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Abstract: This article provides a review of the role of stochastic approaches, in particular Monte Carlo calculations, in the study of aviation-induced contrails at different characteristic lengths, ranging from micrometers to the planetary scale. Pioneered in the 1960s by Bird, Direct Simulation Monte Carlo has for long time been considered unfeasible in extended dispersed-phase systems as clouds. Due to the impressive increase in computational power, Lagrangian Monte Carlo approaches are currently available, even for studying cloud formation and evolution. Some aspects of these new approaches are reviewed after a detailed introduction to the topic of aircraft-induced cloudiness. The role of Monte Carlo approaches in reducing the different source of uncertainty about the contribution of aviation contrails to climate change is introduced. Perspectives on their role in future experimental and theoretical studies are discussed throughout the paper.

Keywords: non-CO<sub>2</sub> emissions; dispersed phase systems; contrails; Monte Carlo simulations



**Citation:** Bianco, D.; Marenna, E.; Loffredo, F.; Quarto, M.; Di Vito, V.; Federico, L. Monte Carlo Simulations in Aviation Contrail Study: A Review. *Appl. Sci.* **2022**, *12*, 5885. https:// doi.org/10.3390/app12125885

Academic Editors: Johannes Schwank and Marco Genovese

Received: 13 February 2022 Accepted: 7 June 2022 Published: 9 June 2022

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# 1. Introduction

Since the second half of the last century, condensation trails (a word usually contracted in *contrails*) originating in aviation have been recognized and studied for their environmental impact [1,2], which could be comparable to that ascribed to CO<sub>2</sub>, although its quantitative evaluation is affected by large uncertainties concerning their formation, persistence, and variability with the scenarios and meteorological conditions of induced radiative forcing. The stochastic nature of these variables naturally suggests the exploitation of statistical approaches and Monte Carlo simulations. In particular, this article aims to review the role of calculations exploited for studying the formation of contrails and their optical characteristics in connection with their microphysical structure. Nonetheless, some insights will be given regarding the statistical approaches used to evaluate the role of contrails on a global scale.

The formation and eventual persistence of contrails, and clouds in general, is a process developed at a length scale to form droplets and crystals, which in atmospheric physics is usually dubbed the microphysics length scale despite including processes from submicron to centimeter lengths. Water and ice can form in clouds through different processes, including nucleation, condensation, collision-coalescence and collision-breakup, freezing, and melting. The (micro)physics of the forming droplets and ice crystals, colliding and interacting with each other, is complicated by the large number of particles, the distribution of their size, and, for ice crystals, their different shapes. Cloud microphysics, however, drives clouds' evolution and persistence and determines their impacts in terms of radiative forcing, that depend on their interaction with electromagnetic radiation.

In modeling contrails and their evolution towards persistence clouds, two challenges have then to be faced: namely, how to consider the contribution of every particle without needing to simulate each of them individually and how to correctly reproduce the microphysics of the system, including the initial size and shape distribution of droplets and crystals. Traditionally, Eulerian schemes have been employed in the field of climate studies and weather prediction modeling. In the last fifteen years, however, due to the incredible increase in available computational power, Lagrangian parameterization has gained attention within the scientific community. These particle-tracking approaches have the advantage of conceptually reproducing the system, made up of particles with different sizes and shapes that move and interact within the fluid. Beyond the need for dedicated algorithms, reducing the overwhelming amount of computational resources that would be otherwise needed for simulating each particle in a cloud, the critical points that still remain to be addressed concern the physics of the interactions between particles and their initial distribution within a condensation trail, which depends on atmospheric, as well as exhaust chemical and physical, parameters.

Coupling direct observation using airborne and remote-sensing probes in space with laboratory experiments would, in principle, help shed some light on the physics of forming condensation trails and their transition into persistent clouds. However, going beyond the actual state-of-the-art will require advances in laboratory and ground-based equipment as well as a new generation of airborne and space observational devices. In fact, the major challenge in observing both laboratory and natural clouds lays in the difficulty of separating the contributions of the different processes that add to the development of the phenomenon. Here, the role of simulations and statistics becomes crucial, conceptually providing the possibility of performing a deconvolution of the observed results with respect to the different acting processes.

