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Gas Turbine Engine Non-volatile Particulate Matter mass emissions: Correlation with Smoke number for Conventional and Alternative Fuel Blends

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18 ABSTRACT

19 This study evaluates the relationship between the emissions parameters of smoke number
20 (SN) and mass concentration of non-volatile particulate matter (nvPM) in the exhaust of a gas
21 turbine engine for a conventional Jet A-1 and a number of alternative fuel blends. The data
22 demonstrate the significant impact of fuel composition on the emissions, and highlight the
23 magnitude of the fuel-induced uncertainty for both SN within the Emissions Data Bank, as
24 well as nvPM mass within the new regulatory standard under development. Notwithstanding
25 these substantial differences, the data show correlation between SN and nvPM mass
26 concentration still adheres to the First Order Approximation (FOA3), and this agreement is
27 maintained over a wide range of fuel compositions. Hence the data support the supposition
28 that the FOA3 is applicable to engines burning both conventional and alternative fuel blends
29 without adaption or modification. The chemical composition of the fuel is shown to impact
30 mass and number concentration as well as geometric mean diameter of the emitted nvPM,
31 however the data do not support assertions that the emissions of black carbon with small
32 mean diameter will result in significant deviations from FOA3.

33

34 INTRODUCTION

35 Emissions from aircraft gas turbine engines include the combustion products carbon
36 dioxide (CO₂) and water (H₂O), combustion by-products: oxides of nitrogen (NO_x) and
37 products of incomplete combustion: carbon monoxide (CO), unburned hydrocarbons (UHC)
38 and soot aerosol (or black carbon, BC). Each of these species are produced in different
39 relative proportions and all impact or contribute to climate forcing and degradation of air
40 quality^{1,2,3,4}.

41 The International Civil Aviation Organization (ICAO) sets regulatory standards for NO_x,
42 CO, UHC and Smoke Number, which are reported for all certified aircraft engine types
43 >26.7kN thrust in the Emissions DataBank⁵ (EDB). Emissions of BC are not currently
44 reported within the ICAO EDB, but may be inferred through the surrogate measurement of
45 Smoke Number (SN) at specific thrust settings that correspond to those used in the Landing
46 and Take-Off (LTO) cycle. SN is an optically based method that quantifies the change in the
47 reflectance of a Whatman #4 filter paper after sampling a fixed mass of engine exhaust per
48 unit area at a given temperature⁶. The ICAO regulation of SN was originally introduced in
49 1981 as means to quantify aircraft exhaust plume visibility and to act as a driver to reduce
50 emissions. SN does not provide a characterization of BC emissions in terms of mass and
51 number concentration, size distribution, or chemical composition, and given its proxy nature,
52 cannot be used to directly determine the environmental impacts of aviation. Currently, there
53 is an initiative within ICAO to replace the SN with a regulatory measurement methodology
54 for non-volatile particulate matter (nvPM) emissions for aircraft engines certified for use in
55 the commercial sector. In the meantime, SN remains the only measurement whereby BC
56 emissions can be estimated for environmental assessment activities.

57 A number of studies have reported the correlation between SN and mass concentration of
58 black carbon (C(BC)) using a range of different hardware: Champagne⁷ reports a correlation
59 derived from exhaust samples extracted from a combustor rig based on a T56 turboprop
60 engine; Whyte⁸ presented a method to convert between SN and C(BC) from a study of
61 kerosene alternative fuels; and Girling et al.⁹ report a correlation from an experimental study
62 using soot generated by a kerosene fuelled smoke generator amongst others^{10,11}. A critical
63 inter-comparison of these and other data, which agree to within 10%, was presented by
64 Wayson et al.¹². These correlations between SN and C(BC) form the basis of a method
65 endorsed by ICAO's Committee for Aviation Environmental Protection to estimate the mass

66 concentration and/or mass-based emission index of BC emitted from aircraft engines, referred
67 to as the ‘First Order Approximation¹³ version 3’ (FOA3). The FOA3 is intended for use as a
68 standard method to estimate PM mass-based emissions from certified commercial aircraft
69 engines within the vicinity of airports, and as an important assessment tool, there is a
70 commitment to improve FOA3 as new data become available until such time that the
71 methodology is rendered obsolete by a fully validated database of PM emission indices for
72 the commercial fleet.

73 Nevertheless, there has been criticism of FOA3, not least because of the potential for the
74 SN measurement to be dependent upon the capture efficiency of the filter and thus particle
75 size distribution of the emitted BC. This potential for error was first alluded to by Dodds et
76 al.¹⁴, followed by Wayson et al.¹², Sevcenco et al.¹⁵, and most recently discussed by Stettler et
77 al.¹⁶, though any suggestion that the SN underestimates C(BC) would mainly be applicable to
78 more recent engine technology due to the reduced mean diameter of the emitted BC.
79 However, as will be demonstrated and discussed later, the correlation between SN and C(BC)
80 remains a good first order approximation, even when the emitted BC particles have a mean
81 geometric diameter of the order of 20nm.

82 Within the emissions inventory and modeling communities, sources of uncertainty in
83 estimating the mass concentration of BC using SN values may arise when there is the need to
84 interpolate between data points at the four specific LTO thrust settings to determine
85 intermediate values, and also more generally, from the error in the reported SN data itself
86 induced by both measurement uncertainty and the use of non-standardized fuel for
87 certification tests on different engine types. Concerning this latter point, the hydrogen to
88 carbon ratio (H:C) and the aromatic content of the fuel used to produce the SN data for the
89 specific engine type and in the specific emissions certification test are (mostly) recorded
90 within the EDB. The spectrum of reported values in the EDB legacy data of 1.85 – 2.00 for

91 H:C ratio and 11.9% – 22.5% for aromatic content, covers a range that extends slightly
92 beyond the current recommended specification for fuel to be used in aircraft engine emission
93 testing of 1.85 – 1.99 and 15% – 23%, respectively¹⁷. Nevertheless, even the current
94 ‘tightened’ specification envelope allows for considerable variation in fuel properties, such
95 that the known impacts of fuel composition upon SN are ostensibly not considered. The
96 variation in the fuel properties reported within the ICAO EDB reveal that aromatic content
97 may vary by $\pm 3\%$ at a given H:C ratio, and H:C ratio may vary by ± 0.05 at a given aromatic
98 content. And whilst a decrease in aromatic content is generally associated with an increase in
99 H:C ratio, the correlation between these two parameters is generally poor and insufficient to
100 define the fuel.

