



Opinion: Eliminating aircraft soot emissions

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1 Abstract

2 Soot from aircraft engines deteriorates air quality around airports and can contribute to climate change primarily 3 by influencing cloud processes and contrail formation. Simultaneously, aircraft engines emit CO₂, nitrogen 4 oxides (NOx) and other pollutants which also negatively affect human health and the environment. While urgent 5 action is needed to reduce all pollutants, strategies to reduce one pollutant may increase another, calling for a 6 need to decrease, for example, the uncertainty associated with soot's contribution to net Radiative Forcing (RF) 7 in order to design targeted policies that minimize the formation and release of all pollutants. Aircraft soot is 8 characterized by rather small median mobility diameters, $d_m = 8 - 60$ nm, and at high thrust, low (< 25%) 9 organic carbon to total carbon (OC/TC) ratios while at low thrust the OC/TC can be quite high. Computational 10 models could aid in the design of new aircraft combustors to reduce emissions, but current models struggle to 11 capture the soot $d_{\rm m}$, and volume fraction, $f_{\rm v}$ measured experimentally. This may be in part due to 12 oversimplification of soot's irregular morphology in models and a still poor understanding of soot inception. 13 Nonetheless, combustor design can significantly reduce soot emissions through extensive oxidation or near-14 premixed, lean combustion. For example, lean premixed prevaporized combustors significantly reduce emissions 15 at high thrust by allowing injected fuel to fully vaporize before ignition while low temperatures from very lean 16 jet fuel combustion limit the formation of NOx. Alternative fuels can be used alongside improved combustor 17 technologies to reduce soot emissions. However, current policies and low supply promote the blending of 18 alternative fuels at low ratios (\sim 1%) for all flights, rather than using high ratios (> 30%) in a few flights which 19 could meaningfully reduce soot emissions. Here, existing technologies for reducing such emissions through 20 combustor and fuel design will be reviewed to identify strategies that eliminate them.

21 1. Introduction

22 Aviation is a growing industry with a significant impact on human health and the environment due to the 23 emission of combustion by-products, including soot aerosols. The latter is one of the most important contributors 24 to climate change (Bond et al., 2013) and a component of air pollution known to cause cancer, cardiovascular 25 and respiratory diseases, and it has been correlated with various other illnesses (Niranjan and Thakur, 2017). 26 Regulations around the world have been limiting soot emissions since the 1970s. The International Civil 27 Aviation Organization (ICAO) until recently limited only the 'smoke number', essentially visible black smoke 28 from aircraft engines which caused dangerous reductions in visibility around airports (George et al., 1972). 29 Modern engines have no visible smoke but still produce invisible nanoparticles (Durdina et al., 2017). In 2020, 30 smoke number was replaced with a limit on the mass concentration of non-volatile Particulate Matter (nvPM) 31 and in 2023 an additional limit was placed on the number concentration of nvPM for all new engines with a rated 32 thrust greater than 26.7 kN (ICAO, 2017). Thus, jet engine manufacturers must design new engines to meet the 33 new nvPM standards without exceeding the regulations limiting nitrogen oxides (NOx), unburned hydrocarbons 34 (UHC) or carbon monoxide (CO) emissions while still maintaining strict safety standards. These regulations are 35 aimed at improving local air quality, so engines are assessed based on a standardized landing and take-off (LTO) 36 cycle most relevant for emissions near the ground.

37 Soot emissions can impact the climate by warming the atmosphere through direct Radiative Forcing 38 (RF) and indirectly by altering cloud processes and decreasing snow albedo (Bond et al., 2013). Aviation is 39 unique in that it emits soot at high altitude with very different atmospheric conditions (e.g., temperature and 40 pressure) from those on the ground. This may influence the formation of contrails (Kärcher, 2018). Lee et al. 41 (2021) estimated the climate forcing contribution of CO₂, contrail cirrus, NOx, soot aerosols, SO₂ aerosols and 42 water vapor from aviation in 2018. By these estimates, contrails account for 57.4 mWm⁻² or 55% of aviation's 43 net radiative forcing but with 95% confidence intervals from 27 - 67% of the net RF illustrating the high 44 uncertainty. The exact RF of contrail cirrus depends on the atmospheric conditions along the flight track and 45 time of day. At night, contrails have an exclusively warming effect while during the day there can be a warming 46 and a cooling effect (Stuber et al., 2006).





47 The estimate of direct RF from soot was relatively low, 0.9 mWm⁻² (Lee et al., 2021). However, 48 inventories of global soot emissions from aircraft can vary by two orders of magnitude (Agarwal et al., 2019). 49 Present inventories are based on the LTO cycle which focuses on landing and take-off at sea-level rather than 50 high-altitude cruise. As these emissions are measured only at ground level for the LTO cycle, the emissions most 51 relevant for climate considerations are only indirectly estimated (Stettler et al., 2013). In addition, the LTO cycle 52 does not exactly match the real time at each thrust for example, the LTO cycle assumes idle/taxi is 7% but real 53 aircraft use between 3 - 17% thrust for these conditions. (Masiol and Harrison, 2014). Estimates of the RF of 54 soot are from climate models which may underestimate the contribution of soot (Kelesidis et al., 2022). While 55 CO₂ remains in the atmosphere for 100 years or more, soot and contrails have short atmospheric lifetimes on the 56 order of a week (Bond et al., 2013) or hours (Bock and Burkhardt, 2016), respectively, so their global warming 57 potential is most important in the short term. This presents an opportunity to make immediate reductions in 58 global warming and 'buying time' for the implementation of technologies to lower CO₂ emissions (Montzka et 59 al., 2011). This may be important for the aviation industry which in 2022, adopted an ambitious goal of net-zero 60 carbon emissions by 2050.

