



- ¹ Aircraft-engine particulate matter emissions from conventional
- ² and sustainable aviation fuel combustion: comparison of
- ³ measurement techniques for mass, number, and size
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- ²¹ 0 AMT Feature: short summary (max. 500 characters incl. spaces)
- ²² The combustion of sustainable aviation fuels in aircraft engines produces
- ²³ particulate matter (PM) emissions with different properties than conventional
- ²⁴ fuels due to changes in fuel composition. Consequently, the response of various
- ²⁵ diagnostic instruments to PM emissions may be impacted. We found no significant
- ²⁶ instrument biases in terms of particle mass, number, and size measurements for
- ²⁷ conventional and sustainable aviation fuel blends despite large differences in the
- 28 magnitude of emissions.

29 1 Abstract

- ³⁰ Sustainable aviation fuels (SAFs) have different compositions compared to
- conventional petroleum jet fuels, particularly in terms of fuel sulphur and
- ³² hydrocarbon content. These differences may change the amount and
- ³³ physicochemical properties of volatile and non-volatile particulate matter (nvPM)
- ³⁴ emitted by aircraft engines. In this study, we evaluate whether comparable nvPM
- ³⁵ measurement techniques respond similarly to nvPM produced by three blends of
- ³⁶ SAFs compared to three conventional fuels. Multiple SAF blends and conventional

³⁷ (Jet A-1) jet fuels were combusted in a V2527-A5 engine, while an additional

³⁸ conventional fuel (JP-8) was combusted in a CFM56-2C1 engine.

- ³⁹ We evaluated nvPM mass concentration measured by three real-time sampling
- techniques: photoacoustic spectroscopy, laser-induced incandescence, and the
- extinction-minus-scattering technique. Various commercial instruments were
- tested including three LII 300s, one PAX, one MSS+, and two CAPS PM_{SSA}. Mass-
- ⁴³ based emission indices (EI_m) reported by these techniques were similar, falling
- within 30% of their geometric mean for EI_m above 100 mg/kg_{fuel} (approximately
- $_{^{45}}$ $\,$ 10 μg PM m $^{\cdot 3}$ at the instrument), this geometric mean was therefore used as a
- reference value. Additionally, two integrative measurement techniques were
- evaluated: filter photometry and particle size distribution (PSD) integration. The
- ⁴⁸ commercial instruments used were one TAP, one PSAP, and two SMPSs. These
- techniques are used in specific applications, such as on-board research aircraft to
- $_{50}$ determine PM emissions at cruise. EI_m reported by the alternative techniques fell
- si within approximately 50 % of the mean aerosol-phase El_m.





- ⁵² In addition, we measured PM-number-based emissions indices using PSDs and
- ⁵³ condensation particle counters. The commercial instruments used included TSI
- 54 SMPSs, a Cambustion DMS500, and an AVL APC, and the data also fell within
- ⁵⁵ approximately 50 % of their geometric mean. The number-based emission indices
- ⁵⁶ were highly sensitive to the accuracy of the sampling-line penetration functions
- ⁵⁷ applied as corrections. In contrast, the EI_m data were less sensitive to those
- ⁵⁸ corrections since a smaller volume fraction fell within the size range where
- ⁵⁹ corrections were substantial. A separate, dedicated experiment also showed that
- the operating laser fluence used in the LII 300 laser-induced incandescence
- instrument for aircraft engine nvPM measurement is adequate for a range of SAF
- ⁶² blends investigated in this study. Overall, we conclude that all tested instruments
- are suitable for the measurement of nvPM emissions from the combustion of SAF
 blends in aircraft engines.
- Keywords: non-volatile particulate matter, aircraft, emissions, sustainable
 aviation fuels, black carbon

67 2 Introduction

Aircraft engine particulate matter (PM) emissions are composed of non-volatile 68 (black carbon, metal ash, oxygenated functional groups) and volatile components 69 (volatile organic compounds, nitrates, sulphates) (Gagné et al., 2021; Masiol and 70 Harrison, 2014; Petzold et al., 2011). The non-volatile particulate matter (nvPM) 71 emissions are formed in the combustor, while volatile particulate matter (vPM) 72 emissions, present in the gas phase at the engine exit, condense after emission. 73 Aircraft engines emit vPM with similar or greater orders of magnitude as nvPM, 74 especially after the vapour pressure of volatile species is lowered by oxidative 75 aging (Kilic et al., 2018) or by cooling (Beyersdorf et al., 2014). The nvPM and vPM 76 are constituents of total PM which affects air quality, health, and climate. The 77 International Civil Aviation Organization (ICAO) has developed standards and 78 recommended practices (SARPs) for measuring the mass- and number-based 79 emissions of nvPM emitted from aircraft engines with maximum rated thrust >26.7 80 kN (ICAO, 2017). Currently, SARPs have not been established for vPM or total PM 81 (Lobo et al., 2020). The SARPs for nvPM specify standardized sampling and 82





- measurement protocols (SAE, 2013, 2018; ICAO, 2017), which have been
- extensively evaluated and validated (Lobo et al., 2015b, 2020; Kinsey et al., 2021).
- 85 The nvPM regulatory limits are applicable for type certification of aircraft engines,
- ⁸⁶ but they do not address the vPM which may have substantial environmental
- 87 impacts.
- 88
- $_{89}$ To reduce CO₂ emissions, mitigate environmental impacts, and make the aviation
- sector more sustainable, a significant effort is underway to develop and deploy
- ⁹¹ sustainable aviation fuels (SAFs). Various feedstocks and different conversion
- pathways can be used to produce SAFs (Hileman and Stratton, 2014), which differ
- ⁹³ in chemical and physical properties compared to conventional petroleum jet fuel
- 94 (Vozka et al., 2019), most notably by lacking aromatic and sulfur species that are
- precursors to nvPM and vPM emissions. New SAF candidates must undergo a
- ⁹⁶ rigorous qualification and approval process (ASTM D4054) prior to being certified
- under the ASTM D7566 standard specification as a blending component. Currently,
- the ASTM D7566 standard allows SAF blend ratios of up to 50% with conventional
- ⁹⁹ fuel for drop-in fuels (Wilson et al., 2013).
- 100
- The combustion of neat SAFs and blends with conventional jet fuel has been shown to result in different PM emissions characteristics as a function of engine type and operating condition (Beyersdorf et al., 2014; Brem et al., 2015; Corporan et al.,
- ¹⁰⁴ 2011; Lobo et al., 2011, 2015a, 2016; Moore et al., 2017; Schripp et al., 2018, 2019;
- ¹⁰⁵ Timko et al., 2010). In addition to changes in PM mass- and number-based
- ¹⁰⁶ emissions, SAF combustion results in changes to particle size distributions (PSD)
- (Beyersdorf et al., 2014; Cain et al., 2013; Kinsey et al., 2012; Lobo et al., 2011,
- 2015a, 2016; Schripp et al., 2018; Timko et al., 2010), chemical composition (Elser
- ¹⁰⁹ et al., 2019; Kinsey et al., 2012; Timko et al., 2013; Williams et al., 2012),
- morphology (Huang and Vander Wal, 2013; Kumal et al., 2020; Liati et al., 2019),
- hygroscopic properties (Trueblood et al., 2018), and optical properties (Elser et al.,
- ¹¹² 2019).
- 113
- The standardized sampling and measurement protocol for aircraft engine nvPM
- emissions was designed and validated for engine certification tests using





conventional jet fuel. The SARP requires that number-based nvPM emissions are 116 measured with a butanol-based condensation-nuclei counter with 10 nm 50% cut-117 size sampling in single-particle-counting mode downstream of a diluter and 118 catalytic stripper. For mass-based nvPM emissions, the instrument must be 119 insensitive to vPM and able to meet performance specifications for repeatability, 120 zero drift, linearity, limit of detection, rise time, sampling interval, accuracy, and 121 applicability. To date, the only commercial instruments that satisfy the SARP 122 number and mass measurement system requirements, respectively, are the AVL 123 Particle Counter (APC) Advanced, and the AVL Micro Soot Sensor (MSS) and the 124 Artium Laser Induced Incandescence LII 300 instrument (LII). Limited information 125 is available on aircraft engine nvPM emissions characteristics measured with the 126 standardized system for different engine types burning SAFs and blends with 127 conventional fuel (Durand et al., 2021; Elser et al., 2019; Lobo et al., 2015a, 2016). 128 129 The standardized system components are not easily adaptable for use on aircraft 130 for measurement of cruise level nvPM emissions. Consequently, there are no 131 comparable in-flight engine-emissions data available for developing and validating 132 models that predict cruise nvPM-emissions based on engine certification data. 133 Particle size distribution measurements are also not included in the standardized 134 system, which are important for assessing the effects of fuels, operating conditions, 135 and engine technologies on the environmental impacts of PM emissions. Thus to 136 advance our understanding of aircraft engine emissions and the factors that 137 control them as well as to develop a large and consistent observational data base, 138 it is important to evaluate the relative performance of other diagnostic 139 instruments that are not prescribed in the standardized protocol but meet these 140 needs. Such instruments must be evaluated for their response to nvPM and total 141 PM emissions from aircraft engines using standardized and non-standardized 142 systems, and for measurements at the engine exit plane and downstream of the 143 engine in the near field, since these instruments are typically used with minimal 144

- change to their operating parameters for a wide range of sampling conditions.
- 146