Numerical sampling and statistics are intimately linked to the development of computing machines. Applications in this field were pioneered at the beginning of the contemporary era of electronic computers, in particular, during theoretical fission reaction studies carried out during the Second World War. The name of the statistical sampling method, "Monte Carlo", recovered by John von Neumann and Stan Ulam immediately after the war, comes from a joke by Nicholas Metropolis, in reference to Ulam's passion for card games and some anecdotes on his family [3].

A Monte Carlo simulation is based on the capability of reproducing randomness using an algorithm. This can be accomplished using one of the so-called pseudo-random number generators, a class of algorithms invented by Ulam, that have now reached a high degree of mathematical sophistication [4]. The term "pseudo" accounts for the impossibility of reproducing, with a deterministic algorithm, a real random distribution. However, pseudorandom methods are devised to provide a set of numbers whose distribution is statistically indistinguishable from the realization of a random variable uniformly distributed in [0, 1].

The availability of uniformly distributed numbers allows reproduction of a generic probability density function (pdf) through an inverse transformation. In fact, the uniform distribution U(0,1) can be associated with the cumulative probability function of a generic stochastic variable, X, whose values are bounded between 0 and 1 by definition. A random process characterized by a vector of random variables,  $V = \{X_1, X_2, ..., X_n\}$ , can be then simulated using the inverse transformation approach. If the random variables are independent, this is trivially possible using n-times the pdf generation algorithm. In the case of dependent random components of the vector X, one can start generating the first random variable, X<sub>1</sub>. Knowledge of the conditional pdf,  $f_2(x_2 | x_1)$ , allows the generation of X<sub>2</sub>. The availability of the remaining n-2 conditional pdf can be exploited for simulation of the whole random process.

Monte Carlo approaches have been found to be particularly fruitful when applied to Markov chains, a class of random processes for which the conditional pdf of  $X_{t+s}$  given  $X_u$  with u less than t is the same as the conditional probability given  $X_t$ . Loosely speaking, Markov processes are those random experiments in which the system does not exhibit long-term memory. Physical examples include radiation transport and water/ice cloud formation, which are both found in the formation, persistence, and radiative forcing evaluation of contrails.

The paper is organized as follows: The next section discusses the thermodynamic conditions under which contrails are thought to form and persist and introduces some empirical aspects of the phenomenon. Section 3 tackles the role of Monte Carlo simulations in understanding the physicochemical details of condensation in the engine plume mixing with the atmosphere, from which the properties of individual contrails, in terms of radiative forcing, arise. The subsequent paragraph is dedicated to discussing statistics applied to the evaluation of the planetary effects of contrails: a statistical approach based on remotesensing to their formation is introduced along with some simulations used for assessing the role of local diversion in airway routes with mitigation purposes. A paragraph summarizing the material presented and offering some seminal perspectives on the experimental and computational studies on contrails' microphysical structure closes the paper.

# 2. Contrails in Aviation: Generalities

Contrails form in the mixing process of an expanding engine plume with the surrounding atmosphere. Exhaust from the propulsion system is generally warmer than air and contains a large amount of water vapor. For this reason, contrails have been mainly observed in the highest part of the troposphere when associated with the turbo-fan engine currently employed in medium and long-haul flights. The numerical criterion for evaluating the trail formation probability is dubbed the Schmidt–Appleman Criterion (SAC) [5] after the two scientists who developed pioneering studies in this field in the early phases of modern aviation.

SAC is a global thermodynamic criterion based on the assumption of an adiabatic and water-conserving mix between the exhaust and the surrounding atmosphere, wherein heat and vapor diffuse at the same rate [6]. Under this condition, the state point representing the mixture in a Temperature-Partial vapor pressure graph evolves along a straight line, whose slope is given by

$$G = \frac{P_v - P_{v\infty}}{T - T_{\infty}},\tag{1}$$

where  $P_v = X_{H_2O}P$  represents the water's partial vapor pressure,  $X_{H_2O}$  represents the water's molar fraction, and the underscript  $\infty$  indicates the completely mixed system. For a turbo-fan engine, the *G* parameter in Equation (1) can be expressed as a function of the engine efficiency and the air pressure only [7]:

$$G = \frac{c_p P_{\infty}}{\epsilon} \frac{E I_{H_2 O}}{(1 - \eta)Q'}$$
(2)

in which  $c_p$  stands for the specific heat capacity of air at constant pressure,  $\epsilon = 0.622 = W_{H_2O}/W_{air}$  is the water/air molar mass ratio,  $EI_{H_2O}$  is the water emission index of the engine (whose test bench measurements in a laboratory are required by ICAO regulation for engines with an output rated greater than 26.7 kN [8–10]), Q is the total heat released per mass of fuel, and  $\eta = (\text{thrust} \times \text{distance per mass of fuel})/Q$  is the engine's efficiency in cruise conditions, in such a way that  $(1 - \eta)Q$  represents the heat conveyed by the engine's exhaust.