61 These uncertainties highlight the importance of further research to better quantify the role of soot in 62 both contrail formation (Marcolli et al., 2021) and direct radiative forcing (Kelesidis et al., 2022). Such 63 uncertainties make it difficult to accurately assess priorities in emission reductions as there are often trade-offs 64 between emissions. For example, reductions in soot often result in an increase in NOx from diesel engines (Kim 65 et al., 2009). Similarly, contrail formation can be avoided by diverting flights to airspace with unfavorable 66 conditions for contrail formation (e.g. warmer temperatures) but may result in higher fuel consumption and, thus, 67 CO₂ emissions (Teoh et al., 2020). The large uncertainty associated with the contribution of soot to climate change is in part due to the oversimplification of soot morphology in climate models which typically assume 68 69 soot to be coated spheres. In reality, soot is an agglomerate composed of polydisperse primary particles (PP), 70 illustrated in Figure 1, with a nanostructure of layered graphene sheets (Fig. 1: inset).

71 The relative amounts of Organic Carbon (OC) or Elemental Carbon (EC) compared to the Total Carbon 72 (TC) is typically used to quantify the chemical composition of the particles. The OC is

73 defined by the ICAO as "...carbon volatilized in Helium while heating a quartz fiber filter sample to 870 °C 74 during thermal optical transmittance analysis including char formed during pyrolysis of some materials". 75 Conversely, EC is "...light absorbing carbon that is not removed from a filter sample heated to 870 °C in an inert 76 atmosphere during thermal optical transmittance analysis, excluding char" (ICAO, 2017). So, while quantifying 77 OC/EC ratios is important for understanding the light absorption of soot (Kelesidis et al., 2021), the split 78 between EC and OC is method-dependent (Cavalli et al., 2010) rather than a discrete property.



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Figure 1: A schematic of a soot nanoparticle highlighting commonly quantified properties which are relevant for 81 assessing the health and climate impact of such particles including the mobility diameter, $d_{\rm m}$ (broken line), primary 82 particle diameter, $d_{\rm p}$ (red solid line) and Organic Carbon (brown shaded area) to Total Carbon ratio, OC/TC. The 83 84 inset shows a high-resolution transmission electron micrograph (HRTEM) of a soot primary particle, from enclosed spray combustion of jet fuel produced at an equivalence ratio of 1.25 (Trivanovic et al., 2022), where the individual 85 graphene layers can be seen. Volatile compounds that may be adsorbed on the surface usually evaporate under the 86 vacuum of the microscope so cannot be visualized easily with HRTEM.





87 The size of irregular agglomerates such as soot is quantified by equivalent diameters such as the 88 mobility diameter, d_m (Fig. 1: broken line). Using a realistic soot morphology rather than equivalent spheres in 89 climate models increases the estimated direct RF by 20% on average revealing large direct RF = 3 – 5 W/m² in 90 hot spot earth regions, in line with field observations (Kelesidis et al., 2022).

91 Furthermore, limited access to real jet engines has made it difficult to assess the efficiency of soot to act 92 as ice condensation nuclei (ICN) and thus to enhance contrail formation. To date, experiments on the ICN 93 activity of soot have been done primarily using commercial carbon blacks or miniCAST soot generated by 94 burning hydrocarbon gases (Gao et al., 2022). MiniCAST particles tend to have much larger $d_{\rm m}$ (> 100 nm) than 95 that produced by real aircraft (< 100 nm) if the organic carbon to total carbon ratio (OC/TC) is sufficiently small 96 (Durdina et al., 2016). Recently, enclosed spray combustion of jet A1 fuel has been shown to be a promising 97 laboratory surrogate for aircraft soot produced at high thrust (i.e. cruise) with sufficiently small d_m and OC/TC 98 (Trivanovic et al., 2022). This is important for the calibration of optical instruments which may be sensitive to 99 the OC/TC ratio in addition to particle morphology (Durdina et al., 2016).

100 Technology for battery-electric or hydrogen-powered planes will not be available in the short-to-101 medium term for long-haul flights (Schäfer et al., 2019). Significant investment in airport infrastructure would 102 be needed to accommodate such changes in technology (Agnolucci et al., 2013). Emissions from aviation need to 103 be addressed urgently to meet climate goals and prevent further health degradation and mortality from air 104 pollution. However, aircraft engines have many competing demands including continued reduction of gaseous 105 emissions, CO₂ net-zero goals, safety requirements and regulations on noise. Thus, a firm understanding of the 106 environmental and health impacts of soot as well as a fundamental understanding of its formation and growth in 107 aircraft engines is essential for weighing the costs and benefits of mitigation strategies. The regulatory term 108 nvPM refers to particles that remain solid when heated to 350 °C. In aircraft emissions, this is primarily soot and 109 concentrations are measured with instruments designed for soot with a low OC/TC ratio (Lobo et al., 2015b) so 110 the terms nvPM and soot will be used interchangeably. Regulations on aircraft emissions apply only to turbofan 111 and turbojet engines with rated thrust > 26.7 kN. As such, most scientific research has been conducted on 112 engines in this category and will also be the category discussed in this paper. However, it is worth noting that 113 small business jets with thrusts < 26.7 kN may produce more nvPM emissions than large aircraft such as the 114 Boeing 737 which do fall under the ICAO regulations and need further research for accurate emissions 115 inventories (Durdina et al., 2019). In addition, while the European Union (EU) voted to ban leaded aviation 116 gasoline (Avgas) used in small piston-engine aircraft in 2022, most other countries still allow its use and it is 117 now considered one of few major sources of ambient lead in the US (National Academies of Sciences 118 Engineering and Medicine, 2021). Possible mechanisms for the formation and dynamics of soot from regulated 119 jet engines will be discussed. Then, strategies already in use or under development for the elimination of jet 120 engine soot emissions will be reviewed.

