

1 **Supporting Information for**  
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3 Comparison of Particle Number Emissions from In-Flight Aircraft Fueled with Jet A1, JP-5 and an  
4 Alcohol-to-Jet Fuel Blend

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11 Number of pages: 25

12 Number of figures: 14

13 Number of tables: 4

## **S1 Particle Transport and Counting Efficiency in the Isokinetic Sampling System and Particle Counters**

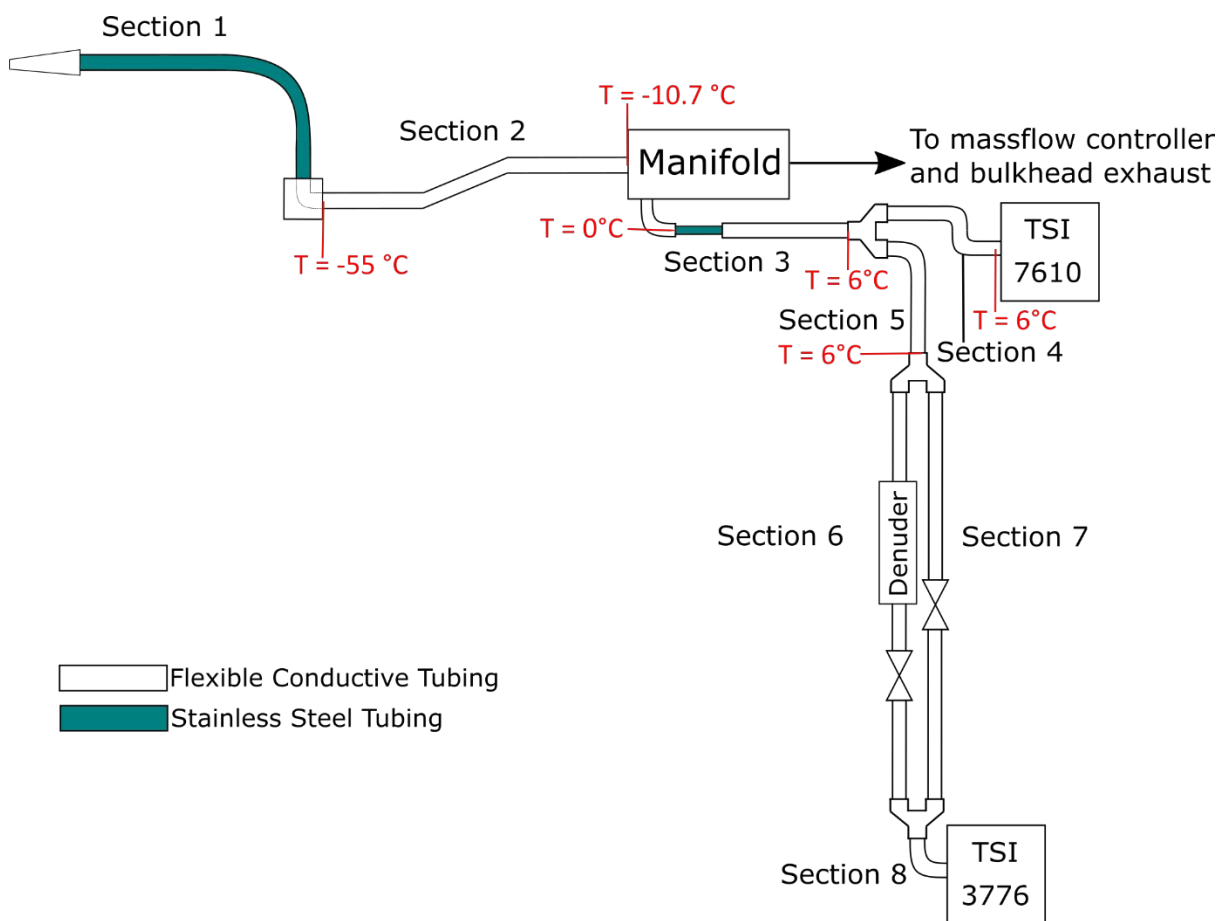
### **S1.1. Introduction**

This section of the supporting information details the calculation of the transport losses and resulting counting efficiencies of the two CPCs. Particle losses through the sampling lines are important because it will affect the particle concentrations measured by the CPCs resulting in fewer particles detected by the CPCs than actual particles in the sample flow. The counting efficiencies of the CPCs can also be affected as particles below a certain size may not be able to fully penetrate the sampling lines which increases the ‘effective’ cut-off diameters of the CPCs.

### **S1.2. Particle Transport Efficiency through the Sampling System**

A schematic of the isokinetic sampling system of the National Research Council Canada (NRC) CT-133 jet aircraft is shown in Figure S1. The sample flow enters the system through an isokinetic probe which consists of a conical tip and a bent tube (Section 1). The conical tip is attached to a stainless-steel tube comprised of a straight length and a 90° bend. The sample flow subsequently enters a coupler after the first section, which has a 90° elbow. The flow is then directed through Section 2 of the sampling system consisting of a conductive flexible tube that is bent at two points with bending angles of 20° and -20°. The sample is drawn through a manifold, where a small fraction of the sample is directed towards the lines leading to both particle counters and the rest of the sample flows to the mass flow controller and bulkhead exhaust. After the manifold, the sample enters Section 3 which consists of an elbow and a straight stainless-steel tubing connected to an additional length of flexible conductive tubing leading to a Y fitting that splits the sample flow to both condensation particle counters (CPC). Section 4 leads the sample to the TSI Model 7610 CPC. Section 5 leads to a second Y fitting which splits the flow between the catalytic denuder and the bypass line to the TSI Model 3776 CPC. Both Sections 6 and 7 contain pinch valves that are used to control whether the sample goes through the catalytic denuder (Section 6) to measure non-volatile particles or the bypass line (Section 7) to measure total particles. Section 8 is the last

section of the particle sampling system which leads the sample to the TSI Model 3776 CPC. Table S1 shows a summary of the total lengths and inner diameters for each section.



**Figure S1.** Schematic of the particle sampling system.

Particle losses in the system will be a function of the particle size. We assume contrail particles that will be measured by the system will consist of a small soot particle core (with diameter less than 100 nm) and a shell of frozen water at very low temperatures at high altitudes. Therefore, it is important to consider the effect of pod temperature on the sampled particles and their estimated size as they enter the particle counter. It is known from previous experimental conditions that at top-of-climb, the pod temperature is  $\sim 20^{\circ}\text{C}$  and at top-of-descent, the pod temperature is  $\sim 6^{\circ}\text{C}$ . A pod temperature of  $6^{\circ}\text{C}$  is considered in this study as the worst-case scenario for the effect of temperature on sampled particles. A heat transfer analysis was conducted on flow temperature as it traveled through the sampling system.

Figure S1 shows the estimated flow temperature for various sections of the sampling system which are also summarized in Table S1. The flow temperature also affects air properties, such as density, viscosity, and volumetric flow rate, in different sections of the sampling system which, in turn, affect particle loss. Figure S1 shows that up to the manifold, the flow temperature is below zero and the water in the contrail particles will not evaporate; however, at the beginning of Section 3, the flow temperature reaches zero and increases up to 6 °C (*i.e.* pod temperature) at the end of that tube. Calculations show that the residence time of particles in Section 3 is much larger than their evaporation time. Therefore, particles reach the particle counter as soot with no condensed water vapour on them.

