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Contrail formation and persistence conditions for alternative fuels

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Abstract
In order to counteract global warming, the European Green Deal was made to improve the journey to a sustainable future. This also has an impact on aviation, because in the future the growth in air traffic must no longer lead to rising emissions, but even all aviation CO_2 emissions have to be reduced to zero to achieve the goal of climate-neutral aviation by 2050. There are several approaches for new propulsion solutions and sustainable vehicle configurations and operations. A promising approach is the use of modern fuels. These include drop-in fuels (kerosene-like fuels) but also revolutionary concepts such as the use of liquid hydrogen or liquid natural gas, electric flying, and mixed forms of these. These approaches have certain advantages regarding the climate impact, but not all processes and effects are fully understood, especially their effects on contrails and their properties, frequency, and lifetime.

In this study, we analyse 10 years of airborne and reanalysis data of temperature and humidity to see, how much more persistent contrails would be formed if kerosene were replaced by alternative fuels of different energy-specific water vapour emission indices, which are generally higher for alternative fuels. It turns out, that the amount of additional persistent contrails is quite minor for drop-in fuels, which are already used nowadays, but it is larger for other kinds of fuels, such as methane and liquid hydrogen.

Keywords: contrails, ice supersaturation, alternative fuels, energy transition

1 Introduction

Aircraft, powered by fossil fuels, emit CO_2 and other gases as well as soot and volatile particles into the atmosphere and leave under certain conditions contrails in the sky, causing changes in radiative fluxes. These flux changes contribute to the climate impact of aviation (for a recent overview, see Lee et al., 2021). The warming effects of aviation exceed the cooling ones and thus contribute to global warming, in particular, due to CO_2 emissions and persistent contrails.

Alternative fuels include not only liquid hydrogen, which is currently the subject of many researches (although it will take many years until airplanes are powered regularly by hydrogen propulsion, see e.g. Klug and Ponater, 2001; Ström and Gierens, 2002; Marquart et al., 2005; Gierens, 2021), but also sustainable aviation fuels (SAFs), which are already added to kerosene nowadays. Alternative fuels, which are not made from fossil sources, can solve several problems if they can be produced sustainably and in sufficient amounts. In particular, their use lessens the chief climate impacts of aviation CO_2 and contrails in flight, because of a higher specific energy content and a higher H/C ratio (ratio of hydrogen to carbon atoms), which they generally have (e.g. Anderson et al., 2011). Moreover, the higher H/C ratio has its origin in a lower fraction of ring molecules (aromatics) compared to linear hydrocarbons. These ring molecules are the initial building blocks for the formation of soot particles; their reduction thus leads to lower soot number concentrations in the exhaust, which in turn leads to lower ice crystal number concentrations in contrails (Bräuer et al., 2021; Voigt et al., 2021). With current soot levels in the fuel, the number of ice crystals is reduced in proportion to the reduction of the soot number emission, up to a reduction factor of about 100. At still larger reduction factors, the ice crystal number can increase again as emitted volatile and ambient particles then take over the role of condensation nuclei (Kärcher, 2018; Kärcher and Yu, 2009). Sulphur, which is present in fossil kerosene in varying amounts (Schumann et al., 2002), is as well strongly reduced or even absent in alternative fuels, which leads to a reduction of the sulphur-based volatile particle number in the exhaust. The kind and number of condensation nuclei in the exhaust affect the microphysical and optical properties of the resulting contrails, but neither their formation nor their persistence. These two issues are purely thermodynamic. Thus, the effect of soot and sulphur on contrails is not considered in this analysis. Eventually, the use of alternative fuels leads to optically thinner and shorter living contrails (Burkhardt et al., 2018; Bock and Burkhardt, 2019) which results in a climate benefit compared to the usual kerosene. The benefit is not

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proportional to the reduction of the ice crystal number and varies with latitude (Bier and Burkhardt, 2019). However, we expect more contrails and in particular more persistent ones according to the theory of contrail formation (see also Caiazzo et al., 2017; Narciso and De Sousa, 2021; Teoh et al., 2022b). Therefore, the actual contrail-related climate benefit from using alternative fuels is the result of counteracting effects: there will be more persistent contrails with individually less impact. The first part of this delicate balance, namely how much more persistent contrails must be expected for alternative fuels, will be studied in the present paper.