Once that the mixing line slope is known, it can be employed as a parameter for evaluating contrail formation and persistency. In particular, the threshold temperature for contrail formation can be derived by considering the temperature dependence of the partial pressure of the liquid water–steam phase transition  $P_{satv}^{liq}$ ; that is, the water liquid–vapour pressure saturation curve, described by the Clausius–Clapeyron relation [11,12]

1:-

$$\frac{dP_{sat_v}^{iiq}}{dT} = \frac{1}{T} \frac{Q^{lat}(T)}{\alpha_v - \alpha_l},\tag{3}$$

in which  $Q^{lat}(T)$  is the latent heat of evaporation/condensation, and  $\alpha_v$ ,  $\alpha_l$ , are the specific volume of the vapour and liquid phases, respectively. The threshold temperature,  $T_{th'}^*$  for

condensation is trivially the temperature of the tangent point between the mixing line and the curve in Equation (5):

$$\left. \frac{dP_{sat_v}^{liq}}{dT} \right|_{T_{th}^*} = G.$$
(4)

Threshold temperature determination is graphically described in [6]. There, the condition for contrail formation and persistence is synthetically described: if the mixing line intersects the liquid–vapour saturation curve, the contrail is formed.

The crucial questions for evaluating the environmental impact regards how long they persist in the atmosphere and which physicochemical processes are involved into their transmutation in clouds [13]. Sight cannot distinguish contrail from natural cirrus clouds [14]. For the condensed trail to persist, a second condition has to occur, which is that the atmosphere–exhaust mixing result has to lay above the vapour–ice saturation curve. Ice formation, however, and the following persistence of the contrail, also depend on the presence in the plume of soot and sulfuric acid aerosol, originated in the engine, acting as condensation nuclei for the microscopical water drops. It has been found that, at the microscopical level, contrail cirrus clouds are distinguishable from their natural counterparts, as evidenced by the bimodal spectrum in the particle dimension distribution occurring when both are observed [15,16]. In ice clouds, sedimentation, deposition growth, and, in particular, radiative properties, depend on the ice crystals' habits [17], and references therein, since ice aggregates scatter shortwave radiation more strongly than pure ice crystals of the same mass.

In accordance with the considerations above, the experimental observation of microphysical properties could be crucial for quantitatively determining the environmental impact, since their statistical distributions could, in principle, be used for developing a Monte Carlo calculation (or eventually as a benchmark for ab initio calculations). Unfortunately, experimental findings available in the literature do not build up a statistically significant sample, and there are still aspects of contrail formation and evolution that need to be further observed [6,18]. In the last three decades, however, an increasing number of articles have been devoted to the discussion of measurement campaign focusing on condensation trails. Some authors [18] distinguish between persistent contrails, which maintain their linear shape, and proper cirrus, which originates from contrails. In general, both these phenomena can be classified as Aircraft-Induced Cloudiness (AIC).

# 2.1. Atmospheric Fluid Dynamics and Early-Stage Contrails

Beyond the general thermodynamic framework discussed above, the actual process that leads to contrail formation and eventual persistence is driven by a complex aerodynamic trade-off involving turbulence generated during the aircraft's passage, the engine's exhaust, and the resulting down- and upward streams of moist air. In fact, for modeling purposes, the aircraft wake–engine plume dynamics are generally divided into four phases, characterized by different time scales:

- 1. (up to  $t \sim 10$  s, *jet regime*) The counter-rotating vortices initiated by the aircraft wings trap the engine plume, whose water vapour content is higher than the atmospheric counterpart, colder, and more rarefied. The water vapour from the plume tends to condensate into water droplets or ice, which are transported by the wake vortices. Condensation is eventually enhanced by the soot released by engines within the plume.
- 2. (up to  $t \sim 100$  s, *vortex regime*) Vortices descend downwards in the atmosphere, creating a secondary wake in the opposite direction. A part of the condensed water vapour is trapped and transported upwards.
- 3. (up to  $t \sim 1000$  s, *dissipation regime*) While primary and secondary vortices dissipate, the condensed phases of water are released into the atmosphere: the contrail has been formed.

