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Impacts of aviation fuel sulfur content on climate and human health

Zarashpe Z. Kapadia[1,2], Dominick V. Spracklen[2], Steve R. Arnold[2], Duncan J. Borman[3], Graham W. Mann[2,4], Kirsty J. Pringle[2], Sarah A. Monks[2,a,b], Carly L. Reddington[2], François Benduhn[5], Alexandru Rap[2], Catherine E. Scott[2], Edward W. Butt[2], and Masaru Yoshioka[2]

[1]Doctoral Training Centre in Low Carbon Technologies, Energy Research Institute, School of Process Environmental and Materials Engineering, University of Leeds, Leeds, UK
[2]Institute for Climate and Atmospheric Science, School of Earth and Environment, University of Leeds, Leeds, UK
[3]Centre for Computational Fluid Dynamics, School of Civil Engineering, University of Leeds, Leeds, UK
[4]National Centre for Atmospheric Science, School of Earth and Environment, University of Leeds, Leeds, UK
[5]Institute for Advanced Sustainability Studies, Potsdam, Germany
[a]now at: Cooperative Institute for Research in Environmental Sciences, University of Colorado, Boulder, Colorado, USA
[b]now at: Chemical Sciences Division, NOAA Earth System Research Laboratory, Boulder, Colorado, USA

Correspondence to: Zarashpe Z. Kapadia (pm08zzk@leeds.ac.uk)

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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 ~ 3600 [95 % CI: 1310–5890] annual premature 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 ~ 620 [95 % CI: 230–1020] mortalities a^{-1} and increase RE_{comb} by $+7.0\,mW\,m^{-2}$.

We explore the impact of varying aviation FSC between 0 and 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.

1 Introduction

Aviation is the fastest growing form of transport (Eyring et al., 2010; Lee et al., 2010; Uherek et al., 2010), with a projected growth in passenger air traffic of 5 % yr^{-1} until 2030 (Barrett et al., 2012; ICAO, 2013), and a projected near doubling of emissions by 2025, relative to 2005 (Eyers et al., 2004). These emissions, and changes to them, have both cli-

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mate and air quality impacts (Lee et al., 2009; Barrett et al., 2010, 2012; Woody et al., 2011).

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 (Holmes et al., 2011; Myhre et al., 2011). Sulfate and nitrate aerosols, formed from aviation sulfur dioxide (SO_2) and NO_x emissions and through altered atmospheric oxidants, lead to a cooling (Unger, 2011; Righi et al., 2013; Dessens et al., 2014), 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 (Lee et al., 2010; Rap 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 $+55\,\text{mW}\,\text{m}^{-2}$ (excluding cirrus cloud enhancement) (Lee et al., 2010).

Previous studies have separately assessed the impacts of aviation through different atmospheric species. Short-term O_3 has been estimated to have a radiative effect ranging between 6 and $36.5\,\text{mW}\,\text{m}^{-2}$ (Sausen et al., 2005; Köhler et al., 2008; Hoor et al., 2009; Lee et al., 2009; Holmes et al., 2011; Myhre et al., 2011; Unger, 2011; Frömming et al., 2012; Skowron et al., 2013; Unger et al., 2013; Khodayari et al., 2014; Brasseur et al., 2016). The aerosol direct effect is highly uncertain [-28 to $+20\,\text{mW}\,\text{m}^{-2}$] (Righi et al., 2013), with the direct aerosol effects for sulfate ranging between -0.9 and $-7\,\text{mW}\,\text{m}^{-2}$ (Sausen et al., 2005; Fuglestvedt et al., 2008; Lee et al., 2009; Balkanski et al., 2010; Unger, 2011; Gettelman and Chen, 2013; Brasseur et al., 2016), nitrate ranging between -4 and $-7\,\text{mW}\,\text{m}^{-2}$ (Unger et al., 2013; Brasseur et al., 2016), BC ranging between 0.1 and $0.3\,\text{mW}\,\text{m}^{-2}$ (Sausen et al., 2005; Fuglestvedt et al., 2008; Lee et al., 2009; Balkanski et al., 2010; Unger, 2011; Gettelman and Chen, 2013; Unger et al., 2013; Brasseur et al., 2016), and for organic carbon (OC) ranging between -0.67 and $-0.01\,\text{mW}\,\text{m}^{-2}$ (Sausen et al., 2005; Fuglestvedt et al., 2008; Lee et al., 2009; Balkanski et al., 2010; Unger, 2011; Gettelman and Chen, 2013; Unger et al., 2013). Few studies estimate the aerosol cloud albedo effect (aCAE) from aviation: Righi et al. (2013) assessed the aCAE to be $-15.4 \pm 10.6\,\text{mW}\,\text{m}^{-2}$ while Gettelman and Chen (2013) estimate $-21 \pm 11\,\text{mW}\,\text{m}^{-2}$.

Aviation emissions can increase atmospheric concentrations of fine particulate matter with a dry diameter of $< 2.5\,\mu\text{m}$ ($PM_{2.5}$). Short-term exposure to $PM_{2.5}$ can exacerbate existing respiratory and cardiovascular ailments, while long-term exposure can result in chronic respiratory and cardiovascular diseases, lung cancer, chronic changes in physiological functions and mortality (Pope et al., 2002; World Health Organisation, 2003; Ostro, 2004). In the US aviation emissions are estimated to lead to adverse health effects in $\sim 11\,000$ people (ranging from mortality, respiratory ailments and hospital admissions due to exacerbated respiratory conditions) and $\sim 23\,000$ work loss days per annum (Ratliff et al., 2009). Landing and take-off aviation emissions increase $PM_{2.5}$ concentrations, particularly around airports (Woody et al., 2011), increasing US mortality rates by ~ 160 per annum.

Previous studies have estimated the number of premature mortalities due to exposure to pollution resulting from aviation emissions. Barrett et al. (2012, 2010) used the methodology of Ostro (2004) to estimate that aviation emissions are responsible for $\sim 10\,000$ premature mortalities a^{-1} due to increases in cases of cardiopulmonary disease and lung cancer. Yim et al. (2015) using the same methodology but with the inclusion of the Rapid Dispersion Code (RDC) to simulate the local air quality impacts of aircraft ground level emissions estimated 13 920 (95 % CI: 7220–20 880) mortalities a^{-1}. Morita et al. (2014), using the integrated exposure–response (IER) model from Burnett et al. (2014) to derive relative risk (RR), estimate that aviation results in 405 (95 % CI: 182–648) mortalities a^{-1} due to increases in cases of lung cancer, stroke, ischemic heart disease, trachea, bronchus, and chronic obstructive pulmonary disease. Jacobson et al. (2013) estimate 310 (95 % CI: -400 to 4300) mortalities a^{-1} from aviation emissions due to cardiovascular effects. Taking these studies in account, the different methodologies applied and modes of mortality investigated aviation is estimated to be responsible for between 310 and 13 920 mortalities a^{-1}.

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 $PM_{2.5}$ is the use of ultra-low sulfur jet fuel (ULSJ), reducing the formation of sulfate aerosol (Barrett et al., 2012, 2010; Ratliff et al., 2009; Hileman and Stratton, 2014). ULSJ fuels typically have a fuel sulfur content (FSC) of 15 ppm, compared with an FSC of between 550 and 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 (Ministry of Defence, 2011; ASTM International, 2012).

Despite the potential for decreased emission of SO_2, 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 desulfurized global aviation fleet would likely contribute a net source of sulfate $PM_{2.5}$. Nevertheless, previous work has shown that the use of ULSJ fuel reduces global aviation-induced $PM_{2.5}$ by $\sim 23\,\%$, annually avoiding ∼ 2300 (95 % CI: 890–4200) mortalities (Barrett et al., 2012).

