employed in air transport. For example, the results of optical depth at contrail trajectory points can be visualized using the computational tool with integrated contrail modeling, as illustrated in Figure 5 for one of the specified routes used to generate the bulk of the data presented and discussed in this section.

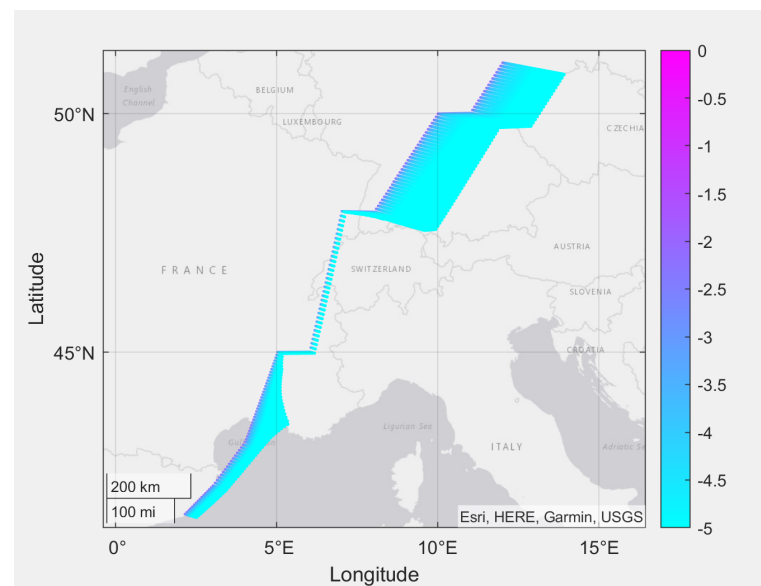


Figure 5. Contrail formation plotted over a typical flight route considered in the analysis of the impact of e-fuels. Colour contours represent the logarithm of the optical depth τ at trajectory points, revealing contrail cirrus for $\tau > 10^{-4}$.

In contrail formation-propitious environments, the impact of FT e-fuels on contrail lifetime and optical depth results was analyzed. Cloud optical depth studies are of paramount importance to assess cloud-radiation interactions, since the optical depth is key for determining shortwave cloud reflectance and absorption, longwave emissivities, and adiabatic heating rates [21]. As portrayed in Figure 6, the use of the mandatory 50% blending limit resulted in an average decrease of approximately 7.6% in contrail lifetime and 14.6% in optical depth for the studied cases. Impact on lifetime is less felt in contrails with short lifetimes, increasing in long-lasting contrail cirrus scenarios, as expected. For comparison, simulations were also carried for HEFA biofuels, as these currently have the highest TRL [1], and are currently subjected to a similar blending limit of 50%. Two fuels from different feedstocks (mixed fats and camelina) were analyzed, with fuels from camelina feedstocks (HEFA-C) showing a larger positive impact in both lifetime and optical depth reduction (7.4 and 14.5%, respectively) than those from mixed fats feedstocks (6.1 and 12.0%).

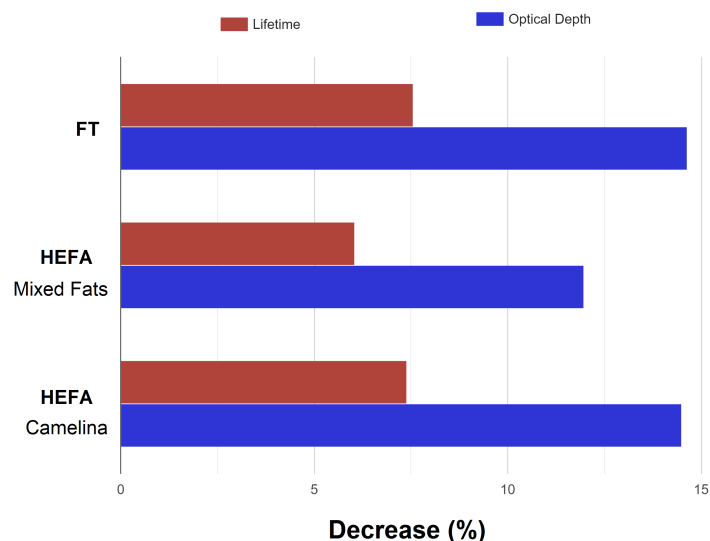


Figure 6. Impact of FT and HEFA fuels in contrail lifetime (red) and optical depth (blue), with respect to simulations for 100% conventional jet fuel, for the maximum blending limits currently allowed.