Three mechanisms for particle loss in the sampling system are considered: Diffusion loss in the tubes, inertial deposition loss in the bends and losses through the catalytic denuder. The extent of particle loss in each mechanism depends on the flow regime in the probe. Therefore, Reynolds number is an important parameter to consider when calculating particle loss in the probe. Because eddies do not form in expansions with a half angle less than 4°, inertial deposition particle loss is negligible in the conical tip<sup>1</sup>. However, diffusion loss must be still considered for the conical tip.

Figure S1 also shows a simplified model of the actual sampling system for the estimation of particle losses. As it will be shown later, diffusion losses depend on tube length; therefore, the overall length of conical tip and the bent tube was considered as 610 mm for diffusion loss calculations. The overall straight section of the stainless-steel tube was 330 mm long, which was used to estimate inertial deposition losses in straight tubes.

Based on flight data, it was assumed that the isokinetic probe was at ambient conditions of –55 °C and 23 kPa. The rest of the sampling system was in a heated pod at 6 °C; however, it took some time for the sampled flow to heat up to the pod temperature. In the particle loss analysis, the flow temperature was assumed to be –55 °C in Section 1, –an average of –33 °C in Section 2, and average of 3 °C in Section 3, and 6 °C in Sections 4 to 8. The density and viscosity of the air was calculated based on the temperature

in each section. Assuming that the mass flow controller regulated the volumetric flow rate at 25 litres per minute at ambient pressure and temperature, Reynolds number was calculated to be 1,274 and 1,326 in Section 1 and Section 2, respectively. Therefore, the flow was laminar in both Sections 1 and 2. The particle counters both drew in a volumetric flow rate of 1.5 L/min (at 23 kPa and 6 °C) via their own pump. Thus, the Reynolds number in the stainless-steel tube and the conductive tubing of Section 3 was 166 and 110, respectively. The Reynold's number in Sections 4 to 8 was 55 and, therefore, the flow regime in all the remaining sections was laminar as well.

**Table S1.** Summary of each section of the sampling system

Section	Total length (mm)	ID (mm)	Flowrate (L/min)	Temperature (°C)	Reynolds number
1	610	10.7	25	-55	1274
2	483	9.5	25	-33	1326
3 (Steel)	190.5	4.3	3	3	166
3 (Conductive tubing)	165	9.5	3	6	110
4	76	9.5	1.5	6	55
5	210	9.5	1.5	6	55
6	585	9.5	1.5	6	55
7	832	9.5	1.5	6	55
8	127	9.5	1.5	6	55

88 **S1.2.1. Diffusion Loss**

89 Kulkarni et al.<sup>1</sup> suggests the following set of equations to calculate transport efficiency (*i.e.* penetration)  
 90 due to diffusional loss:

$$P_{\text{diff}} = \exp(-\xi \cdot Sh) \quad (\text{S1})$$

$$\xi = \frac{\pi DL}{Q} \quad (\text{S2})$$

91 where  $L$  is the tube length,  $Q$  is the volumetric flow rate, and  $D$  is the particle diffusion coefficient.  
 92 Sherwood number,  $Sh$ , for laminar flow is calculated by the following empirical equation

$$Sh_{\text{laminar}} = 3.66 + \frac{0.0668 \frac{d}{L} \text{Re} \cdot \text{Sc}}{1 + 0.04 \left( \frac{d}{L} \text{Re} \cdot \text{Sc} \right)^{2/3}} \quad (\text{S3})$$

93 Reynolds number,  $\text{Re}$ , and Schmidt number,  $\text{Sc}$ , in Eq. S3 is defined as

$$\text{Re} = \frac{\rho U d}{\mu} \quad (\text{S4})$$

$$\text{Sc} = \frac{\mu}{\rho D} \quad (\text{S5})$$

94 where  $d$  is the inner tube diameter. Particle diffusion coefficient,  $D$ , is defined by

$$D = \frac{kTC_c}{3\pi\mu d_p} \quad (\text{S6})$$

95 where,  $C_c$  is the Cunningham slip correction factor (significant for particles in the range of or smaller than  
 96 the mean free path of the air molecules) and  $d_p$  is the particle diameter. The mean free path of air  
 97 molecules,  $\lambda$ , was calculated using<sup>2</sup>

$$\lambda = \lambda_0 \left( \frac{T}{T_0} \right) \left( \frac{P_0}{P} \right) \left( \frac{1 + \frac{S}{T_0}}{1 + \frac{S}{T}} \right) \quad (S7)$$

98 where  $P$  is the pressure,  $T$  is the temperature,  $S$  is the Sutherland constant ( $S = 110$  K for air)<sup>2</sup> and  $\lambda_0$ ,  $P_0$   
 99 and  $T_0$  are the reference mean free path, pressure and temperature respectively. At a reference temperature  
 100 and pressure of  $T_0 = 293.15$  K and  $P_0 = 101.3$  kPa, the mean free path is  $\lambda_0 = 0.0665$   $\mu\text{m}$ .<sup>1</sup> At a pressure of  
 101 23 kPa and temperatures of  $-55$  °C and  $6$  °C, the mean free path of air molecules was estimated to be  
 102  $\sim 199$  nm and  $\sim 275$  nm, respectively. The Cunningham slip correction factor was calculated using<sup>1</sup>

$$C_c = 1 + \frac{\lambda}{d_p} \left( 2.33 + 0.966 \exp \left( -0.499 \frac{d_p}{\lambda} \right) \right) \quad (S8)$$

### 103 **S1.2.2. Inertial Deposition in the Bend**

104 Pui et al.<sup>3</sup> gives the following empirical relation for inertial deposition in a laminar flow through a  
 105 bend:

$$P_{\text{depos, laminar, bend}} = \left( 1 + \left( \frac{Stk}{0.171} \right)^{0.452 \left( \frac{Stk}{0.171} \right) + 2.242} \right)^{-\frac{2}{\pi} \theta} \quad (S9)$$

$$Stk = \frac{\tau U}{d} \quad (S10)$$

106 where  $Stk$  is the Stokes number and  $\theta$  is the angle of the bend in radians. In Eq. S10,  $\tau$  is the relaxation  
 107 time,  $U$  is the flow velocity and  $d$  is the tube diameter.

### S1.2.3. Losses in the Catalytic Denuder

The losses in the Catalytic Instruments, Model CS015 denuder was modeled by using the provided equation in the instrument manual,<sup>4</sup>

$$P = \left( x_1 e^{-\frac{x_2}{d_p^2}} + x_3 e^{-\frac{x_4}{d_p^2}} \right) x_5 \quad (\text{S11})$$

where  $d_p$  is the particle diameter (in nm) and  $x_1 - x_5$  are fitting parameters with the values shown in Table S2.