121 2. Formation and dynamics of aircraft soot

122 Although aircraft combustor design can vary significantly, the soot produced by aircrafts have some 123 morphological and compositional differences from other sources such as diesel engines. Aircrafts tend to 124 produce soot with median d_m in the range of 8 (Durdina et al., 2021) to 60 nm (Abegglen et al., 2015). Such 125 small d_m are associated with greater lung deposition efficiency (Rissler et al., 2012) and translocate from the 126 lungs to other organs more effectively than particles with $d_m > 100$ nm (Cassee et al., 2013). The OC/TC tends to 127 be quite low (< 25%) (Marhaba et al., 2019) when the aircraft operates at high thrust (> 50%) while the reverse 128 is true at low thrust. The OC/TC influences the optical properties of soot and thus its RF (Kelesidis et al., 2021). 129 Aircraft soot has PP diameters, d_p (Fig. 1: solid line), from approximately 5 (Liati et al., 2019) up to 24 nm with 130 lower thrusts tending to produce smaller d_p (Liati et al., 2014) which influences soot reactivity (Messerer et al., 131 2006) and optical properties (Kelesidis et al., 2020). These same properties are also influenced by PP 132 nanostructure which is related to their maturity (Baldelli et al., 2020). Aircraft tend to produce rather disordered 133 soot with a turbostratic structure with more defects on its surface than the bulk (Parent et al., 2016). The 134 conditions under which soot forms determine its final morphology and composition and vice versa (Vander Wal 135 et al., 2010).

Figure 2 depicts the cross-section of a single annular aircraft combustor (SAC), one of the common combustor designs in modern engines. The combustor is typically an annular tube that receives high pressure air from the compressor, adds energy to the system through combustion and uses it to drive the turbine. Liquid jet fuel is injected at one end of the SAC, typically with a swirling mechanism to atomize the fuel, promoting evaporation. However, perfect mixing is not achieved. So locally fuel-rich pockets allow for soot formation even if the global mixture is fuel-lean. Where the fuel is injected, there is significant recirculation allowing soot to grow in these fuel-rich pockets (Gkantonas et al., 2020). When there is insufficient oxygen for complete





143 conversion to carbon dioxide, fuels decompose into radicals and intermediate species, such as acetylene which 144 then grow into small aromatics (Wang, 2011). These aromatic compounds eventually evolve into polycyclic 145 aromatic hydrocarbons (PAHs) which are the key gaseous precursors to soot (Frenklach, 2002). The presence of 146 these soot precursors has been confirmed experimentally with atomic force microscopy (Commodo et al., 2019). 147 With respect to aviation, experimental studies have shown a correlation between jet fuel aromatic content, 148 sooting tendency (Yang et al., 2007) and nvPM emissions (Brem et al., 2015). So, fuel composition plays a key 149 role in the formation of soot and thus provides one possible route for its elimination as discussed in detail in the 150 next section.





155 Although the exact mechanisms of soot nucleation (i.e. the transition from the gas to solid phase) are still an 156 area of active research (Carbone et al., 2023), the dynamics of soot inception (Sharma et al., 2021) and growth 157 from nascent to mature soot (Kelesidis et al., 2017b) leading to its final structure are becoming better 158 understood. Nascent soot particles are as small as dm ~ 2 nm (Camacho et al., 2015), amorphous (Commodo et 159 al., 2017) and liquid-like (Kholghy et al., 2013) with a carbon to hydrogen (C/H) ratio < 2 (Schulz et al., 2019). 160 As they age, nascent soot carbonizes (lose hydrogen) and solidifies (Dobbins, 2002). Soot then simultaneously 161 undergoes surface growth and agglomeration (Kelesidis et al., 2017a). Surface growth of soot is well described 162 by the hydrogen-abstraction carbon-addition (HACA) mechanism (Frenklach, 2002) although other pathways 163 have also been proposed (Wang, 2011). During the first few milliseconds of particle growth, surface growth 164 precursors are depleted then agglomeration takes over as the primary growth mechanism and dm increases 165 markedly while d_p stays approximately constant (Kelesidis et al., 2017a). In the free molecular regime, particles 166 grow into large agglomerates through ballistic cluster-cluster coagulation while in the continuum regime this 167 becomes diffusion-limited cluster agglomeration. Particles which coagulate in the free molecular regime have a 168 slightly more compact structure than those in the continuum regime as shown by their asymptotic mass fractal 169 dimensions of 1.91 and 1.78, respectively (Goudeli et al., 2015).

170 This soot growth sequence has been observed and quantified for soot formation in premixed flames, diesel 171 engines, miniCAST soot generator (Kelesidis et al., 2017b) and even for enclosed spray combustion of Jet A1 172 fuel resulting in aircraft-like soot (Trivanovic et al., 2023). After primary air injection for the initial combustion, 173 dilution air is added at various locations along the combustor length. This oxidizes a sizable portion of the soot 174 which was initially created. Transmission Electron Microscopy (TEM) has shown that aircraft soot is 175 significantly oxidized and the small d_m may be in part due to fragmentation of larger agglomerates after 176 extensive oxidation (Vander Wal et al., 2014). So, in the early stages of the combustor the number and size of 177 soot is likely larger than what is eventually emitted. The final morphology of the particles, including the $d_{\rm p}, d_{\rm m}$ 178 and number of PPs per agglomerate, $n_{\rm p}$, depends on the initial volume fraction, residence time, temperature and 179 pressure (Kelesidis et al. 2023).

180 While conditions can vary significantly depending on the engine, soot in an aircraft combustor 181 experiences both high temperature and pressure. In addition, pressures are increased at high thrust which has 182 been correlated with increased soot concentration and size (Chu et al., 2023). Higher pressures improve the 183 efficiency of engines and so as engine materials have been improved to withstand higher pressures, the pressure 184 ratios in engines have also increased. So, soot may begin growing in the free molecular regime but enters the





transition regime as it grows, in particular at high thrust, when pressures are the highest and soot particles tend to grow to the largest sizes. This is in line with mass-mobility measurements of aircraft soot which shows an increase in the mass-mobility exponent, $D_{\rm fm}$, from 1.86 ± 0.37 to 2.79 ± 0.07 as thrust increases from 7 to 118%, respectively (Abegglen et al., 2015). However, mass-mobility measurements are not part of the regulatory framework for aircraft nvPM.