Since one of the near- to medium-term goals for the alternative fuel industry is to achieve higher values of maximum allowed blending limits, eventually reaching 100% conventional fuel substitution, the effect of different blends should also be analyzed. As shown in Figure 7, a higher volume of SAF naturally yields larger environmental gains regarding both the lifetime and optical depth of contrails, as might be anticipated. For FT fuels, 100% blends show an improvement of 8.9% in shortening the contrail lifetime, and 13.4% in optical depth reduction, with respect to conventional fuel values when compared to those for the 50% maximum limit. On the other hand, differences with respect to the use of HEFA fuel from camelina feedstock in a complete drop-in scenario remain small (less than 0.5% for both lifetime and optical depth), similarly to the case for 50% blends. However, one should not straightforwardly conclude that the usage of both these types of fuels can be deemed identical in terms of environmental benefits. It must be noted that the total environmental impact associated with all other pre-usage stages cannot be discarded, as explored in the review by Cabrera and Sousa [1].

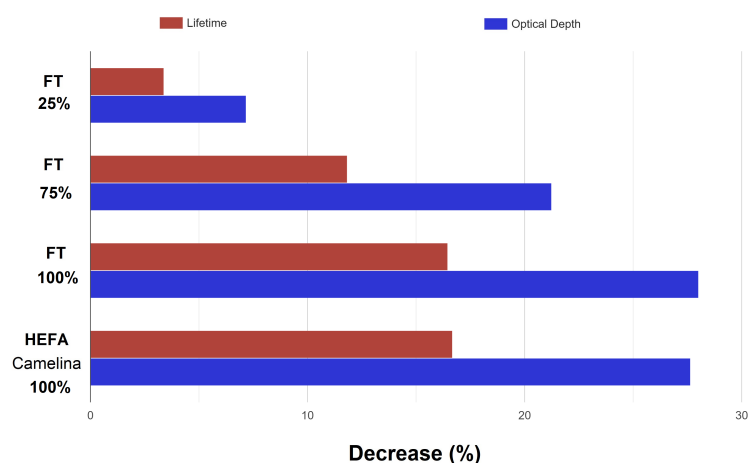


Figure 7. Impact of different blends of FT and HEFA-C fuels in contrail lifetime (red) and optical depth (blue), with respect to simulations for 100% conventional jet fuel.

Irrespective of the present results, the current blending limits for different SAF pathways are determined based on compatibility with current engine requirements and, despite continuous improvements, not all pathways exhibit the same potential to eventually become 100% blend-certified. A possible solution to maximize SAF usage and circumvent

the necessity of conventional fuel blending is to blend multiple alternative fuels in a way that the resulting fuel properties meet all specified requirements for 100% usage since, conceivably, as many blending components as needed could be used to replicate the properties of Jet A/A-1 [22]. However, as more research needs to be conducted regarding the compatibility between different SAFs, this option was not analyzed here.

4.2. Shortcomings of the Original Contrail Model

As previously explained, the original contrail model [16] estimates the early age properties at a stage 1 defined by the wake vortex timescale t_0 . As the LES-based parametrization described in detail in Section 3 provides results for young contrails aged 5 min, the contrail cirrus calculation loop in the original modeling (corresponding to the process of stages $1 \rightarrow 2$ in Figure 2) must be interrupted to ensure physical compatibility between these distinct approaches. Hence, in order to carry out an in-depth analysis of the shortcomings of the original contrail model in the framework of the LES-based parametrization, a time break-loop condition was added at $t > 5$ min.

Although the inputs for both approaches are obtained from the atmospheric model and the aircraft engine model, the initial state of the LES-based parametrization is defined as occurring after the start of the wake vortex, that is, $t_0 > 0$, as depicted in Figure 8. Despite this discrepancy amounting to only a few seconds in wake vortex formation, with most of the quantities therefore experiencing only negligible variations, others are greatly affected, namely, the relative humidity. If contrail formation has already started, then the SAC was satisfied, and so there is a state of supersaturation of the air with respect to ice in the plume mix. Therefore, to accommodate for these differences, the ice relative humidity RH_i was added as a user input in this analysis so that its effect on the results may be duly accounted for.

