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A Methodology to Relate Black Carbon Particle Number and Mass Emissions from Various Combustion Sources

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2018 Cambridge Particle Meeting

15th June 2018

Outline

1. Introduction & Motivation
2. Derivation of a new BC N or EI_n Predictive Model
3. Model Validation
4. Uncertainty & Sensitivity Analysis
5. Conclusions & Further Work



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Why is BC PN Emissions Important?

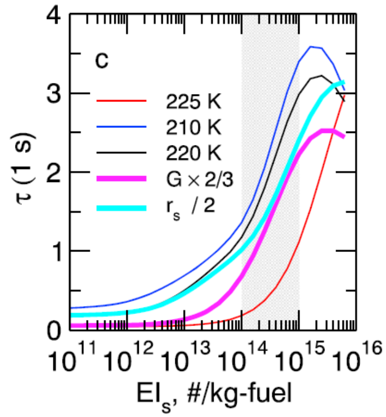
- Health Effects: Ultrafine particles have a higher probability of being deposited into the respiratory system, and translocated towards the circulatory system and internal organs.

- Climate Effects: During cruise, Black Carbon (BC) Particle Number (PN) emissions from aircraft engines act as a condensation nuclei for contrail formation.
  - No. of contrail ice particles = No. of aircraft BC particle emissions per kg-fuel burnt (EI_n in kg^{-1})
  - Different young contrail characteristics influenced by BC EI_n.

- Existing BC EI_n models for aviation emission assumes that BC particle morphologies remain constant irrespective of engine thrust settings.

- BC mass measurements and estimates remain more commonly available than the number metric.

τ = Optical depth



Research Objectives

1) Develop a new model to estimate BC particle number emissions from mass based on the theory of fractal aggregates.

2) Validate the new model using BC measurements from three different emission sources.

3) Perform an uncertainty and sensitivity analysis to understand the accuracy and uncertainty bounds of the newly developed model.

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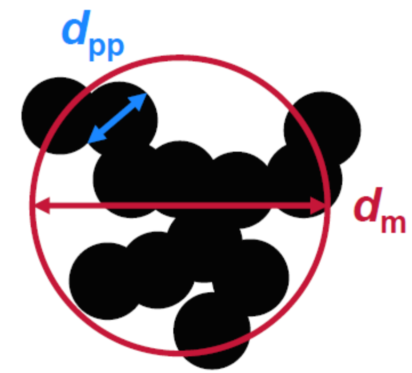
Development of a New BC PN Predictive Model



- Mass of one BC aggregate (m) is the summation of primary particle masses:

$$m = n_{\mathrm{pp}} \rho_0 \left(\frac{\pi}{6}\right) {d_{\mathrm{pp}}}^3$$

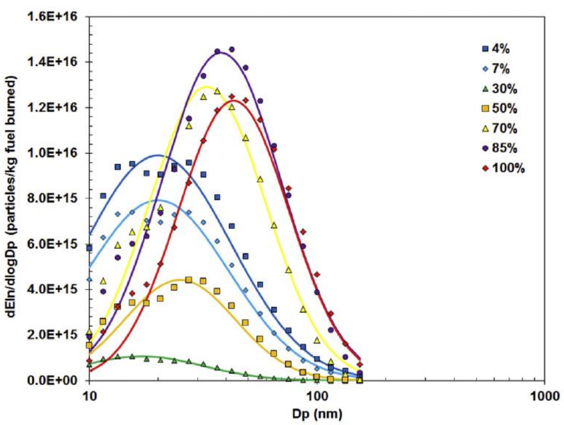
where

n_{pp}	=	Number of primary particles in an aggregate
ρ_0	=	BC material density (1770 kg/m^3)
d_{pp}	=	Primary particle diameter

- The total mass of aggregates (M) is calculated using the integrated product of the aggregate mass and number weighted distribution:

$$M = \int_0^{\infty} m(d_{\mathrm{m}}) n(d_{\mathrm{m}}) \, \mathrm{d}ln d_{\mathrm{m}}$$

where $n(d_{\mathrm{m}}) = N \times p(d_m)$



Source: [11]

$$m = n_{pp}\rho_0\left(\frac{\pi}{6}\right)d_{pp}^{\ 3}$$

Development of a New BC PN Predictive Model

- In the free-molecular regime, the number of primary particles in an aggregate (n_{pp}) [6]:

$$n_{pp} = k_a\left(\frac{d_m}{d_{pp}}\right)^{2D_\alpha} \quad \text{or} \quad n_{pp} = \left(\frac{d_m}{d_{pp}}\right)^{D_{fm}}$$

where d_m = Aggregate mobility diameter

k_a = Scaling pre-factor

D_α = Projected area exponent

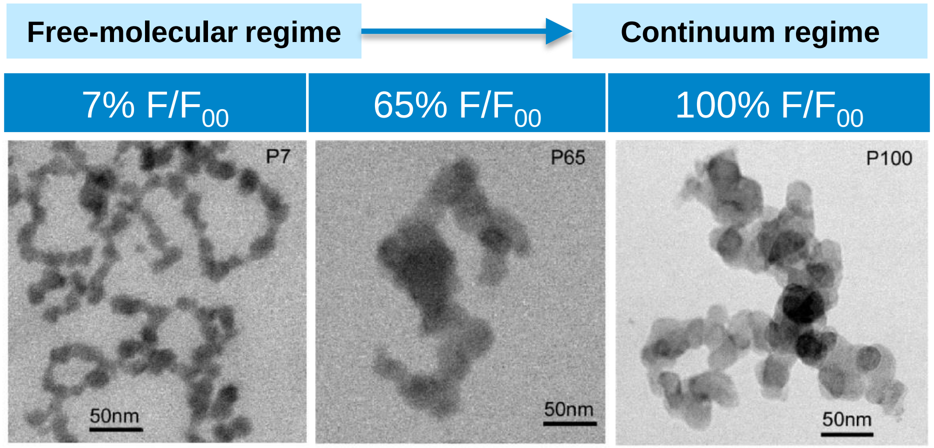
D_{fm} = Mass-mobility exponent

  - Eggersdorfer et al. (2012) [7] suggested universal values of $k_a = 0.998$ and $D_\alpha = 1.069$ for aggregates formed of polydisperse primary particles, irrespective of the state of sintering.

- The Knudsen Number (Kn) is a dimensionless number equal to the ratio of the mean free path (λ) to the particle radius:

$$Kn = \frac{2\lambda}{d}$$

  - Free-molecular regime: $\mathrm{Kn} > 1$
  - Continuum regime: $\mathrm{Kn} \leq 1$
  - Transition regime: $0.1 < \mathrm{Kn} < 10$



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$$m = n_{\mathrm{pp}} \rho_0 \left(\frac{\pi}{6}\right) d_{\mathrm{pp}}{}^{3}$$

Development of a New BC PN Predictive Model

- Relationship between primary particle (d_{pp}) and aggregate mobility diameter (d_m) [9]:

$$d_{\mathrm{pp}} = k_{\mathrm{TEM}} \times d_{\mathrm{m}}{}^{D_{\mathrm{TEM}}}$$

where prefactor-exponent coefficient pairs k_{TEM} & D_{TEM} are fitted with Transmission Electron Microscopy (TEM)

GDI engine:

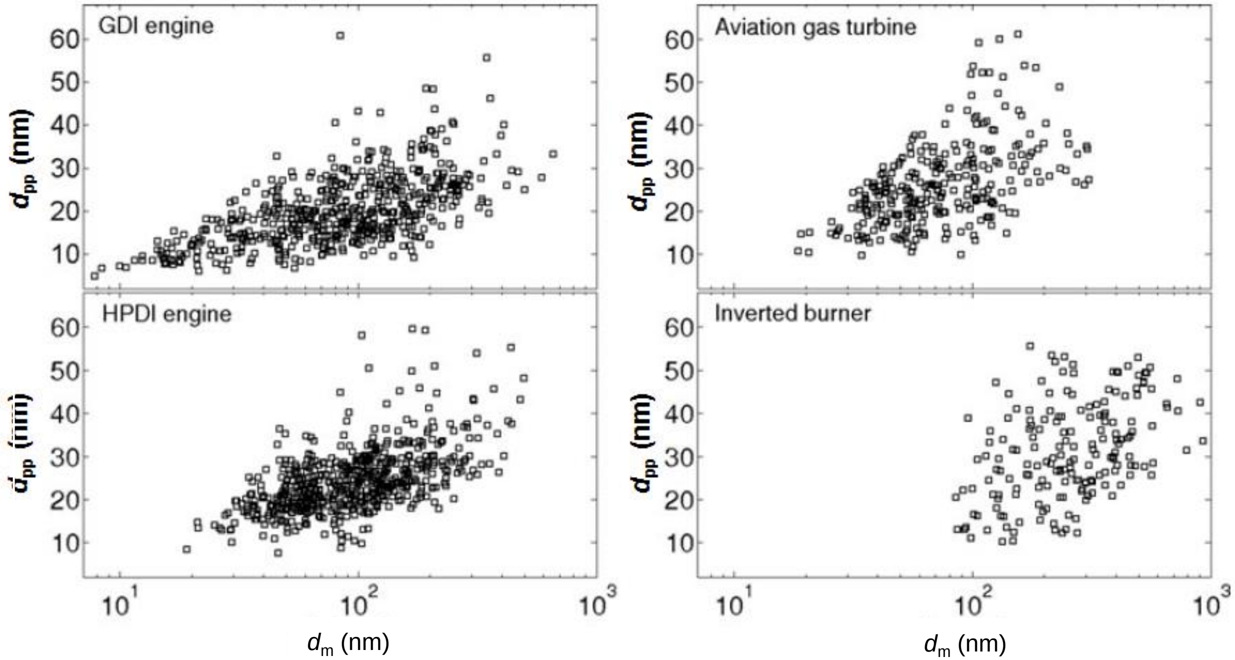
$k_{\mathrm{TEM}} = 2.616 \times 10^{-6}$

