

## Article

# Aircraft emissions, their plume-scale effects, and the nonlinear response of the climate-chemistry system

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## 1. Introduction

Aircraft act as high-altitude emissions vectors, transporting a number of radiatively and chemically active substances across vast regions of the globe. These substances induce a net global warming effect that constitutes 3.5% of the global climate impact due to anthropogenic sources <sup>??</sup>. Two thirds of this impact result from aircraft non-CO<sub>2</sub> emissions, which have a variable impact depending on when and where they are released. This means that avoiding flying in climate-sensitive regions of the atmosphere has the potential to reduce aviation's climate impact significantly <sup>??</sup>. Furthermore, the environmental conditions within the aircraft exhaust plume result in additional chemical and microphysical processing that is not accounted for in global chemistry models. In high-density airspace, such as along the North Atlantic Flight Corridor, aircraft often fly in close proximity, giving rise to the superposition of their plumes and the accumulation of emissions contained within them <sup>??</sup>. It has been shown in <sup>??</sup> that the overlap of four consecutive plumes can influence the nonlinear chemistry and microphysics significantly, leading to saturation effects that can potentially provide a net climate impact reduction.

## 2. Aircraft emissions

Aircraft propulsion systems provide the essential driving force component to enable powerful and efficient flight, however convention has dictated the use of hydrocarbons as the primary source of aviation fuels since the dawn of powered flight. In commercial aviation, responsible for 88% of global aviation fuel usage [1], it is commonplace to use a gas turbine propulsion system operating on kerosene-based jet fuel. Aviation's environmental impact stems from the emission and dispersion of chemically and radiatively active substances, that are generated during the jet fuel combustion process. The combustion of fuel and the generation of emissions is closely coupled with

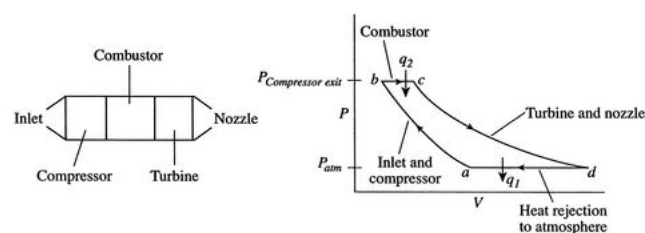
the performance of aircraft, and the forces and dynamics experienced throughout flight. This section explores the coupling between aircraft performance and the generation of aviation-induced climate forcing species, before reviewing the emissions modelling techniques present in the current literature.

### 2.1. The need for propulsion

For an aircraft to achieve and maintain airborne status, it is required that the resistive force due to aerodynamic drag ( $D$ ) and the gravitational force due to aircraft weight ( $W$ ) are sufficiently overcome. The necessary driving force required to counteract drag and maintain forward speed is known as thrust ( $T$ ). The forward speed induces airflow over the aircraft's cambered wings, thus rotating the flow downwards and generating lift ( $L$ ) to counteract the aircraft weight. This constitutes the four fundamental forces of flight [2]. The combination of these forces in varying magnitudes determines the trajectory of an aircraft and its instantaneous velocity and acceleration.

The performance of an aircraft is highly dependent on the optimisation of the four forces, with structural configuration, material utilisation and payload affecting the aircraft's weight, balance and stability, and the aircraft geometry affecting aerodynamics and hence influencing the lift and drag forces acting upon it. The thrust force however, is generated by means of a propulsion system.

Gas turbine engines employ the following four point Brayton thermodynamic cycle (see figure 1): (1) the engine intakes oncoming air flow, (2) the air passes through a compressor to increase its total pressure, (3) the compressed air is mixed with fuel and ignited to cause a combustion reaction and (4) the high temperature, high pressure combustion products are passed through a turbine which extracts some energy from the flow to drive the compressor, before the airflow exits the engine at an accelerated rate, through the exhaust nozzle. The magnitude of thrust generated by the engine is dependent on the mass of air being accelerated, and the difference in velocity of the air mass through the propulsion system.



**Figure 1.** Brayton cycle [1].

### 2.2. The jet fuel combustion process

The widespread usage of kerosene-based jet fuels stems from the need to satisfy strict power-to-weight and safety requirements demanded by commercial aircraft; this is because kerosene has an exceptionally high energy density and wide operating temperature range at a low financial cost, compared to alternative fuel types. The stoichiometric combustion of kerosene with air required to generate

thrust, does however generate a number of additional chemical species which are emitted into the aircraft wake. These aircraft emissions interact with the surrounding atmosphere, perturbing the natural balance of chemistry and contributing to air quality issues and anthropogenic climate change. The mass of emission produced per unit mass of fuel burnt is often referred to as the emission index (EI), measured in grams of emission per kilogram of fuel [g/kg].

The primary products of ideal jet fuel combustion are water vapour ( $\text{H}_2\text{O}$ ), carbon dioxide ( $\text{CO}_2$ ) and, due to fuel impurities, a relatively small amount of sulphur oxides ( $\text{SO}_x$ ). Additionally, a number of secondary combustion products are generated in varying quantities, depending on their formation mechanisms and the nature of the combustion process. The most common of these products are oxides of nitrogen ( $\text{NO}_x = \text{NO} + \text{NO}_2$ ), carbon monoxide (CO), unburnt hydrocarbons (HC), particulate matter (PM) and volatile organic compounds (VOCs) [3].

### 2.3. The generation of aircraft emissions

The generation of primary jet fuel combustion products,  $\text{H}_2\text{O}$ ,  $\text{CO}_2$  and  $\text{SO}_x$ , depend purely on the carbon-hydrogen-sulfur composition of the fuel. This means that for a constant fuel composition, these species have a constant EI throughout the entire range of operating conditions and therefore they are directly coupled to the amount of fuel burnt. The EIs of the secondary combustion products however, are variable depending on the type of aircraft engine, engine operating conditions, and the state of the surrounding atmosphere [4].

The entry of atmospheric nitrogen ( $\text{N}_2$ ) into the high temperature combustion chamber leads to the formation and emission of nitrogen oxides ( $\text{NO}_x = \text{NO} + \text{NO}_2$ ). The levels of  $\text{NO}_x$  emitted from aircraft engines increases with temperature and pressure in the primary combustion zone. Therefore, under the assumption of constant polytropic and combustion efficiencies, the emission index of  $\text{NO}_x$  ( $\text{EINO}_x$ ) can be correlated with aircraft fuel flow [5].

As a result of inefficiencies in the combustion process, products such as carbon monoxide (CO), unburnt hydrocarbons (HC) are formed. The levels of CO and HC on the other hand, are direct products of incomplete combustion, and hence their concentrations are inversely proportional to combustion efficiency. Since combustion efficiency correlates with thrust for sea level static (SLS) conditions (see [5] figure 1.), and fuel flow correlates with thrust, this means that  $\text{EICO}$  and  $\text{EHC}$  decrease with increasing fuel flow.

Trace levels of soot, particulate matter (PM) and volatile organic compounds (VOCs) are also generated, however often in relatively small amounts, and the mechanisms that lead to their production are complex and out of the scope of this review. See [4] for more details on this.

### 2.4. Aircraft emissions modelling

Accurate quantification of aircraft emissions requires the calculation of aircraft performance to estimate the total energy consumed, and hence, the total fuel burnt throughout the duration of a flight. With

123 knowledge of the fuel flow rates experienced throughout flight, flow  
124 rates of aircraft emission species can be deduced based on empirical  
125 engine performance datasets and emissions models.

