This document contains the following for "Impacts of aviation fuel sulfur content on climate and human health" by Z. Z. Kapadia et al.:

a) **Replies to first set of reviewer's comments, and**

b) **Replies to minor comments thereafter.**

The aforementioned is then followed by a marked-up version of the manuscript, with replies to first set of reviewer's comments marked-up in yellow and replies to minor comments marked-up in green.

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

a) Replies to reviewer's comments on "Impacts of aviation fuel sulfur content on climate and human health" by Z. Z. Kapadia et al.

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We would like to thank both of the anonymous reviewers for their helpful and constructive comments.

Reviewer	Comment number		Comment text	How the comment has been addressed/response
1	1	a	***Overall the work appears to be carefully executed. The paper would benefit from a better articulation of how this treatment differs from published previous work (including better explanation of different results).***	We have added a more detailed description of how our work differs from previous studies. See our responses to points 1b and 4 below (from reviewer 1), and point 2 from reviewer 2. We have explicitly mentioned the current broad range of literature estimates in the aerosol direct radiative and aerosol cloud albedo effects (section 1, pp.2, paragraph 3), and how our work is used to re-evaluate these radiative effects using a coupled tropospheric chemistry-aerosol microphysics (section 1, pp.3, paragraph 5). The investigation of the use of desulfurised fuel, ULSJ fuel and variations in FSC above the cruise phase of flight are now explicitly mentioned to identify how this work differs from previous work (section 1, pp.3, paragraph 5).
		b	***and more description/model evaluation for the chemistry results***	The model description has been extended (section 2.1). This has been done by splitting section 2.1 "Coupled chemistry-aerosol microphysics model" in to two parts: 2.1.1 Model Description and; 2.1.2 Model Evaluation. As suggested, we have added a section on model evaluation (section 2.1.2). The model has been extensively evaluated in previous studies and we now describe these previous evaluations in more detail (pp.4, Section 2.1.2). In this study, we focus our new evaluation on aspects of the model that are most pertinent to this study – namely the vertical profile of sulphate and nitrate aerosol. We evaluate simulated vertical profiles of speciated aerosol mass concentrations against aircraft observations from Heald et al., (2011) – section 2.1.2. We add a new figure (new Fig. 1 – shown at the end of this document) that summarises aerosol model-observation comparisons. Overall, we demonstrate that

				globally the model overestimates sulfate, and underestimates nitrate, ammonium and OA. After broad stratification of field campaigns in to polluted, biomass burning and remote regions (as per campaign classifications used by Heald et al., (2011)) we find that the model best performs over polluted regions, with lower skill over remote regions, and greatest underestimation over biomass burning regions. Additionally, we add another new figure (new Fig. 2 – shown at the end of this document) where we demonstrate the model's ability to simulate ozone in the UTLS. In comparison to observational ozonesonde profiles compiled by Tilmes et al., (2012) from 41 different global locations from between 1995 to 2011, we find that globally the model slightly overestimates ozone.
		c	***The mortality methodology needs better explanation and improvement, as it uses out-of-date concentration-response functions, and should include more discussion of the appropriateness of using such factors worldwide.***	We thank the referee for pointing out how better explanation of the mortality methodology could aid the paper. As such we have included an in-depth explanation of the mortality methodology– section 2.5. While the concentration-response function utilised in this study may not be the most recently published function, it allows for this study to provide estimates of aviation-induced mortality directly comparable with previous work from Barrett et al., (2012) and Yim et al., (2015) – please also refer to our response to comment 6. As stated by Butt et al., (2016), the CRFs employed in this study are based on the American Cancer Society Prevention cohort study. WHO recommend the use of a log-linear model (as used here), as linear models could result in unrealistically large RR values when high $PM_{2.5}$ concentrations are considered ($PM_{2.5}$ > 30 μg m^{-3}); as also stated the supplementary information for Barrett et al., (2012). Though the application of the same CRF factor is global (as per Ostro et al., (2004) and recommendation from the WHO), we use regional population data and regional baseline mortalities for both cardiopulmonary disease and lung cancer.
	2		***Specific comments: abstract, line 7, line 16: significant figures***	Taking in to account that the values reported are estimates we have rounded mortality

			on annual mortality? Are 3597 and 624 really the estimates?	estimates up to the nearest 10.
	3		***The introduction could better establish what is not known, and what this study contributes relative to the work that has been done before, especially Barrett et al. 2010, 2012, and Morita et al. 2014.***	To better establish what is known, and the differences in estimates of premature aviation-induced mortality from other studies, we have included a paragraph in the introduction (pp.2, section 1, last paragraph): "Barrett et al. (2012) and Barrett et al. (2010) using the methodology outlined by Ostro (2004), estimated that aviation emissions are responsible for ~10,000 premature mortalities a^{-1} globally, due to increases in cases of cardiopulmonary disease and lung cancer. Yim et al. (2015) revised this estimated, using the same methodology, to 13,920 (95% CI: 7,220–20,880) mortalities a^{-1}. Morita et al. (2014) using the methodology to derive the relative risk (RR) from exposure to surface $PM_{2.5}$ from Burnett et al. (2014) estimate 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) using the methodology from Jacobson (2010) estimate 310 (95% CI: –400 to 4,300) mortalities a^{-1} due to cardiovascular effects. These studies demonstrate that the different methodologies employed and modes of mortality considered produce a wide range in estimated mortalities due to aviation emissions of between 310 – 13,920 mortalities a^{-1}". Additionally, to acknowledge what is not known we have highlighted the large uncertainty in estimates in the aviation-induced cloud albedo effect and how this paper investigates the cloud albedo effect for all FSC cases, through stating in the section 1 (pp.2, paragraph 3): "Few studies estimate the aerosol cloud albedo effect (aCAE) from aviation: Righi et al. (2013) assessed the aCAE to be –15.4±10.6 mW m^{-2} while Gettelman and Chen (2013) estimate –21±11 mW m^{-2}". Along with stating in Section 1 (pp3, paragraph 4) "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.".
	4		***Section 2.1. Some model evaluation would be useful here,***	We thank the reviewer for pointing out the value of model evaluation here. We now

			including information on stratosphere-troposphere exchange.	include speciated aerosol model-observation comparisons from a global synthesis of aerosol mass spectrometer measurements (see response to comment 1b above and our new Fig. 1). It is unclear which datasets could be used to evaluate stratosphere-troposphere exchange directly, however we now include comparisons of model-simulated with vertical profiles of ozone and aerosol in the upper troposphere with observations from ozonesonde and aircraft. The comparisons described show no evidence of stratosphere-troposphere exchange being a problem in the model. Examining the shape of vertical profile comparisons (new Fig. 2), we find no evidence of systematic model bias in the upper troposphere.
	5	a	***Section 2.2. While the authors explain differences in estimates from ranges esp. SO2, OC, CO which are outside previous ranges, a bit more information is warranted here. Specifically, why do the authors think that the fuel burn inventory, or OC emissions index, better reflects reality?***	Our intention was to develop a comprehensive aviation emissions dataset that includes an expansive set of pollution species emitted by aviation. Many previous aviation emission datasets only include a subset of emitted species. For example, the CMIP5 emissions dataset only includes aviation emissions of NOx and BC. We use emissions indices, which are shown to be in agreement and consistent with previous work to produce estimates of additional species emissions (Table 1). There are few previous estimates of global OC aircraft emissions. Wilkerson et al., (2010) using data from Wayson et al., (2009) assume a greater emission index for OC (of 0.015 g kg(fuel)$^{-1}$) but note that "this is intended for airport operations at ground level conditions rather than cruise-related operations" and as such may overestimate OC emissions. We use the relationship between BC and OC of 4:1 (Bond et al., 2004; Hopke 1985) to derive an OC emissions index of 0.00625 g kg(fuel)$^{-1}$. Without developing models for the complex relationship between operating conditions, fuel flow, aircraft type, ambient conditions, and actual flight paths, we see this as a viable and appropriate approach. We add a short statement to the paper (pp.8, section 2.2, paragraph 4): 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.
		b	***Given a fast-growing sector (especially in highly-populated areas such as Asia), why is the year 2000 still relevant? Some comment on the effect this choice has on results would be warranted.***	While we appreciate the fast-growing nature of the sector, investigations on the impacts of aviation for year 2000 are still pertinent given current literature which use year 2000 as a base for comparison for the future impact of aviation e.g. Righi et al., (2015). This choice allows for estimates of the radiative effect from aviation emissions to be compared against current literature.
	6		***Section 2.5. The health effects calculation uses older concentration-response functions that are not state-of-the-science. The authors should revisit their choice here. They should consider using a concentration-response function consistent with previous work to enable comparisons, even if this is just as a sensitivity study.*** ***The functional form should also be given here, and its uncertainty discussed.***	While we acknowledge that the CRF function employed here is older, use of this CRF allows our estimates to be compared to previous studies using the same CRF (Barret et al., (2010), Barret et al., (2012), Yim et al., (2015)). We have added a more detailed description of the function (pp.10, section 2.5). We add the following (pp.22): "Future work needs to estimate the health impacts of aviation using newly available concentration response functions (Burnett et al., 2014)."
	7		***Section 3.1. It is unclear why there are 'increases' under the NORM scenario? Relative to what?***	These are increases relative to a scenario with no aviation emissions, i.e. the NOAVI simulation. We have edited the text to make this clear.
	8		***Figures 1-3 and 5: axes and text a bit too small to read.***	Text size within figures has been increased to enhance readability.
	9		***p 18932 I'm surprised by the strong linearity (R2=1?) of PM2.5 to sulfur content. While I'm not surprised that this is roughly linear, an R2=1 suggests to me that important potentially nonlinear parameters might not have been included in the model. Can the authors comment on this?***	R^2 values of 1 were arrived after rounding to 2 d.p. – these values have been amended in the text to $R^2 > 0.99$ to make clear that these are not precisely unity. The model includes many non-linear processes, including chemistry and aerosol microphysics. We agree with the referee that the linear response is may seem surprising. The near-linear response is likely due to the small emission perturbations that we have applied relative to global aerosol emissions. We add the following statement (pp.11, section 3.1, paragraph 5): "Larger emission perturbations would likely lead to a non-linear response in atmospheric aerosol".