4. (a few hours after the emission, *diffusion regime*) The new condensation trail diffuses within the atmosphere, completely mixing with it within a few hours.

In the first decade of the current millennium, several studies have been developed on this subject. Few research groups have tackled the problem of producing high-fidelity simulations that describe the details of contrail formation from an aerodynamics point of view. In [19], in particular, the coupling of large eddy simulations (LES) with a Lagrangian particle-tracking approach, a class of methods described below, allowed one of the first studies concerning the sensitivity to atmospheric parameters of contrail formation. The work has been extended in [20], with the inclusion of other parameters, notably the considered aircraft linear dimensions.

# 2.2. Experimental Campaigns and Microscophysical Characterization of Contrails

Microphysical parameters identified in experimental campaigns typically include the ice particles' effective radii, size distribution (PSD), shape, and number concentration (Nt) [21]. As natural clouds, a contrail cirrus is often characterized in terms of ice water content (IWC), that is, the cloud's total mass of ice particles, usually expressed as the percentage of total moist air mass, which is computed by adding dry air mass and ice, liquid water, and steam total mass. The total ice content of a cloud is usually employed in models for parameterizing clouds' optical properties and is then an interesting quantity for estimating the radiative forcing induced by cloudiness.

The first studies on the microphysical characteristics of contrail cirrus parameters date back to the 1970s [22–24]. Early campaigns were limited to measuring ice crystals with linear dimensions above 20  $\mu$ m [25–27], even if the existence of a large number of ice particles with a mean diameter below 30  $\mu$ m [28–31] had been demonstrated. The impact of small particles on the radiative transfer through the cirrus is not negligible because particle size is related to the wavelength of the radiation they scatter, and, below 20  $\mu$ m, one finds the thermal infrared region (8 – 15  $\mu$ m), which includes the black-body radiation of an object at room temperature.

An initial breakthrough in terms of small-particles' observation capabilities was achieved in the second half of the 1990s with the Sulfur 4, Sulfur 5, Contrail, and Aerocontrail airborne experiments [5,32], which were performed with the DLR Dassault Falcon 20-E research aircraft and the NASA SUCCESS (Subsonic Aircraft: Contrail and Cloud Effects Special Study) missions [33–54], employing the DC-8 aircraft of the Armstrong (Dryden) Flight Research Center (AFRC). Both of these airplanes (which are still in operation) are multipurpose flying research laboratories equipped with several different instruments. In particular, at the time of the experiments mentioned above, both mounted a number of systems for airborne particles' size and shape measurements, mainly 1D (the Forward Scattering Spectrometer Probe, FSSP [55,56], for size determination) and 2D optical devices (2D optical arrays, used for particles' size measurements and shape recording, which has evolved considerably over time [57–61]).

Optical probes are still the gold standard for instruments for cloud microphysical parameter measurements and have been exploited in other experimental campaigns after those mentioned above, notably the CONCERT (CONtrail and Cirrus ExperRimenT) [62] and ML-CIRRUS [63] experiments carried out by DLR. In particular, CONCERT allowed the collection of new data on contrail formation beyond commercial flights with the innovative measurements of optical properties' distribution within the mixing fluids [62] and of the vertical particle concentration [64], whereas ML-CIRRUS provided one of the first integrated measures on natural and contrail-induced cirrus employing a HALE (high-altitude long dndurance) airplane.

An analysis of the entire set of experimental data measured with airborne probes, complemented with remote sensing [65] and (remote) lidar acquisition [66,67], is out of the scope of this review and has been the subject of some dedicated works [68,69]. What can be observed here is that strong evidence exists in contrails confirming the presence of a large initial number ( $N_t \sim 10^4 - 10^5$  cm<sup>-3</sup>) of very small (mean of PSD < 1 µm) ice particles

with a non-linear evolution in time of particle size and number, respectively decreasing by about four orders of magnitude ( $N_t \sim 10 \text{ cm}^{-3}$ ) and increasing up to a mean size of 10 µm during a one-hour time evolution. The natural logarithm of the IWC, conversely, has been found to evolve linearly against temperature, as reported in [21].