#### 126 2.4.1. Fuel burn estimation

127 Computational modelling of aircraft performance allows the sim-  
128 ulation of aircraft trajectories and the quantification of forces experi-  
129 enced throughout flight, enabling the approximation of thrust and fuel  
130 flow across all phases of flight. Aircraft performance models that are  
131 prevalent in academia and industry, such as the BADA (Base of Air-  
132 craft Data) method [6] and **PIANO-X** [7], emulate aircraft behaviour  
133 by coupling a database of aircraft-specific performance datasets to  
134 mathematical models to calculate useful flight characteristics, such  
135 as fuel flow and fuel burn, at discrete time steps during flight. This  
136 iterative approach accounts for the time-varying nature of aircraft  
137 properties like mass, speed and heading, thus leading to a more accu-  
138 rate analysis of performance and fuel burn estimation. Further to this,  
139 models such as the Aviation Environment Design Tool (AEDT) [8] ex-  
140 ist, which carry out four-dimensional (4-D) physics-based simulations  
141 of aircraft trajectories at an exceptionally high spatial and temporal  
142 resolution, providing highly accurate predictions of fuel consumption  
143 and localised emissions impacts. Such tools do however come at the  
144 expense of high computational and financial cost, and often require  
145 proprietary data that is unavailable to the public domain.

#### 146 2.4.2. Calculating aircraft emissions at reference conditions

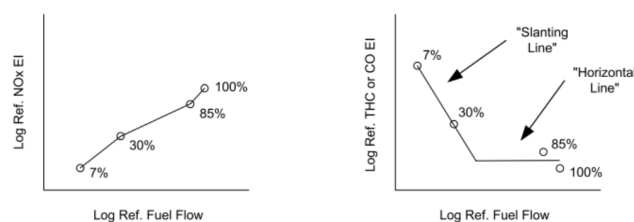
147 As previously alluded to in section 2.3, the primary combustion  
148 products,  $\text{CO}_2$ ,  $\text{H}_2\text{O}$  and  $\text{SO}_x$  have a direct relation to the amount  
149 of fuel burnt and hence a constant EI for a given fuel composition,  
150 whereas the secondary products largely depend on operational and  
151 atmospheric conditions. Emissions of  $\text{NO}_x$ , CO and HC can be cor-  
152 related with fuel flow, meaning estimation of their emission indices  
153 requires knowledge of this relationship across the range of operat-  
154 ing conditions experienced throughout flight. Empirical relationships  
155 have been determined using engine performance datasets, such as the  
156 ICAO engine emissions databank [9]. The ICAO engine emissions  
157 databank was developed for the purpose of engine certification and  
158 compliance with landing and take-off (LTO) cycle emissions standards,  
159 outlined in ICAO Annex 16 Vol. II [10]. Engine test data have been  
160 collected at **SLS**, International Standard Atmosphere (ISA) conditions,  
161 for four reference operating conditions (thrust settings) relevant to the  
162 LTO cycle: take-off (100% thrust), climb out (85%), approach (30%) and  
163 taxi in/out (7%). For every engine at each of the four LTO modes, a ref-  
164 erence fuel flow and corresponding emission index has been derived,  
165 allowing emissions to be estimated for aircraft operating under any of  
166 the four modes, to a reasonable degree of accuracy.

**Table 1:** Example ICAO engine emissions data for Rolls Royce Trent 970-84 engine [9].

	Take-off	Climb out	Approach	Idle
Fuel flow [kg/s]	2.605	2.157	0.720	0.255
EI NO <sub>x</sub> [g/kg]	38.29	29.42	12.09	5.44
EI CO [g/kg]	0.32	0.31	1.16	13.38
EI HC [g/kg]	0.02	0.12	0.08	0.04

#### 2.4.3. Calculating aircraft emissions at non-reference conditions

In reality, aircraft spend the majority of flight outside of the LTO vicinity (above 3,000 ft), and the operating conditions and atmospheric conditions vary considerably. To enable the accurate analysis of aircraft emissions outside of reference conditions, a number of emissions modelling methods have been developed, which apply the necessary adjustments and interpolations to the LTO-limited engine performance datasets, to generate more realistic estimates of aircraft emissions across the whole flight profile. [11] describes a number of methods for calculating aircraft emissions throughout all modes of operation and compares their relative merits. For the primary combustion species, the Fuel Composition method is presented, which determines the EIs from the proportions of carbon, hydrogen and sulfur in the fuel. The set of methods concerning the estimation of EI<sub>NO<sub>x</sub></sub>, EI<sub>CO</sub> and EI<sub>HC</sub> include the ICAO reference method, the Boeing fuel flow method 2 (BFFM2), the DLR fuel flow method and the P3T3 method, in order of increasing fidelity. These methods interpolate engine performance data to determine emissions indices throughout the whole duration of a flight. Furthermore, methods are also presented to account for the remaining key emission species, such as the Derivative Factor method used to approximate the EI values for VOCs and non-methane HC (NMHC) throughout flight and the First Order Approximation method used to estimate PM emission indices. The choice of method generally depends on the emission species to be observed, the compromise between modelling resources and data availability, and the level of accuracy required.

**Figure 2.** Example log-log plots of EI against fuel flow at reference conditions, as prescribed by the BFFM2 [11].

#### 2.4.4. Emissions inventories and integration into large-scale climate models

The estimation of aircraft emissions for a specific flight involves the simulation of aircraft performance across the entire flight profile so as to estimate the fuel flow and engine operating conditions

throughout flight. Knowledge of fuel flows and engine performance characteristics permits the estimation of aircraft emissions, based on the coupling of empirical engine certification data and emissions modelling methods, which interpolate the data to determine the emissions at non-reference conditions. This emissions estimation procedure is commonly carried out on a regional and global scale to determine emissions from a whole range of flights, which are then stored in a so-called emissions inventory [? ]. Aircraft emissions inventories collate data from all flights in the desired range and populate three-dimensional (3-D) latitude-longitude-altitude grid cells (e.g.  $1^\circ \times 1^\circ \times 1000$  ft) with total emissions quantities [12].

Aircraft emissions inventories are utilised to model the atmospheric effects of aviation, using a large-scale chemical transport model (CTM) or a climate-chemistry model (CCM), which captures the chemistry, physics and dynamics of the Earth-atmosphere system. Such models allow one to simulate the perturbation to the state of the atmosphere due to an input of emissions and, in turn, provide quantitative indicators that enable modellers to determine the resultant climate impact (e.g. concentrations of key chemical species, aerosol distribution, cloud processes etc.) [? ]. However, one underlying issue with this conventional approach to aviation climate modelling, is that the use of gridded emissions data inherently assumes the instantaneous dispersion of emissions into the atmosphere [13]. In reality, the emissions released from aircraft are confined to an aircraft exhaust plume, which inhibits mixing with the surrounding atmosphere, for up to a day after emission. Throughout this time, a number of plume-scale processes occur, due to the elevated concentrations of emissions species in the plume, which are neglected in most regional and global aviation climate impact studies [14]. The following section covers the evolution of emissions throughout the lifetime of the aircraft exhaust plume and discusses the modelling approaches present in the literature which are implemented to represent and parametrise plume-scale effects in large-scale models of aviation's impact on the climate.