	10	a	***p 18932 line 20: Why is the estimate of sulfate attributable to NOx so different from Barrett et al. 2010?***	Thank you for pointing this out to us. As far as we can tell, the values are actually rather similar, although the Barrett et al study does not give an exact figure (we estimate a value of 36.2% while Barrett et al., (2010) estimate a value of between ~57–67% as per their supplementary information). We have rephrased the text to: "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." – (Section 3.1, pp.13, paragraph 3).
		b	***What differences are there between the models? Is it more likely to be chemistry or transport parameters?***	There are likely to be a variety of differences between the models: the emissions, transport and chemical schemes, and that GLOMAP-mode is a size resolved model. Without conducting a model comparison experiment that includes sensitivities to different parameters, it is difficult to determine whether differences in transport or chemistry are most important.
	11	a	***3.2. A comparison of how differences in premature mortalities are affected by the choice and assumed slope of CRFs is needed here.***	Thank you highlighting this point. The 95% confidence interval range highlights how the slope of the CRF employed effects estimates in premature mortality. Additionally, we have included a description of the main uncertainties involved and captured within the confidence interval reported evaluating the long-term health effects of exposure to $PM_{2.5}$, referring to Ostro (2004) (pp.14, section 3.2, paragraph 2): "Low-, mid- and high-range cause-specific coefficients (β) are used to account for uncertainty in the health impacts caused by exposure to $PM_{2.5}$ (Section 2.5) (Ostro, 2004)" As such the use of low-, mid- and high-range cause-specific coefficients help and account for uncertainties which are difficult to capture in long-term studies, such as mortality displacement of a few days and disease-relevant times, durations and intensities of exposure (Ostro, 2004)

		b	***Concentration is not the only difference from previous work. Also, what about comparing to the results of Morita et al. (2014) in their present day scenario? Why are the USLJ changes different from Barrett et al. 2012?***	It is true that concentrations will not be the only difference and factors driving differences in aviation-induced mortality estimates. Differences in cause-specific coefficients (β) will also play a role. To acknowledge this, the following statement has been added (pp.14, section 3.2): "Additionally, differences in mortality arise due to the use of different cause-specific coefficients (β) within the same CRF, as well as different population datasets." We cannot confirm the effect of the β values used by Barrett et al., (2012) as these are not provided in their paper or supplementary information. A comparison between estimates in aviation-induced premature mortalities from lung cancer evaluated by this study (390 [95% CI: 150–640] mortalities a^{-1}) and Morita et al., (2014) (41 [95%: 7–67] mortalities a^{-1}) is now discussed. The main reason for the differences in estimates of aviation-induced premature mortality is due to the different CRFs used, with Morita et al., (2014) using the IER (integrated exposure response) methodology outlined by Burnett et al., (2014). The IER methodology is described in order to identify how the two methodologies differ, thus resulting in different estimates in aviation-induced premature mortality – (section 3.2, pp.14, paragraph 3). While we appreciate that through the application of a ULSJ fuel strategy Barrett et al., (2012) estimate that premature 2,300 mortalities could be avoided in comparison to our estimate of 620 mortalities avoided (estimates which differ by a factor of 3.7), we see similar rates of reduction in mortalities avoided when relative values are considered: we estimate a reduction in mortalities of 17.3%, while Barrett et al., (2012) estimates a reduction in aviation-induced mortalities of 23%. Differences in estimates of mortalities avoided between this study and Barrett et al., (2012) for the ULSJ will again be a function of different β values used and differences in simulated surface-layer $PM_{2.5}$ concentrations, in both the base case and ULSJ.
	12		***Figure 8 is perhaps the most unique part of this work and deserves a bit more discussion.***	Additional attention has been given to this figure (now called Fig. 10), linking in to reply to comment 15 from reviewer 2.

2	1		***The main concern with this study is the use of an off-line model to study climate impacts of aircraft emissions. The model description says that that the GLOMAP-mode is embedded within the 3-D off-line Eulerian CTM to make it a coupled chemistry-aerosol microphysics model. It seems that the meteorological and chemical processes are not coupled. A discussion on the justification of an offline CTM to study climate impacts will greatly strengthen this paper.***	When referring to a "coupled chemistry-aerosol" model we mean that the aerosol microphysics and gas-phase chemistry are coupled. The referee is correct that our model is a chemical transport model, using offline meteorology and that there is no coupling between chemistry and meteorology. We now explain this more clearly in the paper (see Section 2.1.1, pp.3, paragraph 6). The advantage is that we can compare our short 1-year simulations with each other directly, since the meteorology in each run is identical, meaning we are only looking at chemical changes due to changes in emissions. We can then calculate the climate effects of our changes offline using the radiative transfer model.
	2		***There is no discussion of evaluation of the model – either for meteorological variables or for air pollutant concentrations. While this is not at the core of this study, model evaluation is an essential prerequisite for any application study like this. It is suggested that the authors include results from the evaluation, and to specifically focus on the model's ability to predict both PM2.5 mass and speciated components in different parts of the world for the year studied.***	We thank the reviewer for highlighting the added value model evaluation can bring to the manuscript. We have added a model evaluation section (section 2.1.2) which details previous model evaluation work conducted on TOMCAT-GLOMAP-mode, and aerosol (sulfate, nitrate, ammonium and organic aerosol) and ozone specific to the nitrate-extended version of the TOMCAT-GLOMAP-mode coupled model used in this study. The model's ability to simulate sulfate, nitrate, ammonium and organic aerosols is evaluated against observations of aerosol mass from aircraft field campaigns compiled by Heald et al., (2011) (see new Fig. 1 and associated text)., Model-simulated ozone profiles are evaluated against ozonesonde profiles compiled by Tilmes et al., (2012). Please also see our response to referee #1 (comments 1b and 4).
	3		***There is no discussion of the emissions inventories used for non-aircraft sources. While documenting the source of these for this study, putting those in context with the other key studies referred in this study is important. Aircraft emissions react with background emissions from other sources such as NH3 to form aviation-attributable PM2.5, specifically inorganic PM2.5 which is at the core of this study. So, a discussion of NH3 emissions used in this study is critical but***	The model description section has been reordered and extended (section 2.1.1, pp.3-4) in order to: 1. Provide a description of the hybrid solver employed to simulate the dissolution of of semi-volatile inorganic gases (such as H2O, HNO3, HCl and NH3) in the aerosol-liquid-phase. 2. Identify where information on cloud fraction and cloud top pressure fields are taken from (ISCCP-D2), and, 3. Highlight the differences between the TOMCAT CTM and p-TOMCAT CTM. 4. Provide a description of sources of non-aviation emissions,

			lacking.	5. Sources of anthropogenic and natural emission sources are now listed in the model description section – stating that NH3 emissions are from the EDGAR inventory (Bouwmann et al., 1997).
	4		***Since Barrett et al (2012) used 3 different models, two of which were applied globally, when comparison are made to Barrett et al (2012), it is helpful to know which of the two models are being referred to in this study.***	We thank the reviewer for making this point. For clarity within the manuscript, and where it seems appropriate we have stated that results from this study are being compared to Barret et al.,'s simulations using GEOS-Chem.
	5		***Suggest including findings from two recent studies, and put these results into context. The first one is by Morita et al, ES&T 2014 which was published last year and is relevant from the health risk aspects of aircraft emissions using the NASA GISS ModelE2, and the other is more recent one by Brasseur et al, BAMS 2015, which is relevant from the climate impact aspects of aircraft emissions using multiple global-scale models.***	We thank the reviewer for this suggestion. We integrated findings from Morita et al., (2014) and Brasseur et al., (2015) in to the paper. Estimates in the ozone and aerosol direct radiative, and cloud albedo effects from Brasseur et al., (2015) have been included in a new paragraph added (pp.2, para 3, section 1) in order to help put in to context and convey current estimates for aviation. Morita et al., (2014) has been mentioned, reporting their estimates in mortality from standard aviation, while making reference to the methodology used as this will impact how directly comparable the values estimated by Morita et al., (2014) are with the mortality estimates derived here (pp.14, section 3.2, paragraph 3).
	6		***Section 1 Pg 18926 Line 10: "A coupled tropospheric chemistry-aerosol microphysics model including nitrate aerosol…" Why the emphasis on nitrate aerosol, and not inorganic PM in general? Can the authors clarify this?***	The explicit mention of nitrate aerosols was included as other versions of the TOMCAT-GLOMAP-mode coupled model do not include the formation of nitrate aerosols. This emphasis has been removed now.
	7		***Section 2.1 Pg 18926 This study has used TOMCAT, and one of the models used by Barrett et al (2012) against which several comparisons are made in this study was p-TOMCAT. Since the names are so close to each other, a brief discussion of how these two models are different will be relevant for the sake of the comparisons presented.***	Both TOMCAT and p-TOMCAT started from the same model version several years ago, but their development has now diverged. The main differences are: our version has a different chemistry scheme, it has coupled online aerosols, a different photolysis scheme, a different dry deposition scheme and different emissions. The 'p' in p-TOMCAT stands for parallel, as it was the first version of the existing TOMCAT to use Message Passing Interface (MPI) to make the model suitable for massively parallel machines. Those modifications are now