In the next section, we show how fuel properties affect contrail formation. In the third section, we then present a data analysis that shows how much more persistent contrails would be formed by a fleet of aircraft using alternative fuels with a higher energy-specific emission index for water vapour (EI_{H_2O}/Q). The results are discussed and summarised and conclusions will be drawn in the final Section 4.

2 Impact of alternative fuels on contrail formation

Contrails form as the consequence of the isobaric mixing of two airmasses, the exhaust plume which initially is hot and moist, and the cold ambient air. It is well known that mixing two air masses of different temperatures can yield supersaturated conditions (e.g. mixing fog). Contrails form if supersaturation with respect to liquid supercooled water is achieved during the mixing process; this condition is known as the Schmidt-Appleman criterion (SAC) after its two developers (Schmidt, 1941; Appleman, 1953). Schumann (1996) presented a detailed derivation and provided a formula for the maximum temperature, $T_{\max}$, at which contrails can form (see below). As soon as the mixture achieves water-supersaturation, emitted and entrained water vapour condenses mainly on emitted soot particles. These droplets freeze quickly in the cold upper-tropospheric air to form ice crystals and thus a contrail. The mixing process that leads to contrail formation is best described with the aid of an e-T diagram (water vapour partial pressure vs temperature) where the phase point of the plume/air mixture follows a straight line that ends at the point that represents ambient conditions (e_a, T_a). The most important property of the mixing trajectory is its slope, G, which depends on ambient conditions, aircraft, and fuel properties, as follows (Schumann, 1996):

$$G = \frac{c_p p}{\varepsilon} \frac{EI_{H_2O}}{(1-\eta)Q}. \tag{2.1}$$

Here, c_p is the specific heat of air at constant pressure, p is pressure, and $\varepsilon = 0.622$, the ratio of the molar masses of water and air. For kerosene the properties are $EI_{H_2O} = 1.24$ kg/kg, the emission index of water vapour (burning one kg of kerosene yields 1.24 kg of water vapour), and $Q \approx 43$ MJ/kg, the lower heating value (combustion energy) of kerosene. Both, EI_{H_2O} and Q, are slightly higher for alternative fuels compared to kerosene. η is the so-called overall propulsion efficiency, which is included here, since only the fraction η of Q propels the aircraft, while the remaining fraction is lost as the heat of the exhaust gases. In this study, we assume $\eta = 0.35$, which corresponds to the overall propulsion efficiency of modern aircraft (Gierens, 2010; Schumann, 2000).

Once G is given for a certain fuel, it is straightforward to compute two threshold temperatures, namely the maximum temperature at which contrail formation is possible at all (Schumann, 1996), $T_{\max}$, and the minimum temperature below which contrails would even form if the ambient air were totally dry ($e_a = 0$), $T_{\min}$:

$$T_{\max} = 226.69 + 9.43\ln(G - 0.053) + 0.72[\ln(G - 0.053)]^2, \tag{2.2}$$

$$T_{\min} = T_{\max} - \frac{e_w^*(T_{\max})}{G}, \tag{2.3}$$

where T is in K and G in Pa/K. e_w^* is the temperature-dependent saturation vapour pressure with respect to liquid water. These threshold temperatures depend, via the factor G, on the fuel properties. We find

$$\frac{\mathrm{d}G}{\mathrm{d}\ln(EI)} = -\frac{\mathrm{d}G}{\mathrm{d}\ln(Q)} = G. \tag{2.4}$$