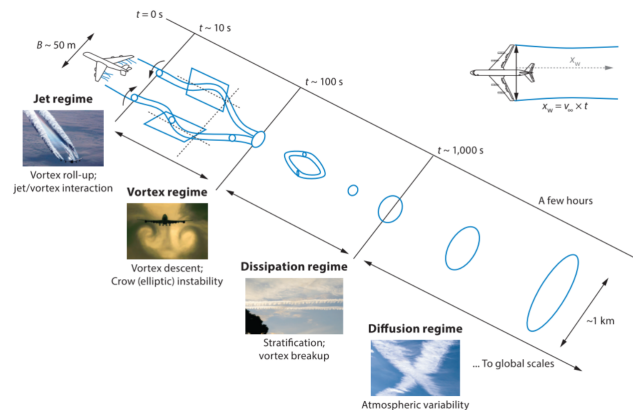
### 3. The dispersion of aircraft emissions and the aircraft exhaust plume

Following their expulsion into the free atmosphere throughout flight, aircraft exhaust gases are confined to a plume that undergoes a series of dynamical regimes (**jet**, **vortex**, **dispersion** and **diffusion** regimes), before becoming fully diluted in the surrounding air. The entrainment of emissions within the plume throughout these dynamical regimes leads to initial species concentrations that are several orders of magnitude higher than background levels [15], giving rise to a number of nonlinear chemical and microphysical effects. These plume-scale effects have considerable implications on the eventual chemical composition of the surrounding atmosphere and lead to the formation of aerosols and ice crystals in the aircraft wake. Therefore, inclusion of plume-scale effects is vital for high fidelity modelling of aviation's impact on the climate.



### 3.1. Plume-scale dynamical regimes

In order to accurately account for nonlinear effects experienced in the aircraft exhaust plume, one must first understand the dynamical response of the plume after combustion, to gauge the length and time scales over which aircraft emissions are entrained within it.



**Figure 3.** Aircraft exhaust plume dynamical regimes [12].

Once emitted from an aircraft's core engines, exhaust gases mix with engine bypass air and cool to around 300 K [3], forming an axisymmetric jet which rapidly diffuses into ambient air. This period, known as the **jet regime**, occurs over a timescale of around 1-20 s, with temperatures cooling to just above ambient throughout. Over the course of the jet regime, the airflow passing over the wings is diverted downwards to generate lift, thus creating a vortex sheet at the trailing edge of the aircraft. This vortex sheet rolls up into a pair of counter-rotating vortices which are shed at the wing tips. The evolving vortex pair then merge together and propagate downwards, due to their mutually induced downwash velocity, trapping the exhaust plume within their cores and signalling the beginning of the vortex regime.

Throughout the **vortex regime**, which occurs between 20 s and 2 minutes after combustion, the primary wake containing the vortices and trapped exhaust plume sinks by around 150–200 m, resulting in a slight temperature increase of 1–3 K, due to the adiabatic heating of exhaust constituents in the sinking vortices. Further to this, the organised vortical structure means the wake does not grow significantly during this time, and hence the concentrations of entrained chemical species remain relatively constant. The adiabatic heating of the exhaust does however lead to baroclinicity at the border between each vortex and the ambient air, which detrains some momentum, heat and exhaust constituents from the primary wake, to form a secondary wake. This secondary wake trails upwards as it is warmer than the surrounding ambient air, and it escapes the influence of the vortex structure, resulting in enhanced mixing with ambient air and thus experiences different chemical and microphysical processes compared to the primary wake [16].

Following this is the **dispersion regime**, in which the aircraft-induced dynamics subside due to the growth of Crow instability [17],

which dissipates and disintegrates the primary and secondary wake vortices [18]. The breakdown of the organised vortical structure and the production of turbulent motion leads to a sudden increase in the rate of entrainment between the exhaust plume and the ambient air by a factor of 10, therefore giving rise to a continuous decay of concentration and temperature within the plume. This regime lasts for 2–5 minutes after combustion.

Lastly, the plume undergoes its final dynamical event, known as the **diffusion** regime. This regime is characterised by the aircraft-induced dynamics becoming negligible (after about 6 minutes [19]), followed by the subsequent dominance of atmospheric processes in the spreading of the aircraft exhaust plume and its constituents. Atmospheric turbulence, radiation transport, stratification are examples of natural phenomena that contribute to the diffusion of the plume, with total dilution to ambient concentrations often occurring over timescales of 2–12 hours post emission. During this time, the plume may spread up to a few kilometres through atmospheric turbulence and shear in the ambient air, diluting the exhaust species over vast volumes of airspace [20].

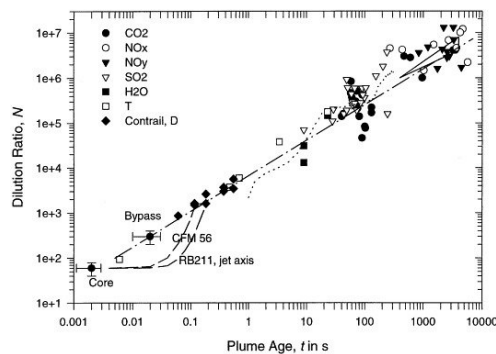
### 3.2. Aircraft plume characteristics

The entrainment of aircraft emissions into the exhaust plume following release into the atmosphere has a considerable effect on the ensuing climate response, through nonlinear chemical and microphysical processing. The magnitude of plume processing is influenced by the spatial and temporal characteristics of the plume. This means that quantitative assessment and modelling of plume-scale effects on the climate requires knowledge of key plume properties such as the characteristic rate of mixing with ambient air, throughout the 4 dynamical regimes, and the lifetime over which the plume typically resides.

#### 3.2.1. Dilution and rate of mixing

The eventual climate impact of aircraft emissions depends on the magnitude of nonlinear processing that occurs within the aircraft plume, and hence depends on the rate at which the plume expands and mixes with ambient air [20]. For this reason, analysis of plume-scale climate effects requires the estimation of plume dilution and mixing rates. In [21], empirical data was collated from over 70 aircraft exhaust plume encounters with research aircraft, to investigate the mixing rate of plumes throughout their typical lifetime. The characteristic property observed in this study is the plume dilution ratio,  $N$ , which is defined as the amount of air mass with which the exhaust plume generated from a unit mass of fuel burn, mixes with, per unit flight distance within the bulk of the plume.





**Figure 4.** Dilution ratio against plume age derived from empirical data [21].

In figure 4, measured dilution ratios from the plume encounters are plotted against plume age. The data was generated by measuring the concentrations of a range of chemical sources across a variety of aircraft types between 0.001 to 10,000s. The significance of these findings is that, in spite of the diverse range of chemical species and aircraft types observed, throughout all four dynamical regimes, a relatively stable logarithmic relationship emerges between dilution ratio and increasing plume age. When interpolating the regression line fitted to the data in figure 4, the following equation can be obtained where  $N$  represents dilution ratio,  $t$  is plume age and  $t_0$  serves as an arbitrary reference scale.

$$N = 7000(t/t_0)^{0.8}, \quad t_0 = 1s \quad (1)$$

It is evident from the figure however, that encounters with plumes older than 50–100 s tend to diverge from the line fit, indicating a reduction in accuracy of the logarithmic approximation over time. This is likely due to the transition from the organised vortex structure present in the vortex regime, to the turbulent dispersion regime, where more unpredictable atmospheric processes begin to take place and become the primary influence on the evolution of the plume. Therefore, this empirical model can only be used reliably up to the vortex regime. Beyond this, a Gaussian approximation to the distribution of species concentrations is typically employed, accounting for dispersion effects experienced at cruising altitudes, such as advection, gravitational sedimentation, anisotropic diffusion, wind shear and stable stratification [22]. See section ?? for more detail on the usage of Gaussian distributions and the two-dimensional diffusion equation in modelling aircraft plumes.

