

# Impacts of aviation fuel sulfur content on climate and human health

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## Abstract

Aviation emissions impact both air quality and climate. Using a coupled tropospheric chemistry-aerosol microphysics model we investigate the effects of varying aviation fuel sulfur content (FSC) on premature mortality from long-term exposure to aviation-sourced PM<sub>2.5</sub> (particulate matter with a dry diameter of <2.5 μm) and on the global radiation budget due to changes in aerosol and tropospheric ozone. We estimate that present-day non-CO<sub>2</sub> aviation emissions with a typical FSC of 600 ppm result in ~3,600 [95% CI: 1,310–5,890] annual premature mortalities globally due to increases in cases of cardiopulmonary disease and lung cancer, resulting from increased surface PM<sub>2.5</sub> concentrations. We quantify the global annual mean combined radiative effect (RE<sub>comb</sub>) of non-CO<sub>2</sub> aviation emissions as –13.3 mW m<sup>-2</sup>; from increases in aerosols (direct radiative effect and cloud albedo effect) and tropospheric ozone.

Ultra-low sulfur jet fuel (ULSJ; FSC = 15 ppm) has been proposed as an option to reduce the adverse health impacts of aviation-induced PM<sub>2.5</sub>. We calculate that swapping the global aviation fleet to ULSJ fuel would reduce the global aviation-induced mortality rate by ~620 [95% CI: 230–1020] mortalities a<sup>-1</sup> and increase RE<sub>comb</sub> by +7.0 mW m<sup>-2</sup>.

We explore the impact of varying aviation FSC between 0–6000 ppm. Increasing FSC increases aviation-induced mortality, while enhancing climate cooling through increasing the aerosol cloud albedo effect (CAE). We explore the relationship between the injection altitude of aviation emissions and the resulting climate and air quality impacts. Compared to the standard aviation emissions distribution, releasing aviation emissions at the ground increases global aviation-induced mortality and produces a net warming effect, primarily through a reduced CAE. Aviation emissions injected at the surface are 5 times less effective at forming cloud condensation nuclei, reducing the aviation-induced CAE by a factor of 10. Applying high FSCs at aviation cruise altitudes combined with ULSJ fuel at lower altitudes results in reduced aviation-induced mortality and increased negative RE compared to the baseline aviation scenario.

## 53 1 Introduction

54 Aviation is the fastest growing form of transport (Eyring et al., 2010; Lee et al., 2010; Uherek et al., 2010), with  
55 a projected growth in passenger air traffic of 5% yr<sup>-1</sup> until 2030 (Barrett et al., 2012; ICAO, 2013), and a  
56 projected near doubling of emissions by 2025, relative to 2005 (Eyers et al., 2004). These emissions, and  
57 changes to them, have both climate and air quality impacts (Lee et al., 2009; Barrett et al., 2010; Woody et al.,  
58 2011; Barrett et al., 2012).

59  
60 Aviation emits a range of gas-phase and aerosol pollutants that can influence climate. Emissions of carbon  
61 dioxide (CO<sub>2</sub>) from aviation warm the climate (Lee et al., 2009; Lee et al., 2010). Emissions of nitrogen oxides  
62 (NO<sub>x</sub>) warm the climate through tropospheric ozone (O<sub>3</sub>) formation, which acts as a greenhouse gas, and cool  
63 climate via a decrease in the lifetime of the well-mixed greenhouse gas methane (CH<sub>4</sub>) through increases in  
64 the OH radical (Holmes et al., 2011; Myhre et al., 2011). Sulfate and nitrate aerosols, formed from aviation  
65 sulfur dioxide (SO<sub>2</sub>) and NO<sub>x</sub> emissions and through altered atmospheric oxidants, lead to a cooling (Unger,  
66 2011; Righi et al., 2013; Dessens et al., 2014), and black carbon (BC) emissions result in a warming (Balkanski  
67 et al., 2010). Additionally, the formation of persistent linear contrails and contrail-cirrus from aircraft leads to  
68 warming (Lee et al., 2010; Rap et al., 2010; Burkhardt and Karcher, 2011). Overall, aviation emissions are  
69 thought to have a warming impact on climate, with net radiative forcing (RF) estimated as +55 mW m<sup>-2</sup>  
70 (excluding cirrus cloud enhancement) (Lee et al., 2010).

71  
72 Previous studies have separately assessed the impacts of aviation through different atmospheric species.  
73 Short-term O<sub>3</sub> has been estimated to have a radiative effect ranging between 6–36.5 mW m<sup>-2</sup> (Sausen et al.,  
74 2005; Köhler et al., 2008; Hoor et al., 2009; Lee et al., 2009; Holmes et al., 2011; Myhre et al., 2011; Unger,  
75 2011; Frömming et al., 2012; Skowron et al., 2013; Unger et al., 2013; Khodayari et al., 2014; Brasseur et al.,  
76 2015). The aerosol direct effect is highly uncertain [–28 to +20 mW m<sup>-2</sup>] (Righi et al., 2013), with the direct  
77 aerosol effects for sulfate ranging between –0.9 to –7 mW m<sup>-2</sup> (Sausen et al., 2005; Fuglestedt et al., 2008;  
78 Lee et al., 2009; Balkanski et al., 2010; Unger, 2011; Gettelman and Chen, 2013; Brasseur et al., 2015), nitrate  
79 ranging between –4 to –7 mW m<sup>-2</sup> (Unger et al., 2013; Brasseur et al., 2015), BC ranging between 0.1–0.3 mW  
80 m<sup>-2</sup> (Sausen et al., 2005; Fuglestedt et al., 2008; Lee et al., 2009; Balkanski et al., 2010; Unger, 2011;  
81 Gettelman and Chen, 2013; Unger et al., 2013; Brasseur et al., 2015), and for organic carbon (OC) ranging  
82 between –0.67 to –0.01 mW m<sup>-2</sup> (Sausen et al., 2005; Fuglestedt et al., 2008; Lee et al., 2009; Balkanski et al.,  
83 2010; Unger, 2011; Gettelman and Chen, 2013; Unger et al., 2013). Few studies estimate the aerosol cloud  
84 albedo effect (aCAE) from aviation: Righi et al. (2013) assessed the aCAE to be –15.4±10.6 mW m<sup>-2</sup> while  
85 Gettelman and Chen (2013) estimate –21±11 mW m<sup>-2</sup>.

86  
87 Aviation emissions can increase atmospheric concentrations of fine particulate matter with a dry diameter of  
88 <2.5 µm (PM<sub>2.5</sub>). Short-term exposure to PM<sub>2.5</sub> can exacerbate existing respiratory and cardiovascular ailments,  
89 while long-term exposure can result in chronic respiratory and cardiovascular diseases, lung cancer, chronic  
90 changes in physiological functions and mortality (Pope et al., 2002; World Health Organisation, 2003; Ostro,  
91 2004). In the U.S. aviation emissions are estimated to lead to adverse health effects in ~11,000 people (ranging  
92 from mortality, respiratory ailments and hospital admissions due to exacerbated respiratory conditions) and  
93 ~23,000 work loss days per annum (Ratliff et al., 2009). Landing and take-off aviation emissions increase PM<sub>2.5</sub>  
94 concentrations, particularly around airports (Woody et al., 2011), increasing US mortality rates by ~160 per  
95 annum.

96  
97 Previous studies have estimated the number of premature mortalities due to exposure to pollution resulting  
98 from aviation emissions. Barrett et al. (2012) and Barrett et al. (2010) used the methodology of Ostro (2004)  
99 to estimate that aviation emissions are responsible for ~10,000 premature mortalities a<sup>-1</sup> due increases in  
100 cases of cardiopulmonary disease and lung cancer. Yim et al. (2015) using the same methodology but with the  
101 inclusion of the Rapid Dispersion Code (RDC) to simulate the local air quality impacts of aircraft ground level  
102 emissions estimated 13,920 (95% CI: 7,220–20,880) mortalities a<sup>-1</sup>. Morita et al. (2014) using the integrated  
103 exposure–response (IER) model from Burnett et al. (2014) to derive relative risk (RR) estimate that aviation  
104 results in 405 (95% CI: 182–648) mortalities a<sup>-1</sup> due to increases in cases of lung cancer, stroke, ischemic heart  
105 disease, trachea, bronchus, and chronic obstructive pulmonary disease. Jacobson et al. (2013) estimate 310

106 (95% CI: -400 to 4,300) mortalities a<sup>-1</sup> from aviation emissions due to cardiovascular effects. Taking these  
107 studies in account, the different methodologies applied and modes of mortality investigated aviation is  
108 estimated to be responsible for between 310–13,920 mortalities a<sup>-1</sup>.

109  
110 The introduction of cleaner fuels and pollution control technologies can improve ambient air quality and  
111 reduce adverse health effects of fossil fuel combustion (World Health Organisation, 2005). One proposed  
112 solution to reduce the adverse health effects of aviation-induced PM<sub>2.5</sub> is the use of ultra-low sulfur jet fuel  
113 (ULSJ), reducing the formation of sulfate aerosol (Barrett et al., 2012; Barrett et al., 2010; Ratliff et al., 2009;  
114 Hileman and Stratton, 2014). ULSJ fuels typically have a fuel sulfur content (FSC) of 15 ppm, compared with an  
115 FSC of between 550–750 ppm in standard aviation fuels (Barrett et al., 2012). The current global regulatory  
116 standard for aviation fuel is a maximum FSC of 3000 ppm (Ministry of Defence, 2011; ASTM International,  
117 2012).

118  
119 Despite the potential for decreased emission of SO<sub>2</sub>, application of ULSJ fuel will not completely remove the  
120 impacts of aviation on PM<sub>2.5</sub>. It is estimated that over a half of aviation-attributable surface-level sulfate is  
121 associated with oxidation of non-aviation SO<sub>2</sub> by OH produced from aviation NO<sub>x</sub> emissions, and not directly  
122 produced from aviation-emitted SO<sub>2</sub> (Barrett et al., 2010). Therefore, even a completely desulfurised global  
123 aviation fleet would likely contribute a net source of sulfate PM<sub>2.5</sub>. Nevertheless, previous work has shown  
124 that the use of ULSJ fuel reduces global aviation-induced PM<sub>2.5</sub> by ~23%, annually avoiding ~2300 (95% CI:  
125 890–4200) mortalities (Barrett et al., 2012).

126  
127 Altering the sulfur content of aviation fuel also modifies the net climate impact of aviation emissions. A  
128 reduction in fuel sulfur content reduces the formation of cooling sulfate aerosols (Unger, 2011; Barrett et al.,  
129 2012), increasing the net warming effect of aviation emissions. The roles of sulfate both in climate cooling and  
130 in increasing surface PM<sub>2.5</sub> concentrations mean that policy makers must consider both health and climate  
131 when considering effects from potential reductions in sulfur emissions from a given emissions sector (Fiore et  
132 al., 2012).

133  
134 In this study, we investigate the impacts of changes in the sulfur content of aviation fuel on climate and human  
135 health. A coupled tropospheric chemistry-aerosol microphysics model is used to quantify global atmospheric  
136 responses in aerosol and O<sub>3</sub> to varying FSC scenarios. Radiative effects due to changes in tropospheric O<sub>3</sub> and  
137 aerosols are calculated using a radiative transfer model the impacts of changes in surface PM<sub>2.5</sub> on human  
138 health are estimated using concentration response functions. Using a coupled tropospheric chemistry-aerosol  
139 microphysics model that includes nitrate aerosol allows us to assess the impacts of nitrate and aerosol indirect  
140 effects in addition to the ozone and aerosol direct effects that have been more routinely calculated.

141

## 142 **2 Methods**

143

### 144 **2.1 Coupled chemistry-aerosol microphysics model**

#### 145 **2.1.1 Model description**

146 We use GLOMAP-mode (Mann et al., 2010), embedded within the 3-D off-line Eulerian chemical transport  
147 model TOMCAT (Arnold et al., 2005; Chipperfield, 2006). Meteorology (wind, temperature and humidity) and  
148 large scale transport is specified from interpolation of 6-hourly European Centre for Medium Range Weather  
149 Forecasts (ECMWF) reanalysis (ERA-40) fields (Chipperfield, 2006; Mann et al., 2010). Cloud fraction and cloud  
150 top pressure fields are taken from the International Satellite Cloud Climatology Project (ISCCP-D2) archive for  
151 the year 2000 (Rossow and Schiffer, 1999).

152

153 GLOMAP-mode is a two-moment aerosol microphysics scheme representing particles as an external mixture  
154 of 7 size modes (4 soluble and 3 insoluble) (Mann et al., 2010). We use the nitrate-extended version of  
155 GLOMAP-mode (Benduhn et al., 2016) which, as well as tracking size-resolved sulfate, BC, OC, sea-salt and  
156 dust components, also includes a dissolution solver to accurately characterise the size-resolved partitioning of

157 ammonia and nitric acid into ammonium and nitrate components in each soluble mode. Aerosol components  
158 are assumed to be internally mixed within each mode. GLOMAP-mode includes representations of nucleation,  
159 particle growth via coagulation, condensation and cloud processing, wet and dry deposition, and in- and  
160 below-cloud scavenging (Mann et al., 2010).