Alternative fuels have a higher hydrogen content than kerosene which leads to a higher emission index for water vapour as well as to an increase of the lower heating value. Thus, these two influences balance each other to a certain degree, but not completely. Generally, G increases by a few percent on the transition from kerosene to alternative fuels, whereby the mixtures of kerosene and SAFs vary with their different blending ratio. This leads to a small increase of $T_{\max}$ of the order of 0.1 K per percent change in G (see Equation (2.2)) and to a similar increase of $T_{\min}$. Contrail persistence is possible if their formation occurs in ice supersaturated regions, whose phase points lie on the left of the critical mixing line between the two saturation curves in the Schmidt-Appleman diagram (Fig. 1). The change from kerosene to alternative fuels is thus accompanied by a change in the amount of ice supersaturation that is accessible during a contrail-forming mixing process. In Fig. 1, ambient ice supersaturation is represented by phase points between the two saturation lines, marked in yellow. Contrails are persistent in air, which is represented by the yellow marked phase points to the left of the critical mixing trajectories (i.e. to the lower temperatures). For standard fuel, this is the yellow region left of the red line. For SAF (or any fuel with a higher G-factor) it is the yellow marked region left of the blue line (including the region left of the red line). Contrail formation does not occur if the ambient air is characterised by phase points on the right of the respective critical mixing trajectory.

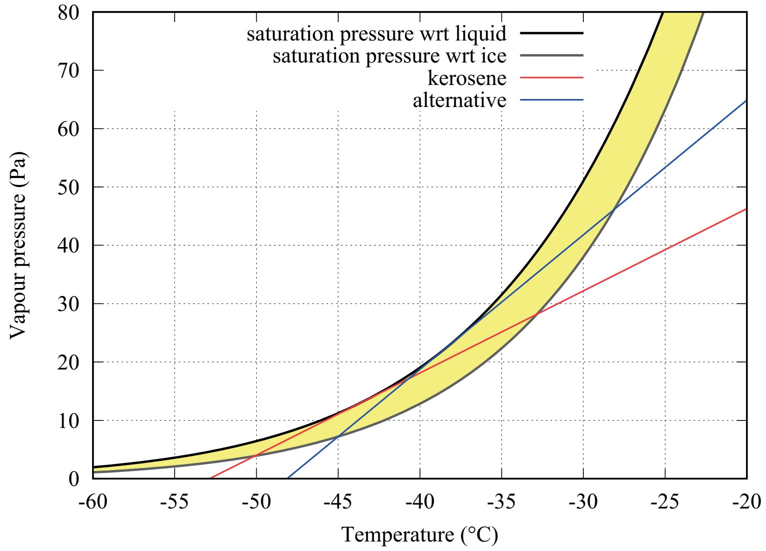

Figure 1: Water vapour pressure vs temperature phase diagram for the isobaric mixing of exhaust gases with ambient air. Shown are the two saturation curves for liquid supercooled water (black) and ice (grey) and a pair of phase trajectories for isobaric mixing that just would allow contrail formation in the case of kerosene combustion (red) and combustion of an alternative fuel (blue). Note that the difference in the slopes of the two mixing lines is exaggerated for a clearer presentation. The yellow region between the two saturation curves represents ice supersaturation, which is a condition for contrail persistence. The region bounded by the saturation curves and the two mixing lines represents cases where flying on kerosene would not produce any contrail, but flying on alternative fuels would lead to persistent contrails.

A higher G-factor not only leads to more contrails but to more persistent contrails as well.

3 Impact of alternative fuels on contrail persistence

The increase in the frequency of contrail formation in ice supersaturated air can be determined in various ways, either by using data from meteorological reanalysis products or by using data obtained from airborne measurements. We deem the latter possibility more appropriate since upper-tropospheric ice supersaturation is not well represented in current weather prediction models (Gierens et al., 2020). But we will use both airborne flight data from the MOZAIC/IAGOS project (Marenco et al., 1998; Petzold et al., 2015), and the corresponding data from the ERA 5 reanalysis (Hersbach et al., 2018; Copernicus Climate Change Service, 2017), interpolated in space and time to the position of the instrumented aircraft, for comparison. Additionally, we scale the ERA 5 values of relative humidity with respect to ice with a factor developed by Teoh et al. (2022a). These authors developed the factor to make the ERA 5 ice supersaturation statistics over the North Atlantic match the corresponding MOZAIC/IAGOS supersaturation statistics. Monthly mean reanalysis data, as used by Narciso and De Sousa (2021), are in our view not appropriate since ice supersaturation will probably be averaged away in monthly means. Hence, we prefer the MOZAIC/IAGOS airborne data and the interpolated ones (from hourly resolved ERA 5) of temperature and relative humidity to perform the analysis.

