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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 $PM_{2.5}$ (particulate matter with a dry diameter of $< 2.5\,\mu m$) and on the global radiation budget due to changes in aerosol and tropospheric ozone. We estimate that present-day non-CO_2 aviation emissions with a typical FSC of 600 ppm result in 3597 (95 % CI: 1307–5888) annual mortalities globally due to increases in cases of cardiopulmonary disease and lung cancer, resulting from increased surface $PM_{2.5}$ concentrations. We quantify the global annual mean combined radiative effect (RE_{comb}) of non-CO_2 aviation emissions as $-13.3\,mW\,m^{-2}$; 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 $PM_{2.5}$. We calculate that swapping the global aviation fleet to ULSJ fuel would reduce the global aviation-induced mortality rate by 624 (95 % CI: 227–1021) mortalities a^{-1} and increase RE_{comb} by $+7.0\,mW\,m^{-2}$.

We explore the impact of varying aviation FSC between 0–6000 ppm. Increasing FSC increases annual mortality, while enhancing climate cooling through increasing the aerosol cloud albedo effect (aCAE). 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 aCAE. Aviation emissions injected at the surface are 5 times less effective at forming cloud condensation nuclei, reducing the aviation-induced aCAE by a factor of 10. Applying high FSCs at aviation cruise altitudes combined with ULSJ fuel at lower altitudes result in reduced aviation-induced mortality and increased negative RE compared to the baseline aviation scenario.

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1 Introduction

Aviation is the fastest growing form of transport (Lee et al., 2010; Uherek et al., 2010; Eyring et al., 2010), with a projected growth in passenger air traffic of 5 % yr^{-1} until 2030 (Barrett et al., 2012), and a projected near doubling of emissions by 2025, relative to 2005 (Eyers et al., 2005). These emissions, and changes to them, have both climate and air quality impacts (Barrett et al., 2012, 2010; Woody et al., 2011; Lee et al., 2009).

Aviation emits a range of gas-phase and aerosol pollutants that can influence climate. Emissions of carbon dioxide (CO_2) from aviation warm the climate (Lee et al., 2009, 2010). Emissions of nitrogen oxides (NO_x) warm the climate through tropospheric ozone (O_3) formation, which acts as a greenhouse gas, and cool climate via a decrease in the lifetime of the well-mixed greenhouse gas methane (CH_4) through increases in the OH radical (Myhre et al., 2011; Holmes et al., 2011). Sulfate and nitrate aerosols, predominantly formed from aviation sulfur dioxide (SO_2) and NO_x emissions and through altered atmospheric oxidants, lead to a cooling (Dessens et al., 2014; Righi et al., 2013; Unger, 2011), and black carbon (BC) emissions result in a warming (Balkanski et al., 2010). Additionally the formation of persistent linear contrails and contrail-cirrus from aircraft leads to warming (Rap et al., 2010; Lee et al., 2010; Burkhardt and Karcher, 2011). Overall, aviation emissions are thought to have a warming impact on climate, with net radiative forcing (RF) estimated as +55 mW m^{-2} (excluding cirrus cloud enhancement) (Lee et al., 2010).

In 2005, aviation was responsible for 3 % of all fossil fuel CO_2 emissions, and for 3.5 % of total anthropogenic radiative forcing when AIC (aviation-induced cloudiness) is excluded (90 % CI: 1.3–10 %), or 4.9 % (90 % CI: 2–14 %) when AIC is included (Lee et al., 2009). However, the atmospheric interactions between climatically relevant aviation emissions and their combined radiative effects are both highly uncertain.

Aviation emissions can increase atmospheric concentrations of fine particulate matter with a dry diameter of < 2.5 µm ($PM_{2.5}$) at the surface. Short-term exposure to fine PM can exacerbate existing respiratory and cardiovascular ailments, while long-term

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exposure can result in chronic respiratory and cardiovascular diseases, lung cancer, chronic changes in physiological functions and mortality (World Health Organisation, 2003; Ostro, 2004; Pope et al., 2002). In the US aviation emissions are estimated to lead to adverse health effects in ∼ 11 000 people (ranging from mortality, respiratory ailments and hospital admissions due to exacerbated respiratory conditions) and ∼ 23 000 work loss days per annum (Ratliff et al., 2009). Landing and take-off aviation emissions increase $PM_{2.5}$ ($D_p > 2.5\,\mu m$) concentrations, particularly around airports (Woody et al., 2011), resulting in 75 premature mortalities in 2005 (Yim et al., 2015; Woody et al., 2011; Levy et al., 2012). Worldwide it is estimated that aviation emissions, are annually responsible for ∼ 10 000 premature mortalities (Barrett et al., 2010, 2012).

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

However, application of ULSJ fuel will not completely remove the impacts of aviation on $PM_{2.5}$. It is estimated that over a half of aviation-attributable surface-level sulfate is associated with oxidation of non-aviation SO_2 by OH produced from aviation NO_x emissions, and not directly produced from aviation-emitted SO_2 (Barrett et al., 2010). Therefore even a completely desulfurised global aviation fleet would likely produce increases in sulfate $PM_{2.5}$. Nevertheless, previous work has shown that the use of ULSJ fuel reduces global aviation-induced $PM_{2.5}$ by ∼ 23 %, annually avoiding ∼ 2300 (95 % CI: 890–4200) mortalities (Barrett et al., 2012).

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

In this study, we investigate the impacts of changes in the sulfur content of aviation fuel on climate and human health. A coupled tropospheric chemistry-aerosol microphysics model including nitrate aerosol is used to quantify global atmospheric responses in aerosol and O_3 to varying FSC scenarios. Radiative effects due to changes in tropospheric O_3 and aerosols are calculated using a radiative transfer model. The impacts of changes in surface $PM_{2.5}$ on human health are estimated using concentration response functions.

2 Methods

2.1 Coupled chemistry–aerosol microphysics model

We use GLOMAP-mode (Mann et al., 2010), embedded within the 3-D off-line Eulerian chemical transport model TOMCAT (Arnold et al., 2005; Chipperfield, 2006). GLOMAP-mode is a two-moment aerosol microphysics scheme representing particles as an external mixture of 7 size modes (4 soluble and 3 insoluble) (Mann et al., 2010). We use the nitrate-extended version of GLOMAP-mode (Benduhn et al., 2015) which, as well as tracking size-resolved sulfate, BC, organic carbon (OC), sea-salt and dust components, also includes a dissolution solver to accurately characterise the size-resolved partitioning of ammonia and nitric acid into ammonium and nitrate components in each soluble mode. Aerosol components are assumed to be internally mixed within each mode. GLOMAP-mode includes representations of nucleation, particle growth via co-

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agulation, condensation and cloud processing, wet and dry deposition, and in- and below-cloud scavenging (Mann et al., 2010).