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 (Unger, 2011; Barrett et al., 2012), increasing the net warming effect of aviation emissions. The roles of sulfate both in climate cooling and in increasing surface $PM_{2.5}$ 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 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, while the impacts of changes in surface $PM_{2.5}$ on human health are estimated using concentration response functions. Using a coupled tropospheric chemistry-aerosol microphysics model that includes nitrate aerosol allows us to assess the impacts of nitrate and aerosol indirect effects in addition to the ozone and aerosol direct effects that have been more routinely calculated.

2 Methods

2.1 Coupled chemistry-aerosol microphysics model

2.1.1 Model description

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). Meteorology (wind, temperature and humidity) and large-scale transport is specified from interpolation of 6-hourly European Centre for Medium Range Weather Forecasts (ECMWF) reanalysis (ERA-40) fields (Chipperfield, 2006; Mann et al., 2010). Cloud fraction and cloud top pressure fields are taken from the International Satellite Cloud Climatology Project (ISCCP-D2) archive for the year 2000 (Rossow and Schiffer, 1999).

GLOMAP-mode is a two-moment aerosol microphysics scheme representing particles as an external mixture of seven size modes (four soluble and three insoluble) (Mann et al., 2010). We use the nitrate-extended version of GLOMAP-mode (Benduhn et al., 2016) which, as well as tracking size-resolved sulfate, BC, 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 coagulation, 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 (Spracklen et al., 2005; Breider et al., 2010; Mann et al., 2010). The tropospheric chemistry is coupled to aerosol as described in Breider et al. (2010).

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

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

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

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 4 months discarded as spin-up time.

2.1.2 Model evaluation

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

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

Overall we find the model overestimates sulfates [NMB = +16.9 %], while underestimating nitrates [NMB = −60.7 %], ammonium [NMB = −47.1 %] and organic aerosols (OA) [NMB = −56.2 %]. Model skill varies depending on the conditions affecting each field campaign. To explore this, we use the broad stratification of the field campaigns into anthropogenic pollution, biomass burning and remote conditions as used by Heald et al. (2011) and shown in Fig. 1. The model underestimates aerosol concentrations in biomass burning regions [sulfate NMB = −14.9 %; nitrate NMB = −79.4 %; ammonium NMB = −68.7 %, and; OA NMB = −74.5 %]. The model performs better in polluted [sulfate NMB = +31.6 %; nitrate NMB = −56.2 %; ammonium NMB = −28.6 %, and; OA NMB = −40.9 %], and remote regions [sulfate NMB = +25.4 %; nitrate NMB = −6.4 %; ammonium NMB = −20.2 %, and; OA NMB = −41.5 %].

The overestimation of sulfate aerosol is likely due to the decline in anthropogenic SO_2 emissions in Europe and the US between 2000 and 2008 (Vestreng et al., 2007; Hand et al., 2012). An underestimation of OA has been reported previously (Heald et al., 2011; Spracklen et al., 2011b) and is likely due to an underestimate in SOA formation in the model. Whitburn et al. (2015) found biomass burning emissions of NH_3 may be underestimated which would affect a number of our comparisons.

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

Figure 2 presents simulated ozone concentration profiles in comparison to ozonesonde observations compiled by Tilmes et al. (2012). Observations were compiled from three networks, comprising 41 stations with continuous sampling from 1995 to 2011: (i) The World Ozone and Ultraviolet Data Center (WOUDC) (http://www.woudc.org/); (ii) the Global Monitoring Division (GMD), and (iii) The Southern Hemisphere ADditional OZonesondes (SHADOZ) (Tilmes et al., 2012).

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

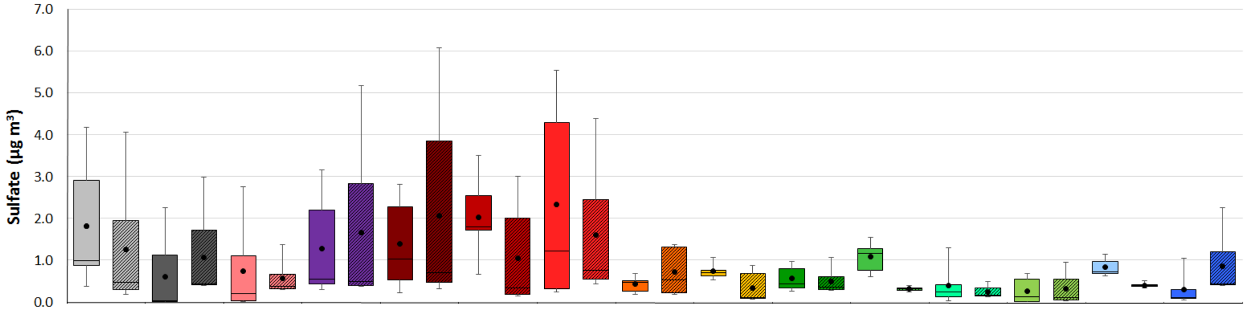
Evaluation of ozone model bias is conducted for the troposphere, using a chemical tropopause definition of 150 ppbv ozone, as previously used by Stevenson et al. (2013), Young et al. (2013) and Rap et al. (2015). We find the model overestimates global ozone concentrations [NMB = + 7.0 %] with overestimates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

Differences between model and observational profiles can in part be explained by the differences in years of simulation and observation, a poor representation of deep convection resulting in model underestimations in the tropics and overestimations downwind (Thompson et al., 1997), in tandem with reductions in anthropogenic NO_x emissions over this time period (Konovalov et al., 2008).

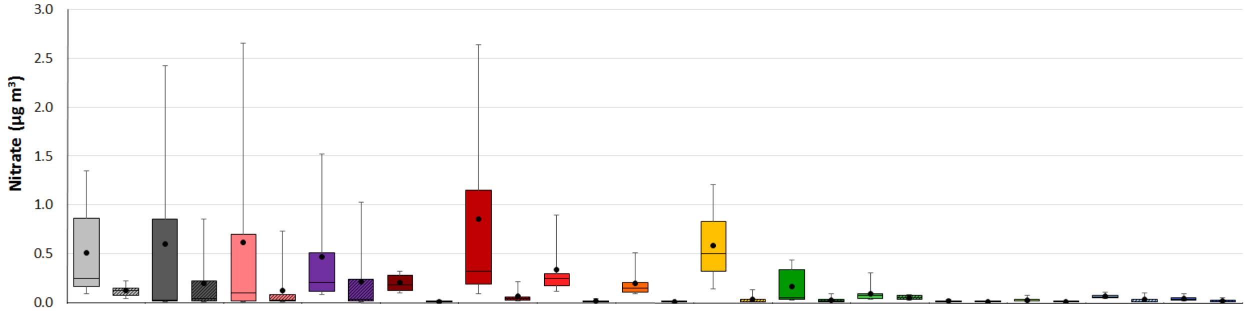
2.2 Aviation emissions

Aircraft emit NO_x, carbon monoxide (CO), SO_2, BC, OC and hydrocarbons (HCs). The historical emissions data set 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 (Eyers et