148	The observations presented in this paper were collected during the NASA/DLR-
149	Multidisciplinary Airborne Experiment (ND-MAX)/ Emission and Climate Impact
150	of Alternative Fuel (ECLIF) 2 campaign that was conducted at Ramstein Air Base,
151	Ramstein-Miesenbach, Germany in January-February 2018 (see overview by
152	(Anderson and NDMAX-Team, 2021)). The campaign included ground-based and
153	in-flight measurements of emissions from the DLR Advanced Technology Research
154	Aircraft (ATRA) A320 aircraft with V2527-A5 engines running on two
155	conventional jet fuels and three blends with SAF. The main objective of the ground-
156	based measurements was to characterize the nvPM, total PM, and hydrocarbon
157	emissions as functions of engine thrust condition and fuel composition. Several
158	identical instruments were included in the in-flight sampling aircraft (NASA DC-8)
159	and ground measurement diagnostic instrument suites to enable comparisons of
160	engine emissions during ground and airborne operations, and create a data set for
161	testing cruise emission models. The NASA DC-8 aircraft with CFM56-2C1 engines
162	was also used as an emissions source to compare various emissions diagnostic
163	instruments during the ground-based measurements.
164	
164 165	Here we present the inter-comparison of real-time measurements of aircraft
	Here we present the inter-comparison of real-time measurements of aircraft engine nvPM emissions in terms of physical characteristics such as mass, number,
165	
165 166	engine nvPM emissions in terms of physical characteristics such as mass, number,
165 166 167	engine nvPM emissions in terms of physical characteristics such as mass, number, and size distributions using different diagnostic instruments and measurement
165 166 167 168	engine nvPM emissions in terms of physical characteristics such as mass, number, and size distributions using different diagnostic instruments and measurement principles. The nvPM mass emissions were evaluated using three real-time
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165 166 167 168 169 170 171 172 173 174 175 176	engine nvPM emissions in terms of physical characteristics such as mass, number, and size distributions using different diagnostic instruments and measurement principles. The nvPM mass emissions were evaluated using three real-time sampling techniques: photoacoustic spectroscopy, the extinction-minus-scattering technique, and laser-induced incandescence (LII), and two alternative measurement techniques widely used in laboratories and on-board aircraft: filter- based photometry and PSD integration. We note that one of the photoacoustic instruments and the LII instruments have been demonstrated to be compliant with the ICAO SARP performance specifications. The PM number-based emissions were measured using a condensation particle counter. The PSD characteristics measured by scanning mobility particle sizers and an electrical mobility spectrometer were also compared. The nvPM and total PM emissions were delineated using a thermal denuder and a catalytic stripper. We also report the
165 166 167 168 169 170 171 172 173 174 175 176 177	engine nvPM emissions in terms of physical characteristics such as mass, number, and size distributions using different diagnostic instruments and measurement principles. The nvPM mass emissions were evaluated using three real-time sampling techniques: photoacoustic spectroscopy, the extinction-minus-scattering technique, and laser-induced incandescence (LII), and two alternative measurement techniques widely used in laboratories and on-board aircraft: filter- based photometry and PSD integration. We note that one of the photoacoustic instruments and the LII instruments have been demonstrated to be compliant with the ICAO SARP performance specifications. The PM number-based emissions were measured using a condensation particle counter. The PSD characteristics measured by scanning mobility particle sizers and an electrical mobility spectrometer were also compared. The nvPM and total PM emissions were





- absorption and consequently LII signals, and hence the derived nvPM mass
- concentration. The impact of fuel composition on PM emissions will be reported
- separately (Schripp and NDMAX-Team, 2021).
- 184 3 Methods

185 3.1 Engine and fuels

In the majority of this work, emissions were sampled from a single IAE 186 V2527-A5 starboard engine of the DLR ATRA aircraft (Airbus A320-232). The 187 engine was operated on two conventional, petroleum jet fuels, referred to as REF3 188 and REF4, and three sustainable aviation fuel blends, referred to as SAF1, SAF2, 189 and SAF3. The abbreviations for the two conventional petroleum fuels are used to 190 avoid confusion with the previous ECLIF campaign (Schripp et al., 2018). 191 A limited number of experiments were also performed with JP-8 fuel, 192 combusted in the starboard CFM56-2C1 engine (#3) of the NASA DC-8 aircraft. 193

¹⁹⁴ Due to limited fuel availability, none of the other five fuels could be combusted in

the CFM56-2C1 engine. The properties of the six fuels are summarized in Table 1.

196 3.2 Ambient conditions

The measurements presented in this manuscript were performed outdoors 197 during winter in western Germany. Detailed meteorology for each test point is 198 given in the supplement. The minimum, median, and maximum temperatures were 199 2.3, 2.9, 8.3 °C, respectively. Conditions were humid (>83 % humidity) and 200 sometimes rainy. Winds ranged from 0 to 15.5 km h⁻¹ and wind direction was 201 sometimes variable. The median wind direction was south-westerly, while the 202 source aircraft was oriented facing to the east. Consequently, winds blowing 203 approximately 45° angle from the right rear of the source aircraft sometimes 204 prevented the engine emissions from reaching the sampling probe at low engine 205 thrust settings. 206





208 3.3 Emissions sampling

209	An extensive suite of aerosol and gas-phase instruments operated by the
210	members of six different institutions were deployed in two different shipping
211	containers to characterize the emissions (Table 2). The complete emission-
212	sampling setup is discussed in companion papers (Anderson and NDMAX-Team,
213	2021; Schripp and NDMAX-Team, 2021). Briefly, emissions were sampled
214	through a probe located 43 m downstream of the starboard engine of the aircraft.
215	The probe was placed in front of a blast fence located on the western side of the
216	Ramstein Air Force Base flight line, and the fence redirected the engine exhaust
217	upwards for safety. The probe was connected to a 18.5-mm ID, 20-m-long
218	electrically-conductive sampling line heated to 60 °C, that transported flow to a
219	sampling plenum maintained at 33 °C. To minimize residence time and particle
220	losses in this sampling line, a pump ensured that a total of at least 137 L min ⁻¹
221	flowed through the sampling manifold at all times. Higher flows produce an
222	unacceptably large pressure drop in the primary sampling line. The majority of
223	this flow was discarded as excess.
224	
225	The plenum was placed inside a modified shipping container (Container 1) behind
226	the blast fence, along with the NRC, DLR, and NASA instruments. The North
227	American Reference System (NARS) was connected to the plenum by a short
228	section of heated line to the NARS dilutor box, which was heated to 60 \pm 15 °C and
229	contained a custom Dekati dilutor with a dilution ratio of approximately 4 (less
230	than the standard Dekati dilutor ratio of 8 to 14). A 25 m line heated to 60 \pm 15 °C
231	transferred sample aerosols flow from the dilutor box to a second shipping
232	container (Container 2), where the MST and ARI instruments were connected in
233	parallel. The NARS components include the 25 m heated line, attached diluters and
234	MST instrument suite; the system is compliant with specifications for the
235	standardized nvPM sampling and measurement system (SAE, 2013; SAE, 2018;
236	ICAO, 2017) and whose performance has been demonstrated and evaluated in
237	previous studies (Lobo et al., 2015b, 2016, 2020). Additional instrumentation
238	installed as part of the NARS included a fast electrical mobility spectrometer
239	(Cambustion DMS500), an Aerodyne Aerosol Mass Spectrometer (results not





- presented here), and a CAPS PMssa monitor (Aerodyne Research Inc.). The details
- of the instruments installed inside these two containers are listed in Table 3.
- 242 3.3.1 Gaseous measurements
- A suite of gaseous emissions was measured in this study, as summarized in Table
- 2. The CO₂ measurements from the NASA LI-COR 7000 were in good agreement
- with those taken by DLR (MKS MultiGas 2030 FTIR Continuous Gas Analyzer) and
- MST (LI-COR model 840A), but had a faster response time and were therefore used
- as the reference for instruments in Container 1. Instruments in Container 2 used
- the MST measurements as reference.
- 3.3.2 nvPM number and particle size distributions (PSDs)

nvPM number concentration was measured directly by a certification-test-250 compliant, particle counter, APC (AVL Inc., which contains a TSI Model 3790E 251 CPC), which was part of the NARS in Container 2. PSDs were measured with two 252 technologies: scanning mobility particle sizers (SMPS, TSI Inc.) and electrical 253 mobility sizers (EMS). Two types of EMS were used; the Cambustion DMS500 (in 254 Container 2, measuring particles 10 to 1000 nm in diameter) and the TSI Engine 255 Exhaust Particle Sizer (EEPS, Container 1). However, the EEPS data were excluded 256 from this analysis due to unidentified problems with the instrument which led to 257 anomalous PSDs. 258 Two SMPSs measured nvPM PSDs. An SMPS operated by NRC measured 259 particles 10 to 278 nm in diameter downstream of a catalytic stripper (Model 260 CS015, Catalytic Instruments GmbH), which heated samples to 350 °C before 261 oxidizing gas-phase VOCs to prevent them from recondensing after exiting the 262 device. Another SMPS operated by NASA measured particles 10 to 278 nm in 263 diameter either directly or downstream of a NASA-constructed thermal denuder 264 (TD) also operated at 350 °C. The TD employs a concentric activated charcoal filter 265 downstream of the sample heater to prevent re-condensation of volatile species. 266 TDs are commonly used on-board aircraft for measuring nvPM number 267 concentration and size distributions (Clarke, 1991; Moore et al., 2017) and have 268 been shown to effectively evaporate nucleation and accumulation mode sulfate 269

and organic aerosols (Beyersdorf et al., 2014; Schripp et al., 2018).