$D_{\mathrm{TEM}} = 0.30$

HPDI engine:

$k_{\mathrm{TEM}} = 2.644 \times 10^{-6}$

$D_{\mathrm{TEM}} = 0.29$



Aviation gas turbine:

$k_{\mathrm{TEM}} = 1.621 \times 10^{-5}$

$D_{\mathrm{TEM}} = 0.39$

Inverted burner:

$k_{\mathrm{TEM}} = 2.465 \times 10^{-6}$

$D_{\mathrm{TEM}} = 0.29$

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Development of a New BC PN Predictive Model

➢ The total mass of aggregates (*M*) for a given particle size distribution:

$$n_{\mathrm{pp}} = k_{\mathrm{a}}\left(\frac{d_{\mathrm{m}}}{d_{\mathrm{pp}}}\right)^{2D_{\alpha}}$$

$$d_{\mathrm{pp}} = k_{\mathrm{TEM}} \times d_{\mathrm{m}}{}^{D_{\mathrm{TEM}}}$$

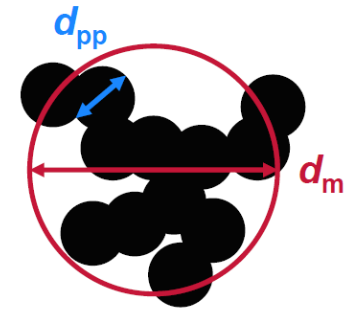
$$m = n_{\mathrm{pp}}\rho_0\left(\frac{\pi}{6}\right)d_{\mathrm{pp}}{}^{3}$$

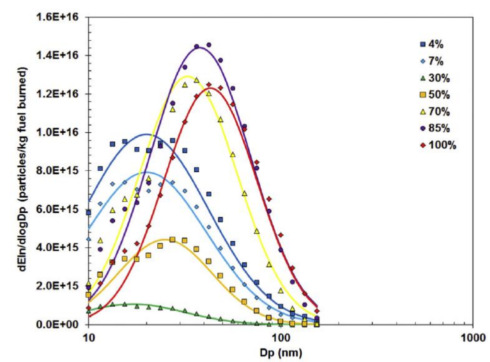
$$n(d_{\mathrm{m}}) = N \times p(d_{m})$$

$$M = \int_0^{\infty} m(d_{\mathrm{m}})n(d_{\mathrm{m}})\,\mathrm{d}\ln d_{\mathrm{m}}$$

$$M = Nk_{\mathrm{a}}\rho_0\left(\frac{\pi}{6}\right)(k_{\mathrm{TEM}})^{3-2D_{\alpha}}\int_0^{\infty} d_{\mathrm{m}}{}^{\varphi}\,\mathrm{p}(d_{\mathrm{m}})\,\mathrm{d}\ln d_{\mathrm{m}}$$

where $\varphi = 3D_{\mathrm{TEM}} + (1 - D_{\mathrm{TEM}})2D_{\alpha}$





Source: [11]

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Development of a new BC PN Predictive Model

- The remaining integral is the φth moment of a log-normal distribution:

$$M = N k_{\mathrm{a}} \rho_0 (\frac{\pi}{6}) (k_{\mathrm{TEM}})^{3-2D_\alpha} \mathrm{GMD}^{\varphi} \exp(\frac{\varphi^2 \ln(\mathrm{GSD})^2}{2})$$

where	GMD	=	Geometric Mean Diameter
	GSD	=	Geometric Standard Deviation

- Rearrange for *N*:

$$N = \frac{M}{k_{\mathrm{a}} \rho_0 (\frac{\pi}{6}) (k_{\mathrm{TEM}})^{3-2D_\alpha} \mathrm{GMD}^{\varphi} \exp(\frac{\varphi^2 \ln(\mathrm{GSD})^2}{2})}$$

New *N* or EI_n Predictive Model, called the Fractal Aggregates (FA) Model.

- Advantages:
  - ✓ New FA model relates BC mass, number and Particle Size Distribution (PSD) in one equation.
  - ✓ Captures the change in particle morphology for different combustion conditions

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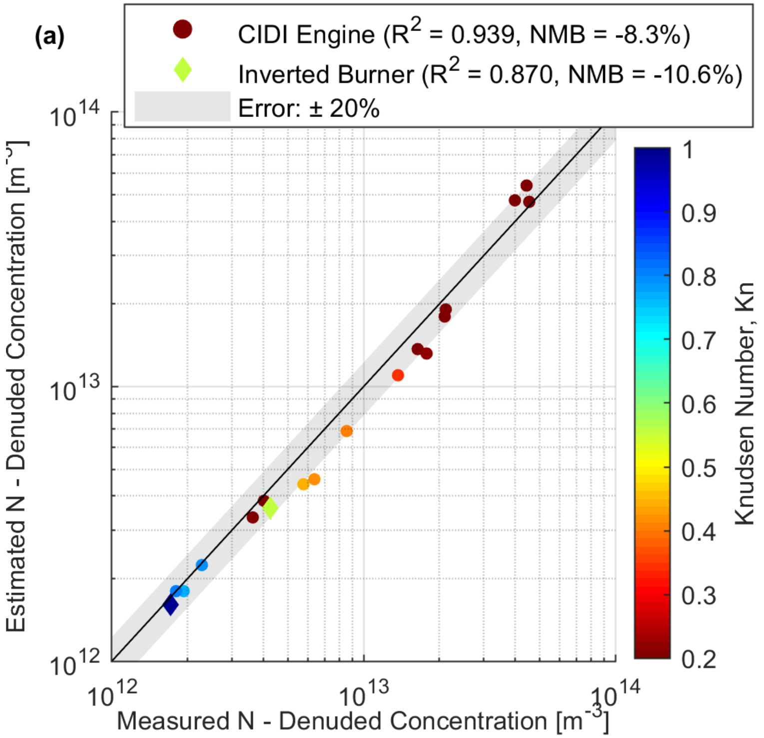
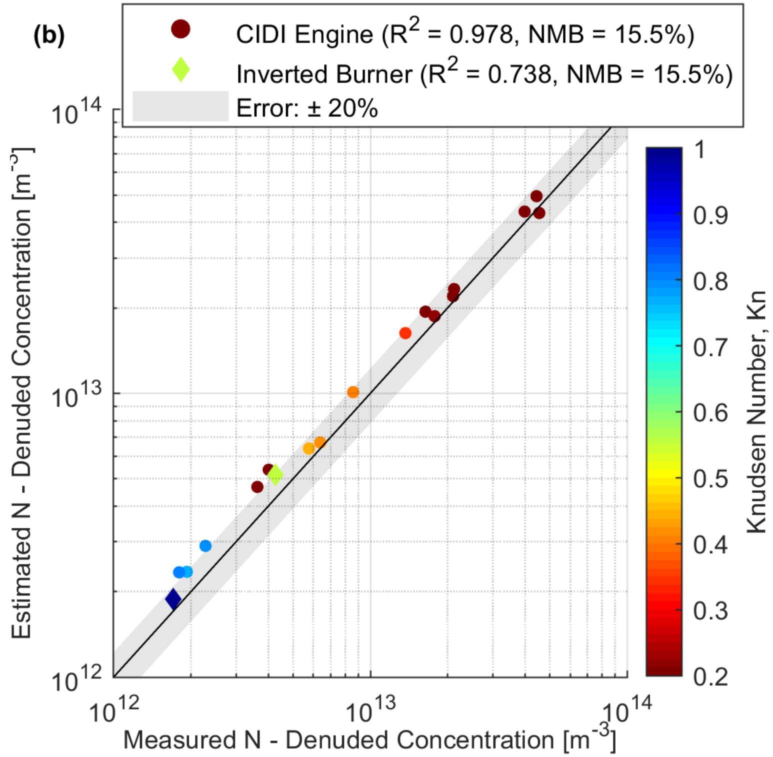
Model Validation – CIDI Engine & Inverted Burner

$$N = \frac{M}{k_a \rho_0 (\frac{\pi}{6})(k_{TEM})^{3-2D_\alpha} \mathrm{GMD}^{\varphi} \exp(\frac{\varphi^2 \ln(\mathrm{GSD})^2}{2})}$$ where $\varphi = 3D_{TEM} + (1 - D_{TEM})2D_\alpha$