TOMCAT includes a tropospheric gas-phase chemistry scheme (inclusive of O_x-NO_Y-HO_x), treating the degradation of C_1-C_3 non-methane hydrocarbons (NMHCs) and isoprene, together with a sulfur chemistry scheme (Breider et al., 2010; Mann et al., 2010; Spracklen et al., 2005). Tracer transport is driven by winds from European Centre for Medium-Range Weather Forecasts (ECMWF) analyses at 6 hourly intervals together with a convective parameterization and boundary layer mixing (see Chipperfield, 2006). Here, we ran simulations at a horizontal resolution of 2.8° × 2.8° with 31 hybrid σ-p levels extending from the surface to 10 hPa.

All simulations were conducted for 16 months from September 1999 to December 2000 inclusive, with the first four months discarded as spin-up time.

2.2 Aviation emissions

Aircraft emit NO_x, carbon monoxide (CO), SO_2, BC, organic carbon (OC) and hydrocarbons (HCs). The historical emissions dataset for the CMIP5 (5th Coupled Model Intercomparison Project) model simulations used by the IPCC 5th Assessment Report only included NO_x and BC aviation emissions (Lamarque et al., 2009). Recently there have been efforts to add HCs, CO and SO_2 emissions to aviation emission inventories (Wilkerson et al., 2010; Eyers et al., 2005; Quantify Integrated Project, 2005–2012).

Here we develop a new 3-D civil aviation emissions dataset for the year 2000, based on CMIP5 historical aviation emissions (Lamarque et al., 2009). The new dataset includes emissions of NO_x, CO, SO_2, BC, OC, and HCs. In contrast to existing datasets which provide a general emissions index for HCs (Eyers et al., 2005) we speciate HCs as formaldehyde (HCHO), ethane (C_2H_6), propane (C_3H_8), methanol (CH_3OH), acetaldehyde (CH_3CHO), and acetone ($(CH_3)_2CO$).

Table 1 describes our new emissions dataset. NO_x and BC emissions are taken directly from Lamarque et al. (2009). We calculate fuelburn from BC emissions data and the BC emissions index (Eyers et al., 2005) as used by Lamarque et al. (2009). Follow-

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ing DuBois and Paynter (2006), we assume that BC emissions scale linearly with fuel consumption. We estimate emissions for other species using our calculated aviation fuelburn in combination with published species-specific emissions indices (EI reported in $g\,kg^{-1}$ of fuel). Emission indices for CO and SO_2 are taken from the FAA's aviation environmental design tool (AEDT) (Wilkerson et al., 2010). OC emissions are calculated using a BC : OC ratio of 4 (Bond et al., 2004; Hopke, 1985); resulting in an EI within the range determined by Wayson et al. (2009). Speciated hydrocarbon emissions are calculated from experimental data following the methodology of Wilkerson et al. (2010) using experimental data from Knighton et al. (2007) and Anderson et al. (2006), in conjunction with operating parameters suggested by the Airbus Flight Crew Training manual (Airbus, 2008).

Our global aviation emissions typically lie within the range of previous studies (Table 1). Our SO_2 emissions are greater than those used by Wilkerson et al. (2010) for 2006, despite the use of the same EI. This is due to the greater global fuelburn considered by the base inventory used to develop our emissions inventory (Lamarque et al., 2010; Eyers et al., 2005). Our estimated OC emissions are lower than the emissions estimated in the AEDT 2006 inventory, due to the lower EI applied here.

We calculate the geometric mean diameter (D_g) for internally mixed BC/OC particles as 50.46 nm from the mean particle mass derived using the particle number emissions index (Eyers et al., 2005) and a constant standard deviation set to $\sigma = 1.59$ nm (Stier et al., 2005).

2.3 Fuel sulfur content simulations

To explore the impact of aviation FSC on climate and air quality we performed a series of 11 global model experiments (Table 2). In 7 of these model experiments FSC values were varied globally between zero and 6000 ppm. Three further simulations varied the vertical distribution of aviation emissions. The first simulation collapses all aviation emissions to ground level (GROUND), in order to compare an equivalent ground emission source and its effects. Two simulations (SWITCH1 and SWITCH2), use a low FSC

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(15 ppm) applied below the cruise phase of flight (< 8.54 km altitude) (Köhler et al., 2013; Lee et al., 2009) combined with a high FSC at altitudes above. The SWITCH1 scenario increases FSC in line with our HIGH scenario above 8.54 km, while in the SWITCH2 scenario, emissions are scaled such that total global sulfur emissions are the same as the standard simulation (NORM), resulting in a FSC of 1420 ppm above 8.54 km. Results from all simulations are compared against a simulation with aviation emissions excluded (NOAVI).

2.4 Radiative impacts

We calculate the aerosol direct radiative effect (aDRE), aerosol cloud albedo effect (aCAE) and tropospheric O_3 direct radiative effect (O3DRE) using the offline Edwards and Slingo (1996) radiative transfer model. The radiative transfer model considers 6 bands in the shortwave (SW) and 9 bands in the longwave (LW), adopting a delta-Eddington 2 stream scattering solver at all wavelengths. The top-of-the-atmosphere (TOA) aerosol aDRE and aCAE are calculated using the methodology described in Rap et al. (2013) and Spracklen et al. (2011), with the method for O3DRE as in Richards et al. (2013). To determine the aCAE we calculated cloud droplet number concentrations (CDNCs) using the monthly mean aerosol size distribution simulated by GLOMAP combined with parameterisations from Nenes and Seinfeld (2003), updated by Fountoukis and Nenes (2005) and Barahona et al. (2010). CDNC were calculated with a prescribed updraft velocity of 0.15 $m\,s^{-1}$ over ocean and 0.3 $m\,s^{-1}$ over land. Changes to CDNC were then used to perturb the effective radii of cloud droplets in low- and mid-level clouds (up to 600 hPa). The aDRE, aCAE and O3DREs for each aviation emissions scenario are calculated as the difference in TOA net (SW + LW) radiative flux compared to the NOAVI simulation.

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2.5 Health effects

We calculate excess premature mortality from cardiopulmonary diseases and increases in cases of lung cancer due to long-term exposure to aviation-induced $PM_{2.5}$ (Ostro, 2004). $PM_{2.5}$ is used as a measure of likely health impacts because chronic exposure is associated with adverse human health impacts including morbidity and mortality (Dockery et al., 1993; Pope and Dockery, 2006).

We relate annual excess mortality to annual mean surface $PM_{2.5}$ via a concentration response-function (Ostro, 2004). This response-function considers concentrations of $PM_{2.5}$ for a perturbed case (defined by aviation emissions scenarios from Table 2) in relation to a baseline case with no aviation emissions (NOAVI). We assume that the cause specific coefficient (β) for cardiopulmonary disease related mortality is 0.155 (95 % CI = 0.056–0.254) and β for lung cancer is 0.232 (95 % CI = 0.086–0.379) (Pope et al., 2002; Ostro, 2004). Baseline mortality is specified via a country-specific baseline mortality rate (Mathers et al., 2008) with population over 30 years of age from the Gridded World Population (GWP; version3) project (Center for International Earth Science Information Network, 2012).