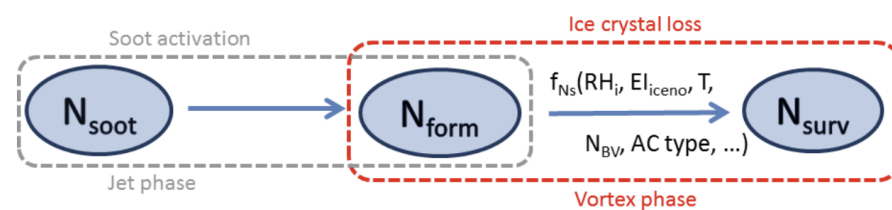


Figure 8. Schematic representation of the process of ice crystal loss between the initial and final stages of the LES-based parametrization, where N_{form} is the number of ice particles at stage 0, and N_{surv} stands for the number of surviving ice crystals at stage 1. The initial state of the original contrail model corresponds to the beginning of the jet phase, where N_{soot} is the number of emitted soot particles from the engine. Adapted from Unterstrasser [18], where *AC type* denotes the aircraft type.

A comparison between the survival factor f_{surv} values produced by the original contrail modeling and the LES-based parametrization for young contrails was performed for a selection of flight route points, as the physical definition of this parameter coincides in both cases. In contrast, the contrail depth values cannot be adequately related between these approaches because of the fundamental differences established in the definition of the latter parameter. Namely, the definition adopted by the LES-based parametrization (Cf. Section 3) incorporates effects related to ice crystal loss via the length scales z_{atm} and z_{emit} . The original contrail model (Cf. Section 2), on the other hand, follows a more simplified viewpoint that does not take into account these effects, thus precluding any relevant conclusions regarding the confidence level in the original modeling from a direct comparison of the contrail depths. However, as noted before in Section 3.5, moderate variations in the contrail depth do not lead to major differences in subsequent contrail evolution calculations.

In the present analysis, the best ice survival factor accordance between the two approaches, corresponding to average deviations of the order of $\pm 10\%$, is found for supersaturation values ranging from 18 to 25%, as shown in Figure 9. For high (up to 40%) supersaturation and close to saturation ($< 10\%$) scenarios, the deviation between survival

factor values increases significantly. Nevertheless, since the routes considered in this analysis mostly represent heavy-traffic flight paths and as such are chosen so as to avoid extreme atmospheric conditions, typical contrail properties should emerge. Hence, moderate levels of supersaturation, say around 20%, are still those most likely to be encountered at mid-latitude routes [23].

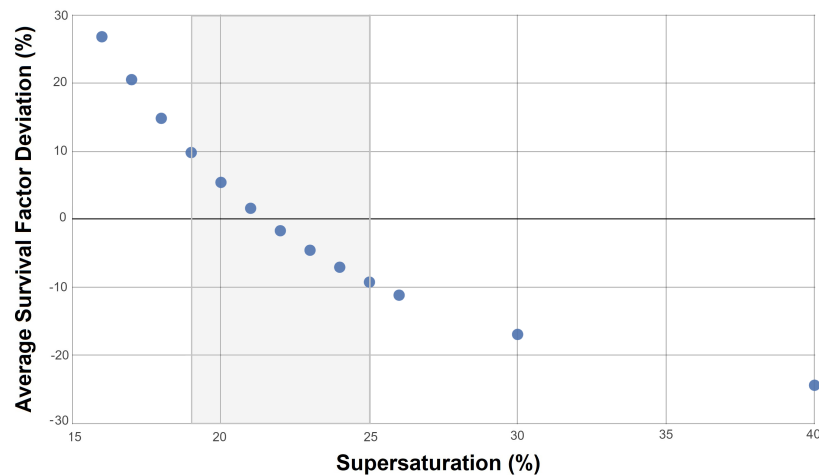


Figure 9. Average deviation of survival factor f_{surv} values from the LES-based parametrization with respect to those in the original contrail model for a range of user-input supersaturation initial states.

5. Conclusions

As the fight against climate change intensifies, the aviation industry must accelerate efforts to mitigate its consequences on the Earth's climate. Such efforts include investing in fundamental research to allow a better understanding of the impact of different pollutants. The detrimental impact of contrails is undeniable, and yet, despite its major contribution to the sector's RF, the phenomena regarding contrail formation and persistence are still not adequately understood. This lacuna in knowledge affects confidence in research conducted with the objective of accurately assessing the impact of climate mitigation measures. To that extent, the use of an LES-based parametrization enables the quantification and reduction of uncertainties in standard contrail models. Specifically, the coupling of a larger-scale model with an LES-based parametrization accounting for processes strongly affecting the contrail evolution during the vortex phase significantly increases the confidence level of the overall computational predictions.