161  
162 TOMCAT includes a tropospheric gas-phase chemistry scheme (inclusive of  $O_x$ - $NO_y$ - $HO_x$ ), treating the  
163 degradation of  $C_1$ - $C_3$  non-methane hydrocarbons (NMHCs) and isoprene, together with a sulfur chemistry  
164 scheme (Spracklen et al., 2005; Breider et al., 2010; Mann et al., 2010). The tropospheric chemistry is coupled  
165 to aerosol as described in Breider et al. (2010).

166  
167 The nitrate-extended version of the TOMCAT-GLOMAP-mode coupled model used in this investigation  
168 employs a hybrid solver to simulate the dissolution of semi-volatile inorganic gases (such as  $H_2O$ ,  $HNO_3$ ,  $HCl$   
169 and  $NH_3$ ) into the aerosol-liquid-phase.

170  
171 Emissions of DMS are calculated using monthly mean sea-water concentrations of DMS from (Kettle and  
172 Andreae, 2000), driven by ECMWF winds and sea-air exchange parameterisations from Nightingale et al.  
173 (2000). Emissions of  $SO_2$  are included from both continuous (Andres and Kasgnoc, 1998) and explosive  
174 volcanoes (Halmer et al., 2002), and wildfires for year 2000 (Van Der Werf et al., 2003; Dentener et al., 2006).  
175 Anthropogenic  $SO_2$  emissions (including industrial, power-plant, road-transport, off-road-transport and  
176 shipping sectors) are representative of the year 2000 (Cofala et al., 2005). Emissions of monoterpenes and  
177 isoprene are from Guenther et al. (1995).  $NH_3$  emissions are from the EDGAR inventory (Bouwman et al., 1997).  
178  $NO_x$  emissions are considered from anthropogenic (Lamarque et al., 2010), natural (Lamarque et al., 2005)  
179 and biomass burning (van der Werf et al., 2010) sources.

180  
181 Annual mean emissions of BC and OC aerosol from fossil fuel and biofuel combustion are from Bond et al.  
182 (2004). Monthly wildfire emissions are taken from the GFED v1 (Global Fire Emissions Database) for the year  
183 2000 (Van Der Werf et al., 2003). For primary aerosol emissions we use geometric mean diameters ( $D_g$ ) with  
184 standard deviations as described by Mann et al. (2010).

185  
186 Here, we ran simulations at a horizontal resolution of  $2.8^\circ \times 2.8^\circ$  with 31 hybrid  $\sigma$ -p levels extending from the  
187 surface to 10 hPa. All simulations were conducted for 16 months from September 1999 to December 2000  
188 inclusive, with the first four months discarded as spin-up time.

189

### 190 **2.1.2 Model evaluation**

191 GLOMAP has been extensively evaluated against observations including comparisons of speciated aerosol  
192 mass (Mann et al., 2010; Spracklen et al., 2011b), aerosol number (Mann et al., 2010; Spracklen et al., 2010)  
193 and cloud condensation nuclei (CCN) concentrations (Spracklen et al., 2011a). TOMCAT simulated fields have  
194 been evaluated against observations, with CO and  $O_3$  evaluated against aircraft observations (Arnold et al.,  
195 2005), Mediterranean summertime ozone against satellite observations (Richards et al., 2013), along with  $O_3$   
196 evaluated against satellite observations (Chipperfield et al., 2015). Benduhn et al. (2016) shows that simulated  
197 surface concentrations of  $NO_3$  and  $NH_4$  are in reasonable agreement with observations in Europe, the U.S. and  
198 East Asia. Here we focus our evaluation on the aerosol vertical profile and as well as nitrate aerosol which has  
199 not been evaluated previously.

200

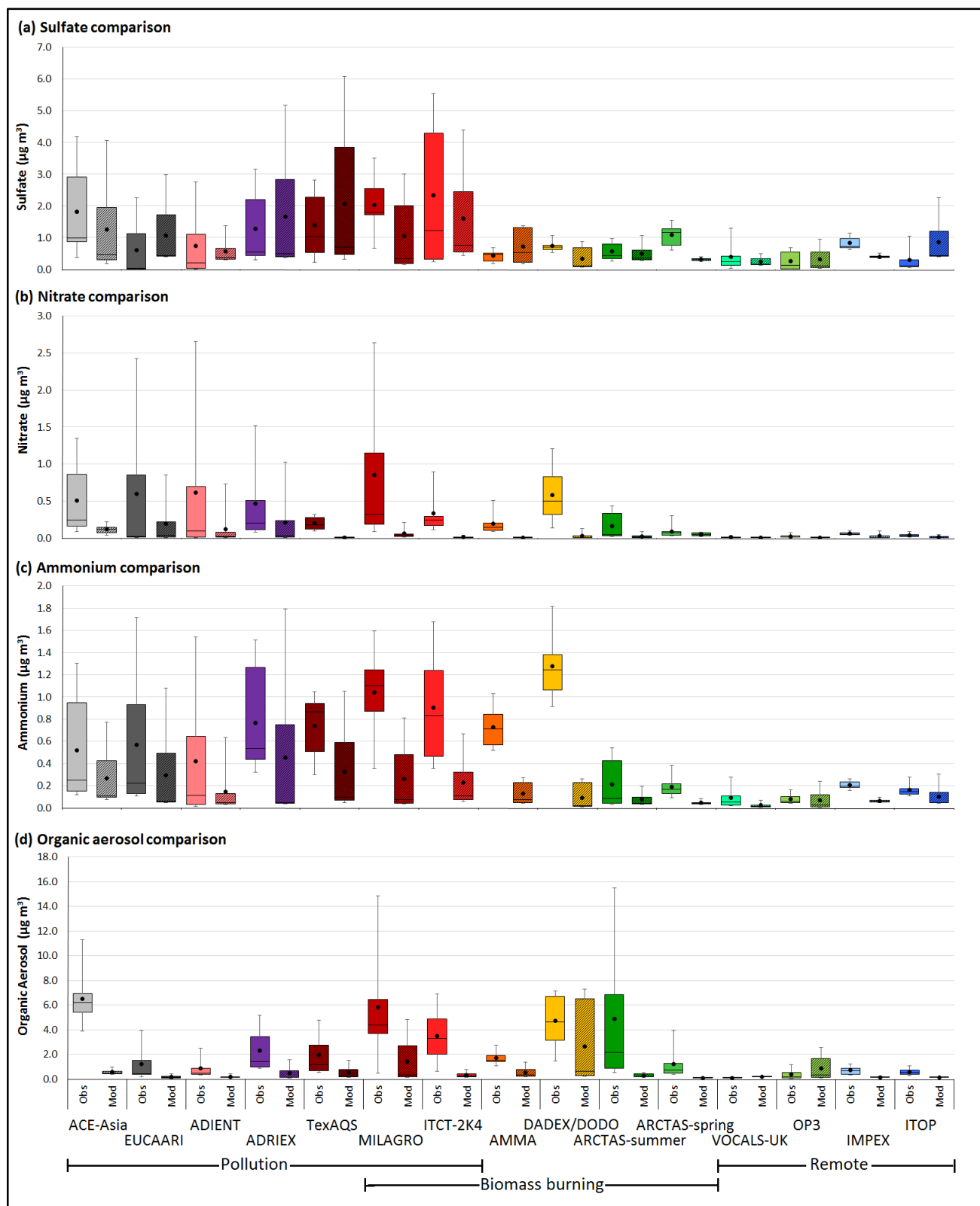
201 Fig. 1 presents simulated sulfate, nitrate, ammonium and organic aerosol mass concentrations in comparison  
202 to airborne observations compiled by Heald et al. (2011). The supplementary information presents the flight  
203 paths of each of the aircraft field campaigns used in the study compiled by Heald et al. (2011) (Figure S1), and  
204 details of each of the aircraft field campaigns used (Table S1). Observations were predominantly made using  
205 an Aerodyne Aerosol Mass Spectrometer (AMS). Simulated profiles are for year 2000, while observational  
206 aerosol profiles are from field campaigns conducted between 2001 and 2008.

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208

209

210 **Fig. 1: Comparison of observed (Obs) and simulated (Mod) (a) sulfate; (b) nitrate; (c) ammonium, and; (d)**  
 211 **organic aerosol mass concentrations. Observations are from airborne field campaigns compiled by Heald et**  
 212 **al. (2011). Mean values are represented by black dots, median values as shown by horizontal lines, while**  
 213 **boxes denote the 25<sup>th</sup> and 75<sup>th</sup> percentiles, and whiskers denote the 5<sup>th</sup> and 95<sup>th</sup> percentile values.**



214 Overall we find the model overestimates sulfates [NMB = +16.9%], while underestimating nitrates [NMB = -  
 215 60.7%], ammonium [NMB = -47.1%] and organic aerosols (OA) [NMB = -56.2%]. Model skill varies dependant  
 216 on the conditions affecting each field campaign. To explore this, we use the broad stratification of the field  
 217 campaigns into anthropogenic pollution, biomass burning and remote conditions as used by Heald et al. (2011)

218 and shown in Fig. 1. The model underestimates aerosol concentrations in biomass burning regions [sulfate  
219 NMB = -14.9%; nitrate NMB = -79.4%; ammonium NMB = -68.7%, and; OA NMB = -74.5%]. The model  
220 performs better in polluted [sulfate NMB = +31.6%; nitrate NMB = -56.2%; ammonium NMB = -28.6%, and;  
221 OA NMB = -40.9%], and remote regions [sulfate NMB = +25.4%; nitrate NMB = -6.4%; ammonium NMB = -  
222 20.2%, and; OA NMB = -41.5%].

223

224 The overestimation of sulfate aerosol is likely due to the decline in anthropogenic SO<sub>2</sub> emissions in Europe and  
225 the US between 2000–2008 (Vestreng et al., 2007; Hand et al., 2012). An underestimation of OA has been  
226 reported previously (Heald et al., 2011; Spracklen et al., 2011b) and is likely due to an underestimate in SOA  
227 formation in the model. Whitburn et al. (2015) found biomass burning emissions of NH<sub>3</sub> may be  
228 underestimated which would affect a number of our comparisons.

229

230 The model underestimation of organic and inorganic aerosol components in biomass burning influenced  
231 regions could partly be due to very concentrated plumes in these regions affecting campaign mean  
232 concentrations. There is a large uncertainty in biomass burning emissions and some evidence that they may  
233 be underestimated (Kaiser et al., 2012), which may contribute to the model bias. Biomass burning emissions  
234 also have large interannual variability (van der Werf et al., 2010; Wiedinmyer et al., 2011), meaning that using  
235 year specific emissions might improve comparison against observations in these regions. Underestimation in  
236 Arctic inorganic aerosol, which will affect the ARCTAS comparisons, is a well-known problem in models,  
237 likely related to problems with model wet deposition and emissions (Shindell et al., 2008; Eckhardt et al.,  
238 2015). The model underestimate over West Africa (AMMA, DADEX and DODO campaigns) is likely due to  
239 a combination of errors in biomass burning emissions and poorly constrained emission sources from  
240 anthropogenic activity (Knippertz et al., 2015).

241

242 Fig. 2 presents simulated ozone concentration profiles in comparison to ozonesonde observations compiled  
243 by Tilmes et al. (2012). Observations were compiled from three networks, comprising of 41 stations with  
244 continuous sampling from 1995 to 2011: (i) The World Ozone and Ultraviolet Data Center (WOUDC)  
245 (<http://www.woudc.org/>); (ii) the Global Monitoring Division (GMD, [ftp:// ftp.cmdl.noaa.gov/ozwv/ozone/](ftp://ftp.cmdl.noaa.gov/ozwv/ozone/)),  
246 and (iii) The Southern Hemisphere ADDitional OZonesondes (SHADOZ) (Tilmes et al., 2012).

247

248 Regional model-observation comparison profiles presented in Fig. 2 demonstrate good agreement between  
249 the model and ozonesonde profiles, while demonstrating regional variations driven by variations in  
250 tropopause height, showing no evidence of systematic model bias in the upper troposphere. Notable  
251 differences are seen between simulated and observed ozone profiles over the Praha launch site in Western  
252 Europe, with the model greatly overestimating observed ozone.

253

254 Evaluation of ozone model bias is conducted for the troposphere, using a chemical tropopause definition of  
255 150 ppbv ozone, as previously used by Stevenson et al. (2013), Young et al. (2013) and Rap et al. (2015). We  
256 find the model overestimates global ozone concentrations [NMB = +7.0%] with overestimates in Western  
257 Europe [+18.9%] and the Northern Hemisphere Polar West [NMB = +14.4%] regions and underestimates over  
258 the Atlantic/Africa [NMB = -11.0%] and Southern Hemisphere Polar [NMB = -4.6%] regions.

259

260 Differences between model and observational profiles can in part be explained by the differences in years of  
261 simulation and observation, a poor representation of deep convection resulting in model underestimations in  
262 the tropics and overestimations downwind (Thompson et al., 1997), in tandem with reductions in  
263 anthropogenic NO<sub>x</sub> emissions over this time period (Konovalov et al., 2008).