3 Results

3.1 Surface $PM_{2.5}$

Figure 1 shows the simulated impact of aviation emissions with standard FSC (FSC = 600 ppm; NORM) on surface $PM_{2.5}$ concentrations. The greatest absolute increases (up to 80 ng m^{-3}) in annual mean $PM_{2.5}$ concentrations occur over central Europe and eastern China (Fig. 1a). Aviation emissions result in largest fractional changes in annual mean $PM_{2.5}$ concentrations (up to 0.8 %) over North America and Europe (Fig. 1b).

Figure 2 shows the impact of aviation emissions on global and regional mean $PM_{2.5}$ concentrations, as a function of FSC. With standard FSC (FSC = 600 ppm), aviation

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increases global mean surface $PM_{2.5}$ concentrations by 3.9 ng m^{-3}; with increases in $PM_{2.5}$ dominated by sulfates (56.2 %), nitrates (26.0 %) and ammonium (16.0 %). Aviation emissions increase European annual mean $PM_{2.5}$ concentrations by 20.3 ng m^{-3} (Fig. 2b), substantially more than over North America (Fig. 2c) where an annual mean increase of 6.3 ng m^{-3} is simulated. Increased $PM_{2.5}$ is dominated by nitrates, both over Europe (55.5 %) and over North America (44.4 %). Sulfates contribute up to 44.6 % of increases in $PM_{2.5}$ over North America, and 30.0 % over Europe.

The use of ULSJ fuel (FSC = 15 ppm) reduces global annual mean surface aviation-induced $PM_{2.5}$ concentrations (in relation to the NORM case) by 35.7 % (1.41 ng m^{-3}) (Fig. 2); predominantly due to changes in sulfate (−1.37 ng m^{-3}; −62.1 %) and ammonium (−0.24 ng m^{-3}; −37.9 %), which are marginally offset by increases in nitrates ($+3.17 \times 10^{-3}$ ng m^{-3}; +0.3 %). In comparison to the global mean, larger absolute reductions in $PM_{2.5}$ are simulated over Europe [−4.21 ng m^{-3}] and North America (−3.38 ng m^{-3}) (Fig. 2b and c). Over North America, swapping to ULSJ fuel reduces aviation-induced $PM_{2.5}$ by 53.4 %, while a smaller reduction of 2.5 % is simulated over Europe. The smaller fractional change in $PM_{2.5}$ over Europe is caused by smaller reductions in aviation-induced sulfate (−55.9 %) and ammonium (−18.4 %) compared to over North America, which sees a reduction in ammonium of 41.6 % and a reduction in sulfates of 103 % indicating that over the US the ULSJ fuel scenario sees a reduction in sulfates in relation to a NOAVI scenario.

Complete desulfurisation of jet fuel (FSC = 0 ppm; DESUL) reduces global mean aviation-induced surface $PM_{2.5}$ concentrations by 36.5 % (−1.43 ng m^{-3}), with changes in sulfates (−1.40 ng m^{-3}; −63.5 %) and ammonium (−0.24 ng m^{-3}; −38.8 %) dominating. Under this scenario the reductions in surface sulfate $PM_{2.5}$ from aviation are 57.3 % over Europe and 105 % over North America. ULSJ fuel therefore gives similar results to complete desulfurisation, due to the very small sulfur emission from ULSJ fuel (Table 2).

In summary, increases in FSC result in increased surface $PM_{2.5}$, due to increased sulfate outweighing the small reductions in nitrate. Simulated changes in sulfate, ni-

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trate, ammonium and total $PM_{2.5}$ are linear (R^2 = 1.00, p value < 0.001 globally and for all individual regions) with respect to FSC (Fig. 2). The impact of variations in FSC on $PM_{2.5}$ are regionally variable; over Europe changes in $PM_{2.5}$ concentrations are observed to be more sensitive to changes in FSC than over North America, and the global domain.

Figure 3 shows the impact of changing to ULSJ fuel on zonal mean sulfate and nitrate concentrations relative to standard fuel (NORM). Table 3 reports the global aerosol burden from aviation under different emission scenarios. With standard FSC (FSC = 600 ppm), the global aviation-induced aerosol burden is 16.9 Gg, dominated by sulfates (76.3 %) and nitrates (33.4 %). The use of ULSJ (FSC = 15 ppm) reduces the global aerosol burden from aviation by 26.8 %. Complete desulfurisation of aviation fuel reduces the global aerosol burden from aviation by 28.4 %, with the global sulfate burden from aviation reduced by 71.6 % (Table 3). When aviation emissions contain no sulfur, aviation-induced sulfate is formed through aviation NO_x-induced increases in OH concentrations, resulting in the increased oxidation of SO_2 from non-aviation sources (Barrett et al., 2010; Unger et al., 2006).

We found that for standard FSC, 36.2 % of aviation-attributable sulfate formed at the surface is associated with aircraft NO_x emissions and not directly with aviation sulfur emissions. This is less than the estimate, of more than half, from Barrett et al. (2010). We find desulfurisation increases the aviation nitrate burden by 5.1 % (Table 3); although much of this increase occurs at altitudes well above the surface (Figure 3) and so is not reflected in surface $PM_{2.5}$ concentrations.

We explored the impacts of NO_x emission reductions in combination with fuel desulfurisation. A scenario with desulfurised fuel and zero NO_x emissions reduces the global aviation-induced aerosol burden by 88.3 % (Table 3), in comparison to a desulfurised only case (DESUL), where the aviation-induced aerosol burden in reduced by 28.4 %. Removal of aviation NO_x and SO_2 emissions results in a 95. % reduction in aviation-induced global mean surface level aviation-induced $PM_{2.5}$. These results imply that only limited sulfate reductions are achieved through reducing FSC alone, with further

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reductions in aviation-induced $PM_{2.5}$ sulfates requiring additional controls on aviation NO_x emissions.

3.2 Premature mortality

Figure 4 shows estimated annual premature mortalities (from cardiopulmonary disease and lung cancer) due to aviation-induced changes in $PM_{2.5}$ as a function of FSC. We estimate that aviation emissions with standard FSC (FSC = 600 ppm) cause 3597 (95 % CI: 1307–5888) premature mortalities each year. Low-, mid- and high-range cause-specific coefficients (β) are used to account for uncertainty (Sect. 2.5). Our mid-range aviation-induced premature mortality estimate represents 36 % of that estimated by Barrett et al. (2012) ($\sim 10\,000$ mortalities a^{-1}). Our estimated global mortality due to aviation emissions is greatest in the Northern Hemisphere, which accounts for 98.7 % of global mortalities. Europe and North America account for 42.3 and 8.4 % of mortality due to aviation emissions respectively.