101 Since the introduction of SN, engine technology has made significant progress and certified
102 SN’s at take-off power have decreased from the 25 – 35 range in early data, to values for
103 newer engine technology that typically occupy the 0 – 5 range. However, a SN of zero is
104 clearly a problem for the application of FOA3 in air quality and climate models as it implies
105 that the mass concentration of BC is also zero. For these reasons, ICAO has committed to
106 develop a new direct nvPM standard, but with typical engine lifetimes exceeding 20 years,
107 older legacy engines will continue to contribute to overall emission levels and so both SN and
108 FOA3 may not be fully transitioned for some years to come.

109 The new ICAO regulatory standard under development for the measurement of aircraft gas
110 turbine engine nvPM number and mass-based emissions uses the standard methodology
111 specified in the Society of Automotive Engineers (SAE) Aerospace Information Report
112 (AIR) 6241¹⁸. The development of this standard methodology for engine nvPM emission
113 measurement was born out of the Aircraft Particle Emissions eXperiment (APEX) campaigns
114 and many other similar studies^{19,20,21,22,23,24,25}. These studies highlighted the complexity of BC
115 emissions measurement, and in particular the difficulty in obtaining repeatable and reliable

116 measurement data. The data presented here have been obtained using the AIR6241 compliant
117 system North American Mobile Reference System that has been developed and robustly
118 characterized over several years through international collaboration²⁶.

119 The objective of this work was to compare SN measured using a SAE Aerospace
120 Recommended Practice (ARP) 1179d⁶ compliant system with the nvPM mass concentration
121 measured using the SAE AIR6241 compliant system for a conventional Jet A-1 and a number
122 of alternative fuel blends. In contrast to earlier FOA3 analysis where the correlation was
123 examined in terms of engine technology applicability, here we analyze the FOA3 correlation
124 from a fuel composition perspective. The gas turbine engine used in this study, a Garrett
125 Honeywell GTCP85-129 auxiliary power unit (APU), is not included within the EDB as its
126 rated output is <26.7kN. It is however a suitably close analogue to aircraft main engines that
127 provides a good model soot aerosol, and advances the methodologies previously used in the
128 development of the FOA3.

129 The chemical composition of the test fuels was managed by introducing various blends of Jet
130 A-1 and a Used Cooking Oil derived Hydrotreated Esters and Fatty Acids (UCO-HEFA)
131 kerosene. A complete range of fuels was investigated to allow full characterization of the data
132 within the FOA3 model, from low blend ratios (0 – 20%) that may be considered to be
133 essentially Jet A-1 variants, through to very high blend ratios that are distinctly paraffinic and
134 alternative in composition. Through this careful management of the fuel composition, it was
135 possible to vary SN in the range from 4 to >40, although the size distribution of the soot
136 aerosol is also present as a co-variable.

137 The significant impact of fuel aromatic content and/or fuel H:C ratio on nvPM emissions
138 and measured SN, is highly relevant to both the recommended specification for fuel to be
139 used in aircraft engine certification testing and the downstream effect on accurate emission
140 estimates due to regional variability in commercially available aviation fuel. Furthermore, the

141 potential impact of fuel compositional change becomes considerably more pronounced and
142 pertinent within the context of alternative fuels, and to the projected scale-up of sustainable
143 alternative aviation fuel use (eg. EU Flightpath 2020²⁷), together with future fuel
144 certification, fuel diversification, and long-term fuel security. Sustainable alternative fuels are
145 anticipated to play a sizeable role in decarbonizing the aviation industry, and currently there
146 are no methods to quantify the much-reduced atmospheric burden of BC that results from
147 their use²⁸. Any future update to FOA3 may need to incorporate a SN-fuel composition
148 response function.

149

150 BACKGROUND

151 Soot aerosol

152 Unfortunately the term ‘soot aerosol’ is rather imprecise in its definition, and terms such as
153 particulate matter, soot, black carbon, graphitic carbon, refractive carbon and non-volatile
154 particulate matter are often used synonymously. On occasions even the term carbon black is
155 used, even though this is distinct in that it is a manufactured product²⁹. Efforts to develop
156 precise nomenclature to distinguish between these terms are on-going, but these are often
157 based on particular measurement techniques or light-absorbing properties^{30,31,32,33} and lack
158 universal acceptance.

159 In recent years, the term black carbon (BC) has gained widespread usage within the climate
160 and emissions measurement communities, although it is recognized that BC is in itself a
161 generic term that describes a wide range of carbonaceous combustion derived substances
162 from partly charred residues to highly graphitized soot³⁴. BC particles have highly variable
163 physical properties and chemical compositions that very much depend upon their source^{35,36}.
164 Indeed the disparate nature of BC from different sources is well established and has even
165 been used in source apportionment studies. Physical properties such as size, morphology,

166 heterogeneity, surface area, isotopic ratio and density are all variable, as is chemical
167 composition with solvent extractable organic matter, and total carbon sometimes being
168 primarily elemental carbon (EC), but more often existing as complex mixtures of EC and
169 organic carbon (OC), with volatile and semi-volatile hydrocarbons, and other non-carbon
170 species such as ionic species, sulphates, moisture and trace metals^{37,38,39,40}.

171 Laboratory-generated ultrafine EC particles such as those created in a diffusion flame are
172 yet another distinct form of carbonaceous material. Overall, scientific studies need to clearly
173 distinguish between these highly disparate EC-containing particles with care and precision to
174 forestall the unwarranted extrapolation of properties and the transposition of inappropriate
175 study conclusions from one material to another. Black carbon from one combustion source is
176 not necessarily a model particle that is representative of the characteristics of an entirely
177 different combustion source.