264

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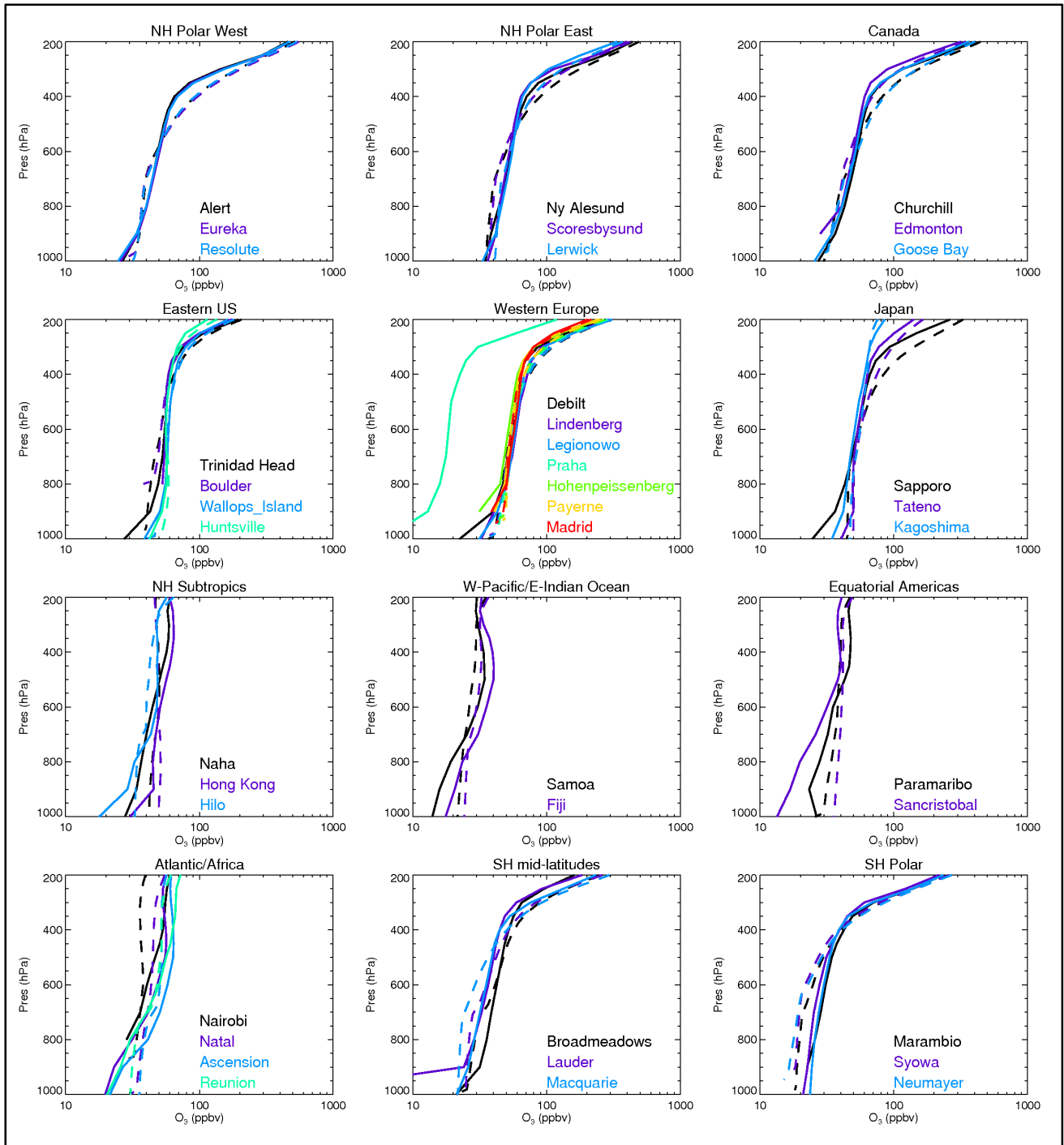
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**Fig. 2: Comparison of observed (solid lines) and simulated (dashed lines) ozone profiles. Observations are taken from ozonesonde observations, and arranged by launch location regions according to Tilmes et al. (2012).**



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## 283 2.2 Aviation emissions

284 Aircraft emit NO<sub>x</sub>, carbon monoxide (CO), SO<sub>2</sub>, BC, OC and hydrocarbons (HCs). The historical emissions dataset  
 285 for the CMIP5 (5<sup>th</sup> Coupled Model Intercomparison Project) model simulations used by the IPCC 5<sup>th</sup> Assessment  
 286 Report only included NO<sub>x</sub> and BC aviation emissions (Lamarque et al., 2009). Recently there have been efforts  
 287 to add HCs, CO and SO<sub>2</sub> emissions to aviation emission inventories (Eyers et al., 2004; Quantify Integrated  
 288 Project, 2005-2012; Wilkerson et al., 2010).

289  
 290 Here we develop a new 3-D civil aviation emissions dataset for the year 2000, based on CMIP5 historical  
 291 aviation emissions (Lamarque et al., 2009). The new dataset includes emissions of NO<sub>x</sub>, CO, SO<sub>2</sub>, BC, OC, and  
 292 HCs. In contrast to existing datasets which provide a general emissions index for HCs (Eyers et al., 2004) we  
 293 speciate HCs as formaldehyde (HCHO), ethane (C<sub>2</sub>H<sub>6</sub>), propane (C<sub>3</sub>H<sub>8</sub>), methanol (CH<sub>3</sub>OH), acetaldehyde  
 294 (CH<sub>3</sub>CHO), and acetone ((CH<sub>3</sub>)<sub>2</sub>CO).

295  
 296 Table 1 describes our new emissions dataset. NO<sub>x</sub> and BC emissions are taken directly from Lamarque et al.  
 297 (2009). We calculate fuelburn from BC emissions data and the BC emissions index (Eyers et al., 2004) as used  
 298 by Lamarque et al. (2009). Following DuBois and Paynter (2006), we assume that BC emissions scale linearly  
 299 with fuel consumption. We estimate emissions for other species using our calculated aviation fuelburn in  
 300 combination with published species-specific emissions indices (EI reported in g kg<sup>-1</sup> of fuel). Emission indices  
 301 for CO and SO<sub>2</sub> are from the FAA's aviation environmental design tool (AEDT) (Wilkerson et al., 2010). OC  
 302 emissions are calculated using a BC:OC ratio of 4 (Hopke, 1985; Bond et al., 2004); resulting in an EI within the  
 303 range determined by Wayson et al. (2009). Speciated hydrocarbon emissions are calculated from experimental  
 304 data following the methodology of Wilkerson et al. (2010) using experimental data from Knighton et al. (2007)  
 305 and Anderson et al. (2006), in conjunction with operating parameters suggested by the Airbus Flight Crew  
 306 Training manual (Airbus, 2008).

307  
 308 **Table 1: Aviation emissions indices and total annual emissions for year 2000**  
 309

Species	Emissions index (g kg <sup>-1</sup> of fuel)	Global emissions for year 2000 (Tg of species)	Range of annual global emissions from previous studies (Tg of species)
NO <sub>x</sub>	13.89 <sup>a</sup>	2.786	1.98–3.286 <sup>a,b,j,h,i,k,l</sup>
CO	3.61 <sup>b</sup>	0.724	0.507–0.679 <sup>b,h,i,j</sup>
HCHO	1.24 <sup>c,d</sup>	0.249	0.01205 <sup>b</sup>
C <sub>2</sub> H <sub>6</sub>	0.0394 <sup>e</sup>	0.007899	0.00051 <sup>b</sup>
C <sub>3</sub> H <sub>8</sub>	0.03 <sup>e</sup>	0.006014	0.00444 <sup>b</sup>
CH <sub>3</sub> OH	0.22 <sup>d</sup>	0.044	0.00177 <sup>b</sup>
CH <sub>3</sub> CHO	0.33 <sup>d</sup>	0.066	0.00418 <sup>b</sup>
(CH <sub>3</sub> ) <sub>2</sub> CO	0.18 <sup>d</sup>	0.036	0.00036 <sup>b</sup>
SO <sub>2</sub>	1.1760 <sup>b</sup>	0.236	0.182–0.221 <sup>a,b,h,i,j</sup>
BC	0.0250 <sup>a</sup>	0.005012	0.0039–0.0068 <sup>a,b,h,i,j,k</sup>
OC	0.00625 <sup>f,g</sup>	0.001253	0.003 <sup>b,i</sup>

<sup>a</sup>(Eyers et al., 2004), <sup>b</sup>(Wilkerson et al., 2010), <sup>c</sup>(Spicer et al., 1994), <sup>d</sup>(Knighton et al., 2007),  
<sup>e</sup>(Anderson et al., 2006), <sup>f</sup>(Bond et al., 2004), <sup>g</sup>(Hopke, 1985), <sup>h</sup>(Olsen et al., 2013), <sup>i</sup>(Unger, 2011),  
<sup>j</sup>(Lee et al., 2010), <sup>k</sup>(Lamarque et al., 2010), <sup>l</sup>(Quantify Integrated Project, 2005-2012)

310  
 311 Our global aviation emissions typically lie within the range of previous studies (Table 1). Our SO<sub>2</sub> emissions are  
 312 greater than those used by Wilkerson et al. (2010) for 2006, despite the use of the same EI. This is due to the  
 313 greater global fuelburn considered by the base inventory used to develop our emissions inventory (Eyers et  
 314 al., 2004; Lamarque et al., 2010). Our estimated OC emissions are lower than the emissions estimated in the  
 315 AEDT 2006 inventory, due to the lower EI applied here. The lower EI<sub>OC</sub> applied here (in comparison to  
 316 Wilkerson et al. (2010)) is a due to the phase of flight considered when deriving the AEDT emissions inventory;  
 317 where they derive EI<sub>OC</sub> focusing on airport operations at ground level condition acknowledging the risk of



318 overestimating aviation OC emissions, while in comparison we consider aircraft operations after ground idle  
319 conditions which risks underestimating aviation OC emissions.

320

321 We calculate the geometric mean diameter ( $D_g$ ) for internally mixed BC/OC particles as 50.5 nm from the mean  
322 particle mass derived using the particle number emissions index (Eyers et al., 2004) and a constant standard  
323 deviation set to  $\sigma = 1.59$  nm.

324

### 325 2.3 Fuel sulfur content simulations

326 To explore the impact of aviation FSC on climate and air quality we performed a series of 11 global model  
327 experiments (Table 2). In 7 of these model experiments FSC values were varied globally between zero and  
328 6000 ppm. Three further simulations varied the vertical distribution of aviation emissions. The first simulation  
329 collapses all aviation emissions to ground level (GROUND), in order to compare an equivalent ground emission  
330 source and its effects. Two simulations (SWITCH1 and SWITCH2), use a low FSC (15 ppm) applied below the  
331 cruise phase of flight (<8.54 km altitude) (Lee et al., 2009; Köhler et al., 2013) combined with a high FSC at  
332 altitudes above. The SWITCH1 scenario increases FSC in line with our HIGH scenario above 8.54 km, while in  
333 the SWITCH2 scenario, emissions are scaled such that total global sulfur emissions are the same as the  
334 standard simulation (NORM), resulting in a FSC of 1420 ppm above 8.54 km. Results from all simulations are  
335 compared against a simulation with aviation emissions excluded (NOAVI).

336

337 **Table 2: FSC and global SO<sub>2</sub> emissions applied in each model experiment.**

Scenario name	Description	FSC (ppm)	Total SO <sub>2</sub> emitted (Tg)
NOAVI	No aviation emissions	n/a	0.0
NORM	Standard aviation emissions scenario	600	0.236
DESUL	Desulfurised case	0	0.0
ULSJ	Ultra low sulfur jet fuel	15	0.006
HALF	Half FSC of normal case	300	0.118
TWICE	Twice FSC of normal case	1200	0.472
HIGH	FSC at international specification limit	3000	1.179
OVER	Twice FSC specification limit	6000	2.358
GROUND	All emissions emitted at surface level (FSC as NORM)	600	0.236
SWITCH1	ULSJ FSC to 8.54 km, HIGH FSC content above	15/3000	0.491
SWITCH2	ULSJ FSC to 8.54 km, FSC = 1420 ppm above	15/1420	0.236

338

### 339 2.4 Radiative impacts

340 We calculate the aerosol direct radiative effect (aDRE), aerosol cloud albedo effect (aCAE) and tropospheric  
341 O<sub>3</sub> direct radiative effect (O3DRE) using the offline Edwards and Slingo (1996) radiative transfer model. The  
342 radiative transfer model considers 6 bands in the shortwave (SW) and 9 bands in the longwave (LW), adopting  
343 a delta-Eddington 2 stream scattering solver at all wavelengths. The top-of-the-atmosphere (TOA) aerosol  
344 aDRE and aCAE are calculated using the methodology described in Rap et al. (2013) and Spracklen et al.  
345 (2011a), with the method for O3DRE as in Richards et al. (2013). To determine the aCAE we calculated cloud  
346 droplet number concentrations (CDNCs) using the monthly mean aerosol size distribution simulated by  
347 GLOMAP combined with parameterisations from Nenes and Seinfeld (2003), updated by Fountoukis and  
348 Nenes (2005) and Barahona et al. (2010). CDNC were calculated with a prescribed updraft velocity of 0.15 m  
349 s<sup>-1</sup> over ocean and 0.3 m s<sup>-1</sup> over land. Changes to CDNC were then used to perturb the effective radii of cloud  
350 droplets in low- and mid-level clouds (up to 600 hPa). The aDRE, aCAE and O3DREs for each aviation emissions  
351 scenario are calculated as the difference in TOA net (SW + LW) radiative flux compared to the NOAVI  
352 simulation.