190 Low thrusts lead to the longest soot residence time in the combustor but tend to produce the smallest 191 particles both in terms of $d_{\rm m}$ and $d_{\rm p}$ which can be attributed to the smaller amount of fuel resulting in a lower 192 volume fraction of nascent soot (i.e. less nucleation) and allowing for a longer residence time in oxygen rich 193 zones which oxidizes the soot reducing both the number and size of particles (Durdina et al., 2014). At the same 194 time, the OC/TC increases at low thrust which could be attributed to the poor combustion efficiency at these 195 conditions. At high thrust the residence time is short but initial number concentrations are higher due to high fuel 196 flow. The time in oxidating zones is reduced also, resulting in a larger number concentration, d_p (Liati et al., 197 2014) and d_m (Abegglen et al., 2015). Simulations of aircraft combustors have shown that soot forms 198 intermittently in locally rich regions of the flame and, due to recirculation, soot spends 4-5 times longer in the 199 combustor than the fluid time scales (Chong et al., 2018b). The high-temperature residence time of soot in a 200 combustor can only be estimated from simulations that account for the geometry, fluid flow rates, temperature 201 and pressure in a given combustor.

202 Modeling soot emissions accurately remains a challenge (Chong et al., 2018a) because soot formation in 203 combustors is intermittent. So, simulations must take place over a large time frame to achieve a statistically 204 representative time-averaged result (Franzelli et al., 2023). Furthermore, the transport and chemistry of soot must 205 be solved simultaneously in order to capture the real volume fraction, f_v , and particle size distributions (PSD) 206 (Gkantonas et al., 2020). The most detailed simulations to date have utilized laboratory combustors such as the 207 Cambridge Rich Quench Lean (RQL) burner (Gkantonas et al. 2020, Fig. 2). These laboratory burners are 208 optically accessible for laser diagnostics allowing for a detailed comparison to the evolution of soot f_v and PSD. 209 However, the laboratory burners use ethylene, a gas, instead of liquid jet fuel and pressures are up to 5 bar 210 (Chong et al., 2018a). Modern aircraft engines may have pressures up to an order of magnitude higher than this 211 at certain conditions (Nguyen et al., 2019). Nonetheless, such simulations can give insight into the formation and 212 growth of soot in aircraft combustors capturing some of the trends observed experimentally. Specifically, 213 simulations show that soot forms near the shear layers between the fuel and oxidizer streams and then enters an 214 inner recirculation zone where it grows further (Gkantonas et al., 2020). Fuel rich pockets can also break off 215 from the main jet and become entrained in the recirculation zone driving the intermittent soot growth within the 216 combustor (Chong et al., 2018a). Soot was shown to grow by both acetylene-based surface growth (e.g., HACA) 217 and condensation via aromatics (Gkantonas et al., 2020). Simultaneously, significant oxidation reduces the 218 particle size and can induce fragmentation increasing the number concentration (Gkantonas et al., 2020) which is 219 supported by experimental data (Vander Wal et al., 2014). Introduction of dilution air part way through the 220 burner oxidizes soot in the lean combustion zone as well as lowers the rate of soot formation near the nozzle 221 (Chong et al., 2018a). Higher pressures in the model combustor result in larger soot f_v , a trend which was 222 captured by simulations but the total f_v for the high pressure condition was underpredicted by a factor of 4 223 (Chong et al., 2018a). Therefore, simulations can give insight into the formation of soot in aircraft combustors 224 but significant improvements are needed to have truly predictive models which can aid in combustor design 225 (Franzelli et al., 2023). It is worth noting that these simulations focus on capturing the number and mass 226 emissions from combustors, but do not seem to account for the realistic morphology of soot particles which are 227 highly irregular agglomerates rather than spheres. The assumption that soot is spherical rather than an 228 agglomerate with polydisperse primary particles can significantly change the resulting estimate of soot $d_{\rm m}$, 229 number and, most importantly, f_v (Kelesidis and Goudeli, 2021).

230 3. Means for the elimination of aircraft soot

231 3.1 Sustainable aviation fuels

232 Alternative fuels are attractive due to their unique position as a drop-in solution for reducing CO₂ emissions as 233 these fuels can be used directly in existing engines. The ICAO specifies fuels must be "completely 234 interchangeable and compatible with conventional jet fuel" in order to prevent the safety risks of mishandling 235 and high costs of additional infrastructure (ICAO, 2018). Sustainable Aviation Fuels (SAF) are produced from 236 biological feedstocks (e.g. soybeans, sugarcane, biomass, etc.) (Staples et al., 2018). These are converted into 237 liquid hydrocarbon fuels through processes such as Hydroprocessed Esters and Fatty Acids (HEFA), Fischer-238 Tropsch (F-T) or Alcohol-to-Jet (ATJ) to name a few (Brooks et al., 2016). Lower Carbon Aviation Fuels 239 (LCAF) or e-fuels use CO₂ capture and sustainable energy sources such as solar to produce synthetic jet fuels





240 (Schäppi et al., 2022). The CO_2 reduction from such fuels comes primarily from the synthetic or biological CO_2 241 captured during the production process. Actual CO₂ released from the engine remains about the same as 242 conventional jet fuel. So, a Life Cycle Analysis (LCA) is needed to account for the so-called Well-to-Wake 243 emissions (Han et al., 2013). The total reduction in Green House Gas (GHG) emissions will depend on both the 244 GHG emissions associated with production of the petroleum based jet fuel as well as the net GHG emissions 245 from growing, transporting and burning the SAF fuels. The ICAO certifies alternative fuels as SAF or LCAF 246 based on a standardized LCA. While the exact reduction in GHGs will change as technologies evolve, an LCA 247 of the best case scenarios show up to a 68% reduction in CO_2 emissions if SAFs account for > 85% of all 248 aviation fuels (Staples et al., 2018).