Given the limited amount of experimental data available, in forthcoming years there will still need to be improvements in laboratory and model studies for more accurate reproduction of the microphysical properties of the mixing fluid to correctly address the role of the large number of small ice crystals (mean PSD  $\sim 1-1000$  nm), which evolve towards a limited amount of larger particles through aggregation, nucleation, and sublimation loss phenomena.

In particular, the observation of contrails forming beyond an innovative propulsion system, such as a hydrogen fuel cell, may benefit of the exploitation of large test facilities, such as the Icing Wind Tunnel (IWT) at CIRA [70]. Originally built for studying ice formation processes on aerodynamical surfaces, IWT and similar wind tunnels can easily house a fuel cell and its exhaust within a test chamber, an experimental setup not easily achievable with a full-size turbo-fan engine. In Figure 1, taken from [70], an aerial overview of the CIRA facility is shown along with a picture of the main test section.



Figure 1. (a) Icing Wind Tunnel aerial view; (b) IWT main test section. Taken from [70].

#### 3. Monte Carlo Simulations for Contrails Formation and Evolution

Contrails' formation and transformation into cirrus is believed to be linked to the mutual thermodynamic state, the water-vapor content of the atmosphere, and the engine exhaust, as discussed above. This section is devoted to analyzing the role of computational, stochastic approaches in simulating the formation of the microscopical ice aggregates that characterize a trail's evolution into a persistent cirrus.

From a chemical point of view, a contrail originating in the exhaust–atmosphere mixing is a dispersed phase system, containing aerosols and solid particles in a continuum phase. In practice, simulating the origin and evolution of a contrail is strongly linked to the capabilities of computationally reproducing the lifecycle of an (ice) cloud. Historically, clouds were first simulated using Eulerian, density-based approaches [71–76] with equations that described the time evolution of bulk properties, including mass (one-moment scheme), number concentration (two-moments scheme), and, recently, radar-reflectivity (three-moments scheme). These models are still at the foundation of planetary scale and climate simulations [77–94], given their relatively low computational complexity, which is suitable for insertion into codes with a high spatial resolution with respect to their length scale, which usually covers the entire planet.

The starting point for a theoretical study of contrail formation is the stochastic collection equation (SCE) (see, for examples, the introduction given in [79] and references therein), which describes the time evolution of a system of particles (representing droplets, ice crystals, or both) colliding and subsequently forming a larger structure (*coalescence*):

$$\frac{\partial n(v,t)}{\partial t} = \frac{1}{2} \int_{0}^{v} n(v-v',t)n(v',t)K(v-v',v)dv' - n(v,t) \int_{0}^{\infty} n(v',t)K(v,v')dv' = I_1 - I_2,$$
(5)

where n(v) is the number density function of the particle with volume v (equivalent formulation in terms of mass is possible), and K(v, v') is the coalescence kernel of particles with volumes v and v'. In Equation (5),  $I_1$  and  $I_2$  refer to the integral source and sink term for the coalescence process. Moment methods, in fact, provide a solution to the SCE by modeling each integral term as a whole.

Beyond the simplistic early studies on clouds, Equation (5) has been exploited since the 1970s in a class of methods called bin approaches [81,82], which are based on the discretization of the particle spectra. The discretized form of Equation (5) reads as:

$$\frac{\partial N_i(t)}{\partial t} = \sum_{j,k}^{j \ge k} (1 - \frac{1}{2} \,\delta_{j,k}) \, K_{j,k} N_j(t) N_k(t) - N_i(t) \, \sum_k K_{i,k} N_k(t), \tag{6}$$

where  $N_i(t) = \int_{v_i}^{v_i+1} n(v,t) dv$  is the number of particles in the *i*-th bin, and  $K_{j,k}$  is the discretized form of the coalescence kernel. In fact, this wide class of computational methods is still Eulerian in nature because the concept of a particle is not introduced except for populating the different bins in which the spectrum has been divided. Nonetheless, bin models allow calculation to a good approximation of the development in time of the particle concentrations, which usually exploits a finite difference approach for describing the time variations of Equation (6). Bin models were extremely appealing at the beginning of the electronic computer era since they were capable of producing a reasonable time-evolution calculation even with a limited number of bins. Their accuracy, however, is strongly linked to the chosen number of bins, whose increase is linked quadratically to the complexity of the problem to be solved. The result, then, is too heavy for both weather prediction, when employing a limited number of bins, and for detailed studies at cloud-resolving scales, when the number of volume (mass) intervals is increased.