353

## 354 2.5 Health effects

355 We calculate excess premature mortality from cardiopulmonary diseases and increases in cases of lung cancer  
356 due to long-term exposure to aviation-induced PM<sub>2.5</sub> (Ostro, 2004). Using this function allows us to compare  
357 directly with previous studies (Barrett et al., 2012; Yim et al., 2015); in future work estimates are required with  
358 updated methodologies (Burnett et al., 2014). PM<sub>2.5</sub> is used as a measure of likely health impacts because  
359 chronic exposure is associated with adverse human health impacts including morbidity and mortality (Dockery  
360 et al., 1993; Pope and Dockery, 2006).

361  
362 We relate annual excess mortality to annual mean surface PM<sub>2.5</sub> via a concentration response-function (CRF)  
363 (Ostro, 2004). This response-function considers concentrations of PM<sub>2.5</sub> for a perturbed case (X) (defined by  
364 aviation emissions scenarios from Table 2) in relation to a baseline case with no aviation emissions (X<sub>0</sub>)  
365 (NOAVI). To calculate excess mortality, the relative risk (RR) for both cardiopulmonary disease and lung cancer  
366 are calculated according to Ostro (2004) using a function of baseline (X<sub>0</sub>) and perturbed (X) PM<sub>2.5</sub>  
367 concentrations, and the disease specific cause-specific coefficient (β):

$$370 \text{ RR} = \left[ \frac{(X+1)}{(X_0+1)} \right]^\beta$$

369 (1)

371  
372 β coefficients for cardiopulmonary disease mortality of 0.15515 [95% CI = 0.05624–0.2541] and lung cancer of  
373 0.232 [95% CI = 0.086–0.379] are used (Pope et al., 2002; Ostro, 2004). The 95% confidence interval (CI) in β  
374 allow low-, mid- and high-range mortality values to be calculated. The attribution factor (AF) from the  
375 exposure to air pollution is calculated using equation (2):

$$377 \text{ AF} = (\text{RR} - 1) / \text{RR}$$

378 (2)

379 Excess mortality (E) for both cardiopulmonary disease and lung cancer are calculated using baseline mortality  
380 rates (B), the fraction of the population over 30 years old (P<sub>30</sub>), along with the AF:

$$382 \text{ E} = \text{AF} \times \text{B} \times \text{P}_{30}$$

383 (3)

384 Global population data is taken from the Gridded World Population (GWP; version3) project (Center for  
385 International Earth Science Information Network, 2012) with country specific data on the fraction of the  
386 population under 30.

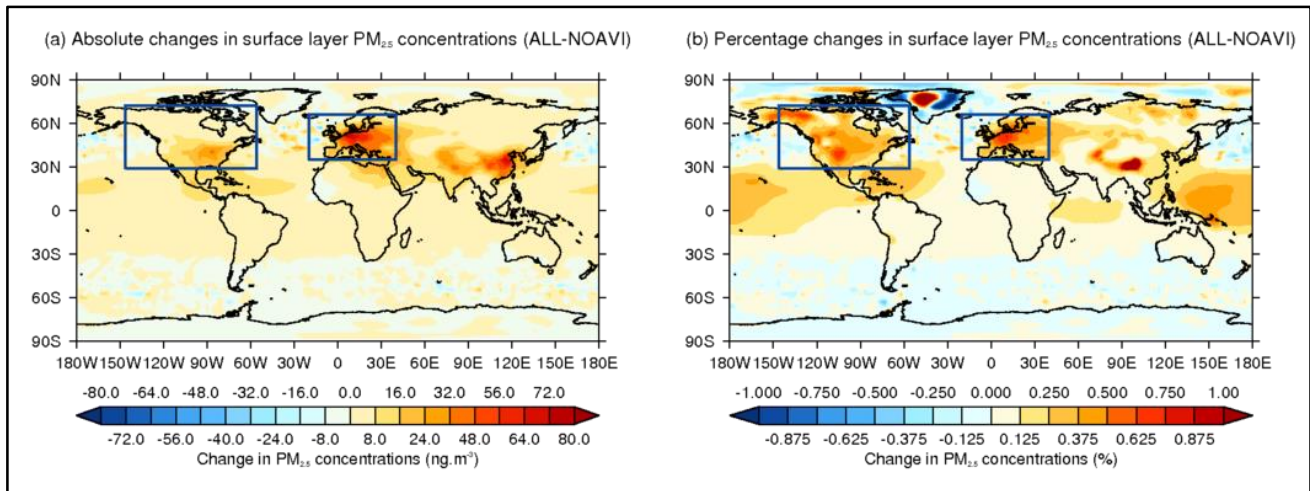
## 388 3 Results

### 390 3.1 Surface PM<sub>2.5</sub>

391 Fig. 3 shows the simulated impact of aviation emissions with standard FSC (FSC = 600 ppm; NORM) on surface  
392 PM<sub>2.5</sub> concentrations. Aviation increases annual mean PM<sub>2.5</sub> concentrations by up to ~80 ng m<sup>-3</sup> (relative to  
393 the NOAVI simulation) over Central Europe and Eastern China (Fig. 3(a)). Aviation emissions result in largest  
394 fractional changes in annual mean PM<sub>2.5</sub> concentrations (up to 0.8%) over North America and Europe (Fig.  
395 3(b)).

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403 **Fig. 3: Impact of aviation emissions (FSC = 600 ppm) on surface annual mean PM<sub>2.5</sub> concentrations. (a)**  
 404 **absolute (NORM–NOAVI) and (b) percentage changes. Boxes show the European (20°–40°E, 35°N–66°N) and**  
 405 **North American (146°W–56°W, 29°N–72°N) regions.**



406 Fig. 4 shows the impact of aviation emissions on global and regional mean PM<sub>2.5</sub> concentrations, as a function  
 407 of FSC. With standard FSC (FSC = 600 ppm), aviation increases global mean surface PM<sub>2.5</sub> concentrations by  
 408 3.9 ng m<sup>-3</sup>; with increases in PM<sub>2.5</sub> dominated by sulfates [56.2%], nitrates [26.0%] and ammonium [16.0%].  
 409 Aviation emissions increase European annual mean PM<sub>2.5</sub> concentrations by 20.3 ng m<sup>-3</sup> (Fig. 4(b)),  
 410 substantially more than over North America (Fig. 4(c)) where an annual mean increase of 6.3 ng m<sup>-3</sup> is  
 411 simulated. Increased PM<sub>2.5</sub> is dominated by nitrates, both over Europe [55.5%] and over North America  
 412 [44.4%]. Sulfates contribute up to 44.6% of increases in PM<sub>2.5</sub> over North America, and 30.0% over Europe.  
 413

414 The use of ULSJ fuel (FSC = 15 ppm) reduces global annual mean surface aviation-induced PM<sub>2.5</sub> concentrations  
 415 (in relation to the NORM case) by 35.7% [1.4 ng m<sup>-3</sup>] (Fig. 4); predominantly due to changes in sulfate [-1.4 ng  
 416 m<sup>-3</sup>; -62.1%] and ammonium [-0.2 ng m<sup>-3</sup>; -37.9%], which are marginally offset by very small increases in  
 417 nitrates [+3.2x10<sup>-3</sup> ng m<sup>-3</sup>; +0.3%]. Aviation emissions also leads to small changes to other aerosol components  
 418 of +0.2 ng; which includes natural aerosols such as dust [+0.3 ng m<sup>-3</sup>; +61.8%], sodium [-19.5%] and chloride  
 419 from sea-salt [-19.5%] with the changes due to changes in aerosol lifetimes, along with changes in BC [-7.9%]  
 420 and OC [-19.3%].  
 421

422 In comparison to the global mean, switching to the use of ULSJ fuel in aviation larger absolute reductions in  
 423 PM<sub>2.5</sub> of -4.2 ng m<sup>-3</sup> are simulated over Europe [ $\Delta$ sulfate = -3.4 ng m<sup>-3</sup>;  $\Delta$ nitrate = +0.1 ng m<sup>-3</sup>;  $\Delta$ ammonium =  
 424 -0.8 ng m<sup>-3</sup>; and  $\Delta$ others = -0.1 ng m<sup>-3</sup>] and of -3.4 ng m<sup>-3</sup> over North America [ $\Delta$ sulfate = -2.9 ng m<sup>-3</sup>;  $\Delta$ nitrate  
 425 = +0.02 ng m<sup>-3</sup>;  $\Delta$ ammonium = -0.5 ng m<sup>-3</sup>; and  $\Delta$ others = -0.01 ng m<sup>-3</sup>] (Fig. 4 (b,c)). Over North America,  
 426 swapping to ULSJ fuel reduces aviation-induced PM<sub>2.5</sub> by 53.4%, while a smaller reduction of 20.5% is simulated  
 427 over Europe. The smaller fractional change in PM<sub>2.5</sub> over Europe is caused by smaller reductions in aviation-  
 428 induced sulfate [-55.9%] and ammonium [-18.4%] compared to over North America, which sees a reduction  
 429 in ammonium of 41.6% and a reduction in sulfates of 103% indicating that over the US the ULSJ fuel scenario  
 430 sees a reduction in sulfates in relation to a NOAVI scenario.  
 431

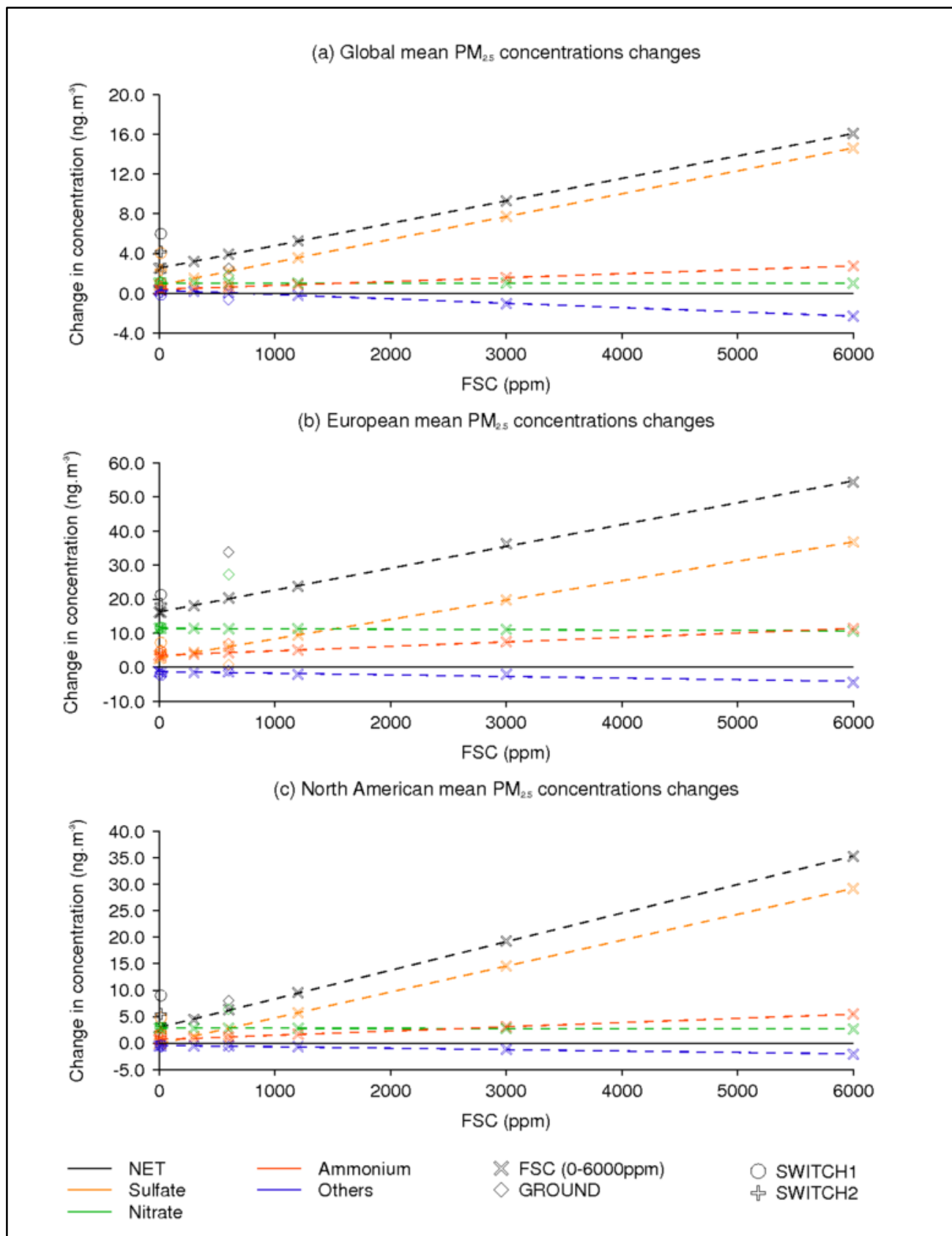
432 Complete desulfurisation of jet fuel (FSC = 0 ppm; DESUL) reduces global mean aviation-induced surface PM<sub>2.5</sub>  
 433 concentrations by 36.5% [-1.43 ng m<sup>-3</sup>], with changes in sulfates [-1.40 ng m<sup>-3</sup>; -63.5%] and ammonium [-0.24  
 434 ng m<sup>-3</sup>; -38.8%] dominating. Under this scenario the reductions in surface sulfate PM<sub>2.5</sub> from aviation are 57.3%  
 435 over Europe and 105% over North America. ULSJ fuel therefore gives similar results to complete  
 436 desulfurisation, due to the very small sulfur emission from ULSJ fuel (Table 2).  
 437