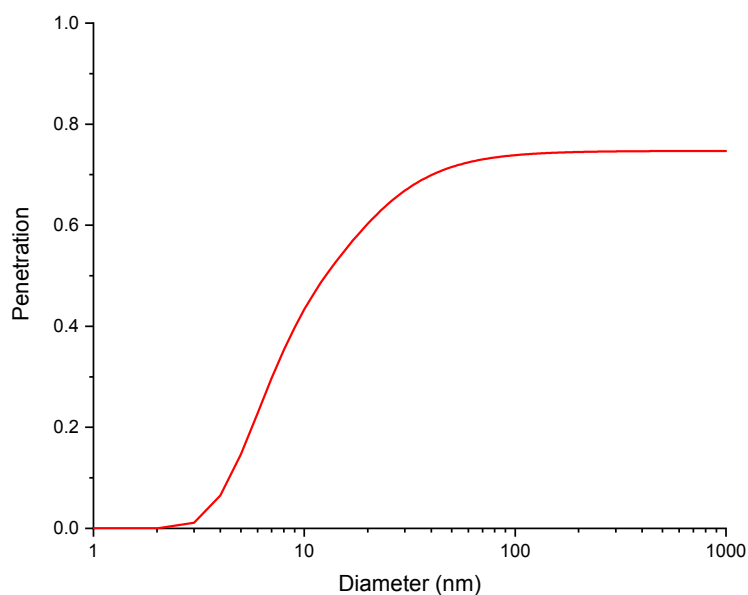
**Table S2.** Fit parameters for Equation S11

Parameter	Value
$x_1$	0.190
$x_2$	499
$x_3$	0.925
$x_4$	36.0
$x_5$	0.670

The resulting penetration efficiency curve is shown in Figure S2. As can be seen in Figure S2, the catalytic denuder has a maximum penetration efficiency of 75% once particles reach 100 nm or larger.



120 Therefore, the measurement of non-volatile particles will experience a large amount of losses due to the  
 121 denuder.



122  
 123 **Figure S2.** Penetration efficiency of particles in the catalytic denuder.

124  
 125

### 126 **S1.3. Counting Efficiency of the Particle Counters**

127 The cut-off diameters of the counting efficiency of the TSI 3776 CPC and TSI 7610 CPC were found  
 128 by fitting data provided in the TSI Model 3776 manual<sup>5</sup> and data estimated from Zhang and Liu<sup>6</sup> for  
 129 standard condition operation; respectively. A sigmoidal logistic fit was applied with the form shown in  
 130 Eq. S12 and the parameters for both CPCs are shown in Table S3.

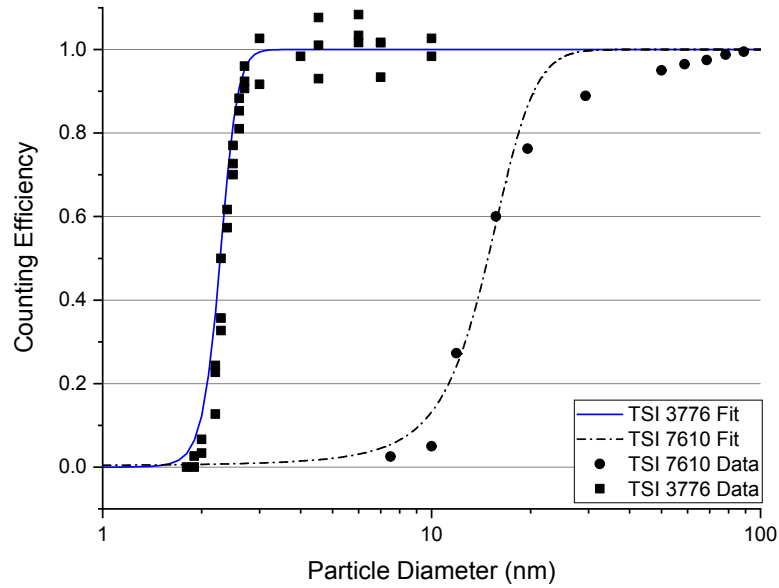
$$y = \frac{1}{1 + e^{-k(x - x_c)}} \quad (\text{S12})$$

131 **Table S3.** Parameters for logistic fit for both CPCs.

Parameters	TSI 3776	TSI 7610
$k$	7.11741	0.38999

$$x_c \quad 2.27708 \quad 14.82943$$

The resulting counting efficiencies of the TSI 3776 CPC and the TSI 7610 CPC are shown in Figure S3. The 3776 CPC also has a much smaller cut-off diameter,  $d_{50}$ , of 2.5 nm compared to the 7610 CPC  $d_{50}$  of approximately 15 nm.



**Figure S3.** Counting efficiency of TSI 3776 and 7610 CPCs.

#### S1.4. Overall Counting Efficiency of the Sampling System and Particle Counters

The overall penetration of particles through the sampling system takes into account the three different particle loss mechanisms in each tube section and bends, the catalytic denuder as well as the counting efficiency of the CPCs and was calculated using

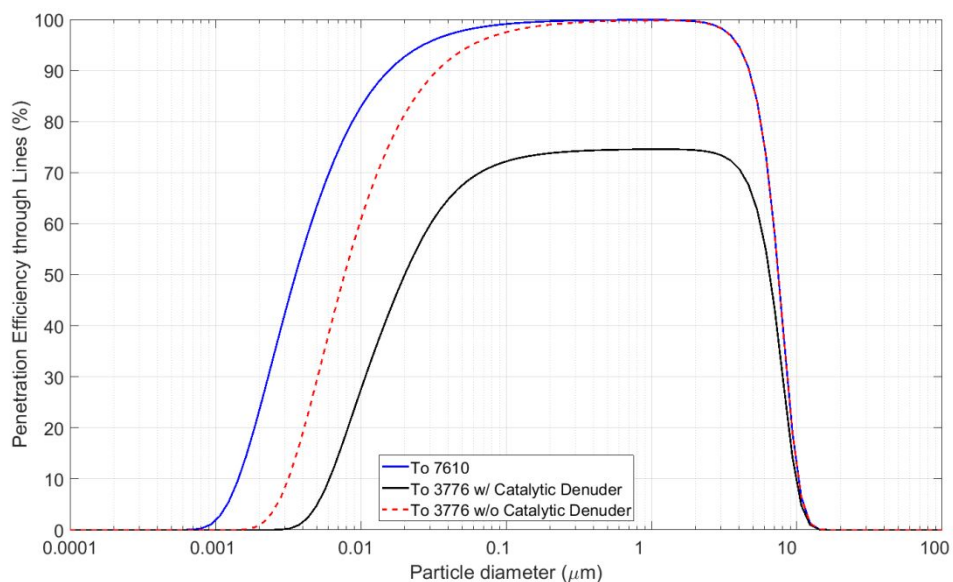
$$P_{\text{overall}} = \frac{n_{\text{out}}}{n_{\text{in}}} = P_{\text{diff}} \times P_{\text{depos, bend}} \times P_{\text{denuder}} \times P_{\text{CPC}} \quad (\text{S13})$$

This assumes that the particle concentration is uniformly distributed across the tube at the entrance to each section.