249 In addition to reducing net-CO2 emissions, SAFs and LCAFs also have the potential to reduce soot 250 emissions and thus the health impact and non-CO₂ radiative forcing of aircraft emissions which is typically 251 excluded from LCA analysis (Staples et al., 2018). These fuels tend to have a lower aromatic content than fossil 252 fuels which has been correlated to the number of particles emitted by an aircraft (Brem et al., 2015). As 253 discussed previously, aromatic species are key precursors to soot formation and thus a decrease in fuel aromatics 254 may reduce the rate of soot nucleation. The hydrogen-to-carbon ratio (H/C) of the fuel has been shown to have 255 an even greater anti-correlation with aircraft soot emissions than fuel aromatic content (Brem et al., 2015). While 256 H/C has long been associated with the sooting tendency of a fuel (Yang et al., 2007), the mechanism for this is 257 less clear as it is difficult to separate from effects such as lower flame temperatures (Xue et al., 2019). Blends of 258 a HEFA-based SAF with Jet A1 up to 50% (the current upper limit for a SAF blend) showed a ~35% reduction 259 in number based nvPM and ~60% reduction in mass based nvPM (Lobo et al., 2015a). These reductions 260 correlated best with the H/C content of the blends. The size distributions of the soot produced shifted to smaller 261 mobility diameters from $d_m = 49$ to 22.5 nm and narrowed the distribution from a geometric standard deviation, 262 $\sigma_{\rm g} = 1.99$ to 1.58 with pure Jet A1 and a 50% blend, respectively (Lobo et al., 2015a). With pure Jet A1, the $\sigma_{\rm g}$ 263 approaches that of the self-preserving limit for agglomerates coagulating in the free-molecular regime (Goudeli 264 et al., 2015) while the σ_g produced with the SAF blends are significantly smaller. This could be due to the 265 decreased number concentration from extended surface growth and less agglomeration. Currently, alternative 266 fuels are designed primarily with the goals of reducing life-cycle CO₂ emissions and matching the properties of 267 conventional jet fuels. However, there is an opportunity to also optimize jet fuel composition for minimum soot 268 emissions. Schripp et al. (2021) showed that different SAF could be blended to obtain a desired H/C, while 269 maintaining regulatory specifications for jet fuels. Soot emissions of these fuels were first tested in a laboratory 270 flame, then the optimal mixture was used in a real jet engine to confirm the trends seen in the laboratory 271 resulting in emission reductions of particle mass and number by 29 and 37%, respectively, when using a 38% 272 SAF blend with Jet A1 (Schripp et al., 2021). Laboratory tests are essential for speeding up the design of 273 alternative fuels since real jet engines are inaccessible to many researchers and too costly to operate for initial 274 screening tests. A standardized flame for assessing the sooting properties of jet fuels would assist in the 275 development of alternative fuels however, there is currently no standardized method for such experiments. 276 Enclosed spray combustion is a promising unit for such in lab approaches (Trivanovic et al., 2022).

277 Several publications have shown that the benefits of a SAF blend are thrust-dependent. For example, a 278 32% blend of HEFA-synthetic paraffinic kerosene and Jet A1 at idle operation showed a 60 and 70% reduction 279 in number- and mass-based nvPM, respectively (Durdina et al., 2021). The same blend at 65% thrust resulted in 280 only a 12% reduction in number-based nvPM and at take-off the reduction was only 7%. In this case, the use of 281 such SAF blends may improve local air quality by reducing emissions in the vicinity of airports but may not 282 make a significant impact on cruise conditions which are most concerning for climate change. It is worth noting 283 that the majority of studies on aircraft soot emissions are done at ground level which has significantly different 284 atmospheric conditions than cruise in the upper atmosphere. Ideally, cruise emissions should be measured behind 285 an aircraft in-flight, but this is rarely done due to the cost and logistical challenges. One of the few in-flight 286 studies comparing conventional jet fuel to a 50% HEFA blend showed a 50 and 70% reduction in particle 287 number and mass emissions, respectively, behind an aircraft with a medium thrust setting of ~ 50% (Moore et 288 al., 2017). At the high thrust setting, the particle number reduction was only 25% (Moore et al., 2017), 289 supporting the trend observed on the ground. The wide range of values listed here highlights the need for more 290 studies both at the ground level and in-flight.

291 Currently, SAF must be blended with conventional jet fuel (up to 50%) for safety reasons although 292 100% blends may be allowed by 2030. In practice, supply issues keep the use of SAF low accounting for an 293 estimated 0.1 - 0.15% of global jet fuel use in 2022 despite a tripling in the supply of SAF from 2021 to 2022. If 294 the SAF supply is limited and individual flights only have a very small fraction of SAF in the fuel, there will 295 likely be no effect on the soot emissions (Lobo et al., 2015a). So, while alternative fuels could provide a short-296 term solution to reducing aircraft emissions, the speed at which this is adopted is still limited. Targeted use of the





297 limited SAF supply could be used in the short term to maximize the benefits of such fuels while supply is 298 limited. For example, contrails with the greatest warming effect are commonly at dusk during the winter (Teoh et 299 al., 2022a) so fueling flights at such times with high SAF blends could have the biggest benefit. One analysis 300 found that compared to a 1% SAF blend for all transatlantic flights, fueling the 2% of flights producing the 301 highest RF with a 50% SAF blend could take the total RF reduction from 0.6% up to 6% (Teoh et al., 2022b). 302 The European Commission and the US have implemented policies to mandate the uptake of SAF which may 303 prohibit the targeted use of SAF. For example, starting in 2025 it will be required that "all aviation fuel supplied 304 to aircraft operators at (European) Union airports contains a minimum share of SAF" (European Commission, 305 2021). Thus, while the supply of SAF is limited, it will be used in more aircraft at lower blending ratios missing 306 an opportunity to reduce soot emissions. Intelligent changes to policy on the use of alternative fuels could thus 307 reduce the net-RF of aviation without needing to increase the supply of SAF and LCAF.