(a) Experimentally fitted k_a and D_α values for each operating mode [14]



(b) Constant $k_a = 0.998$ and $D_\alpha = 1.069$ for all operating mode [7]



CIDI Engine Data Source: [12]
Inverted Burner Data Source: [13]

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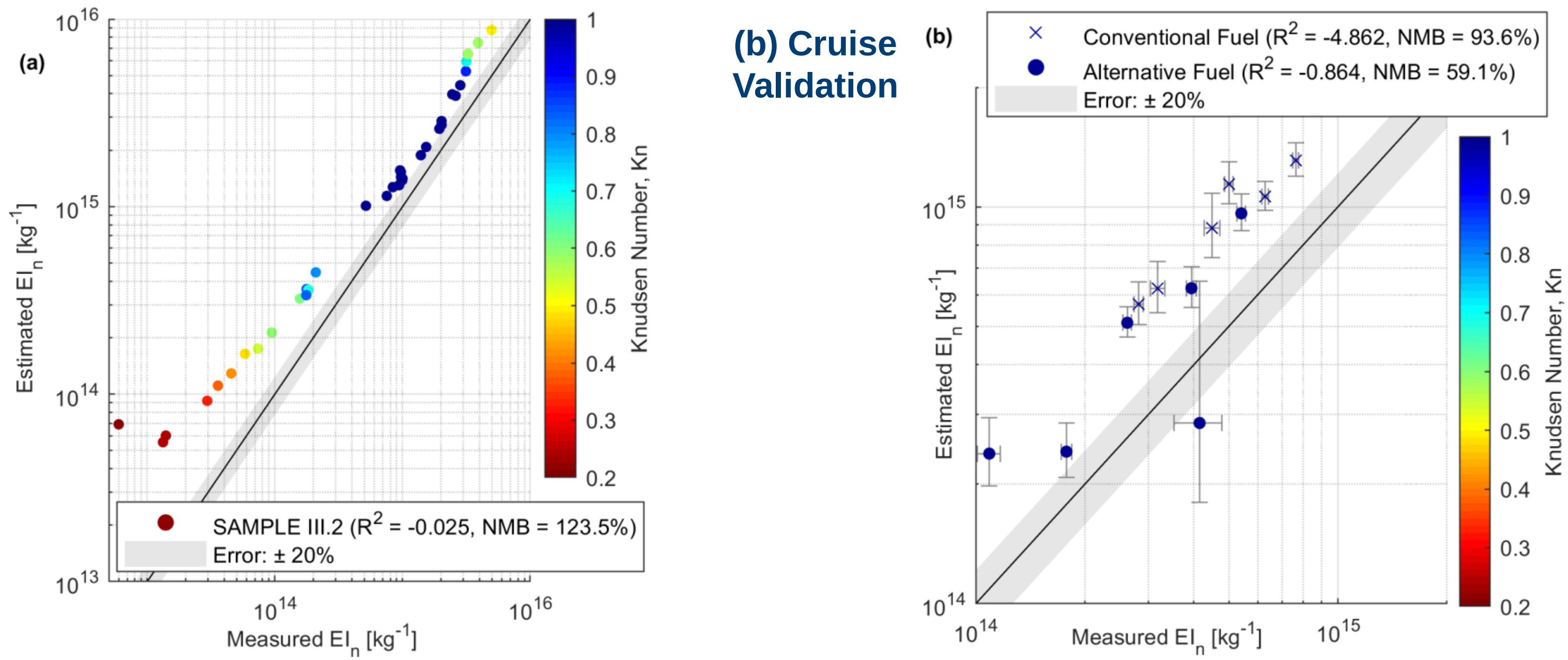
Model Validation – Aircraft Gas Turbine Engines

Constant $k_a = 0.998$ and $D_\alpha = 1.069$ for all operating mode [7]

$$\mathrm{EI_n} = \frac{\mathrm{EI_m}}{k_a \rho_0 (\frac{\pi}{6})(k_{\mathrm{TEM}})^{3-2D_\alpha} \mathrm{GMD}^{\varphi} \exp(\frac{\varphi^2 \ln(\mathrm{GSD})^2}{2})} \quad \text{where} \quad \varphi = 3D_{TEM} + (1 - D_{TEM})2D_\alpha$$

(a) Ground Validation **(b) Cruise Validation**



- When k_a and D_α values of 0.998 and 1.069 [7] are applied to aircraft datasets, on average, we obtain negative R^2 and > 100% NMB values

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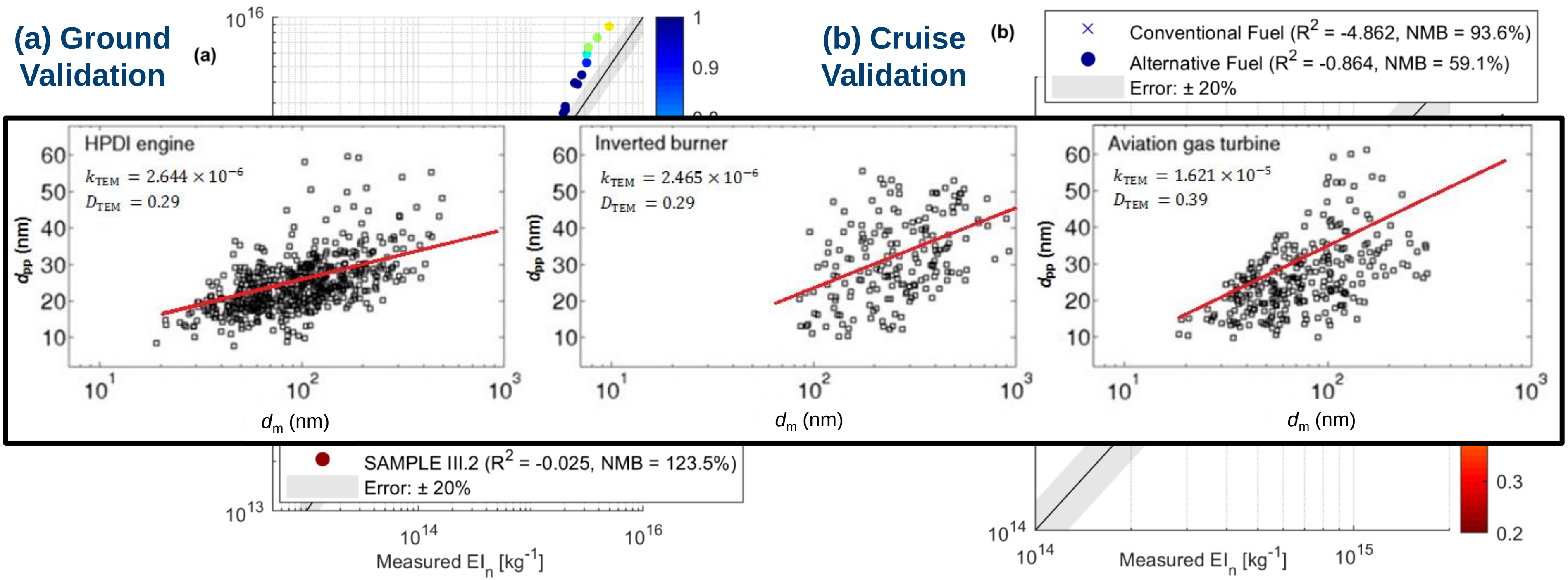
Model Validation – Aircraft Gas Turbine Engines

Constant $k_a = 0.998$ and $D_\alpha = 1.069$ for all operating mode [7]

$$\mathrm{EI_n} = \frac{\mathrm{EI_m}}{k_a \rho_0 (\frac{\pi}{6})(k_{\mathrm{TEM}})^{3-2D_\alpha} \mathrm{GMD}^{\varphi} \exp(\frac{\varphi^2 \ln(\mathrm{GSD})^2}{2})} \quad \text{where} \quad \varphi = 3D_{TEM} + (1 - D_{TEM})2D_\alpha$$

(a) Ground Validation

(b) Cruise Validation



- When k_a and D_α values of 0.998 and 1.069 are applied to aircraft datasets, on average, we obtain negative R^2 and > 100% NMB values

