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Public Health, Climate, and Economic Impacts of Desulfurizing Jet Fuel

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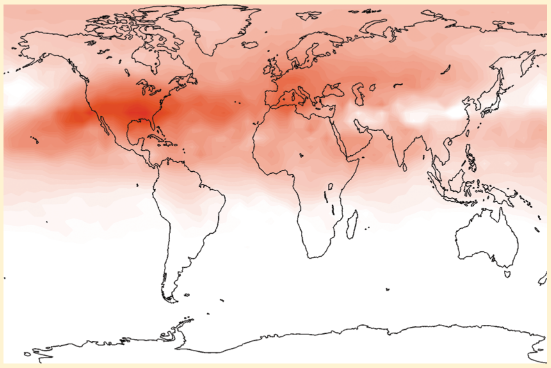
ABSTRACT: In jurisdictions including the US and the EU ground transportation and marine fuels have recently been required to contain lower concentrations of sulfur, which has resulted in reduced atmospheric SO_x emissions. In contrast, the maximum sulfur content of aviation fuel has remained unchanged at 3000 ppm (although sulfur levels average 600 ppm in practice). We assess the costs and benefits of a potential ultra-low sulfur (15 ppm) jet fuel standard ("ULSJ"). We estimate that global implementation of ULSJ will cost US$1−4bn per year and prevent 900−4000 air quality-related premature mortalities per year. Radiative forcing associated with reduction in atmospheric sulfate, nitrate, and ammonium loading is estimated at +3.4 mW/m^2 (equivalent to about 1/10th of the warming due to CO_2 emissions from aviation) and ULSJ increases life cycle CO_2 emissions by approximately 2%. The public health benefits are dominated by the reduction in cruise SO_x emissions, so a key uncertainty is the atmospheric modeling of vertical transport of pollution from cruise altitudes to the ground. Comparisons of modeled and measured vertical profiles of CO, PAN, O_3, and ^{7}Be indicate that this uncertainty is low relative to uncertainties regarding the value of statistical life and the toxicity of fine particulate matter.



1. INTRODUCTION

1.1. Context. Aircraft emissions impact the environment by perturbing the climate[1] and reducing air quality,[2] which leads to adverse health impacts including increased risk of premature mortality.[2−4] Aircraft landing and takeoff (LTO) emissions − i.e. emissions below 3000 ft above ground level − have been estimated to cause ∼200 air quality-related premature mortalities per year in the US.[4,5] While only LTO emissions have been regulated, Barrett et al.[2] estimated that full-flight aircraft emissions result in ∼10,000 premature mortalities per year globally, with the majority of impacts due to non-LTO emissions. With aviation demand forecast to grow at an average of 5% per year through 2030,[6] aircraft emissions and

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associated impacts may more than double within 20 years[3] with an even greater increase in public health impacts given population growth and changing non-aviation emissions influencing secondary particulate matter formation.[7]

As aircraft have a service life of approximately 25 years, technological improvements alone are unlikely to result in emissions reductions in the near-term. However, the US Federal Aviation Administration has an aspirational goal of halving aviation's significant health impacts by 2018 relative to a 2005 baseline.[8] If such a goal is to be approached, operational and fuel-related options also need to be considered. In this paper we assess the implications of a potential ultra-low sulfur jet fuel standard. We denote the policy option as "ULSJ" for brevity.

In 2006, UK Jet A-1, US Jet A, and US Department of Defense JP-8 fuel had an average fuel sulfur content (FSC) between 550 to 750 ppm (by mass),[9] well below the specification limit of 3000 ppm.[10,11] In 2006 the US introduced an ultra-low sulfur standard for highway diesel of 15 ppm.[12] Jurisdictions including Australia, Canada, New Zealand, Mexico, Japan, India, Argentina, Brazil, Chile, Peru, and the European Union all have instituted fuel sulfur standards of 50 ppm or less, effective by 2016 for road transportation. We adopt a baseline FSC for civil aviation of 600 ppm and assess the implications of a ULSJ policy case of 15 ppm.