Greater aviation-induced mortality simulated by (Barrett et al., 2012), can be attributed to greater aviation-induced surface $PM_{2.5}$ concentrations simulated in their study, particulary over highly populated areas. Their study simulated maxmium aviation-induced $PM_{2.5}$ concentrations over Europe, eastern China and eastern North America greater than those in our simulations by factors of 5 for Europe and eastern China and 2.5 over eastern North America. Our aviation-induced sulfate concentrations compare well with (Barrett et al., 2012), indicating that the resulting differences in aviation-induced surface $PM_{2.5}$ concentrations are a result of other aerosol components.

We estimate that aviation emissions with ULSJ fuel result in 2973 (95 % CI: 1080–4867) premature mortalities globally per annum from increases in $PM_{2.5}$. Therefore changing from standard FSC to ULSJ would result in 624 (95 % CI: 227–1021) 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 178 over Europe and by 107 over North America.

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Barrett et al. (2012) estimated that swapping to ULSJ fuel could result in ∼ 2300 (95 % CI: 890–4200) fewer premature mortalities globally per annum; a reduction of 23 %. In their work, the use of ULSJ reduces global mean $PM_{2.5}$ concentrations by 0.89 ng m^{-3}. In comparison we calculate a greater reduction of 1.61 ng m^{-3}, when considering the same three aerosol components (sulfates, nitrates and ammonium). When considering all aerosol components we calculate a net reduction in surface $PM_{2.5}$ of 1.41 ng m^{-3}; due to an increase in other aerosol species (BC, OC, Na^+, dust and Cl^-) of +0.20 ng m^{-3}.

Despite the greater reductions in global mean surface layer $PM_{2.5}$ concentrations simulated here, Barrett et al. (2012) simulate greater reductions in $PM_{2.5}$ over populated regions, resulting in greater reductions of aviation-induced mortality under the ULSJ scenario.

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 6034, an increase of 67.8 % in relation to standard aviation (NORM; FSC = 600 ppm).

3.3 Sensitivity of cloud condensation nuclei to aviation FSC

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-cloud level (879 hPa; 0.96 km) by 0.9 % (2.3 cm^{-3}) (Fig. 5a). Increases in CCN concentrations are greater in the Northern Hemisphere (+3.9 cm^{-3}; +1.4 %) compared to the Southern Hemisphere (+0.7 cm^{-3}; +0.5 %). Maximum increases in low-level CCN are simulated over the Pacific, central Atlantic and Arctic Oceans.

The use of ULSJ (FSC = 15 ppm) reduces global mean low-level CCN concentrations by 0.4 cm^{-3}, (−18.2 %) relative to the NORM case (Fig. 5). Northern Hemisphere

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CCN concentrations are reduced by 0.8 cm^{-3} (−19.4 %), while Southern Hemisphere concentrations are reduced by 0.1 cm^{-3} (−11.5 %) (Fig. 5).

Figure 6 shows the sensitivity of low level CCN concentrations to FSC. As with $PM_{2.5}$, we find simulated changes in CCN are linear with respect to FSC (R^2 = 1.00 and p value < 0.001 globally and for all individual regions).

ULSJ fuel reduces global mean CCN by −0.42 cm^{-3} with largest reductions over the Atlantic Ocean (−0.81 cm^{-3}), North America (−0.55 cm^{-3}), and the Pacific Ocean (−0.51 cm^{-3}), i.e. in relation to standard aviation (ULSJ–NORM). The complete desulfurisation of aviation fuel results in reductions in CCN in relation to standard aviation (DESUL–NORM) which follow the same regional trends (Fig. 6a).

3.4 Sensitivity of aerosol and ozone radiative effect to FSC

Figure 7 shows the calculated global mean net RE due to non-CO_2 aviation emissions. For standard FSC (FSC = 600 ppm) emissions the global mean combined RE is −13.3 $mW\,m^{-2}$.

This combined radiative effect (RE_{comb}) results from a balance between a positive aDRE of +1.4 $mW\,m^{-2}$ and O3DRE +8.9 $mW\,m^{-2}$, and a negative aCAE of −23.6 $mW\,m^{-2}$ (Fig. 7).

Our estimated aviation aerosol DRE (+1.4 $mW\,m^{-2}$) lies in the middle of the range given by previous work. The aviation aerosol DRE has been previously assessed as highly uncertain, ranging between −28 to +20 $mW\,m^{-2}$ (Righi et al., 2013). Our estimated aviation-induced aCAE (−23.6 $mW\,m^{-2}$) lies within the range of uncertainty from previous literature: Righi et al. (2013) estimated −15.4 ± 10.6 $mW\,m^{-2}$ and Gettelman and Chen (2013) estimated −18 ± 11 $mW\,m^{-2}$.

Our O3DRE estimate (+8.9 $mW\,m^{-2}$), normalised by global aviation NO_x emission to +10.5 $mW\,m^{-2}\,Tg(N)^{-1}$, is at the lower end of current estimates (7.4–37.0 $mW\,m^{-2}\,Tg(N)^{-1}$) (Myhre et al., 2011; Holmes et al., 2011; Lee et al., 2009; Sausen et al., 2005; Frömming et al., 2012; Hoor et al., 2009; Unger, 2011; Unger

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et al., 2013; Köhler et al., 2008; Khodayari et al., 2014; Skowron et al., 2013). This can be attributed to the lower net O_3 chemical production efficiency (OPE) within our model (1.33). Unger (2011) estimated an O3DRE of 7.4 mW m^{-2} Tg(N)$^{-1}$ with a model OPE of ∼ 1, while the ensemble of models considered by Myhre et al. (2011) have an OPE range of 1.5–2.4, resulting in an O3DRE range of 16.2–25.4 mW m^{-2} Tg(N)$^{-1}$.

We calculate that an aviation fleet utilising ULSJ fuel would result in a in a global annual mean RE_{comb} of −6.3 mW m^{-2} (aDRE = +1.8 mW m^{-2}; aCAE = −16.8 mW m^{-2}; and O3DRE = +8.7 mW m^{-2}). Thus, swapping from standard aviation fuel to ULSJ fuel reduces the net cooling effect from aviation-induced aerosol and O_3 by 7.0 mW m^{-2}, in comparison to the reduction of 3.3 mW m^{-2} estimated by Barrett et al. (2012). In our model, this change is primarily due a reduction in cooling from the aCAE of +6.7 mW m^{-2} combined with smaller contributions from an increased aDRE of +0.4 mW m^{-2}, and reduction in warming from the O3DRE of −0.12 mW m^{-2} (Fig. 7).