### 3.2.2. Plume lifetime

The lifetime of a plume is typically signified by the end of the diffusion regime and eventual return towards ambient background concentrations, around 2–12 h post emission. However, numerous distinct definitions have become apparent over the past few decades, which enable the concept of plume lifetime to be measured and used to determine the duration over which plume-scale processes should be accounted for when modelling the atmospheric effects of aviation. In [12], three definitions of plume lifetime are explored.

358 The first of these is the so-called “dispersion” time  $t_{ref}$  [13], de-  
 359 fined as the time it takes for the plume to occupy the dimensions of  
 360 a given reference area  $A_{ref}$ . This can be a useful metric to determine  
 361 the time taken for a plume to become fully immersed in a known  
 362 volume such as a latitude-longitude grid box of a global atmospheric  
 363 model or the boundaries of a flight corridor. The second definition  
 364 of plume lifetime is known as the “mixing” time  $t_{mix}$ , as [23?, 24].  
 365 This is the time in which the chemical conversion rate of  $\text{NO}_x$  in the  
 366 plume becomes sufficiently close to that of the surrounding ambient  
 367 air, falling below a very small threshold value. The dependency on  
 368 the state of the background atmosphere means that this lifetime can  
 369 change significantly with time and location. The final definition is from  
 370 [14] where the lifetime  $t_l$  is determined by the exhaust concentrations  
 371 dropping below a threshold mixing ratio and the excess exhaust mass  
 372 dropping to zero.

373 The second lifetime definition,  $t_{mix}$ , is entirely dependent on the  
 374 state of the background atmosphere, meaning the lifetime is subject  
 375 to change significantly given a change in time and/or location of  
 376 emission, as shown in figure 1 of [23]. Moreover, the first and third def-  
 377 initions not only vary with atmospheric conditions, but also depend on  
 378 the threshold values set by the observer, meaning consistent thresholds  
 379 must be maintained throughout a given study to maintain validity.  
 380 Results from the aforementioned studies produced estimates of plume  
 381 lifetime that lie in the range of 10–20 h. Given that plume lifetime and  
 382 rate of mixing are calculable and measurable concepts, it is now possi-  
 383 ble to model the temporal and spatial scales of the chemically active  
 384 plume, so as to investigate the extent to which nonlinear chemistry  
 385 and microphysics influence the ensuing climate impact throughout the  
 386 lifetime of the plume.

### 387 3.3. Plume-scale modelling

388 Convention amongst the climate modelling community is to as-  
 389 sume instantaneous dispersion of aircraft emissions into the atmo-  
 390 sphere, neglecting aircraft-induced dynamics and the presence of air-  
 391 craft plumes, to which emissions are confined to for hours after release  
 392 into the atmosphere. This approach therefore excludes plume-scale  
 393 effects and thus leads to notable discrepancies in the chemical and  
 394 physical response of the atmosphere in the model, compared to real  
 395 life. To tackle the issue of neglected plume-scale effects, a number of  
 396 plume modelling methods of varying fidelity have been theorised in  
 397 the literature, and their outputs have been parametrised into global  
 398 models to reduce uncertainties in the calculation of the climate impact  
 399 of aviation.

#### 400 3.3.1. Plume modelling methods

401 A common method found in [13] is known as the single plume  
 402 (SP) model which approximates the time-evolving concentration field  
 403 of an aircraft exhaust plume using a Gaussian distribution acting  
 404 perpendicular to the flight path [25]. The SP model solves two sets  
 405 of ordinary differential equations that describe the diffusion using  
 406 horizontal ( $D_h$ ), vertical ( $D_v$ ) and shear ( $D_s$ ) diffusion coefficients. One

set of equations is for the mean concentration inside the plume and the other is for the background concentration. From this, the plume volume and concentration can be deduced as a function of time, thus serving as input to atmospheric models in the form of plume mixing ratios and concentrations of key chemical species.

A generalisation to the SP model is the **Multi-layered Plume** model [12?]. This model discretises the concentration distribution in the radial direction into a number of concentric rings that each have a constant concentration equal to that of the mean concentration within the ring according to the Gaussian distribution. The primary benefit of this model over the SP model is the reduced computational demand from assuming discrete and homogeneous concentration rings, compared to the SP approach with a continuous concentration distribution.

Models such as the Aircraft Plume Chemistry, Emissions, and Microphysics Model (APCEMM) from [26] further increase the accuracy of the SP and **Multi-layered Plume** models by capturing additional effects that occur within the plume. This includes effects such as plume anisotropy and asymmetry which impact the eventual spatial distribution of the plume, and the inclusion of modelling of the microphysical processes that strongly influence contrail formation and persistence.

Finally, the plume dynamical evolution can be most accurately captured using high-resolution LESs over the entire lifetime of the plume. LESs can model dynamics on a scale of several millions of grid points for a few seconds to a few minutes of plume age, providing unmatched levels of accuracy at the cost of extremely high computational demand. For this reason, LESs are usually limited to case studies from which the data obtained can be used to derive and calibrate plume parametrisations for use in the lower fidelity methods [27].

### 3.3.2. Effective emissions and parametrisation into global models

- Attempt to parametrise into global models EEIs, ECFs, EERs
- Issues with resolving between plume and global models
- Nonlinear chemistry subsection in climate section will cover exact impact on chemical concs caused by plume effects (Ozone drop due to high conversion rate to reservoir in early plume)

## 4. Air traffic distribution

The previous section discussed the forces of flight and the generation and dispersion of aircraft emissions, from the perspective of a single aircraft. However, the state of the atmosphere is inhomogeneous with respect to space and time, meaning that the climate sensitivity to aircraft emissions differs considerably, depending on the time and location of their release. Therefore, to begin to understand the relationship between aircraft emissions and their resulting climate impact, one must first gain insight into the characteristic operation of aircraft within the wider air transportation system. Key factors that shape the distribution of air traffic, and hence aircraft emissions, include the management and control of air traffic flows and the obligation to meet geographical and temporal demand.

#### 4.1. *Air traffic management*

The air traffic management (ATM) system is responsible for overseeing the network-wide implementation of safe, orderly and efficient air traffic flows, providing assistance to aircraft in transit from departure to destination aerodrome. It is the role of the air traffic control (ATC) team to manage and monitor air traffic in their respective airspace in real time, ensuring that the safety, order and efficiency of aircraft operations are maintained at all times [28]. This means that air traffic controllers must constantly monitor the state of the airspace, swiftly identifying and resolving any unfavourable circumstances by issuing the appropriate clearances or instructions to rectify the situation.

##### 4.1.1. *Air traffic safety*

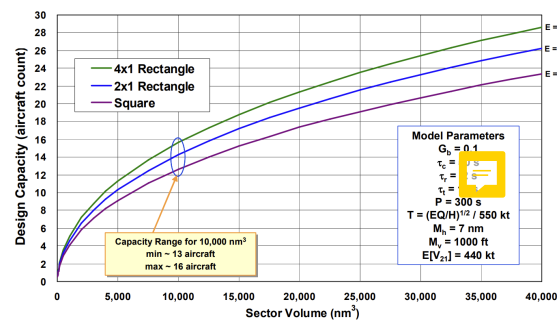
The inherent risk involved in the transportation of vast numbers of passengers at near transonic speeds through the upper atmosphere means that aviation safety is of paramount importance. A safe aircraft operation takes the path of least danger, primarily influenced by the need to avoid unfavourable atmospheric conditions and to prevent conflicts with other aircraft.