271 3.3.3 nvPM mass measurements

272	In this study, most of the nvPM mass data were derived from light
273	absorption coefficients (units of m ⁻¹), either determined in flow-through sample
274	cells (the CAPS PM_{SSA} , PAX, and MSS introduced below) or after collecting particles
275	onto a filter (the TAP and PSAP introduced below). Such absorption measurements
276	can be converted to equivalent black carbon or eBC mass concentrations (eBC,
277	units of g m ⁻³ ; Petzold et al. (2013)) by dividing them by a reference mass
278	absorption cross-section (MAC, units of $m^2 g^{-1}$). The LII measurements also rely on
279	light absorption, although the measurand is not absorption but incandescence at
280	two wavelengths and is termed rBC (Petzold et al., 2013; Michelsen et al., 2014).
281	The reference MAC used to report eBC represents an assumed physical
282	property of the nvPM emitted by the engine at a given time. The extensive review
283	of Bond and Bergstrom (2006) concluded that the MAC at 550 nm of externally-
284	mixed BC from a variety of sources could be summarized as 7.5 \pm 1.2 $m^2g^{\text{-1}}$; the
285	more recent review of in-situ measurements by (Liu et al., 2020) recommended
286	$8.0\pm0.7\ m^2g^{\cdot1}$ at 550 nm. In this study, we have used the Bond and Bergstrom
287	value of 7.5 $m^2 g^{-1}$ for consistency with earlier work and instrument software.
288	These values are assumed to vary inversely with wavelength, with an Angstrom
289	(power) exponent of 1; for example, the 660 nm CAPS PM_SSA monitor data were
290	processed with a MAC of 7.5 m ² g ⁻¹ × (550 nm / 660 nm) ¹ = 6.5 m ² g ⁻¹ .
291	One eBC technique, the CAPS PM_{SSA} monitor (Aerodyne Research Inc.; Onasch et al.,
292	2015) derives absorption coefficients as the difference between measured aerosol
293	extinction and scattering coefficients, from which eBC concentrations were
294	calculated as described above. The CAPS PM_{SSA} measures light extinction by the
295	calibration-free cavity attenuation phase shift (CAPS) technique and light
296	scattering with an integrating nephelometer. The CAPS technique measures the
297	lifetime of photons in a high-finesse optical cavity comprised of two high
298	reflectivity mirrors, from which the extinction coefficient can be calculated. An
299	integrating nephelometer captures light scattered from a section of this cavity, and
300	is calibrated using the measured extinction of small (Rayleigh regime) non-
301	absorbing particles. In this study, two CAPS PM_{SSA} were present, one operated at
302	630 nm wavelength by ARI and the other at 660 nm wavelength by NRC. The





- 303 scattering channel of the NRC CAPS PMssA was calibrated on-site using nebulized
- $_{\tt 304}$ $\,$ and dried ammonium sulfate particles; the other instruments were similarly
- ³⁰⁵ calibrated prior to the campaign at the manufacturer using 200 nm ammonium
- ³⁰⁶ sulfate. For the sub-200 nm particles measured in this study, no truncation
- ³⁰⁷ corrections (Modini et al., 2021) were necessary.
- Two other eBC instruments were based on photoacoustic spectroscopy, namely 308 the Photoacoustic Extinctiometer (PAX, DMT Inc,; Nakayama et al., 2015) and the 309 Micro Soot Sensor (MSS; AVL GmbH; Schindler et al., 2004). In both of these 310 instruments, aerosol absorption is measured by the periodic heating of particles 311 using a modulated laser, resulting in the generation of pressure waves which are 312 amplified by an acoustic cell and detected by a microphone. The PAX was 313 calibrated using nebulized ammonium sulfate as well as graphitic nanoparticles 314 (Aquadaq). 315
- During on-site calibration of the PAX using graphitic Aquadag nanoparticles, the PAX signals were observed to drifted slowly upwards after each baseline. We were nevertheless able to obtain useful data by configuring the PAX to auto-baseline every 180 seconds, and only using the first 15 seconds of measurements after each baseline. After the campaign, it was found that a component of the circuit board was damaged during the initial shipment. In spite of this electrical problem, the PAX data do not represent outliers in the following analysis.
- Two additional pairs of eBC instruments were deployed at the ground site and on-323 board the NASA DC-8 that measured aerosol absorption coefficients based on filter 324 attenuation, namely a Particle Soot Absorption Photometer (PSAP, Radiance 325 Research; Bond et al., 1999) and Tricolor Absorption Photometer (TAP, Brechtel 326 Manufacturing Inc, ; Ogren et al., 2017). These instruments were designed as low-327 cost, low-maintenance devices for monitoring aerosol optical properties in the 328 background atmosphere (i.e., at low concentrations) and have been used 329 previously in airborne and ground-based studies (Moore et al., 2017). In these 330 instruments, particles are continuously collected onto an internal filter while its 331 light attenuation is measured. The change in light attenuation over time is used to 332 calculate absorption coefficients. This calculation requires post-processing to 333





- ³³⁴ correct for filter loading effects (which do not require independent measurements)
- and may also be corrected for light attenuation due to scattering rather than
- absorption (which requires an independent nephelometer measurement)
- (Virkkula, 2010). Other sources of error include nonlinearities due to size-
- dependent penetration of particles into the filter media and the evaporation of
- volatile species over time (Lack et al., 2014; Nakayama et al., 2010). We note that
- the TAP automatically advances its filter when its transmission drops below 80%,
- ³⁴¹ whereas the PSAP requires a manual filter change. The PSAP filter was therefore
- changed manually before each set of experiments herein, to ensure that its filter
- transmission remained above 80% during all measurements.
- ³⁴⁴ Finally, three Artium LII 300 (Artium Technologies) instruments measured rBC,
- based on two-colour pulsed laser–induced incandescence (LII) (Snelling et al.,
- ³⁴⁶ 2005). These instruments heat nvPM using a 1064 nm pulsed laser and measure
- the resulting incandescence at two wavelength bands. From this measurement,
- rBC temperature and mass concentrations can be calculated. One of the LII 300s
- was a component of the NARS. Of the other two, one was dedicated to an
- experiment where its operating conditions were varied (Section 4.6). Therefore,
- $_{351}$ only two LII 300s were measuring real-time nvPM mass concentration
- simultaneously at any given time. The MSS+ and the LII 300s were calibrated by
- reference to the elemental carbon mass (defined by thermal–optical analysis)
- produced by a laboratory diffusion-flame combustion aerosol source using
- $_{\tt 355}$ measurements at three mass concentrations spanning 0.1 to 0.5 mg m $^{-3}$ (SAE,
- ³⁵⁶ 2018).

357 3.4 Data analysis

358 3.4.1 Emission index calculations

The raw data were analysed over comparable time intervals and crosschecked by independent calculations. The general analysis proceeded as described in this section. First, the time series of measured CO₂ concentrations was used as a reference against which to synchronize all time series, based on rapid rises and

³⁶³ falls in the observed concentrations (measured at 1 Hz) when the engine thrust





- ³⁶⁴ condition underwent large changes (as shown at 08:00 in Figure 2). All
- instruments were synchronized against the NASA CO₂ sensor except the
- ³⁶⁶ instruments in container 2, which was synchronized against the MST LI-COR CO₂
- 367 sensor, because of the additional dilution stage. The time synchronization
- accounted for different lag times due to differences in the response times and clock
- accuracy of each instrument.
- ³⁷⁰ Second, the CO₂ concentrations [CO₂] were baseline-subtracted and filtered as
- $_{371}$ follows. The CO2 baseline ([CO₂]_b) was calculated as the mean of the CO₂
- $_{372}$ concentrations measured before ([CO₂]₀) and after ([CO₂]₁) each test. The
- uncertainty in this baseline value was calculated as either $([CO_2]_b [CO_2]_0)$ or
- $([CO_2]_b [CO_2]_1)$, whichever was greater.
- ³⁷⁵ Due to the prevailing crosswind mentioned above, unstable CO₂ concentrations
- occurred during from some test points at the idle engine thrust condition. These
- unstable conditions were identified and filtered using two separate methods. In
- the first method, the SMPS PSDs were inspected for reproducibility. In the second
- ³⁷⁹ method, an algorithm was used to reject any test points with CO₂ uncertainties
- greater than 50%, CO₂ signals less than a factor of ten greater than uncertainty, or
- ³⁸¹ CO₂ signals less than 20% above baseline. We found that the first method rejected
- all of the points rejected by the algorithm, in addition to a few additional points.
- ³⁸³ The analysis presented uses the first method.
- Third, all data were arithmetically averaged over the test point periods defined in
- Table S1. For each instrument, the averaging periods were refined by inspection of
- the data since sampling-line residence times varied. The averaged data were
- typically at 1 Hz sampling frequency initially, although the SMPS instruments
- measured PSDs at 45 second intervals (NRC instrument) or 30 second intervals
- (NASA). Emission indices (EIs) were then calculated from the averaged data
- ³⁹⁰ following (SAE, 2013):

$$\mathsf{EI}_{\mathrm{m}} = \mathsf{PM}_{\mathrm{m}} \frac{RT_{m}}{[\mathsf{CO}_{2}](M_{c} + \alpha M_{H})P_{m}} \tag{1}$$





$$\mathsf{EI}_{\mathrm{num}} = \mathsf{PN} \times 10^6 \frac{RT_m}{[\mathsf{CO}_2](M_c + \alpha M_H)P_m}$$
(2)

392

³⁹³ Where EI_m and EI_{num} are mass and number-based EIs, respectively; PM_m ³⁹⁴ and PN are mass and number concentrations, respectively, at standard reference ³⁹⁵ temperature (T_m ; 273.15 K) and pressure (P_m ; 1 atm); α is the hydrogen to carbon ³⁹⁶ ratio of the fuel; M_c and M_H are the molar masses of carbon and hydrogen, ³⁹⁷ respectively; and R is the ideal gas constant (0.082 L.atm.K⁻¹.mol⁻¹).