## S1.5. Results

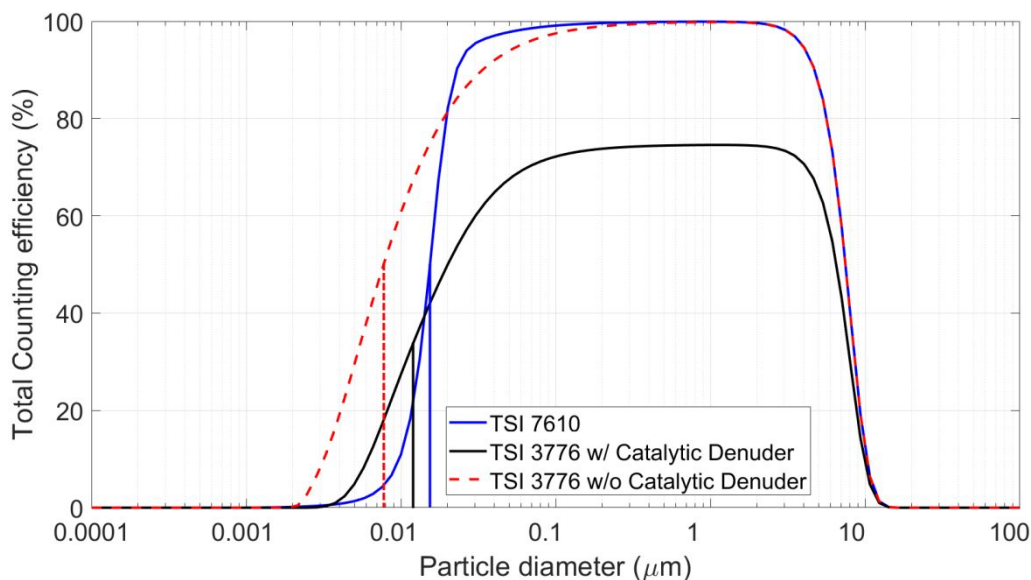
Figure S4 shows the overall penetration of particles through the system before reaching the CPCs. It can be seen that with the current geometry, particle losses in the lines leading to the TSI 7610 CPC are the lowest with a  $d_{50}$ , the diameter where the penetration efficiency is less than 50%, of 3.5 nm. For the bypass line leading to the TSI 3776 CPC, the  $d_{50}$  is slightly larger at 7.7 nm. Finally, the catalytic denuder has the largest impact on the penetration efficiency by reducing the maximum penetration efficiency to 75%, which leads to the largest  $d_{50}$  of 13.3 nm.

It should be noted that particle loss is negligible for the straight sections because the flow regime is laminar. Nevertheless, particle loss due to deposition in bends is still important mainly for particles larger than 10  $\mu\text{m}$ . We expect particles in Sections 1 and 2 of the sampling system to contain contrail particles larger than 1  $\mu\text{m}$  because these sections are not sufficiently warm to evaporate the condensed water; however, deposition loss in the bends is negligible for particle sizes up to 3  $\mu\text{m}$ . Therefore, diffusional losses are the most significant contributor to the penetration efficiency of most particles through the sampling system. Due to the higher temperature of the flow in Sections 3–8, all water will evaporate and only soot particles survive, which generally have a median size less than 100 nm<sup>7,8</sup>. As seen in Figure S4, the sections leading to the TSI 7610 CPC and the bypass line leading to the TSI 3776 CPC have negligible particle losses for particles larger than 100 nm but the penetration efficiency begins to drop as the particle diameter decreases. As mentioned, the line including the catalytic denuder and leading to the TSI 3776 CPC has a penetration efficiency of 75% and has greater particle losses compared to the other two lines.



**Figure S4.** Overall particle penetration efficiency in the sampling system before the CPCs

Figure S5 shows the total counting efficiency of the sampling system including the counting efficiency of the CPCs. The resulting effective  $d_{50}$  of the TSI 3776 CPC and sampling system became 7.7 nm and 13.3 nm for the bypass line and catalytic denuder line, respectively. On the other hand, the effective  $d_{50}$  of the TSI 7610 CPC and sampling system remained relatively unchanged at 15.4 nm.



**Figure S5.** Total overall counting efficiency as measured by the CPCs.

## S2 Uncertainty Calculation

As shown in the manuscript, the calculation of the ratio of particle emissions indices was determined by:

$$EI_{\text{Ratio}} = \frac{EI_{N_{\text{Alt}}}}{EI_{N_{\text{Jet A1}}}} = \frac{\left( \frac{N - N_{\text{bg}}}{P_{\text{CPC}} \times (y_{\text{NO}_x} - y_{\text{NO}_x\text{bg}})} \right)_{\text{Alt}}}{\left( \frac{N - N_{\text{bg}}}{P_{\text{CPC}} \times (y_{\text{NO}_x} - y_{\text{NO}_x\text{bg}})} \right)_{\text{Jet A1}}} \quad (\text{S14})$$

A Monte Carlo simulation was done using the bias uncertainties for the particle number,  $N$ , background particle number,  $N_{\text{bg}}$ ,  $\text{NO}_x$  concentration,  $y_{\text{NO}_x}$ , and the  $\text{NO}_x$  background concentration,  $y_{\text{NO}_x\text{bg}}$ . The uncertainty in the CPC pressure,  $P_{\text{CPC}}$ , was assumed to be negligible because the uncertainty in  $N$  was expected to be much larger. The precision uncertainty was accounted for by combining multiple segments from the same flight where possible. For each segment (or flight) a Monte Carlo simulation was performed where the top and bottom parts of Eq. S14 were calculated separately 10,000 times and were then used to calculate 10,000 ratios. Since the same instrument was used for both the alternate and Jet A1 fuel measurements, by taking the ratio, the bias uncertainties would ideally cancel out (ie. There would be no uncertainty in the particle number,  $N$ , and  $\text{NO}_x$  concentration,  $y_{\text{NO}_x}$ ). By including the uncertainties for  $N$  and  $y_{\text{NO}_x}$ , a worst case scenario is being assumed where there is some variation in the performance of the instruments during the back-to-back measurements of the exhaust plumes from the different fuels.

The uncertainty in the TSI 7610 CPC was taken from the manufacturer's specifications and was assumed to have a Gaussian distribution with  $2\sigma = \pm 10\%$  of reading for the Monte Carlo simulation. The TSI 3776 CPC on the other hand was assumed to have an uncertainty with a Gaussian distribution with  $2\sigma = \pm 30\%$  of reading. This higher value was chosen instead of the manufacturer specified 10% because a study by Takegawa et al.<sup>9</sup> found that the 3776 CPC overestimated particle concentrations by 20–40% for particles with mobility diameters of 11 and 48 nm at absolute inlet pressures of 102 and 60 kPa. Also, in one flight segment (Dec 21 with Jet A1 fuel) the CPC concentration sometimes reached concentrations

near  $1 \times 10^6$  where coincidence error also increases. Thus, the increased uncertainty ( $2\sigma = \pm 30\%$  of reading) is chosen to account for errors due to low pressure operation and coincidence.

The uncertainty in the background was calculated by taking half of the difference between the minimum and maximum background value immediately before or after the test section, represented as  $\varepsilon_{N_{bg}}$ . The background concentration is not expected to be constant throughout the atmosphere may be changing during the flights. A square distribution was used for the background uncertainty as it was assumed that there was an equal chance for the background value to fall anywhere between the upper and lower limits.

In the past, NRC had calibrated the Thermo 42i NO<sub>x</sub> analyzer over a range of NO<sub>x</sub> concentrations and ambient pressures and provided the following calibration equations:

$$y_{NO_x} = f_1(P)x + f_2(P) \quad (S15)$$

$$f_1(P) = (-4.39 \times 10^{-7})P^3 + (3.35 \times 10^{-4})P^2 - (9.13 \times 10^{-2})P + 9.58 \quad (S16)$$