178 The focus of this work is to evaluate the correlation between current and forthcoming
179 regulated measurement techniques using the soot aerosol emitted from a gas turbine engine
180 burning a conventional Jet A-1 and a number of alternative fuel blends. The precise bounds
181 and classification of the emitted soot aerosol is therefore operationally defined by the
182 measurement technique employed. Within this text, the term black carbon is used to define
183 the measurand associated with the measurement of smoke number through SAE ARP1179d,
184 whilst the term non-volatile particulate matter is used to define the measurand associated with
185 the measurement of mass concentration, number concentration and size distribution through
186 SAE AIR6241, although it is recognized that size distribution is not a formal part of this
187 standard. The term soot aerosol is used elsewhere in the broader discussion to represent less
188 defined states.

189

190 **Impact of fuel chemistry on soot aerosol formation**

191 Aviation Jet A-1 is a complex cocktail of thousands of different hydrocarbon component
192 molecules, though these molecules are often categorized into four principal groupings: n-
193 paraffins, iso-paraffins, cyclo-paraffins and aromatics⁴¹. The former two groupings of n- and
194 iso-alkanes typically dominate the class composition of all-fit-for-purpose petroleum derived
195 fuels⁴².

196 Variability in the chemical composition of Jet A-1 (and other kerosene specifications such
197 as Jet A, JP4, JP8, etc.) over both region and time is commonplace. The extent of this
198 variation is largely reflective of variability in the feedstock crude and localized demand for
199 other petrochemical distillation fractions. It is assessed on a regional level within fuel survey
200 data such as Rickard⁴³ or the Petroleum Quality Information System⁴⁴ (PQIS). Furthermore,
201 this diversity in the chemical composition of aviation kerosene is set to increase as alternative
202 fuels from a variety of sources enter the market as blend components or substitute fuels.

203 Perhaps the most notable impact of low aromatic kerosene fuels, including Jet A-1 blended
204 with Fischer-Tropsch (F-T) or Hydro-processed Esters and Fatty Acids (HEFA) alternative
205 fuels, is the very strong reduction in black carbon emissions^{45,46,47,48,49,50,51}. For example, the
206 Alternative Aviation Fuel Experiment (AAFEX) study using a CFM56-2C1 engine reported
207 concentrations of BC at the engine exit nozzle may be reduced by as much as 90% using F-T
208 fuels⁵⁰. These reductions affect the mass concentration, number concentration and size of the
209 emitted BC aerosol^{46,47,51}. A detailed evaluation of the impact of small variations in the Jet A-
210 1 / HEFA fuel blend ratio on the emission of nvPM is given in Lobo⁵². Evidence that the
211 reduction in soot aerosol occurs due to the lower aromatic content of the fuel is becoming
212 established, and aromatics are attributed as the class of compounds that primarily influence
213 the tendency to form BC and soot precursors during combustion^{42,45,53}. For example,
214 DeWitt⁴⁵ in an investigation of fuel composition, material compatibility and its relation to
215 emission characteristics showed that BC emissions increase with both increasing fuel

216 aromatic content and increased aromatic molecular weight when evaluated in a T63 turbo
217 shaft engine. This increase in BC emissions was attributed to an increase in soot precursors.

218

219 **FOA3: smoke number – mass concentration correlation model**

220 The FOA3 model endorsed by ICAO, is often used to predict the mass concentration of BC
221 in the exhaust emissions of a gas turbine engine from the surrogate smoke number
222 measurement¹³. Such data is routinely required by atmospheric modelers and for the
223 development of emission inventories.

224 For an engine with $SN < 30$, the mass concentration of BC (mg/m^3) is predicted from the
225 measured smoke number using the following FOA3 equation¹²:

$$226 \quad C(BC) = 0.0694 (SN)^{1.24} \quad (1)$$

227 Whereas for $SN > 30$, the mass concentration of BC (mg/m^3) is predicted from the measured
228 smoke number and using the following FOA3 equation¹²:

$$229 \quad C(BC) = 0.0297 (SN)^2 - 1.802(SN) + 31.94 \quad (2)$$

230 In both of these equations, $C(BC)$ is reported at standard temperature (273.15 K) and
231 pressure (101.325 kPa), and the bounds of uncertainty for the correlation are dominated by
232 the error in the measurement of the SN as errors in measurement of mass concentration are
233 small in comparison¹².

234

235 **EXPERIMENTAL METHOD**

236 **Gas turbine engine & operating conditions**

237 The Garrett Honeywell GTCP85-129 gas turbine engine used in this study is often operated
238 as an auxiliary power unit (APU) on Boeing 737 aircraft. APU gas turbine engines offer a
239 good model of aircraft main engine combustion characteristics whilst being considerably
240 more manageable and less costly to operate.

241 In this work, three APU operating conditions were investigated: No Load (NL),
242 Environmental Control Systems (ECS), and Main Engine Start (MES). These conditions
243 correspond to the normal operating conditions for an APU. For each experimental run the
244 APU was put through a warm up sequence using Jet A-1 before switching to the test fuel
245 without interruption, and then stabilizing at the first condition. The test matrix followed a
246 successive step down in power from MES to ECS to NL condition, which represented 1 test
247 cycle. For each of the fuel blends evaluated, this test cycle was twice sequenced without
248 shutdown. The sequence stepped down in power to minimize possible differences in
249 operating temperature and therefore potential differences in the fuel vaporization rates that
250 could feasibly manifest themselves as measurement uncertainties. For each engine condition,
251 the emissions data were recorded over a 6 minute window once the APU was determined to
252 be stable (ie. when engine EGT, RPM, and fuel flow were established as consistent).

253 The different fuel blends of Jet A-1 and Used Cooking Oil based HEFA (UCO-HEFA)
254 used for the study were selected at random to mitigate possible systematic bias and drift.
255 Experimental runs with Jet A-1 were conducted at the beginning and end of the study, as well
256 as several times in between runs with different fuel blends to reaffirm baseline conditions.
257 Engine parameters such as fuel flow rate, RPM, air fuel ratio, and exhaust gas temperature
258 were also recorded. The engine was very stable at each operating condition and the
259 reproducibility of engine parameters was good due to the on-board engine management
260 system.