398 3.4.2 Loss correction

Particles may be lost to the walls of sampling lines or to deposits on those 399 walls. The fraction of particles penetrating a given system varies with size, 400 according to a characteristic penetration function. Four penetration functions were 401 applied in this study: 1) from the probe to the sampling plenum, 2) from the 402 plenum to the NARS, 3) within the TD, and 4) within the CS (Figure 4). Function 1 403 was measured on site as described below. Function 2 was calculated using the 404 standard loss calculation methodologies provided in SAE documents AIR6504 405 (SAE, 2017) and ARP6481 (SAE, 2019). Function 3 was experimentally determined 406 in the laboratory by NASA. Function 4 was obtained from theoretical estimates and 407 experimental measurements (Catalytic Stripper manual, 2014). 408 409 Penetration function 1 (probe-to-plenum penetration) was measured 410 experimentally by nebulizing ammonium sulfate particles at the probe while all 411 instruments were sampling and all heated lines had reached thermal equilibrium. 412 (Function 1 therefore also includes the smaller instrument sampling lines 413 downstream of the plenum in its correction as well; however, these were 414 considered negligible relative to the longer probe-to-plenum and plenum-to-415 Container-2 transport lengths.) For this measurement, the NRC SMPS was moved 416 to the probe, while the NASA instrument remained in its standard position. The 417

- ratio of the NASA to NRC PSDs then provided a first estimate of the penetration
- ⁴¹⁹ function. However, this first estimate was not accurate, as the measurements were
- performed on a cold day (measured as approximately 5 °C outdoors and 15 °C in
- the instrument container) and as it does not account for performance differences





between the NASA and NRC SMPSs. Therefore, two corrections were made. First, 422 both measurements were corrected to standard temperature and pressure. 423 Second, differences between the two instruments were directly measured by 424 moving the NRC SMPS just outside of the sampling container (to keep it at 5 °C) 425 and connecting it to an identical sampling line as the NASA SMPS. The ratio of the 426 two measured PSDs in this setup was defined as equal to unity at all sizes, and 427 used to correct the initial penetration function. Therefore, no further correction 428 was made for sampling lines in Container 1. Losses in this additional line were 429 negligible (calculated penetrations of 0.997 at 100 nm and 0.98 at 20 nm) relative 430 to the long NARS line to Container 2 (i.e., Function 2). 431

432 3.5 Uncertainties

All reported uncertainties and error bars represent standard errors, 433 propagated through the calculation as necessary. When two independent sources 434 of uncertainty were available (for example, the standard error in the 10 second 435 averages of $[CO_2]$ and the uncertainty in the baseline value) they were added in 436 guadrature. Our bottom-up calculations of uncertainty can be compared with the 437 spread of the data points in our El comparisons below. This spread represents a 438 top-down uncertainty, and is similar in magnitude to the bottom-up uncertainties 439 (i.e. error bars). This similarity lends confidence to our uncertainty estimates. In 440 most figures, error bars have generally been omitted for clarity, but uncertainties 441 are given for each instrument at each test point in Table S1. 442

443 4 Results and discussion

444 4.1 Experiment overview

A typical time series obtained when the emissions from the IAE V2527-A5 engine were sampled is shown in Figure 2. Nominal low-pressure turbine fan speeds (N1) expressed as a percentage of maximum continuous thrust, are shown by the labels at the top of the figure. Percent N1 (along with engine fuel flow rate) is another metric for representing the different engine thrust conditions and is used as a primary independent variable in this study. The CO₂ concentrations (red line) were

highly variable at N1 = 23% as the ambient wind shifted the aircraft exhaust plume





- toward and away from the sampling probe. Correspondingly, both nvPM mass and
- ⁴⁵³ PSD measurements were highly variable, as shown by the blue trace and black
- 454 symbols, respectively.
- 455
- $_{456}$ As shown in Figure 3a, nvPM mass concentrations, represented by EI_{m} , increased
- with increasing N1 before decreasing slightly at the highest N1, similar to the
- trends for other engine types reported by Lobo et al. (2015b, 2020). Figure 3b
- shows that the relationship for EI_{num} is less clear, with a slight increase at
- 460 moderate N1 followed by a greater decrease at high N1. As discussed below
- $_{\tt 461}$ (Section 4.2.3), the higher EI_m at higher N1 thrust was associated with larger
- ⁴⁶² particle sizes, and therefore smaller penetration-function corrections (Section
- 463 4.2.1). An effect of fuel composition is evident, and discussed in detail in Schripp et
- al. (Schripp and NDMAX-Team, 2021).
- 465 4.2 Size distributions and penetration functions

466 4.2.1 Penetration function

- A typical PSD, and corresponding PVD, are shown in Figure 4, in the context of the
- ⁴⁶⁸ penetration functions applied in this work. The PVD was calculated by assuming
- spherical particles, which incurs negligible error for aircraft-engine nvPM due to
- the small diameter of particles produced by such engines (Durdina et al., 2014;
- ⁴⁷¹ Saffaripour et al., 2020). For the example PSD and PVD in Figure 4 (shading), it is
- clear that a substantial fraction of the particle number was corrected for
- ⁴⁷³ penetrations (lines) of roughly 0.5. In contrast, the larger mode of the PVD
- ⁴⁷⁴ corresponds to penetrations larger than 0.8 in most cases. These differences led to
- a median number- and mass-based correction factors of 1.51 and 1.19, respectively
- ⁴⁷⁶ for penetration Function 1 (probe to plenum) labelled in the figure. The remaining
- instrument-specific penetration corrections were applied according to the position
- of each instrument in the sampling system, as specified in Table 2. The magnitude
- of each correction is given in Table S1.
- Figure 5 shows selected PSDs from the IAE V2527-A5 engine operated with SAJF1
- (Figure 5a) and REF4 (Figure 5b) fuels. The plot illustrates a lower (40 %) and a





higher thrust point (60 %) from the available data for two fuels. Note that the
ordinate scales are harmonized across the upper and lower rows only. All abscissa
scales are harmonized. The figure indicates roughly comparable PSDs from these
two fuels. The companion paper (Schripp and NDMAX-Team, 2021) compares the
effects of fuel composition in detail.

487 4.2.2 PSDs of CFM56-2C1

The CFM56-2C1 engine on the DC-8 burning JP-8 emitted an order of magnitude 488 more total particles per unit fuel burned than any of the fuels combusted in the 489 ATRA. We attribute this difference to the relatively high sulfur content of the JP-8 490 fuel (1490 ppm sulfur versus \leq 105 ppm for the other fuels). The CFM56-2C1 491 engine also emitted a factor of three lower nvPM mass and nvPM number than the 492 V2527-A5 engine. The presence of extremely small particles with $d_m < 10$ nm was 493 evident in the two nvPM PSDs (not shown due to the extremely large penetration 494 function at these sizes; Figure 4). The CS-SMPS data extended to smaller diameters, 495 and showed that the size range measured by these two instruments was 496 insufficient to capture the full PSD for the CFM56-2C1 engine data at 22% N1 as 497 well as 63% N1. The d_m < 10nm mode was not as prominent in the V2527-A5 498 engine exhaust at any thrust, although some evidence was observed for it (e.g. 499 number distribution at 40% N1 in Figure 5b). 500

Since the CFM56-2C1-with-JP-8 data were strongly influenced by a nucleation 501 mode, and were therefore not well described by the GMD and GSD of the data, 502 these measurements have been omitted from all subsequent PSD analysis in this 503 manuscript. Bimodal fits to the data were not possible as the nucleation mode was 504 not captured by our size distributions. However, the nvPM mass measurements 505 are much less sensitive to these small particles (Hinds, 1999) and have therefore 506 been retained. PSDs from all instruments, test points, and fuels from both the 507 CFM56-2C1 and V2527-A5 engines are included in the supplement. 508