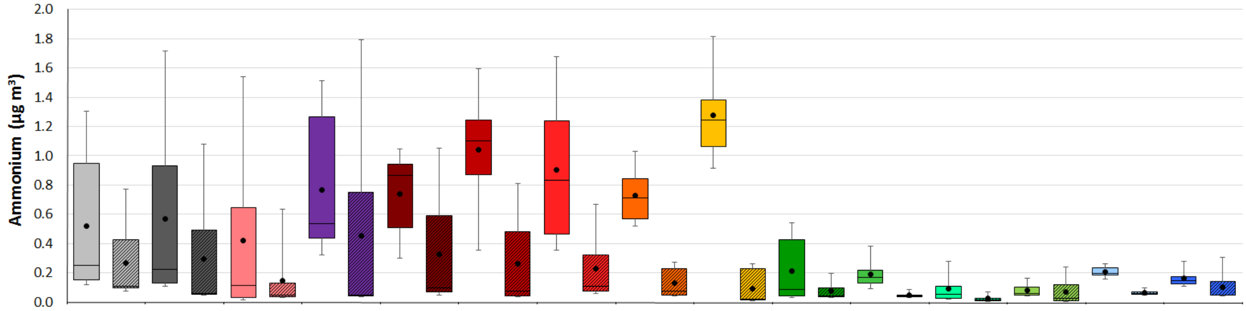
(a) Sulfate comparison



(b) Nitrate comparison



(c) Ammonium comparison



(d) Organic aerosol comparison

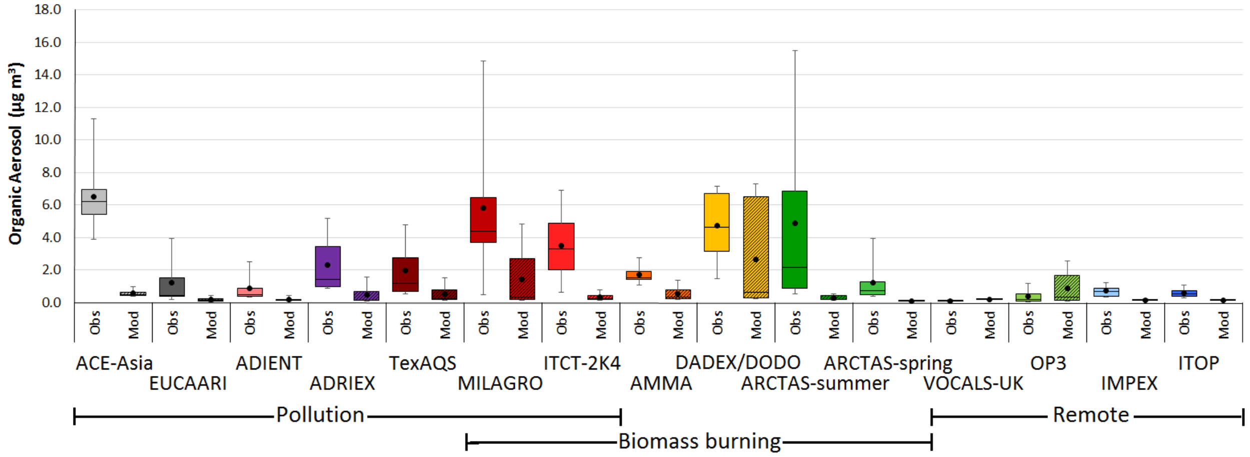


Figure 1. Comparison of observed (Obs) and simulated (Mod) **(a)** sulfate; **(b)** nitrate; **(c)** ammonium, and **(d)** organic aerosol mass concentrations. Observations are from airborne field campaigns compiled by Heald et al. (2011). Mean values are represented by black dots, median values as shown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

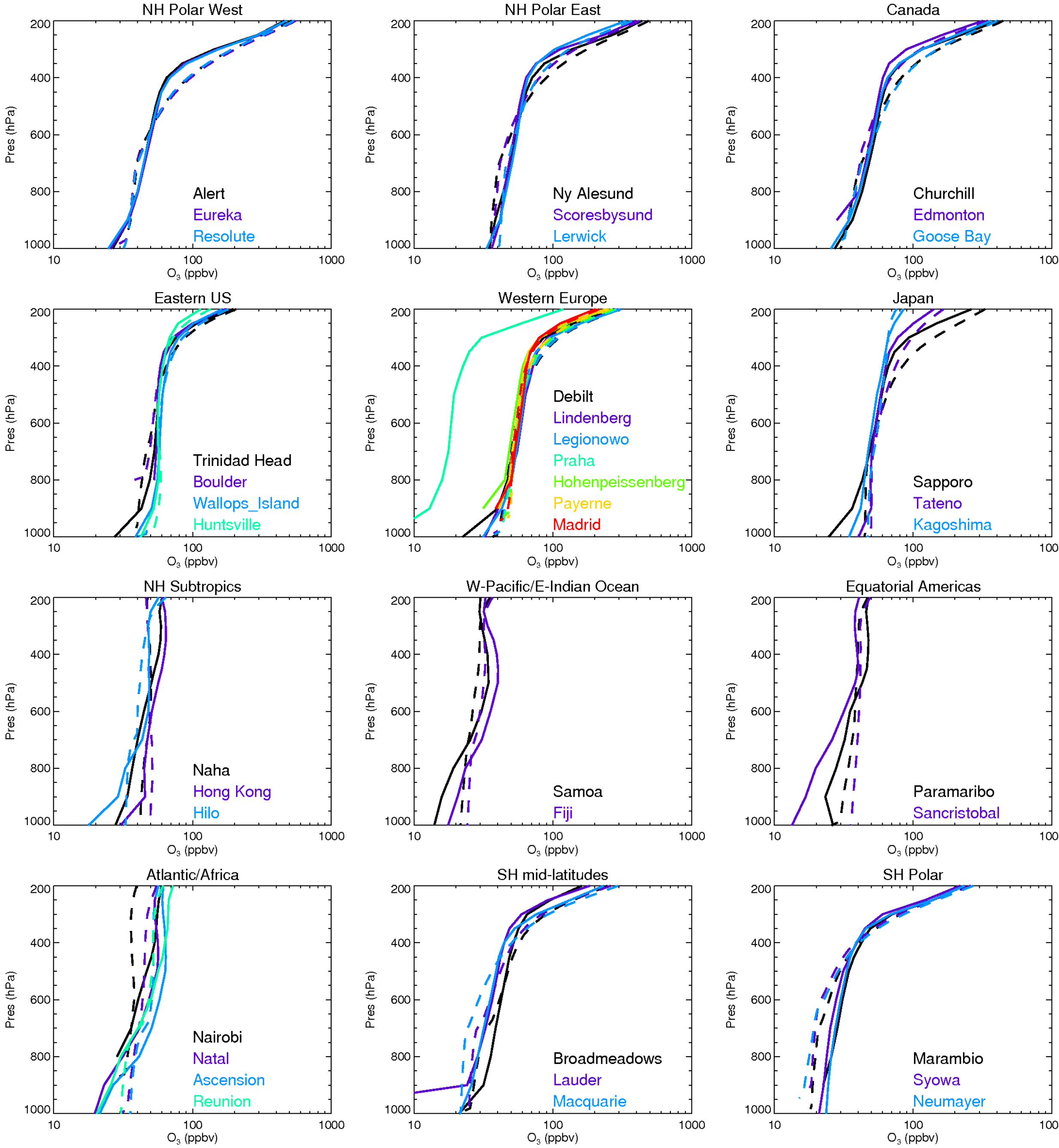


Figure 2. Comparison of observed (solid lines) and simulated (dashed lines) ozone profiles. Observations are taken from ozonesonde observations, and arranged by launch location regions according to Tilmes et al. (2012).

al., 2004; Quantify Integrated Project, 2005–2012; Wilkerson et al., 2010).

Here we develop a new 3-D civil aviation emissions data set for the year 2000, based on CMIP5 historical aviation emissions (Lamarque et al., 2009). The new data set includes emissions of NO_x, CO, SO_2, BC, OC, and HCs. In contrast to existing data sets which provide a general emissions index for HCs (Eyers et al., 2004) 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 data set. NO_x and BC emissions are taken directly from Lamarque et al. (2009). We calculate fuel burn from BC emissions data and the BC emissions index (Eyers et al., 2004) as used by Lamarque et al. (2009). Following DuBois and Paynter (2006), we assume

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,b,j,h,i,k,l]
CO	3.61[b]	0.724	0.507–0.679[b,h,i,j]
HCHO	1.24[c,d]	0.249	0.01205[b]
C_2H_6	0.0394[e]	0.007899	0.00051[b]
C_3H_8	0.03[e]	0.006014	0.00444[b]
CH_3OH	0.22[d]	0.044	0.00177[b]
CH_3CHO	0.33[d]	0.066	0.00418[b]
$(CH_3)_2CO$	0.18[d]	0.036	0.00036[b]
SO_2	1.1760[b]	0.236	0.182–0.221[a,b,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[b,i]

[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).

that BC emissions scale linearly with fuel consumption. We estimate emissions for other species using our calculated aviation fuel burn in combination with published species-specific emissions indices (EI reported in $g\,kg^{-1}$ of fuel). Emission indices for CO and SO_2 are 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); 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).