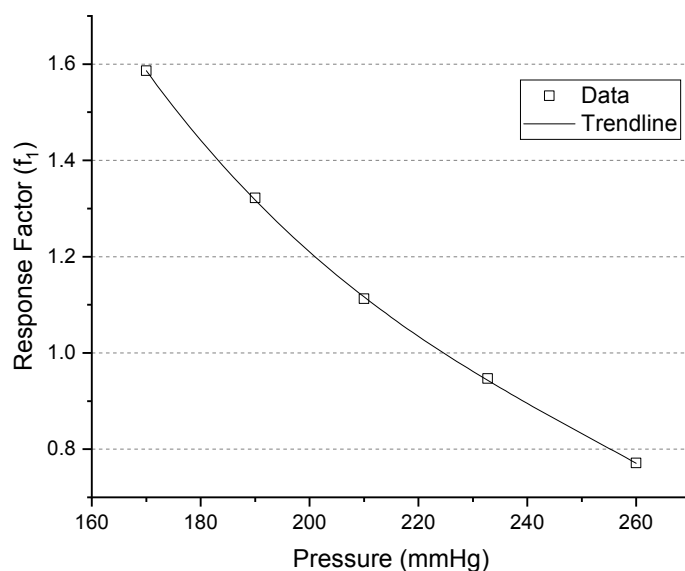
$$f_2(P) = (7.87 \times 10^{-7})P^3 - (4.85 \times 10^{-4})P^2 + (9.3010^{-2})P - 4.98 \quad (S17)$$

where  $x$  is the measured NO<sub>x</sub> concentration and  $P$  is the ambient pressure in mmHg. Eq. S16 and S17 were trend lines fitted to data for the slopes (response factor) and intercepts (zero offset) of the instrument response at different pressures. These equations were applied to the measured NO<sub>x</sub> concentrations before being used to calculate the particle number emission index ratio. The uncertainty in the calibration becomes:<sup>10</sup>

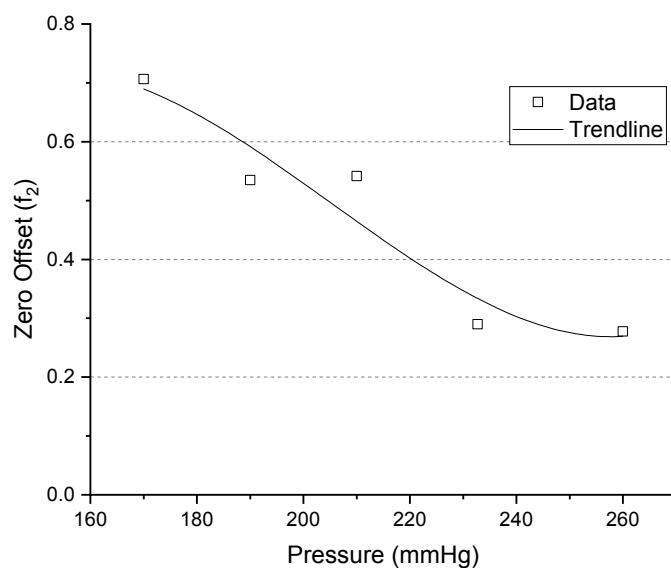
$$\varepsilon_y^2 = \left( \frac{\partial y}{\partial f_1} \frac{\partial f_1}{\partial P} \right)^2 \varepsilon_{f_1}^2 + \left( \frac{\partial y}{\partial f_2} \frac{\partial f_2}{\partial P} \right)^2 \varepsilon_{f_2}^2 + \frac{\partial y^2}{\partial x} \varepsilon_x^2 \quad (S18)$$

where  $\varepsilon_{f_1}$  and  $\varepsilon_{f_2}$  are the uncertainties in Eq. S16 and S17 respectively. The values for  $\varepsilon_{f_1}$  and  $\varepsilon_{f_2}$  were determined by finding the largest difference between the trend lines and actual data points and were found

to be  $4.34 \times 10^{-3}$  and  $7.65 \times 10^{-2}$ , respectively. The calibration curves are shown in Figure S6 and Figure S7. The uncertainty in the measured  $\text{NO}_x$  concentration,  $\varepsilon_x$  was taken as the uncertainty of the calibration gas used for the calibration of the Thermo 42i  $\text{NO}_x$  analyzer ( $\varepsilon_x = 1\%$ ). The uncertainty in the measured  $\text{NO}_x$  concentration,  $\varepsilon_{y\text{NO}_x}$ , was then calculated using Eq. S18. For the test conditions in this study  $\varepsilon_{y\text{NO}_x}$  ranged between 1.3% and 1.4%. In the Monte Carlo analysis, it was assumed that this uncertainty was Gaussian.



**Figure S6.** The  $\text{NO}_x$  calibration response factor as a function of pressure.



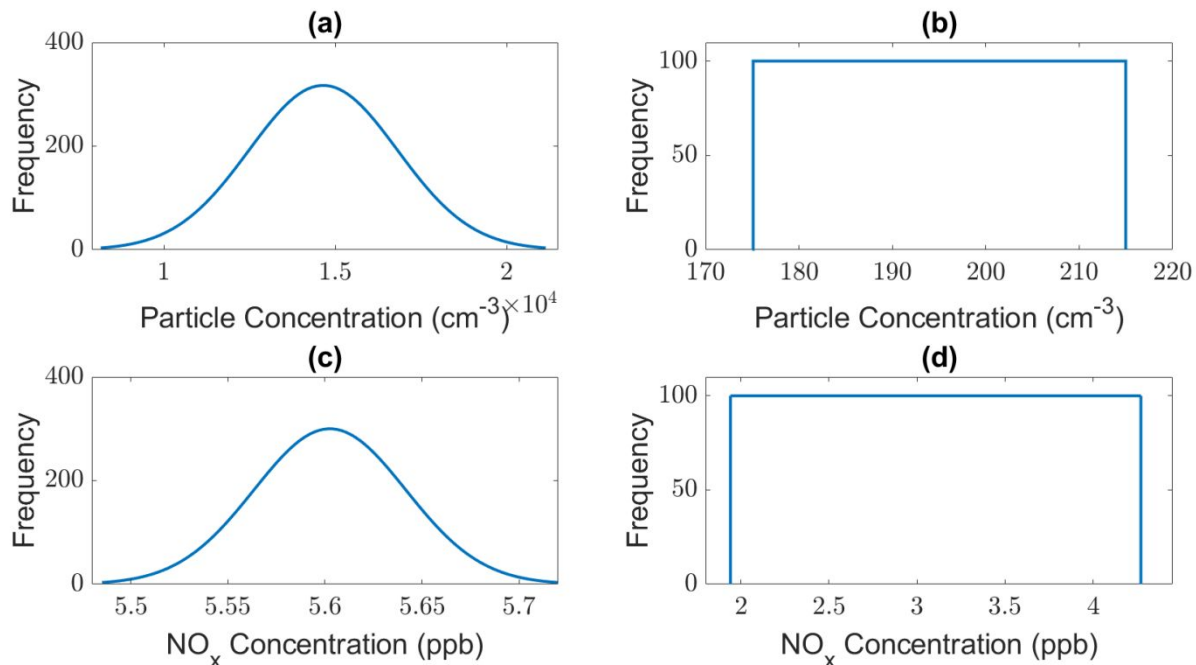
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221 **Figure S7.** The  $\text{NO}_x$  calibration zero offset as a function of pressure.

222 The  $\text{NO}_x$  background uncertainty was determined by taking half of the difference between the  
 223 minimum and maximum of the trend lined fitted to the approximated background data. Thus, a square  
 224 distribution was used for the Monte Carlo simulation.