1.2. Overview. We approach assessing the implications of ULSJ by determining the change in emissions and costs associated with desulfurization, where costs are estimated using two approaches. Changes in emissions are propagated to three atmospheric chemistry-transport models (CTMs) to estimate the effect of ULSJ on atmospheric composition, where the policy "delta" is identified as the difference in atmospheric composition between simulations with all emissions at their nominal values and simulations where aviation SO_x emissions have been reduced by 585/600 = 97.5%. The reduction in atmospheric sulfate loading due to lower aviation SO_x emissions is mapped to a radiative forcing (RF) and the change in lifecycle CO_2 emissions is estimated. Changes in fine particulate matter ($PM_{2.5}$) concentrations at the surface are overlaid on population density data to determine changes in human exposure to $PM_{2.5}$ by country. [This is on the basis that the majority of air quality-related health impacts are captured by considering $PM_{2.5}$.[4]] Epidemiological concentration–response functions (CRFs) relate changes in $PM_{2.5}$ exposure to expected changes in premature mortality, with consideration of variability across countries. Country-specific values of statistical life (VSL) are estimated to monetize air quality benefits of ULSJ. Short- and long-term climate impacts are monetized using a simplified impulse-response climate model and damage functions. Monetized costs, benefits, and disbenefits are aggregated into an overall benefit-cost analysis.

Where possible, uncertainties in parameters are estimated and propagated throughout the analysis, for which a Monte Carlo approach is used. An important atmospheric modeling uncertainty considered in detail is that of vertical transport of pollution from cruise altitudes to the surface, which we assess by comparing model simulations to measurements of relevant tracers.

2. METHODS AND ASSUMPTIONS

2.1. Fuel Properties. We define baseline jet fuel as having a FSC of 600 ppm and to be 86.2 wt % C.[9] ULSJ fuel with a FSC of 15 ppm is assumed to be obtained from baseline jet fuel by hydrodesulfurization (HDS), requiring H_2 created from refinery operations and steam reformation of natural gas. With assumptions detailed in section 5 of the Supporting Information (SI), we estimate the change in operating energy inputs to produce ULSJ instead of conventional petroleum. The largest share of this is an increase of refinery and natural gas consumption of 0.018 scm/L (standard cubic meters of gas per liter of jet fuel). Accounting for both CO_2 produced from HDS – which is assumed to be vented to the atmosphere – and overall process energy efficiency, we estimate a 2% increase in lifecycle CO_2 emissions for jet fuel associated with desulfurization, with bounds of 0–4%.

ULSJ fuel has 1% lower energy density and 0.3% higher specific energy relative to baseline fuel[9] and may need a corrosion inhibitor/lubricity improver (CI/LI) additive. ULSJ fuel could also have reduced aromatic composition, relative to conventional jet fuel, and this could result in reduced black carbon emissions. Implications of these factors for emissions are neglected since they are not expected to significantly impact the overall benefit-cost analysis results.

2.2. Emissions. Baseline civil aviation emissions of NO_x, SO_x, hydrocarbons (HC), black carbon (BC), and organic carbon (OC) are calculated using the FAA's aviation environmental design tool (AEDT)[13] for 2006. We estimate total fuel burn for 2006 at 188 Tg. This corresponds to 0.11 Tg of fuel-S, of which 98% is assumed to be emitted as SO_2 and the remaining 2% as S(VI).[14] Other assumptions are detailed in section 2.1 of the SI and are similar to Barrett et al.[2]

Hourly emissions are gridded from raw AEDT output for each CTM applied. For the ULSJ policy case, aircraft SO_x emissions are reduced by 97.5%. In addition to the baseline and ULSJ cases, corresponding cases are also assessed where only LTO emissions are included, to understand the relative contribution of LTO versus cruise emissions. Lifecycle CO_2 emissions are included in climate modeling. Background emissions in GEOS-Chem are described in Donkelaar et al.[15]

2.3. Chemistry-Transport Modeling. Three CTMs are used – GEOS-Chem,[16] CMAQ,[17] and p-TOMCAT.[18] GEOS-Chem is driven by GEOS-5 meteorological data from the NASA Global Modeling and Analysis Office (GMAO) with $0.5° \times 0.667°$ horizontal resolution. We apply it at a horizontal resolution of $4° \times 5°$ globally but with native $0.5° \times 0.667°$ resolution in a nested domain encompassing the contiguous US. Time-varying boundary conditions for the nested domain are taken from the global $4° \times 5°$ simulation. CMAQ is applied at a 36 km resolution for the contiguous US with time-varying boundary conditions from GEOS-Chem. A ± 60% uncertainty in population-weighted $PM_{2.5}$ is applied.[2]

MM5 is used to generate meteorology for CMAQ with GEOS-5 data as boundary and initial conditions. As such meteorology for CMAQ is consistent with GEOS-Chem. p-TOMCAT is used in an ancillary way (see section 2.2 of SI). All simulations were for 15 months, using October 2005 to December 2006 (inclusive) meteorology. The first three months are discarded as spin-up time so that steady state impacts are considered.