4.2.3 Particle size statistics; GMD and GSD

⁵¹⁰ Figure 6 summarizes the PSDs measured by three instruments in terms of their





- 511 GMD and GSD. The data sets labelled SMPS and TD-SMPS were both obtained from
- NASA's SMPS, which was manually switched to a bypass line and the TD at each
- test point. The data set labelled CS-SMPS was obtained with NRC's SMPS.
- ⁵¹⁴ Total PM is represented by the data sets labelled DMS500 and SMPS. However, the
- two are not directly comparable because the DMS500 measurements were
- obtained after an additional dilution by a factor of 4 in the NARS and the DMS500
- was not operated behind a volatile particle remover (CS or TD). Moreover, the
- inversion of DMS500 data requires more assumptions about the particle size
- distribution than the analogous SMPS calculation. Either volatiles or this inversion
- procedure may have caused the 10% larger GSDs observed for the DMS500 for
- some data (some measurements with GMDs over 35 nm) relative to the SMPS.
- 522 Since volatiles would affect both GMD and GSD, but we primarily observed
- discrepancies in the DMS500 GSD, we suggest that the inversion was the major
- ⁵²⁴ source of bias in these data.
- 525
- nvPM is represented by the open circles and filled squares in Figure 6. These two 526 data sets show a different relationship (slope) between GMD and GSD, reflecting 527 systematic differences in the corresponding PSDs. Relative to the mean of the two 528 instruments, the NRC GMDs were higher (Figure 7a) while the NRC GSDs were 529 higher at GSD < 1.75 but lower at GSD > 1.75 (Figure 7b). Inspection of the 530 corresponding PSDs showed that the NASA and NRC instruments agreed at higher 531 d_m but that NRC number concentrations were higher at smaller d_m . This trend 532 suggests that a bias in the penetration functions applied to each instrument 533 (Figure 4, Table 2) led to the discrepancy in GMD and GSD. Such a bias would affect 534 the nvPM concentration estimated from these PSDs (Figure 8b) and will be 535 discussed further below. 536
- In spite of these trends in GMD and GSD, the PSD measurements agreed to within
- ⁵³⁸ 20% (Figure 7a) for nvPM GMDs and within 5% for nvPM GSDs (Figure 7b).
- 539 Furthermore, these measurements are consistent with previous measurements by
- Lobo et al. (2015c), as illustrated by the line in Figure 6, which reproduces the
- ⁵⁴¹ polynomial best-fit line reported by those authors.





- 4.3 Consistency between number-based emission indices of nvPM and
- 543 VPM

544	Figure 7c compares the measured vPM and $nvPMEI_{num}$ with the mean $nvPMEI_{num}$
545	(i.e., mean of the NRC CS-SMPS, NASA TD-SMPS, and NARS APC. The grey shading
546	shows that all instruments agreed to within a factor of 2. The APC and DMS500
547	$n\nu\text{PM}\text{EI}_{num}$ were both typically higher than the two similar SMPSs. The APC has a
548	50% efficiency at its cut-off diameter of 10 nm, reaching 100% efficiency above
549	this size and 0% below it. Therefore, relative to the SMPSs, which measured down
550	to approximately 10 nm with 100% efficiency, the APC should measure lower than
551	the SMPSs since it will underperform at sizes close to 10 nm. (This expectation
552	requires that there are no particles present above the SMPS upper detection limit
553	of 280 nm in our study, which was verified by our PSD analysis in Section 3 and
554	Table 2). However, the APC measured approximately 50% larger $nvPM EI_{num}$
555	under all conditions, and our measured PSDs rule out the possibility that 50% of
556	particles were not seen by the SMPS. Therefore, we attribute the difference
557	between APC and SMPS results to uncertainties in the APC or SMPS penetration
558	correction functions. Since the two SMPSs agreed, the APC measurements were
559	likely overcorrected when the SARP correction procedures were applied.
	We also attribute the larger by DM EL
560	We also attribute the larger nvPM EI_{num} measured by the DMS500 to the same
561	cause; to which a similar penetration function as the APC applies (Section 3.4.2).

- ⁵⁶² 4.4 Consistency between mass-based emission indices
- 563 4.4.1 EI_m measurements by real-time sampling instruments

 $_{\tt 564}$ $\,$ Figure 8a presents scatterplots of the real-time EI_m measurements acquired during

 $_{\tt 565}$ this study for all fuels and both engine types. In Figure 8a, the individual EI_m are

- ⁵⁶⁶ plotted against the geometric mean of the instruments shown in the caption: three
- LII 300 instruments, two CAPS instruments, one PAX and one MSS+. The geometric
- mean was chosen over the more-common arithmetic mean because the data are
- not normally distributed; the arithmetic mean would therefore have over-
- ⁵⁷⁰ emphasized outliers.





Figure 9a presents the same data as Figure 8a except that the measurements have 571 been normalized to the geometric-mean EI_m from Figure 8a. Most data fall within 572 30 % of the mean (inner dashed lines) above 100 mg / kg_{fuel}. We note that exhaust 573 samples were diluted with background air by a factor of 40 or more before 574 reaching the inlet probe, so at this lower limit, the actual concentration observed 575 by the instruments was approximately 10 μ g m⁻³ (the exact conversion factor 576 varies with CO₂ concentration and fuel properties), which is close to their 577 detection limits, as expected. This lower limit may have been influenced by the 578 ambient measurement conditions, where background nvPM concentrations were 579 non-negligible. 580 The agreement of the real-time measurements to within 30 % is notable 581

considering the different types of instruments used. The scatter at lower EIm 582 values reflects the noise levels of the instruments. Both of these observations are 583 consistent with data reported previously for different engine types by Lobo et al. 584 (2016, 2020). The LII 300 and MSS+ from the North American Reference System 585 (NARS) have been widely used to characterize aircraft engine nvPM emissions. The 586 two CAPS instruments were independently calibrated and operated. The MSS+ and 587 PAX represent two photoacoustic spectrometers from different manufacturers, 588 operated by different teams, with different principles of calibration. The PAX was 589 also operated with a damaged capacitor on its printed circuit board. As noted in 590 Methods, these instruments operate on a variety of physical principles, including 591 photoacoustic spectroscopy (with two different designs), extinction-minus-592 scattering, and laser-induced incandescence (cf. Section 3.3.3). Agreement 593 between these various principles also suggests that factors such as volatile 594 coatings on nvPM did not influence the instrument responses. 595

596 4.4.2 SMPS-based EI_m

⁵⁹⁷ Figure 8b and Figure 9b are analogous to Figure 8a and Figure 9a, but for the

⁵⁹⁸ integrative nvPM measurements that do not fall into the real-time sampling

category. These data are plotted against the same geometric mean from Figure 8a.

- ⁶⁰⁰ The dashed lines in Figure 9b represent the same ratios as in Figure 9a.
- 601 Considering that the real-time instruments in Figure 8a were either calibrated to





- ⁶⁰² aerosol absorption or to aviation nvPM, we consider their accuracy as greater than
- the instruments in Figure 8b and consider departures from the 1:1 line as due to
- 604 inaccuracy.
- Most of the instruments in Figure 8b were accurate to within 30% of the reference,
- similar to Figure 8a, with the exception of the CS-SMPS and PSAP. This is
- summarized in Table 3, which shows the mean ratios of all data except engine idle
- (23% N1) with the geometric mean. Table 3 also includes the results of a linear
- regression against the geometric mean to facilitate comparison of our
- measurements with Kinsey et al. (2021), who performed linear regressions against
- simultaneous elemental carbon (EC) measurements (in our study, mass
- concentrations were too low to obtain EC measurements). The PSAP data are
- discussed in the next section. The CS-SMPS data were systematically higher than
- the geometric mean, potentially due to an overcorrection of the penetration of
- ⁶¹⁵ large particles to the SMPS.
- ⁶¹⁶ Since the spread of nvPM EI_m reported by the two SMPS systems was smaller than
- $_{\tt 617}$ the bias, their difference relative to the reference EI_m cannot be attributed to
- measurement imprecision. Since the two SMPS systems showed different
- accuracies, their differences cannot be ascribed to a lack of constraints on the
- effective density of the nvPM particles (Momenimovahed and Olfert, 2015), which
- may vary with the monomer diameter (Abegglen et al., 2014; Durdina et al., 2014)
- and/or shape of soot aggregates. With respect to the real-time measurements, the
- TD-SMPS data are also consistent with previous measurements of aviation engine
- PSDs, which, however, were not corrected for diffusional particle loss (Lobo et al.,
- ⁶²⁵ 2015b, 2020). Careful measurement of the penetration functions used in these
- calculations would be required to confirm our interpretation.
- 627 4.4.3 Filter photometer-based EI_m from TAP and PSAP
- ⁶²⁸ Figure 8b and Figure 9b show that the TAP measurements were within the 30 %
- range observed for the real-time instruments, with a relative standard deviation
- (RSD) of 14 % (Table 3) for all data excluding the engine idle condition (23% N1).
- ⁶³¹ This provides high confidence for the use of the TAP for in-flight or field





- measurements of aircraft-engine nvPM mass emissions, when filter-loading
- ⁶³³ corrections (Section 3.3.3) are correctly applied.
- The PSAP, on the other hand, showed much greater variability, with an RSD of 36% 634 (Table 3). This is substantially higher than the variability reported by a laboratory 635 intercomparison of PSAP and CAPS PM_{SSA} (Perim De Faria et al., 2021) (that study 636 did not report a statistic comparable to RSD). Although the PSAP has been 637 observed to deviate up to a factor of two higher in cases of high organic aerosol 638 loading or reduced filter transmission (Lack et al., 2013), our data are restricted to 639 transmissions above 0.8. The fact that the PSAP shows great variability rather than 640 a fixed offset indicates that the issue is not due to a systematic error such as an 641 inaccurate MAC or flow rate calibration. We note that the TAP and PSAP were 642 operated with reduced sample flow rates of 0.05 L min⁻¹ and 0.1 L min⁻¹, 643 respectively, (5 to 10% of nominal settings) to extend the life of their filter media 644 while sampling the high soot concentrations in the aircraft exhaust. Under these 645 conditions, detector noise and small fluctuations in sample flow have a magnified 646 effect on resulting derived absorption coefficients. We suspect that the 647 measurements would have been significantly more precise if the instruments had 648 been operated at nominal flows, although this would have required changing 649 filters after each test point. Consistent with our hypothesis, we note that 650 Nakayama et al. (2010) observed substantially larger variability in PSAP 651 measurements at 0.3 than at 0.7 standard litres per minute. We also note that Bond 652 et al. (1999) did not observe an impact of flow rate when changing from 1 to 2 653 litres per minute. 654 Figure 10 plots as a function of particle GMD the same relative TAP and PSAP EI_m 655 data shown in Figure 9b. No clear trend of this ratio with size is evident, although 656 the measurements become somewhat more scattered at smaller sizes for the SAF1 657
- data set, where signal to noise is lower (GMD and EI_m were correlated, see the
- ⁶⁵⁹ below discussion of Figure 12). Figure 10b includes the size-dependent PSAP
- correction function reported by Nakayama et al. (2010) (their Equation 8), with
- $_{\tt 661}$ $\,$ shading representing a 1σ uncertainty. Those authors predicted the true
- absorption values using Mie theory for nigrosin particles of diameter 100 to
- 663 600 nm and refractive index 1.685–0.285*i*. Thus, their correction factor is