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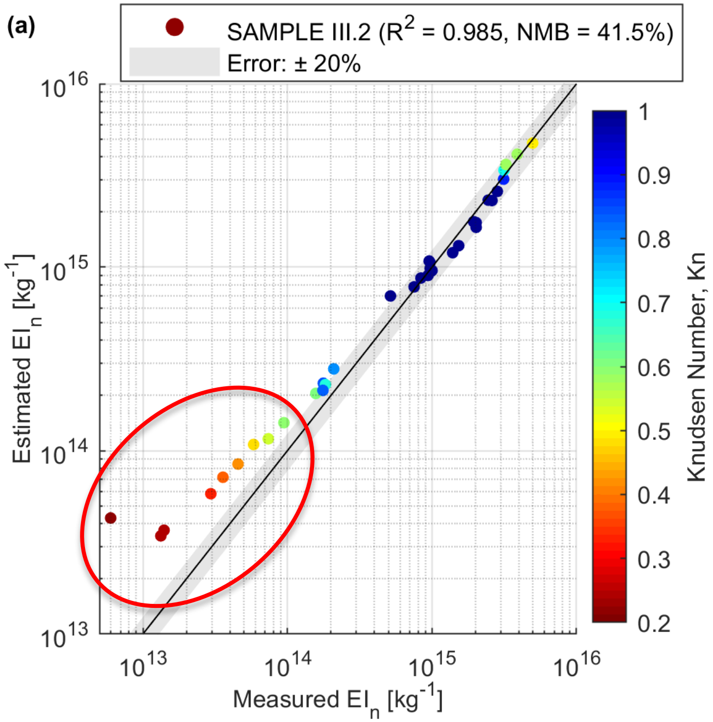
Model Validation – Aircraft Gas Turbine Engines

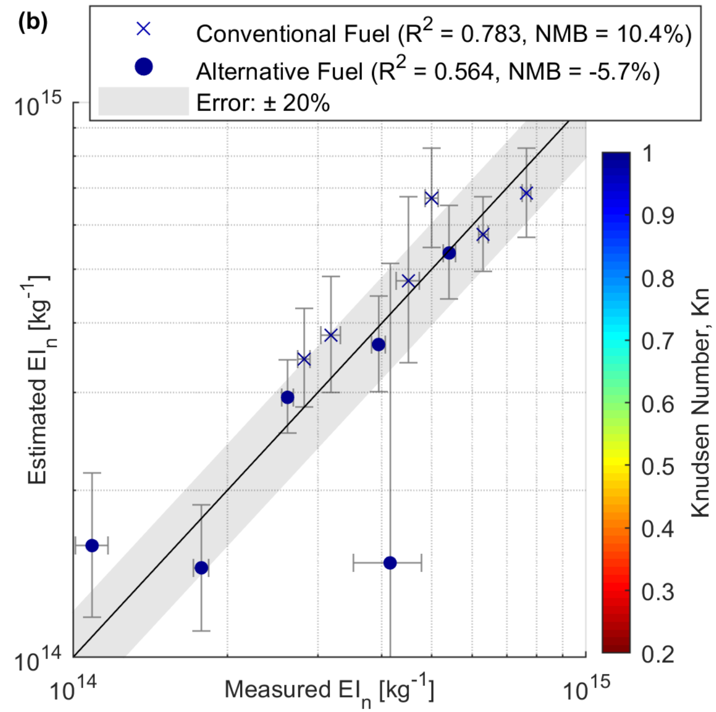
$$\mathrm{EI_n} = \frac{\mathrm{EI_m}}{1 \times \rho_0 (\frac{\pi}{6})(k_{\mathrm{TEM}})^{3-D_{fm}} \mathrm{GMD}^{\varphi} \exp(\frac{\varphi^2 \ln(\mathrm{GSD})^2}{2})}$$ where $\varphi = 3D_{TEM} + (1 - D_{TEM})D_{\mathrm{fm}}$

- **Recall**: $n_{pp} = k_a (\frac{d_m}{d_{pp}})^{2D_\alpha}$ or $n_{pp} = (\frac{d_m}{d_{pp}})^{D_{fm}}$. Hence, we assume that $k_a = 1$ and $D_\alpha = \frac{1}{2} D_{\mathrm{fm}}$ [7]

(a) Ground Validation



(b) Cruise Validation



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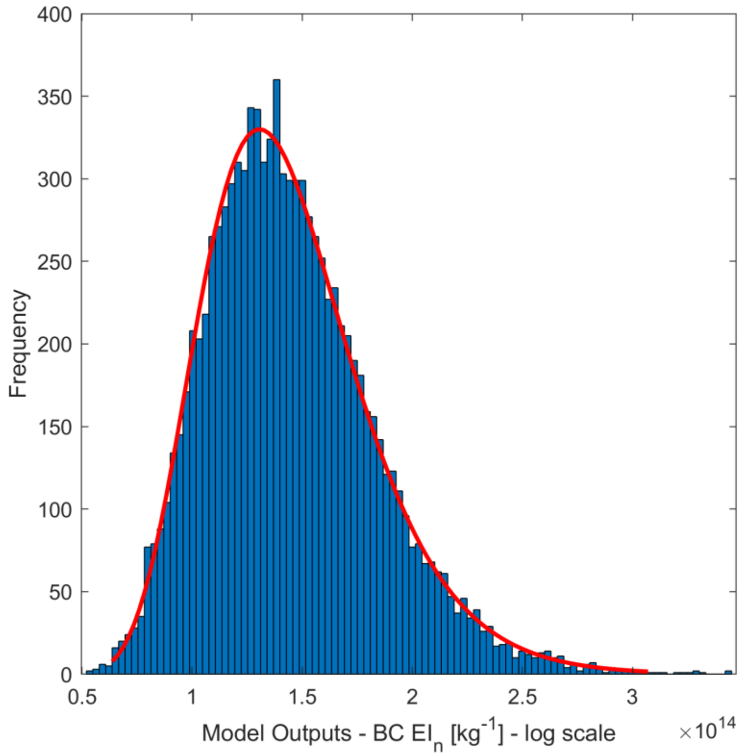
Results: Uncertainty Analysis

- Uncertainty analysis is performed using the Monte Carlo 1000-member ensembles [16].

- The uncertainty of the estimated EI_n are asymmetrically distributed (-37%, +55%) at 1.96σ.

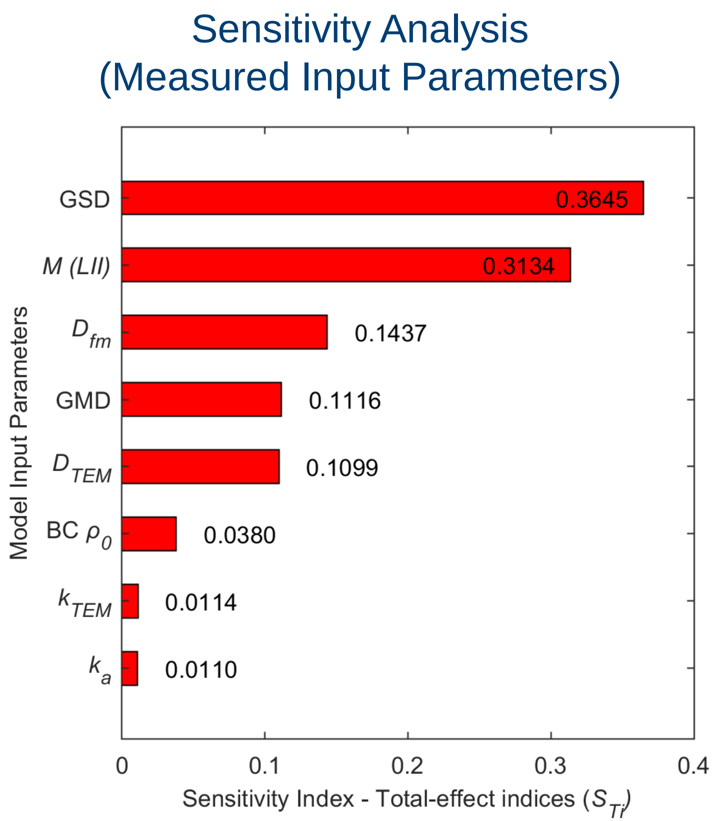
- The asymmetrical distribution is due to the non-linearity of the FA model.

- An uncertainty analysis on the estimated EI_n was not conducted in previous aviation PN methodologies.



Results: Sensitivity Analysis

- A variance-based global sensitivity analysis identified that the uncertainties in GSD contribute to the largest sensitivity in the FA model output.
- A prioritisation can be recommended for future research to measure certain variables (such as M, D_{fm}, GMD and GSD) more accurately to reduce the uncertainty bounds of the FA model outputs.
- Given that k_a contributes to the lowest sensitivity to the estimated EI_n, the assumption of $k_a = 1$ across all engine types & F/F_{00} would not significantly affect the FA model outputs.