Moreover, the simulations carried out in this work regarding the use of e-fuels successfully demonstrated their potential in mitigating contrail-related adverse effects. Both the contrail lifetime and optical depth showed significant decreases for the regulated blending limits. Additional simulations also enabled it to be concluded that should maximum blending limits be adjusted or even lifted in the future, the mitigation effects will be substantially larger, thus leading to even greater environmental gains.

Author Contributions: Conceptualization, E.C. and J.M.M.d.S.; methodology, E.C.; investigation, E.C.; resources, E.C. and J.M.M.d.S.; writing—original draft preparation, E.C.; writing—review and editing, E.C. and J.M.M.d.S.; supervision, J.M.M.d.S.; project administration, J.M.M.d.S.; funding acquisition, J.M.M.d.S. All authors have read and agreed to the published version of the manuscript.

Funding: This work was supported by national funding through FCT—Fundação para a Ciência e a Tecnologia, I.P., in the framework of project UIDB/50022/2020.

Data Availability Statement: The original contributions presented in the study are included in the article, further inquiries can be directed to the corresponding author.

Conflicts of Interest: The authors declare no conflicts of interest.

Abbreviations

The following abbreviations are used in this manuscript:

CoCiP	Contrail Cirrus Prediction Model
CTL	Coal-to-Liquid
FT	Fischer–Tropsch
GHG	Greenhouse Gases
GTL	Gas-to-Liquid
HEFA	Hydroprocessed Esters and Fatty Acids
LES	Large Eddy Simulation
PtL	Power-to-Liquid
RF	Radiative Forcing
SAC	Schmidt–Appleman Criterion
SAF	Sustainable Aviation Fuel
SPK	Synthetic Paraffinic Kerosene
TAS	True Air Speed
TRL	Technological Readiness Level

Nomenclature

η	Propulsion efficiency
ϵ	Eddy dissipation rate (J/(kg s))
Γ_0	Initial vortex circulation (m ² /s)
Γ_d	Dry adiabatic lapse rate (K/km)
ν	Water vapor emission (kg/m)
ρ	Density (kg/m ³)
ρ_{emit}	Emitted “concentration” (kg/m ³)
τ	Optical depth
θ	Non-dimensional total temperature
A	Area (m ²)
B	Contrail width (m)
b_0	Initial vortex separation (m)
c_p	Specific heat of air at constant pressure (J/(kg K))
D	Contrail depth (m)
e_{sat}	Partial water pressure at saturation (Pa)
EI_{H_2O}	Water emission index (kg-water/kg-fuel)
EI_{ice}	Ice crystal emission index (kg-fuel ^{−1})
EI_N	Particle number emission index (kg-fuel ^{−1})
f_{surv}	Fraction of surviving ice crystals
G	Mixing line gradient (Pa/k)
g	Gravitational acceleration (m/s ²)
$\hat{H}$	Parametrized depth for a 5 min old contrail (m)
I	Ice mass ratio
m_f	Mass flow of fuel per flight distance (kg/(s m))
m_H	Hydrogen mass ratio in the fuel
M_a	Aircraft mass (kg)
M_H	Molar mass of hydrogen (kg/mol)
M_{H_2O}	Molar mass of water (kg/mol)
N	Number of ice crystals per flight distance (m ^{−1})
N^*	Normalized Brunt–Väisälä frequency
N_{BV}	Brunt–Väisälä frequency (s ^{−1})
$\hat{n}_{mean}$	Parametrized mean ice crystal number concentration (m ^{−3})
P	Pressure (Pa)
Q	Fuel combustion heat (J/kg)
q	Absolute humidity (kg/kg)
R_{air}	Gas constant for air (J/(kg K))
R_{H_2O}	Gas constant for water vapor (J/(kg K))
r	Radius (m)

RH_i	Ambient relative humidity with respect to ice
S	Vertical wind shear (s^{-1})
s_a	Wing span of aircraft (m)
s_i	Ambient supersaturation
T	Temperature (K)
t	Time (s)
U	Ambient relative humidity
$\dot{W}$	Estimated horizontal spreading rate of the contrail (m/s)
$\hat{W}$	Parametrized width for a 5 min old contrail (m)
w_0	Initial descent speed of wake vortex (m/s)
z	Altitude (m)
z_Δ	Balance length scale (m)
z_{atm}	Supersaturation length scale (m)
z_{desc}	Vortex length scale (m)
z_{emit}	Emission length scale (m)

Appendix A. Fuel Properties

Table A1. Required fuel properties from the computational model's database.

