Aircraft Black Carbon Particle Number Emissions – New Predictive Method & Uncertainty Analysis

Marc Stettler[1], Roger Teoh[1], Ulrich Schumann[2]

[1] Centre for Transport Studies, Department of Civil and Environmental Engineering Imperial College London, London, UK

[2]Deutsches Zentrum für Luft- und Raumfahrt, Institut für Physik der Atmosphäre, 82234 Oberpfaffenhofen, Germany

Cambridge Particle Meeting 2017
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Outline

1. Introduction and motivation

2. New method to estimate particle number emissions

3. Results
    1. Validation
    2. Uncertainty analysis
    3. Application to a sample of aircraft activity
    4. Implication for contrail properties

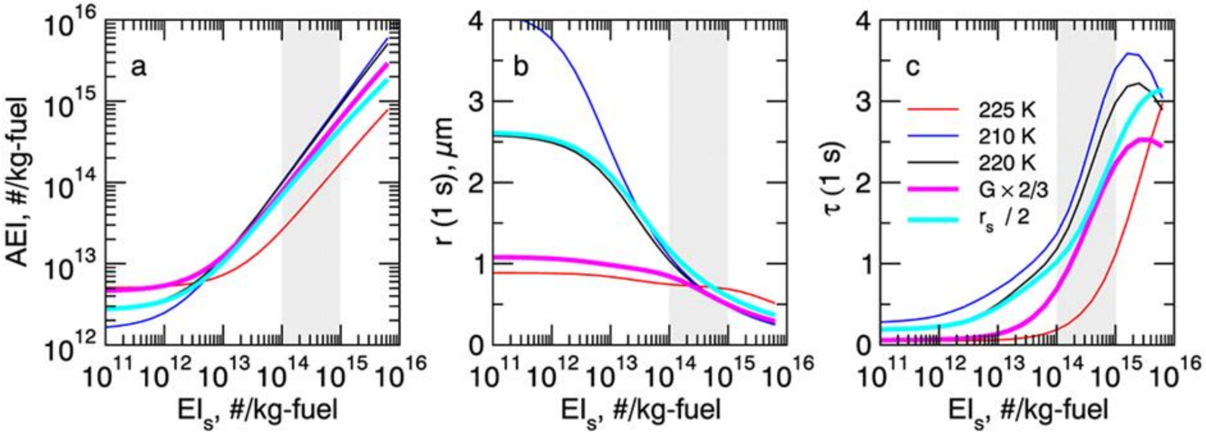
4. Conclusions

Motivation

- Knowledge of soot PSDs and ice nucleation properties is important to accurately predict visible contrail formation

- Higher EI_n leads higher number of ice particles
  - More smaller particles
  - Greater optical depth

- Measurements of ice particles in contrails and lifetimes of contrails suggests EI_n is underpredicted by up to a factor of 3



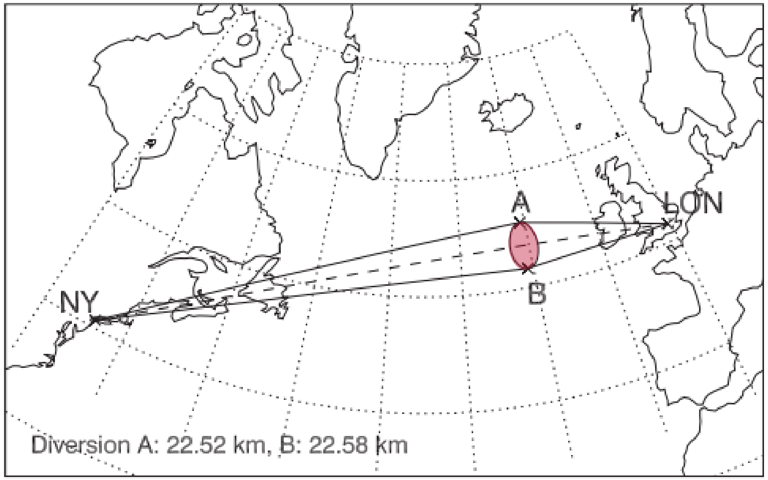
AEI – Ice nuclei EI

r – mean particle radius

τ– optical depth

Kärcher (2016). Journal of Geophysical Research: Atmospheres. 121(7), 3497-3505. http://onlinelibrary.wiley.com/doi/10.1002/2015JD024696/

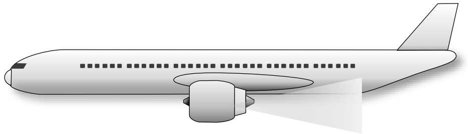
Motivation



Irvine et al. (2014). Environ. Res. Lett., vol. 9, no. 6, p. 64021.

Q: Is it worth diverting flights to avoid contrail formation?

- **Quantify BC particle number emissions (and uncertainties)**
- Contrail model needed to evaluate trade-off with CO_2
- Additional demands on air traffic control and management



CO2, PM, PN,…

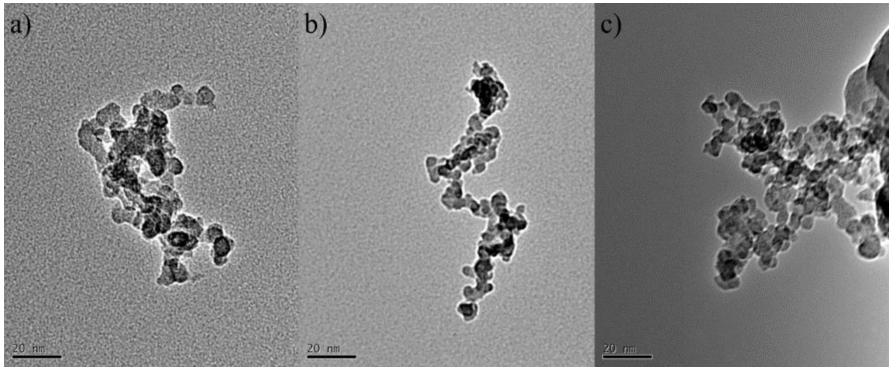
Contrail model

Radiative forcing

Air traffic control

Aircraft particle emissions

- Characterised as an **emissions index**: mass/number of particles emitted per kg of fuel burned.
- Mass (**EI_m**) and number (**EI_n**) emissions indices are dependent on the engine thrust setting.
- Size distribution (**GMD** and **GSD**) of particles is dependent on the engine thrust setting and characterised by a lognormal distribution.
- Morphology (**D_{fm}** and **d_{pp}**) of particles is dependent on the engine thrust setting.



d_m* = 15 nm d_m* = 15 nm d_m* = 50 nm

Boies, A. M. et al. (2015). Aerosol Science and Technology, 49(9), 842–855. http://doi.org/10.1080/02786826.2015.1078452

Aircraft particle mass (EI_m) estimates

- Smoke number based [1,2]
  - Limited by accuracy of certification smoke numbers
- FOX method [3]
  - Smoke numbers are discarded
  - Semi-empirical model
  - Improved BC mass estimates at ground level and at cruise
- ImFOX [4]
  - ‘Improved FOX’ method
  - Accounts for hydrogen content of fuel (*H*)
  - Different equations for global air to fuel ratio (*AFR*) at cruise and ground
- From 2020: ICAO regulation
  - Certification test and limits on non-volatile particulate matter
  - Mass and number

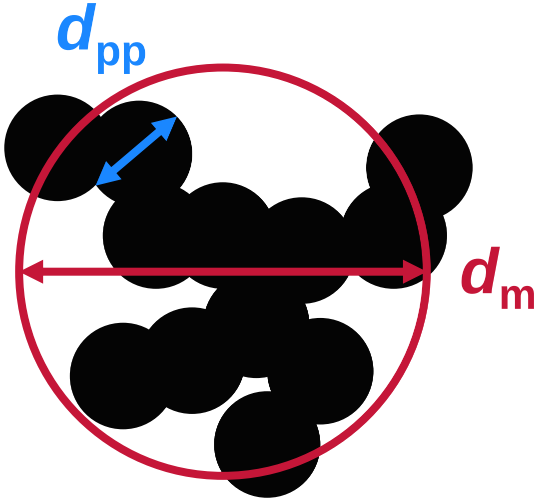
[1] Wayson et al. (2009). Journal of the Air & Waste Management Association, 59(1), 91–100. http://doi.org/10.3155/1047-3289.59.1.91
[2] Peck et al. (2013). Journal of the Air & Waste Management Association, 63(3), 367–375. http://doi.org/10.1080/10962247.2012.751467
[3] Stettler et al. (2013). Environmental Science & Technology. http://doi.org/10.1021/es401356v
[4] Abrahamson et al. (2016). Environmental Science & Technology. http://pubs.acs.org/doi/abs/10.1021/acs.est.6b03749