Interesting approaches, based on a Lagrangian Monte Carlo simulation of a single or, more often, a bunch of particles at one time, was developed in the last fifteen years by researchers from different backgrounds who proposed similar strategies in different applied contexts [83–92]. Lagrangian approaches to aerosol/cloud microphysics, conceptually deriving from Direct Simulation Monte Carlo (DSMC, [93]), have the intriguing feature of allowing, at least in principle, single particle tracking, a fundamental step for comprehension of the fluid-dynamical condition for the formation and persistence of contrails and cloudiness in general. In order to limit the demand of computational power, these models usually introduce an ideal object, referred to as a super particle, computational particle, or simulation particle, representing the bunch of particles mentioned above. Lagrangian particles represent a given number (called weighting factor or multiplicity) of real particles characterized by the same volume (mass). Particles belonging to the same collective representation are supposed, within the approximation introduced, to occupy the same position in the grid, and move with the transport equation associated with the fluid motion.

Monte Carlo simulations are still a niche in cloud microphysics research and have been applied to the study of aircraft-induced contrails in a relatively small number of papers. In [91], in particular, the technical details of a Lagrangian code dedicated to the microphysical aspects of ice cloudiness, EULAG-LCM, are described. It was pointed out, there, that the number of computational particles to be employed for statistical convergence changes with the microphysical process considered and that the ice nucleation is the most demanding one. The code developed was used in [92] to study the differences in the formation of contrails for different aircrafts and how they are linked with the transformation into cirrus clouds. Six different aircraft, ranging from a small regional airplane to the largest A380, were considered, and the peculiarities of their vortices and engine exhausts were described with large eddy simulations in order to determine the microphysical differences of the forming cloud. Larger vehicles were associated with an enhanced number of ice crystals, that was reduced, however, by the associated stronger vortices in the few seconds after plume emission. Nonetheless, the cirrus associated with the AIC of the largest aircraft were not as persistent as those of the regional one, which lasted from 1.5 to 2.5 times less. Assessments such as the one described above represent the future of the discipline and are crucial to evaluating the non- $CO_2$  environmental impact of the new configurations and propulsion systems that will be introduced in aviation towards 2050. In this perspective, these kinds of Monte Carlo calculations will become the gold standard for cloud-resolving simulations and, probably, even for larger length scales. The capability of correctly reproducing the role of aircraft-induced cloudiness in the climate equilibrium of the planet is still a challenging task, as described below, and could benefit from the introduction of high-fidelity approaches into models of the atmosphere dynamics.

# 4. Statistical Approaches and Monte Carlo Simulations for Global Evaluation of Contrail Effect

At the end of the last century, the Intergovernmental Panel for Climate Change (IPCC) promoted an assessment of aviation's contribution to global warming [94], from which a series of uncertainties in the quantitative evaluation arise. Apart from the microphysical characteristics of cirrus that directly come from air transport, the second source of uncertainties can be found in the dynamics of the aircraft-generated cloudiness. An interesting statistical approach to derive the actual contrail lifetimes, based on geostationary satellite observations, was presented in [95] and is briefly reviewed here.

The method in [95] exploits the availability of an automatic contrail tracking algorithm, capable of extracting a trail's evolution information from remote-sensing images. Two examples of such algorithms are available in [96,97]. Contrail tracking is limited by satellite spatial and wave-length resolutions, which affect the initial detection of a trail and its disappearance, respectively. In fact, both determine a time bias between (i) a contrail's first observation and its actual genesis (pre-detection time) and (ii) contrail undetectability in images and its radiative forcing contribution becoming negligible.