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 fuel burn considered by the base inventory used to develop our emissions inventory (Eyers et al., 2004; Lamarque et al., 2010). Our estimated OC emissions are lower than the emissions estimated in the AEDT 2006 inventory, due to the lower EI applied here. The lower EI_{OC} applied here (in comparison to Wilkerson et al., 2010) is a due to the phase of flight considered when deriving the AEDT emissions inventory; where they derive EI_{OC} focusing on airport operations at ground-level condition acknowledging the risk of overestimating aviation OC emissions, while in comparison we consider aircraft operations after ground idle conditions which risks underestimating aviation OC emissions.

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

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 (15 ppm) applied below the cruise phase of flight (< 8.54 km altitude) (Lee et al., 2009; Köhler et al., 2013) combined with a high FSC at altitudes above cruise level. 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 six bands in the shortwave (SW) and nine 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. (2011a), with the method for O3DRE as in Richards et al. (2013). To determine the aCAE we calculated cloud droplet number concentrations

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	Desulfurized 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

(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 $0.15\,\mathrm{m\,s^{-1}}$ over ocean and $0.3\,\mathrm{m\,s^{-1}}$ 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.

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). Using this function allows us to compare our premature mortality estimates with those from previous studies (Barrett et al., 2012; Yim et al., 2015) using the same concentration function; in future work estimates are required with updated methodologies (Burnett et al., 2014). $PM_{2.5}$ 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 (CRF) (Ostro, 2004). This response function considers concentrations of $PM_{2.5}$ for a perturbed case (X) (defined by aviation emissions scenarios from Table 2) in relation to a baseline case with no aviation emissions (X_0) (NOAVI). To calculate excess mortality, the relative risk (RR) for both cardiopulmonary disease and lung cancer is calculated according to Ostro (2004) using a function of baseline (X_0) and perturbed (X) $PM_{2.5}$ concentrations, and the disease-specific, cause-specific coefficient (β):

$$\mathrm{RR} = \left[\frac{(X+1)}{(X_0+1)}\right]^{\beta}. \quad (1)$$

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

$$\mathrm{AF} = (\mathrm{RR} - 1)/\mathrm{RR}. \quad (2)$$

Excess mortality (E) for both cardiopulmonary disease and lung cancer is calculated using baseline mortality rates (B), the fraction of the population over 30 years old (P_{30}), along with the AF:

$$E = \mathrm{AF} \times B \times P_{30}. \quad (3)$$

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

3 Results

3.1 Surface $PM_{2.5}$

Figure 3 shows the simulated impact of aviation emissions with standard FSC (FSC = 600 ppm; NORM) on surface $PM_{2.5}$ concentrations. Aviation increases annual mean $PM_{2.5}$ concentrations by up to $\sim 80\,\mathrm{ng\,m^{-3}}$ (relative to the NOAVI simulation) over Central Europe and Eastern China (Fig. 3a). Aviation emissions result in largest fractional changes in annual mean $PM_{2.5}$ concentrations (up to 0.8 %) over North America and Europe (Fig. 3b).

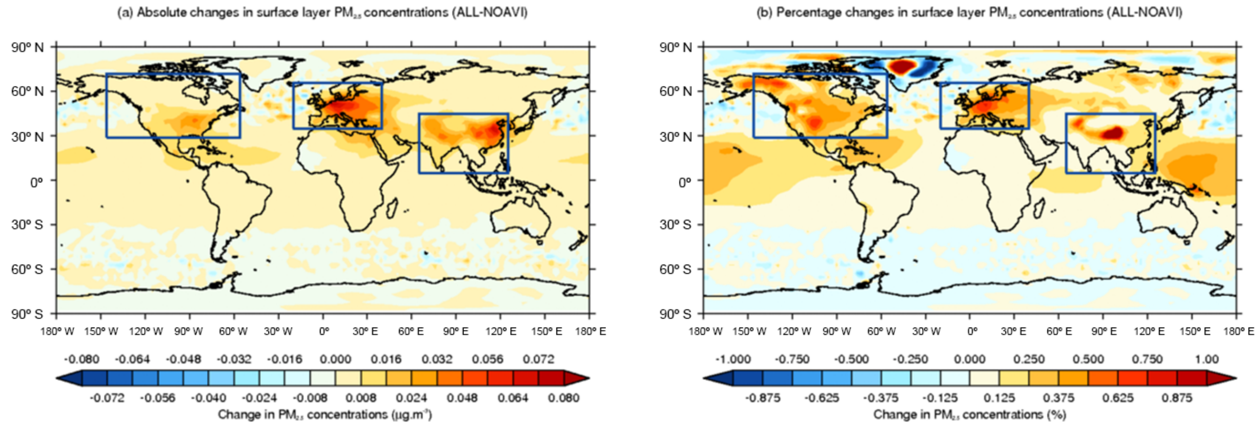


Figure 3. Impact of aviation emissions (FSC = 600 ppm) on surface annual mean $PM_{2.5}$ concentrations. **(a)** absolute (NORM–NOAVI) and **(b)** percentage changes. Boxes show the European (20–40° E, 35–66° N) and North American (146–56° W, 29–72° N) regions.

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 %)

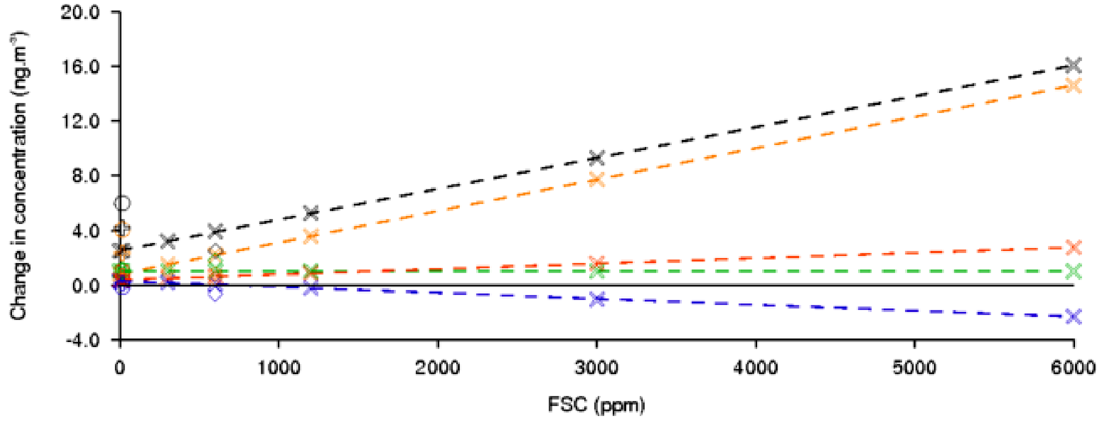
Figure 4 shows the impact of aviation emissions on global and regional mean $PM_{2.5}$ concentrations as a function of FSC. With standard FSC (FSC = 600 ppm), aviation increases global mean surface $PM_{2.5}$ concentrations by 3.9 ng m^{-3}; with increases in $PM_{2.5}$ dominated by sulfates [56.2 %], nitrates [26.0 %] and ammonium [16.0 %]. Aviation emissions increase European annual mean $PM_{2.5}$ concentrations by 20.3 ng m^{-3} (Fig. 4b), substantially more than over North America (Fig. 4c) 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 % [1.4 ng m^{-3}] (Fig. 4); predominantly due to changes in sulfate [−1.4 ng m^{-3}; −62.1 %] and ammonium [−0.2 ng m^{-3}; −37.9 %], which are marginally offset by very small increases in nitrates [$+3.2 \times 10^{-3}$ ng m^{-3}; +0.3 %]. Aviation emissions also lead to small changes to other aerosol components of +0.2 ng; which includes natural aerosols such as dust [+0.3 ng m^{-3}; +61.8 %], sodium [−19.5 %] and chloride from sea salt [−19.5 %] with the changes due to changes in aerosol lifetimes, along with changes in BC [−7.9 %] and OC [−19.3 %].