3.1 Data from real flights

The MOZAIC/IAGOS data come from automated measuring instruments installed on commercial passenger aircraft as part of the “Measurement of Ozone and Water Vapour on Airbus In-service Aircraft” program (MOZAIC) (Marenco et al., 1998), which was transferred into the new European Research Infrastructure “In-service Aircraft for a Global Observing System” (IAGOS) (Petzold et al., 2015) in 2011. This data includes measurements of ozone, water vapour, and other atmospheric variables and has already been used for many studies on contrails, their persistence, or contrail radiative forcing (see e.g. Wilhelm et al., 2021, 2022). In the present study, temperature, relative humidity with respect to ice, and pressure data from all flights from 2000 to 2009 in the area 30° N to 70° N and 125° W to 145° E and in the altitude range between 310 hPa and 190 hPa are used. The MOZAIC/IAGOS data together with the corresponding ERA 5 data are the same as in the studies of Wilhelm et al. (2021, 2022); their treatment is described in these papers. An important property of this data set is the avoidance of autocorrelation; the individual data records have been selected randomly and comprise about one percent of the whole data set. We use the humidity data as given in ERA 5 and additionally with the scaling by Teoh et al. (2022a).

3.2 Method and results

To find out how much more persistent contrails are caused by alternative fuel-driven aircraft compared to conventional kerosene-powered aircraft, the MOZAIC/

IAGOS data and corresponding ERA 5 data (as described in Chapter 3.1) are used. For each of the measuring points, it is calculated whether the Schmidt-Appleman criterion is fulfilled or not by using the relative humidity with respect to ice, the temperature, and the respective pressure level of the aircraft. If there is ice supersaturation measured by the aircraft, and the SAC is fulfilled, a persistent contrail can be assumed for this specific measuring point.

For the reference case (conventional kerosene engine), G is calculated for every aircraft measurement according to Equation (2.1) We assume an overall propulsion efficiency of 0.35 in all cases. Based on these results, Equation (2.2) is used to calculate the maximum temperature at which a contrail can just form under water saturation with the corresponding value for G. For the calculation of the relative humidity with respect to water, the saturation vapour pressure over liquid water and over ice are needed. These are computed for the measured temperatures using the formulation by Murphy and Koop (2005). With the saturation vapour pressure over liquid water and ice and the measured relative humidity with respect to ice, the relative humidity with respect to water can be calculated for every measurement point:

$$RH_w = \frac{RH_i e_i^*}{e_w^*}. \tag{3.1}$$

Next, we compute the relative humidity at the phase point of the actual mixing trajectory when it reaches $T_{\max}$; this is labelled $RH_{\max}$. With this quantity, the fulfilment of the SAC is equivalent to $RH_{\max} \geq 1$, that is, contrails are formed in this case. If instead $RH_{\max} < 1$, contrail formation is not possible. (Please note that $RH_{\max}$ is just an operand; it can even be negative in quite warm situations). $RH_{\max}$ is determined as follows: Let us write $e_{\max}$ for $e_w^*(T_{\max})$. We then calculate

$$RH_{\max} = \frac{e_a + G\,(T_{\max} - T_a)}{e_{\max}}. \tag{3.2}$$

Here, e_a is the ambient saturation vapour pressure and T_a is the ambient temperature. For each data point of the reference case (i.e. kerosene assumed), it is determined whether the SAC is fulfilled (i.e. whether $T_a < T_{\max}$ & $RH_{\max} \geq 1$) and whether there is ice supersaturation. The total number of events for which these conditions are met corresponds to the number of points at which persistent contrails produced by kerosene-powered aircraft are possible.