Aircraft particle number (EI_n) estimates

- **EI_n**
  - 10^{14} to 10^{15} per kg fuel burned
  - E.g. Kärcher et al. (2016) [1].
- **EI_n/EI_m ratios**
  - 5×10^{15} to 1.6×10^{15} particles per g(BC)
  - E.g. Döpelheuer (2002) [2].
- **Assumed particle diameter or size distribution**
  - Assume a particle diameter and density
  - E.g. d = 38 nm and ρ = 1000 kg/m^3 (Barrett et al., 2010) [3]

[1] Kärcher (2016). Journal of Geophysical Research: Atmospheres. 121 (7), 3497-3505. http://onlinelibrary.wiley.com/doi/10.1002/2015JD024696/full
[2] Döpelheuer (2002) Anwendungsorientierte Verfahren Zur Bestimmung Von CO, HC Und Ruß Aus Luftfahrttriebwerken.
[3] Barrett, S. R. H. et al. (2010). Guidance on the use of AEDT gridded aircraft emissions in atmospheric models. http://lae.mit.edu/uploads/LAE_report_series/2010/LAE-2010-008-N.pdf

Mobility of fractal aggregates



Number of primary particles:

$$n_{pp} \approx \left(\frac{d_m}{d_{pp}}\right)^{D_{fm}}$$

Mass of the aggregate is the sum of the mass of primary particles:

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

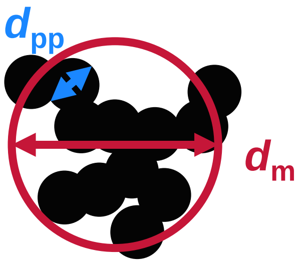
Free-molecular regime:
Kn = λ/d_m >> 1 (mean free path >> d)

Diffusion limited cluster aggregation (DLCA):
Aerosols aggregate via random Brownian motion

D_{fm} is the **mass-mobility exponent**
n_{pp} is the **number of primary particles**
m is the **mass of the aggregate**
ρ_0 is the **material density of black carbon (1770 ± 8%)**

Sorensen, C. M. (2011). Aerosol Science and Technology, 45(7), 765–779. http://doi.org/10.1080/02786826.2011.560909

Mass of aggregates



Number of primary particles: $n_{\mathrm{pp}} = \left(\frac{d_{\mathrm{m}}}{d_{\mathrm{pp}}}\right)^{D_{\mathrm{fm}}}$

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

Primary particle diameter: $d_{\mathrm{pp}}[\mathrm{m}] = a\ d_{\mathrm{m}}^b$

$$\boxed{\therefore m = \rho_0 \left(\frac{\pi}{6}\right) d_{\mathrm{m}}^{\phi} a^{3-D_{\mathrm{fm}}}}$$

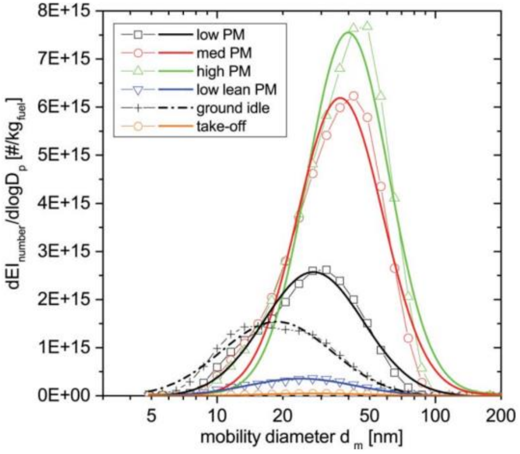
where $\phi = D_{\mathrm{fm}} + b(3 - D_{\mathrm{fm}})$

Number and mass of aggregate population

Mass of a collection of aggregates with size distribution $n(d_\mathrm{m})$:

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

$$= \rho_0 \left(\frac{\pi}{6}\right) a^{3-D_\mathrm{fm}} \int_0^\infty d_\mathrm{m}^{\phi}\, n(d_\mathrm{m}) \mathrm{dlog} d_\mathrm{m}$$



Lobo, et al. (2015).

$n(d_\mathrm{m})$ for non-volatile aircraft PM is typically lognormal (single mode). This then becomes the ϕ-th moment of the lognormal distribution:

$$\boxed{M = N \rho_0 \left(\frac{\pi}{6}\right) a^{3-D_\mathrm{fm}} \mathrm{GMD}^{\phi} \exp\left(\frac{\phi^2 \log(\mathrm{GSD})^2}{2}\right)}$$

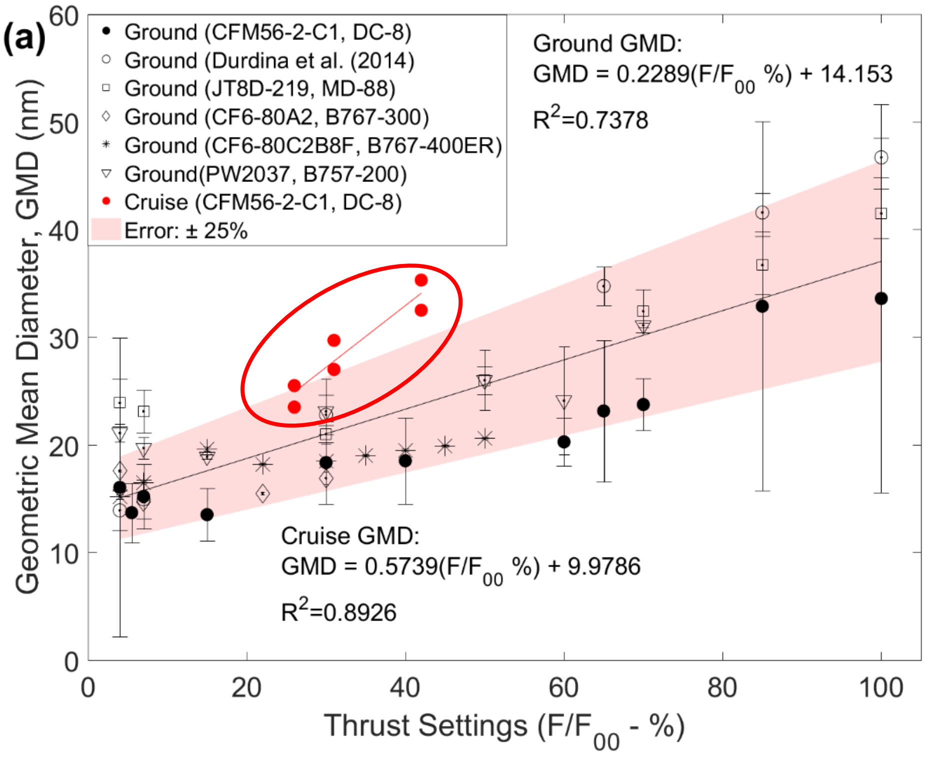
Re-arrange for N…

where $\phi = D_\mathrm{fm} + b(3 - D_\mathrm{fm})$

Lobo, P., et al. (2015). Aerosol Science and Technology, 49(7), 472–484. http://doi.org/10.1080/02786826.2015.1047012