2.4. Climate Impacts. Warming related to (i) increased lifecycle CO_2 emissions from HDS and (ii) decreased SO_x emissions resulting in decreased direct aerosol cooling are considered. GEOS-Chem online calculations of aerosol optical depth for sulfates, nitrates, and ammonium (collectively called "sulfates") are related to radiative forcing (RF) (see section 3.1 of SI). This short-lived sulfate RF is incorporated into the

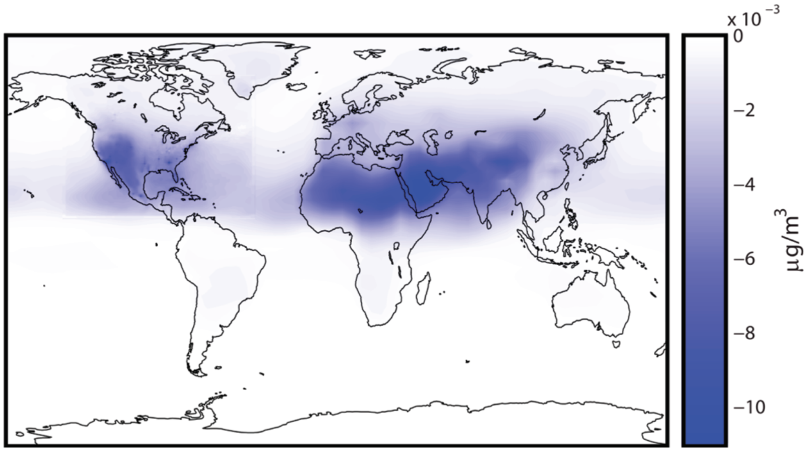

Figure 1. The annual average surface sulfate concentration change due to ULSJ as calculated by GEOS-Chem at 4° × 5°, with nested 0.5° × 0.667° results superimposed over the contiguous US.

aviation environmental portfolio management tool (APMT)-Impacts Climate Module,[18] which is used to assess the difference in climate impacts of a one year pulse of emissions under the baseline and ULSJ scenarios. The forcing associated with sulfate is assumed to decay instantaneously after the one year pulse of emissions ends,[19] while the carbon cycle implemented in APMT means that the CO_2 RF survives hundreds of years after the policy year being assessed. APMT monetizes climate impacts as described in Mahashabde et al.[19]

2.5. Health Impacts. The metric chosen for health impacts is premature mortalities due to long-term exposure to $PM_{2.5}$ as this is likely to capture >95% of the monetized health impacts of air pollution.[5] As done elsewhere,[20] we derive a CRF of an approximate 1% decrease in all-cause mortality in the US per 1 $\mu g/m^3$ decrease in annual average concentrations of $PM_{2.5}$, based on a Weibull distribution fit to the two major cohort studies in the US.[21,22] This value is comparable to the average value across the median estimates from experts in a recent EPA expert elicitation study.[23,24] We use lower and upper bound values of 0.4% and 1.8%, respectively, reflecting the uncertainty bounds from the Weibull distribution and comparable to the corresponding percentiles in the expert elicitation study. Results are similar to an EU expert elicitation study.[25]

Two issues arise in applying this CRF outside of the US. First, disease patterns may differ significantly from the US. To adjust for differences in contributors to baseline mortality, we assume that air quality-related premature mortalities are dominated by cardiopulmonary disease and lung cancer. We derive disease-specific CRFs in a manner that correspond to the aforementioned all-cause mortality CRF and apply these to other countries with different baseline disease rates. The method is detailed in section 4.2 of the SI.

Second, the slope of the CRF may not be linear through the range of concentrations observed globally. Prior work by the WHO in the context of global burden of disease modeling[26] used a log−linear rather than linear CRF to yield lower slope at higher concentrations, providing more realistic burden of disease estimates in developing countries. As the EPA CRF described above reflects more recent interpretations of health evidence in the US, we use it for our study but test the sensitivity of our findings to the use of the WHO methodology.

2.6. Benefit-Cost Analysis. HDS of jet fuel is considered as a cost and monetized climate disbenefits due to reduction of sulfate (direct) cooling and increased lifecycle CO_2 are accounted for. Benefits due to reduction in premature mortality globally are monetized.

Valuation of avoided premature mortalities in the US is based on a Weibull distribution fit to 26 wage-risk and contingent valuation studies, as done elsewhere.[20] The resulting VSL distribution (in 2006US$, based on 1990 income levels) had a mean of $7.4 m with lower and upper bounds of $1 m and $12 m, respectively. To develop appropriate VSL estimates for other countries, we used the gross national income for each country and an income elasticity range of 1−2, except where the resulting VSL would be less than the net present value of half lifetime earnings.[27] The EPA mortality lag structure is used in this analysis.[20] It assumes that 30% of avoided mortalities are seen in the year of implementation, 50% in years 2−5, and the remaining 20% spread out over years 6−20.