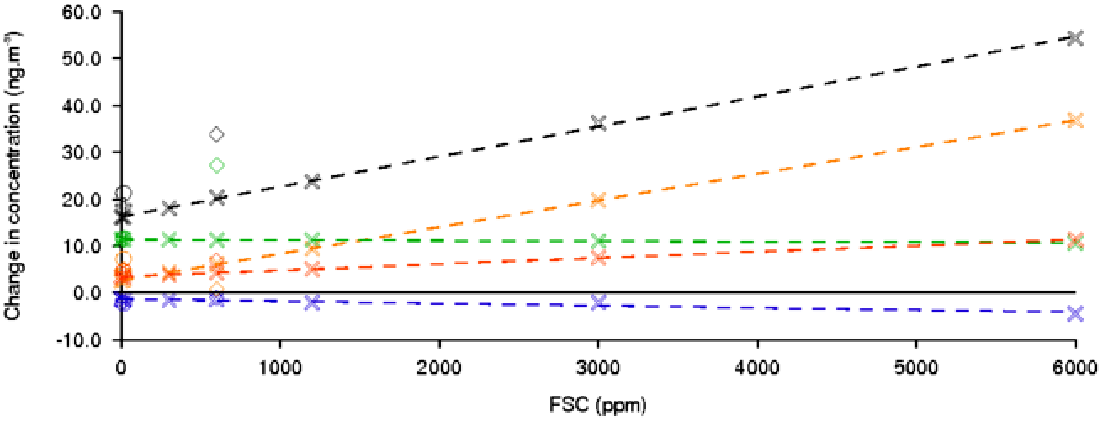
In comparison to the global mean, switching to the use of ULSJ fuel in aviation larger absolute reductions in $PM_{2.5}$ of −4.2 ng m^{-3} are simulated over Europe [Δsulfate = −3.4 ng m^{-3}; Δnitrate = +0.1 ng m^{-3}; Δammonium = −0.8 ng m^{-3}; and Δothers = −0.1 ng m^{-3}] and of −3.4 ng m^{-3} over North America [Δsulfate = −2.9 ng m^{-3}; Δnitrate = +0.02 ng m^{-3}; Δammonium = −0.5 ng m^{-3}; and Δothers = −0.01 ng m^{-3}] (Fig. 4b, c). Over North America, swapping to ULSJ fuel reduces aviation-induced $PM_{2.5}$ by 53.4 %, while a smaller reduction of 20.5 % 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 desulfurization 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 desul-

(a) Global mean $PM_{2.5}$ concentrations changes



(b) European mean $PM_{2.5}$ concentrations changes



(c) North American mean $PM_{2.5}$ concentrations changes

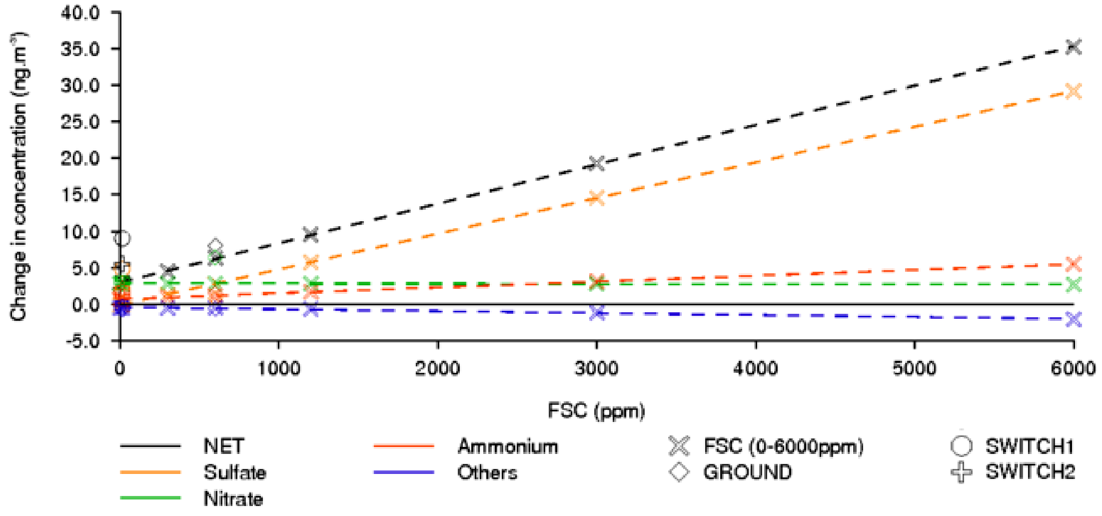


Figure 4. Impact of aviation FSC on **(a)** global, **(b)** European (20–40° E, 35–66° N), **(c)** North American (146–56° W, 29–72° N) surface annual mean $PM_{2.5}$ mass concentrations: FSC variations (×), GROUND (◇), SWITCH1 (−), and SWITCH2 (+) simulations. Solid lines demonstrate the linear relationship between FSC and $PM_{2.5}$.

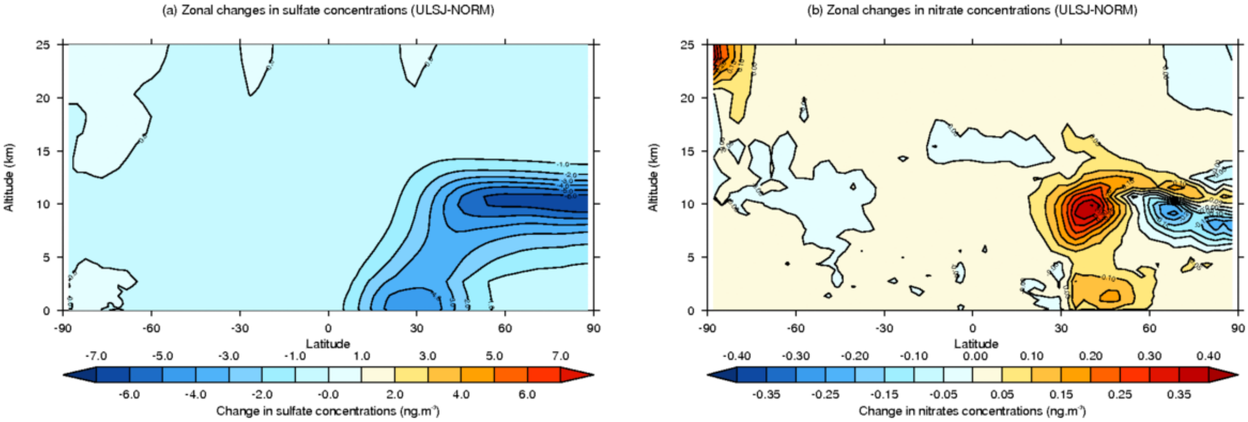


Figure 5. Simulated differences in zonal annual mean sulfate **(a)** and nitrate **(b)** concentrations from the use of ULSJ fuel relative to standard fuel (ULSJ–NORM).