438 In summary, increases in FSC result in increased surface PM<sub>2.5</sub>, due to increased sulfate outweighing the small  
 439 reductions in nitrate. Simulated changes in sulfate, nitrate, ammonium and total PM<sub>2.5</sub> are linear ( $R^2 > 0.99$ ,  $p$ -  
 440 value < 0.001 globally and for all individual regions) with respect to FSC (Fig. 4). Larger emission perturbations  
 441 would likely lead to a non-linear response in atmospheric aerosol. The impact of variations in FSC on PM<sub>2.5</sub> are

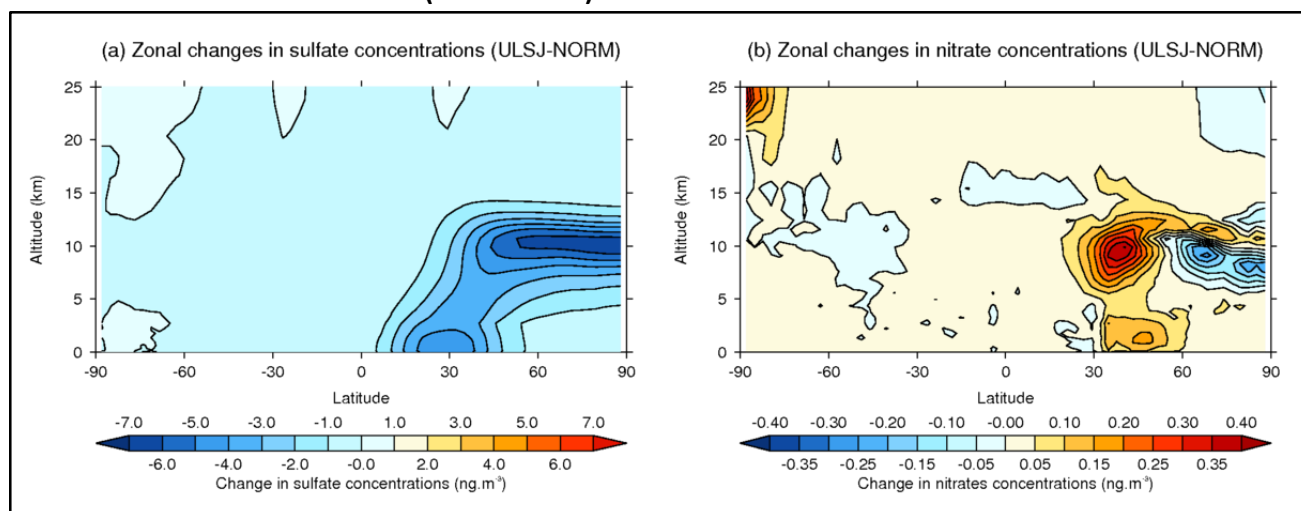
442 regionally variable; over Europe changes in PM<sub>2.5</sub> concentrations are observed to be more sensitive to changes  
 443 in FSC than over North America, and the global domain.

444

445 **Fig. 4: Impact of aviation FSC on (a) global, (b) European (20°–40°E, 35°N–66°N), (c) North American (146°W–**  
 446 **56°W, 29°N–72°N) surface annual mean PM<sub>2.5</sub> mass concentrations: FSC variations (×), GROUND (◇),**  
 447 **SWITCH1 (–), and SWITCH2 (+) simulations. Solid lines demonstrate the linear relationship between FSC**  
 448 **and PM<sub>2.5</sub>.**



450 **Fig. 5: Simulated differences in zonal annual mean sulfate (a) and nitrate (b) concentrations from the use of**  
 451 **ULSJ fuel relative to standard fuel (ULSJ–NORM).**



452

453 Fig. 5 shows the impact of changing to ULSJ fuel on zonal mean sulfate and nitrate concentrations relative to  
 454 standard fuel (NORM). Table 3 reports the global aerosol burden from aviation under different emission  
 455 scenarios. With standard FSC (FSC = 600 ppm), the global aviation-induced aerosol burden is 16.9 Gg,  
 456 dominated by sulfates (76.3%) and nitrates (33.4%). The use of ULSJ (FSC = 15 ppm) reduces the global aerosol  
 457 burden from aviation by 26.8%. Complete desulfurisation of aviation fuel reduces the global aerosol burden  
 458 from aviation by 28.4%, with the global sulfate burden from aviation reduced by 71.6% (Table 3). When  
 459 aviation emissions contain no sulfur, aviation-induced sulfate is formed through aviation NO<sub>x</sub>-induced  
 460 increases in OH concentrations, resulting in the oxidation of SO<sub>2</sub> from non-aviation sources (Unger et al., 2006;  
 461 Barrett et al., 2010).

462

463 **Table 3: Global aviation-induced aerosol mass burdens for different emission scenarios. Values in**  
 464 **parentheses show percentage change relative to NORM case.**

Scenario	All components (Gg)	Sulfates (Gg)	Nitrates (Gg)
NORM	16.9	12.9	5.7
ULSJ	12.4 (-26.8%)	4.0 (-69.1%)	5.9 (+4.5%)
DESUL	12.1 (-28.4%)	3.7 (-71.6%)	6.0 (+5.1%)
No NO <sub>x</sub> and SO <sub>2</sub>	2.0 (-88.3%)	0.3 (-97.5%)	0.1 (-97.9%)

465

466 In line with previous work, we find a substantial fraction of aviation sulfate can be attributed to aviation NO<sub>x</sub>  
 467 emissions and not directly to aviation SO<sub>2</sub> emissions. We estimate that 36% aviation-attributable sulfates  
 468 formed at the surface are associated with aviation NO<sub>x</sub> emissions, compared to ~63% estimated by Barrett et  
 469 al. (2010) using the GEOS-Chem model (both estimates for FSC = 600 ppm). Differences between model  
 470 estimates can be attributed to differences in model chemistry and microphysics, and different aviation NO<sub>x</sub>  
 471 emissions. We find desulfurisation increases the aviation nitrate burden by 5.1% (Table 3); although much of  
 472 this increase occurs at altitudes well above the surface (Fig. 5) and so is not reflected in surface PM<sub>2.5</sub>  
 473 concentrations.

474

475 We explored the impacts of NO<sub>x</sub> emission reductions in combination with fuel desulfurisation. A scenario with  
 476 desulfurised fuel and zero NO<sub>x</sub> emissions reduces the global aviation-induced aerosol burden by 88.3% (Table  
 477 3), in comparison to a desulfurised only case (DESUL), where the aviation-induced aerosol burden is reduced  
 478 by 28.4%. Removal of aviation NO<sub>x</sub> and SO<sub>2</sub> emissions results in a 95.0% reduction in aviation-induced global  
 479 mean surface level aviation-induced PM<sub>2.5</sub>. These results imply that only limited sulfate reductions can be

480 achieved through reducing FSC alone, with further reductions in aviation-induced PM<sub>2.5</sub> sulfates requiring  
481 additional controls on aviation NO<sub>x</sub> emissions.  
482

### 483 **3.2 Premature mortality**

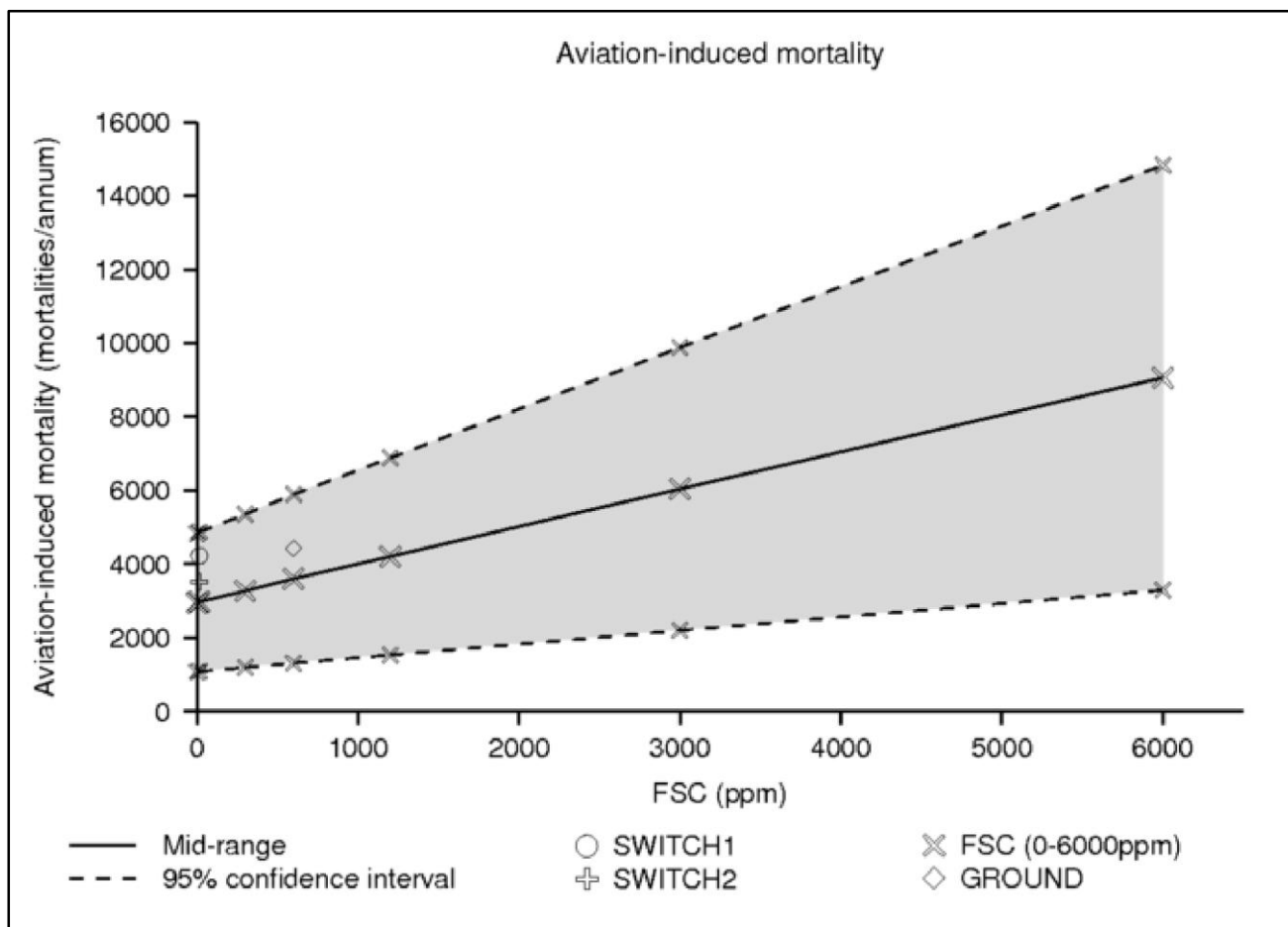
484 Fig. 6 shows estimated annual premature mortalities (from cardiopulmonary disease and lung cancer) due to  
485 aviation-induced changes in PM<sub>2.5</sub> as a function of FSC. We estimate that aviation emissions with standard FSC  
486 (FSC = 600 ppm) cause 3,600 [95% CI: 1,310–5,890] premature mortalities each year, with 3210 [95% CI: 1160–  
487 5250] mortalities a<sup>-1</sup> due to increases in cases of cardiopulmonary disease and 390 [95% CI: 150–640]  
488 mortalities a<sup>-1</sup> due to increases in cases of lung cancer. Low-, mid- and high-range cause-specific coefficients  
489 ( $\beta$ ) are used to account for uncertainty in the health impacts caused by exposure to PM<sub>2.5</sub> (Section 2.5) (Ostro,  
490 2004). Our estimated global mortality due to aviation emissions is greatest in the Northern Hemisphere, which  
491 accounts for 98.7% of global mortalities. Europe and North America account for 42.3% and 8.4% of mortality  
492 due to aviation emissions respectively.  
493

494 Our estimate of the premature mortality due to aviation lies within the range of previous estimates (310–  
495 13,920 mortalities a<sup>-1</sup>) (Barrett et al., 2010; Barrett et al., 2012; Jacobson et al., 2013; Morita et al., 2014; Yim  
496 et al., 2004). Barrett et al. (2012) estimated ~10,000 mortalities a<sup>-1</sup> due to aviation, almost a factor 3 higher  
497 than our central estimate. The greater aviation-induced mortality simulated by Barrett et al. (2012), can be  
498 attributed to greater aviation-induced surface PM<sub>2.5</sub> concentrations simulated in their study, particularly over  
499 highly populated areas. Their study simulated maximum aviation-induced PM<sub>2.5</sub> concentrations over Europe,  
500 eastern China and eastern North America greater than those in our simulations by factors of 5 for Europe and  
501 eastern China and 2.5 over eastern North America. Our aviation-induced sulfate concentrations compare well  
502 with Barrett et al. (2012), indicating that the resulting differences in aviation-induced surface PM<sub>2.5</sub>  
503 concentrations are a result of other aerosol components. Additionally, differences in mortality arise due to the  
504 use of different cause-specific coefficients ( $\beta$ ) within the same CRF, as well as different population datasets.  
505 Morita et al. (2014) estimate that aviation is responsible for 405 [95% CI: 182–648] mortalities a<sup>-1</sup>. This lower  
506 estimate is primarily due to the mortality functions used, with Morita et al. (2014) using the integrated  
507 exposure response (IER) function as described by Burnett et al. (2014). The IER function considers a PM<sub>2.5</sub>  
508 concentration below which there is no perceived risk, reducing estimated impacts of aviation in regions of low  
509 PM<sub>2.5</sub> concentrations.  
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**Fig. 6: Estimated global aviation-induced mortality as a function of FSC, and changes in vertical aviation-emissions distributions for year 2000 (Shaded region denotes the 95% confidence through application of low- and high-range cause-specific coefficients).**



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We estimate that aviation emissions with ULSJ fuel result in 2,970 [95% CI: 1,080–4,870] premature mortalities globally per annum. Therefore, changing from standard FSC to ULSJ would result in 620 [95% CI: 230–1,020] fewer premature mortalities globally per annum; a reduction in aviation-induced mortalities of 17.4%. Regionally we find the implementation of an ULSJ fuel reduces annual mortality by 180 over Europe and by 110 over North America.