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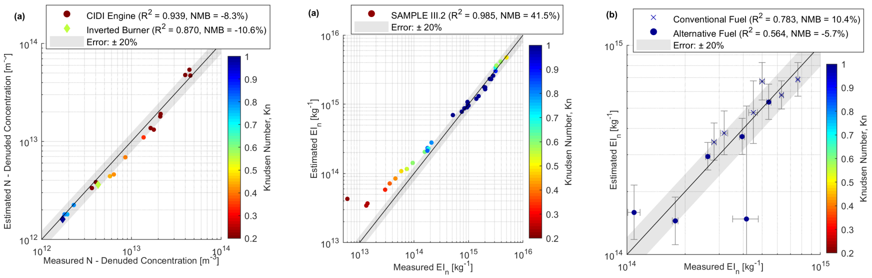
Conclusions

$$N = \frac{M}{k_a \rho_0 (\frac{\pi}{6})(k_{\mathrm{TEM}})^{3-2D_\alpha} \mathrm{GMD}^{\varphi} \exp(\frac{\varphi^2 \ln(\mathrm{GSD})^2}{2})}$$

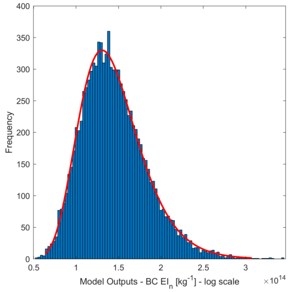
- A new methodology to relate BC Particle Number and Mass emissions is developed based on the theory of fractal aggregates.

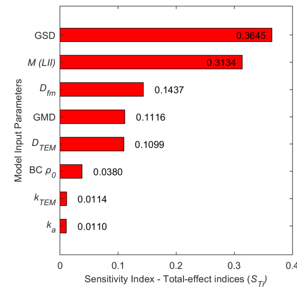
- The new FA Model is validated with three different emission sources:
  - An internal combustion engine (CIDI)
  - An inverted burner
  - Two aircraft gas turbine engines



- An uncertainty analysis estimates *N* or $\mathrm{EI_n}$ to have an asymmetrical uncertainty bound (-36%, +54%) at 1.96σ.

- A sensitivity analysis shows that GSD is the most critical input parameter, followed by the *M*, D_{fm} and GMD.





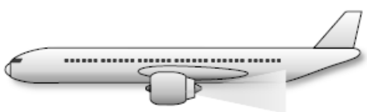
Future Work

FA Model Application to Aviation Emissions:

Is there a net climate benefit in diverting flights to avoid contrail formation?

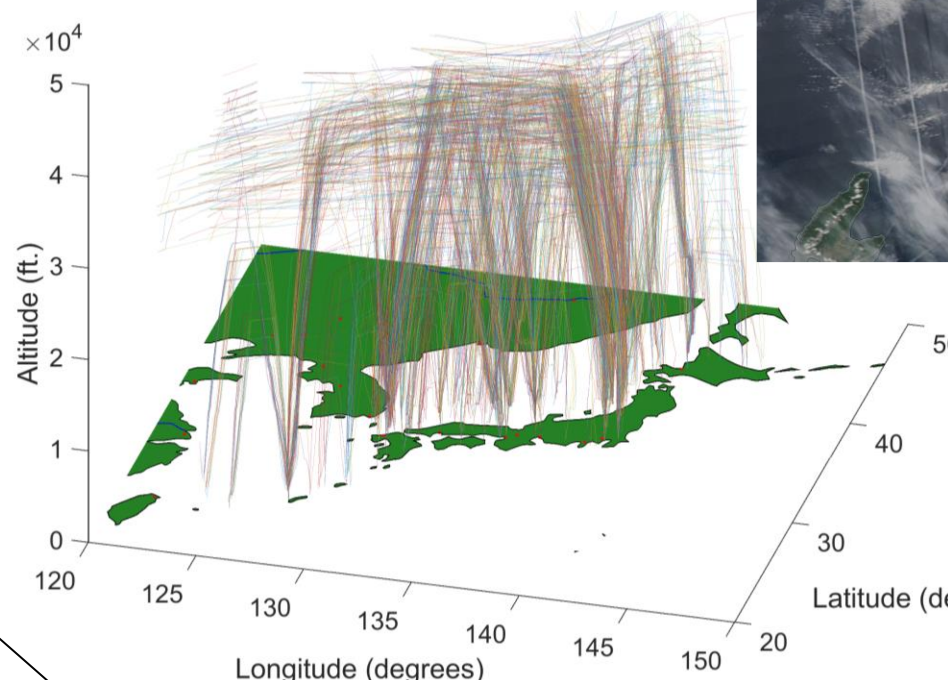
Aircraft Activity Dataset:

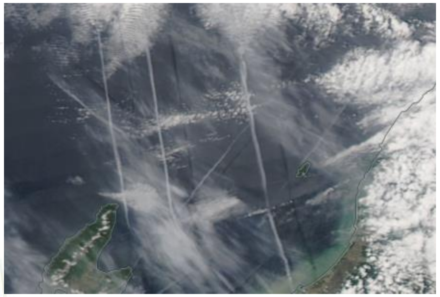
CARATS Open Data, Japan



Emissions:

CO_2, BC EI_n (FA Model) & EI_m,…





Ambient atmospheric conditions:

ECMWF Dataset

Contrail Model:

CoCiP

Climate Impacts

Air Traffic Management

Thank you, questions?

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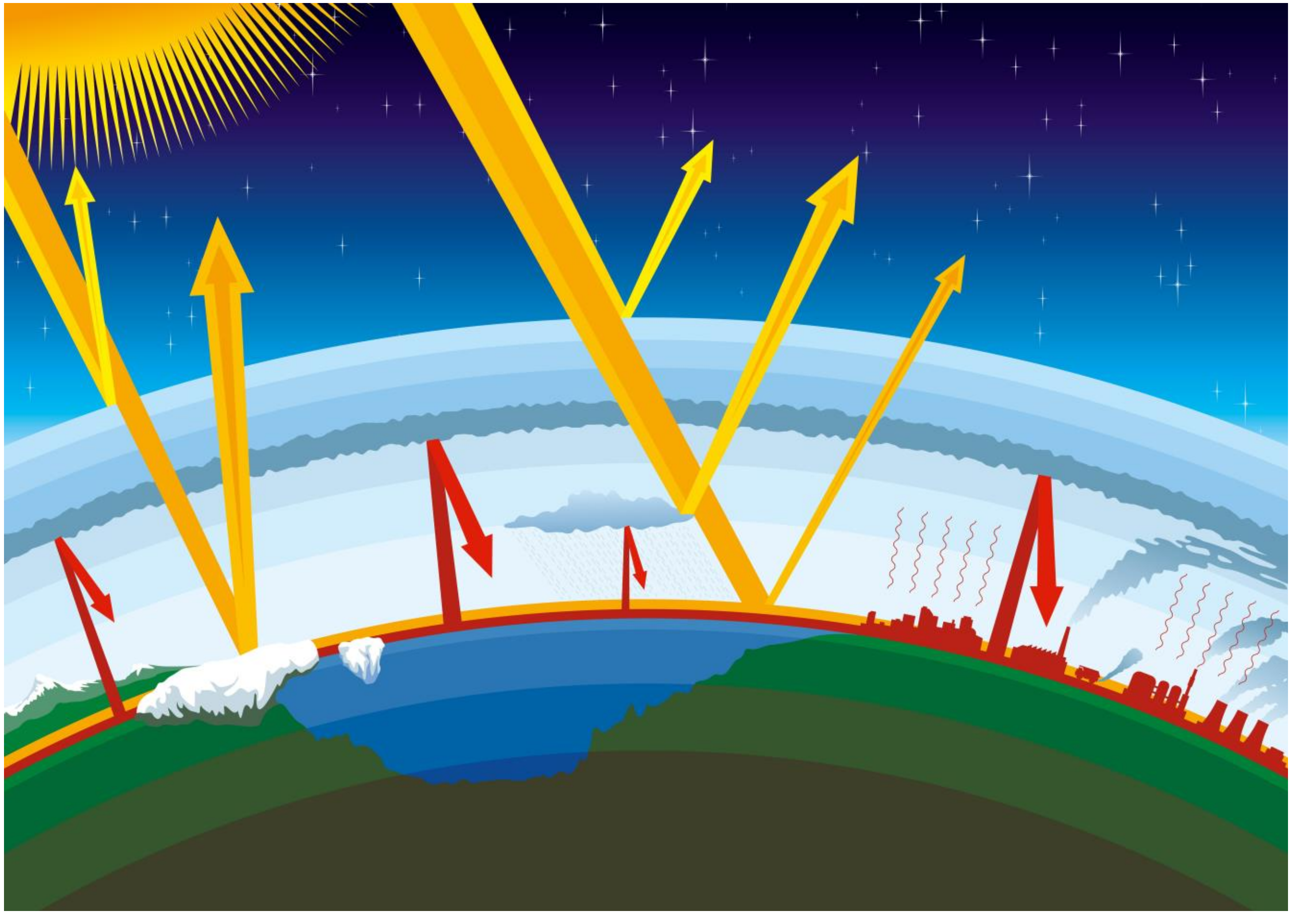
Global Warming