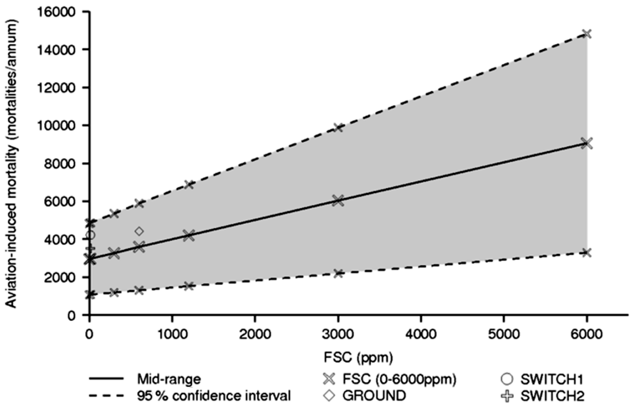


Figure 6. Estimated global aviation-induced mortality as a function of FSC, and changes in vertical aviation emission distributions for year 2000 (Shaded region denotes the 95 % confidence through application of low- and high-range cause-specific coefficients).

furization, 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, nitrate, ammonium and total $PM_{2.5}$ are linear ($R^2 > 0.99$, p value < 0.001 globally and for all individual regions) with respect to FSC (Fig. 4). Larger emission perturbations would likely lead to a non-linear response in atmospheric aerosol. 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 5 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 desulfurization 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 oxidation of SO_2 from non-aviation sources (Unger et al., 2006; Barrett et al., 2010).

In line with previous work, we find that a substantial fraction of aviation sulfate can be attributed to aviation NO_x emissions and not directly to aviation SO_2 emissions. We estimate that 36 % aviation-attributable sulfates formed at the surface are associated with aviation NO_x emissions, compared to ~ 63 % estimated by Barrett et al. (2010) using the GEOS-Chem model (both estimates for FSC = 600 ppm). Differences between model estimates can be attributed to differences in model chemistry and microphysics, and different aviation NO_x emissions. We find desulfurization increases the aviation nitrate burden by 5.1 % (Table 3); although much of this increase occurs at altitudes well above the surface (Fig. 5) and so is not reflected in surface $PM_{2.5}$ concentrations.

We explored the impacts of NO_x emission reductions in combination with fuel desulfurization. A scenario with desulfurized fuel and zero NO_x emissions reduces the global aviation-induced aerosol burden by 88.3 % (Table 3), in comparison to a desulfurized only case (DESUL), where the aviation-induced aerosol burden is reduced by 28.4 %. Removal of aviation NO_x and SO_2 emissions results in a 95.0 % reduction in aviation-induced global mean surface level aviation-induced $PM_{2.5}$. These results imply that only limited sulfate reductions can be achieved through reducing FSC alone, with further reductions in aviation-induced $PM_{2.5}$ sulfates requiring additional controls on aviation NO_x emissions.

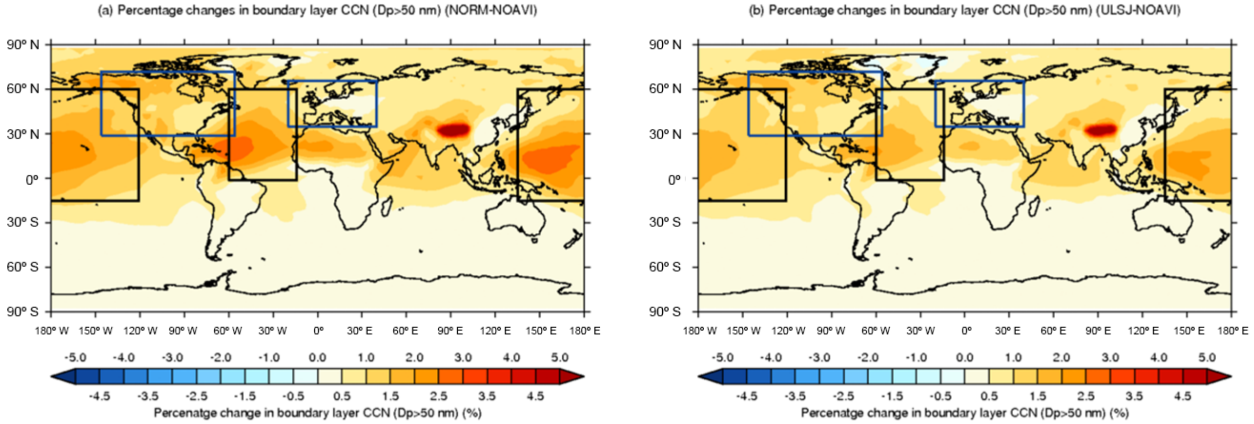


Figure 7. Impact of aviation emissions on low-cloud level (879 hPa) CCN ($D_\mathrm{p} > 50$ nm) concentrations: **(a)** standard FSC (NORM–NOAVI) and **(b)** FSC = 15 ppm (ULSJ–NOAVI). Blue boxes define North American and European regions, and black boxes define Atlantic (60–14° W, 1.4° S–60° N) and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

3.2 Premature mortality

Figure 6 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 3600 [95 % CI: 1310–5890] premature mortalities each year, with 3210 [95 % CI: 1160–5250] mortalities a^{-1} due to increases in cases of cardiopulmonary disease and 390 [95 % CI: 150–640] mortalities a^{-1} due to increases in cases of lung cancer. Low-, mid- and high-range cause-specific coefficients (β) are used to account for uncertainty in the health impacts caused by exposure to $PM_{2.5}$ (Sect. 2.5) (Ostro, 2004). 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.

Our estimate of the premature mortality due to aviation lies within the range of previous estimates (310–13 920 mortalities a^{-1}) (Barrett et al., 2010, 2012; Jacobson et al., 2013; Morita et al., 2014; Yim et al., 2004). Barrett et al. (2012) estimated ∼ 10 000 mortalities a^{-1} due to aviation, almost a factor of 3 higher than our central estimate. The 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 maximum 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. Additionally, differences in mortality arise due to the use of different cause-specific coefficients (β) within the same CRF, as well as different population data sets. Morita et al. (2014) estimate that aviation is responsible for 405 [95 % CI: 182–648] mortalities a^{-1}. This lower estimate is primarily due to the mortality functions used, with Morita et al. (2014) using the integrated exposure response (IER) function as described by Burnett et al. (2014). The IER function considers a $PM_{2.5}$ concentration below which there is no perceived risk, reducing estimated impacts of aviation in regions of low $PM_{2.5}$ concentrations.

We estimate that aviation emissions with ULSJ fuel result in 2970 [95 % CI: 1080–4870] premature mortalities globally per annum. Therefore, changing from standard FSC to ULSJ would result in 620 [95 % CI: 230–1020] 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 ∼ 2300 [95 % CI: 890–4200] fewer premature mortalities globally per annum; a reduction of 23 %. In their work (using GEOS-Chem), the use of ULSJ reduces global mean $PM_{2.5}$ concentrations (sulfates, nitrates and ammonium) by 0.89 ng m^{-3}, less than the 1.61 ng m^{-3} reduction in $PM_{2.5}$ simulated here). 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. Additionally, the GRUMPv1 population data set that Barrett et al. (2012) use resolves population data on a finer scale compared to the resolution of GPWv3 population data set used here (Center for International Earth Science Information Network, 2012); differences which could contribute to differences in estimates of mortality.