When we assume fully desulfurised aviation jet fuel (DESUL; FSC = 0 ppm), the RE_{comb} induced by aviation-induced aerosol and O_3 is very similar to that for ULSJ fuel and is estimated as −6.1 mW m^{-2} (aDRE = +1.8 mW m^{-2}; aCAE = −16.6 mW m^{-2}; and O3DRE = +8.7 mW m^{-2}).

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_{comb} 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_{comb}; increasing in magnitude by a factor of ∼ 5 (−16.6 mW m^{-2} for FSC = 0 ppm to −82.1 mW m^{-2} for FSC = 6000 ppm) (Fig. 7). Therefore we find that increases in FSC provide a cooling effect due to the dominating effect from aviation-induced aCAE.

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3.5 Relationship between aviation-induced radiative effects and mortality due to aviation non-CO_2 emissions

Figure 8 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(mortalities)/d(FSC) (mortalities ppm^{-1}) and d(RE)/d(FSC) (mW m^{-2} ppm^{-1}). We calculate the sensitivity of global premature mortality to be 1.0 mortalities ppm^{-1} (95 % CI = 0.4 to 1.6 mortalities ppm^{-1}, where the range is due to uncertainty in β). The global mean RE_{comb} has a sensitivity of -1.2×10^{-2} mW m^{-2} ppm^{-1}, dominated by large changes to the aCAE (-1.1×10^{-2} mW m^{-2} ppm^{-1}), and much smaller changes in the aDRE (-6.9×10^{-4} mW m^{-2} ppm^{-1}) and O_3 RE ($+4.4 \times 10^{-5}$ mW m^{-2} ppm^{-1}).

To assess how the vertical distributions of aviation SO_2 emissions influence human health and climate effects, we performed three additional simulations where we altered the vertical distribution of aviation SO_2 emissions (GROUND, SWITCH1 and SWITCH2 simulations). In these simulations the relationships between mortality and RE_{comb} deviate from the linear relationship seen when varying FSC between 0–6000 ppm (Fig. 8).

In relation to the standard aviation emissions simulation (FSC = 600 ppm; NORM), when we release all aviation emissions at the surface (GROUND; FSC = 600 ppm) aviation-induced surface $PM_{2.5}$ concentrations increase by +13.5 ng m^{-3} (+65.7 %) over Europe and by +1.7 ng m^{-3} (+27.1 %) over North America, but decrease by −1.4 ng m^{-3} (−36.7 %) globally (Fig. 2). Greater surface layer $PM_{2.5}$ perturbations (GROUND–NORM) over populated regions increase aviation-induced annual mortality by +22.9 % (+825 mortalities a^{-1}) (Fig. 4).

Releasing aviation emissions at the surface (GROUND case) increases global mean cloud level CCN by only 0.4 cm^{-3} relative to NOAVI; providing a reduction in CCN of 82.1 % (−1.89 cm^{-3}) relative to the NORM case (i.e. GROUND–NORM). That is, injecting aviation emissions into the free troposphere in the standard scenario is over 5 times

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more efficient at increasing CCN concentrations compared to when the same emissions are released at the surface (GROUND CCN = 0.4 cm^{-3}; NORM CCN = 2.3 cm^{-3}); both in relation to the NOAVI scenario. Similar behaviour has been demonstrated previously for volcanic SO_2 emissions (Schmidt et al., 2012), where volcanic SO_2 emissions injected into the free troposphere (FT) were more than twice as effective at producing new CCN compared to boundary layer emissions of DMS. Injection of aviation SO_2 emissions at the surface will increase both deposition rates and aqueous phase oxidation of SO_2; the latter resulting in the growth of existing CCN, but not the formation of new CCN. In contrast, when SO_2 is emitted into the FT the dominant oxidation mechanism is to H_2SO_4, leading to the formation of new CCN through particle formation and the condensational growth of particles to larger sizes. Subsequent entrainment of these new particles into the lower atmosphere results in enhanced CCN concentrations in low level clouds. Reduced CCN formation when aviation emissions are injected at the surface has implications for the aCAE. When aviation emissions are released at the surface we calculate an aCAE of $-2.3\,mW\,m^{-2}$; a factor of 10 smaller than the standard aviation scenario. This demonstrates that low-level CCN concentrations and the aCAE are particularly sensitive to aviation emissions, because of the efficient formation of CCN when SO_2 emissions are injected into the FT. Injecting aviation emissions at the surface also results in an increase in the aDRE of $+5.9\,mW\,m^{-2}$, resulting in a RE_{comb} of $+5.\,mW\,m^{-2}$ (Fig. 7).

Surface O_3 concentrations are also less sensitive to aviation when emissions are located at the surface. Global mean aviation-induced surface O_3 concentrations are reduced from 0.15 ppbv (NORM) to 0.3 ppbv when all emissions are in the surface layer. Releasing aviation emissions at the surface also reduces the global O_3 burden by 3.1 Tg. These perturbations in O_3 concentrations result in a reduction in the O_3 radiative effect from $+8.9\,mW\,m^{-2}$ (NORM; FSC = 600 ppm) to $+1.5\,mW\,m^{-2}$ (GROUND; FSC = 600 ppm) (Fig. 7). This is a reflection of increases in the OPE of NO_x with increases in altitude due to lower background NO_x and NMHC (non-methane hydrocar-

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bon) concentrations (Skowron et al., 2013; Köhler et al., 2008; Stevenson and Derwent, 2009; Snijders and Melkers, 2011).

We investigated altering FSC between the take-off/landing and the cruise phases of flight using two scenarios (SWITCH1 and SWITCH2) (Table 2). Our SWITCH1 scenario increases global mean aviation-induced surface layer $PM_{2.5}$ concentrations by $+2.1\,ng\,m^{-3}$ (52.2 %), European mean concentrations by $+0.9\,ng\,m^{-3}$ (+4.5 %), and North American concentrations by $+2.7\,ng\,m^{-3}$ (+42.2 %) relative to NORM (Fig. 2). These changes increase aviation-induced mortality by +17.4 % (+625 mortalities a^{-1}) (Fig. 4). This scenario results in greater global mean increases in CCN (relative to NORM) of $+1.2\,cm^{-3}$ (+51.2 %), a larger cooling aCAE ($-42.4\,mW\,m^{-2}$), larger warming aDRE ($2.07\,mW\,m^{-2}$), resulting in additional $-18.1\,mW\,m^{-2}$ (136 %) of aviation-induced cooling (SWITCH1 RE_{comb} of $-31.4\,mW\,m^{-2}$).