				incorporated into the main TOMCAT model used here. The version of the TOMCAT CTM used in this study can be considered as an updated version of TOMCAT, on which p-TOMCAT was based.
	8		***Section 2.2 Pg 18927-18928 While discussing the aircraft emissions inventories in Table 1, providing comparisons against previous global inventories as ranges is helpful. However, given the subsequent multiple comparisons of air quality and health risk estimates from this study with Barrett et al (2012), which used the Wilkerson et al (2010) inventories, having an additional column with Wilkerson et al numbers is suggested. Further, the EIs are listed for 6 explicit VOC species – formaldehyde, ethane, propane, methanol, acetaldehyde and acetone, and in the last column, no comparison is shown and N/A is stated. Two suggestions are offered to improve this table. Wilkerson et al report total hydrocarbons in their Table 4. The authors could compare their estimate of total HCs against that of Wilkerson, or use the speciation information of TOG in Wilkerson's Table 9, and compare explicitly for each of these 6 HCs. Since both Barrett et al (2012), and Morita et al (2014) use the Wilkerson et al inventories, providing this comparison upfront for all key species including HCs is of special relevance.***	As suggested we have used speciation information from Wilkerson et al., (2010) to provide information on the annual emissions of speciated HCs to allow CMIP5-extended annual emissions to be compared to annual emissions from Wilkerson et al., (2010). We have taken the suggestion to have an additional column presenting Wilkerson et al., (2010)'s numbers. Though a separate column has not been added to Table 1 presenting their data (Wilkerson et al., (2010)) specifically, global annual VOC emissions from Wilkerson et al has been incorporated in to the last column of Table 1.
	9		***Section 2.5 Pg 18930 Some justification of why they chose a somewhat outdated C-R function for PM2.5 is helpful. The literature has evolved, and more recent functions including those used in the Global Burden of Disease, 2010 are available now.***	While we acknowledge that this methodology is not the most recent, we used methodology based on Ostro (2004) to allow us to be directly comparable with previous literature (Barrett et al., (2010) and Barrett et al., (2012)) that used the same CRF (please see our response to comment 1b and 4 from reviewer 1). We add a statement to the paper (pp.10, section 2.5, paragraph 1) to explain our choice and to acknowledge that newer functions are now available "Using this

				function allows us to compare directly with previous studies (Barrett et al., 2012; Yim et al., 2015); in future work estimates are required with updated methodologies (Burnett et al., 2014)".
	10		***Section 3.1 Pg 18931-18932 The authors acknowledge that the response of modelled inorganic PM2.5 is very non-linear and do a nice job illustrating examples where even "when aviation emissions contain no sulfur, aviation-induced sulfate is formed through aviation NOx-induced increases in OH concentrations, resulting in the increased oxidation of SO2 from non-aviation sources". However, this does not align with the fairly linear response of aviation-attributable PM2.5 to changes in FSC, as presented in Figure 2. A reconciliation of the non-linear response discussed above with the linear response in Figure 2 warrants additional explanation.***	This comment has brought to our attention that through the inclusion of the word "increased" within the associated explanation the wrong impression was given. We intended for that sentence to put across that even when aviation emissions contain no SO_2, sulfates are still formed through aviation-NO_X induced increases in OH concentrations. To clear this up we have removed the use of the word "increased", so as not imply there are any changes in the rates of aviation-induced sulfates, irrespective of whether the source SO_2 emissions are from aviation or other sources. This hopefully clears any confusion about our message and helps clarify that a fairly linear response is still seen in Fig. 4 (previously Fig. 2), with our desulfurised case (FSC = 600 ppm) creating a "baseline" level of aviation-induced sulfates.
	11	a	***Section 3.2 Pg 18933 The comparison with Barrett et al (2012) can be improved here, and provide more insights to the reader on the differences being seen, especially if Barrett et al estimates are higher by factors of 5 and 2.5 in different parts of the world.***	Further analysis cannot be provided here as we are unable to compare changes surface nitrate and ammonium concentrations, as these are not shown by Barrett et al., (2012). A statement to this effect has been added (section 3.2, pp.14, paragraph 2): "Additionally, differences in mortality arise due to the use of different cause-specific coefficients (β) within the same CRF, as well as different population datasets." Along with the following statement (section 3.2, pp.15, paragraph 2) "Additionally, the GRUMPv1 population dataset that Barrett et al. (2012) use resolves population data on a finer scale compared to the resolution of GPWv3 population dataset used here (Center for International Earth Science Information Network, 2012); differences which could contribute to differences in estimates of mortality."
		b	***In lines 20-22, when they attribute some of these differences to "other aerosol components", a quantitative comparison for each of these other components along with some explanation would be***	We acknowledge that this comparison would be helpful and aid further understanding the differences between these two pieces of work, but mean aviation-induced $PM_{2.5}$ changes from normal aviation are not reported by Barrett et al., (2012). Plots of aviation-attributable ground-level $PM_{2.5}$

			helpful.	concentrations for standard aviation using standard aviation fuel are presented by Barrett et al., (2012)'s supplementary information, but values are not reported in the text. A breakdown of changes in 'other' aerosol species is provided in section 3.1. when discussing the use of ULSJ fuel (pp.11, paragraph 2).
	12		***Section 3.2 Pg 18934 Lines 1-8: ULSJ reduces global mean PM2.5 concentrations by 1.41 ng/m3 and 0.89 ng/m3 in this study and Barrett et al (2012). For inorganic PM2.5 components, this study estimates 1.61 ng/m3. How does this compare with Barrett et al (2012)?***	Unfortunately, we cannot make this comparison as Barrett et al. (2012) do not report all the necessary aerosol components.
	13		***Again, when using ULSJ, if the authors see a net reduction in surface PM2.5 of 1.41 ng/m3, what is causing an increase in other aerosol species of +0.20 ng/m3? Showing the aviation-attributable speciated PM2.5 will be helpful, perhaps as global average, and again for each of the major regions studied – Europe, North America, and Asia for key ULS scenarios.***	In section 3.1 (pp.12, paragraph 2) a breakdown of the global average changes in speciated aerosol from using ULSJ fuel is provided; providing mass and relative changes for the largest changes and relative changes for the species which see the smaller changes in mass.
	14		***Lines 25-28: This study shows a 17.4% reduction in global premature mortality, while Barrett et al (2012) show a 23% reduction. The authors attribute this to larger changes in PM2.5 in populated regions of the world. Can the authors comment on potential differences in the population datasets used in the two studies?***	Both Barrett et al., (2012) and this study use population data from the Center for International Earth Science Information Network (CIESIN). We use the "Gridded Population of the World, Version Three (GPWv3)", while Barrett et al., (2012) use the Global Rural-Urban Mapping Project, Version One (GRUMPv1) version. It is acknowledged that the GRUMPv1 dataset that Barrett et al., (2012) *"which provides a higher- resolution data product that moves populations out of thinly settled large administrative units into settlements"*, thus giving a better resolution at an urban scale. The differences between the population dataset has been acknowledged in section 3.2 (pp.14, paragraph 2 and pp.15, paragraph 2)
	15	a	***Section 3.5 Pg 18937 Figure 8 presents an interesting relationship between changes in***	The difference in the three slopes are due to the different cause-specific coefficients used (beta coefficients) which aim to highlight the

			mortality versus net radiative effect for the low, mid and high ranges of mortality sensitivities for various FSC scenarios. What would explain the differing slopes for the 3 ranges?	uncertainties that are present when trying to evaluate premature mortality from long-term exposure to $PM_{2.5}$ – linked to comment 11 from reviewer 1. While addressing this comment and how the choice of cause-specific coefficient affect the mortality line and its associated relationship with net radiative effect, fig 8 (now fig 10) has also been paid more attention.
		b	***While it is appreciated that the authors have performed this analysis, additional discussion here would be helpful to understand the implications of the fairly stiff response for mortality at low range versus almost linear change at high range.***	The implications of the fairly stiff responses seen at the low range in comparison of the almost linear response seen at the high range are discussed in the response to reviewer 1's comment 11a, i.e. the range created by the low and high CRFs (driven by different β-values) are employed to try and account for uncertainties which are difficult to capture in long-term studies, such as mortality displacement of a few days and disease-relevant times, durations and intensities of exposure (Ostro, 2004). The following has been added to the manuscript on (section 3.2, pp.14, paragraph 1): "Low-, mid- and high-range cause-specific coefficients (β) are used to account for uncertainty in the health impacts caused by exposure to $PM_{2.5}$ (Ostro, 2004)".

Fig. 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.