Source: http://globalwarming-facts.info/wp-content/uploads/shutterstock_91110830.jpg

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Existing BC EI_n Models for Aviation Emissions

1) **Average BC Particle Mass** [2]

- Total BC mass divided by an average mass of each BC particle
- $\text{BC EI}_n = \frac{\text{BC EI}_m}{(3.2\times10^{-17})}$

2) **EI_n/EI_m Ratio with Altitude Variations** [3]

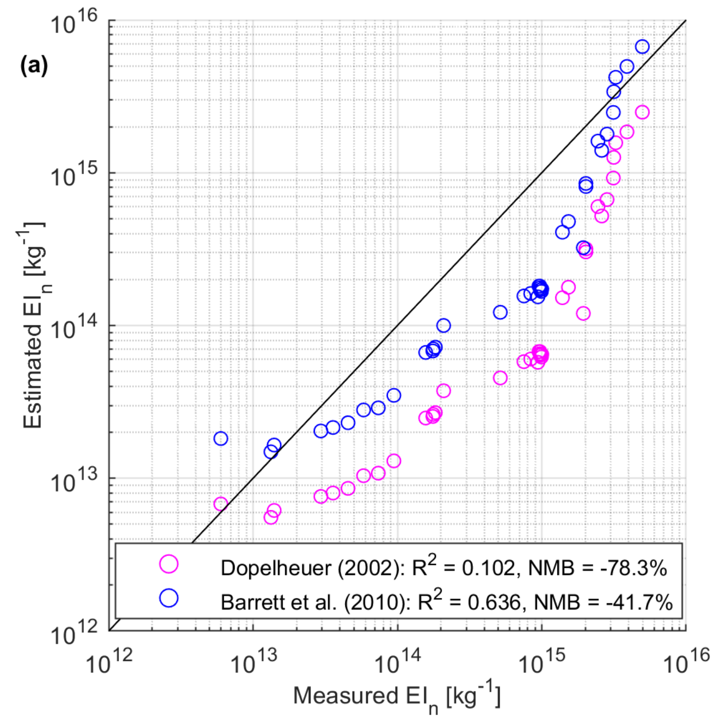
- ≈ $5\text{x}10^{15}$ to $1.6\text{x}10^{16}$ particles per g(BC)
- Used in the Aero2K Global Aviation Emissions Inventories

3) **Assumed Particle Diameter** [4]

- $\text{EI}_n = \frac{\text{EI}_m}{(\frac{\pi}{6})\rho_{NV}\times\text{GMD}^3\times\exp(\frac{9}{2}(\ln\text{GSD})^2)}$
- Assume log-normal distribution, GMD and GSD fixed at 38nm and 1.6 respectively.

4) **BC EI_n Range** [1]

- BC EI_n ≈10^{14} to 10^{15} per kg fuel burned



KEY LIMITATION:
Existing BC EI_n models do not include a dependence on the change in BC aggregate morphologies vs engine thrust settings

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Knudsen Number, Kn

Knudsen Number, $Kn = \frac{2\lambda}{d}$

- Mean free path, λ = Average distance travelled by an aggregate between successive collisions with gas molecules.

- The mean free path of at a given pressure (P_1) can be estimated using standard atmospheric conditions (P_0, λ_0) as a reference:

  $\lambda_1 = \lambda_0 \frac{P_0}{P_1}$, where P_0 = 1 atm & $\lambda_0 = 0.066\ \mu m$

- For the emissions from an internal combustion engine and aircraft gas turbine, the pressure in the combustor is used for P_1

- For inverted burner emissions, we assume that P_1 = 1 atm

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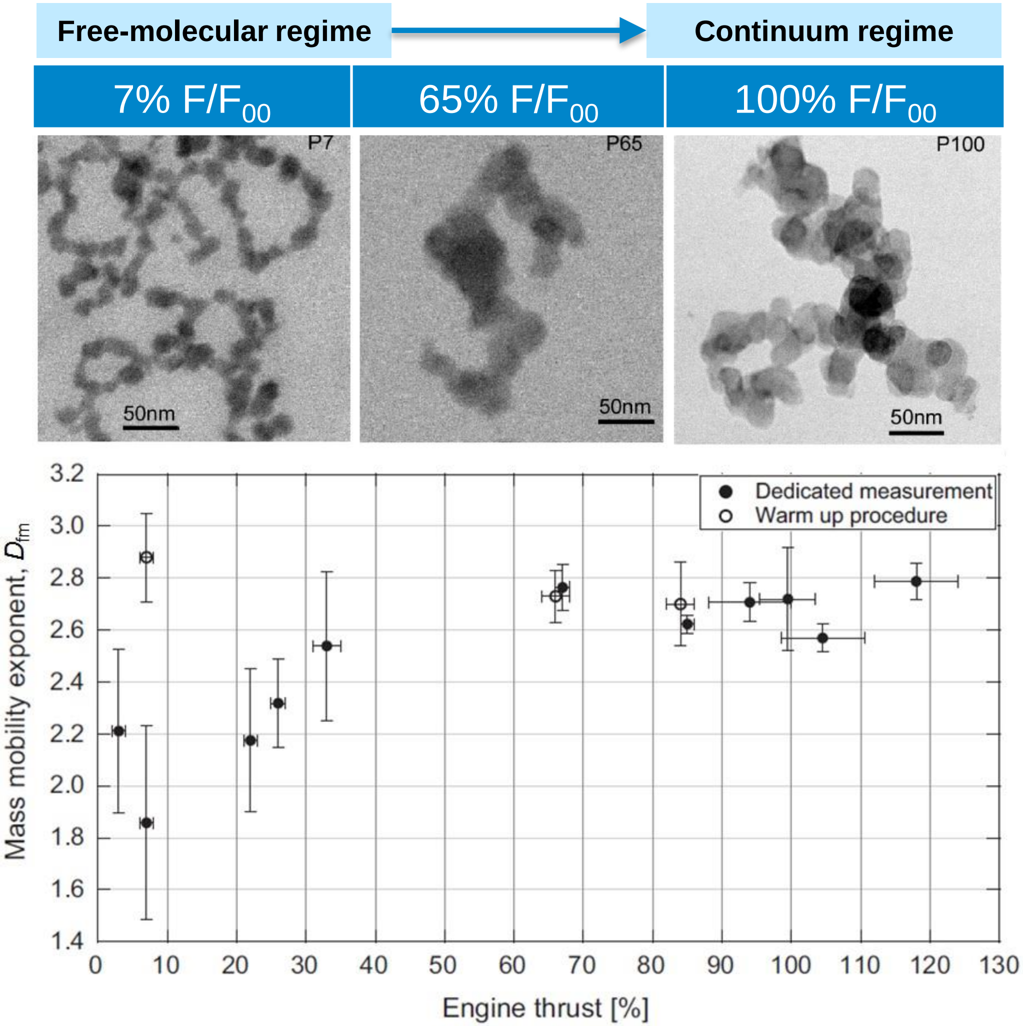
Knudsen Number, Kn

- Free-molecular regime: $\mathrm{Kn} > 1$
- Continuum regime: $\mathrm{Kn} \leq 1$
- Transition regime: $0.1 < \mathrm{Kn} < 10$



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How is k_a and D_α Experimentally Fitted?

Dastanpour et al. (2016) [14]

METHOD 1: Estimation of k_a and D_α using Optimisation

- Combining equations $n_{pp} = k_a(\frac{d_m}{d_{pp}})^{2D_\alpha}$ and $m = n_{pp}\left(\frac{\pi}{6}\right) d_{pp}{}^3 \rho_0$ to obtain:

$$d_{pp} = \left[\frac{k_a \pi \rho_0}{6m}(d_m)^{2D_\alpha}\right]^{\frac{1}{2D_\alpha - 3}}$$

- Optimum k_a and D_α values are calculated using regression to minimise the difference between the TEM determined d_{pp} and the above equation.

METHOD 2: Estimation of k_a and D_α using Experimental Measurements

- Combining equations $n_{pp} = k_a(\frac{d_m}{d_{pp}})^{2D_\alpha}$ and $n_{pp} = \frac{m}{m_{pp}}$ to obtain:

$$\text{In}\left(\frac{m}{m_{pp}}\right) = 2D_\alpha \ln\left(\frac{d_m}{d_{pp}}\right) + \ln(k_a)$$

- Mass-mobility data (obtained from CPMA and DMA), and TEM-obtained d_{pp} and m_{pp} were used in the above equation to obtain k_a and D_α values.