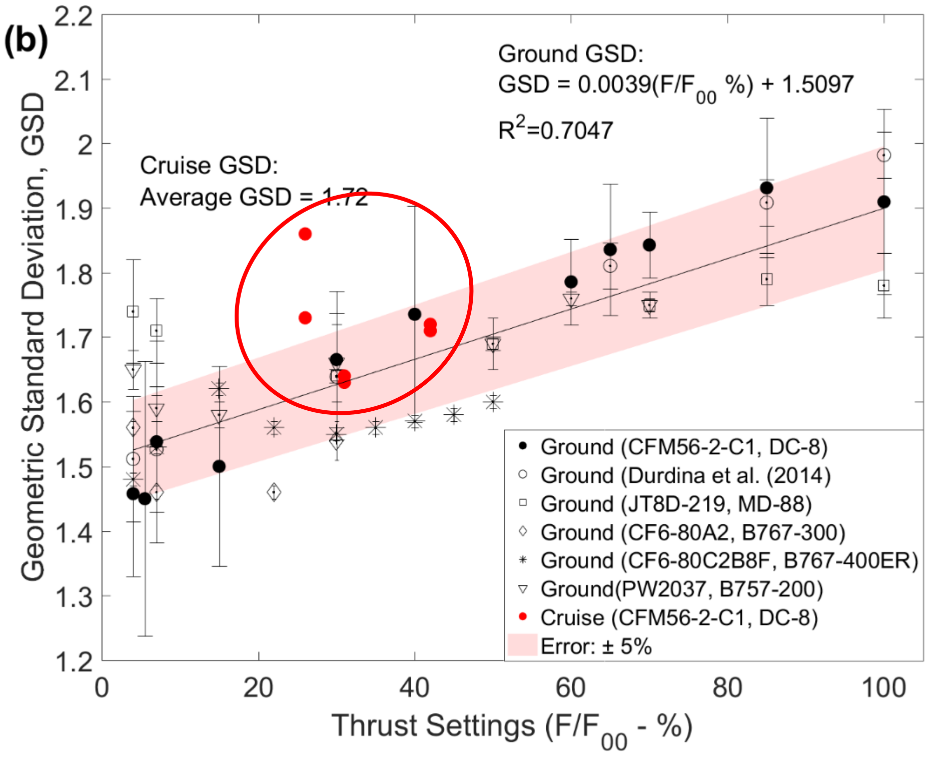
Aircraft PM GMD and GSD

GMD



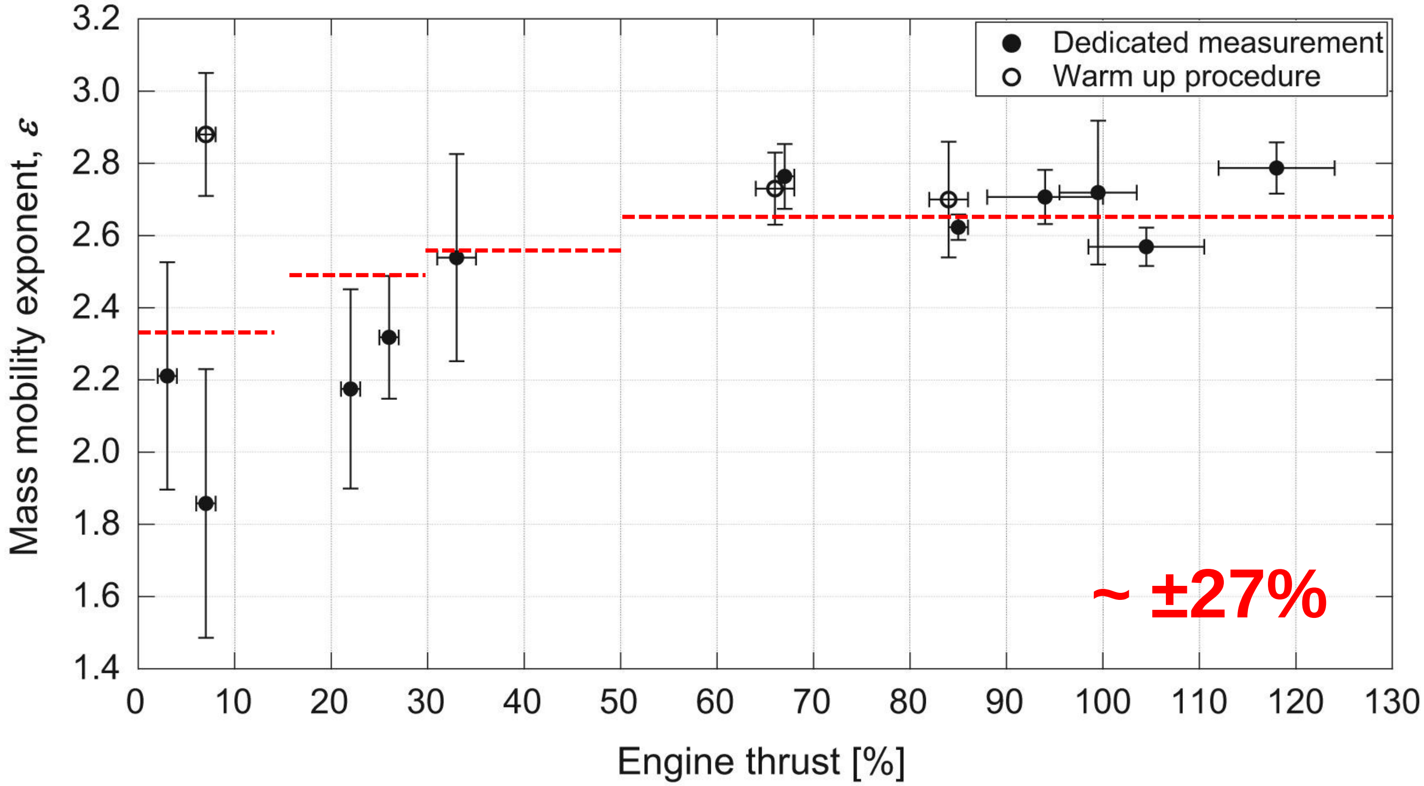
~ ±25%

GSD



~ ±14.5%

Mass-mobility exponent - D_{fm}



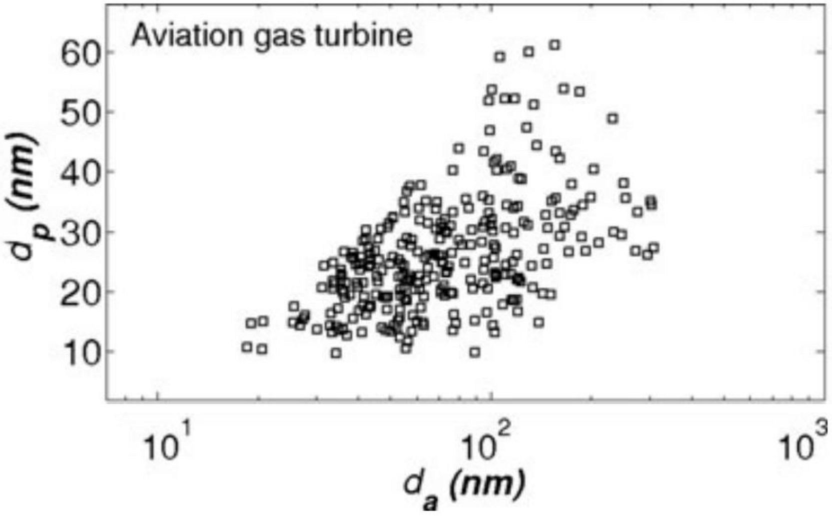
$$D_{\mathrm{fm}}(\mathrm{SAC}) = 2.37 \quad for\ 0.03 \ll \frac{F}{F_{00}} < 0.15$$
$$D_{\mathrm{fm}}(\mathrm{SAC}) = 2.50 \quad for\ 0.15 \ll \frac{F}{F_{00}} < 0.30$$
$$D_{\mathrm{fm}}(\mathrm{SAC}) = 2.57 \quad for\ 0.30 \ll \frac{F}{F_{00}} < 0.50$$
$$D_{\mathrm{fm}}(\mathrm{SAC}) = 2.64 \quad for\ 0.50 \ll \frac{F}{F_{00}} \ll 1.00$$

Abegglen, M. et al. (2015). Journal of Aerosol Science, 88, 135–147. http://doi.org/10.1016/j.jaerosci.2015.06.003

Primary particle diameter - $d_{\mathrm{pp}}[\mathrm{m}] = a\, d_{\mathrm{m}}^{b}$