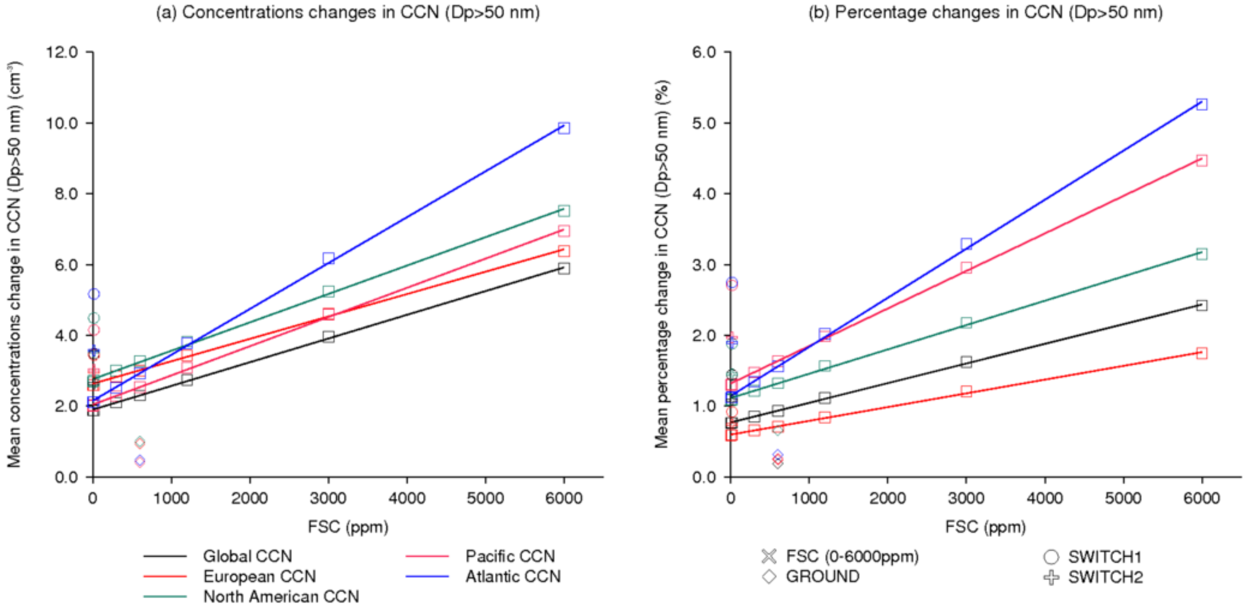


Figure 8. Global and regional variations in low-cloud level (879 hPa) CCN ($D_p > 50$ nm): **(a)** changes in mean concentrations and **(b)** percentage changes. See Fig. 5 for definitions of regions.

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 6030, 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. 7a). 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. 7). Northern Hemisphere 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. 7).

Figure 8 shows the sensitivity of low level CCN concentrations to FSC. As with $PM_{2.5}$, we find simulated changes in CCN are near linear with respect to FSC ($R^2 > 0.99$ 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 desulfurization of aviation fuel results in reductions in CCN in relation to standard aviation (DESUL–NORM), which follow the same regional trends (Fig. 8a).

3.4 Sensitivity of aerosol and ozone radiative effect to FSC

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. 9).

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 and +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 −21 ± 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}] (Sausen et al., 2005; Köhler et al., 2008; Hoor et al., 2009; Lee et al., 2009; Holmes et

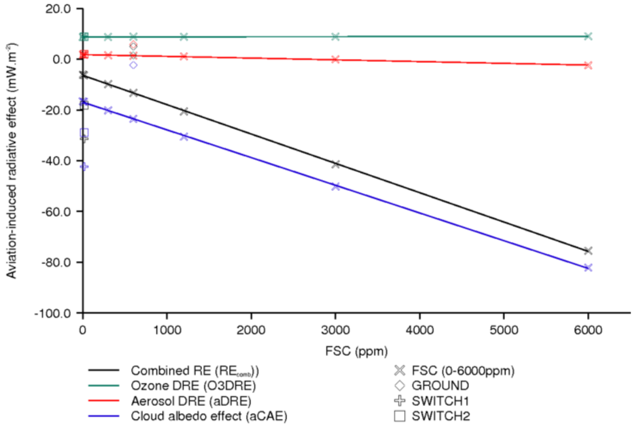


Figure 9. Aviation-induced radiative effects due to variations in fuel sulfur content (FSC), the ground release of aviation emissions (GROUND), and variations in the vertical distribution of aviation SO_2 emissions (SWITCH1 and SWITCH2 simulations).

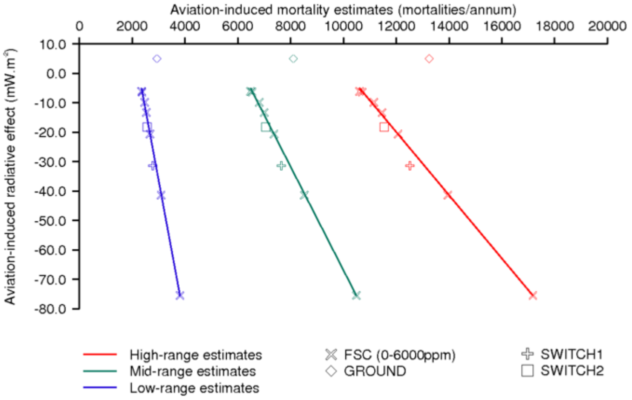


Figure 10. Relationship between net radiative effect (sum of ozone direct (O3DRE), aerosol direct radiative (aDRE) and aerosol cloud albedo (aCAE) effects) and annual mortality rates: for low- mid- and high-range mortality sensitivities.

al., 2011; Myhre et al., 2011; Unger, 2011; Frömming et al., 2012; Skowron et al., 2013; Unger et al., 2013; Khodayari et al., 2014). 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 $7.4\,\mathrm{mW\,m^{-2}\,Tg(N)^{-1}}$ 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 $16.2\text{–}25.4\,\mathrm{mW\,m^{-2}\,Tg(N)^{-1}}$.

We calculate that an aviation fleet utilising ULSJ fuel would result in a global annual mean $\mathrm{RE_{comb}}$ of $-6.3\,\mathrm{mW\,m^{-2}}$ [$\mathrm{aDRE} = +1.8\,\mathrm{mW\,m^{-2}}$; $\mathrm{aCAE} = -16.8\,\mathrm{mW\,m^{-2}}$; and $\mathrm{O3DRE} = +8.7\,\mathrm{mW\,m^{-2}}$]. Thus, swapping from standard aviation fuel to ULSJ fuel reduces the net cooling effect from aviation-induced aerosol and O_3 by $7.0\,\mathrm{mW\,m^{-2}}$, in comparison to the reduction of $3.3\,\mathrm{mW\,m^{-2}}$ estimated by Barrett et al. (2012). In our model, this change is primarily due a reduction in cooling from the aCAE of $+6.7\,\mathrm{mW\,m^{-2}}$ combined with smaller contributions from an increased aDRE of $+0.4\,\mathrm{mW\,m^{-2}}$, and reduction in warming from the O3DRE of $-0.12\,\mathrm{mW\,m^{-2}}$ (Fig. 9).

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

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 ($\mathrm{FSC} > 1200$ 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 $\mathrm{RE_{comb}}$ is dominated by these changes to the aCAE. Increases in FSC from 0 to 6000 ppm result in a greater negative (cooling) aviation-induced $\mathrm{RE_{comb}}$; increasing in magnitude by a factor of ~ 5 ($-16.6\,\mathrm{mW\,m^{-2}}$ for $\mathrm{FSC} = 0$ ppm to $-82.1\,\mathrm{mW\,m^{-2}}$ for $\mathrm{FSC} = 6000$ 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_2 emissions

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

The different slopes in the relationship between estimated RE and mortality (Fig. 10) are driven by the range of coefficients used in the CRF. This highlights the considerable uncertainty in the health impacts caused by exposure to $PM_{2.5}$. We note that uncertainty in the RE due to aerosol and ozone exists, but is not included in Fig. 9.

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 net RE deviate from the linear relationship seen when varying FSC between 0 and 6000 ppm (Fig. 10).