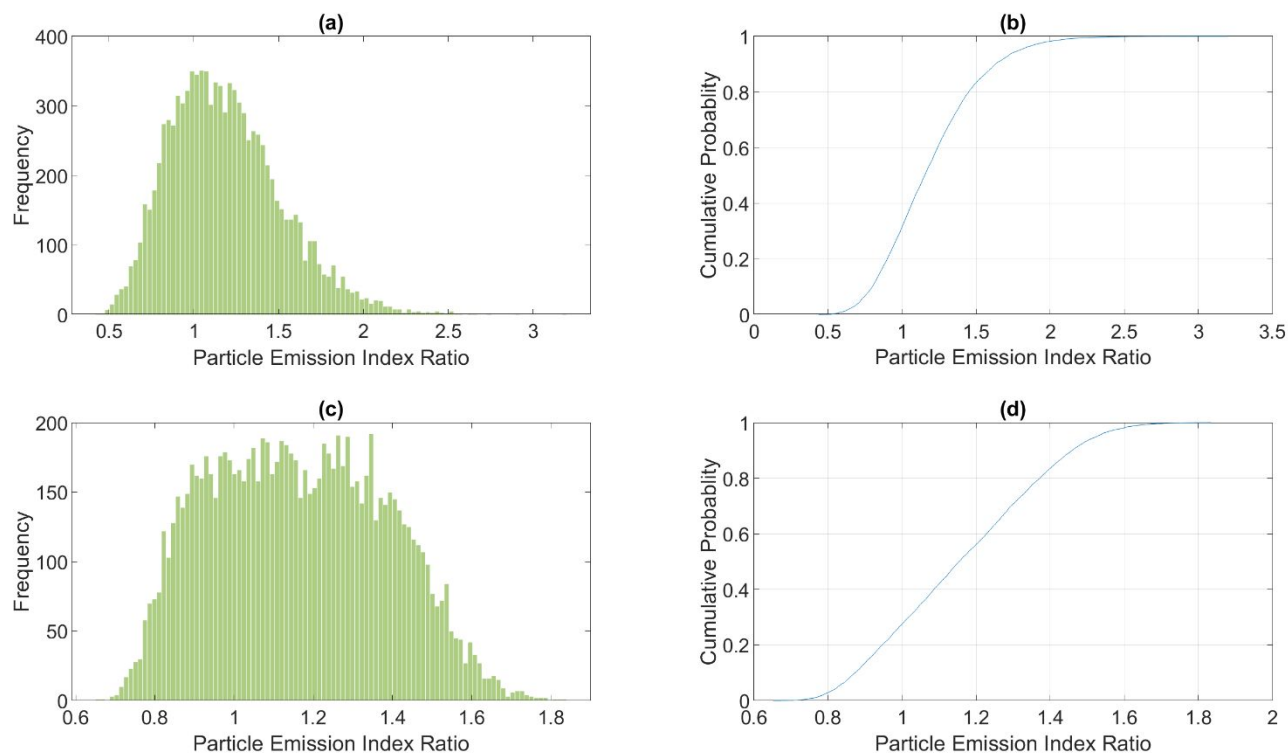
225 Sample histograms plots for the particle and  $\text{NO}_x$  concentration and background results for one segment  
 226 of the Dec 21 Jet A1 flight are shown in Figure S8. Only sample inputs for one type of fuel is shown  
 227 because all fuels had similar inputs.





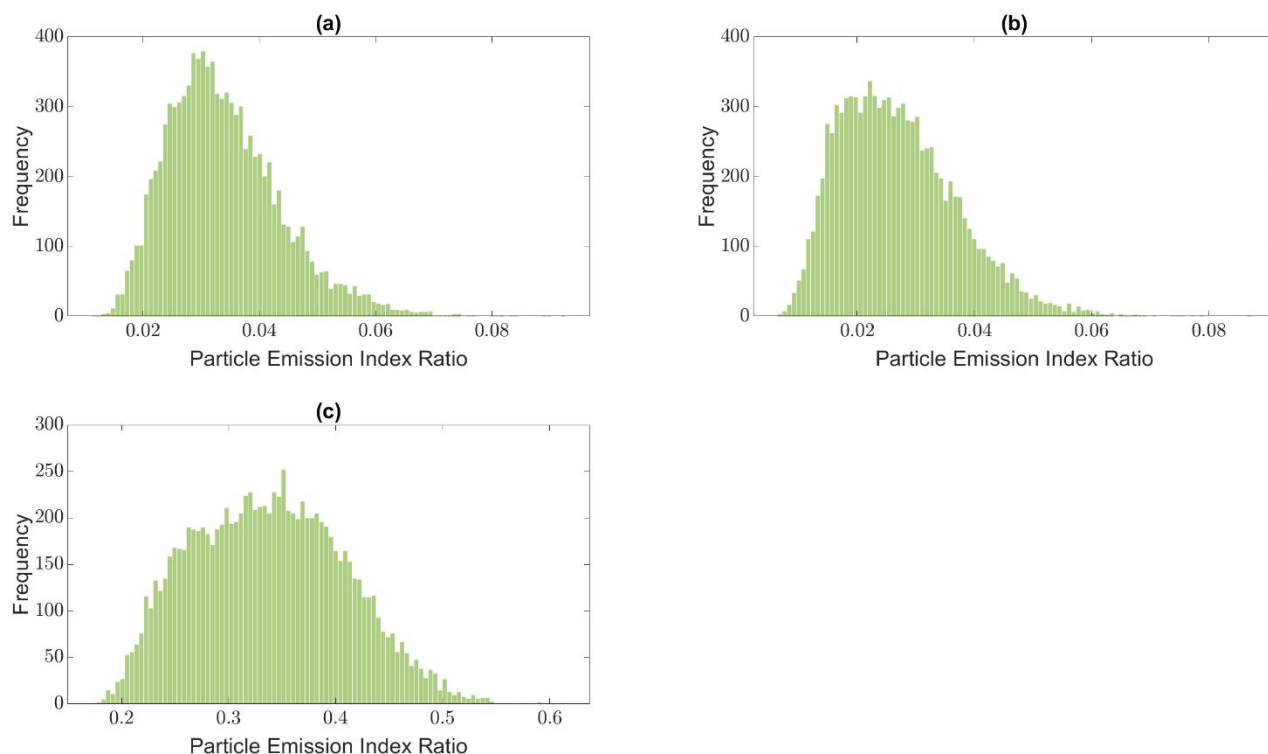
228  
 229 **Figure S8.** Sample inputs for the Monte Carlo Simulation for one segment of the Dec 21 Jet A1  
 230 flight (alternative fuels had similar inputs as well); (a) particle number concentration (TSI 3776),  
 231 (b) background particle concentration (TSI 3776), (c)  $\text{NO}_x$  concentration and (d) background  
 232  $\text{NO}_x$  concentration.

233 For the Nov 20 flight, the ratio of particle emission indices between JP-5 and Jet A1 were determined.  
 234 The resulting histogram plots and cumulative distribution plots for the particle emission index ratios of  
 235 JP-5/Jet A1 from the 3776 CPC and 7610 CPC are shown in Figure S9. By using the cumulative  
 236 distribution plots, the upper and lower limits were taken at the 95% and 5% levels respectively.



**Figure S9.** Monte Carlo results for Nov 20 total particle emission index ratio. (a) distribution for the TSI 3776 (b) cumulative distribution for the TSI 3776 (c) distribution for the TSI 7610 (d) cumulative distribution for the TSI 7610

The Dec 21 flights included a total of 2 separate denuded and non-denuded segments for the ATJ-SPK blend fuel case and 1 of each denuded and non-denuded segments for the Jet A1 case. For the 2 denuded and non-denuded segments of the ATJ-SPK blend fuel case, the average of the two separate segments was first taken before a ratio was calculated. A summary of the results for all cases is shown in Table S4.



**Figure S10.** Monte Carlo results for Dec 21 flight particle emission index ratio between ATJ and Jet A1. (a) distribution for total particles from the TSI 3776 (b) distribution for total particles from the TSI 7610 (c) distribution for non-volatile particles from the TSI 3776.