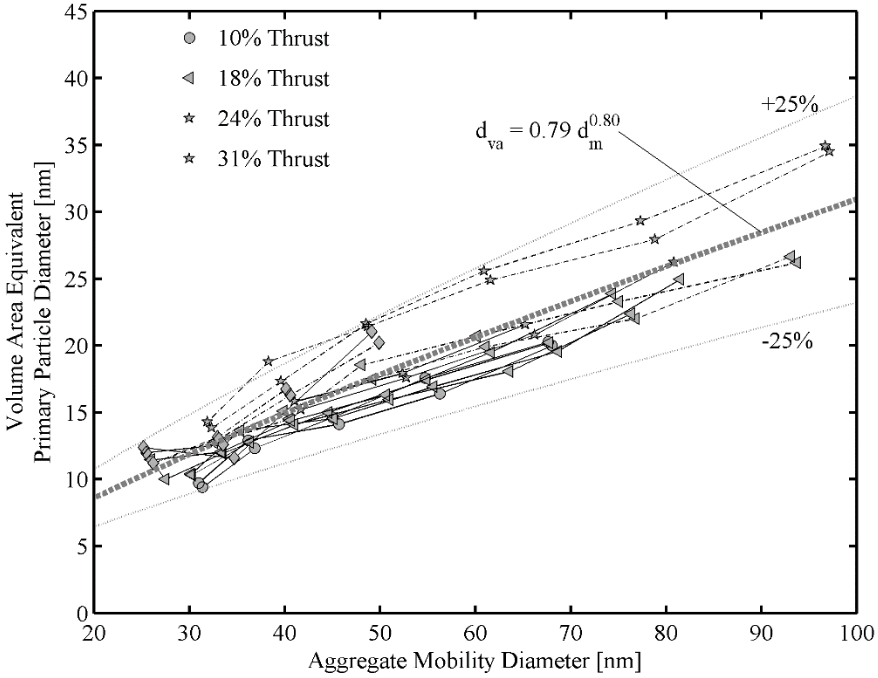
$$d_{\mathrm{pp}}[\mathrm{m}] = 1.62 \times 10^{-5}\, d_{\mathrm{m}}^{0.39}$$

$$d_{\mathrm{pp}}[\mathrm{m}] = 0.0125\, d_{\mathrm{m}}^{0.80}$$



Dastanpour & Rogak

[$d_a = d_m$ in free-molecular and transition regimes]*



Boies et al.

Boies, A. M. et al. (2015). Aerosol Science and Technology, 49(9), 842–855. http://doi.org/10.1080/02786826.2015.1078452
Dastanpour & Rogak (2014). Aerosol Science and Technology, 48(10), 1043-1049. http://www.tandfonline.com/doi/full/10.1080/02786826.2014.955565
*Dastanpour et al. (2016). Aerosol Science and Technology, 50(2), 101-109. http://www.tandfonline.com/doi/abs/10.1080/02786826.2015.1130796

Primary particle diameter - $d_{\mathrm{pp}}[\mathrm{m}] = a\, d_{\mathrm{m}}^{b}$

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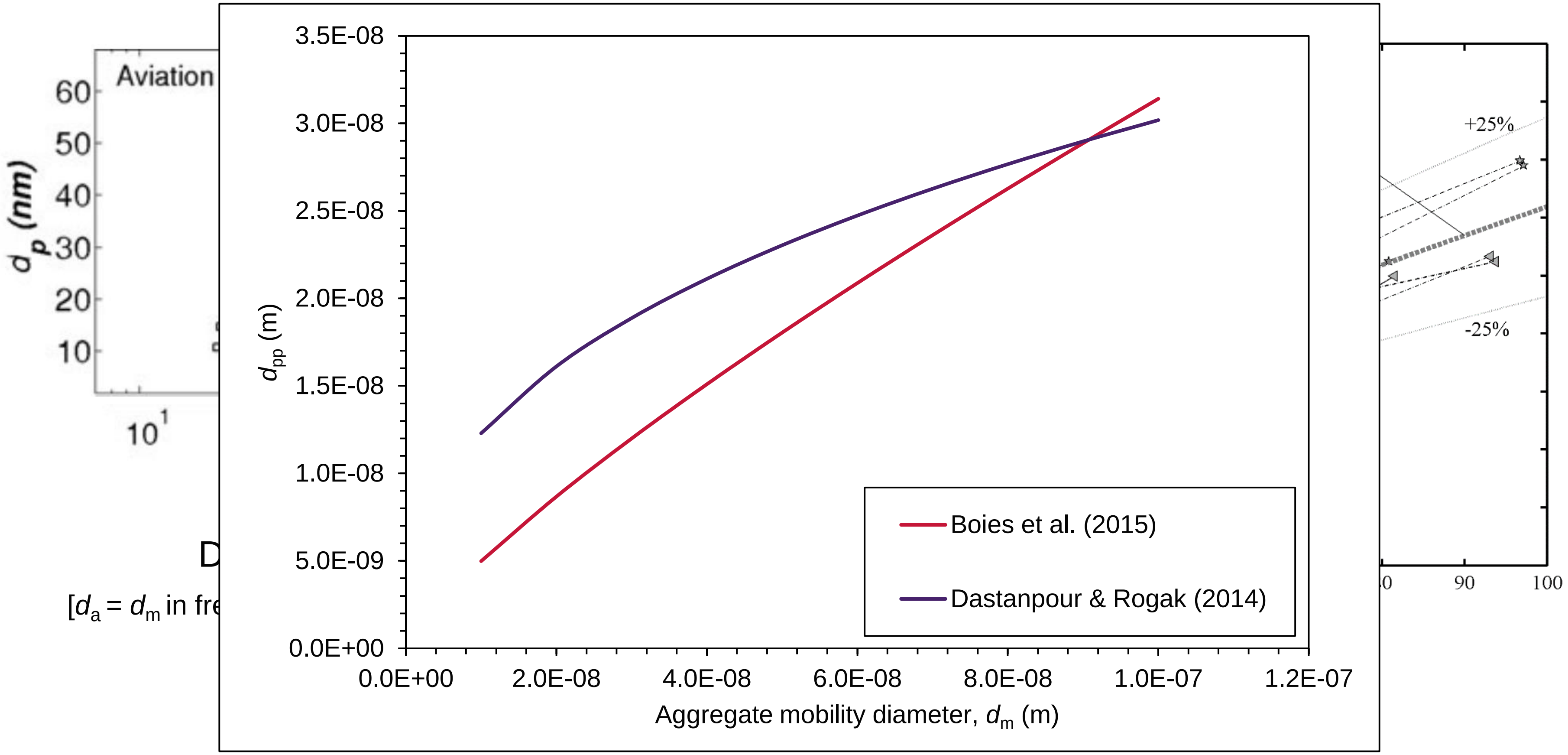


D

[d_a = d_m in fre

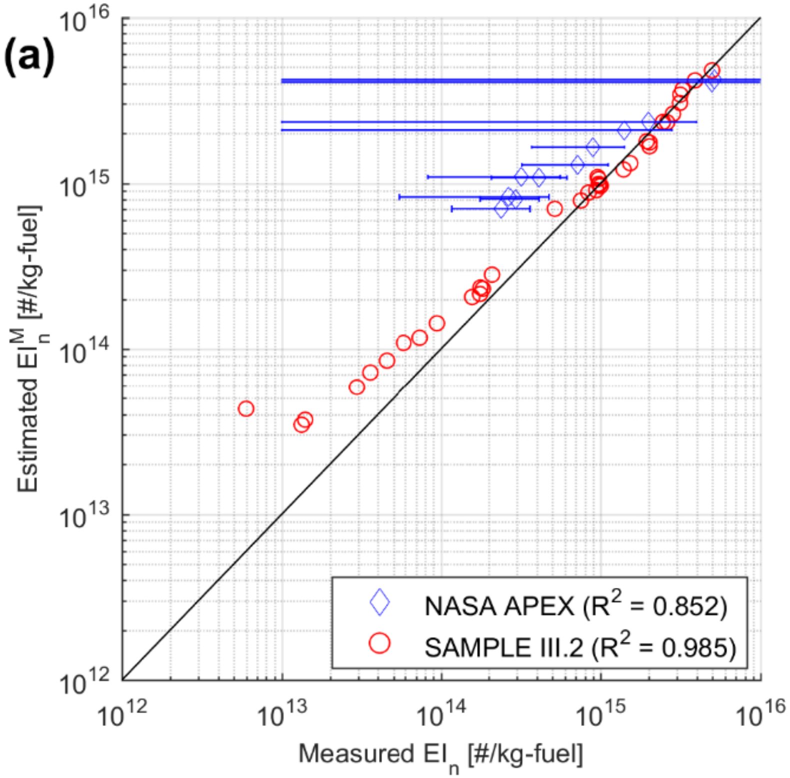
EI$_n$ estimates are within ~ ±20%

Boies, A. M. et al. (2015). Aerosol Science and Technology, 49(9), 842–855. http://doi.org/10.1080/02786826.2015.1078452
Dastanpour & Rogak (2014). Aerosol Science and Technology, 48(10), 1043-1049. http://www.tandfonline.com/doi/full/10.1080/02786826.2014.955565
*Dastanpour et al. (2016). Aerosol Science and Technology, 50(2), 101-109. http://www.tandfonline.com/doi/abs/10.1080/02786826.2015.1130796