261 Ambient conditions of temperature, pressure, and relative humidity were also recorded
262 throughout, and the range of values for these parameters was: 14.0 – 20.6 °C, 102.47 – 103.11
263 kPa, and 61 – 85%, respectively.

264

265 **Sampling system and instrumentation**

266 Two identical and almost collocated single-point probes, one for gaseous emissions and SN
267 measurement, and the second for nvPM emissions measurement were placed within $\frac{1}{2}$ nozzle
268 diameter of the engine exit plane (~15 cm).

269 The sample line for gaseous emissions and SN was compliant with the specifications in
270 ICAO¹⁷ Annex 16 Volume 2 and maintained at a temperature of 160°C. Gaseous species
271 were determined using a Binos Non-Dispersive Infrared Sensor (CO), a Signal Flame
272 Ionization Detector (UHC), and an Eco Physics Chemi-Luminescence Analyser (NO_x), each
273 using appropriate span and zero gases between measurements. The SN was determined in
274 accordance with SAE ARP1179d⁶ using a Richard Oliver smoke meter to collect at least
275 three filter samples for each fuel and at each engine condition. The reflectance of the filter
276 samples was determined pre and post sampling using a BOSCH reflectometer. Reported SN
277 data are the arithmetic mean of measurements from 6 filters (2 test cycles x 3 filters at each
278 condition) and uncertainty is conservatively estimated as ± 2 SN. This estimate of uncertainty
279 is consistent with the measured variability, with due recognition that the accuracy of an
280 individual SN measurement is considered to be ± 3 SN⁶.

281 The nvPM emissions were measured using the AIR6241 compliant North American mobile
282 reference system^{18,26}. The probe line used to extract nvPM emissions sample was connected
283 to a 3-way splitter using a 7.5 m long, 7.9 mm internal diameter thin-walled stainless steel
284 tubing maintained 160°C. The nvPM sample was diluted with particle-free nitrogen gas via a
285 Dekati ejector diluter and carried to the measurement suite along a 25 m long, 7.9 mm
286 internal diameter, carbon-loaded and electrically grounded PTFE tube maintained at 60°C in
287 accordance with SAE AIR6241. The nvPM number-based emissions were measured using an
288 AVL Advanced Particle Counter, while nvPM mass-based emissions measurements were
289 obtained using an Artium Laser Induced Incandescence and an AVL Micro Soot Sensor
290 (MSS). Only the nvPM mass data obtained using the MSS is used in this analysis. The

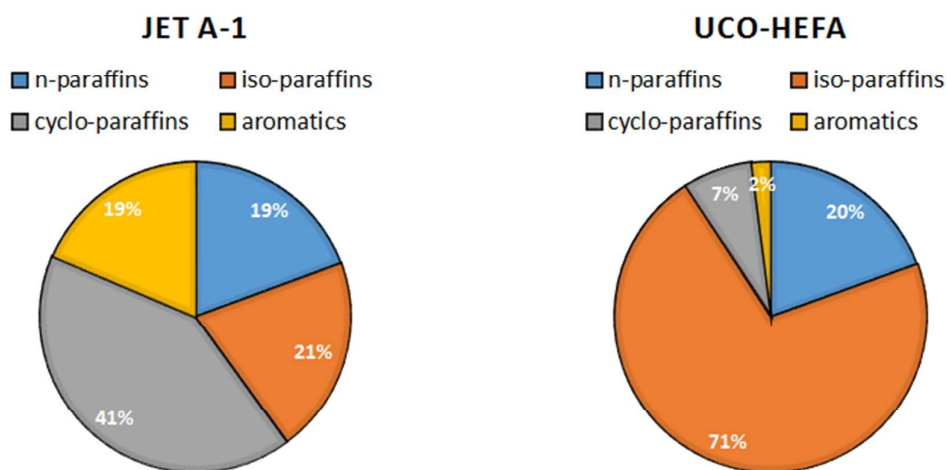
291 particle size distributions of the nvPM, which are not specified in AIR6241, were measured
292 using the Cambustion DMS500. The CO₂ concentration in the diluted nvPM line was
293 measured using a LiCor NDIR detector. The nvPM emissions data are reported at standard
294 temperature and pressure (273.15 K and 101.325 kPa), which is equivalent to mass
295 concentration data reported via FOA3. All nvPM emission concentration data was corrected
296 for dilution and thermophoretic loss in the sampling system. Measurement uncertainties in
297 nvPM emissions were calculated using 1 σ standard deviation of the average data.

298

299 **Properties of test fuels**

300 The two kerosene fuels used in this study were Jet A-1 and UCO-HEFA. The Jet A-1 was
301 straight-run kerosene obtained from Air BP (Kingsbury, UK), while the UCO-HEFA was
302 provided by SkyNRG (Amsterdam, NL). A GC x GC chemical analysis was used to quantify
303 the paraffinic and aromatic chemical composition of the two fuels, a summary of which is
304 shown in Figure 1. The figure shows the significant difference in the composition of the two
305 fuels: the Jet A-1 contains a substantial fraction of cyclo-paraffins and aromatics, whereas
306 these are much reduced for the UCO-HEFA fuel that is dominated by iso-paraffins.