Analyzing the ACTA (automatic contrail tracking algorithm) database [98], the authors of [95] found that the contrails' survival function,

2

$$S(t) = \frac{n(t)}{n(0)},$$
 (7)

with n(t) representing the number of contrails still observable in satellite images at time t, is well fitted by a generalized (Weibull [99]) exponential law

$$S(t) = \exp\left[\left(-\lambda \ t\right)^k\right].$$
(8)

Apart from reproducing the decreasing number of observed contrails, Equation (8) can be used for estimating their unobserved persistency times. Consider that the percentage of contrails observed at time  $T = \tau$  survives for a further time lapse  $\delta$  given by

$$\frac{n(\tau+\delta)}{n(\tau)} = \frac{S(\tau+\delta)}{S(\tau)} = P \ (T = \tau + \delta \mid T = \tau), \tag{9}$$

from which the conditional probability density function

$$f(\delta|\tau) = -\frac{d}{d\delta} \left( \frac{S(\tau+\delta)}{dS(\tau)} \right),\tag{10}$$

can be derived. Via exploitation of a regression to the database with Equation (8), and via a rule-of-thumb that suggests a slow ( $\sim$  5 km/h), constant contrail spreading speed—from which a Gaussian-like distribution of their width before detection can be postulated—it has been possible to build up a Monte Carlo calculation for extracting the actual life-time of observed contrails. In fact, three random, uniformly distributed numbers are generated and used to sample the three available distributions, that is, the time in which a contrail is revealed by satellite, the pre-detection time, and the time a contrail survives after its disappearance from satellite.

The example discussed above shows once again the preeminent role of statistics and a Monte Carlo simulation in the study of contrails in aviation, even on a different length scale. Apart from microphysical characteristics and the actual lifetime of aircraftinduced cloudiness, another source of the uncertainty discussed in the IPCC report lies in the contrails' contribution to climate. As a matter of fact, assessing mitigation options for contrail-induced climate effects, in terms of the adoption of both technology and local diversion strategies despite large uncertainties regarding atmospheric processes, is currently a theme of scientific debate [100–102], involving different future air-traffic scenarios. A detailed, fully-coupled calculation evaluating the effect of aviation-induced cloudiness on a global scale presents the same complexity of the quantitative estimation of the whole radiative forcing due to natural stratiform clouds. As mentioned in the previous section, computational models that treat dynamics of the whole atmosphere have a coarser spatial resolution with respect to the characteristic dimensions of clouds, either natural or artificially induced, which are then not resolved in these large-scale calculations.

General circulation models (GCM) cope with this sub-resolution issue by introducing the concept of fractional cloud coverage, which originates from subgrid scale variability in humidity and/or in temperature (on which the water saturation mixing ratio,  $r_s$ , depends). In advanced GCM, as in ECHAM5 [103] (ECWMF model, Hamburg version), which simulates the atmosphere and its chemical and physical processes, statistical approaches offer a solution for calculating fractional cloud coverage [104]. Focusing on the fluctuations in total water content, only, this class of methods writes the percentage *C* of cloud coverage in each model cell as

$$C = \int_{r_s}^{\infty} G(r_t) dr_t, \tag{11}$$

where  $G(r_t)$  represents the probability density function of the total water mixing ratio in the cell considered, including vapor, liquid, and ice phases. Tompkins [104] used a cloud-resolving model for determining the best a priori distribution, the Beta function with boundaries *a*, *b* and parameters *p*, *q* as

$$G(t) = \frac{1}{B(p,q)} \frac{(t-a)^{p-1}(b-t)^{q-1}}{(b-a)^{p+q-1}},$$
(12)

where  $B(p,q) = \frac{\Gamma(p)\Gamma(q)}{\Gamma(p+q)}$  is a normalizing combination of gamma functions.

An example of a fully-coupled calculation of AIC effects with a GCM model was provided in [105,106], where a study of temporal dynamics and radiative forcing of contrails' cirrus has been performed with a modified version of the GCM ECHAM5, the ECHAM5-CCMod. This code, however, was based on a different class of methods for tackling the fractional cloud coverage, dubbed relative humidity schemes [107], and conceived of the use of Eulerian moments methods in describing the subgrid cloud dynamics. ECHAM5-CCMod was used in Monte Carlo calculations for assessing the effects of atmospheric uncertainties on mitigation options for air traffic scenarios [100], even though in association with the climate-response model AirClim [108], employed for reducing the complexity of the model calculation and making the Monte Carlo simulation feasible. In 2020, a calculation employing the most advanced version of ECHAM5-CCMod [109] evidenced a consistent reduction of the estimated radiative forcing due to AIC. Whether the radiative effect of contrails is really less intense than that estimated up to now, or whether such detailed simulations need to properly include other elements, as the coupling with oceans, still needs to be understood in order to be quantitatively reliable on an absolute scale.