Results: Validation of $\mathrm{EI_n}$

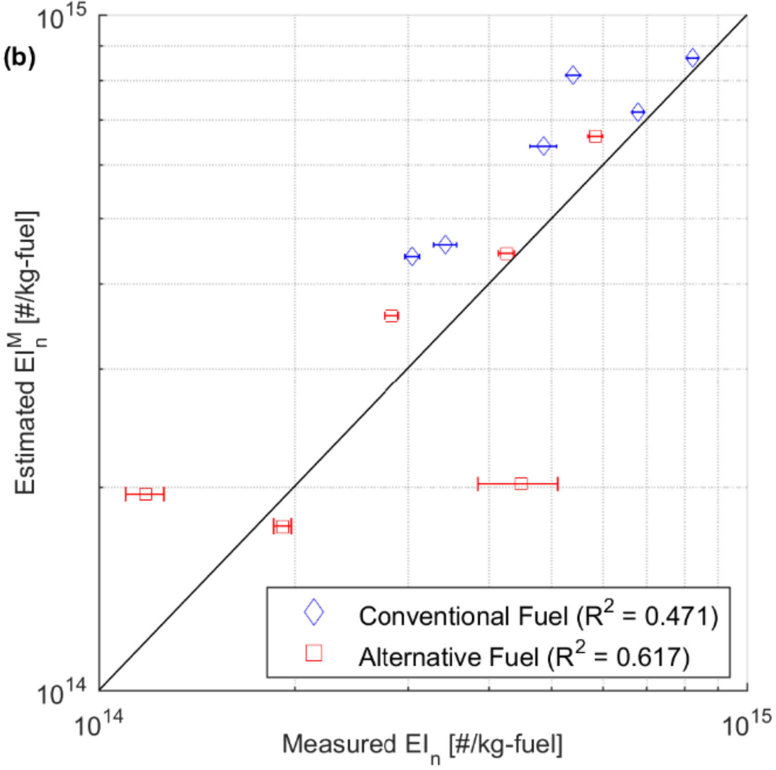
$$\mathrm{EI_n} = \frac{\mathrm{EI_m}}{\rho_0 \left(\frac{\pi}{6}\right) a^{3-D_{\mathrm{fm}}} \mathrm{GMD}^{\phi} \exp\left(\frac{\phi^2 \log(\mathrm{GSD})^2}{2}\right)}$$

Ground



c.f. R^2 = [-0.635, 0.102]

Cruise



c.f. R^2 = [-0.304, 0.156]

NASA APEX: Wey et al. (2006). Aircraft particle emissions experiment (APEX). NASA.
SAMPLE III.2: Boies et al. (2015). Aerosol Science and Technology, 49(9), 842–855. http://doi.org/10.1080/02786826.2015.1078452
Cruise: Moore et al. (2017). Nature. 543 (7645), 411-415. http://www.nature.com/nature/journal/v543/n7645/full/nature21420.html

Application to fleet

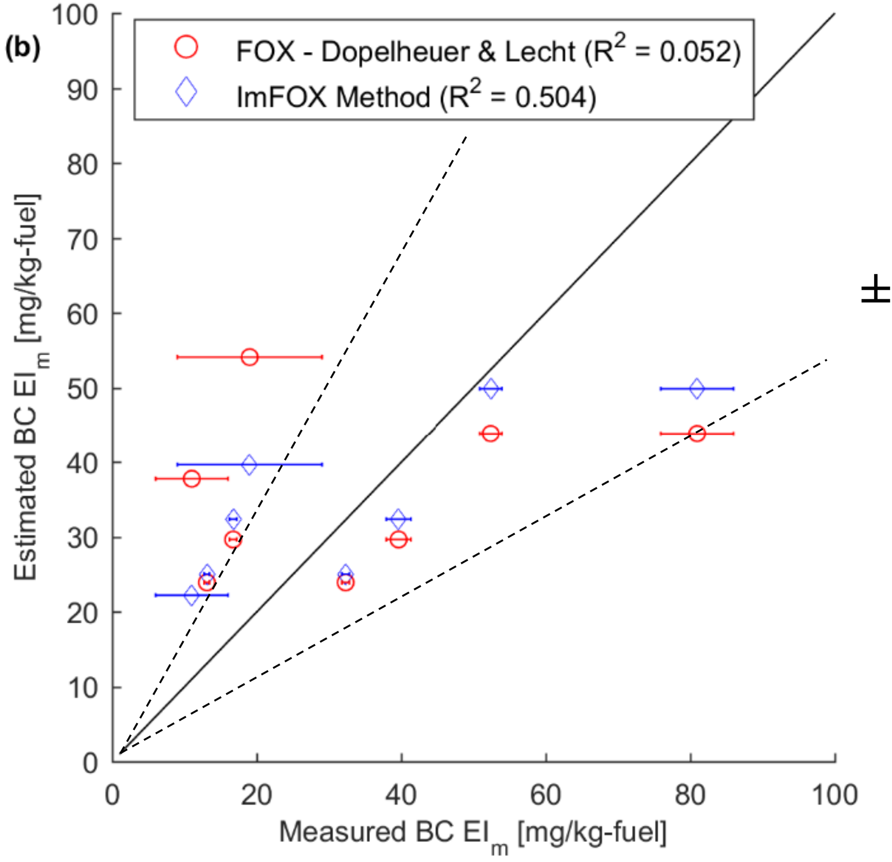
- Need to estimate the BC mass emissions (EI_m)

$$EI_n = \frac{EI_m}{\rho_0 \left(\frac{\pi}{6}\right) a^{3-D_{fm}} GMD^{\phi} \exp\left(\frac{\phi^2 \log(GSD)^2}{2}\right)}$$

- Sample of Aviation Environmental Design Tool (AEDT) dataset
  - 9th March 2006 23:15:04 to 11th March 2006 08:09:35
  - Flight levels from 15,000 ft to 43,000 ft
  - 3371 flights
    - US Flights ≈ 3258
    - Asian Flights ≈ 10
    - Transatlantic & EU Flights ≈ 103

Results: Estimates of EI_m at cruise

$$\mathrm{EI_n} = \frac{\mathrm{EI_m}}{\rho_0 \left(\frac{\pi}{6}\right) a^{3-D_{\mathrm{fm}}} \mathrm{GMD}^{\phi} \exp\left(\frac{\phi^2 \log(\mathrm{GSD})^2}{2}\right)}$$



±50%

Results: Uncertainty analysis

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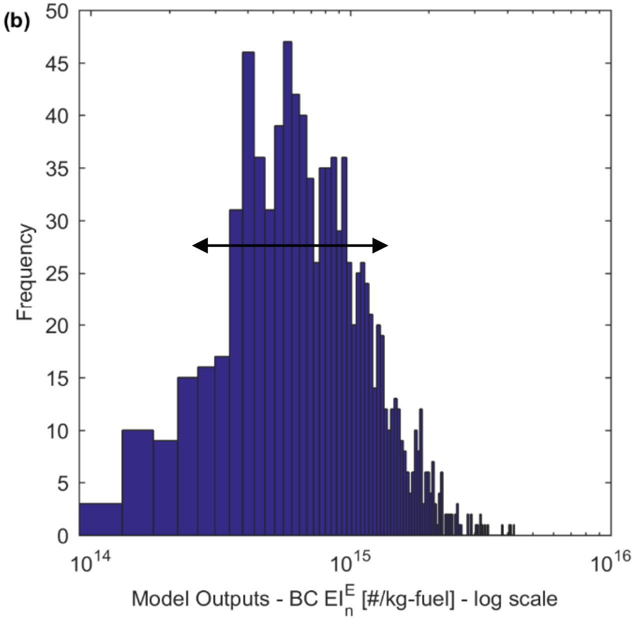
$$\mathrm{EI_n} = \frac{\mathrm{EI_m}}{\rho_0 \left(\frac{\pi}{6}\right) a^{3-D_{\mathrm{fm}}} \mathrm{GMD}^{\phi} \exp\left(\frac{\phi^2 \log(\mathrm{GSD})^2}{2}\right)}$$