- ⁶⁶⁴ conceptually equivalent to our El_m/mean-El_m. Extrapolating their correction
- function down from 100 nm to 15 nm gives values ranging from 4 to 8, whereas
- our measurements are close to 1.0. This discrepancy may be attributed primarily
- to the extrapolation, and possibly also to the fact that we have measured solid
- nvPM particles rather than liquid nigrosin. Overall, it is clear that the variability in
- our PSAP data is not sufficiently predicted by the GMD.
- ⁶⁷⁰ Overall, our data show that any possible size dependency in the TAP and PSAP
- ⁶⁷¹ response is smaller than the observed variability between samples. The TAP and
- ⁶⁷² PSAP data exhibit relative standard deviations (RSD) of 19% and 16%,
- ⁶⁷³ respectively, for samples with GMD > 25 nm. Future studies may consider
- 674 correcting PSAP and TAP measurements by the ratios shown in Table 3, which
- ⁶⁷⁵ represent the ratio between the calibrated aerosol-phase nvPM mass
- ⁶⁷⁶ measurements and the previously uncalibrated PSAP and TAP measurements, for
- data above 25 mg kg_{fuel}⁻¹ and N1 > 40%.
- 4.5 Instrument performance for fuels with different composition
- ⁶⁷⁹ Figure 11 shows a category plot of the ratio El_m/mean-El_m (that is, the ordinate of
- $_{680}$ Figure 9) for the different instruments. Data below 100 mg / kg_{fuel} have been
- excluded as this ratio reflects only noise in that region (Figure 9). The symbols
- have been sized by mean N1. The data have been coded by symbol and colour to
- reflect the 6 fuels used in this study, although JP-8 measurements are few in
- $_{\rm 684}$ number due to the EIm of the data set (CFM56-2C1 with JP-8) being typically below
- 685 25 mg / kg_{fuel}.
- ⁶⁸⁶ Figure 11 shows that no substantial difference can be seen for these instruments
- for the nvPM EIm for fuels with different composition; the spread in the data for a
- given fuel is larger than the difference between fuels. Outliers tend to be associated
- with low N1 (small symbols). Because low N1 corresponds to both lower
- concentrations (lower signal-to-noise) and lower exhaust velocities relative to
- ambient wind speeds, these outliers are not surprising.
- ⁶⁹² The instruments in Figure 11 show a linear response to nvPM mass and operate on
- a range of physical principles. This observation indicates that no instrument was





- ⁶⁹⁴ uniquely sensitive to changes in particle size over the observed range, since EIm
 ⁶⁹⁵ was correlated with GMD (Figure 12), as is typical of aviation engines (Saffaripour
 ⁶⁹⁶ et al., 2020). We note that the response of all of these instruments is proportional
- to the MAC of the sample, so that it remains possible that the sample MAC changed with GMD or El_m.
- 4.6 Influence of LII laser fluence
- An additional experiment was performed to test the hypothesis that the laser 700 fluence of the LII 300 may not be sufficiently high to heat nvPM to incandescence 701 in aircraft-engine PM emissions from SAFs at different engine thrusts. This 702 hypothesis is related to electron microscopy evidence (Vander Wal et al., 2014) 703 showing that the degree of graphitization of aircraft-engine soot may be 704 substantially lowered at low thrusts. A lower degree of graphitization may result in 705 a lower LII signal if the 1064 nm MAC is lower (resulting in a lower maximum 706 temperature being reached) or if part of the laser energy leads to carbon annealing 707 rather than thermal excitation (Botero et al., 2021; Ugarte, 1992; Vander Wal and 708 Choi, 1999). If correct, this hypothesis would mean that the nvPM concentrations 709 reported by an LII 300 operated at reduced fluence would be lower than those of a 710 reference LII 300. Higher fluences are also required for nvPM internally mixed 711 with volatile PM, as some laser energy may be lost to volatile evaporation 712 (Michelsen et al., 2015). 713
- Figure 13a illustrates the experiment we performed to test this hypothesis. The 714 figure presents data for SAF1 only; results for other fuels were similar. One 715 "reduced-fluence" LII 300 was programmed to change its Q-switch delay from 716 140 µs to 240 µs, with a randomized order. In this experiment, lower Q-switch 717 delays corresponded to higher laser fluence; the lowest Q-switch delay was the 718 optimal one for this system. Another "reference" LII 300 operated with no change 719 to its Q-switch delay. Figure 13a shows that the reduced-fluence LII reported lower 720 mass concentrations when its Q-switch delay was increased, but returned to the 721 expected values when its Q-switch delay was reduced. 722
- We defined R_{LII} as the ratio of nvPM mass concentrations reported by the reduced-





- fluence and reference LII 300 instruments. Figure 13b shows that R_{LII} was a
- ⁷²⁵ function of Q-switch delay, and therefore laser fluence, for all engine thrust
- conditions. This observation is expected, since LII signals are lower at lower
- ⁷²⁷ fluence (Michelsen et al., 2015) and since we calculated R_{LII} without taking this
- effect into account. We have verified in our laboratory that Q-switch delay is
- ⁷²⁹ inversely proportional to laser fluence for this system and that saturation effects⁷³⁰ are negligible.
- A trend of decreasing R_{LII} with decreasing N1 is evident at moderate and low Q-
- ⁷³² switch delays, which can be interpreted as indicating that the nvPM was more
- graphitic at higher N1 conditions. However, R_{LII} reached a plateau at high fluence
- (smaller Q-switch delay), which is the region where the LII 300 normally operates.
- This plateau was reached at all engine thrusts, with a broader range for the plateau
- at higher thrusts and a decreasing range as the thrust was lowered. Therefore, the
- LII 300 has sufficient fluence and can be expected to perform well for SAF blends
- 738 at all engine thrust conditions.

739 5 Conclusions

- For multiple instruments measuring nvPM number, size, and mass, we observed 740 no evidence of anomalous instrument responses to the exhaust emissions 741 produced by SAF blends relative to reference fuels (REFs) combustion in an IAE 742 V2527-A5 engine. The GMD, GSD, and EI_{num} data for all fuels fell within 20%, 5%, 743 and a factor of 2 of their mean, respectively. Anomalous instrumental responses 744 would have resulted in two groups of data for these parameters, which was not 745 observed. However, a difference between Elnum for instruments located on 746 different-length sampling lines was noted and attributed to a greater sensitivity of 747 EI_{num} than EI_m to the penetration function. 748
- 749
- The majority of nvPM mass measurements by the real-time instruments (CAPS
- PM_{SSA}, LII 300, MSS+, PAX) agreed to within 30% of their geometric mean
- (reference mean), for EI_m above 100 mg/ kg_{fuel}. This lower limit corresponded to a
- mass concentration of approximately 10 μ g m⁻³ (the conversion of EI_m to mass
- varies because the emitted [CO₂] varies), which was the noise level of these





- instruments. The ratio of each real-time measurement with the reference mean was close to unity (maximally 1.24, minimally 0.78) and indicated good precision (all RSDs \leq 17%).
- 758

Integrative nvPM EI_m, calculated from PSD measurements or filter attenuation (TAP and PSAP), fell within a factor of two of the reference mean. The ratio of each integrative measurement with the reference mean was further from unity (maximally 1.50, minimally 0.88) and variability was higher precision (all RSDs \leq 36%). The variability in TAP data was notably low at 14%, and the variability in PSAP data was notably high at 36%, likely due to its operation at a reduced flow rate.

- 766
- Two other instrument- and fuel composition-specific observations were made. A
- dedicated experiment showed that changing the laser fluence of an LII 300 could
- ⁷⁶⁹ influence its reported nvPM mass concentrations at low to moderate fluences. By
- maintaining sufficiently high fluence a plateau region was established, irrespective
- $_{771}$ of thrust or fuel, where reported nvPM mass concentrations were stable and not
- influenced by experimental conditions. Second, additional measurements of
- emissions from JP-8 fuel combusted in a CFM56-2C1 engine indicated the presence
- of very high concentrations of volatile nucleation-mode particles with
- diameter < 20 nm. These measurements reflect a different engine, as well as a fuel
- $_{\rm 776}$ $\,$ with a factor 20 higher sulfur content, and the increased total PM number
- concentration is most likely attributable to the sulfur.
- 778
- Overall, this study found that real-time instruments for the measurement of nvPM
- emissions in aviation turbine engines are comparable whether conventional fuels
- or SAFs are used. Since all real-time measurements were influenced by the MAC
- and no independent measurement of nvPM mass was made, no conclusions about
- the variability thereof can be made from this study.