The SWITCH2 scenario was designed to have the same global total sulfur emission as the normal aviation simulation. SWITCH2 increased global mean surface aviation-induced $PM_{2.5}$ concentrations by $+0.3\,ng\,m^{-3}$ (+6.6 %), but reduces mean surface $PM_{2.5}$ concentrations over Europe ($-1.8\,ng\,m^{-3}$; −8.7 %) and North America ($-0.8\,ng\,m^{-3}$; −12.8 %) compared to NORM. Under this scenario global aviation-induced mortality is decreased by 2.4 % (−87 mortalities a^{-1}) compared to the standard aviation simulation (Fig. 4). The SWITCH2 scenario results in a RE_{comb} of $-18.2\,mW\,m^{-2}$, providing an additional $-4.9\,mW\,m^{-2}$ (36.6 %) cooling in relation to standard aviation emissions (NORM; FSC = 600 ppm).

4 Discussion and conclusions

We have used a coupled chemistry–aerosol microphysics model to estimate the impact of aviation emissions on aerosol and O_3 concentrations, premature mortality and radiative effect on climate.

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We calculated the top-of-atmosphere (TOA) tropospheric O_3 radiative effect (O3DRE), aerosol direct RE (aDRE) and aerosol cloud albedo effect (aCAE). We find that these non-CO_2 REs result in a net cooling effect on climate as has been found previously (Unger et al., 2013; Lee et al., 2009; Sausen et al., 2005; Gettelman and Chen, 2013; Righi et al., 2013). For year 2000 aviation emissions with a standard fuel sulfur content (FSC = 600 ppm), we calculate a global annual mean net TOA RE of $-13.3\,mW\,m^{-2}$, due to a combination of O3DRE ($+8.9\,mW\,m^{-2}$), aDRE ($+1.4\,mW\,m^{-2}$) and aCAE ($-23.6\,mW\,m^{-2}$).

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

Our estimate of aviation-induced aCAE ($-23.6\,mW\,m^{-2}$) lies just outside the range provided by Gettelman and Chen (2013) and Righi et al. (2013) (−15.4 to $-18\,mW\,m^{-2}$). Our estimated aDRE ($+1.4\,mW\,m^{-2}$) lies within the middle of the range given by previous work (Balkanski et al., 2010; Fuglestvedt et al., 2008; Unger, 2011; Unger et al., 2013; Lee et al., 2009; Gettelman and Chen, 2013; Sausen et al., 2005; Righi et al., 2013).

We estimate that standard aviation (NORM; FSC = 600 ppm) is responsible for 3597 premature mortalities a^{-1} due to increased surface layer $PM_{2.5}$, in line with previous work (Barrett et al., 2012). We find that aviation-induced mortalities are highest over Europe, eastern North America and eastern China; reflecting larger regional perturbations in surface layer $PM_{2.5}$ concentrations. Comparing these estimates with total global premature mortalities from ambient air pollution from all anthropogenic sources (Lim et al., 2012), aviation is responsible for 0.1 % (0.4–0.18 %) of annual premature mortalities.

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We investigated the impact of varying aviation FSC over the range 0–6000 ppm. Increases in FSC lead to increases in surface $PM_{2.5}$ concentrations and subsequent increases in aviation-induced mortality. Increases in FSC also lead to a more negative RE_{comb} due to an enhanced aCAE. We estimate that the use of ultra-low sulfur jet (ULSJ) fuel, with a FSC of 15 ppm, could prevent 624 (227–1021) mortalities annually compared to standard aviation emissions. Swapping to ULSJ fuel increases the global mean net RE by $+7.0\,mW\,m^{-2}$ compared to standard aviation emissions, largely due to a reduced aCAE. We calculate a larger warming effect from switching to ULSJ fuel than that assessed by Barrett et al. (2012), who did not evaluate changes in aCAE.

Absolute reductions in FSC result in limited reductions in aviation-induced surface layer $PM_{2.5}$. We estimate that aviation-NO_x emissions are responsible for 36.2 % of aviation-induced sulfate perturbations. Thus further reductions in aviation-induced $PM_{2.5}$ can potentially be achieved if NO_x emission reductions are implemented in tandem with reductions to fuel sulfur content.

In line with previous work (Köhler et al., 2008; Stevenson and Derwent, 2009; Snijders and Melkers, 2011; Frömming et al., 2012; Skowron et al., 2013), decreasing the altitude at which O_3 forming species are emitted results in a reduction in aviation-induced O_3, and resulting O3DRE. This is due to the relationship between altitude and OPE, and the inverse relationship between altitude and background pollutant concentrations. We also explored the sensitivity of emission injection altitude on aerosol, mortality and aerosol RE. Injecting aviation emissions at the surface results in a reduction in global mean concentrations of $PM_{2.5}$ (relative to NORM), but with higher regional concentrations over central Europe and eastern America; resulting in higher annual mortalities due to aviation. We find that aviation emissions are a factor 5 less efficient at creating CCN when released at the surface, resulting in an aCAE of $-2.3\,mW\,m^{-2}$, a reduction of 90.1 % in relation to the standard aviation scenario. When aviation SO_2 emissions are injected into the free-troposphere, the dominant oxidation pathway is to H_2SO_4 followed by particle formation and condensational growth of new particles to larger sizes. Subsequent entrainment of these new particles into the lower atmosphere

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leads to increased CCN concentrations and impacts on cloud albedo. Aviation SO_2 emissions are therefore particularly efficient at forming CCN with resulting impacts on cloud albedo.

We explored the impact of applying altitude dependent variations in aviation FSC. We tested a scenario with high FSC in the free troposphere and low FSC near the surface, resulting in the same global aviation sulfur emission as the standard aviation scenario. In this scenario, aviation-induced premature mortalities were reduced by 2.4 % ($-87\,\text{mortalities}\,\text{a}^{-1}$) and the magnitude of the negative RE_{comb} was increased by 36.6 %, providing an additional cooling impact of climate of $-4.88\,\text{mW}\,\text{m}^{-2}$.

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

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Table 1. Aviation emissions indices and total annual emissions for year 2000.

Species	Emissions index ($g\,kg^{-1}$ of fuel)	Global emissions for year 2000 (Tg of species)	Range of annual global emissions from previous studies (Tg of species)
NO_x	13.89[a]	2.786	1.98–3.286[a, j, h, i, k, l]
CO	3.61[b]	0.724	0.507–0.679[h, i, j]
HCHO	1.24[c, d]	0.249	n/a
C_2H_6	0.0394[e]	0.007899	n/a
C_3H_8	0.03[e]	0.006014	n/a
CH_3OH	0.22[d]	0.044	n/a
CH_3CHO	0.33[d]	0.066	n/a
$(CH_3)_2CO$	0.18[d]	0.036	n/a
SO_2	1.1760[b]	0.236	0.182–0.221[a, h, i, j]
BC	0.0250[a]	0.005012	0.0039–0.0068[a, b, h, i, j, k]
OC	0.00625[f, g]	0.001253	0.003[l]

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

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Table 2. FSC and global SO_2 emissions applied in each model experiment.