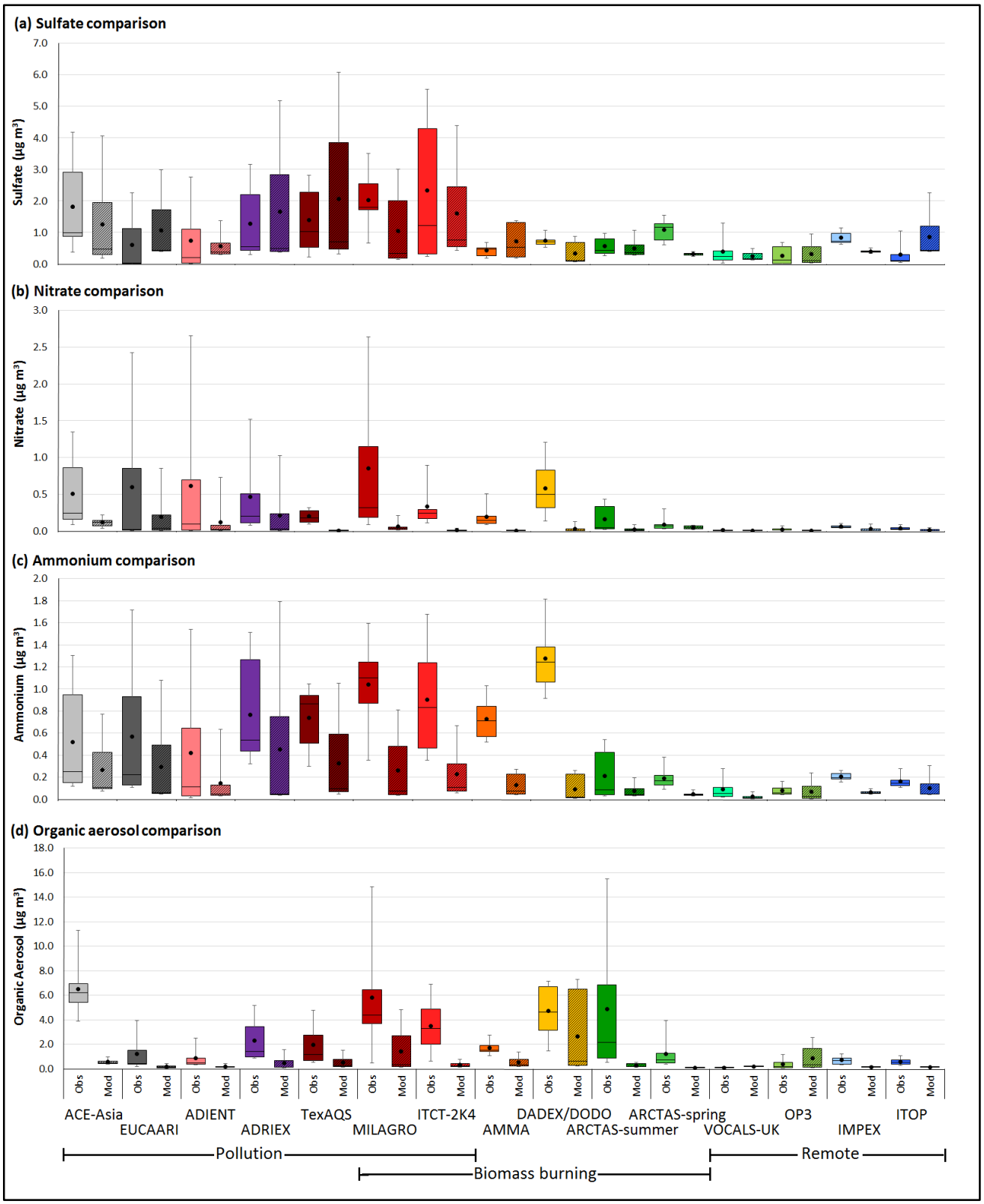
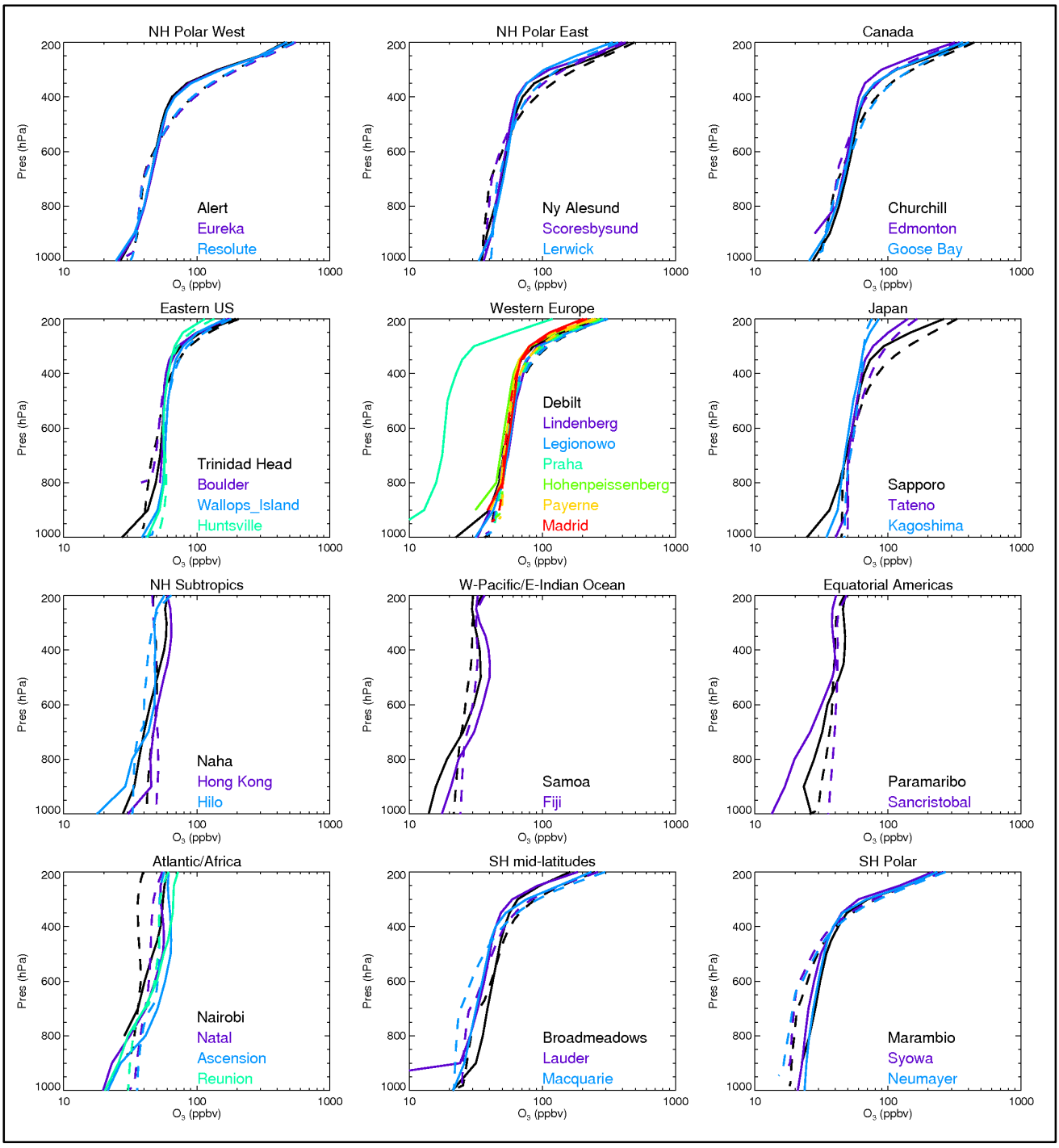


Fig. 2: Comparison of observed (solid lines) and simulated (dashed lines) ozone profiles. Observations are taken from ozonesonde observations, and arranged by launch location regions as arranged by Tilmes et al. (2012).



b) Reply to editor's decision and to referee #2's minor comments on "Impacts of aviation fuel sulfur content on climate and human health" by Z. Z. Kapadia et al.

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

We would like to thank both anonymous referees and the editor for their time in reviewing this paper.

As per the editor's decision and anonymous referee #2's minor comments (below), the following changes have been made (as listed under each revision suggested).

Minor revisions suggested as per Report #2:

The only remaining comments are on the model evaluation section, specifically Figure 1.

a) ***Suggest adding a table and figure, showing brief details of the 17 airborne campaign datasets used in the model evaluation, perhaps as Supplemental information. Not everyone is familiar with the acronyms and details of the field campaigns used in Figure 1.***

   As per this suggestion we have created a supplementary information document which reproduces a figure and adapts a table from Heald et al., (2011) (Figure S1 and Table S1 respectively). Figure S1 shows the flight paths for each of the aircraft field campaigns collated by Heald et al., (2011), while Table S1 is again adapted from Heald et al., (2011) providing details on each campaign, aircraft type used, location, date of campaign, regional class and relevant references for each campaign.

   As such at line 202 we insert the following:

   "The supplementary information presents the flight paths of each of the aircraft field campaigns used in the study compiled by Heald et al. (2011) (Figure S1), and details of each of the aircraft field campaigns used (Table S1)."

b) ***Also, while explaining model performance, I appreciate the grouping by 3 source regions, on the lines of Heald et al. However, I am not convinced with the authors' explanation about the poor model performance in the biomass burning regions. The model performance is particularly poor for all inorganic PM2.5 components (nitrate and ammonium in particular) in the DADEX/DODO and ARCTAS campaigns. What explains such large underpredictions for inorganic PM2.5 components, where it is hypothesized that the model is missing the narrow concentrated plumes in biomass burning regions? Given the focus on ULS fuel for aviation and its potential health and climate benefits, it will help addressing this underprediction to provide context for the impact assessments.***

   The referee correctly highlights that the model underestimates both organic and inorganic components in biomass burning influenced regions. We agree that there are additional reasons that we did not mention previously. As suggested by the referee we have added a short discussion of likely reasons for this underestimation in biomass burning regions.

   We delete "These model underestimations could partly due to very concentrated plumes in these regions affecting campaign mean concentrations" and add the following discussion at line 230:

   "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)."

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 µm) 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 ~3,600 [95% CI: 1,310–5,890] annual premature mortalities globally due to increases in cases of cardiopulmonary disease and lung cancer, resulting from increased surface $PM_{2.5}$ 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 0–6000 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 climate and air quality impacts (Lee et al., 2009; Barrett et al., 2010; Woody et al., 2011; Barrett et al., 2012).

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; Lee et al., 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 mW 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 6–36.5 mW 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., 2015). The aerosol direct effect is highly uncertain [–28 to +20 mW m^{-2}] (Righi et al., 2013), with the direct aerosol effects for sulfate ranging between –0.9 to –7 mW 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., 2015), nitrate ranging between –4 to –7 mW m^{-2} (Unger et al., 2013; Brasseur et al., 2015), BC ranging between 0.1–0.3 mW 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., 2015), and for organic carbon (OC) ranging between –0.67 to –0.01 mW 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±10.6 mW m^{-2} while Gettelman and Chen (2013) estimate –21±11 mW m^{-2}.

Aviation emissions can increase atmospheric concentrations of fine particulate matter with a dry diameter of <2.5 µm ($PM_{2.5}$). 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 U.S. 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 $PM_{2.5}$ 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) and Barrett et al. (2010) used the methodology of Ostro (2004) to estimate that aviation emissions are responsible for ~10,000 premature mortalities a^{-1} due 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: 7,220–20,880) 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 4,300) 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 310–13,920 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; Barrett et al., 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 550–750 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 desulfurised 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 ~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 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 7 size modes (4 soluble and 3 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).