- 784 5.1 Author contributions
- BEA, PLC, TS, PL, GJS, PDW, and RML designed the study. JCC, TS, PLC, GJS, ECC, SA,
- PDW, RML, ZY, AF, MT, DS, WL, CR, PO, MS, and PL took the measurements. JCC, TS,
- ⁷⁸⁷ BEA, RHM, MAS, ECC, SA, ZY analyzed the data with input from GJS, PL, RML, and
- AF. JCC prepared the figures. JCC and PL drafted the manuscript. All authors
- ⁷⁸⁹ discussed the data interpretation and presentation.
- 790 5.2 Competing interests
- ⁷⁹¹ RML and AF are employed by ARI, which produces the CAPS PM_{SSA} commercially.
- ⁷⁹² ZY was employed by ARI at the time of the study.
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812 5.5 Data availability

- ⁸¹³ The data presented in Figures 4 to 10 are available in the Zenodo repository at
- https://sandbox.zenodo.org/record/950512 as a spreadsheet file. Other data are
- available from the authors upon request.

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1060 7 Figures and Tables

- 1061 Table 1. Properties of the fuels used for the ground-based measurements (fuel
- samples acquired from wing-tank after test).

Property	Method	JP-8	REF3	REF4	SAF1	SAF2	SAF3
Aromatics	ASTM	19.9	18.6	16.5	9.6	10.8	15.2
[vol%]	D1319						
Hydrogen H	ASTM	13.86	13.65	14.08	14.40	14.51	14.04
[mass%]	D7171						
Sulphur, total	ISO	1240	105	5.7	56.8	4.1	58.6
[ppm]	20884						
Naphthalenes	ASTM	1.49	1.17	0.13	0.61	0.05	0.64
[mass%]	D1840						
Smoke point	ASTM	23.0	23.0	27.0	30.0	30.0	28.0
[mm]	D1322						



(sanado	Sunding		Lenell allon
Š	oerator	Operator Instrument	Acronym	measured	duration [s]	Units	functions ^d
Ź	NASA	Particle soot absorption photometer	PSAP	nvPM ^a mass	~	hg m ⁻³	-
		Tricolor absorption photometer	TAP	nvPM ^a mass	-	hg m ⁻³	-
		Scanning mobility particle sizer	SMPS	Total PSD (10 to 278 nm)	45	particles cm ⁻³ , and µg m ⁻³	←
		Thermo-denuder with SMPS	TD-SMPS	nvPM PSD ^b (10 to 278 nm)	45	hg m ⁻³	1, 4
		CO ₂ sensor	LI-COR 7000	CO ₂	-	bpmv	ı
NF	NRC	Cavity-attenuated phase shift PMssA monitor (660 nm)	CAPS (NRC)	nvPM ^a mass	, -	hg m ⁻³	
		Photoacoustic extinctiometer	PAX	nvPM ^a mass	-	hg m- ³	-
		Laser-induced-incandescence	LII 300 (NRC; 2x)	nvPM ^b mass	-	hg m- ³	-
		Catalytic stripper SMPS	CS-SMPS	nvPM PSD (8.6 to 278 nm)	45	particles cm ⁻³	1, 3
MST	ST	AVL Particle Counter Advanced	APC	nvPM number	-	particles cm ⁻³	1, 2
Z	(NARS)	Micro Soot Sensor	MSS Plus	nvPM ^a mass	-	hg m- ³	1, 2
		Laser-induced-incandescence	LII-300 (NARS)	nvPM ^c mass	1	hg m ⁻³	1, 2
		CO ₂ sensor	LI-COR 840A	CO2	-	mdd	
		Differential mobility spectrometer	DMS500	Total PSD (5 to 1000 nm)	-	particles cm- ³	1, 2
ARI	2	Cavity-attenuated phase shift PMsea monitor (630 nm)	CAPS (ARI)	nvPM ^a mass	-	hg m ⁻³	1, 2





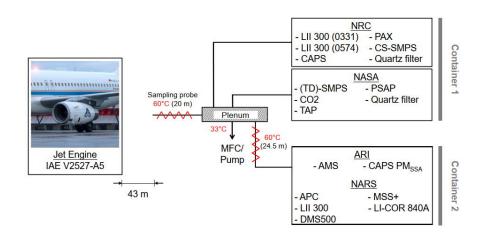


- 1068 Table 3. Summary of the ratios between the Elm of individual instruments and the
- 1069 geometric mean of the Group 1 (real time) instruments. The corresponding raw
- data are shown in Figure 11. Regression: linear regression against Group 1
- $_{1071}$ geometric mean weighted by standard deviations, with k = 2 uncertainties from fit.
- ¹⁰⁷² SD: standard deviation. RSD: Relative SD. Group 1: real time instruments. Group 2:
- ¹⁰⁷³ integrative instruments.
- 1074

		El _{mass} Ratio			Regression		
		vs. Group 1			vs. Group 1		
				RSD			
Group	Instrument	Mean	SD	[%]	Intercept	Slope	
1	CAPS PM _{SSA} (ARI)	0.84	0.08	10	12 ± 19	0.8 ± 0.1	
1	CAPS PM _{SSA} (NRC)	0.99	0.09	9	-0.3 ± 0.8	1.01 ± 0.04	
1	LII (NARS)	1.24	0.18	15	27 ± 6	1.03 ± 0.04	
1	LII (NRC-0331)	1.07	0.1	9	-15 ± 42	1.17 ± 0.16	
1	LII (NRC-0574)	0.78	0.08	10	-17.1 ± 2	0.88 ± 0.08	
1	MSS+	1.07	0.14	13	17.8 ± 5	0.92 ± 0.04	
1	ΡΑΧ	1.06	0.18	17	-15 ± 1	1.21 ± 0.02	
2	CS-SMPS	1.50	0.27	18	12 ± 22	1.02 ± 0.12	
2	TD-SMPS	1.14	0.26	23	-5 ± 1	1.47 ± 0.04	
2	PSAP	0.89	0.32	36	8 ± 16	0.82 ± 0.08	
2	ТАР	0.88	0.12	14	6 ± 6	0.75 ± 0.02	

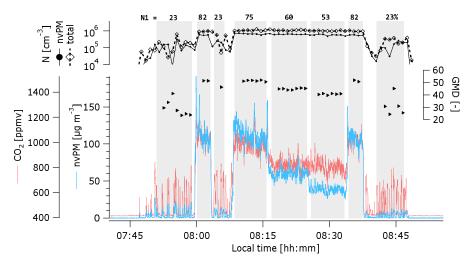






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Figure 1. Schematic of sampling configuration behind the DLR ATRA aircraft. The length and flow rate of sampling lines from the manifold to the various instruments varied as described in the text. The NRC and NASA instruments were all placed within Container 1, while the NARS and ARI instruments were placed in Container 2. For simplicity, the figure omits a short heated line connecting the first plenum to the NARS. The ARI instruments were downstream of all NARS instruments except the DMS500 (see Lobo et al., 2016 for detailed NARS diagram). NARS = North American Reference System.

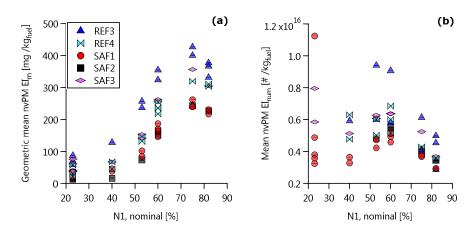


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Figure 2. Illustration of a typical test run. Variation in the CO₂ concentration was not due to instrument noise, as illustrated by the CO₂ measurements prior to and following sampling. A representative nvPM mass instrument is shown by the blue trace. Sizing information (GMD) is shown by the black symbols (triangles: GMD; diamonds with dashed line: total PM number; spheres with solid line: nvPM number measured with the CS-SMPS).



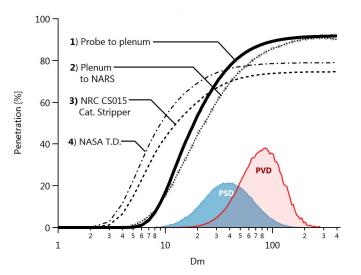




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Figure 3. Relationship between nvPM (a) EI_m and (b) EI_{num} with N1 for all data obtained with the V2527-A5 engine. The trends shown in this plot are discussed further in the companion article (Schripp and NDMAX-Team, 2021). The ordinate values are the geometric mean discussed in the text.

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Figure 4. Penetration functions for the main probe-to-plenum sampling line as well as
 other components in the sampling system. Shaded areas illustrate a representative
 particle size (PSD) and volume (PVD) distribution measurement with GMD 34 nm and GSD
 1.72. PSD data for all test points and instruments are provided in the supplement. NARS:

North American Reference System; CS015: Catalytic Stripper; T.D.: thermodenuder.