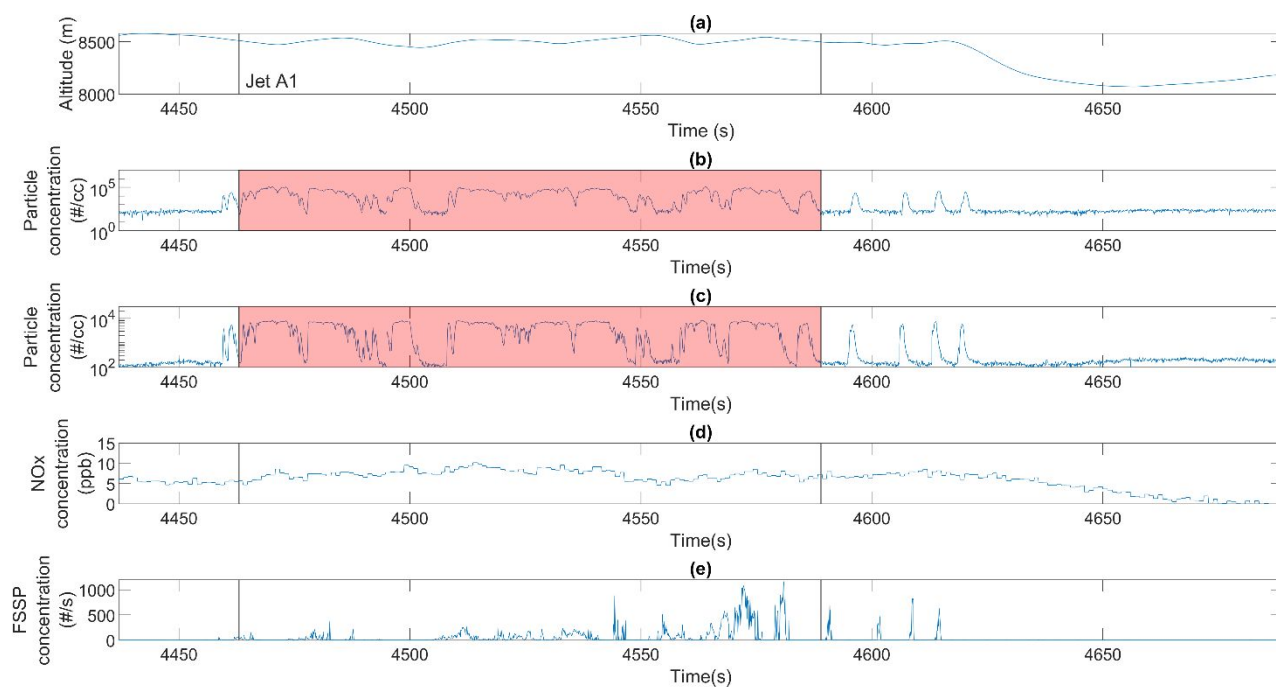
**Table S4.** Summary of the final particle number emission index ratios for the CAAFCEB campaign. The particle number emission index ratios were calculated using integrals and the confidence intervals were determined from the Monte Carlo simulation

Flight Day	Fuel Types	Particle Type	Effective Cut-Off Diameter (nm)	Particle Number Emission Index Ratio	Uncertainty Upper limit (95%)	Uncertainty Lower limit (95%)
Nov 20	JP-5/Jet A1	Total	7.7	0.96	1.80	0.71
Nov 20	JP-5/Jet A1	Total	15.4	0.96	1.53	0.83
Dec 21	ATJ/Jet A1	Total	7.7	0.033	0.052	0.021
Dec 21	ATJ/Jet A1	Total	15.4	0.34	0.46	0.12
Dec 21	ATJ/Jet A1	Non-Volatile	13.3	0.030	0.045	0.013

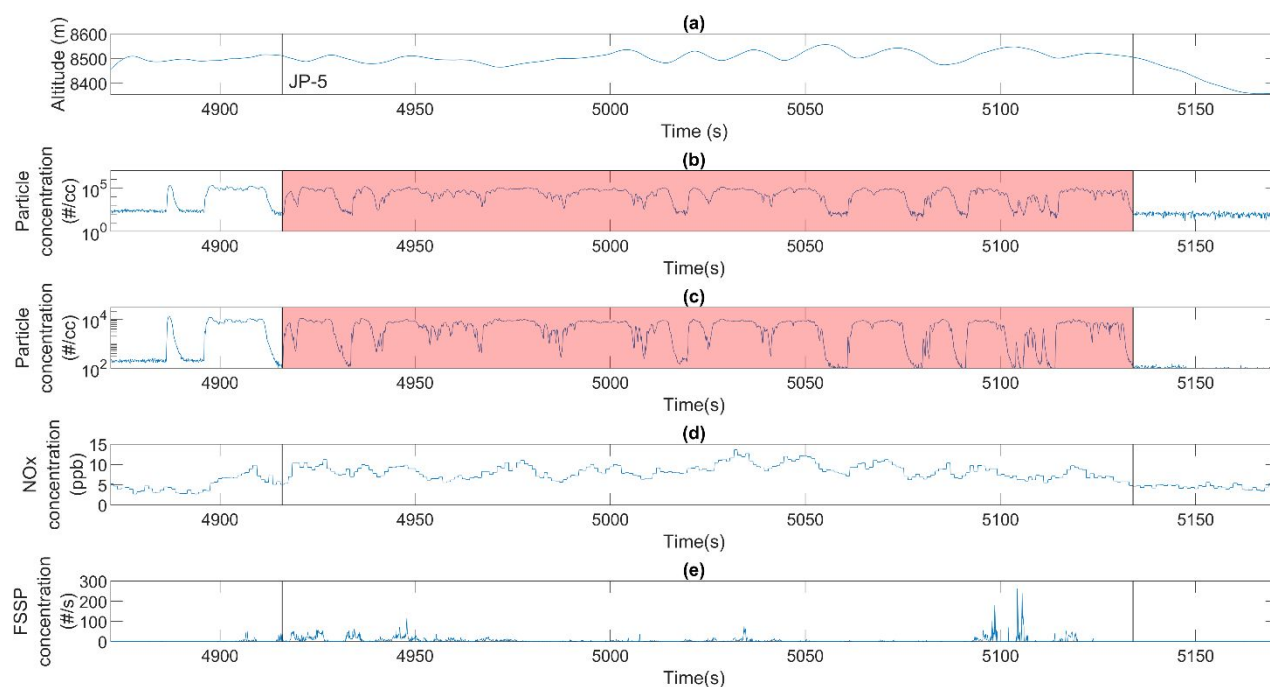
### **S3 Raw data sample**

Sample raw data plots for all flights are shown in Figure S11 to Figure S14. The highlighted sections represent the segments that were analyzed: red represent when the by-pass line was used to measure total particles and blue represents when the denuder was used to measure non-volatile particles. First data from the forward scattering spectrometer probe (Particle Measuring Systems Inc., FSSP-100) was used to identify when the CT-133 was in the contrails of the Falcon 20. Then the data for the particle and NO<sub>x</sub> concentration was examined to determine periods where the concentrations were above background which meant that the exhaust plume was being sampled. The status of the catalytic denuder was also recorded and used to determine when it was in-line or when the bypass line was used. The altitude data was used as a reference to identify when the CT-133 was at a steady altitude (thus at cruise) in the contrail of the Falcon 20.