Variable	Fixed F/F$_{00}$	Mean (μ)	Error
$\mathrm{EI_m}$	0.4	14.2 mg/kg	±50%
ρ_0	0.4	1770 kg/m^3	±8%
D_{fm}	0.4	2.57	±27%
GMD	0.4	18.52 nm	±25%
GSD	0.4	1.736	±14.5%

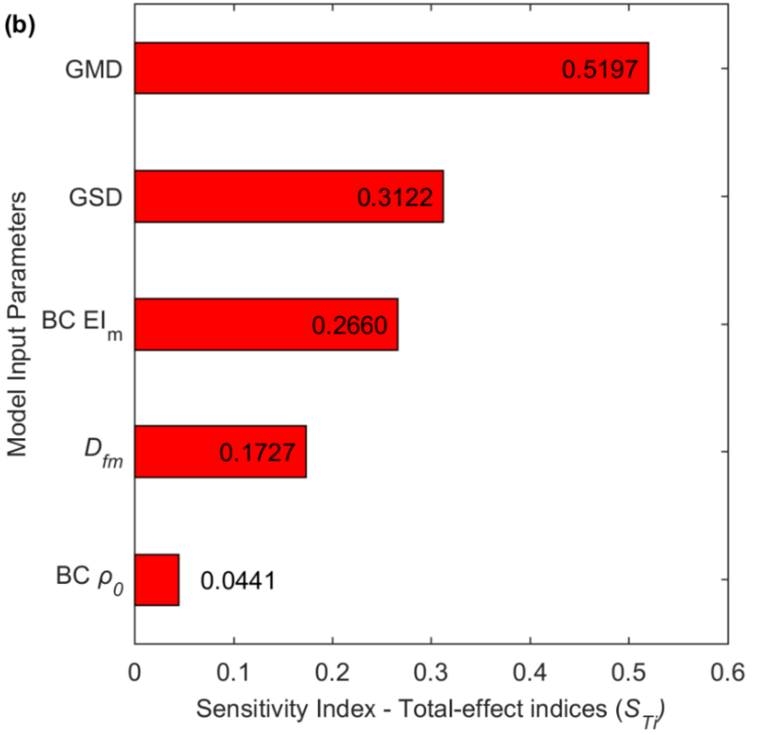


Propagated uncertainty (σ/μ) ~ ±64%

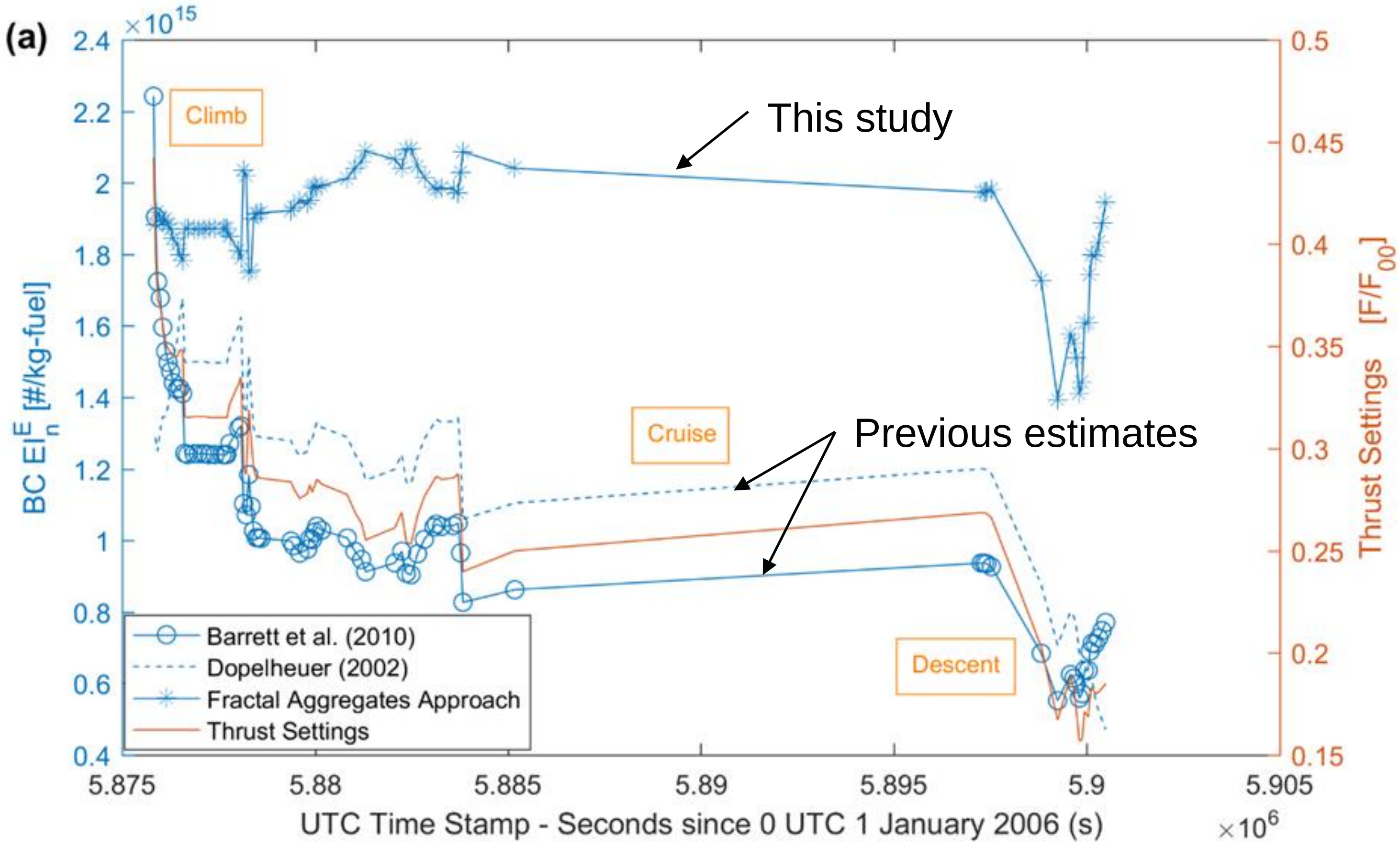
Results: Sensitivity analysis

$$\mathrm{EI_n} = \frac{\mathrm{EI_m}}{\rho_0\left(\frac{\pi}{6}\right) a^{3-D_{\mathrm{fm}}}\,\mathrm{GMD}^{\phi}\exp\left(\frac{\phi^2 \log(\mathrm{GSD})^2}{2}\right)}$$

- Uncertainty in inputs propagates to uncertainty in $\mathrm{EI_n}$
- GMD, GSD and $\mathrm{EI_m}$ are the most critical parameters
- A ±10% change in GMD will result in $\mathrm{EI_n}$ varying by approximately -23% to +33%