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 $+13.5\,\mathrm{ng\,m^{-3}}$ [+65.7 %] over Europe and by $+1.7\,\mathrm{ng\,m^{-3}}$ [+27.1 %] over North America, but decrease by $-1.4\,\mathrm{ng\,m^{-3}}$ [−36.7 %] globally (Fig. 4). Greater surface layer $PM_{2.5}$ perturbations (GROUND–NORM) over populated regions increase aviation-induced annual mortality by +22.9 % [+830 mortalities $\mathrm{a^{-1}}$] (Fig. 6).

Releasing aviation emissions at the surface (GROUND case) increases global mean cloud level CCN by only $0.4\,\mathrm{cm^{-3}}$ relative to NOAVI; providing a reduction in CCN of 82.1 % [$-1.89\,\mathrm{cm^{-3}}$] 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 more efficient at increasing CCN concentrations compared to when the same emissions are released at the surface [GROUND CCN = $0.4\,\mathrm{cm^{-3}}$; NORM CCN = $2.3\,\mathrm{cm^{-3}}$]; both in relation to the NOAVI scenario. Similar behaviour has been demonstrated previously for volcanic SO_2 emissions by 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 $-2.3\,\mathrm{mW\,m^{-2}}$; 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 $+5.9\,\mathrm{mW\,m^{-2}}$, resulting in an RE_{comb} of $+5.0\,\mathrm{mW\,m^{-2}}$ (Fig. 9).

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 0.03 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 $+8.9\,\mathrm{mW\,m^{-2}}$ (NORM; FSC = 600 ppm) to $+1.5\,\mathrm{mW\,m^{-2}}$ (GROUND; FSC = 600 ppm) (Fig. 9). 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 hydrocarbon) concentrations (Köhler et al., 2008; Stevenson and Derwent, 2009; Snijders and Melkers, 2011; Skowron et al., 2013).

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 $+2.1\,\mathrm{ng\,m^{-3}}$ [52.2 %], European mean concentrations by $+0.9\,\mathrm{ng\,m^{-3}}$ [+4.5 %], and North American concentrations by $+2.7\,\mathrm{ng\,m^{-3}}$ [+42.2 %] relative to NORM (Fig. 4). These changes increase aviation-induced mortality by +17.4 % [+630 mortalities $\mathrm{a^{-1}}$] (Fig. 6). This scenario results in greater global mean increases in CCN (relative to NORM) of $+1.2\,\mathrm{cm^{-3}}$ [+51.2 %], a larger cooling aCAE [$-42.4\,\mathrm{mW\,m^{-2}}$], larger warming aDRE [$2.07\,\mathrm{mW\,m^{-2}}$], resulting in additional $-18.1\,\mathrm{mW\,m^{-2}}$ [136 %] of aviation-induced cooling [SWITCH1 RE_{comb} of $-31.4\,\mathrm{mW\,m^{-2}}$].

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 $+0.3\,\mathrm{ng\,m^{-3}}$ [+6.6 %], but reduces mean surface $PM_{2.5}$ concentrations over Europe [$-1.8\,\mathrm{ng\,m^{-3}}$; −8.7 %] and North America [$-0.8\,\mathrm{ng\,m^{-3}}$; −12.8 %] compared to NORM. Under this scenario global aviation-induced mortality is decreased by 2.4 % [−90 mortalities $\mathrm{a^{-1}}$] compared to the standard aviation simulation (Fig. 6). The SWITCH2 scenario results in a RE_{comb} of $-18.2\,\mathrm{mW\,m^{-2}}$, providing an additional $-4.9\,\mathrm{mW\,m^{-2}}$ [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.

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 (Sausen et al., 2005; Lee et al., 2009; Gettelman and Chen, 2013; Righi et al., 2013; Unger 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 $-13.3\,\mathrm{mW\,m^{-2}}$, due to a combination of O3DRE [$+8.9\,\mathrm{mW\,m^{-2}}$], aDRE [$+1.4\,\mathrm{mW\,m^{-2}}$] and aCAE [$-23.6\,\mathrm{mW\,m^{-2}}$].

Our O3DRE [$+8.9\,\mathrm{mW\,m^{-2}}$] when normalised to represent the impact of the emissions of 1 Tg(N) [$+10.45\,\mathrm{mW\,m^{-2}\,Tg(N)^{-1}}$] is at the lower end of the range provided by previous studies [7.39–36.95 $\mathrm{mW\,m^{-2}\,Tg(N)^{-1}}$] (Sausen et al., 2005; Hoor et al., 2009; Lee et al., 2009; Holmes et al., 2011; Myhre et al., 2011; Unger, 2011; Frömming et al., 2012; 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 [$-23.6\,\mathrm{mW\,m^{-2}}$] lies just outside the range provided by Gettelman and Chen (2013) and Righi et al. (2013) [-15.4 to $-21\,\mathrm{mW\,m^{-2}}$]. Our estimated aDRE [$+1.4\,\mathrm{mW\,m^{-2}}$] lies within the middle of the range given by previous work (Sausen et al., 2005; Fuglestvedt et al., 2008; Lee et al., 2009; Balkanski et al., 2010; Unger, 2011; Gettelman and Chen, 2013; Righi et al., 2013; Unger et al., 2013).

We estimate that standard aviation (NORM; FSC = 600 ppm) is responsible for approximately 3600 premature mortalities annually due to increased surface layer $\mathrm{PM_{2.5}}$, 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 $\mathrm{PM_{2.5}}$ 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 % [0.04–0.18 %] of annual premature mortalities.

We investigated the impact of varying aviation FSC over the range 0–6000 ppm. Increases in FSC lead to increases in surface $\mathrm{PM_{2.5}}$ concentrations and subsequent increases in aviation-induced mortality. Increases in FSC also lead to a more negative $\mathrm{RE_{comb}}$ due to enhanced aCAEs. We estimate that the use of ultra-low sulfur jet (ULSJ) fuel, with a FSC of 15 ppm, could prevent 620 [230–1020] mortalities annually compared to standard aviation emissions. Swapping to ULSJ fuel increases the global mean net RE by $+7.0\,\mathrm{mW\,m^{-2}}$ 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 $\mathrm{PM_{2.5}}$. We estimate that aviation-NO_x emissions are responsible for 36.2 % of aviation-induced sulfate perturbations. Thus further reductions in aviation-induced $\mathrm{PM_{2.5}}$ 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 $\mathrm{O_3}$-forming species are emitted results in a reduction in aviation-induced $\mathrm{O_3}$, 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 $\mathrm{PM_{2.5}}$ (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 of 5 less efficient at creating CCN when released at the surface, resulting in an aCAE of $-2.3\,\mathrm{mW\,m^{-2}}$, a reduction of 90.1 % in relation to the standard aviation scenario. When aviation $\mathrm{SO_2}$ emissions are injected into the free-troposphere, the dominant oxidation pathway is to $\mathrm{H_2SO_4}$ followed by particle formation and condensational growth of new particles to larger sizes. Subsequent entrainment of these new particles into the lower atmosphere leads to increased CCN concentrations and impacts on cloud albedo. Aviation $\mathrm{SO_2}$ 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 % [-90 mortalities $\mathrm{a^{-1}}$] and the magnitude of the negative $\mathrm{RE_{comb}}$ was increased by 36.6 %, providing an additional cooling impact of climate of $-4.88\,\mathrm{mW\,m^{-2}}$.

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 desulfurization 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. Future work needs to estimate the health impacts of aviation using newly available concentration response functions (Burnett et al., 2014).

5 Data availability

Ozone observations used in this work can be acquired from Tilmes et al. (2012). Aerosol data are described in

Heald et al. (2011). CMIP5 aviation emissions are available from http://tntcat.iiasa.ac.at:8787/RcpDb/dsd?Action=htmlpage&page=about. Model outputs are available on request from the corresponding author.

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