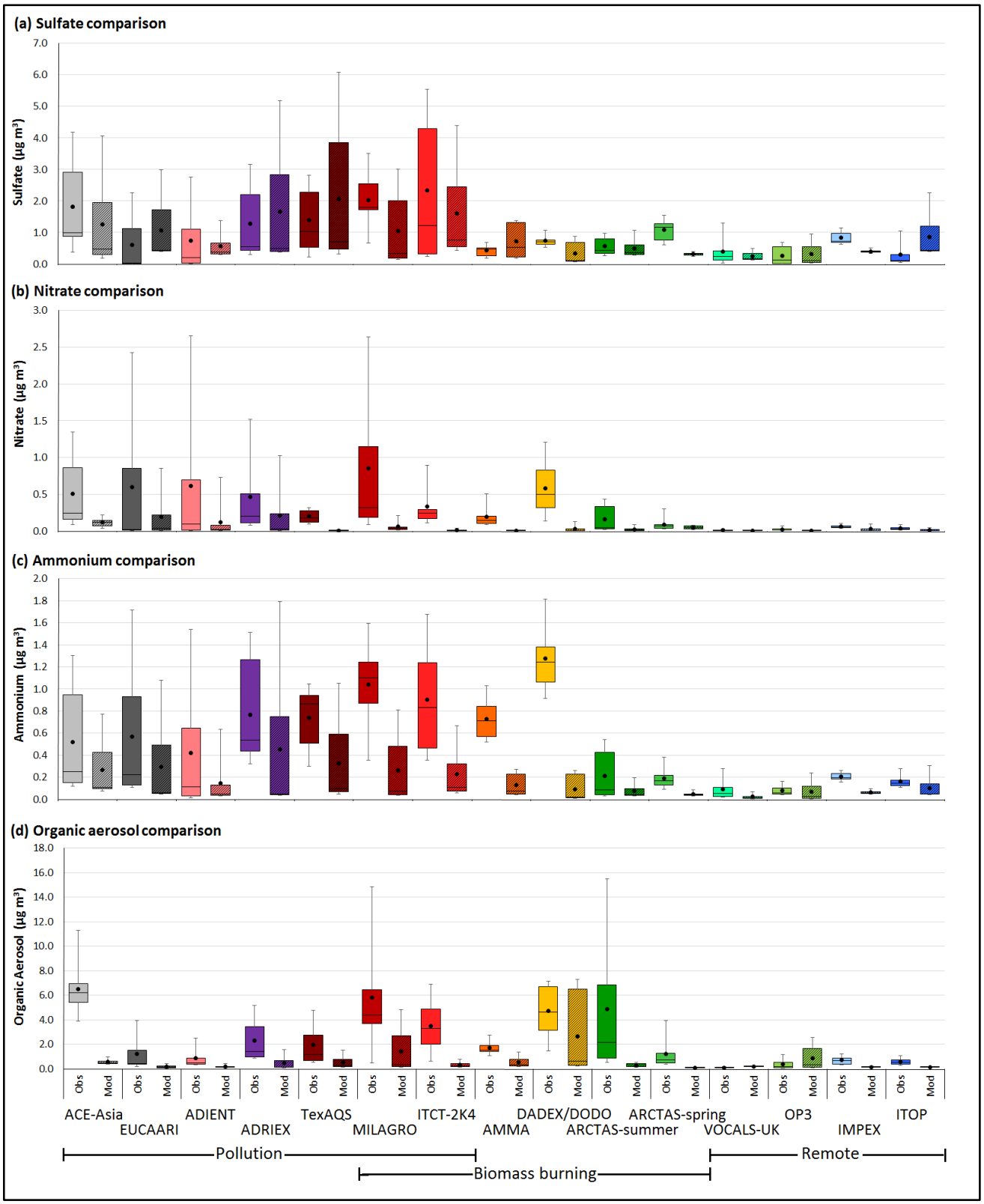
Here, we ran simulations at a horizontal resolution of 2.8° x 2.8° with 31 hybrid σ-p levels extending from the surface to 10 hPa. All simulations were conducted for 16 months from September 1999 to December 2000 inclusive, with the first four months discarded as spin-up time.

2.1.2 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 U.S. and East Asia. Here we focus our evaluation on the aerosol vertical profile and as well as nitrate aerosol which has not been evaluated previously.

Fig. 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) (Figure S1), and details of each of the aircraft field campaigns used (Table S1). 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.

Fig. 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.



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 dependant 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 2000–2008 (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).

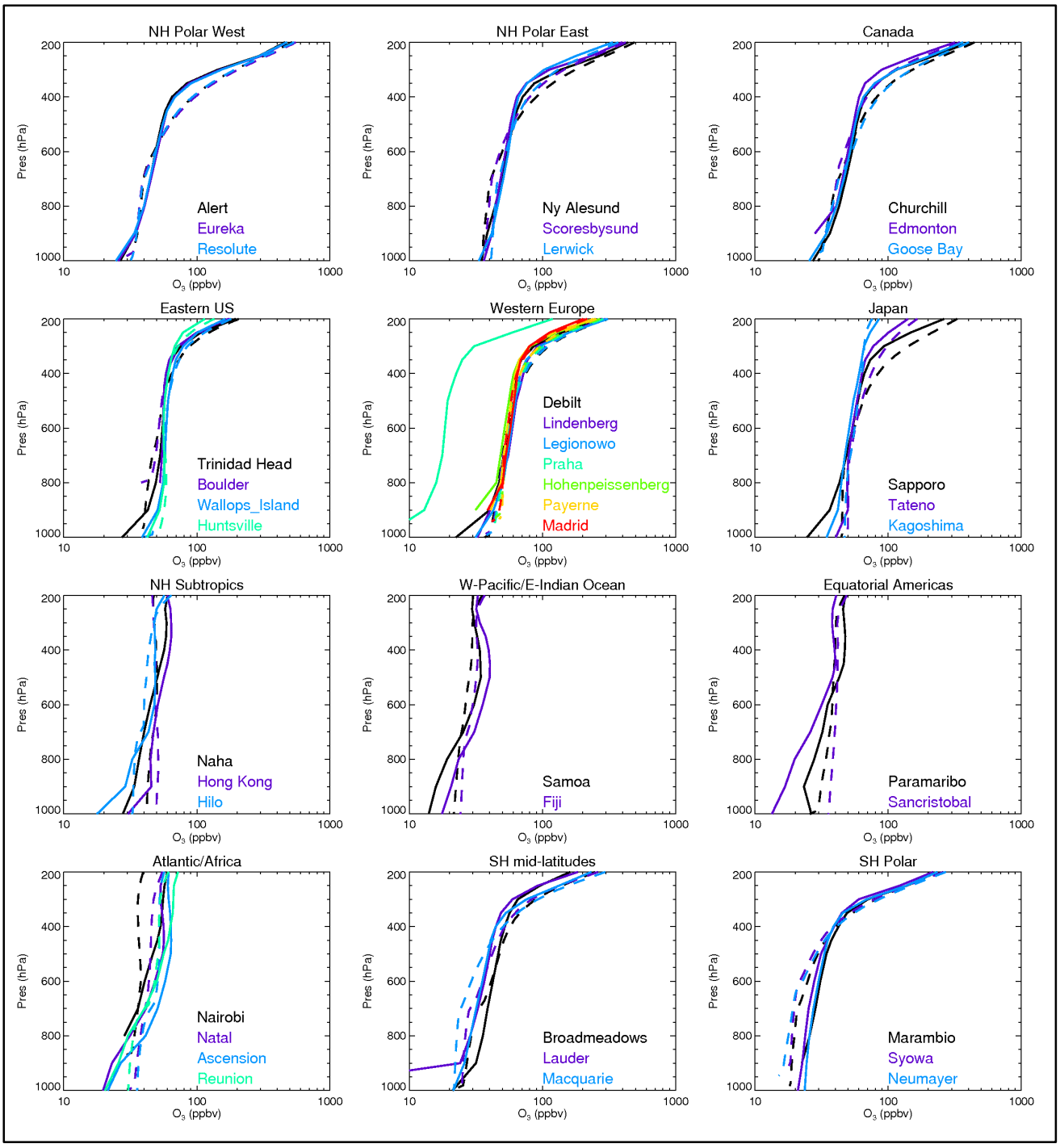
Fig. 2 presents simulated ozone concentration profiles in comparison to ozonesonde observations compiled by Tilmes et al. (2012). Observations were compiled from three networks, comprising of 41 stations with continuous sampling from 1995 to 2011: (i) The World Ozone and Ultraviolet Data Center (WOUDC)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

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



2.2 Aviation emissions

Aircraft emit NO_X, carbon monoxide (CO), SO_2, BC, 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 (Eyers et al., 2004; Quantify Integrated Project, 2005-2012; Wilkerson et al., 2010).

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., 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 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., 2004) as used by Lamarque et al. (2009). Following 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 from the FAA's aviation environmental design tool (AEDT) (Wilkerson et al., 2010). OC emissions are calculated using a BC:OC ratio of 4 (Hopke, 1985; 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), in conjunction with operating parameters suggested by the Airbus Flight Crew Training manual (Airbus, 2008).

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)

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 (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 σ = 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. 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).

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

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. (2011a), 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.

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 directly with previous studies (Barrett et al., 2012; Yim et al., 2015); 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 are 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):

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

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 equation (2):

$$AF = (RR - 1)/RR \quad (2)$$

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

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

Global population data is 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 $PM_{2.5}$

Fig. 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 ~80 ng m^{-3} (relative to the NOAVI simulation) over Central Europe and Eastern China (Fig. 3(a)). Aviation emissions result in largest fractional changes in annual mean $PM_{2.5}$ concentrations (up to 0.8%) over North America and Europe (Fig. 3(b)).

Fig. 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°N–66°N) and North American (146°W–56°W, 29°N–72°N) regions.