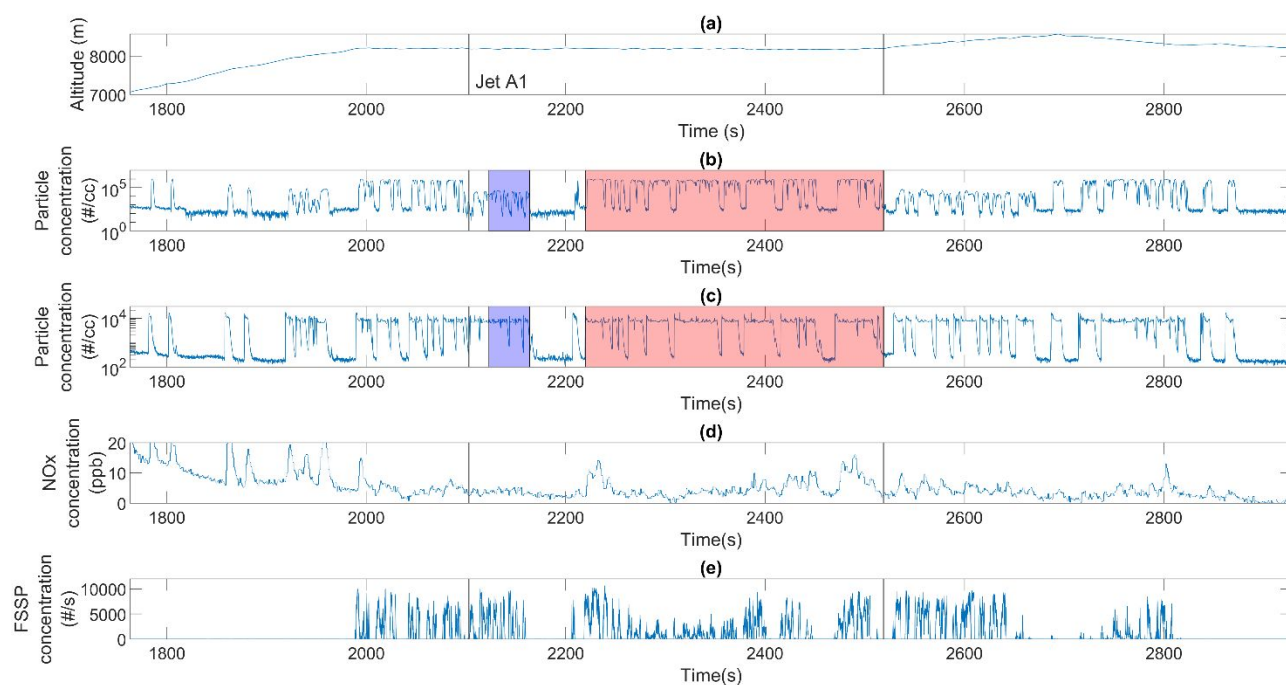
As seen in Figure S11 to Figure S14, the particle concentrations varied between high and low concentrations as the CT-133 flew in and out of the contrails. As shown in the figures, the NO<sub>x</sub> concentrations have broader peaks and do not appear to vary at the same rate as the particle concentrations. This is due to the two CPCs having much faster response times (a little less than 1 second and 2 seconds for the TSI 3776 and TSI 7610 CPC; respectively) than the NO<sub>x</sub> analyzer (tens of seconds for the Thermo 42i). Thus, in the analysis of the particle number emission index ratios, the integrals of the particle and NO<sub>x</sub> concentrations over each segment was used instead of individual data points similar to the method of Schulte P. et al. (1997).<sup>11</sup>



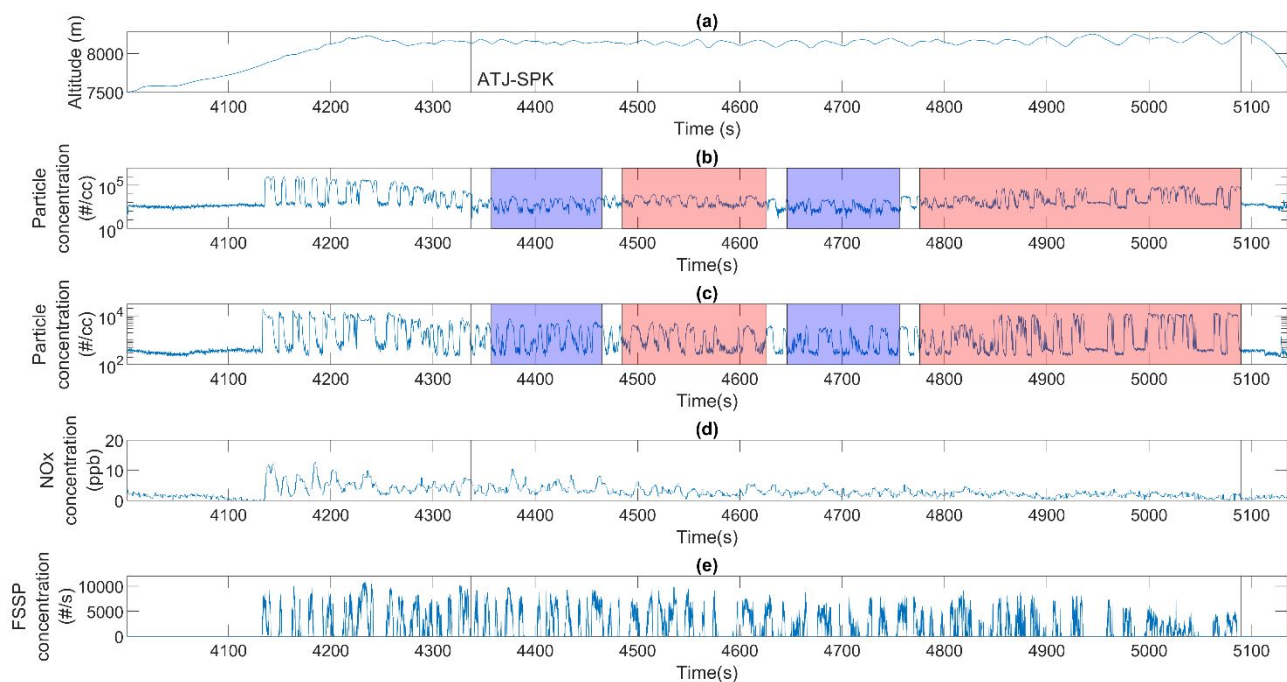
**Figure S11.** Raw data plot for the Jet A1 flight on Nov 20 with lines indicating the segments chosen for analysis: (a) altitude measurements from the CT-133 (b) 3776 CPC particle concentration, (c) 7610 CPC particle concentration, (d) NO<sub>x</sub> concentration, (d) ice particle concentration.



**Figure S12.** Raw data plot for the JP-5 flight on Nov 20 with lines indicating the segments chosen for analysis: (a) altitude measurements from the CT-133 (b) 3776 CPC particle concentration, (c) 7610 CPC particle concentration, (d)  $\text{NO}_x$  concentration, (d) ice particle concentration.



**Figure S13.** Raw data plot for the Jet A1 flight on Dec 21 with lines indicating the segments chosen for analysis: (a) altitude measurements from the CT-133 (b) 3776 CPC particle concentration, (c) 7610 CPC particle concentration, (d) NO<sub>x</sub> concentration, (d) ice particle concentration.



**Figure S14.** Raw data plot for the ATJ-SPK flight on Dec 21 with lines indicating the segments chosen for analysis: (a) altitude measurements from the CT-133 (b) 3776 CPC particle concentration, (c) 7610 CPC particle concentration, (d) NO<sub>x</sub> concentration, (d) ice particle concentration.



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