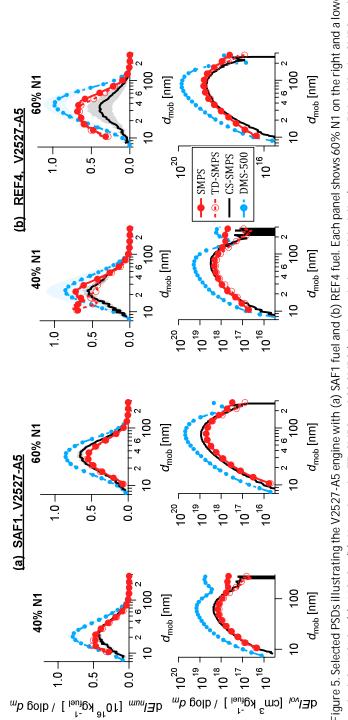
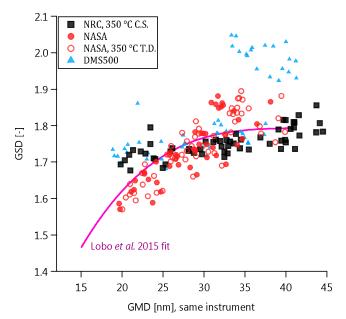


Figure 5. Selected PSDs illustrating the V2527-A5 engine with (a) SAF1 fuel and (b) REF4 fuel. Each panel shows 60% N1 on the right and a lower N1 on the left: 40% for (a), 60% for (b). Note that the TD-SMPS and CS-SMPS (red open circles and black line) represent nvPM, while the SMPS and DMS500 represent vPM. 1106 1107 1108

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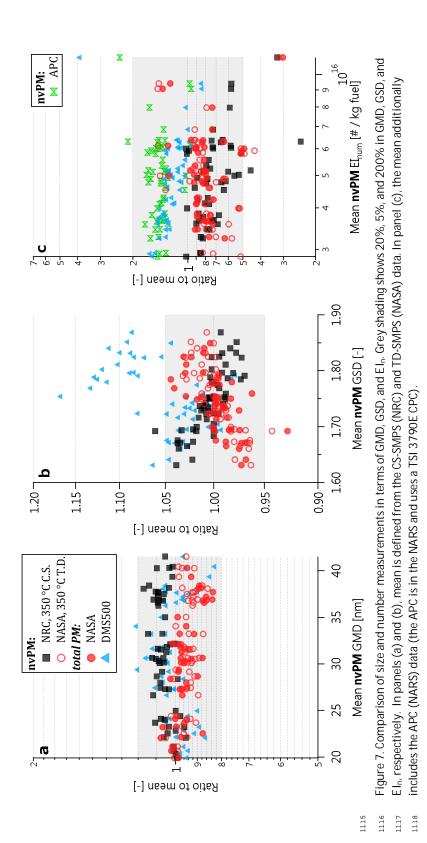




1110	Figure 6. GSD versus GMD data as by measured by each particle sizer for all test points.
1111	Higher GSDs for the DMS500 correspond to bimodal PSDs (non-volatile and volatile
1112	modes). Note that size-dependent particle losses (see penetration functions in Figure 4)
1113	may affect both GSD and GMD. Based on Figure 12, the TD-SMPS (NASA) data may be more
1114	accurate than the CS-SMPS data (see text). Fit is from Lobo et al. (2015c).







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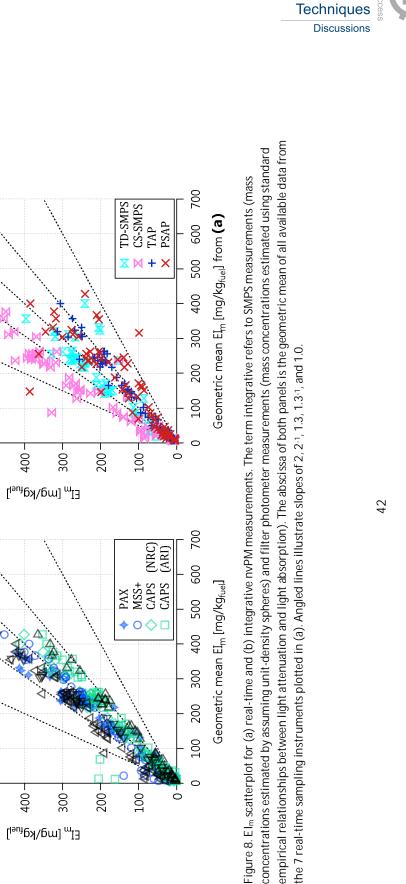
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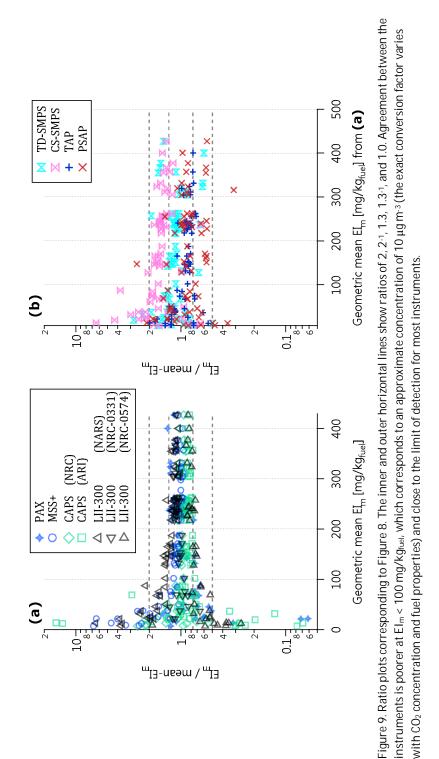
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Atmospheric 9

Measurement

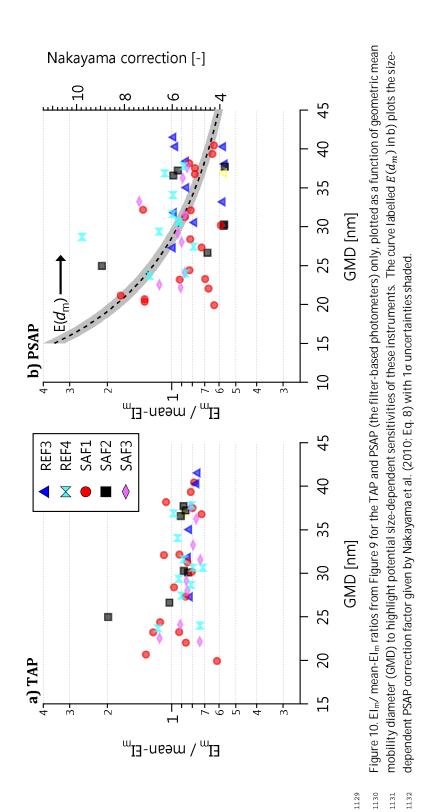






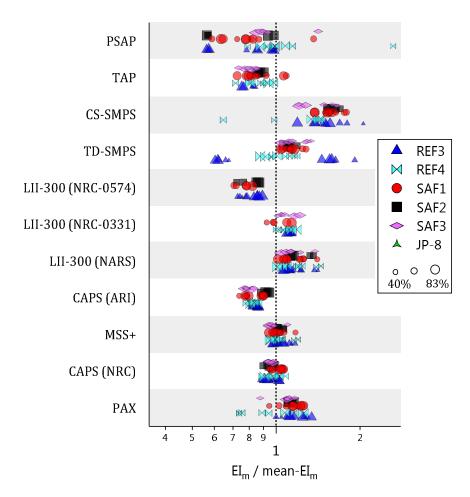












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Figure 11. Ratios of Figure 9 grouped by fuel. All fuels except JP-8 were combusted in the V2527-A5 engine; JP-8 was combusted in the CFM56-2C1 engine. Shading is to guide the

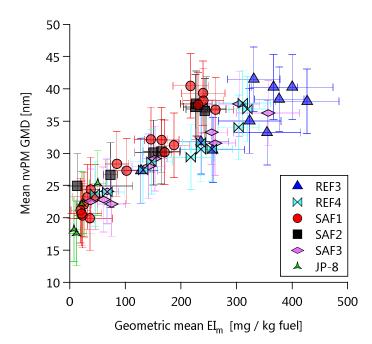
eye. Symbols are sized by N1 thrust. Plot excludes data where $EI_m < 25 \text{ mg/kg}_{fuel}$ and N1

thrust below 40% to minimize the effects of instrument noise and wind speed,

¹¹³⁸ respectively, on the ratios.







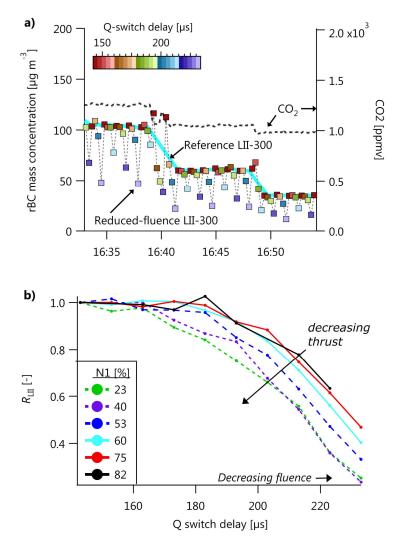
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- Figure 12. Scatterplot of the mean nvPM GMD within test points against geometric
- mean nvPM EI_m from Figure 8a. The correlation with GMD and EI_m indicates that Figure 9
- implicitly represented different particle sizes.

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Figure 13. (a) LII 300 experiment time series, where one LII 300 was operated with increased Q-switch delays to reduce its laser fluence (squares) and the other was operated at standard fluence (solid line). CO_2 data are also shown for context. (b) The ratio R_{LII} of the concentration reported by the reduced-fluence LII divided by the reference LII. It is evident from (b) that the standard high-fluence conditions generate data that are independent of N1 thrust, and that moderate- and low-fluence conditions (Q-switch delays greater than about 165 to 185 μ s) display a weak dependence on thrust.