Properties	Symbol	Jet A-1	GTL	CTL	HEFA R-8	HEFA C
Net Heat of Combustion, MJ/kg	Q	43.2	44.2	44	44.1	44.3
Density at 15 °C, kg/m	ρ_{15}	802	737	762	763	751
Viscosity at −20 °C, kg/m	ν	3.91	2.6	3.6	5.5	3.3
Surface Tension at 25 °C, mm ² /s	σ	27.4	23.8	25.2	25.8	24.8
Initial Boiling Point, °C	$T_{b,i}$	151	146	149	156	151
10% Recovered, °C	$T_{10\%}$	169	162	166	178	161
50% Recovered, °C	$T_{50\%}$	199	169	180	218	182
90% Recovered, °C	$T_{90\%}$	243	184	208	263	237
Final Boiling Point, °C	$T_{b,f}$	262	198	228	274	259
Hydrogen Content, % weight	$H\%$	13.87	15.6	15.1	15.3	15.4
Hydrogen to Carbon Molar Ratio	h/c	1.919	2.203	2.119	2.153	2.169
Molecular Weight, kg/kmol	M	160.5	146	156	177	160
Critical Temperature, QC	T_c	392.3	346.5	364.5	394.2	367.1
Critical Pressure, bar	P_c	21.88	20.95	21.27	17.8	19.79
Smoke Point, mm %	SP	23	40	40	40	50

Appendix B. LES Approach

The 3-D LES simulations of the contrail vortex phase were performed using the EULAG-LCM model system developed at DLR. The EULAG base model is utilized to solve the momentum and energy equations by employing the Multidimensional Positive Definite Advection Transport Algorithm (MPDATA). The EULAG-LCM model is then completed by coupling EULAG to a microphysics module with Lagrangian ice particle tracking [19]. The new explicit aerosol and ice microphysics model allows for a complete definition and accurate simulations of the ice microphysical processes related to cirrus formation.

Following flow field initialization, turbulent fields are created in runs for a defined stratification and eddy dissipation rate, and then superimposed with a counter-rotating vortex pair. For the wake vortex simulations, a variant of a nonlinear predictor scheme is employed in order to minimize the computational burden.

The LES approach leading to the simulations that are the basis for the parametrization used in this work involves lengthy formulations, which are beyond the scope of the present

study. Hence, for more specific details regarding the LES simulations, the reader is referred to Unterstrasser [19], and Sölch and Kärcher [24].