307



308

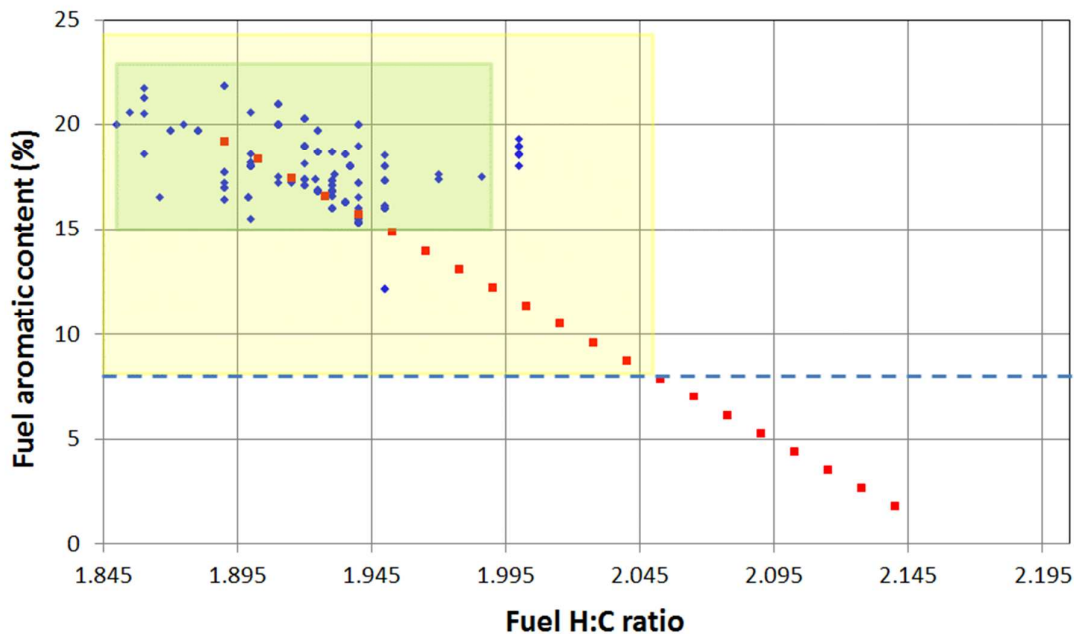
309 Figure 1. Summary of the GC x GC compositional analysis for the Jet A-1 and UCO-HEFA
310 kerosene fuels that were used to formulate the test blends.

311

312 A number of Jet A-1 / UCO-HEFA kerosene fuel blends were formulated in-house through
313 careful weighing and thorough mixing (blend ratios of 5%, 10%, 15%, 20%, 25%, 30%, 40%,
314 50%, 60%, 70%, 75%, 80%, 85%, 90%, and 95% by mass). The chemical composition of the
315 fuels varied linearly with fuel blend ratio, and test fuel H:C ratio varied from 1.89 to 2.14
316 whilst aromatic content correspondingly varied from 19.2% to 1.8% by mass. The Jet A-1
317 and UCO-HEFA fuels were fully miscible and the blended fuels were formulated at least 48
318 hours prior to use. It is recognized that several of these blends are outside of current ASTM
319 certification limits for HEFA fuel blends in operational aircraft, however these limits are no
320 longer applicable to the now ground based APU used within this study. Further details of the
321 fuel properties for neat Jet A-1 and UCO-HEFA fuels are given in Lobo et al.⁵².

322 By introducing the hypothetical concept of an aromatic - H:C ratio space, these fuels can be
323 compared with fuels in the EDB, a world survey of the available JP8 fuels, and the nominal
324 bounds for JP8 jet fuel. The specification for JP8, a military grade kerosene made to more
325 exacting specifications than commercial jet fuel, is used in this context as a proxy, since H:C
326 ratio is not defined for checklist Jet A-1. This comparison is shown in Figure 2.

327



328

329 Figure 2. Comparison of aromatic content and H:C ratio of different fuels: Blue points: EDB
 330 engine certification data; Red points: Experimental fuel blends; Green shaded area: Bounds
 331 of ICAO engine test fuel specification¹⁷; Yellow shaded area: Bounds of PQIS world JP8
 332 2013 survey⁴⁴; Chart area: Nominal bounds for JP8 jet fuel; Blue dashed line: ASTM D7566
 333 minimum aromatic limit⁵⁴.

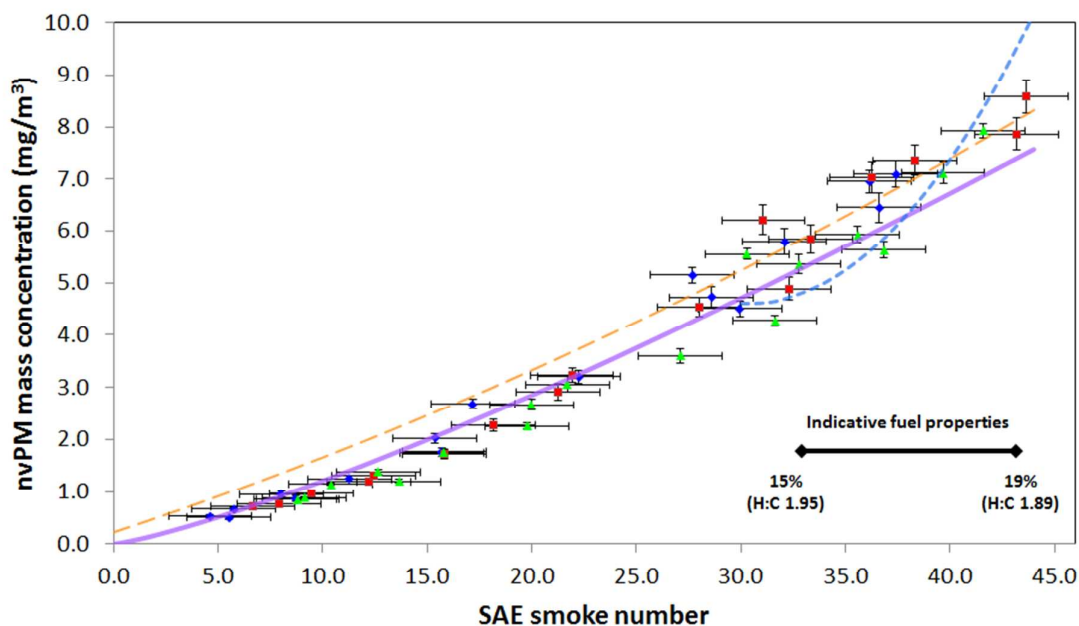
334

335 RESULTS AND DISCUSSION

336 Correlation between nvPM mass concentration and SN

337 Figure 3 shows the measured nvPM mass concentration, corrected for dilution and
 338 thermophoretic loss¹⁸, as a function of SN. The different colored data points in the plot
 339 indicate the three different engine conditions, and the BC mass concentration as a function
 340 SN predicted by FOA3 for both SN<30 and SN>30 are also overlaid.