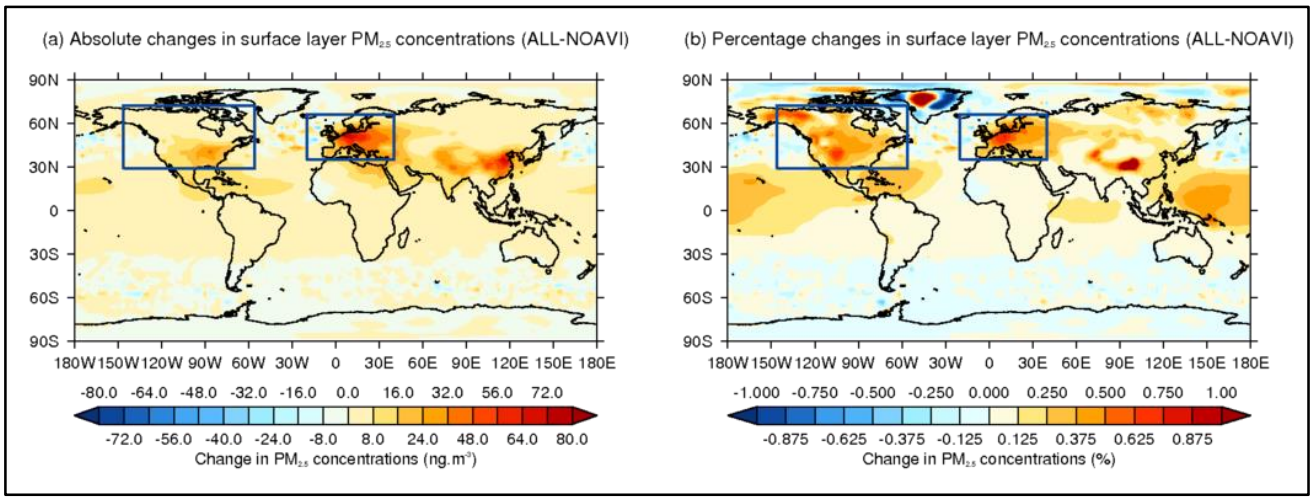


Fig. 4 shows the impact of aviation emissions on global and regional mean $PM_{2.5}$ concentrations, as a function of FSC. With standard FSC (FSC = 600 ppm), aviation increases global mean surface $PM_{2.5}$ concentrations by 3.9 ng m^{-3}; with increases in $PM_{2.5}$ dominated by sulfates [56.2%], nitrates [26.0%] and ammonium [16.0%]. Aviation emissions increase European annual mean $PM_{2.5}$ concentrations by 20.3 ng m^{-3} (Fig. 4(b)), substantially more than over North America (Fig. 4(c)) 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 [+3.2x10^{-3} ng m^{-3}; +0.3%]. Aviation emissions also leads 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. 4 (b,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 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, 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.

Fig. 4: Impact of aviation FSC on (a) global, (b) European (20°–40°E, 35°N–66°N), (c) North American (146°W–56°W, 29°N–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 $PM_{2.5}$.

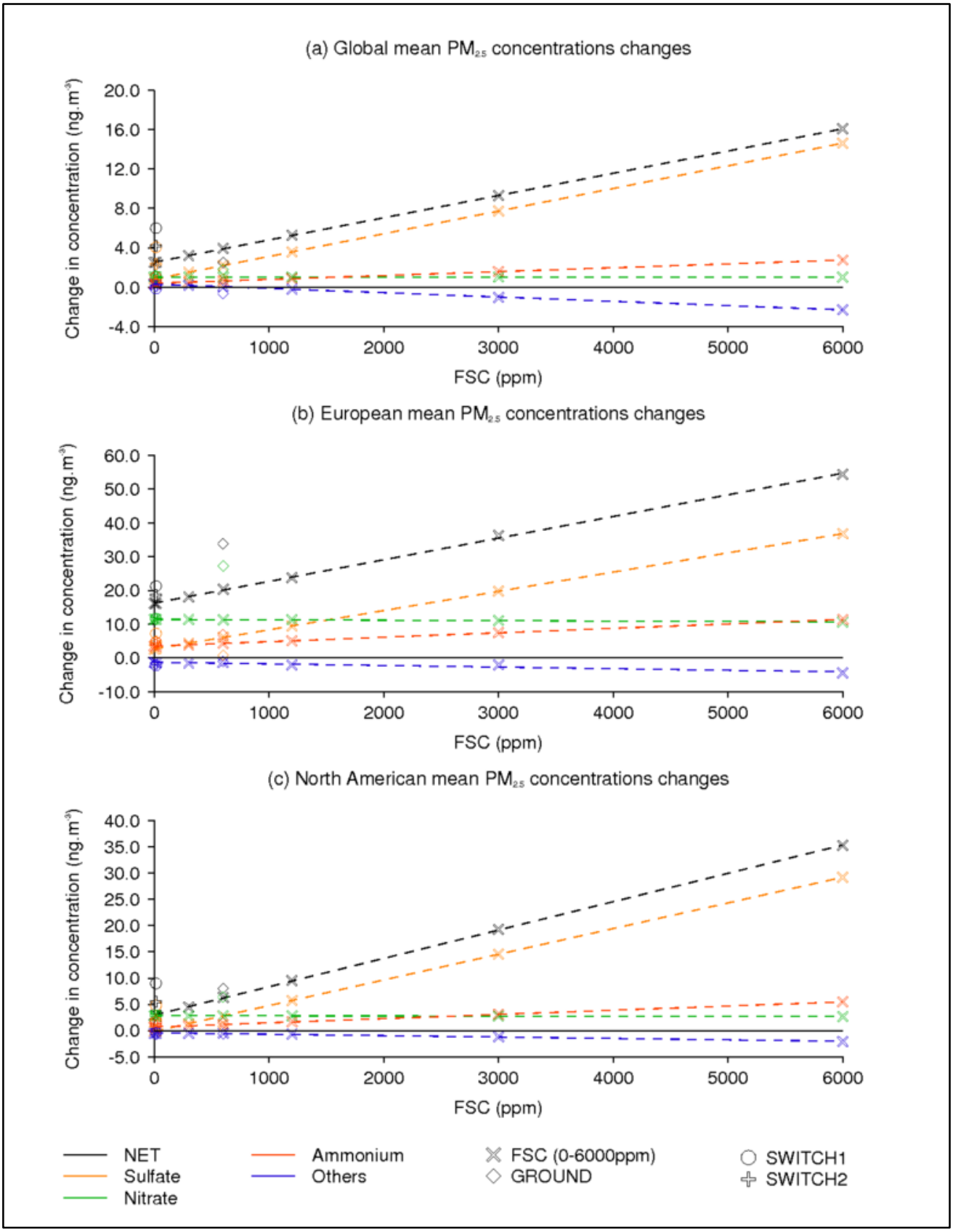


Fig. 5: Simulated differences in zonal annual mean sulfate (a) and nitrate (b) concentrations from the use of ULSJ fuel relative to standard fuel (ULSJ–NORM).

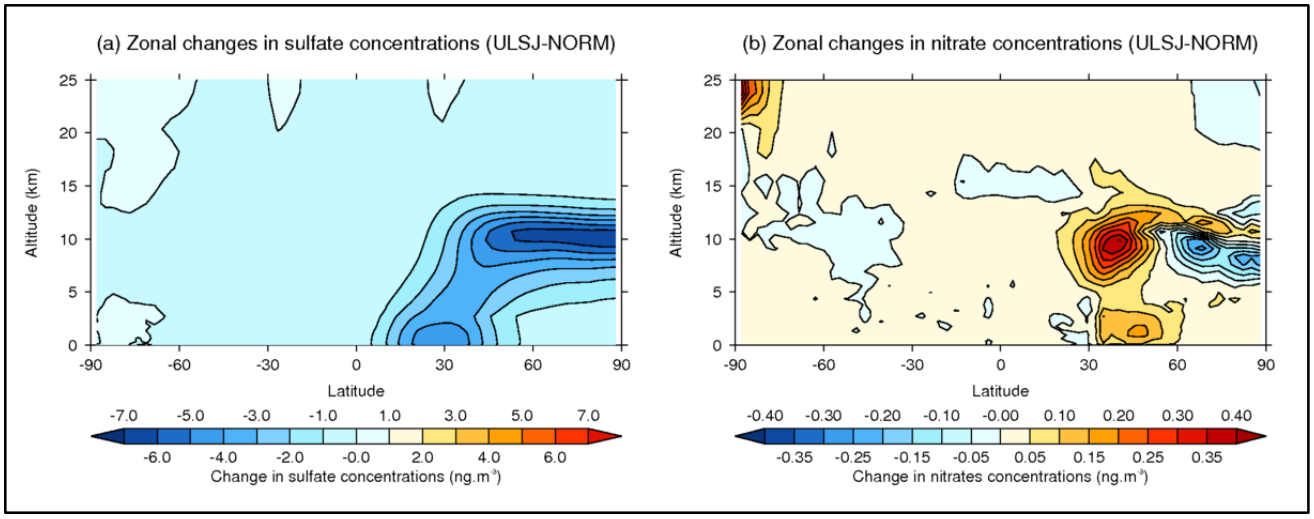


Fig. 5 shows the impact of changing to ULSJ fuel on zonal mean sulfate and nitrate concentrations relative to standard fuel (NORM). Table 3 reports the global aerosol burden from aviation under different emission scenarios. With standard FSC (FSC = 600 ppm), the global aviation-induced aerosol burden 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 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 oxidation of SO_2 from non-aviation sources (Unger et al., 2006; Barrett et al., 2010).