A range for the cost of HDS is estimated using two methods. First, historical US highway diesel prices are analyzed to determine the spread between ultra-low sulfur (<15 ppm) and low sulfur (15−500 ppm) fuel and low sulfur and high sulfur (>500 ppm) fuel. Second, the cost of natural gas and capital investment required for HDS is estimated.

Monetized climate disbenefits are calculated by APMT for a one-year pulse of emissions with discount rates of 2%, 3%, and 7%. Discount rate choice affects the valuation of lifecycle CO_2 disbenefits and health benefits of ULSJ. Discount rate is treated as a policy choice − not an uncertainty − because it is a quantitative expression of the extent to which costs and benefits in the future are valued relative to costs and benefits now.

Costs, benefits, and disbenefits are aggregated using a Monte Carlo analysis with input variables assigned triangular distributions corresponding to the lower, nominal, and upper values described. Results are given as an expectation with a 95% confidence interval (CI). Sensitivities to individual parameters are calculated with all other values held at their nominal value, and main and total effect indices are calculated.

2.7. Vertical Transport. As CTMs have rarely been used to assess the impact of cruise altitude emissions on surface air quality, we apply two approaches to evaluate the performance of GEOS-Chem (the model used for nominal results) with regard to vertical transport from the upper troposphere/lower stratosphere (UT/LS) to the surface. First, we compare vertical profiles of CO, O_3, and peroxyacetyl nitrate (PAN) from NASA aircraft missions to GEOS-Chem simulation results for 2006. The observations are averaged over chemically and geo-

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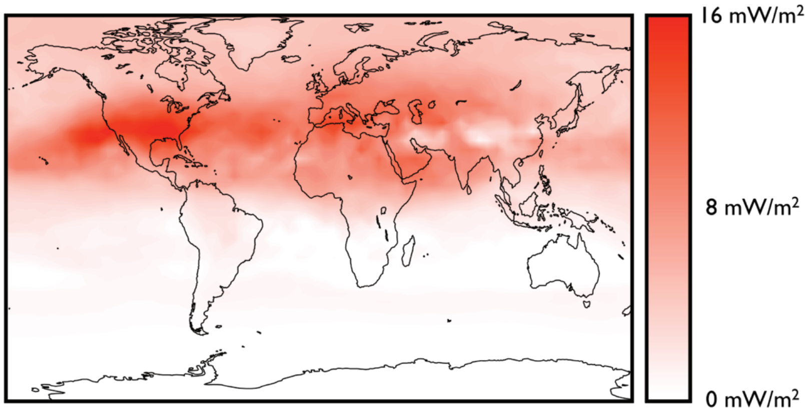

Figure 2. Change in sulfate direct climate forcing due to 97.5% removal of aviation fuel-S (i.e., a warming due to a decrease in cooling).

graphically coherent regions described by Wang et al.[28] and Bey et al.,[16] with updates from a more recent campaign TRACE-P.[29] Although all of these aircraft missions took place before 2006, the interannual variability of regionally averaged concentrations is sufficiently small that these observations are still useful to test model vertical transport.[16]

Second, we simulate beryllium-7 (^7Be) production and scavenging using GEOS-Chem. ^7Be is produced by cosmic ray spallation of N_2 and O_2 in the UT/LS,[30] is immediately taken up by aerosol particles, and is subsequently transported until loss by radioactive decay (half-life 53.3 d) or deposition to the surface. Its source distribution is relatively well-known, and there are extensive climatological observations from a global network of surface sites and from aircraft originally designed by the US Department of Energy (DOE) to monitor radioactive fallout. ^7Be has been used in numerous global model studies to test the simulation of vertical transport.[31−35] Here we conducted a 6-year GEOS-Chem simulation of ^7Be using 2004−2009 GEOS-5 meteorological data and the ^7Be source parametrization from Usoskin and Kovaltsov.[36] The ^7Be source depends on solar activity in a predictable manner, and we correspondingly scale the ^7Be observations following Koch et al.[32]

3. RESULTS

3.1. Surface $PM_{2.5}$ Impacts. Figure 1 shows results from GEOS-Chem for the change in aviation-attributable annual average surface sulfate concentration for 2006. High resolution nested GEOS-Chem results are superimposed on the US, where it can be seen that boundaries match most closely in upwind directions. Localized (negative) peaks in the US correspond to locations of airports, the impacts of which are resolved by the high resolution nested domain. Elsewhere the widespread impacts are dominated by cruise emissions, which occur primarily over North America and Europe, but impact the surface by subsidence. The largest effects are in the strongly subsiding arid regions of the subtropics. Impacts do not penetrate the intertropical convergence zone (ITCZ).