Scenario name	Description	FSC (ppm)	Total SO_2 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

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Table 3. Global aviation-induced aerosol mass burdens for different emission scenarios. Values in 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_x and SO_2	2.0 (−88.3 %)	0.3 (−97.5 %)	0.1 (−97.9 %)

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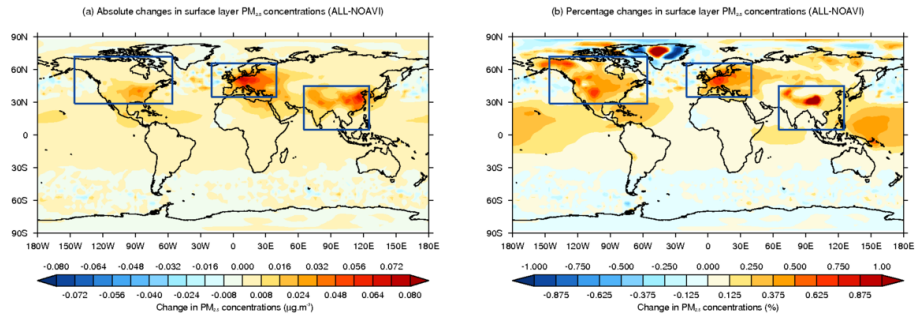

Figure 1. Impact of aviation emissions (FSC = 600 ppm) on surface annual mean $PM_{2.5}$ concentrations. **(a)** absolute (NORM–NOAVI) and **(b)** percentage changes. Boxes show the European (20–40° E, 35–66° N) and North American (146–56° W, 29–72° N) regions.

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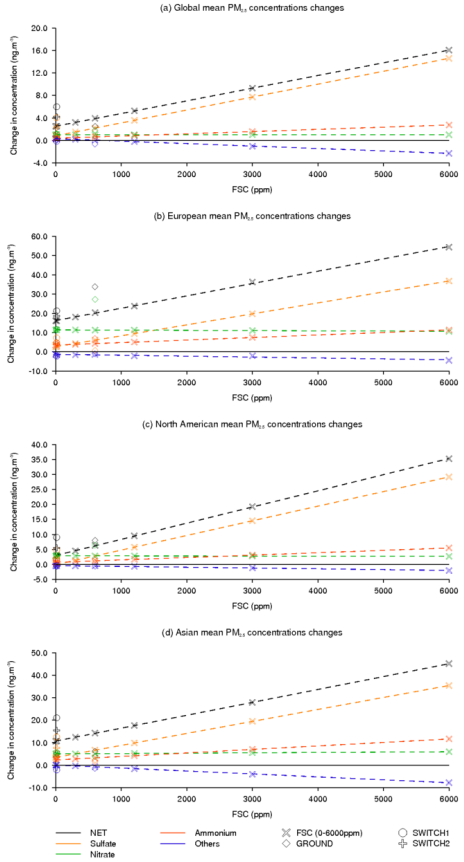

Figure 2. Impact of aviation FSC on **(a)** global, **(b)** European (20–40° E, 35–66° N), **(c)** North American (146–56° W, 29–72° N) surface annual mean $PM_{2.5}$ mass concentrations: FSC variations (X), GROUND (◊), SWITCH1 (−), and SWITCH2 (+) simulations. Dashed lines demonstrate the linear relationship between FSC and $PM_{2.5}$.

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Impacts of aviation fuel sulfur content on climate and human health

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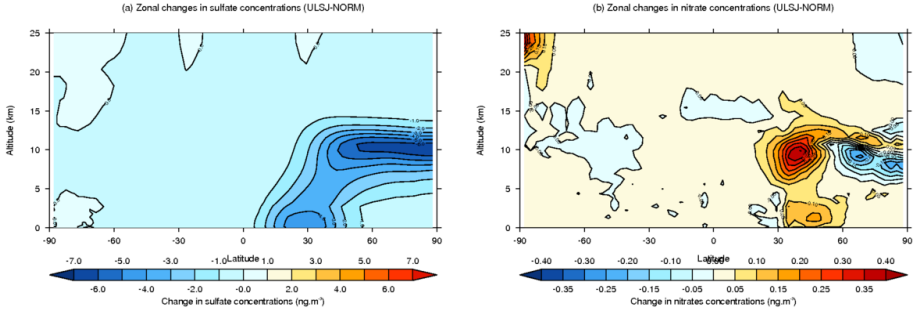

Figure 3. Simulated differences in zonal annual mean sulfate **(a)** and nitrate **(b)** concentrations from the use of ULSJ fuel relative to standard fuel (ULSJ–NORM).

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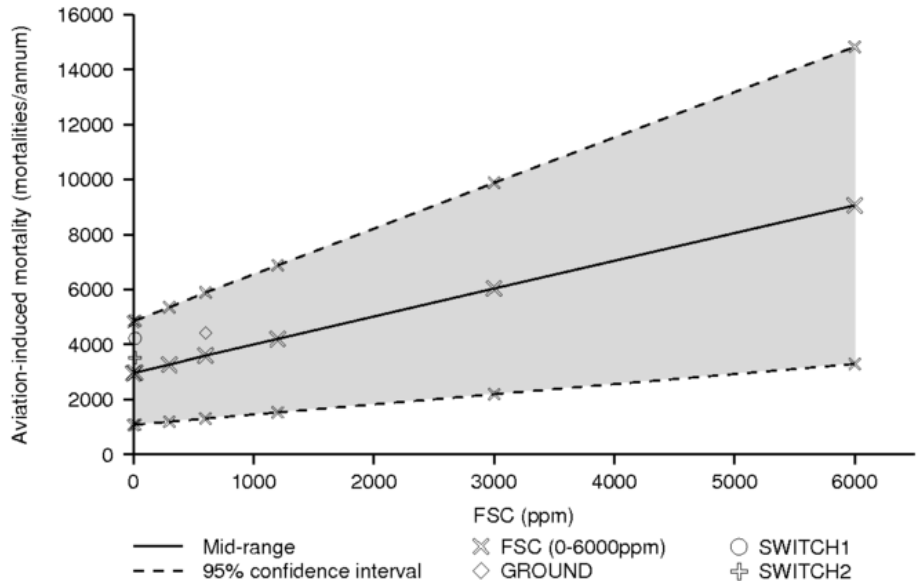

Figure 4. Estimated global aviation-induced mortality as a function of FSC, and changes in vertical aviation-emissions distributions for year 2000 (Shaded region denotes the 95 % confidence through application of low- and high-range cause-specific coefficients).