Results: Example flight

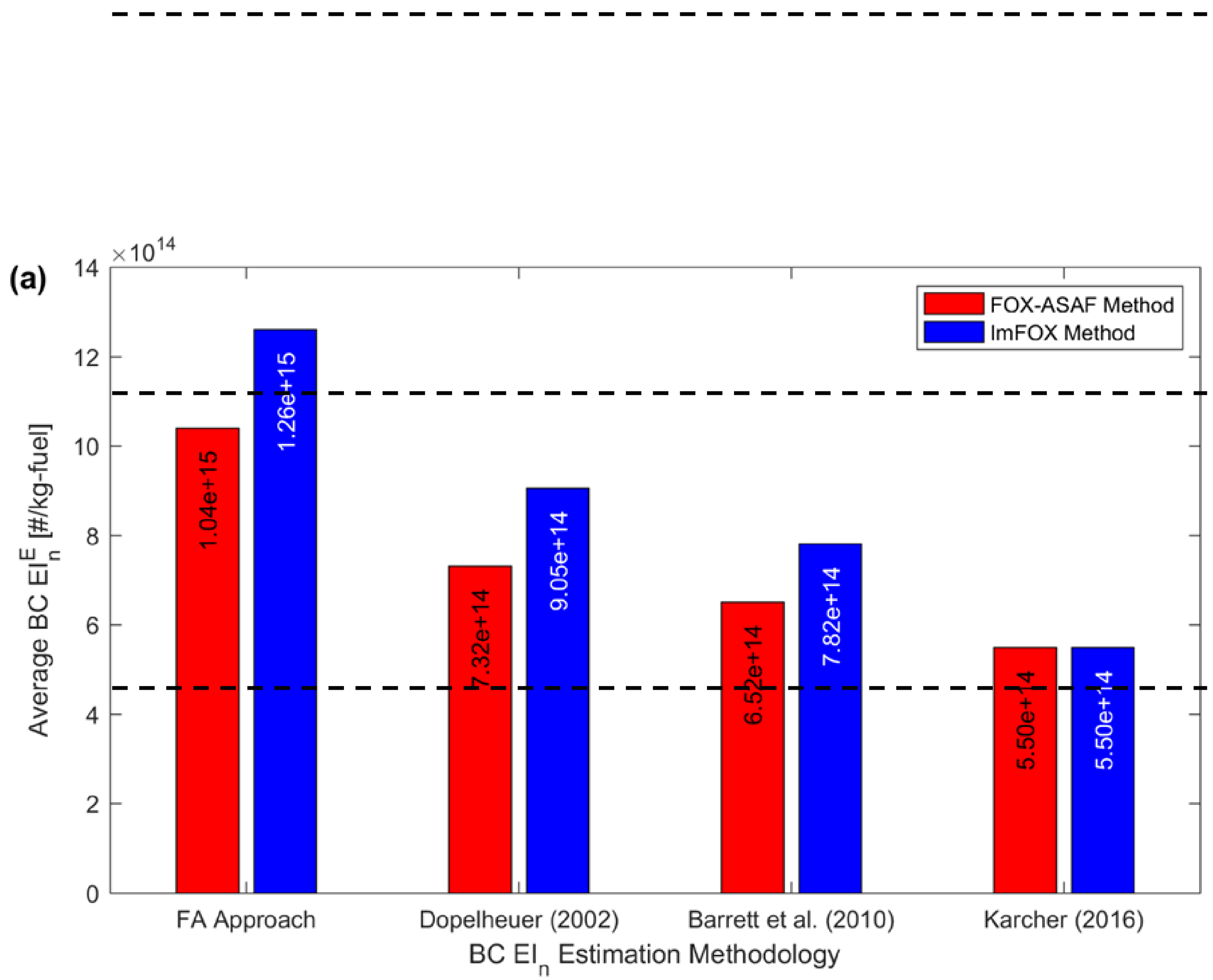


This study

Previous estimates

- New fractal aggregate approach leads to estimates that are ~2x higher than previous estimates
- There is additional dependence on particle size and morphology

Results: Comparison of fleet average EI_n



Upper 1.9×10^{15} kg^{-1}

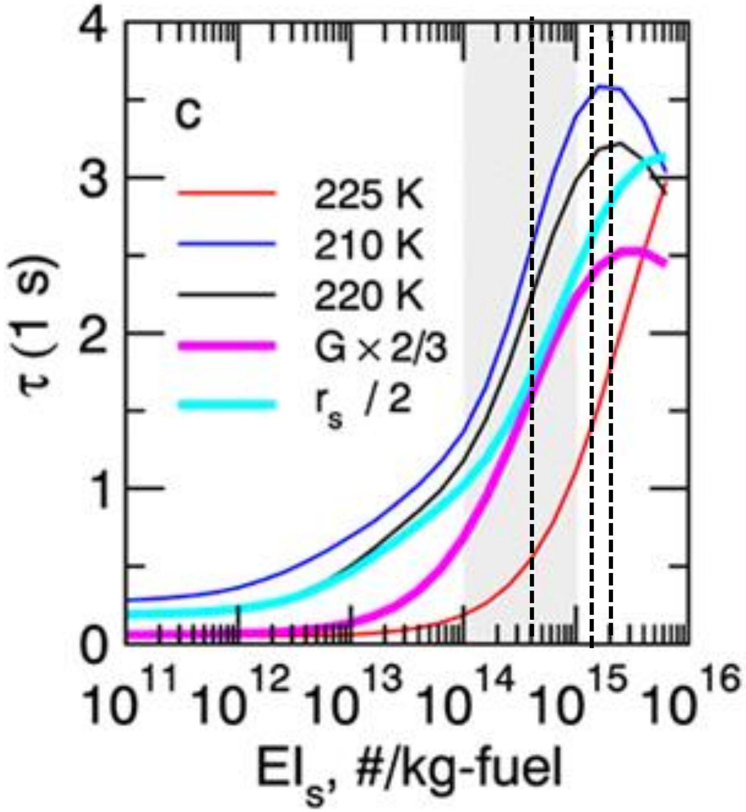
Mean 1.2×10^{15} kg^{-1}

Lower 4.4×10^{14} kg^{-1}

Implications for contrails

220 K [210 K, 225 K]

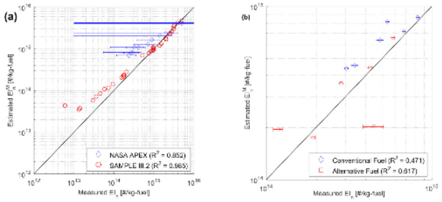
Input Scenario	BC EI_n (kg^{-1})	Mean Visible Optical Depth (τ)	Difference
Mean	1.2×10^{15}	3 [3.5, 1.3]	
Lower Bound	4.4×10^{14}	2.36 [2.68, 0.61]	-21% [-23%, -53%]
Upper Bound	1.9×10^{15}	3.20 [3.6, 1.68]	+7% [+3%, +29%]
Previous estimates	$\sim 6 \times 10^{14}$	~2.5	



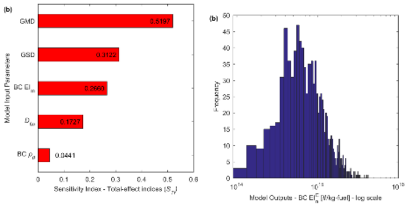
Contrail optical depth may be ~20% higher than with previous estimates

Conclusions

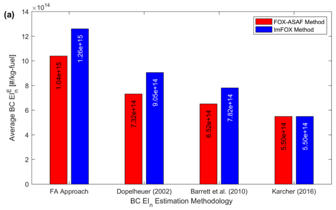
- New approach based on mobility of fractal aggregates provides more accurate EI_n estimates than previous methods.



- Large uncertainties remain and sensitivity analysis show that GMD, GSD and EI_m are most important parameters.

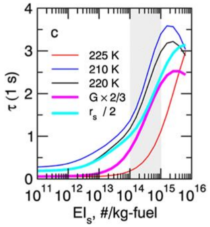


- Fleet average EI_n of 1.2 [0.44, 1.9] $\times 10^{15}$ per kg of fuel.



- Application to a sample of aircraft activity data suggests average EI_n are revised upwards by a factor of ~2. Previous estimates are within the lower bound.

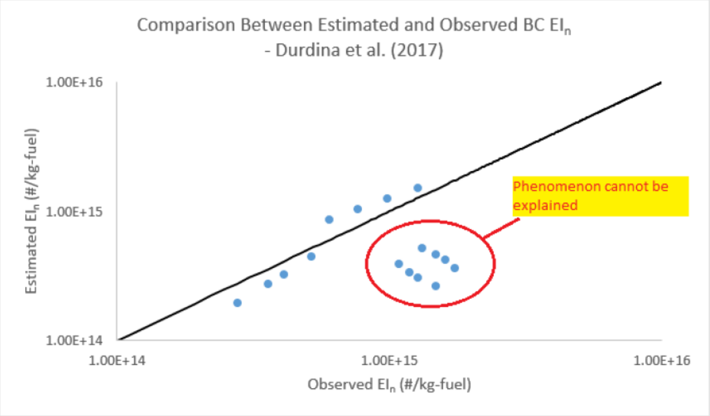
- Contrail optical depth may be ~20% higher than previous estimates

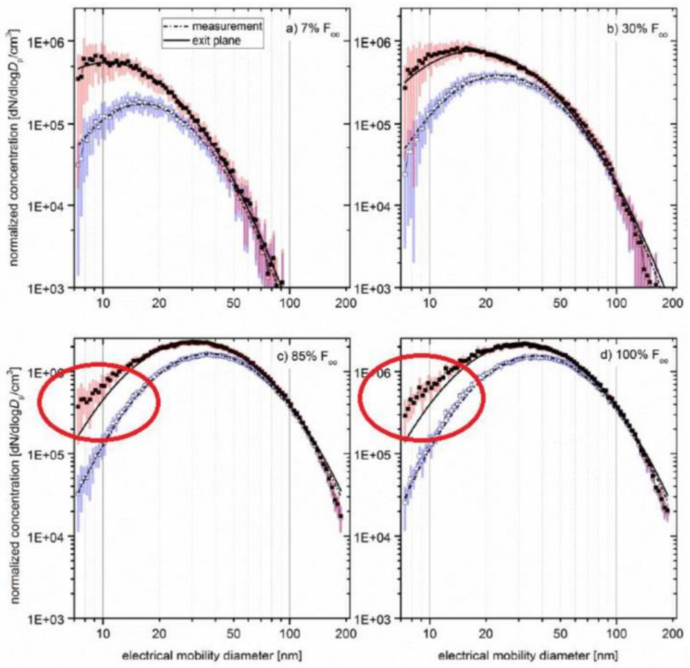


Future work

- Further validation.
- Integrate method into contrail model to quantify any change in contrail properties.
- Are models of ice nucleation in contrails appropriate for fractal aggregates?