Overall ULSJ decreases the surface average sulfate concentration by 9.6×10^{-4} $\mu g/m^3$ and ammonium by 2.54×10^{-4} $\mu g/m^3$. It increases nitrate by 2.7×10^{-5} $\mu g/m^3$ due to the greater availability of ammonia to form ammonium nitrate when sulfate decreases.

Figure 1 shows that reductions in ground-level aviation-attributable sulfate concentrations due to ULSJ are greater in North Africa and the Middle East than Europe. This is because the average circulation means that emissions (and therefore emissions reductions) at cruise altitudes over both Europe and North America impact surface air quality relatively strongly in North Africa and the Middle East.[2] Specifically, emissions at cruise altitude occur in westerly winds of >10 m/s on average, while air subsides along the subtropical ridge (∼35°N), and particularly strongly in North Africa and the Middle East as shown in the SI (section 2.3.2).

3.2. Health Impacts. Using the EPA-derived CRF and global GEOS-Chem results, ULSJ causes a reduction of ∼2300 premature mortalities per year (95% CI: 890−4200), of which ∼120 are in the US (95% CI: 46−210) when changes in total ground-level $PM_{2.5}$ concentrations are considered. Ostro[26] estimated that ∼800,000 premature mortalities per year are attributable to fine particulate matter air pollution. ULSJ represents 0.3% of this figure, although the methods used in our estimate differ significantly from the Ostro estimate.

Using results from nested GEOS-Chem in the contiguous US increases the mortalities avoided by ULSJ by ∼20 or 17%. Contiguous US CMAQ calculations indicate 85% more avoided premature mortalities in the US than nested GEOS-Chem, or 92% greater than global GEOS-Chem, where CMAQ predicts a total of ∼230 avoided mortalities. Applying the older WHO CRF as a sensitivity, ULSJ results in ∼1500 avoided premature mortalities globally per year, of which ∼140 are in the US. The global mortality estimate is reduced given the lower CRF applied in countries with higher ambient $PM_{2.5}$ concentrations, such as China and India.

Avoided premature mortalities for selected other countries using the EPA-derived CRF are as follows: India, ∼870; China, ∼220; Pakistan, ∼95; Germany, ∼83; Russia, ∼73; Egypt, 39; UK, ∼25; France, ∼21; and Saudi Arabia, ∼11. Countries where aviation-attributable baseline $PM_{2.5}$ is dominated by nitrates benefit from ULSJ relatively less than sulfate-rich countries. For example, baseline aviation-attributable $PM_{2.5}$ exposure in China is 73% nitrate (excluding ammonium mass), and ULSJ results in a 1% reduction in aviation-attributable $PM_{2.5}$ exposure. On the other hand, baseline exposure in Saudi Arabia is 80% sulfate and ULSJ results in a 47% reduction. Globally, aviation-attributable $PM_{2.5}$ exposure is reduced by 6% by ULSJ. We also note that population density plays an important role. For example, the peak sulfate concentration reduction under ULSJ occurs in Saudi Arabia − which has a population density of 12/km² − where ∼11 premature mortalities per year are avoided. This can be compared to Pakistan − which has a population density of 214/

km^2 – where ∼95 premature mortalities per year are avoided even though the sulfate concentration reduction is lower than Saudi Arabia.

3.3. Climate Impacts. Figure 2 shows the (warming) radiative forcing due to the reduction in aircraft SO_x emissions and resultant reduction in sulfate direct climate forcing. The average ULSJ-attributable warming due to this mechanism is +3.3 mW/m^2 (95% CI: 1.4–6.0) globally or +6.1 mW/m^2 (95% CI: 2.6–11.2) for the northern hemisphere.

Applying a one-year pulse of emissions in the APMT-Impacts Climate Module, the time-integrated forcing out to +800 years of ULSJ is +3.1 mW/m^2·yrs due to lifecycle CO_2 changes and +3.3 mW/m^2·yrs due to SO_4 reduction. The equivalent central estimates for temperature response are +2.3 mK.yrs and +2.5 mK·yrs. This indicates that discounting future climate impacts will increasingly weight the importance of the sulfate direct climate-forcing component of the climate disbenefit of ULSJ.