Barrett et al. (2012) estimated that swapping to ULSJ fuel could result in ~2,300 [95% CI: 890–4,200] fewer premature mortalities globally per annum; a reduction of 23%. In their work (using GEOS-Chem), the use of ULSJ reduces global mean PM<sub>2.5</sub> concentrations (sulfates, nitrates and ammonium) by 0.89 ng m<sup>-3</sup>, less than the 1.61 ng m<sup>-3</sup> reduction in PM<sub>2.5</sub> simulated here). Despite the greater reductions in global mean surface layer PM<sub>2.5</sub> concentrations simulated here, Barrett et al. (2012) simulate greater reductions in PM<sub>2.5</sub> over populated regions, resulting in greater reductions of aviation-induced mortality under the ULSJ scenario. Additionally, the GRUMPv1 population dataset that Barrett et al. (2012) use resolves population data on a finer scale compared to the resolution of GPWv3 population dataset used here (Center for International Earth Science Information Network, 2012); differences which could contribute to differences in estimates of mortality.

We also estimate how aviation-induced mortality would change if FSC was increased. We find that increasing FSC to 3000 ppm (HIGH) would increase annual aviation-induced mortalities to 6,030, an increase of 67.8% in relation to standard aviation (NORM; FSC = 600 ppm).

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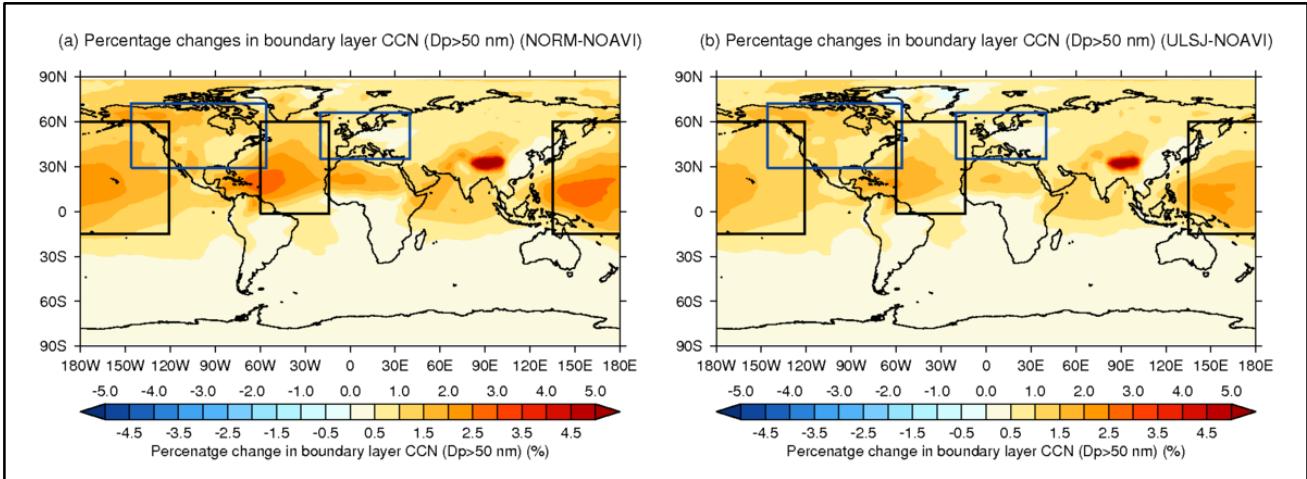
### 3.3 Sensitivity of cloud condensation nuclei to aviation FSC

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Aviation emissions with standard FSC (NORM; FSC = 600 ppm) increase global annual mean cloud condensation nuclei (CCN), here taken as the number of soluble particles with a dry diameter greater than 50 nm, at low-

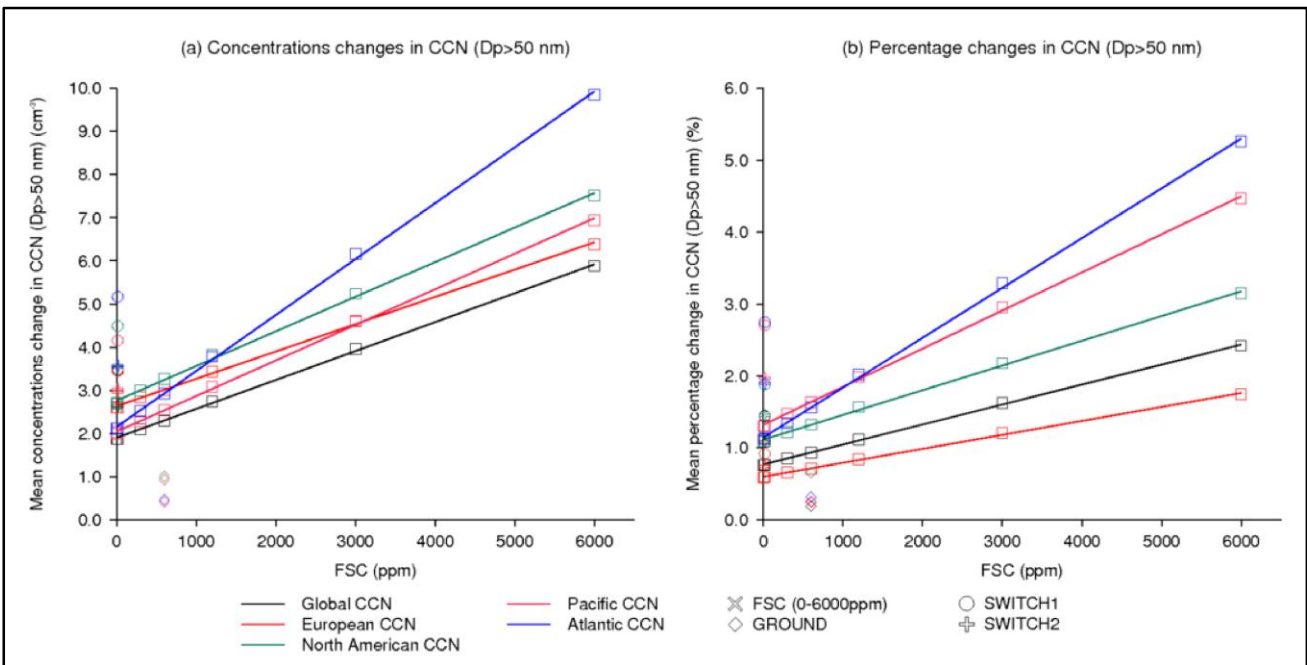
559 cloud level (879hPa; 0.96km) by 0.9% ( $2.3 \text{ cm}^{-3}$ ) (Fig. 7(a)). Increases in CCN concentrations are greater in the  
 560 Northern Hemisphere [ $+3.9 \text{ cm}^{-3}$ ;  $+1.4\%$ ] compared to the Southern Hemisphere [ $+0.7 \text{ cm}^{-3}$ ;  $+0.5\%$ ]. Maximum  
 561 increases in low-level CCN are simulated over the Pacific, central Atlantic and Arctic Oceans.

562  
 563 **Fig. 7: Impact of aviation emissions on low-cloud level (879 hPa) CCN ( $D_p > 50 \text{ nm}$ ) concentrations: (a)**  
 564 **standard FSC (NORM–NOAVI) and (b) FSC = 15 ppm (ULSJ–NOAVI). Blue boxes define North American and**  
 565 **European regions, and black boxes define Atlantic ( $60^\circ\text{W}$ – $14^\circ\text{W}$ ,  $1.4^\circ\text{S}$ – $60^\circ\text{N}$ ) and Pacific regions ( $135^\circ\text{E}$ –**  
 566  **$121^\circ\text{W}$ ,  $15^\circ\text{S}$ – $60^\circ\text{N}$ ) referred to in the text.**



567 The use of ULSJ (FSC = 15 ppm) reduces global mean low-level CCN concentrations by  $0.4 \text{ cm}^{-3}$ , [ $-18.2\%$ ]  
 568 relative to the NORM case (Fig. 7). Northern Hemisphere CCN concentrations are reduced by  $0.8 \text{ cm}^{-3}$  [ $-19.4\%$ ],  
 569 while Southern Hemisphere concentrations are reduced by  $0.1 \text{ cm}^{-3}$  [ $-11.5\%$ ] (Fig. 7).

570  
 571 **Fig. 8: Global and regional variations in low-cloud level (879 hPa) CCN ( $D_p > 50 \text{ nm}$ ): (a) changes in mean**  
 572 **concentrations and (b) percentage changes. See Fig. 5 for definitions of regions.**



573 Fig. 8 shows the sensitivity of low level CCN concentrations to FSC. As with  $\text{PM}_{2.5}$ , we find simulated changes  
 574 in CCN are near linear with respect to FSC ( $R^2 > 0.99$  and  $p$ -value  $< 0.001$  globally and for all individual regions).  
 575

576 ULSJ fuel reduces global mean CCN by  $-0.42 \text{ cm}^{-3}$  with largest reductions over the Atlantic Ocean [ $-0.81 \text{ cm}^{-3}$ ],  
 577 North America [ $-0.55 \text{ cm}^{-3}$ ], and the Pacific Ocean [ $-0.51 \text{ cm}^{-3}$ ], i.e. in relation to standard aviation (ULSJ–  
 578 NORM). The complete desulfurisation of aviation fuel results in reductions in CCN in relation to standard  
 579 aviation (DESUL–NORM), which follow the same regional trends (Fig. 8(a)).



580

### 581 3.4 Sensitivity of aerosol and ozone radiative effect to FSC

582

583 Fig. 9 shows the calculated global mean net RE due to non-CO<sub>2</sub> aviation emissions. For standard FSC (FSC = 600  
584 ppm) emissions the global mean combined RE is  $-13.3 \text{ mW m}^{-2}$ . This combined radiative effect ( $\text{RE}_{\text{comb}}$ ) results  
585 from a balance between a positive aDRE of  $+1.4 \text{ mW m}^{-2}$  and O3DRE  $+8.9 \text{ mW m}^{-2}$ , and a negative aCAE of  $-$   
586  $23.6 \text{ mW m}^{-2}$  (Fig. 9).

587

588 Our estimated aviation aerosol DRE [ $+1.4 \text{ mW m}^{-2}$ ] lies in the middle of the range given by previous work. The  
589 aviation aerosol DRE has been previously assessed as highly uncertain, ranging between  $-28$  to  $+20 \text{ mW m}^{-2}$   
590 (Righi et al., 2013). Our estimated aviation-induced aCAE [ $-23.6 \text{ mW m}^{-2}$ ] lies within the range of uncertainty  
591 from previous literature: Righi et al. (2013) estimated  $-15.4 \pm 10.6 \text{ mW m}^{-2}$  and Gettelman and Chen (2013)  
592 estimated  $-21 \pm 11 \text{ mW m}^{-2}$ .

593

594 Our O3DRE estimate ( $+8.9 \text{ mW m}^{-2}$ ), normalised by global aviation NO<sub>x</sub> emission to  $+10.5 \text{ mW m}^{-2} \text{ Tg(N)}^{-1}$ , is at  
595 the lower end of current estimates [ $7.4$ – $37.0 \text{ mW m}^{-2} \text{ Tg(N)}^{-1}$ ] (Sausen et al., 2005; Köhler et al., 2008; Hoor et  
596 al., 2009; Lee et al., 2009; Holmes et al., 2011; Myhre et al., 2011; Unger, 2011; Frömming et al., 2012; Skowron  
597 et al., 2013; Unger et al., 2013; Khodayari et al., 2014). This can be attributed to the lower net O<sub>3</sub> chemical  
598 production efficiency (OPE) within our model (1.33). Unger (2011) estimated an O3DRE of  $7.4 \text{ mW m}^{-2} \text{ Tg(N)}^{-1}$   
599 with a model OPE of  $\sim 1$ , while the ensemble of models considered by Myhre et al. (2011) have an OPE range  
600 of  $1.5$ – $2.4$ , resulting in an O3DRE range of  $16.2$ – $25.4 \text{ mW m}^{-2} \text{ Tg(N)}^{-1}$ .