- Particle losses when measuring EI_n through long sample lines:





Durdina et al., (2017). http://pubs.acs.org/doi/abs/10.1021/acs.est.6b05801

Acknowledgements

- 2006 AEDT sample dataset used for this study was made available for climate research by FAA within the ACCRI research project.

- A. Boies for SAMPLE III.2 data (and all other collaborators) and comments.

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Imperial College London

Thanks, questions?

m.stettler@imperial.ac.uk | www.imperial.ac.uk/people/m.stettler

Centre for Transport Studies | Department of Civil and Environmental Engineering Imperial College London

@TransEnvLab_IC

Aircraft particle mass ($\mathrm{EI_m}$) estimates

- Smoke number based
    - First Order Approximation v3 [1]
    - Aerodyne Research Inc. method [2]
    - Limited by accuracy of certification smoke numbers

- FOX method [3]
    - Smoke numbers are discarded
    - Semi-empirical model fitted to measurements
    - Improved BC mass estimates at ground level and at cruise

$$C_{\mathrm{BC}}\left[\frac{\mathrm{mg}}{\mathrm{m}^3}\right] \approx \dot{m}_f \left(\underbrace{A_{form}\, e^{\left(-\frac{6390}{T_{fl}}\right)}}_{\text{Formation}} - \underbrace{\mathrm{AFR}\, A_{ox}\, e^{\left(-\frac{19778}{T_{fl}}\right)}}_{\text{Oxidation}}\right)$$

[1] Wayson et al. (2009). Journal of the Air & Waste Management Association, 59(1), 91–100. http://doi.org/10.3155/1047-3289.59.1.91
[2] Peck et al. (2013). Journal of the Air & Waste Management Association, 63(3), 367–375. http://doi.org/10.1080/10962247.2012.751467
[3] Stettler et al. (2013). Environmental Science & Technology. http://doi.org/10.1021/es401356v
[4] Abrahamson et al. (2016). Environmental Science & Technology. http://pubs.acs.org/doi/abs/10.1021/acs.est.6b03749

Aircraft particle mass ($\mathrm{EI_m}$) estimates

- ImFOX [4]
  - 'Improved FOX' method
  - Accounts for hydrogen content of fuel (*H*)
  - Quadratic equation for A_{form} and different equation for T_4
  - Different equations for global air to fuel ratio (*AFR*) at cruise and ground

$$C_{\mathrm{BC}}\left[\frac{\mathrm{mg}}{\mathrm{m}^3}\right] \approx \dot{m}_f\, e^{13.6-H}\left(A_{\mathrm{form}}\, e^{\left(-\frac{6390}{T_4}\right)} - \mathrm{AFR}\, A_{\mathrm{ox}}\, e^{\left(-\frac{19778}{T_{\mathrm{fl}}}\right)}\right)$$

- From 2020: ICAO regulation
  - Certification test and limits on non-volatile particulate matter
  - Mass and number

[1] Wayson et al. (2009). Journal of the Air & Waste Management Association, 59(1), 91–100. http://doi.org/10.3155/1047-3289.59.1.91
[2] Peck et al. (2013). Journal of the Air & Waste Management Association, 63(3), 367–375. http://doi.org/10.1080/10962247.2012.751467
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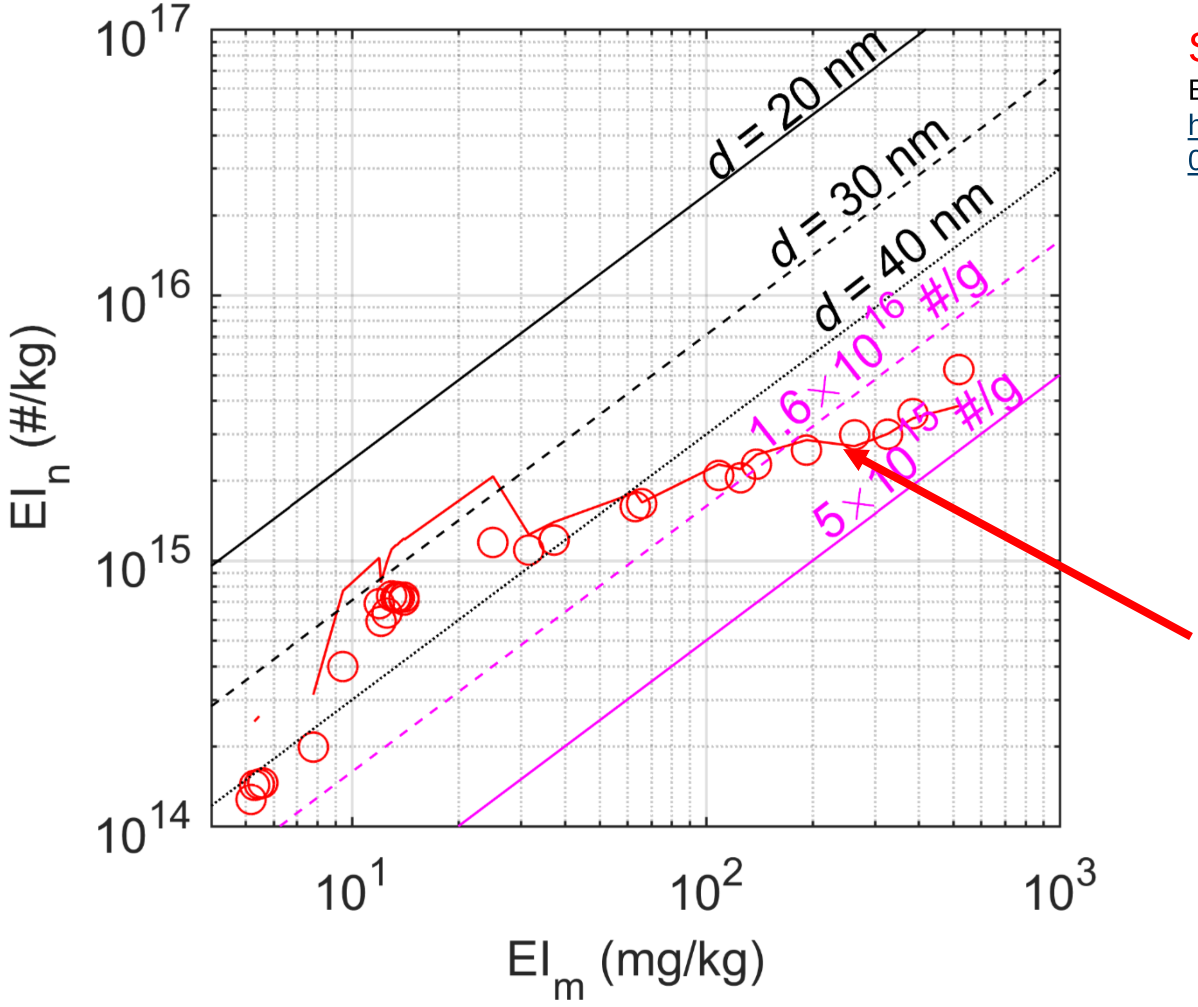
[4] Abrahamson et al. (2016). Environmental Science & Technology. http://pubs.acs.org/doi/abs/10.1021/acs.est.6b03749

Results: EI_n versus EI_m



SAMPLE III.2

Boies, A. M. et al. (2015).
http://doi.org/10.1080/02786826.2015.1078452

Modelled EI_n
R^2 = 0.985
SAMPLE III.2

EI_n dependence on engine thrust and altitude