The total number of possible persistent contrail points for an assumed alternative fuel is calculated in the same way. The only difference is that G is assumed slightly larger than in the reference case. This has been achieved by multiplying the original G-values with factors 1.01 to 1.20 (in 0.01-steps) to represent the increased energy-specific emission index of water vapor or the higher H/C ratio for various mixtures of alternative fuels and kerosene, which is ≈ 1.88 for kerosene, 2.12 for fuels derived from coal, or 2.19 for fuels from natural gas for instance (Anderson et al., 2011). Gierens et al. (2016) found that neat SPK and HEFA fuels (synthetic paraffinic kerosene and hydro-processed esters and fatty acids) have an emission index for water vapour higher by about 9 % than for Jet A-1, with a simultaneous increase of the lower heating value of about 3.5 %. The corresponding increase of G would be about 5.5 %. Two related studies (Caiazzo et al., 2017; Teoh et al., 2022b) assume larger increases for a transition to modern fuels, namely 8 % and 9 %. As we do not know what will be possible in the future, we chose to consider in this study increases of G up to 20 % for sustainable aviation fuels.

The results of the calculations are shown in Fig. 2. The x-axis describes the change factor by which the reference-G for kerosene is multiplied. The y-axis shows the relative increase in the number of cases for which persistent contrails are possible (in %). The more G is increased, the more persistent contrails can form. The increase is stronger for the airborne than for both the unmodified and the modified reanalysis data. For example, for a 20 % increase in G (1.20 on the x-axis), there are almost 4.0 % (3.0 %, 3.4 %) more persistent contrail cases for alternative fuels than for kerosene in the airborne (unmodified and modified ERA 5) data.

Furthermore, this effect is not only analysed for SAFs, which are already used but also for other kinds of fuels, such as methane and liquid hydrogen. Therefore, the relative increase in the number of cases for which persistent contrails are possible is examined for these fuels, which have a considerable higher energy-specific emission index for water vapour, which is 0.029 kg/MJ for kerosene, 0.045 kg/MJ for methane and 0.075 kg/MJ for hydrogen (IPCC, 1999). So, the G factor increases by 55 % (for methane) to more than 158 % (for liquid hydrogen). Here, the same overall propulsion efficiency is assumed, since in the transition to methane and liquid hydrogen the change in the overall propulsion efficiency is probably not too large (see Gierens, 2021).

For methane ≈ 7.1 % (5.0 %, 5.7 %) and for hydrogen-powered aircraft ≈ 9.1 % (6.0 %, 7.1 %) more cases with persistent contrails compared to the reference case kerosene can be observed in the airborne (unmodified and modified ERA 5) data. But in these cases, T_a often exceeds −38 °C so that droplets formed initially will not necessarily freeze. However, since our data set already ends at 310 hPa (lowest altitude), we cannot evaluate this temperature threshold.

4 Discussion, summary, and conclusions

In this paper, we have analysed 10 years of airborne and reanalysis temperature and humidity data to see, how much more persistent contrails would be formed if kerosene was replaced with alternative fuels of different energy-specific vapour emission indices. The theoretical