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 SO2	2.0 (–88.3%)	0.3 (–97.5%)	0.1 (–97.9%)

In line with previous work, we find 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 desulfurisation 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 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 95.0% reduction in aviation-induced global mean surface level aviation-induced $PM_{2.5}$. These results imply that only limited sulfate reductions can be

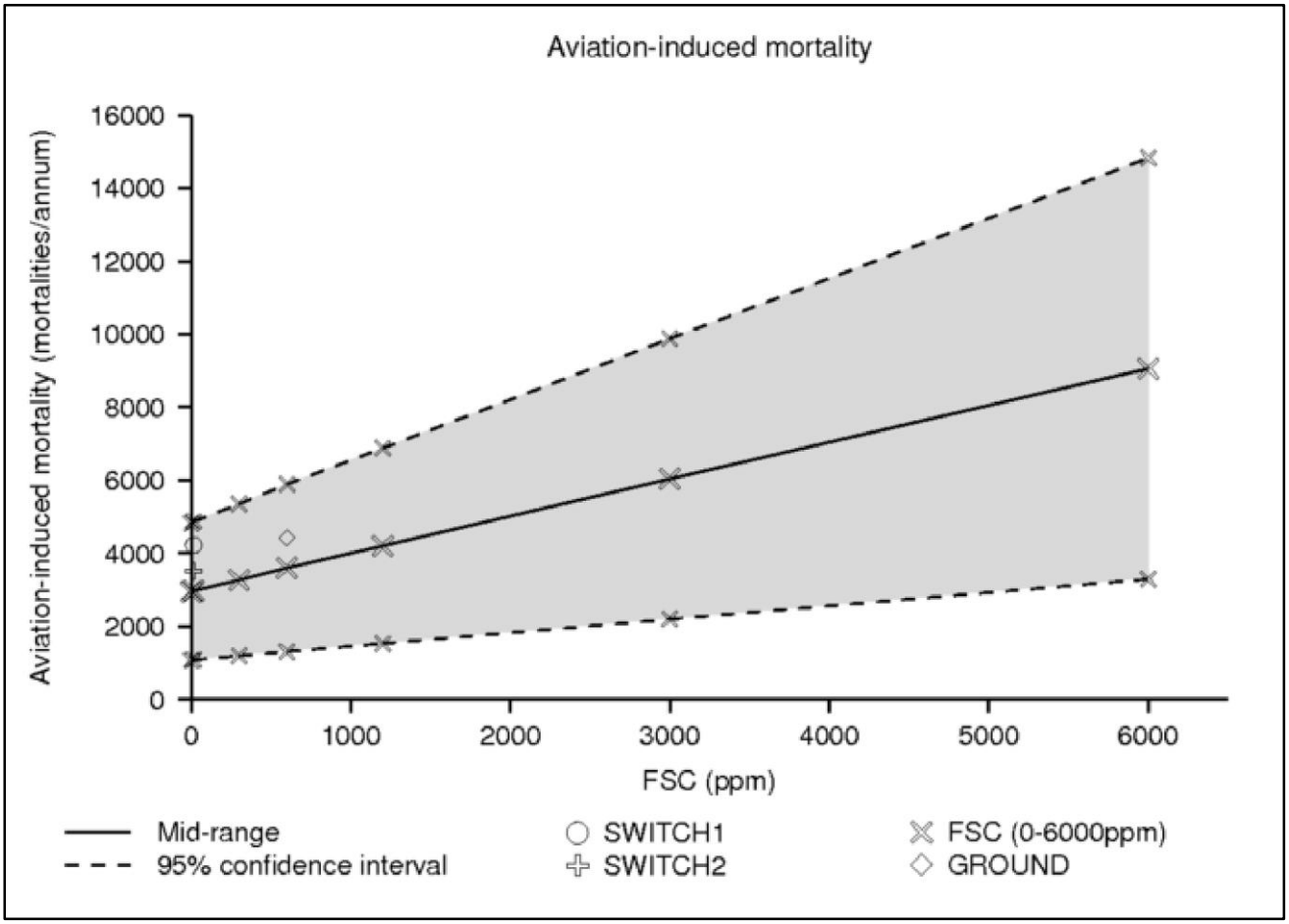
achieved through reducing FSC alone, with further reductions in aviation-induced $PM_{2.5}$ sulfates requiring additional controls on aviation NO_x emissions.

3.2 Premature mortality

Fig. 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 3,600 [95% CI: 1,310–5,890] 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 $PM_{2.5}$ (Section 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; Barrett et al., 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 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 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. Additionally, differences in mortality arise due to the use of different cause-specific coefficients (β) within the same CRF, as well as different population datasets. 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.

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



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

Barrett et al. (2012) estimated that swapping to ULSJ fuel could result in ~2,300 [95% CI: 890–4,200] fewer premature mortalities globally per annum; a reduction of 23%. In their work (using GEOS-Chem), the use of ULSJ reduces global mean $PM_{2.5}$ 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 dataset that Barrett et al. (2012) use resolves population data on a finer scale compared to the resolution of GPWv3 population dataset used here (Center for International Earth Science Information Network, 2012); differences which could contribute to differences in estimates of mortality.

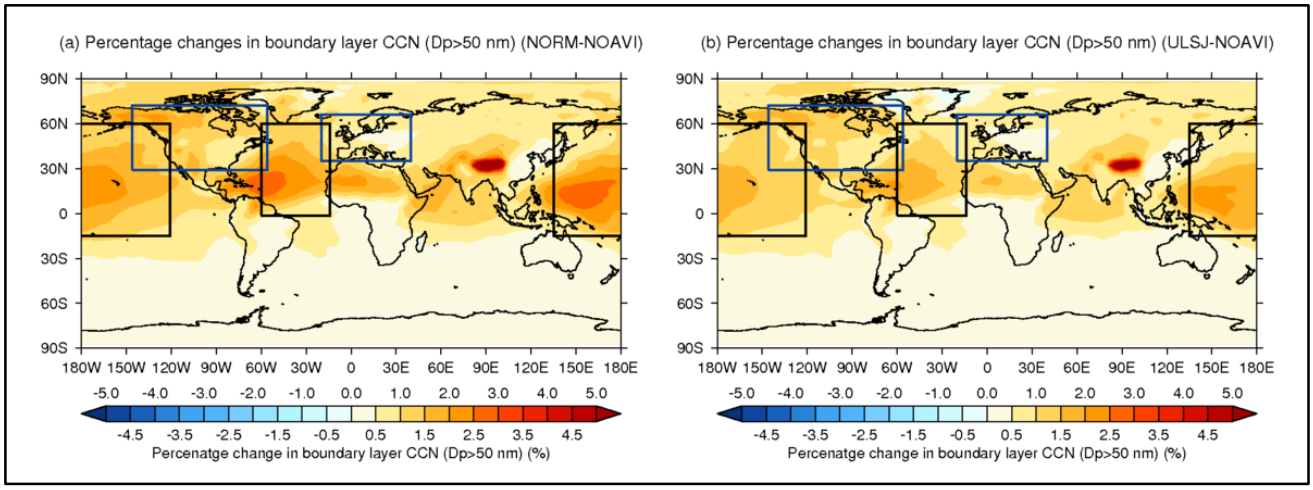
We also estimate how aviation-induced mortality would change if FSC was increased. We find that increasing FSC to 3000 ppm (HIGH) would increase annual aviation-induced mortalities to 6,030, an increase of 67.8% in relation to standard aviation (NORM; FSC = 600 ppm).

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 (879hPa; 0.96km) by 0.9% (2.3 cm^{-3}) (Fig. 7(a)). 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.

Fig. 7: Impact of aviation emissions on low-cloud level (879 hPa) CCN (Dp>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°W–14°W, 1.4°S–60°N) and Pacific regions (135°E–121°W, 15°S–60°N) referred to in the text.



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).

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

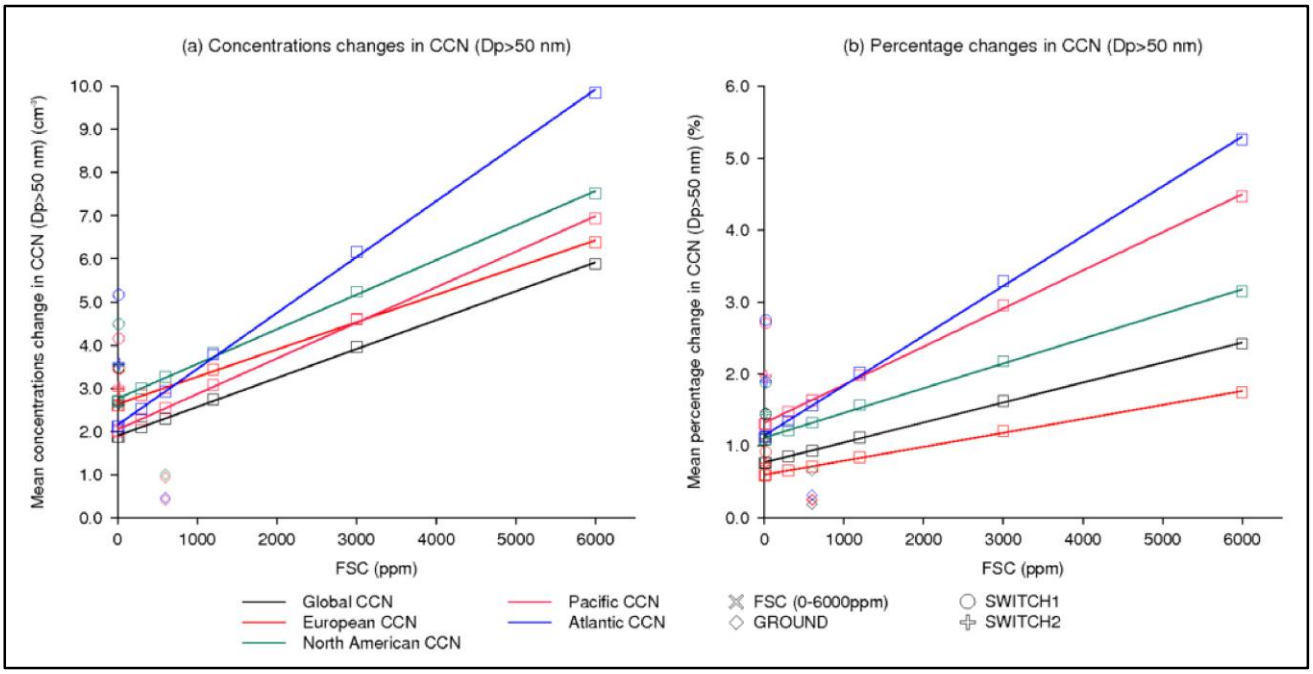


Fig. 8 shows the sensitivity of low level CCN concentrations to FSC. As with $PM_{2.5}$, we find simulated changes in CCN are near linear with respect to FSC ($R^2 > 0.99$ and p-value < 0.001 globally and for all individual regions).

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 desulfurisation of aviation fuel results in reductions in CCN in relation to standard aviation (DESUL–NORM), which follow the same regional trends (Fig. 8(a)).

3.4 Sensitivity of aerosol and ozone radiative effect to FSC

Fig. 9 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).