We note that calculations by Unger[37] using ModelE indicate that reducing aircraft SO_x emissions may result in net cooling due to increased nitrate loading resulting from increased free ammonia, which is inconsistent with our findings. Unger did not include the increased CO_2 emissions associated with ULSJ and performed 10-year climate simulations (with a 2 year spin-up), and so results are not directly comparable to the present study. Unger finds that under the baseline (600 ppm fuel sulfur content) scenario, aviation results in a nitrate warming of approximately +6 mW/m^2, i.e. that aviation emissions result in nitrate aerosol reduction on net. Under ULSJ, this switches to a nitrate aerosol increase due to aviation and a nitrate RF of −1 mW/m^2. The reason for this change in sign is not described, but Unger notes that future work will concentrate on the tropospheric distribution of NH_3. Our calculations, on the other hand, do not indicate that the nitrate "bounce-back" RF exceeds the reduction in sulfate RF or a change in sign of the nitrate RF. Specifically, we estimate that reducing aircraft SO_x emissions (i.e., ULSJ) results in a total sulfate-ammonium-nitrate RF of +3.4 mW/m^2, whereas sulfate alone contributes +5.3 mW/m^2. This can be compared to the central estimate for aviation's sulfate RF by Lee et al.[1] of −4.8 mW/m^2 (where here only the magnitude is important), implying that our sulfate results are consistent with other studies. Our calculations therefore indicate a nitrate bounce-back RF of −1.9 mW/m^2, compared to −7 mW/m^2 by Unger. Unger's baseline aviation sulfate RF is approximately −8 mW/m^2, which is about 60% higher in magnitude than our central estimate or the Lee et al. review. We finally note that there is empirical evidence to suggest that (at least over a period of years) high altitude SO_x emissions result in cooling, i.e. that reducing high altitude SO_x emissions would be warming (consistent with our result). Specifically, in an analysis of the atmosphere after the 1991 eruption of Mt. Pinatubo, McCormick et al.[38] found that mean tropospheric temperatures decreased through 1993 due to increases in sulfuric acid aerosol. We did not quantitatively account for Unger's result that reducing aircraft SO_x emissions would be net cooling in our assessment of ULSJ due to the empirical evidence, our calculations, and that the finding was noted by Unger to require further work, but we note that the warming climate implications of ULSJ may be overestimated in our calculations.

3.4. Additional Production Costs. Analyzing US Energy Information Administration price history data (from 2001 to 2011) for the introduction of ultra-low sulfur highway diesel fuel – which has similar properties to Jet-A/A1 – we estimate that desulfurizing fuel costs 3.7–6.6 ¢/gal. (1¢ = US2006 $0.01 and 1 gal. = 3.785 L). This can be compared to estimates by QinetiQ – an additional production cost of 4.5–6.7 ¢/gal.[39]

As an alternative approach, we estimate capital and feedstock costs directly using a representative refinery (see section 5.2 of the SI). Natural gas is estimated to cost of 1–3 ¢/gal. of Jet A/A-1 produced with a corresponding capital cost (with depreciation over 30 years) of 0.6 ¢/gal. This gives a total of 1.6–3.6 ¢/gal. Combining this range with the price history data listed above, we determine a nominal value of 3.7 ¢/gal., with lower and upper bounds of 1.6 and 6.6 ¢/gal., respectively.

Scaling this to total civil aviation fuel burn, ULSJ will cost $2.5bn (95% CI: 1.3–3.8) in US2006$ globally or $0.89bn (95% CI: 0.5–1.4) for the US portion of fuel burn.

In this analysis, implementation costs are based on US refining prices given the availability of product pricing data. We estimate a potential error of −7 to 4% in global desulfurization costs relative to US prices (see section 5.3 of the SI).

3.5. Benefit-Cost Analysis. The central nondiscounted public health benefit of ULSJ is estimated at $2.5bn/year when only mortality impacts are considered, but when the same discount rate as applied to climate costs is applied to health benefits, the central monetized health benefit estimate is $[1.8, 1.8, 1.6]bn/yr, while the central monetized climate damage estimate is $[2.1, 1.5, 0.7]bn/year for a [2, 3, 7]% discount rate choice. Corresponding lower and upper bounds for health benefits are $[0.21, 0.20, 0.18] and $[7.6, 7.3, 6.7] bn/yr, respectively, and for climate disbenefits are $[0.13, 0.10, 0.06]bn/year and $[6.3, 4.3, 2.1]bn/year, respectively. There is a [46, 57, 77]% chance that public health benefits exceed climate disbenefits. (See section 6.4 of the SI for tabulated components of costs and benefits with confidence intervals.)

Figure 3 depicts the probability distribution of the overall net difference between costs and benefits of ULSJ (i.e., benefits minus costs and disbenefits). There is a [78, 77, 77]% chance that the policy has a net negative benefit when including implementation costs and climate disbenefits.