308 3.2 Aircraft Combustor Design & Operation

309 The limitations of alternative jet fuels highlight the continued need for improved and novel engine technologies 310 which could be used also with alternative fuels to minimize the total impact of aviation on the environment. 311 Here, only combustion engines will be considered as electric aircrafts are estimated to account for only a quarter 312 of all passenger-miles in 2050 (Prabhakar et al., 2022). Since nvPM regulations only recently came into effect, 313 most aircraft combustors are designed primarily to lower NOx, but some designs can also reduce soot. 314 Alternative fuels have not been shown to reduce NOx emissions compared to conventional jet fuel (Moore et al., 315 2017). Combustor designs must balance limits for all regulated gas and particulate emissions, fuel efficiency, safety and cost. Rich Quench Lean (RQL) combustors have been used by the aviation industry since at least the 316 317 1980s to reduce NOx emissions while maintaining sufficient combustion stability (Novic et al., 1983). Today, 318 they are the most common type of combustor listed in the ICAO emissions database (ICAO Aircraft Engine 319 Emissions Databank, 2023). Briefly, RQL combustors have three zones, depicted in Figure 3. First, there is a 320 fuel-rich zone that allows for more stable combustion which is important for the safety of the aircraft. Rich 321 conditions have lower combustion efficiency and promote the formation of soot, UHCs, and CO. In the 322 quenching zone, a large volume of cool air is injected to provide oxygen for completing the conversion of UHCs 323 and CO to CO_2 while lowering the temperature to minimize NOx formation. The air flow for the rich combustion 324 stage and quenching zone are controlled separately and further dilution air may be added before the gases are 325 sent to the turbine. Although the mixing and residence times in RQL combustors were originally optimized for 326 reducing NOx (Rizk and Mongia, 1990) proper design and operation can also reduce soot emissions through 327 oxidation during the lean burn stage. In fact, it was shown that a judicious injection of fresh oxygen in a manner 328 similar to RQL combustors can promote oxidation of soot removing up to 99.6% of the initial soot volume 329 fraction from jet fuel combustion (Kelesidis et al., 2023b). When quenching air is introduced farther downstream 330 in the combustor, soot has more time to form and grow. Hence, oxidation is less effective. Earlier injection of air 331 with sufficient turbulent mixing has the opposite effect, minimizing soot emissions (El Helou et al., 2021). 332 However, if quenching air is injected too early this could increase NOx emissions or reduce combustion stability.





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Figure 3: A simplified schematic of a Rich Lean Quench (RQL) aircraft combustor adapted from Rizk and Mongia 335 (1990) where there is first a fuel rich combustion zone, followed by a large flow of quenching air to lower the 336 temperature and dilute to a globally lean combustion zone. The dynamics of soot are qualitatively depicted from 337 inception to surface growth, agglomeration and oxidation.

338 In 1995, the first Double Annular Combustor (DAC) was used commercially. This combustor design 339 has two stages as the name implies, depicted in Figure 4. At low thrust (e.g., idle) only the pilot stage is used 340 with a low air to fuel ratio and low flowrate to ensure good ignition and to reduce CO and UHC emissions.



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341 When sufficiently high thrust is achieved, both the pilot and main stage are ignited with a high air to fuel ratio 342 (lean burn) and high flowrates (Boies et al., 2015). This had the desired effect of reducing the NOx emissions 343 over the LTO cycle by ~30% compared to a single annular combustor on the same engine (Mongia, 2007). Soot 344 emissions from a DAC equipped engine vary significantly with thrust. At low thrust, when only the pilot stage is 345 ignited, soot emissions are high, and increase with increasing thrust in both number and mobility diameter (Boies 346 et al., 2015). When both stages are ignited at thrust ~25%, the soot concentration and size drops significantly 347 (Boies et al., 2015). Similarly, a DAC using only the pilot stage showed an increased mass concentration of 348 organic particulate matter compared to when both stages were used (Lobo et al., 2015b). The morphology of soot 349 produced in both stages is in the range observed in other combustors.





As demonstrated by the low emissions of DAC when operated in the lean combustion mode, lean burn 353 354 engines have the potential for extremely low emissions if the combustion stability issues can be overcome. In 355 fact, lean combustion technologies typically produce an order of magnitude less soot than an RQL combustor 356 (Liu et al., 2017). Lean burn combustors were first developed for stationary gas turbines used for energy 357 generation where safety requirements are less strict and are now being transferred to aviation as technology 358 improves. Such technologies include Lean Direct Injection (LDI) or the Multipoint Lean Direct Injection concept 359 (MLDI) (Liu et al., 2017). Direct injection is used to reduce the risk of autoignition that comes with premixed 360 combustion. The use of multiple injectors, depicted in Figure 5, along with intense mixing creates conditions similar to lean, premixed combustion. In an LDI combustor a central pilot injector is surrounded by multiple 361 362 main fuel injectors with little to no dilution added after the initial air supply near the fuel injectors. The MLDI concept is similar to the LDI combustor with an altered injector layout. Globally lean combustion with good 363 364 mixing is unfavorable for soot production as there are few locally fuel-rich areas. At the same time, low 365 temperatures from the lean burn reduce NOx emissions significantly (Liu et al., 2017). Regulatory measurements



Figure 5: A simplified schematic of a Lean Direct Injection (LDI) combustor adapted from Fric (1995) which features
 a central pilot injector surrounded by multiple main injectors. These combustors usually have most or all the air flow
 into the combustor around the fuel injectors without subsequent dilution to provide intense mixing for lean
 combustion with close to premixed combustion.





of nvPM emissions from an LDI combustor show nvPM mass and number emission levels on par with RQL
combustors with similar rated thrusts (ICAO Aircraft Engine Emissions Databank, 2023). To the best of our
knowledge, no studies have characterized the size, morphology or chemical composition of soot from an LDI
equipped engine. The limited data for such combustors makes the real emissions performance of such an engine
difficult to assess.