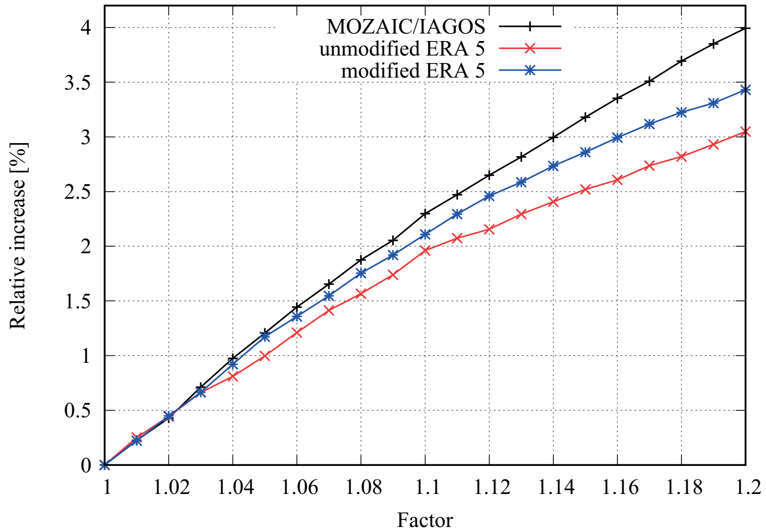

Figure 2: Relative increase of cases with persistent contrails with a relative increase of G (with the factor indicated) for airborne data from MOZAIC/IAGOS (black), unmodified (red), and modified (blue) ERA 5 reanalysis data. The point (1, 0) represents the kerosene case while points at higher G represent alternative fuels with a higher energy-specific emission index for water vapour.

expectation is that alternative fuels lead to earlier (that is slightly lower and warmer layers in the troposphere) contrail formation and also lead to slightly more persistent contrails than kerosene.

For this purpose, we have used humidity and temperature data from MOZAIC/IAGOS on one hand and corresponding ERA 5 (interpolated to the position of the measuring aircraft) on the other hand and computed the number of cases where persistent contrails would have formed, assuming $\eta = 0.35$ and an energy-specific water vapour emission index (EI_{H_2O}/Q) for kerosene. In order to repeat this calculation for alternative fuels, we multiplied the original G-values with factors 1.01 to 1.20. It turns out that indeed up to almost 4.0 % (3.0 %, 3.4 %) more persistent contrails occur when alternative fuels are used in the case of MOZAIC/IAGOS (unmodified and modified ERA 5). In any case, the increase is small for drop-in fuels, for which the energy-specific water vapour emission index increases only by a few percent.

But for other fuels, such as methane or liquid hydrogen, the effects become greater because the G factor increases by 55 to more than 158 % for these kinds of fuels. Therefore, ≈ 7.1 % (5.0 %, 5.7 %) more persistent contrail cases could be found for methane compared to kerosene, and ≈ 9.1 % (6.0 %, 7.1 %) more for liquid hydrogen in the MOZAIC/IAGOS (unmodified and modified ERA 5) cases. But contrails will probably not persist if T_a exceeds −38 °C. The increased occurrence of persistent contrails with alternative fuels in this study is consistent with modelling results for biofuels (Caiazzo et al., 2017) and sustainable alternative fuels (Teoh et al., 2022b). Caiazzo et al. (2017) assumed biofuels with an energy-specific emission index about 9 % higher than our standard kerosene case. They simulated the formation of contrails over the United States under various emission scenarios and observed an increase in the occurrence of contrails by about ≈ +8 %, a much larger value than we obtained from the MOZAIC/IAGOS data. However, it is not clear how far their numbers and ours are comparable since it is not clear whether they counted contrails or evaluated contrail total length or coverage. This is however important. Teoh et al., 2022b, with about an 8 % higher energy-specific emission index, made this important distinction: they found a 5 % increase in total contrail length, but only a 1.6 % increase in the number of persistent contrails. This latter number is almost identical to our result. As mentioned above, we use only about one percent of the data records in order to avoid autocorrelation. In this way, our result is closer to contrail counting than to measuring their length, for which all data along a flight would have been required.

An increase in contrails would imply a higher climate impact at first glance. However, it would be premature to draw any conclusions about their climate impact only based on the frequency of their occurrence. Climate impact does not only depend on the degree of coverage, or the frequency of this phenomenon, but it also depends on the microphysical and in particular on the optical properties of persistent contrails.

Contrail formation of alternative fuels occurs already at higher temperatures, as we saw, but these contrails are optically thinner than kerosene contrails, their crystals are on average larger, and thus, the lifetime of such contrails is on average shorter than the lifetime of kerosene contrails. It seems that these effects more than outweigh the slightly higher formation and persistence occurrences so eventually, contrails from alternative fuels are less climate-affecting than contrails from kerosene (Bock and Burkhardt, 2019; Teoh et al., 2022b).

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