References

1. Cabrera, E.; Sousa, J.M.M. Use of Sustainable Fuels in Aviation—A Review. *Energies* **2022**, *15*, 2440. [CrossRef]
2. AIN. GE Honda Aero Continues Neat SAF Tests. Available online: <https://www.ainonline.com/aviation-news/business-aviation/2023-10-20/ge-honda-aero-continues-neat-saf-tests> (accessed on 15 November 2023).
3. AIN. Gulfstream Completes First Transatlantic Flight on 100% SAF. Available online: <https://www.ainonline.com/aviation-news/business-aviation/2023-11-20/gulfstream-completes-first-transatlantic-flight-100-saf> (accessed on 27 November 2023).
4. Schumann, U. On conditions for contrail formation from aircraft exhausts. *Meteorol. Z.* **1996**, *5*, 4–23. [CrossRef]
5. Gierens, K.; Spichtinger, P.; Schumann, U. Ice Supersaturation. In *Atmospheric Physics: Background–Methods–Trends*; Springer: Berlin/Heidelberg, Germany, 2012; pp. 135–150.
6. Schumann, U. Formation, properties and climatic effects of contrails. *Comptes Rendus Phys.* **2005**, *6*, 549–565. [CrossRef]
7. Burkhardt, U.; Kärcher, B. Global radiative forcing from contrail cirrus. *Nat. Clim. Chang.* **2011**, *1*, 54–58. [CrossRef]
8. Lee, D.S.; Fahey, D.W.; Skowron, A.; Allen, M.R.; Burkhardt, U.; Chen, Q.; Doherty, S.J.; Freeman, S.; Forster, P.M.; Fuglestedt, J.; et al. The contribution of global aviation to anthropogenic climate forcing for 2000 to 2018. *Atmos. Environ.* **2021**, *244*, 117834. [CrossRef] [PubMed]
9. Schumann, U. A contrail cirrus prediction model. *Geosci. Model Dev.* **2012**, *5*, 543–580. [CrossRef]
10. Schumann, U.; Baumann, R.; Baumgardner, D.; Bedka, S.T.; Duda, D.P.; Freudenthaler, V.; Gayet, J.-F.; Heymsfield, A.J.; Minnis, P.; Quante, M.; et al. Properties of individual contrails: A compilation of observations and some comparisons. *Atmos. Chem. Phys.* **2017**, *17*, 403–438. [CrossRef]
11. Wong, H.-W.; Yelvington, P.E.; Timko, M.T.; Onasch, T.B.; Miake-Lye, R.C.; Zhang, J.; Waitz, I.A. Microphysical Modeling of Ground-Level Aircraft-Emitted Aerosol Formation: Roles of Sulfur-Containing Species. *J. Propuls. Power* **2008**, *24*, 590–602. [CrossRef]
12. Paoli, R.; Shariff, K. Contrail Modeling and Simulation. *Annu. Rev. Fluid Mech.* **2016**, *48*, 393–427. [CrossRef]
13. Kärcher, B.; Yu, F. Role of aircraft soot emissions in contrail formation. *Geophys. Res. Lett.* **2009**, *36*, 1–5. [CrossRef]
14. Kärcher, B. Formation and radiative forcing of contrail cirrus. *Nat. Commun.* **2018**, *9*, 1824. [CrossRef] [PubMed]
15. Grimonprez, S.; Wu, J.; Faccineto, A.; Gosselin, S.; Riber, E.; Cuenot, B.; Cazaunau, M.; Pangui, E.; Formenti, P.; Doussin, J.-F.; et al. Hydrophilic properties of soot particles exposed to OH radicals: A possible new mechanism involved in the contrail formation. *Proc. Combust. Inst.* **2021**, *38*, 6441–6450. [CrossRef]
16. Narciso, M.; Sousa, J.M.M. Influence of Sustainable Aviation Fuels on the Formation of Contrails and Their Properties. *Energies* **2021**, *14*, 5557. [CrossRef]
17. Gaspar, R.M.P.; Sousa, J.M.M. Impact of alternative fuels on the operational and environmental performance of a small turbofan engine. *Energy Convers. Manag.* **2016**, *130*, 81–90. [CrossRef]
18. Unterstrasser, S. Properties of young contrails—A parametrisation based on large-eddy simulations. *Atmos. Chem. Phys.* **2016**, *16*, 2059–2082. [CrossRef]
19. Unterstrasser, S. Large-eddy simulation study of contrail microphysics and geometry during the vortex phase and consequences on contrail-to-cirrus transition. *J. Geophys. Res. Atmos.* **2014**, *119*, 7537–7555. [CrossRef]
20. Unterstrasser, S.; Görsch, N. Aircraft-type dependency of contrail evolution. *J. Geophys. Res. Atmos.* **2014**, *119*, 14015–14027.
21. Kärcher, B.; Burkhardt, U.; Unterstrasser, S.; Minnis, P. Factors controlling contrail cirrus optical depth. *Atmos. Chem. Phys.* **2009**, *9*, 6229–6254. [CrossRef]
22. Kramer, S.; Andac, G.; Heyne, J.; Ellsworth, J.; Herzig, P.; Lewis, K. Perspectives on Fully Synthesized Sustainable Aviation Fuels: Direction and Opportunities. *Front. Energy Res.* **2022**, *9*, 782823. [CrossRef]
23. Dekoutsidis, G.; Groß, S.; Wirth, M.; Krämer, M.; Rolf, C. Characteristics of supersaturation in mid-latitude cirrus clouds and their adjacent cloud-free air. *Atmos. Chem. Phys. Discuss.* **2023**, *23*, 3103–3117. [CrossRef]
24. Sölch, I.; Kärcher, B. A large-eddy model for cirrus clouds with explicit aerosol and ice microphysics and Lagrangian ice particle tracking. *Q. J. R. Meteorol. Soc.* **2010**, *136*, 2074–2093. [CrossRef]

Disclaimer/Publisher’s Note: The statements, opinions and data contained in all publications are solely those of the individual author(s) and contributor(s) and not of MDPI and/or the editor(s). MDPI and/or the editor(s) disclaim responsibility for any injury to people or property resulting from any ideas, methods, instructions or products referred to in the content.