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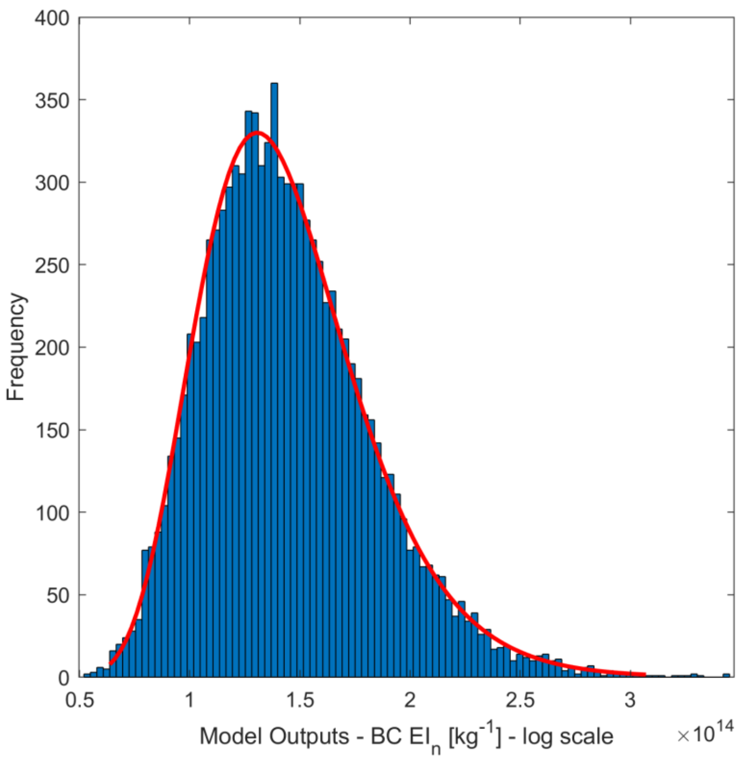
Uncertainty & Sensitivity Analysis

$$EI_n = \frac{EI_m}{k_a \rho_0 (\frac{\pi}{6})(k_{TEM})^{3-2D_\alpha} \mathrm{GMD}^{\varphi} \exp(\frac{\varphi^2 \ln(\mathrm{GSD})^2}{2})}$$

Variable	Fixed F/F$_{00}$	Mean (μ)	Std Dev (σ)
BC EI$_m$ (LII)	0.4	2.7 mg/kg	(25%/1.96)*μ
ρ_0	0.4	1770 kg/m^3	70
k_a	0.4	1	(2.4%/1.96)*μ
D_{fm}	0.4	2.76	(7.9%/1.96)*μ
k_{TEM}	0.4	1.621x10^{-5}	(7.2%/1.96)*μ
D_{TEM}	0.4	0.39	(7.9%/1.96)*μ
GMD	0.4	18.49 nm	(6.5%/1.96)*μ
GSD	0.4	1.73	(7.6%/1.96)*μ

- Uncertainty Distribution for all Parameters: Normal



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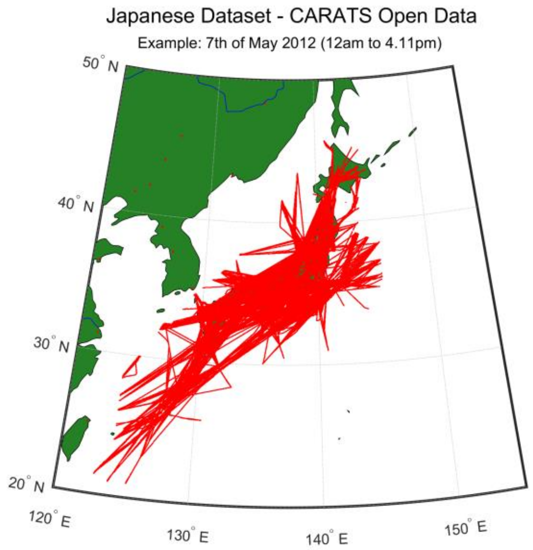
Model Application to Aviation Emissions

Can we apply the new FA model to estimate BC EI_n for global civil aviation, or at an individual flight level?

$$\mathrm{EI_n} = \frac{\mathrm{EI_m}}{\rho_0\left(\frac{\pi}{6}\right)(k_{\mathrm{TEM}})^{3-D_{\mathrm{fm}}}\mathrm{GMD}^{\varphi}\exp\left(\frac{\varphi^2\ln(\mathrm{GSD})^2}{2}\right)}$$

where $\varphi = 3D_{\mathrm{TEM}} + (1 - D_{\mathrm{TEM}})D_{\mathrm{fm}}$



Requirements:

- Estimate different input variables (BC EI_m, GMD, GSD and D_{fm}) versus engine thrust settings (F/F_{00}).

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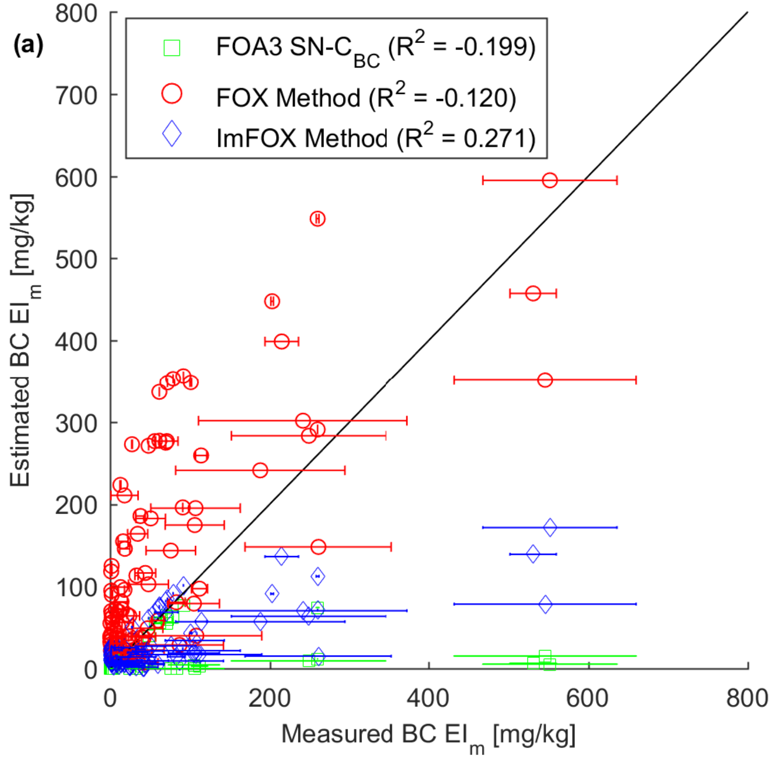
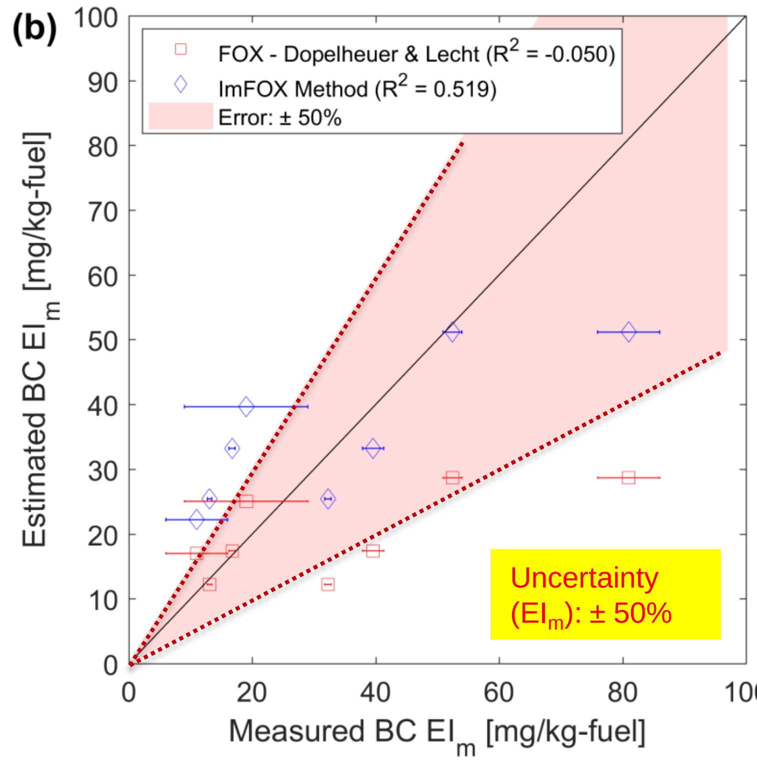
Model Inputs – (1) Existing BC EI_m Models

$$\mathrm{EI_n} = \frac{\mathbf{EI_m}}{\rho_0(\frac{\pi}{6})(1.621\times10^{-5})^{3-D_{\mathrm{fm}}}\mathrm{GMD}^{\varphi}\exp(\frac{\varphi^2\ln(\mathrm{GSD})^2}{2})}$$

(a) Ground Conditions



(b) Cruise Conditions



Source: FOA3 [16] FOX [17] ImFOX [18]