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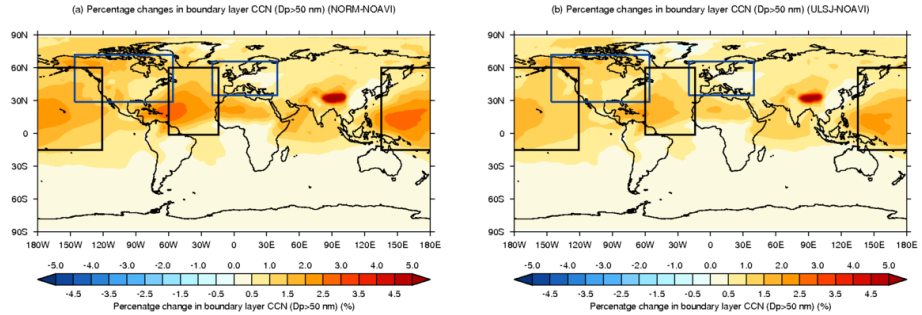

Figure 5. Impact of aviation emissions on low-cloud level (879 hPa) CCN ($D_p > 50$ nm) concentrations: **(a)** standard FSC (NORM–NOAVI) and **(b)** FSC = 15 ppm (ULSJ–NOAVI). Blue boxes define North American and European regions, and black boxes define Atlantic (60–14° W, 1.4° S–60° N) and Pacific regions (135° E–121° W, 15° S–60° N) referred to in the text.

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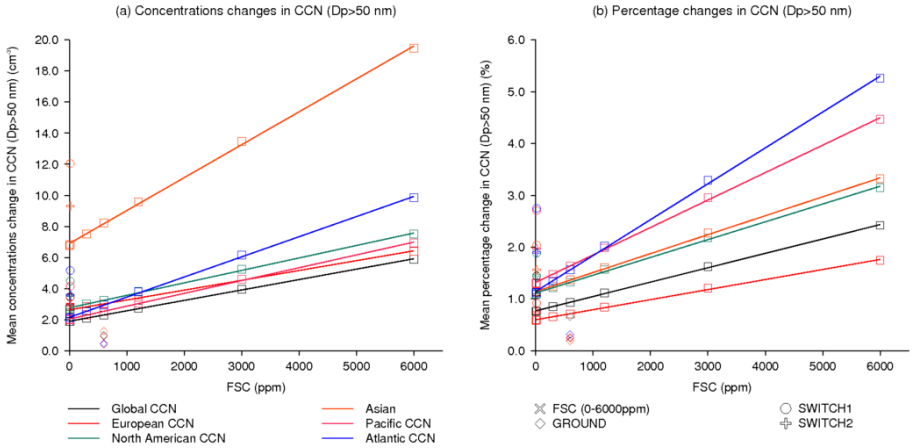

Figure 6. Global and regional variations in low-cloud level (879 hPa) CCN ($D_p > 50$ nm): **(a)** changes in mean concentrations and **(b)** percentage changes. See Fig. 5 for definitions of regions.

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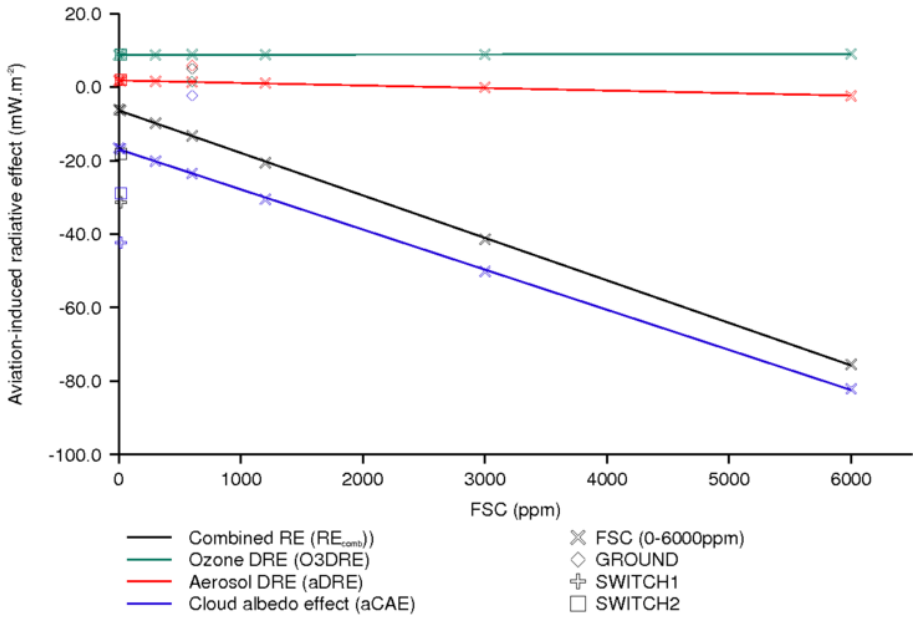

Figure 7. Aviation-induced radiative effects due to variations in fuel sulfur content (FSC), the ground release of aviation emissions (GROUND), and variations in the vertical distribution of aviation SO_2 emissions (SWITCH1 and SWITCH2 simulations).

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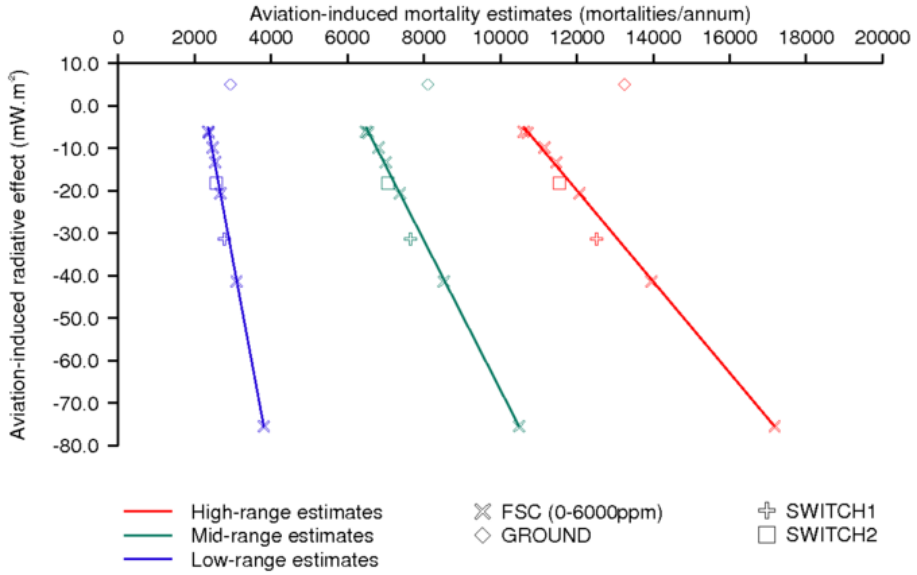

Figure 8. Relationship between net radiative effect [sum of ozone direct (O3DRE), aerosol direct radiative (aDRE) and aerosol cloud albedo (aCAE) effects] and annual mortality rates: for low- mid- and high-range mortality sensitivities.

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