In-flight atmospheric conditions susceptible to icing, turbulence or the presence of hazardous convective weather can all be classified as unfavourable for aircraft, with the latter presenting the greatest constraint on aircraft routing [29]. The enhanced risk resulting from flight through weather-affected regions means that aircraft must re-route, leading to restrictions on available airspace and deviations from the optimal flight profile, thus increasing flight-times, fuel burn and delays [30].

The safety risks associated with aircraft-to-aircraft collisions necessitate air traffic controllers to impose safe separation standards between aircraft in the lateral, longitudinal and vertical direction, as specified in [31]. The stated minimum separation distances between aircraft are 5 nautical miles (NM) laterally, 20 NM longitudinally and 1,000 ft in the vertical direction under the most lenient scenarios. Enforcement of separation minima introduces a theoretical upper limit on airspace density within a fixed volume of airspace, in which a breach of separation in more than one direction, known as a conflict, must be resolved as quickly as possible.

The true upper limit on airspace density is determined by a parameter known as airspace capacity, defined as the maximum number of aircraft permitted in a region of airspace over a given time frame. The effective management of air traffic depends on the human cognition of air traffic controllers, to make difficult decisions and carry out complex tasks in a time-critical dynamic environment. As density and complexity levels of air traffic increase, so does the mental workload of the air traffic controller, up until a threshold level is reached where the controller can no longer safely handle the situation. This threshold mental workload is therefore a key determinant of airspace capacity, and is driven by the airspace situation, state of equipment being used, and the controller's own mental state [32]. To estimate capacity, controller workload models are used, which model ATC tasks

to determine a safe upper limit on workload. In [33], a macroscopic workload model is proposed which generalises ATC tasks into four distinct categories: background, transition, recurring and conflict tasks. This provides an objective basis for estimating capacity and enables the formulation of an analytical relationship between airspace capacity and sector volume, as seen in figure 5.



**Figure 5.** Aircraft capacity estimation against sector volume for a range of airspace scenarios [33].

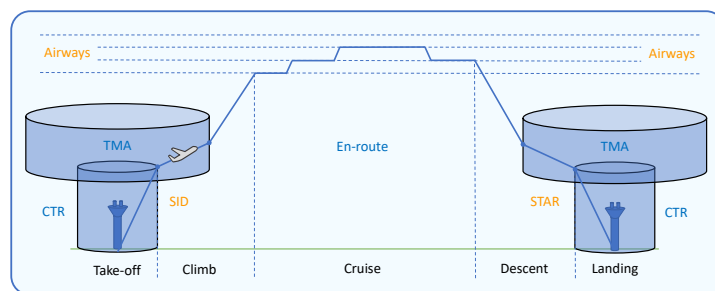
#### 4.1.2. Air traffic order

To keep air traffic flows organised within controlled airspace, aircraft are ordered to follow the traditional fixed-route air traffic network, limiting the possible trajectories flown and further restricting airspace availability. The air traffic network is constructed from four key airspace elements that facilitate the air traffic management process [28]:

- **airports/aerodromes** - an area of land or water intended to be used for the arrival, departure and surface movement of aircraft;
- **waypoints** - a specified geographical location used to define the flight path of an aircraft, representing either a navigational aid (navaid) or a reference coordinate that the aircraft must fly-by or fly-over;
- **airways** - a controlled portion of airspace established in the form of a corridor (usually 8-10NM wide) between two waypoints;
- **sectors** - a region of airspace managed by a single ATC team, stratified into various levels to accommodate a wide variety of traffic.

In [34], the notion of optimising air traffic flows for a given demand and capacity is explored, in which air traffic flows are represented using the four key elements. **Airports** represent the sources and sinks of the flow, **airways** are the arcs along which the flow travels, **waypoints** are the network nodes at which airways intersect, merge or diverge, and **sectors** are a collection of waypoints and contiguous segments of airways. Each airspace element has a corresponding capacity which is declared by the air traffic controller in charge of the airspace. If traffic volumes within respective airspace approach or exceed capacity levels, this can result in significant delays and congestion in the flow of air traffic, resulting in ATM efficiency losses, increased passenger dissatisfaction, and a greater environmental burden.

Controlled airspace is divided into the control zone (CTR), the terminal manoeuvring area (TMA) and en-route airspace. The control zone is the region of protected airspace in the immediate vicinity of an aerodrome, established to ensure the safe departure and arrival of aircraft [35]. The TMA is a larger control area situated above the control zone, which deals with aircraft in the landing and take-off (LTO) cycle, often serving as a junction for airways leading towards the control zone. Aircraft within this region must comply with so-called standard instrument departures (SIDs) and standard arrival routes (STARs) that are designated to a flight to minimise potential conflicts through the use of specific routing, speeds and altitudes. En-route airspace is defined as the volume of airspace outside terminal control areas, beginning at the termination of an SID and ending at the initiation of a STAR, encompassing the climb, cruise and descent phases of flight [36]. In en-route airspace, aircraft follow airways which are situated at a given flight level (FL), where flight levels are a measure of pressure altitude, expressed in hundreds of feet (e.g. FL 360 corresponds to a pressure altitude of 36,000 ft). Figure 6 portrays a typical vertical flight profile of an aircraft from departure to destination, under the constraints of controlled airspace.



**Figure 6.** Typical aircraft trajectory subject to airspace constraints.

#### 4.1.3. Air traffic efficiency

The third and final component of effective air traffic management is the optimisation of flight trajectories, subject to the prioritisation of safety and the compliance with the fixed-route airspace structure. Flight trajectory optimisation is an essential step in ensuring maximum airspace utilisation and efficiency, so that revenue is maximised and demand levels are sufficiently met. Trajectory optimisation is a multi-faceted problem, requiring consideration of nonlinear aircraft performance, wind and weather forecasts, payload, departure fuel load, reserve fuel load and ATM constraints that restrict aircraft operations and routing [37]. This requires an exhaustive assessment to be carried out at the flight planning stage, to test all possible combinations of route, payload, fuel load and operating approach, involving tens to hundreds of thousands of calculations per flight. The most optimal scenarios are then ranked in order of optimality, with the final route selected based on operator preference and/or the occurrence of unexpected circumstances, such as sudden adverse weather conditions or aircraft conflicts [38].



578 In an ideal airspace situation where the atmosphere is calm and  
579 constant; aircraft are not constrained to a fixed route; and there is  
580 no risk of conflict with other aircraft, the least-time and least-energy  
581 aircraft operation would be to fly the great-circle arc between departure  
582 and destination. The vertical profile of the aircraft would consist of  
583 a continuous climb out to the most efficient cruise altitude, then to  
584 cruise at constant speed, with the ability to cruise-climb continuously  
585 as the aircraft burns fuel and loses mass.