601

602 We calculate that an aviation fleet utilising ULSJ fuel would result in a in a global annual mean  $\text{RE}_{\text{comb}}$  of  $-6.3$   
603  $\text{mW m}^{-2}$  [aDRE =  $+1.8 \text{ mW m}^{-2}$ ; aCAE =  $-16.8 \text{ mW m}^{-2}$ ; and O3DRE =  $+8.7 \text{ mW m}^{-2}$ ]. Thus, swapping from  
604 standard aviation fuel to ULSJ fuel reduces the net cooling effect from aviation-induced aerosol and O<sub>3</sub> by  $7.0$   
605  $\text{mW m}^{-2}$ , in comparison to the reduction of  $3.3 \text{ mW m}^{-2}$  estimated by Barrett et al. (2012). In our model, this  
606 change is primarily due a reduction in cooling from the aCAE of  $+6.7 \text{ mW m}^{-2}$  combined with smaller  
607 contributions from an increased aDRE of  $+0.4 \text{ mW m}^{-2}$ , and reduction in warming from the O3DRE of  $-0.12$   
608  $\text{mW m}^{-2}$  (Fig. 9).

609

610 When we assume fully desulfurised aviation jet fuel (DESUL; FSC = 0 ppm), the  $\text{RE}_{\text{comb}}$  induced by aviation-  
611 induced aerosol and O<sub>3</sub> is very similar to that for ULSJ fuel and is estimated as  $-6.1 \text{ mW m}^{-2}$  [aDRE =  $+1.8 \text{ mW}$   
612  $\text{m}^{-2}$ ; aCAE =  $-16.6 \text{ mW m}^{-2}$ ; and O3DRE =  $+8.7 \text{ mW m}^{-2}$ ].

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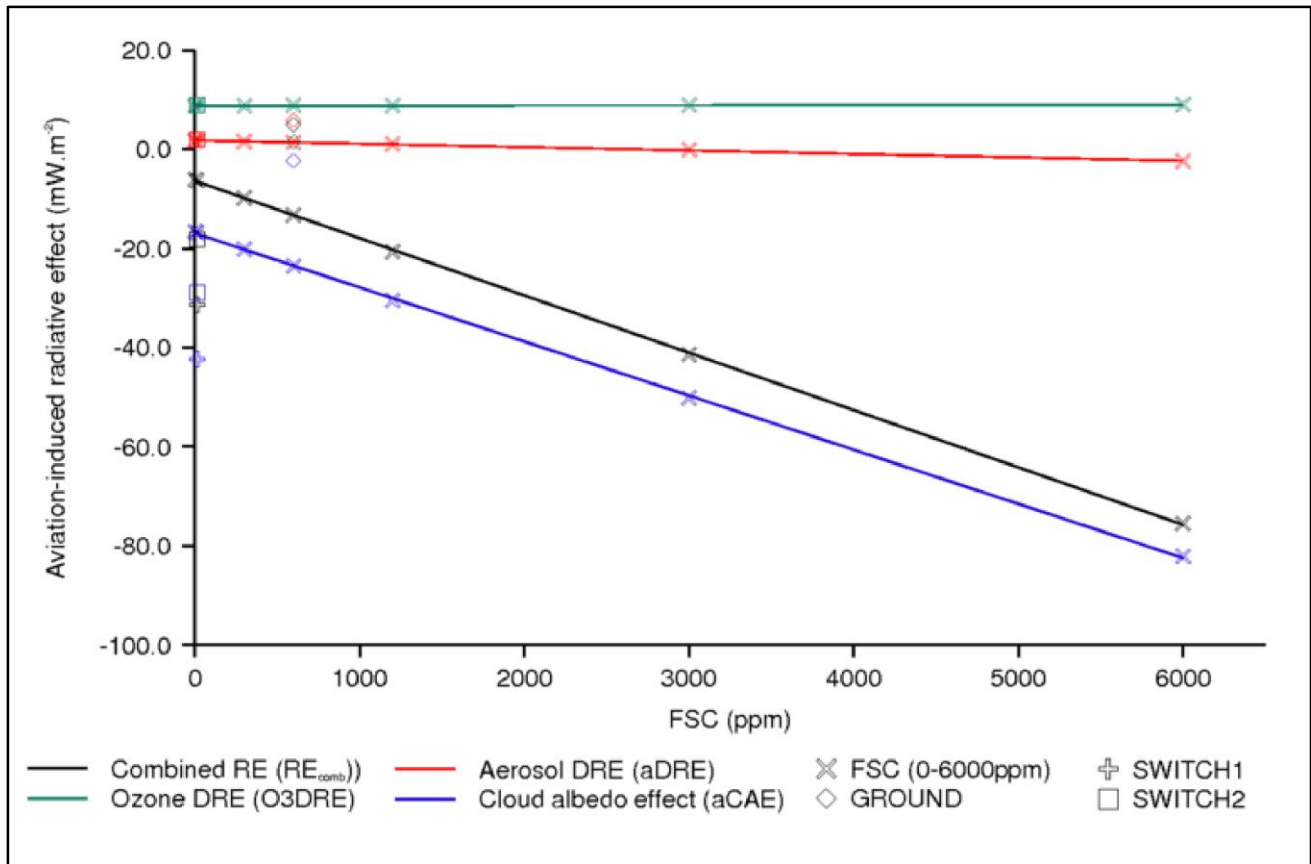
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**Fig. 9: Aviation-induced radiative effects due to variations in fuel sulfur content (FSC), the ground release of aviation emissions (GROUND), and variations in the vertical distribution of aviation SO<sub>2</sub> emissions (SWITCH1 and SWITCH2 simulations).**



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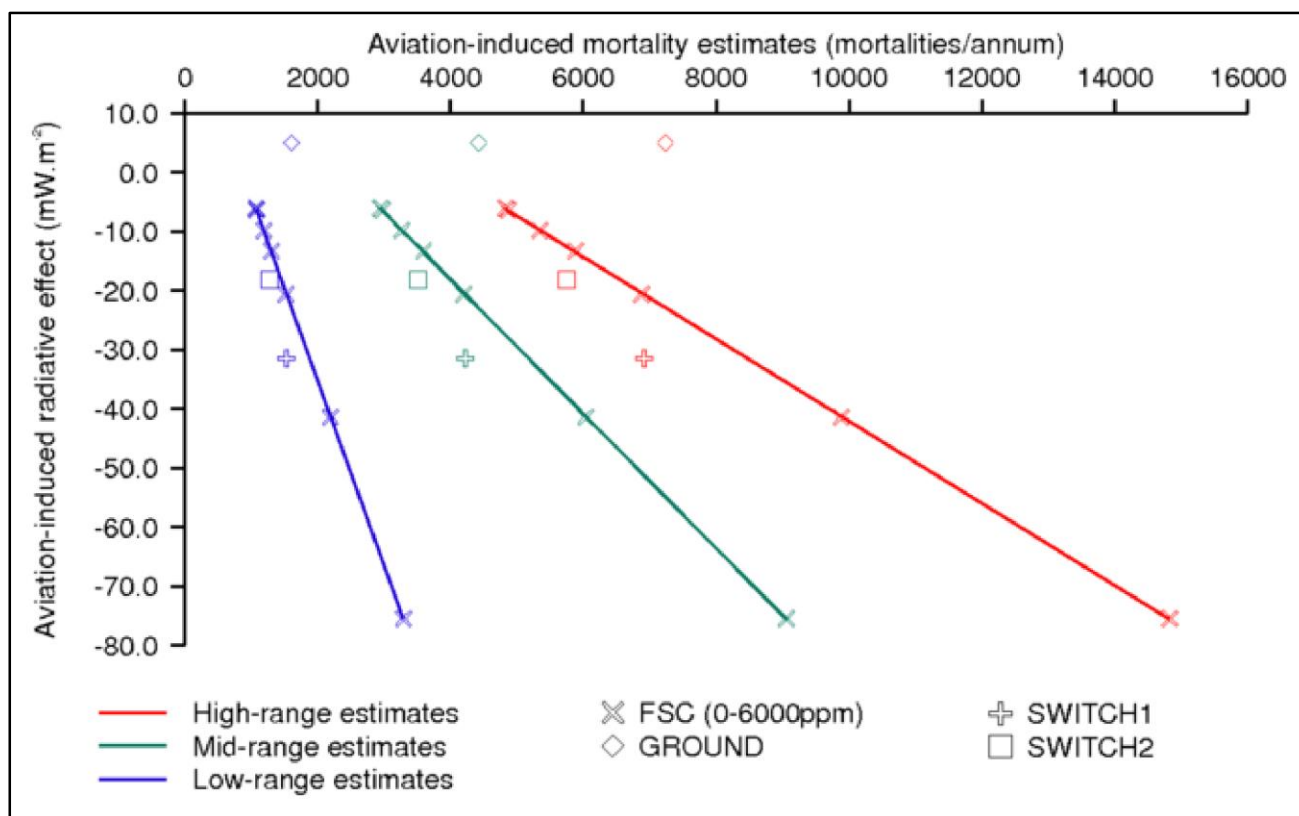
Increases in FSC result in reductions in the aerosol DRE (aDRE), changing from a positive aerosol DRE for low FSC scenarios, to a negative aerosol DRE for high FSC (FSC > 1200 ppm). As FSC is increased, we find the aCAE exhibits a larger cooling effect, i.e. becoming more negative with increases in FSC, increasing by a factor ~5 as FSC is increased from 0 to 6000 ppm. The RE<sub>comb</sub> is dominated by these changes to the aCAE. As a result increases in FSC from 0–6000 ppm, result in a greater negative (cooling) aviation-induced RE<sub>comb</sub>; increasing in magnitude by a factor of ~5 (–16.6 mW m<sup>-2</sup> for FSC = 0 ppm to –82.1 mW m<sup>-2</sup> for FSC = 6000 ppm) (Fig. 9). Therefore, we find that increases in FSC provide a cooling effect due to the dominating effect from aviation-induced aCAE.

### 3.5 Relationship between aviation-induced radiative effects and mortality due to aviation non-CO<sub>2</sub> emissions

Fig. 10 shows the net RE and premature mortality for different aviation emission scenarios. Increases in FSC lead to approximately linear increases in both estimated mortality and the negative net RE. We quantify the impact of FSC on mortality and REs in terms of  $d(\text{mortalities})/d(\text{FSC})$  [mortalities ppm<sup>-1</sup>] and  $d(\text{RE})/d(\text{FSC})$  [mW m<sup>-2</sup> ppm<sup>-1</sup>]. We calculate the sensitivity of global premature mortality to be 1.0 mortalities ppm<sup>-1</sup> [95% CI = 0.4 to 1.6 mortalities ppm<sup>-1</sup>, where the range is due to uncertainty in  $\beta$ ]. The global mean RE<sub>comb</sub> has a sensitivity of  $-1.2 \times 10^{-2}$  mW m<sup>-2</sup> ppm<sup>-1</sup>, dominated by large changes to the aCAE [ $-1.1 \times 10^{-2}$  mW m<sup>-2</sup> ppm<sup>-1</sup>], and much smaller changes in the aDRE [ $-6.9 \times 10^{-4}$  mW m<sup>-2</sup> ppm<sup>-1</sup>] and O<sub>3</sub> RE [ $+4.4 \times 10^{-5}$  mW m<sup>-2</sup> ppm<sup>-1</sup>].

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661 **Fig. 10: Relationship between net radiative effect [sum of ozone direct (O3DRE), aerosol direct radiative**  
 662 **(aDRE) and aerosol cloud albedo (aCAE) effects] and annual mortality rates: for low- mid- and high-range**  
 663 **mortality sensitivities.**



664 The different slopes in the relationship between estimated RE and mortality (Fig. 10) are driven by the range  
 665 of coefficients used in the CRF. This highlights the considerable uncertainty in the health impacts caused by  
 666 exposure to PM<sub>2.5</sub>. We note that uncertainty in the RE due to aerosol and ozone exists, but is not included in  
 667 Fig. 9.

668  
 669 To assess how the vertical distributions of aviation SO<sub>2</sub> emissions influence human health and climate effects,  
 670 we performed three additional simulations where we altered the vertical distribution of aviation SO<sub>2</sub> emissions  
 671 (GROUND, SWITCH1 and SWITCH2 simulations). In these simulations the relationships between mortality and  
 672 net RE deviate from the linear relationship seen when varying FSC between 0–6000 ppm (Fig. 10).

673  
 674 In relation to the standard aviation emissions simulation (FSC = 600 ppm; NORM), when we release all aviation  
 675 emissions at the surface (GROUND; FSC = 600 ppm) aviation-induced surface PM<sub>2.5</sub> concentrations increase by  
 676 +13.5 ng m<sup>-3</sup> [+65.7%] over Europe and by +1.7 ng m<sup>-3</sup> [+27.1%] over North America, but decrease by -1.4 ng  
 677 m<sup>-3</sup> [-36.7%] globally (Fig. 4). Greater surface layer PM<sub>2.5</sub> perturbations (GROUND–NORM) over populated  
 678 regions increase aviation-induced annual mortality by +22.9% [+830 mortalities a<sup>-1</sup>] (Fig. 6).