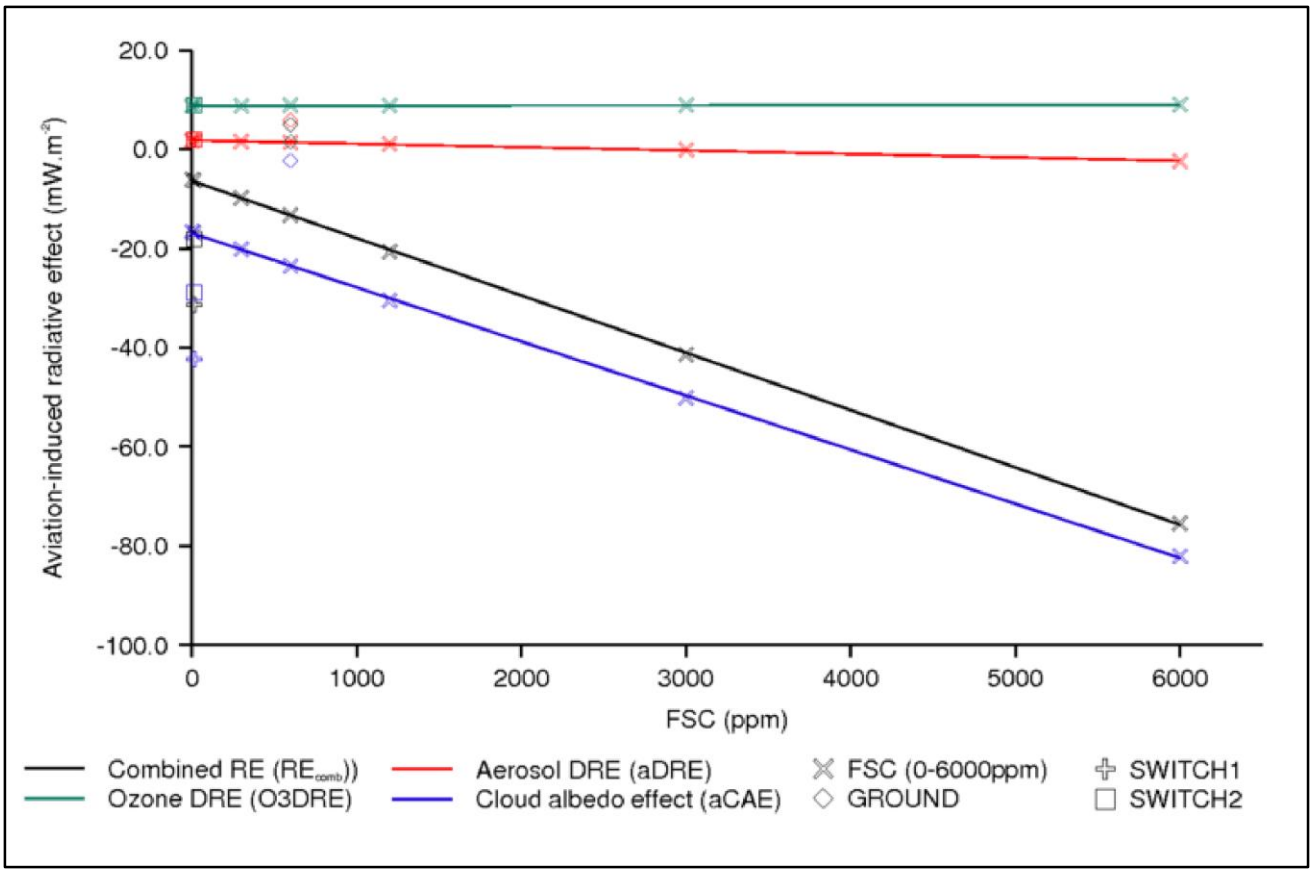
Our estimated aviation aerosol DRE [+1.4 mW m^{-2}] lies in the middle of the range given by previous work. The aviation aerosol DRE has been previously assessed as highly uncertain, ranging between –28 to +20 mW m^{-2} (Righi et al., 2013). Our estimated aviation-induced aCAE [–23.6 mW m^{-2}] lies 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 –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 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 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. 9).

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}].

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

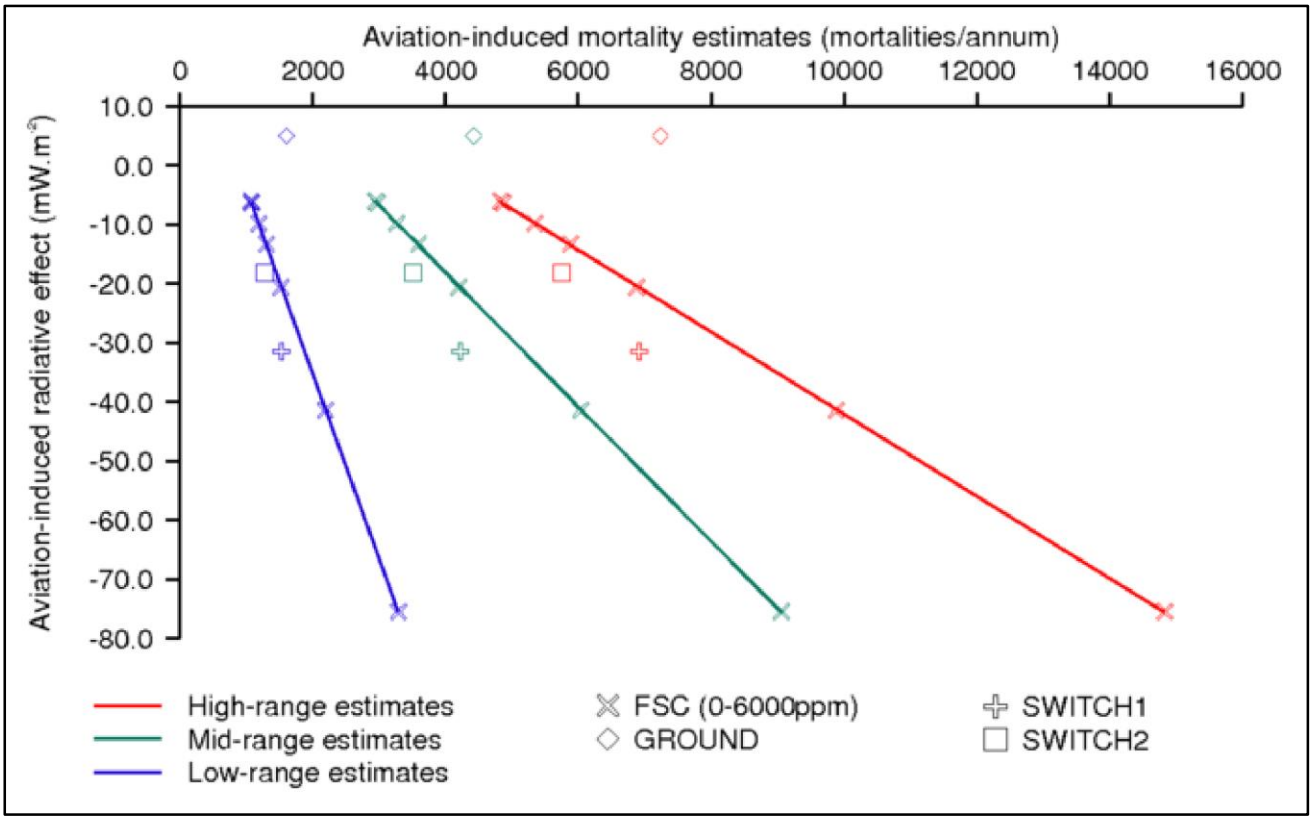


Increases in FSC result in reductions in the aerosol DRE (aDRE), changing from a positive aerosol DRE for low FSC scenarios, to a negative aerosol DRE for high FSC (FSC > 1200 ppm). As FSC is increased, we find the aCAE exhibits a larger cooling effect, i.e. becoming more negative with increases in FSC, 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. 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

Fig. 10 shows the net RE and premature mortality for different aviation emission scenarios. Increases in FSC lead to approximately linear increases in both estimated mortality and the negative net RE. We quantify the impact of FSC on mortality and REs in terms of d(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 $–1.2x10^{-2}$ mW m^{-2} ppm^{-1}, dominated by large changes to the aCAE [$–1.1x10^{-2}$ mW m^{-2} ppm^{-1}], and much smaller changes in the aDRE [$–6.9x10^{-4}$ mW m^{-2} ppm^{-1}] and O_3 RE [$+4.4x10^{-5}$ mW m^{-2} ppm^{-1}].

Fig. 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.



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 0–6000 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 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. 4). Greater surface layer $PM_{2.5}$ perturbations (GROUND–NORM) over populated regions increase aviation-induced annual mortality by +22.9% [+830 mortalities a^{-1}] (Fig. 6).

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 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 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 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 +5.0 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 mW m^{-2} (NORM; FSC = 600 ppm) to +1.5 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 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. 4). These changes increase aviation-induced mortality by +17.4% [+630 mortalities a^{-1}] (Fig. 6). 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% [–90 mortalities a^{-1}] compared to the standard aviation simulation (Fig. 6). 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.

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 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 1Tg(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} $Tg(N)^{-1}$] (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 mW m^{-2}] lies just outside the range provided by Gettelman and Chen (2013) and Righi et al. (2013) [–15.4 to –21 mW m^{-2}]. Our estimated aDRE [+1.4 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 3,600 premature mortalities annually 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% [0.04–0.18%] 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 $PM_{2.5}$ concentrations and subsequent increases in aviation-induced mortality. Increases in FSC also lead to a more negative RE_{comb} due to an enhanced aCAEs. We estimate that the use of ultra-low sulfur jet (ULSJ) fuel, with a FSC of 15 ppm, could prevent 620 [230–1,020] 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 of 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 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% [–90 mortalities a^{-1}] and the magnitude of the negative RE_{comb} was increased by 36.6%, providing an additional cooling impact of climate of –4.88mW 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. Future work needs to estimate the health impacts of aviation using newly available concentration response functions (Burnett et al., 2014).

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