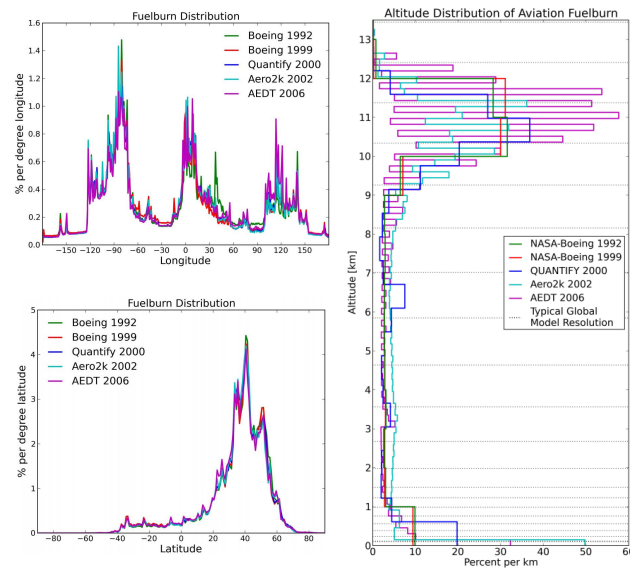
586 In reality, the true optimal route can deviate considerably from  
587 the great-circle arc, depending on the state of the atmosphere and the  
588 airspace situation along the route. Horizontal trajectory optimisation  
589 involves taking the path which minimises risk of bad weather encoun-  
590 ters and collisions, abides by the fixed-route airspace structure, whilst  
591 also flying a route which is optimised with respect to wind and temper-  
592 ature. The magnitude and direction of wind and the localised variation  
593 in temperature experienced by the aircraft throughout flight, can have  
594 a drastic impact on route optimality, with tailwinds and colder tem-  
595 perature regions being favourable [39]. [40] found for a wide range  
596 of wind-optimal flight scenarios that domestic flight saved up to 3%,  
597 and international flights saving up to 10% on both fuel burn and travel  
598 time, despite flying longer routes. Trajectory optimisation in the verti-  
599 cal direction involves climbing to the optimum cruise altitude where  
600 air density is low so drag is minimised, whilst maintaining sufficient  
601 airflow to provide adequate thrust and lift to achieve steady level flight.  
602 In fixed-route airspace however, the aircraft must conform to flight  
603 level allocations, meaning that as the aircraft climbs as it burns fuel,  
604 it must remain at allocated flight levels, further condensing air traffic  
605 and its corresponding emissions into narrow bands of altitude.

#### 606 4.2. Aviation demand

607 Flight routes are constrained by the need to avoid hazardous  
608 situations, abide by ATC standards and optimise aircraft operations,  
609 however another factor which significantly affects air traffic distri-  
610 bution is aviation demand. There are inconsistencies in the levels of  
611 demand for aviation both geographically and temporally, depending  
612 on the demographic of the area and the preferences of those who do  
613 fly.

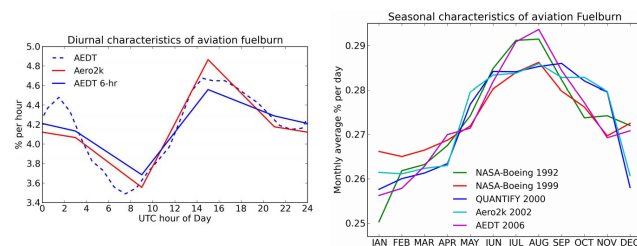
##### 614 4.2.1. Global air traffic and emissions distribution

615 The nature of air traffic and emissions distribution was investi-  
616 gated in [41] where a range of global aircraft emissions datasets are  
617 compared (NASA-Boeing 1992, NASA-Boeing 1999, QUANTIFY 2000,  
618 Aero2k 2002, AEDT 2006 and aviation fuel usage estimates from the  
619 International Energy Agency) to show distribution patterns in the  
620 latitudinal, longitudinal and vertical sense. Further to this, temporal  
621 variations with respect to both the diurnal (time of day) and seasonal  
622 (time of year) cycles are explored.



**Figure 7.** Spatial (latitude, longitude and altitude) distribution of global aviation emissions from a range of aircraft emissions datasets [41].

Figure 7 shows the spatial distribution of fuel burn across the range of datasets in the longitudinal, latitudinal and vertical directions. The longitudinal distribution shows three emissions peaks around the densely-populated regions of the US, Europe and East Asia, with the largest situated above the North American land mass. It is evident in the latitudinal distribution, that the Northern Hemisphere dominates, with a strong peak in the northern mid-latitudes that appears due to high volumes of air traffic above the US and Europe, as well as along the connecting region of airspace, the North Atlantic flight corridor (NAFC). Contrarily, there are almost no emissions present in southern latitudes below 40°S, with the region between 40°S and the equator constituting only a small percentage. The altitudinal distribution on the other hand, experiences emissions peaks around both the low altitude LTO area and the high-altitude cruising regions between 9 to 13 km, with relatively low emission intensities at mid-altitudes. Furthermore, the peak around cruising altitude is discretised into peaks every other flight level, due to the vertical separation constraints and the allocation of aircraft to specific flight levels, thus owing to further increases in emissions intensities at these altitudes.



**Figure 8.** Temporal (diurnal and seasonal) distribution of global aviation emissions from a range of aircraft emissions datasets [41].

642 The temporal variation of atmospheric parameters evident in  
643 figure 8 (e.g. background concentrations and reaction rates) means  
644 that the climate sensitivity to aircraft emissions is highly dependent on  
645 when the emission is released. Therefore, understanding the temporal  
646 emissions distribution with respect to the diurnal and seasonal trends  
647 is crucial. The diurnal cycle of global aviation fuel burn, as seen on the  
648 left hand side of figure 8 displays a peak at around 15:00UTC, which  
649 decreases through the night until around 09:00UTC where total fuel  
650 burn begins to increase again. With regards to seasonal variation, there  
651 is significant variance between emissions datasets, however in general,  
652 all display a wintertime minimum between December and January,  
653 and a summertime maximum between June and September.

#### 654 4.2.2. Local air traffic and emissions distribution

655 Due to the fixed-route nature of airspace, aircraft tend to fly along  
656 common airways or flight corridors, and pass common waypoints  
657 along their journey, leading to exceptionally high flux densities of air-  
658 craft through these regions at peak times. This has implications on the  
659 nonlinear chemical and physical effects occurring at the plume scale,  
660 due to the intersection of aircraft plumes and the elevated exhaust gas  
661 concentrations entrained within them. A prime example of a high-  
662 density airspace region is the NAFC, made up of a series of tracks that  
663 aircraft traversing the North Atlantic must follow, updated daily to  
664 allow for convective weather avoidance, tracking of the North Atlantic  
665 Jet Stream and favourable tailwinds to maximise efficiency [42]. The  
666 annually-averaged number of aircraft traversing the NAFC per day  
667 has increased from 800 in 1997 [43] to around 2,500 in recent years  
668 [35], owing to the tripling of passenger demand since then [44]. With  
669 North Atlantic air traffic being confined to a limited number of tracks  
670 (usually 3 or 4), it can be assumed that aircraft separation distances  
671 and airspace capacities are pushed to their limit on a regular basis  
672 along this flight corridor.

673 Previous experimental work on air traffic emissions in the NAFC  
674 was carried out in the late 1990's, through campaigns such as the  
675 Pollution from Aircraft Emissions in the North Atlantic Flight Corri-  
676 dor (POLINAT) and Subsonic Assessment Ozone and Nitrogen Oxide  
677 Experiment (SONEX) [45]. At least 20 follow up papers were pub-  
678 lished following these campaigns, in which POLINAT/SONEX data  
679 are utilised to provide insight on a number of major scientific issues  
680 [46]. A noteworthy publication with regards to localised emissions  
681 impacts is [47], which carries out an in-situ investigation of air traffic  
682 emissions signatures (nitrogen oxides ( $\text{NO}_x$ ), sulfur dioxide ( $\text{SO}_2$ ) and  
683 cloud condensation nuclei ( $\text{CCN}$ )) in the NAFC using experimental data  
684 from a POLINAT research aircraft. The research aircraft flew perpendic-  
685 ular to the major eastbound corridor tracks and took measurements of  
686 various chemical concentration fields and meteorological parameters  
687 throughout. The results show that the superposition of aircraft exhaust  
688 plumes led to peak concentrations of  $\text{NO}_x$ ,  $\text{SO}_2$  and  $\text{CCN}$  above back-  
689 ground levels by factors of 30, 5 and 3, respectively. This is because  
690 plume dispersion timescales greatly exceed the daily frequency with  
691 which aircraft emissions are input into the flight corridor, resulting in

an inhomogeneous concentration field with narrow and sharp peaks over a relatively low and smooth background level [45].