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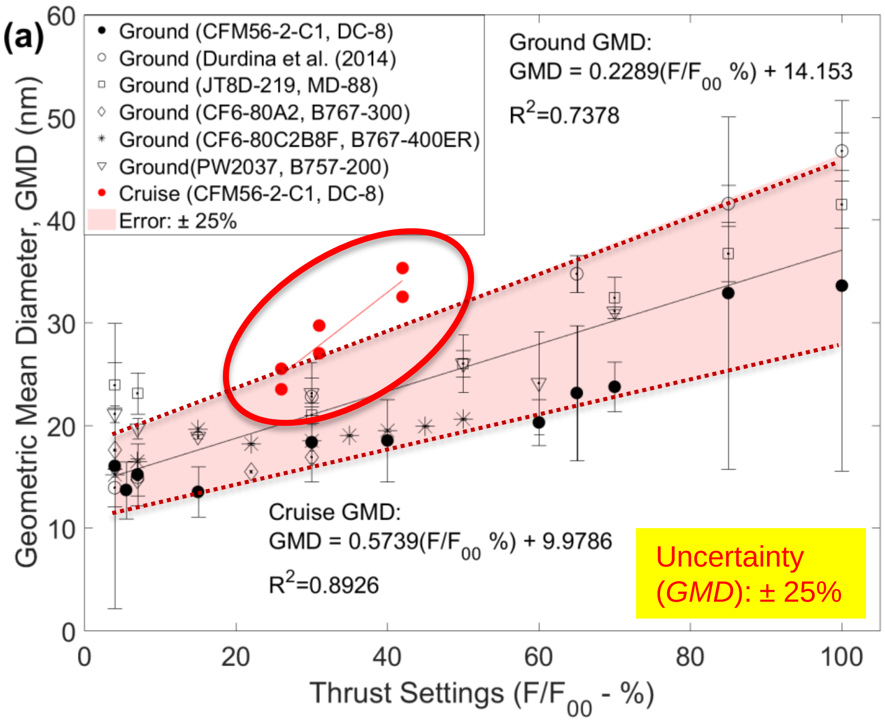
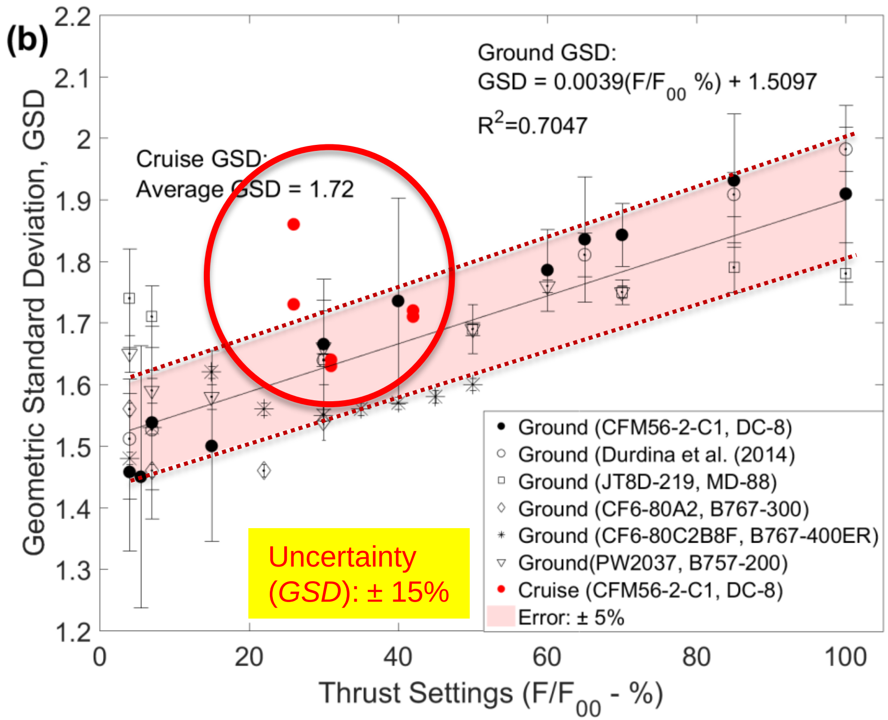
Model Inputs – (2) GMD & (3) GSD

$$\mathrm{EI_n} = \frac{\mathrm{EI_m}}{\rho_0(\frac{\pi}{6})(1.621 \times 10^{-5})^{3-D_{\mathrm{fm}}}\mathbf{GMD}^{\varphi}\exp(\frac{\varphi^2 \ln(\mathbf{GSD})^2}{2})}$$

(a) BC GMD vs. Thrust Settings (F/F_{00})



(b) BC GSD vs. Thrust Settings (F/F_{00})



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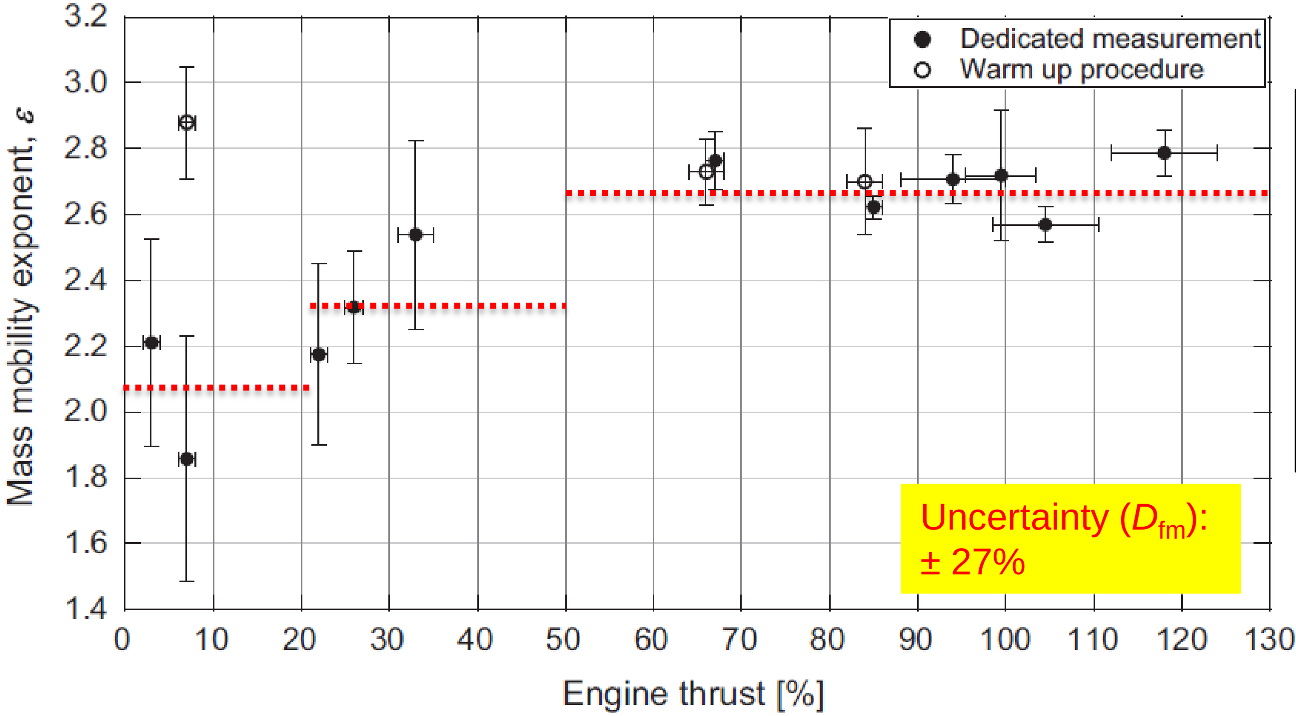
Model Input Parameters – (4) D_{fm}

$$\mathrm{EI_n} = \frac{\mathrm{EI_m}}{\rho_0(\frac{\pi}{6})(1.621\times10^{-5})^{3-D_{fm}}\mathrm{GMD}^{\varphi}\exp(\frac{\varphi^2\ln(\mathrm{GSD})^2}{2})}$$

where $\varphi = 1.17 + 0.61 D_{fm}$



$D_{fm} = 2.04$, for $0.03 \le \frac{F}{F_{00}} < 0.2$

$D_{fm} = 2.35$, for $0.2 \le \frac{F}{F_{00}} < 0.50$

$D_{fm} = 2.64$, for $0.5 \le \frac{F}{F_{00}} < 1$

*D_{fm} values are for Singular Annular Combustor (SAC) turbofan engines only

Source: [19]

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FA Model Validation – Estimated Inputs

- Cruise BC EI_n and EI_m measurements were available from the SULFUR experimental campaigns. [20] [21]
- No GMD, GSD and D_{fm} measurements were available.
- Estimated inputs for GMD, GSD and D_{fm} versus F/F_{00} (specified in previous slides) were used.
- Validation results justify the use of the GMD, GSD and D_{fm} predictive relations as model inputs to the FA model.