341



342

343 Figure 3. nvPM mass concentration as a function of smoke number. Colored data points
344 indicate the three different engine operating conditions: Blue=MES; Red=ECS; Green=NL.
345 The C(BC) as predicted by FOA3 using SN data is overlaid: Purple line = FOA Equation 1
346 (nominally applicable for SN<30); Dashed blue line = FOA Equation 2 (nominally applicable
347 for SN>30); Dashed orange line = upper bound for Equation 1 generated using +3 SN error
348 [Wayson et al.¹²]. An indication of the change in fuel aromatic content (H:C ratio) for ECS
349 operating condition is inset.

350

351 The experimental data for the correlation between SN and nvPM mass concentration show
352 close agreement with FOA3, particularly at SN<30. Furthermore this agreement is
353 maintained over a wide range of kerosene compositions, and is largely independent of the
354 engine operating condition. Lines of regression for the datasets representing the three engine
355 conditions are practically coincident (not shown in the figure). The location of specific
356 emissions data on the FOA3 curve is merely dependent upon the chemical properties of the
357 fuel. Data points towards the left in Figure 3 represent measurements from fuel blends with

358 lower aromatic content and correspondingly greater H:C ratio. It is clear that the chemical
359 composition of the kerosene has a significant impact on the tendency to form nvPM. Fuel
360 aromatics have been identified as compounds that primarily influence the tendency to form
361 soot aerosol during combustion^{45,46}, although it has long been suggested that fuel hydrogen
362 content may be a more fundamental parameter that is independent of molecular structure^{58,59}.
363 The data presented here cannot be used to differentiate between the impact of aromatics and
364 the impact of H:C ratio since both vary linearly in the two component fuel blends.
365 Experimental data using multi-component blends or surrogate fuels to adjust these parameters
366 independently is necessary to explore their relative authority. The magnitude of the
367 reductions in SN and/or nvPM emissions are comparable with data reported elsewhere for
368 other gas turbine engines burning paraffinic fuels^{38,44,45,47,48}.

369 The nvPM mass concentration ($C(\text{nvPM})$) and BC mass concentration ($C(\text{BC})$) as defined
370 by their respective measurement methodologies are not identical, and generally $C(\text{nvPM}) \geq$
371 $C(\text{BC})$ since the former encompasses line loss correction factors that are not inherent in latter.
372 The two standards are however closely related and these data support the supposition that
373 $C(\text{nvPM})$ can be estimated from FOA3, but more significantly, that FOA3 can be used with
374 alternative fuel blends of varying chemical composition without adaption or modification.

375 The data indicate that the relation between $C(\text{nvPM})$ and SN is foremost represented by
376 FOA3 equation (1), even at $\text{SN} > 30$. Using all data points in Figure 3 and a power law fit to
377 be consistent with FOA3 equation (1), the line of regression is given by:

$$378 \quad C(\text{nvPM}) = 0.048 (\text{SN})^{1.35} \quad (3)$$

379 Whilst using a constrained range of data points up to $\text{SN} < 30$, the line of regression is given
380 by:

$$381 \quad C(\text{nvPM}) = 0.058 (\text{SN})^{1.27} \quad (4)$$

382 with the correlation coefficients of $R^2 = 0.979$ ($n=51$) and $R^2 = 0.965$ ($n=33$), respectively
383 (to simplify the representation of data, these lines of regression are not included in Figure 3).

384 Figure 3 also shows a marker to indicate the 15% - 19% fuel aromatic range for the ECS
385 engine operating condition (markers for other engine conditions are of comparable magnitude
386 but offset relative to the SN axis). This marker corresponds to the mid-range and the lower
387 bound for aromatic content in the ICAO specification for fuel to be used for aircraft engine
388 certification testing. For this modest shift in fuel composition, the SN decreased by 30% and
389 C(nvPM) decreased by 45%. Hence the fuel-induced uncertainty in EDB SN or C(nvPM)
390 derived through FOA3 is potentially twice this number, when considered in respect of the
391 limits of fuel used for engine certification testing¹⁷ and typical commercial fuel variability⁴³.
392 SN data for a particular engine in the EDB is strictly only correct for the stated certification
393 test fuel and will increase or decrease in magnitude for fuel of different chemical
394 composition.