376 Lean Premixed Prevaporized (LPP) combustors aim to completely vaporize the jet fuel prior to ignition 377 in order to have lean, premixed combustion (Figure 6). Without locally fuel-rich conditions, little to no soot will 378 form. As with the LDI combustors, there is little dilution after the initial injection of primary air for combustion. 379 Premixed combustion with high pressures comes with a risk of autoignition in the mixing zone so careful design 380 of the combustor is needed to prevent such instabilities. These combustors use special fuel injectors to achieve 381 near-premixed lean combustion conditions which tend to form significantly less soot. Both the LDI and LPP 382 combustor designs achieve stable combustion through complex combustor design which could lead to increased 383 cost and maintenance. So, lean conditions are favorable for emissions reduction but come with engineering 384 challenges. Theoretically, new jet fuels with lower lean blow-off (LBO) limits could extend the lean operating 385 range of an engine and conversely, fuels with an insufficient LBO could pose a safety risk (Undavalli et al., 386 2023).

387 Recently, a novel research engine called the Lean Azimuthal Flame (LEAF) combustor (not yet in 388 commercial use) using "flameless oxidation" has been developed for soot-free and low NOx combustion 389 (Oliveira et al., 2021). This concept can be further improved through co-combustion of small amounts of 390 hydrogen which extends the operating window (Miniero et al., 2023). The use of hydrogen helps to stabilize the 391 combustion without the use of a fuel-rich pilot flame that can increase soot production as with the DAC combustors. Such concepts which require an additional fuel that cannot be used in all engines require 392 393 significantly more capital to implement because additional infrastructure needs to be built to support, for 394 example, hydrogen storage and fueling. Furthermore, such parallel infrastructure poses a safety risk if an aircraft 395 is filled with the wrong fuel and therefore such solutions are not promoted by the ICAO (2018). So, combustors 396 which achieve lean, premixed conditions are promising for achieving both low soot and low NOx emissions but 397 pose design challenges.





Figure 6: A simplified schematic of a Lean Premixed Prevaporized (LPP) combustor adapted from Foust et al. (2012) which contains multiple injectors that spray fuel into the premix zone where the jet fuel completely vaporizes without ignition. Then, in the combustion zone the premixed fuel is ignited under fuel-lean conditions which nearly eliminate soot while low temperatures prevent the formation of NOx.

403 The ICAO provides a public database of regulated emissions with the earliest nvPM emission test dates 404 starting in 2014 (ICAO Aircraft Engine Emissions Databank, 2023). These data are collected and reported by the 405 engine manufacturers following the standards laid out in the ICAO Annex 16 for engine emissions certification 406 (ICAO, 2017). Emissions are tested across the entire LTO cycle which includes idle/taxi (7% thrust), approach 407 (30%), climb-out (80%) and take-off (100%) for both nvPM mass and number. Figure 7 shows the nvPM 408 number emissions normalized by the fuel flow (#/kg) at (a) idle/taxi and (b) take-off for simplicity, although 409 approach and climb-out data are also available (ICAO Aircraft Engine Emissions Databank, 2023). Mass nvPM 410 data shows similar trends. Values for approach and climb-out tend to fall between those measured at the 411 extremes for both number and mass nvPM. Combustor names are provided for all entries in the database and can 412 be grouped by type if sufficient information is given by the manufacturer. The RQL combustors make up the 413 majority of reported data (134 entries), followed by SAC (38), LPP (27), LDI (7) and DAC (2). The SAC 414 (squares) have some of the highest emissions in the database, but a group of SAC are approximately an order of





415 magnitude lower at idle/taxi (Fig. 7a). These lower emission SAC are modified for better performance (CFM 416 Tech Insertion) which seems to improve emissions at low (7 and 30%) thrust with little change at high (80 and 417 100%) thrust. The data for RQL combustors (circles) have the most variation quite likely due to the fact that 418 there are significantly more entries for ROL combustors compared to all other combustor types. This highlights 419 the fact that RQL burners can have quite low particulate emissions if designed and operated properly, 420 particularly for engines with lower static rated thrust. At take-off (Fig. 7b), the LPP combustors (triangles) 421 clearly outperform all other combustors in the database. At idle/taxi (Fig. 7a) LPP combustors still perform well 422 but some RQL and modified-SAC combustors have similar or lower emissions. In an LPP combustor, all 423 injectors are on during high thrust operation and premixed combustion can be achieved resulting in lower 424 emissions. Conversely, at low thrust only some of the injectors are used to lower the power output without 425 creating conditions which are too lean for stable combustion which may explain the higher emissions at idle/taxi 426 compared to take-off. A similar phenomenon has been observed in scientific studies of DAC engines where 427 emissions were reduced significantly when both combustor stages were in use at approximately thrusts > 30%428 (Boies et al., 2015) The small number of entries for DAC and LDI combustors makes it difficult to draw 429 conclusions about such combustors but the data that are provided for both fall in approximately the middle of the 430 nvPM emission range. So, at present LPP combustors seem to perform at least as well as other combustors at 431 idle/taxi and significantly reduce emissions at take-off resulting in the lowest overall emissions in the ICAO 432 database. It is worth noting that engine operation can also reduce emissions, for example reduced thrust take-off 433 has been shown to reduce fuel consumption, NOx and black carbon (soot) emissions by 1.0-23.2%, 10.7-47.7%, 434 and 49.0-71.7% respectively (Koudis et al., 2017).



435 436

Figure 7: The nvPM number as a function of an engine's rated thrust at (a) idle/taxi (7% thrust), (b) take-off (100%).
Combustor types represented in the database include SAC (squares), DAC (diamonds), RQL (circles), LDI (inverted triangles), LPP (triangles). The total nvPM number is normalized by the fuel flow (kg).

439 While the ICAO database provides information on the mass and number of nvPM emissions, it does not 440 include any morphological or chemical characterization of the particles. Furthermore, the data are collected by 441 the engine manufacturers, rather than independent researchers. Thus far, the vast majority of academic studies on 442 soot emissions from aircraft engines have been conducted on large commercial aircraft (rated thrust >26.7 kN) 443 most with SAC combustors (Liati et al., 2014; Beyersdorf et al., 2014; Marhaba et al., 2019; Parent et al., 2016; 444 Abegglen et al., 2015; Johnson et al., 2015; Elser et al., 2019). A few studies have explored soot from DAC 445 (Lobo et al., 2015b; Boies et al., 2015; Johnson et al., 2015) and RQL (Saffaripour et al., 2017; Delhaye et al., 446 2017; Brem et al., 2015) engines. The limited number of studies characterizing soot emissions from 'low 447 emission' engine technology highlights the need for more research on such engines if they will be adopted in the 448 future. Commercial deployment of new engine technologies takes a significant amount of time and money and 449 so, when a new technology is deployed it remains in use for many years with the life span of an average aircraft 450 spanning from 20 - 30 years (Ceruti et al., 2019). This makes it essential to identify which technologies offer the 451 best emissions profile before it is commercially scaled up, for example through the use of computational fluid 452 dynamics (CFD).