The management and optimisation of air traffic directly influences the distribution of aircraft, and hence their associated emissions, throughout the atmosphere at cruising altitudes. The importance of safety, order and efficiency in the air traffic management process and the characteristics of global and local air traffic flows have been discussed. The following section will explore the atmospheric response to the input of aviation emissions on a global and local scale, through evaluation of the climate forcing mechanisms that result from emissions and analysis of their relative global warming contributions. Further to this, the nonlinear chemistry and microphysics within the exhaust plume is covered in great detail.

## 5. The climate impact of aviation

### 5.1. Atmospheric chemistry in the UTLS

As figure 7 suggests, the vast majority of aviation fuelburn and hence aviation emissions, occur between 9 and 13km altitude. The region of the atmosphere encompassing this volume of airspace is known as the Upper Troposphere and Lower Stratosphere (UTLS), with around 20-40% of total aircraft emissions being released in the LS [48]. The greenhouse effect due to the release of chemically-active substances in the UTLS is considerably greater than that of emissions at the surface, due to the increased sensitivity of the climate at these altitudes. The climate in the UTLS is more sensitive due to increased residence times of pollutants, lower background concentrations meaning emissions have a greater influence on the chemistry, lower temperatures, and a higher radiative efficiency ?? The climate effects associated with water vapour and NO<sub>x</sub> emissions are particularly sensitive to the atmospheric conditions of the UTLS. Water vapour emissions released into the sufficiently cold and humid conditions of the upper atmosphere form condensation trails, which trap heat more effectively than they reflect inbound solar radiation, leading to a net global warming effect. Moreover, NO<sub>x</sub> emissions constitute a 30 times greater climate impact compared to equivalent surface emissions, due to the absence of direct deposition and slower conversion to stable reservoir species at aircraft cruising altitudes [? ].

With the enhanced climate sensitivity of the UTLS and the highly variable chemical and meteorological state of the atmosphere at these altitudes, the climate impact induced by an aircraft depends not only on the mass of emissions it releases, but also on the time and location of their release. Therefore, aircraft emissions climate effects are spatio-temporally sensitive and require comprehensive climate metrics to quantify their atmospheric effects.

### 5.2. Radiative forcing aircraft emissions

The most common climate metric used to compare the magnitudes of climate impact from a range of emission species is radiative forcing (RF). RF is defined as the perturbation to the net energy balance of the Earth-atmosphere system due to natural or anthropogenic factors of climate change, measured in watts per square metre [W/m<sup>2</sup>]

[49]. The emission of harmful chemical species into the UTLS, as is the case for aviation, induces a number of climate forcing mechanisms that exhibit a radiative forcing effect. [50] presents an updated analysis of the global effective radiative forcing (ERF) contributions for aviation-induced climate change. ERF is a newly proposed climate modelling framework that builds upon the RF concept by removing rapid atmospheric adjustments that bear no relation to the long-term surface temperature response that occurs over decadal timescales. ERF serves as a more suitable equivalency metric to compare the global warming response of heterogeneously distributed short-lived climate forcers and uniformly distributed long-lived climate forcers. Figure 9 displays the ERF and RF contributions for each of the key aviation-induced climate forcers.

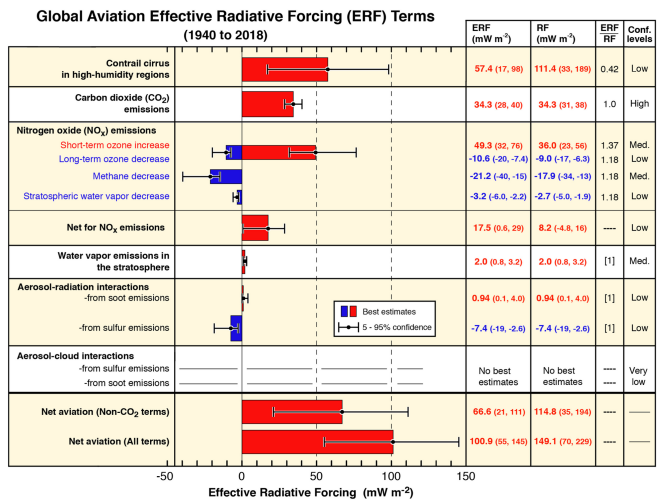


Figure 9. Radiative forcing contributions from global aviation between 2000 and 2018 [50].

5.2.1. CO<sub>2</sub>

The RF of aviation CO<sub>2</sub> emissions is direct and well understood. Due to its thermodynamic and photochemical stability, CO<sub>2</sub> has a relatively long atmospheric lifetime on the order of 100 to 1,000 years. This means that aviation CO<sub>2</sub> emitted into the atmosphere simply serves to increase its atmospheric concentration, leading to the eventual distribution over global spatial scales and a steady, uniform increase in the planetary greenhouse effect. The ubiquitous and intuitive nature of CO<sub>2</sub>-related warming deems it a suitable benchmark to compare warming from non-CO<sub>2</sub> climate forcers against.

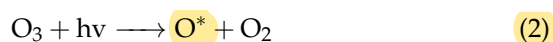
5.2.2. Contrail cirrus

Water vapour and particulate emissions released from jet-powered aircraft in sufficiently cold and humid conditions give rise to the generation of anthropogenic vapour trails, known as contrails. Such trails often have a significant level of opacity and occur at high altitudes, tending to absorb outbound terrestrial radiation more efficiently than they reflect inbound solar radiation, thus inducing a net global heating effect. The extent of contrail-related warming is highly dependent on the ambient conditions of the surrounding atmosphere – in particular,

contrails generated in regions which are supersaturated with respect to ice are thought to create the greatest warming effect. The ice crystals they are composed of grow with age and spread over vast expanses of the atmosphere, often leading to the formation of additional cirrus clouds, or the consolidation of existing ones, at high-altitudes in the upper atmosphere.

### 5.2.3. Net-NO<sub>x</sub>

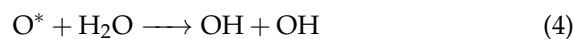
Tropospheric chemistry is dominated by ozone (O<sub>3</sub>), a potent greenhouse gas which contributes to climate change and air quality issues. O<sub>3</sub> is an oxidant in its own right, reacting with reactive volatile organic compounds (VOCs, see later) but is the precursor to the OH radical, which is known as the chemical detergent of the lower atmosphere. The OH radical reacts with VOCs and allows them to be oxidised and removed from the atmosphere. Photolysis of ozone (absorption of sunlight in the UV region 290-330 nm and the breaking of an O-O bond) starts the process of OH production, depicted by reaction (1), where  $h\nu$  denotes a photon of light;



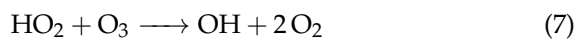
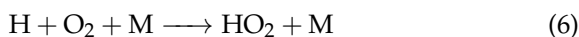
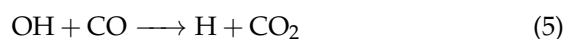
Reaction (1) makes a reactive oxygen atom O\*, that can either collide with N<sub>2</sub> or O<sub>2</sub> molecules (denoted as M) and be quenched to ground state O atoms in reaction (2)