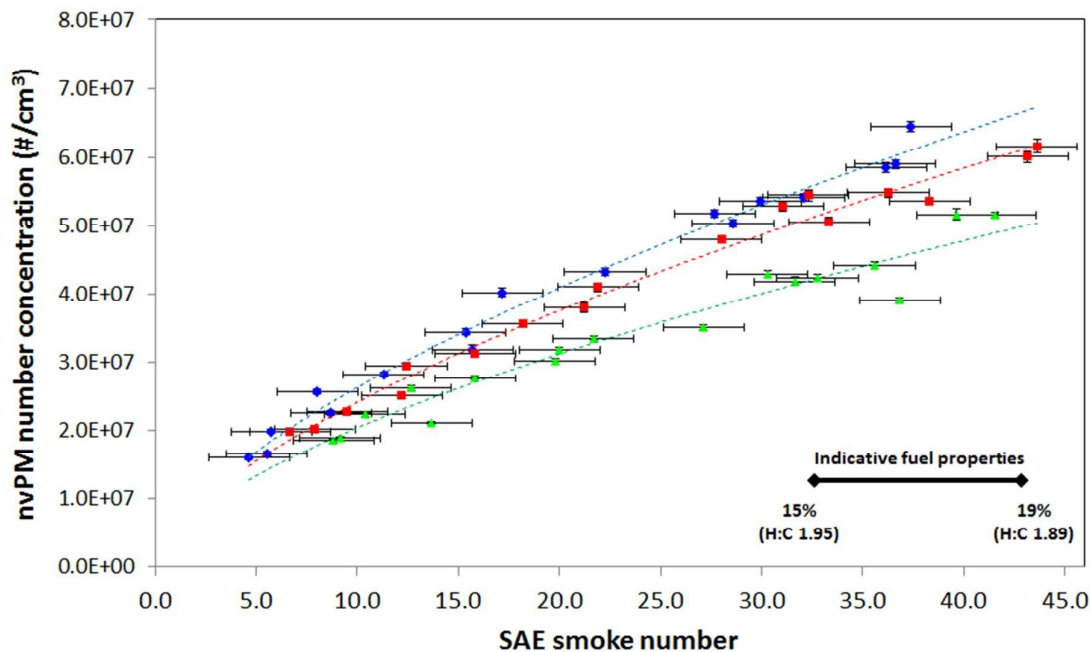
395 The data suggest that for engines with relatively large reported SNs the fuel-induced
396 uncertainty could be significant and markedly greater than the nominal ± 3 SN uncertainty
397 associated with the measurement of SN, whilst for engines with relatively small reported SNs
398 the fuel-induced uncertainty will be captured within this same ± 3 SN measurement
399 uncertainty. The proportional reduction in nvPM mass are consistent with data reported by
400 Brem et al.⁵⁵ in a study evaluating the impact of fuel aromatic content on nvPM emissions
401 from an in-production gas turbine engine.

402

403 **nvPM number concentration and size distribution**

404 Figure 4 shows the measured nvPM number concentration corrected for dilution and
405 thermophoretic loss¹⁸ as a function of the measured SN. Measurement uncertainties are as

406 previously described, and similarly the different colored data points in the plot indicate the
 407 three different engine conditions.



408
 409 Figure 4. nvPM number concentration as a function of smoke number. Colored data points
 410 indicate the three different engine operating conditions: Blue=MES; Red=ECS; Green=NL.
 411 An indication of the change in fuel aromatic content (H:C ratio) for ECS operating condition
 412 is inset.

413

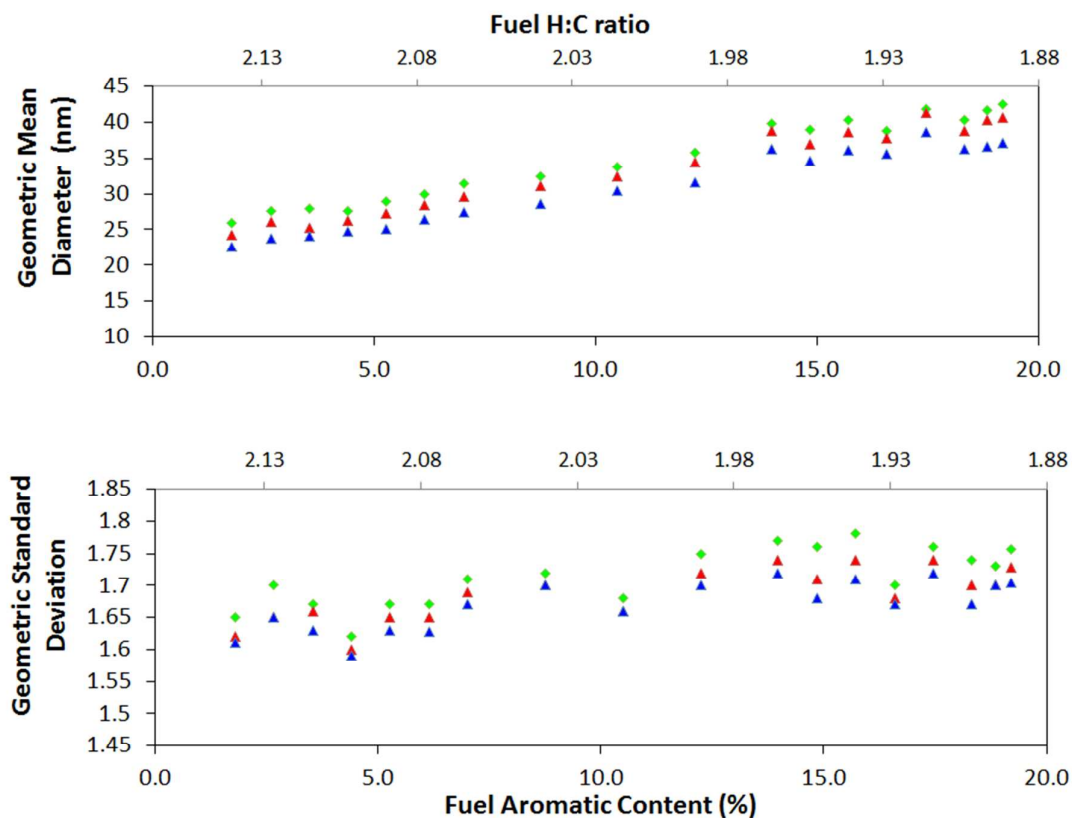
414 Data points towards the left in Figure 4 represent measurements from kerosene fuel blends
 415 of lower aromatic content and show a progressive reduction in the nvPM number
 416 concentration. In this case there is some distinction between lines of regression for the three
 417 datasets (shown in the figure) indicating that the relation between nvPM number
 418 concentration and SN may be dependent upon the engine operating condition.

419 Figure 4 also shows a marker to indicate the 15% – 19% fuel aromatic range for the ECS
 420 engine operating condition corresponding to the mid-range and the lower bound for aromatic
 421 content in the ICAO specification for fuel to be used in aircraft engine certification testing.

422 For this shift in fuel composition, SN decreased by 30% and the nvPM number concentration
423 decreased by 22%. This would suggest that nvPM number concentration is also a strong
424 function of fuel composition, an observation that is consistent with data reported
425 elsewhere^{52,55}.