If all avoided premature mortalities are valued using the aforementioned US VSL range – instead of country-specific VSLs – then there is an 84% chance that ULSJ is net beneficial

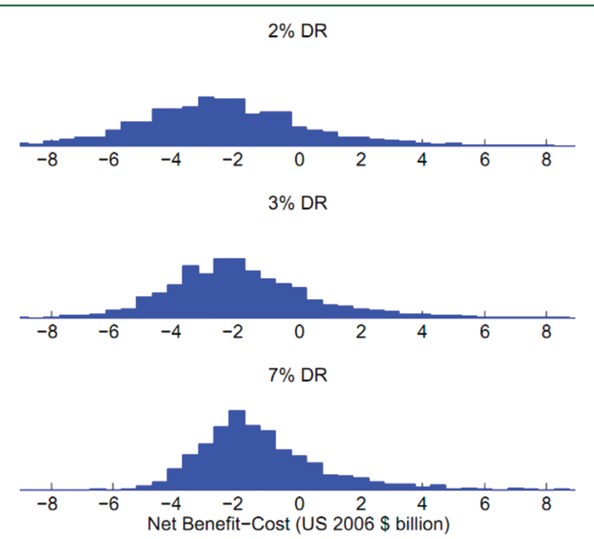

Figure 3. The probability distribution for yearly net cost (-) or net benefit (+) of ULSJ under global implementation for discount rates of 2%, 3%, and 7%. Country-specific VSLs are used.

(taking a discount rate of 3%) with an equal likelihood that the net benefit is higher or lower than \$12bn/year.

For the US, public health benefits also exceed climate disbenefits, and there is a greater than even chance of a net policy cost when including implementation costs.

The greatest quantified contributors to uncertainty in rank order by total effect index (see section 7.2 of the SI) are the US VSL, $PM_{2.5}$ mortality CRF, modeled aviation-attributable $PM_{2.5}$, the cost of HDS, and the climate damage function in APMT. A significant issue is that if only LTO emissions and benefits associated with the reduction in LTO SO_x emissions are accounted for − as is conventional when considering aviation's impact on air quality − our results show that ULSJ has a statistically significant net cost (see section 6 of the SI). This implies that the ability of CTMs to correctly capture vertical transport from cruise altitudes and scavenging is of central importance.

3.6. Vertical Transport Assessment. Figure 4 compares model results with the climatological observations of ^{7}Be averaged over 10° latitude bins. The model is sampled at the month and location of the observations (solid lines), and the zonal mean is also given (dotted lines). The top panel evaluates the model ^{7}Be emissions source by comparing with the UT/LS aircraft observations of the DOE Radionuclide Database (RANDAB).[40] The middle panel compares model results with the ^{7}Be wet deposition fluxes aggregated by Koch et al.,[32] which provide an additional test of the model source since the dominant ^{7}Be removal in the troposphere is by wet deposition. The bottom panel compares the model surface air concentrations with long-term observations from the DOE Surface Air Sampling Program.[41] We see from Figure 4 that GEOS-Chem reproduces successfully the magnitudes and latitudinal patterns of the ^{7}Be observations. Comparison to RANDAB indicates a model source bias of $-4 \pm 2\%$. Comparison to observed surface air concentrations indicates a bias of $-18 \pm 6\%$ globally and <10% over the US. The ^{7}Be source on average is 60% stratospheric and 40% tropospheric, and Dutkiewicz and Husain[42] deduced from observed ^{90}Sr/^{7}Be ratios that ~25% of surface ^{7}Be at northern midlatitudes is of stratospheric origin. We find the same fraction in GEOS-Chem, which tests the model simulation of stratosphere-troposphere exchange and implies that the model biases estimate above should be insensitive to the precise distribution of the aerosol source within the UT/LS. Simulated vertical profiles of CO, O_3, and PAN are compared to measurements in section 9.1 of the SI, demonstrating that GEOS-Chem captures the vertical profiles of these species.