453 4. Conclusions

454 Soot from aviation has a negative effect on human health and can contribute to climate change through direct 455 radiative forcing and increasing the formation of persistent contrails. New regulations have been put into place to 456 limit soot emissions in addition to other pollutants such as NOx, UHC and CO. The strategies for reducing one 457 type of pollutant may increase another with soot and NOx emissions often at odds with one another. Non-CO₂ 458 aircraft emissions are estimated to be two thirds of aviation's net-RF, but the uncertainties associated with the 459 non- CO_2 terms are very high. The difficulty in reducing soot emissions from aviation comes primarily from the 460 competing requirements which include safety, reduction of gaseous pollutants and cost. A better understanding 461 of the role of soot and other non-CO₂ emissions is needed to properly assess trade-offs between design 462 requirements and avoid improving emissions of one pollutant while increasing another's or compromising 463 safety.

464 Aircrafts tend to produce soot with relatively small $d_{\rm m}$ which has greater health impacts than larger soot 465 particles. Soot nucleates in locally fuel-rich zones (created by the jet fuel spray) then grows through surface 466 growth, condensation and agglomeration. The OC/TC ratio of aircraft soot depends on thrust. Low thrust is 467 associated with high OC/TC and high with low OC/TC. Extensive oxidation reduces the number concentration 468 and size of soot resulting in smaller particles than other combustion sources (e.g. diesel). Significant progress is 469 still needed to accurately quantify this process in realistic aircraft combustors. Some progress has been made in 470 recent years matching experimental data from laboratory combustors but there are important differences between 471 laboratory combustors and real aircraft combustors and simulations are not yet able to match the output of these 472 simplified combustors at all conditions. The high cost and 20 - 30 year lifespan of aircraft necessitates robust 473 models to aid in combustor design and operation for further technological advancements.

474 Alternative fuels have the potential to significantly reduce soot emissions due to the lower aromatic and 475 H/C content typically associated with these fuels in addition to reductions in lifetime CO₂ emissions. Alternative 476 jet fuels can be created through processing of biological feedstocks (e.g., soybeans), typically referred to as 477 Sustainable Aviation Fuels (SAF), or through conversion of energy (e.g., solar electricity) into a liquid fuel, 478 typically referred to as Low Carbon Aviation Fuels (LCAF). Although most literature on the use of such fuels 479 does show that it reduces soot emissions, the reduction appears to be thrust dependent. So, it has the greatest 480 effect on reducing low-thrust emissions which are important for local air quality (e.g., idle) although modest 481 reductions have also been observed at high altitude cruising conditions. Several SAFs are approved for 482 commercial use but lack of sufficient supply makes it a tiny proportion of the global jet fuel supply (0.1-0.15% 483 in 2022). If SAFs are blended at small proportions with conventional jet fuel, the soot reduction benefits might 484 be hardly seen. Targeted use of high SAF blends on certain flights rather than low SAF blends for all flights 485 could be the best use of a limited resource. Supply issues likely will not be overcome soon, so policies 486 mandating the use of SAF and LCAF fuels should be designed in a way that encourages the use of a targeted 487 approach that will also lower soot emissions, not just life cycle CO₂.

488 Soot is primarily produced during fuel-rich combustion. So, throughout the years efforts have been made 489 to move toward fuel-lean combustion processes. The RQL combustors use a lean quenching stage after an initial 490 rich burn to ensure good combustion stability while still reducing NOx and in some cases soot. The design of the 491 quenching stage is essential for balancing combustion efficiency, NOx, and soot emissions from such engines. 492 The DAC combustors similarly take advantage of a pilot stage with low air to fuel ratios for combustion stability 493 at low thrust and a second main stage combustor which can be used at medium to high thrust for lean 494 combustion with a high air to fuel ratio. When both stages are in use, DAC combustors have very low soot 495 emissions but when only the pilot stage is used, soot emissions can be higher than in a traditional burner 496 particularly at medium-low thrust (e.g., ~20%). More recently, advances have been made on truly lean engine 497 technologies. This can be achieved either by using multiple injectors and high mixing rates to achieve nearly 498 premixed combustion or through mixing zones which allow for full evaporation of fuel before ignition. These 499 lean burn engines promise the lowest emissions of soot and NOx due to the lower temperatures and lack of fuel-500 rich zones. High complexity in such burners may result in higher maintenance costs. Finally, hydrogen can be 501 used to help stabilize lean combustion such as in the LEAF combustor which is both soot-free and low NOx but 502 is still under development in academic laboratories. However, the ICAO is discouraging such solutions which 503 require fuels that are not "drop-in" (e.g. hydrogen), as incompatibilities between engines and fuels could pose 504 safety risks and require significant capital investment in infrastructure.

The combined use of fuels with low sooting propensity and operating at lean combustion conditions have the potential to reduce or even eliminate soot emissions from aircraft engines. However, caution should be used whenever there is a trade-off with other emissions (i.e. NOx) as there is still significant uncertainty in the contribution of soot to direct RF and its role in contrail formation. The development of computational models





- 509 which can accurately predict soot production from various combustor designs and modes of operation will be 510 essential for minimizing soot emissions from aircraft while balancing other considerations. This will rely on 511 further fundamental research to better understand soot nucleation rates to close the soot mass balance and match
- 512 field data.
- 513 Competing interests
- 514 The contact author has declared that none of the authors has any competing interests.
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