426 The nvPM size distribution parameters of geometric mean diameter (GMD) and geometric
427 standard deviation (GSD) for the fuel blends tested at each of the three APU operating
428 conditions are shown in Figure 5. The nvPM exhibited a characteristic lognormal size
429 distribution, which narrows and shifts the geometric mean diameter to smaller sizes as the
430 aromatic content of the fuel blend is decreased (correspondingly increased H:C ratio). For a
431 given fuel, the succession of nvPM GMD tracked the sequence NL>ECS>MES. Overall the
432 GMD varied from a minimum of 22nm for 1.8% aromatic fuel in the MES engine condition
433 to 42nm for 19.2% aromatic fuel in the NL engine condition. The corresponding GSD ranged
434 from 1.58 to 1.79. Hence on the microscopic scale, the fuel-induced reduction in the mass of
435 emitted nvPM corresponds to the emission of fewer and smaller units of particulate matter.
436 These data are consistent with those reported for other gas turbine engines burning
437 conventional and alternative fuels^{21,26,47,49,50}.

438



439

440 Figure 5. Geometric mean diameter and geometric standard deviation for nvPM emission
 441 from selected fuel blends. Colored data points indicate the three different engine operating
 442 conditions: Blue=MES; Red=ECS; Green=NL. In both cases the upper secondary axis shows
 443 the corresponding fuel H:C ratio.

444

445 When these nvPM GMD data are considered in relation to the $C(\text{nvPM})$ in Figure 3, it is
 446 evident that the small nvPM with GMD $\sim 22\text{nm}$ that are characteristic of modern aircraft
 447 engine emissions, do not result in significant deviations in the FOA3 estimation of mass
 448 concentration. Previously Stettler et al.¹⁶ published data that appears to show that the relation
 449 between SN and $C(\text{nvPM})$ deviates significantly from FOA3 for nvPM with a GMD of the
 450 order of 20nm, with deviations of up to a factor 3. The data presented here do not support this
 451 finding. The data presented by Stettler et al.¹⁶ do not represent a fair comparison with the
 452 FOA3 method in three principal ways. Firstly, the applied methodology did not have a direct

453 measurement of nvPM mass concentration, but rather estimated it from size distribution and
454 effective density measurements. In the current work, we directly measure nvPM mass.
455 Secondly, Stettler's experiments were based on laboratory measurements of propane
456 diffusion flame combustion, and the black carbon generated from a propane burner is not a
457 model particle that is representative of the soot aerosol produced by a gas turbine engine.
458 Propane burners produce black carbon via a different mechanistic route (as chemically
459 dissimilar) that result in high EC fraction particulate matter with different physical and
460 chemical properties. This assertion is supported by experimental data from Durdina et al.⁵⁶.
461 Thirdly, the SN measurement methodology employed by Stettler was not comparable with the
462 methodology that has been used to populate the data in the ICAO EDB. The use of a catalytic
463 stripper to remove the semi-volatile OC from the line is not compliant with SAE ARP1179d⁶
464 and will result in a relatively 'clean' source of soot aerosol to be impingent upon the SN
465 filter. The impact of volatiles to the measurement of SN was demonstrated by Rye et al⁶⁰.

466 The data in Stettler et al.¹⁶ do demonstrate that a 'clean' black carbon from a propane
467 burner is captured with a progressively decreasing efficiency as the geometric mean diameter
468 is reduced. However, the extrapolation that these data are applicable to the emission of nvPM
469 from an aircraft gas turbine engine cannot be justified because of the differences in both the
470 modeled source for BC / nvPM and the measurement methodologies employed. This is
471 important since Stettler et al.¹⁶ claim that the FOA3 significantly underestimates aircraft
472 emissions of BC by a factor of 2.5 – 3 for SN ≤ 15 , and consequentially, propose a factor ~ 3
473 upwards revision of aircraft BC radiative forcing which would make it equivalent to $\sim 1/3$ of
474 the aviation CO₂ radiative forcing⁵⁷. On the basis of the measurements presented here and
475 critique of the Stettler et al¹⁶ methodology, such conclusions and extrapolations cannot be
476 supported.

477 Significantly, this work develops a comparative framework between current and future
478 regulatory standards for the measurement of soot aerosol from a gas turbine that incorporates
479 the quantitatively distinct emission from the combustion of alternative fuels, and places these
480 within the ICAO endorsed and widely accepted FOA3 methodology. With typical engine
481 lifetimes exceeding 20 years, older legacy engines will continue to contribute to overall
482 emission levels and so both SN as a surrogate measurement of BC, and FOA3 as a vital
483 assessment tool, may not be fully transitioned for some years to come. The importance of fuel
484 composition and the impact of its attendant variability may be particularly acute in the
485 application of EDB data to air quality modeling and the development of emission inventories.

486

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496

497 ABBREVIATIONS

498 BC, Black Carbon; C(BC), Concentration of Black Carbon; C(nvPM), Concentration of
499 non-volatile Particulate Matter; EDB, Emissions Data Bank; FOA3, First Order
500 Approximation (version 3); GMD, Geometric Mean Diameter; GSD, Geometric Standard
501 Deviation; ICAO, International Civil Aviation Organization; LTO, Landing and Take Off;
502 nvPM, non-volatile Particulate Matter; SN, Smoke Number; UCO-HEFA, Used Cooking Oil
503 derived Hydrotreated Esters and Fatty Acids.

504

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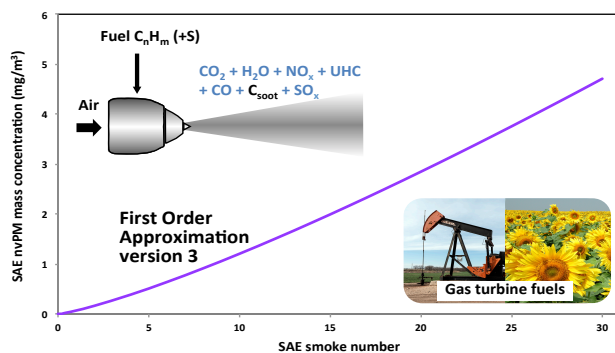
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