Or it can react with water to form HO radicals

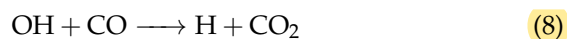


OH radicals can then participate in chemical cycles that preserve levels but also remove pollutants, CO, in the following examples. First, under clean conditions (low NO<sub>x</sub> = NO and NO<sub>2</sub>) the following scheme (1) takes place:

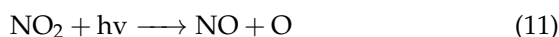
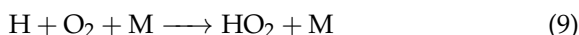


Net reaction  $\text{CO} + \text{O}_3 \longrightarrow \text{CO}_2 + \text{O}_2$

Here, ozone is removed and CO is oxidised to form CO<sub>2</sub>, which, in a pre-industrial world, was taken up by plants and the oceans on relatively short timescales. This clean condition scheme removes pollutants and keeps ozone levels under control. However, industrial activity through fossil fuel combustion has increased levels of NO<sub>x</sub> in the atmosphere, changing from scheme (1) to scheme (2):







Net reaction:  $\text{CO} + 2\text{O}_2 \longrightarrow \text{CO}_2 + \text{O}_3$

Now, ozone is formed as CO is oxidised, and one can assume that adding more  $\text{NO}_x$  would further increase ozone production. However, if NO and  $\text{NO}_2$  levels surpass a threshold, they can compete with VOC for reaction with OH radicals:



Reactions (13) and (14) are called termination reactions, removing radicals from the atmosphere and preventing  $\text{O}_3$  formation. In polluted environments, OH collapse can occur, and so ...

#### 4. Stratospheric water vapour

##### 5.2.5. Aerosol effects

##### 5.3. Nonlinear chemistry and microphysics in the aircraft exhaust plume

#### 6. Plume superposition

Following expulsion into the free atmosphere, exhaust species become entrained in the aircraft wake, forming a plume of elevated chemical concentrations which persist for 2 to 12 hours ??, before fully dispersing into ambient air. The build-up of non- $\text{CO}_2$  emissions in the plume give rise to a number of nonlinear chemical and microphysical effects, which influence the ensuing atmospheric response by altering the net production rates of radiatively active gases and affecting contrail formation and persistence. It is often the case however, that in large-scale atmospheric models, emissions are instantaneously diluted into the volume of the smallest resolved grid cell, with dimensions according to the model's spatial resolution. The instantaneous dilution (ID) approach neglects the plume-scale processes, thus leading to discrepancies in the calculated climate response such as the overestimation of  $\text{O}_3$  production,  $\text{CH}_4$  and CO destruction, and the increased rate of  $\text{NO}_x$  conversion to nitrogen reservoir species ?. Furthermore, it is stated in ?? that the formation of ice in aircraft exhaust plumes may result in additional heterogeneous chemical reactions, that are not captured in global atmospheric models.

##### 6.1. Plume superposition

##### 6.2. Atmospheric response to plume superposition

In ?? the notion of plume superposition and the extent to which it influences the atmospheric response was explored, by comparing

results from an expanding plume model against an atmospheric model which assumes instant dilution. The analysis was carried out firstly on a single aircraft at cruise altitude, then on four aircraft flying in the same direction each separated by 1 hr. Observing the single aircraft's plume 10 hrs after emission, it was found that the instantaneous dilution approach overestimated O<sub>3</sub> production by 33%, CH<sub>4</sub> destruction by 30% and CO destruction by 32%. When observing the response due to the four overlapping plumes however, the overestimation of O<sub>3</sub> production increases to 77%, CH<sub>4</sub> destruction to 68% and CO destruction to 74%. The marked difference in measured response for four overlapping plumes compared to a single plume serves as evidence that emissions saturation in superimposed plumes can significantly alter the chemical response of the atmosphere. However, further work needs to be done to clarify the direction and magnitude of the net change in climate impact resulting from these changes to the chemical composition. Moreover, the sensitivity of the chemical and climate response to plume superposition needs to be tested for a range of aircraft transit frequencies and ambient atmospheric conditions, so as to build up a visual representation over the phase space of the atmosphere. In addition to chemical changes in the atmosphere, plume overlap can also influence the formation and persistence of aircraft contrails and the subsequent generation of aviation-induced cirrus clouds. ?? explored the effect of recurrent water vapour emission on contrail formation and persistence at typical aircraft cruising altitudes. It was discovered that continual contrail generation actually leads to dehydration of the surrounding atmosphere and a diminishing radiative forcing effect of 15% in the affected areas. This outcome provides further impetus to explore the potential effects of plume superposition on the global warming induced by aviation.

## 7. Mitigation potential

### 7.1. Formation flight

Formation flight involves the flight of two or more aircraft in aerodynamic formation, with the follower aircraft positioned in the smooth updraft of the leader aircraft's wake, reducing required lift and thrust, hence reducing fuel and CO<sub>2</sub> emissions by 5-8% per trip ???. A fortuitous outcome of formation flight is however, the saturation of emissions in the trailing exhaust plumes, which can lead to the aforementioned nonlinear chemical and microphysical response that reduces ozone and contrail formation. The non-CO<sub>2</sub> climate effects of a twin-aircraft formation flight are observed in ??, where a climate model was used to determine the changes in NO<sub>x</sub>-related ozone production and contrail formation processes. The case study found that, despite a 1-3% increase in flown distance, CO<sub>2</sub> was reduced by 6% and NO<sub>x</sub> was reduced by 11%. This resulted in a 5% reduction in ozone production efficiency due to NO<sub>x</sub> saturation and a 48% contrail reduction due to mutual competition for available water vapour, resulting in a total climate impact reduction of approximately 23%. While part of the alleviated climate impact can be related to the decrease in total emissions due to wake energy retrieval of the follower aircraft, there is also further emphasis due to the saturation of emissions in the

overlapping plumes of both aircraft involved. This demonstrates great potential for reducing both CO<sub>2</sub>- and non-CO<sub>2</sub>-induced climate effects from aviation, if such a scheme were to be carried out on a global scale.

## 7.2. *Climate-optimal aircraft routing*

Fleet-wide implementation of formation flight would however, pose a number of research obstacles, that must be overcome to maximise its cost-benefit potential, whilst ensuring the safe and orderly operation of aircraft in controlled airspace. Optimising aircraft trajectories to minimise fuel burn requires the consideration of nonlinear aircraft performance, wind and weather, payload, fuel load, and constraints set out by air traffic control ???. Formation flight adds another variable to the fuel burn optimisation problem, maximising the time spent flying in formation whilst minimising deviation from the true optimal route. This is a research topic covered extensively in the literature ???. Optimising formation flight trajectories to minimise climate impact on the other hand, is a relatively novel concept, which requires deeper understanding of the sensitivity of the climate response to emissions under a range of different atmospheric states.

## 7.3. *Alternative propulsion technologies*

# 8. **Plume-scale processes**

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## 8.1. *Plume superposition*

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## Abbreviations

The following abbreviations are used in this manuscript:

MDPI	Multidisciplinary Digital Publishing Institute
DOAJ	Directory of open access journals
TLA	Three letter acronym
LD	Linear dichroism

## Appendix A

### *Appendix A.1*

The appendix is an optional section that can contain details and data supplemental to the main text—for example, explanations of experimental details that would disrupt the flow of the main text but nonetheless remain crucial to understanding and reproducing the research shown; figures of replicates for experiments of which representative data are shown in the main text can be added here if brief, or as Supplementary Data. Mathematical proofs of results not central to the paper can be added as an appendix.

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