Different approaches, based on simpler evaluation of contrails' effects, though uncoupled with the atmospheric system, have been developed. An example is the CERM (contrail evolution and radiation model) [110], an evolution of the CoCiP (contrail cirrus prediction model) [111]. Beyond evaluation of the accuracy of these simplified calculations with respect to detailed GCM models, the questions to be addressed concern the magnitude of the coupling of cloudiness, in general, and contrail cirrus, in particular, with the other constituents of the planetary climate system. This general question goes well beyond the scope of this review; from one point of view, however, it could be argued that the evolution of the Lagrangian Monte Carlo-based calculations will help to shed some light on physical details and, hopefully, on coupling phenomena in planetary-scale models of the atmosphere.

#### 5. Challenges and Perspectives

The study of aircraft-induced cloudiness still suffers of some fundamental knowledge gaps, particularly as they relate to microphysical aspects of the formation process. The understanding of contrail formation and persistence, that in last decades has been partly elucidated through laboratory experimentation, field observations, and theoretical and simulation developments, continues to be affected by large uncertainties related to the ice-phase microphysics, including nucleation, aggregation, and direct growth from vapor phase, which are represented in Equation (5) by the interaction kernels K(v, v'). The source of the poor understanding of these processes largely lays in the complication related to particle size distribution and shape, resulting in a low capability of modeling persistency, which is linked to the transformation of the contrail into an ice cirrus.

Nucleation, or the growth of ice crystals upon solid/liquid particles acting as condensation nuclei, is extremely sensitive to onset conditions, including particle surface shape and size, and their morphological, mechanical, and chemical characteristics that represent a source of uncertainty. Ice growth from vapor, on the other hand, is a relatively homogeneous process through which ice crystals of considerable dimensions can be formed in a relatively short amount of time. The model's sensitivity to parameters, which generates large fluctuations in the possible outcome, must found here, in the vapor diffusion and frost processes, which determine the crystal's shape and dimension. In particular, modeling is limited by the unknown, microscopical distribution of vapor upon a growing crystal surface. Heterogeneity of this distribution determines the evolution of crystals with irregular shapes, which is difficult to predict. This uncertainty about the crystal shape, apart from that regarding their size distribution, induces doubts about the role and dynamics of ice–ice collision break-up, whose rate should depend also on partial water vapor pressure and temperature.

Even if Lagrangian particle-tracking schemes have represented a breakthrough in cloud modeling, then, the lack of a clear understanding of the underlying physics represents the first challenge for future developments in this field. Monte Carlo approaches are crucial for quantitatively estimating the impact of a multi-scale, complex process as contrails form in aviation, but much more effort will be devoted to modeling and computation, as well as to airborne and laboratory experiments, for validating and feeding simulations. From this viewpoint, large facilities are a valuable tool that can be used for the experimental measurements of the contrail microphysics, in particular for evaluating the impact of new propulsion systems. Experimental data can then be compared with in-flight observations and simulations, for study of the microphysics of forming contrails.

Beyond the physics, however, particle-tracking schemes are computationally expensive because the number of considered computational particles increases in order to deal with larger, more complex systems and obtain a significant statistic. This is the case when the transport of computational particles between two grids is considered, for instance, in the modeling of the exchange of ice and water vapor between clouds and surrounding atmosphere.

In [112], it was shown that the computational effort can be reduced by introducing condensation nuclei, which circumvents the need to consider advection outside the cloud. In the same article, it was roughly estimated that between 50 and 200 computational particles per grid are needed for properly simulation of a cloud; the range is due to the number of variables considered. However, Lagrangian approaches show a dependence on computational particle initialization that does not always correctly represent the triggering

of real atmospheric processes [113], as is the case with contrails that transition to ice cirrus. Moreover, the statistical variability introduced is expected to be higher than in nature since minority processes are represented with a relatively low number of computational particles [114].

Gaining a deeper understanding of the natural processes that underlie cloud dynamics will have a positive impact on aviation. This benefit to the sector will result in the capability to estimate the impact of circulation and newly-designed aircraft, as well as in understanding how air-traffic management (ATM) principles may be applied to the reduction in aircraft -induced cloudiness production and radiative forcing.

Author Contributions: Literature review, D.B. and E.M.; conceptualization, D.B. and L.F.; writing original draft preparation, D.B. and F.L.; writing—review and editing, E.M., M.Q., V.D.V. and L.F. All authors have read and agreed to the published version of the manuscript.

Funding: This research received no external funding.

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

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