679  
 680 Releasing aviation emissions at the surface (GROUND case) increases global mean cloud level CCN by only 0.4  
 681 cm<sup>-3</sup> relative to NOAVI; providing a reduction in CCN of 82.1% [-1.89 cm<sup>-3</sup>] relative to the NORM case (i.e.  
 682 GROUND–NORM). That is, injecting aviation emissions into the free troposphere in the standard scenario is  
 683 over 5 times more efficient at increasing CCN concentrations compared to when the same emissions are  
 684 released at the surface [GROUND CCN = 0.4 cm<sup>-3</sup>; NORM CCN = 2.3 cm<sup>-3</sup>]; both in relation to the NOAVI  
 685 scenario. Similar behaviour has been demonstrated previously for volcanic SO<sub>2</sub> emissions by Schmidt et al.  
 686 (2012), where volcanic SO<sub>2</sub> emissions injected into the free troposphere (FT) were more than twice as effective  
 687 at producing new CCN compared to boundary layer emissions of DMS. Injection of aviation SO<sub>2</sub> emissions at  
 688 the surface will increase both deposition rates and aqueous phase oxidation of SO<sub>2</sub>; the latter resulting in the  
 689 growth of existing CCN, but not the formation of new CCN. In contrast, when SO<sub>2</sub> is emitted into the FT the  
 690 dominant oxidation mechanism is to H<sub>2</sub>SO<sub>4</sub>, leading to the formation of new CCN through particle formation

691 and the condensational growth of particles to larger sizes. Subsequent entrainment of these new particles into  
692 the lower atmosphere results in enhanced CCN concentrations in low level clouds. Reduced CCN formation  
693 when aviation emissions are injected at the surface has implications for the aCAE. When aviation emissions  
694 are released at the surface we calculate an aCAE of  $-2.3 \text{ mW m}^{-2}$ ; a factor of 10 smaller than the standard  
695 aviation scenario. This demonstrates that low-level CCN concentrations and the aCAE are particularly sensitive  
696 to aviation emissions, because of the efficient formation of CCN when  $\text{SO}_2$  emissions are injected into the FT.  
697 Injecting aviation emissions at the surface also results in an increase in the aDRE of  $+5.9 \text{ mW m}^{-2}$ , resulting in  
698 a  $\text{RE}_{\text{comb}}$  of  $+5.0 \text{ mW m}^{-2}$  (Fig. 9).

700 Surface  $\text{O}_3$  concentrations are also less sensitive to aviation when emissions are located at the surface. Global  
701 mean aviation-induced surface  $\text{O}_3$  concentrations are reduced from 0.15 ppbv (NORM) to 0.03 ppbv when all  
702 emissions are in the surface layer. Releasing aviation emissions at the surface also reduces the global  $\text{O}_3$   
703 burden by 3.1 Tg. These perturbations in  $\text{O}_3$  concentrations result in a reduction in the  $\text{O}_3$  radiative effect from  
704  $+8.9 \text{ mW m}^{-2}$  (NORM; FSC = 600 ppm) to  $+1.5 \text{ mW m}^{-2}$  (GROUND; FSC = 600 ppm) (Fig. 9). This is a reflection  
705 of increases in the OPE of  $\text{NO}_x$  with increases in altitude due to lower background  $\text{NO}_x$  and NMHC (non-  
706 methane hydrocarbon) concentrations (Köhler et al., 2008; Stevenson and Derwent, 2009; Snijders and  
707 Melkers, 2011; Skowron et al., 2013).

709 We investigated altering FSC between the take-off / landing and the cruise phases of flight using two scenarios  
710 (SWITCH1 and SWITCH2) (Table 2). Our SWITCH1 scenario increases global mean aviation-induced surface  
711 layer  $\text{PM}_{2.5}$  concentrations by  $+2.1 \text{ ng m}^{-3}$  [52.2%], European mean concentrations by  $+0.9 \text{ ng m}^{-3}$  [+4.5%], and  
712 North American concentrations by  $+2.7 \text{ ng m}^{-3}$  [+42.2%] relative to NORM (Fig. 4). These changes increase  
713 aviation-induced mortality by  $+17.4\%$  [+630 mortalities  $\text{a}^{-1}$ ] (Fig. 6). This scenario results in greater global mean  
714 increases in CCN (relative to NORM) of  $+1.2 \text{ cm}^{-3}$  [+51.2%], a larger cooling aCAE [ $-42.4 \text{ mW m}^{-2}$ ], larger  
715 warming aDRE [ $2.07 \text{ mW m}^{-2}$ ], resulting in additional  $-18.1 \text{ mW m}^{-2}$  [136%] of aviation-induced cooling  
716 [SWITCH1  $\text{RE}_{\text{comb}}$  of  $-31.4 \text{ mW m}^{-2}$ ].

718 The SWITCH2 scenario was designed to have the same global total sulfur emission as the normal aviation  
719 simulation. SWITCH2 increased global mean surface aviation-induced  $\text{PM}_{2.5}$  concentrations by  $+0.3 \text{ ng m}^{-3}$   
720 [+6.6%], but reduces mean surface  $\text{PM}_{2.5}$  concentrations over Europe [ $-1.8 \text{ ng m}^{-3}$ ;  $-8.7\%$ ] and North America  
721 [ $-0.8 \text{ ng m}^{-3}$ ;  $-12.8\%$ ] compared to NORM. Under this scenario global aviation-induced mortality is decreased  
722 by  $2.4\%$  [ $-90$  mortalities  $\text{a}^{-1}$ ] compared to the standard aviation simulation (Fig. 6). The SWITCH2 scenario  
723 results in a  $\text{RE}_{\text{comb}}$  of  $-18.2 \text{ mW m}^{-2}$ , providing an additional  $-4.9 \text{ mW m}^{-2}$  [36.6%] cooling in relation to standard  
724 aviation emissions (NORM; FSC = 600 ppm).

725

## 726 4 Discussion and Conclusions

727 We have used a coupled chemistry-aerosol microphysics model to estimate the impact of aviation emissions  
728 on aerosol and  $\text{O}_3$  concentrations, premature mortality and radiative effect on climate.

729

730 We calculated the top-of-atmosphere (TOA) tropospheric  $\text{O}_3$  radiative effect (O3DRE), aerosol direct RE (aDRE)  
731 and aerosol cloud albedo effect (aCAE). We find that these non- $\text{CO}_2$  REs result in a net cooling effect on climate  
732 as has been found previously (Sausen et al., 2005; Lee et al., 2009; Gettelman and Chen, 2013; Righi et al.,  
733 2013; Unger et al., 2013). For year 2000 aviation emissions with a standard fuel sulfur content (FSC = 600 ppm),  
734 we calculate a global annual mean net TOA RE of  $-13.3 \text{ mW m}^{-2}$ , due to a combination of O3DRE [ $+8.9 \text{ mW m}^{-2}$ ],  
735 aDRE [ $+1.4 \text{ mW m}^{-2}$ ] and aCAE [ $-23.6 \text{ mW m}^{-2}$ ].

736

737 Our O3DRE [ $+8.9 \text{ mW m}^{-2}$ ] when normalised to represent the impact of the emissions of  $1\text{Tg(N)}$  [ $+10.45 \text{ mW}$   
738  $\text{m}^{-2} \text{Tg(N)}^{-1}$ ] is at the lower end of range provided by previous studies [ $7.39\text{--}36.95 \text{ mW m}^{-2} \text{Tg(N)}^{-1}$ ] (Sausen et  
739 al., 2005; Hoor et al., 2009; Lee et al., 2009; Holmes et al., 2011; Myhre et al., 2011; Unger, 2011; Frömming  
740 et al., 2012; Unger et al., 2013; Khodayari et al., 2014). This can be attributed to our model's lower OPE of  
741 1.33, in comparison to the range of 1–2.4 from other models (Myhre et al., 2011; Unger, 2011).

742

743

744 Our estimate of aviation-induced aCAE [ $-23.6 \text{ mW m}^{-2}$ ] lies just outside the range provided by Gettelman and  
745 Chen (2013) and Righi et al. (2013) [ $-15.4$  to  $-21 \text{ mW m}^{-2}$ ]. Our estimated aDRE [ $+1.4 \text{ mW m}^{-2}$ ] lies within the  
746 middle of the range given by previous work (Sausen et al., 2005; Fuglestedt et al., 2008; Lee et al., 2009;  
747 Balkanski et al., 2010; Unger, 2011; Gettelman and Chen, 2013; Righi et al., 2013; Unger et al., 2013).

748  
749 We estimate that standard aviation (NORM; FSC = 600 ppm) is responsible for approximately 3,600 premature  
750 mortalities annually due to increased surface layer  $\text{PM}_{2.5}$ , in line with previous work (Barrett et al., 2012). We  
751 find that aviation-induced mortalities are highest over Europe, eastern North America and eastern China;  
752 reflecting larger regional perturbations in surface layer  $\text{PM}_{2.5}$  concentrations. Comparing these estimates with  
753 total global premature mortalities from ambient air pollution from all anthropogenic sources (Lim et al., 2012),  
754 aviation is responsible for 0.1% [0.04–0.18%] of annual premature mortalities.

755  
756 We investigated the impact of varying aviation FSC over the range 0–6000 ppm. Increases in FSC lead to  
757 increases in surface  $\text{PM}_{2.5}$  concentrations and subsequent increases in aviation-induced mortality. Increases in  
758 FSC also lead to a more negative  $\text{RE}_{\text{comb}}$  due to an enhanced aCAEs. We estimate that the use of ultra-low sulfur  
759 jet (ULSJ) fuel, with a FSC of 15 ppm, could prevent 620 [230–1,020] mortalities annually compared to standard  
760 aviation emissions. Swapping to ULSJ fuel increases the global mean net RE by  $+7.0 \text{ mW m}^{-2}$  compared to  
761 standard aviation emissions, largely due to a reduced aCAE. We calculate a larger warming effect from  
762 switching to ULSJ fuel than that assessed by Barrett et al. (2012), who did not evaluate changes in aCAE.

763  
764 Absolute reductions in FSC result in limited reductions in aviation-induced surface layer  $\text{PM}_{2.5}$ . We estimate  
765 that aviation- $\text{NO}_x$  emissions are responsible for 36.2% of aviation-induced sulfate perturbations. Thus further  
766 reductions in aviation-induced  $\text{PM}_{2.5}$  can potentially be achieved if  $\text{NO}_x$  emission reductions are implemented  
767 in tandem with reductions to fuel sulfur content.

768  
769 In line with previous work (Köhler et al., 2008; Stevenson and Derwent, 2009; Snijders and Melkers, 2011;  
770 Frömming et al., 2012; Skowron et al., 2013), decreasing the altitude at which  $\text{O}_3$  forming species are emitted  
771 results in a reduction in aviation-induced  $\text{O}_3$ , and resulting O3DRE. This is due to the relationship between  
772 altitude and OPE, and the inverse relationship between altitude and background pollutant concentrations. We  
773 also explored the sensitivity of emission injection altitude on aerosol, mortality and aerosol RE. Injecting  
774 aviation emissions at the surface results in a reduction in global mean concentrations of  $\text{PM}_{2.5}$  (relative to  
775 NORM), but with higher regional concentrations over central Europe and eastern America; resulting in higher  
776 annual mortalities due to aviation. We find that aviation emissions are a factor of 5 less efficient at creating  
777 CCN when released at the surface, resulting in an aCAE of  $-2.3 \text{ mW m}^{-2}$ , a reduction of 90.1% in relation to the  
778 standard aviation scenario. When aviation  $\text{SO}_2$  emissions are injected into the free-troposphere, the dominant  
779 oxidation pathway is to  $\text{H}_2\text{SO}_4$  followed by particle formation and condensational growth of new particles to  
780 larger sizes. Subsequent entrainment of these new particles into the lower atmosphere leads to increased CCN  
781 concentrations and impacts on cloud albedo. Aviation  $\text{SO}_2$  emissions are therefore particularly efficient at  
782 forming CCN with resulting impacts on cloud albedo.

783  
784 We explored the impact of applying altitude dependent variations in aviation FSC. We tested a scenario with  
785 high FSC in the free troposphere and low FSC near the surface, resulting in the same global aviation sulfur  
786 emission as the standard aviation scenario. In this scenario, aviation-induced premature mortalities were  
787 reduced by 2.4% [ $-90$  mortalities  $\text{a}^{-1}$ ] and the magnitude of the negative  $\text{RE}_{\text{comb}}$  was increased by 36.6%,  
788 providing an additional cooling impact of climate of  $-4.88 \text{ mW m}^{-2}$ .

789  
790 Our simulations suggest that the climate and air quality impacts of aviation are sensitive to FSC and the altitude  
791 of emissions. We explored a range of scenarios to maximise climate cooling and reduce air quality impacts.  
792 Use of ULSJ fuel (FSC = 15 ppm) at low altitude combined with high FSC in the free troposphere results in  
793 increased climate cooling whilst reducing aviation mortality. More complicated emission patterns, for  
794 example, use of high FSC only whilst over oceans might further enhance this effect. However, we note that  
795 the greatest reduction in aviation-induced mortality is simulated for complete desulfurisation of aviation fuel.  
796 Given the uncertainty in both the climate and air quality impacts of aerosol and ozone, additional simulations  
797 from a range of atmospheric models are required to explore the robustness of our calculations. Finally, we

798 note that our calculations are limited to calculation of aviation-induced RE. Future work needs to assess the  
799 complex climate impacts of altering aviation FSC. Future work needs to estimate the health impacts of aviation  
800 using newly available concentration response functions (Burnett et al., 2014).  
801

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