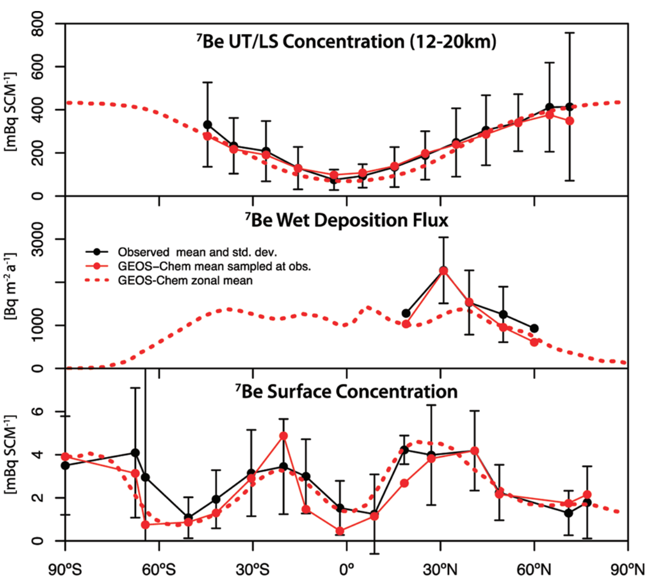

Figure 4. Latitudinal profiles of cosmogenic ^{7}Be as a test of the GEOS-Chem model simulation of vertical transport of aerosols from the UT/LS to the surface. Observations (black lines) are averaged over 10° latitude bins. GEOS-Chem results for 2004−2009 are sampled at the month and location of observations (red lines) and the model zonal mean is also given (dotted lines). The top panel shows DOE RANDAB UT/LS aircraft data from 1957 to 1983, the middle panel shows annual mean wet deposition flux data compiled by Koch et al.,[32] and the bottom panel shows DOE SASP surface air concentration data for 1957−1999. Error bars indicate the variability ($\pm\sigma$) across sites in the wet deposition flux data and across the spatial, seasonal, and interannual variability of the RANDAB and SASP samples for each bin. The GEOS-Chem simulation is conducted for average solar activity conditions (Φ = 670 MV with Usokin and Kovaltsov[35]). The RANDAB and wet deposition flux data are adjusted for average solar activity following Koch et al.,[32] while the surface air observations are filtered for average solar activity (Φ = 520−820 MV from Usoskin et al.[43]). Concentrations are represented by the S.I. unit for radioactivity per cubic meter air at 0 °C and 1 atm.

4. DISCUSSION

Jet A/A-1 is unusual as a transportation fuel in not being subject to a current or planned ultra-low sulfur standard in developed countries. Within the context of the FAA's aspirational goal of reducing aviation's significant health impacts in 2018 by 50% relative to 2005, and considering the time constants of technology changes combined with anticipated growth, ULSJ may be a suitable option. Without ULSJ, the FAA will likely have increased difficulty meeting this 2018 goal because of the anticipated increase in jet fuel sulfur content that will be a natural result of increasing sulfur content in petroleum.[44] In the US, the final rule mandating ultra-low sulfur diesel fuel was made in 2001, with 80% of the fuel being imported/produced required to meet a 15 ppm specification in 2006 and 100% in 2010. This implies that progress toward ULSJ implementation may be possible by 2018 in a US context. However, the implications of ULSJ are intrinsically international due to the intercontinental nature of aircraft pollution caused by cruise emissions.[2] Furthermore, as aircraft refuel in different countries (e.g., flights from Europe to North America will be fueled in Europe), consideration of ULSJ at an international level may be justified.

We have shown that the net benefit of ULSJ may be positive or negative given the uncertainties captured, with a greater than 50% chance that the additional feedstock and capital costs coupled with climate disbenefits exceed public health benefits when country-specific VSLs are applied. 900−4000 premature mortalities per year will be averted under a ULSJ scenario. There are appreciable uncertainties that indicate the possibility of either positive or negative net benefits. This indicates that ULSJ may be justifiable.

Although there were many contributors to uncertainty, the greatest quantified contributor was the VSL, which has significant uncertainty within a US context and heightened uncertainty in a global application. We note that if the US VSL were applied to all countries, then ULSJ would be cost-beneficial with global public health benefits at ~\$12bn. There is no economic rationale for doing this, given significant differences in national income. However, policy-makers may be more comfortable with approaches that use a single VSL across countries. An alternative strategy would be to avoid valuation of mortality and conduct a cost-effectiveness analysis. When doing so, we find a central estimate of $1.97 million per premature mortality averted (95% interval: 0.73−5.66), where the costs include both implementation costs and climate disbenefits for a 3% discount rate.

Our analysis also reinforced the importance of cruise emissions to public health impacts of aviation. If only LTO emissions were included, the public health benefits are significantly outweighed by the costs of implementation and climate disbenefits. This emphasizes the importance of appropriately capturing vertical transport from cruise altitudes, and our comparisons between modeled and simulated tracers indicate that vertical transport and wet removal rates are captured in the model applied with an uncertainty that is small relative to other modeling uncertainties. Climate-feedbacks and indirect effects of reduced atmospheric sulfate and CO_2 concentrations have not been assessed but are potentially significant. Additionally, two factors may mean that we have overestimated the warming associated with ULSJL: (i) new results indicate that the nitrate bouce-back may offset the decrease in sulfate RF and (ii) the 2% of fuel-S emitted as S(VI) may increase the warming due to aviation black carbon emissions by optical focusing, implying that ULSJ may decrease the (highly uncertain) black carbon RF.

Finally, we note that the pubic health benefits and sulfate-related climate impacts of desulfurizing jet fuel calculated here would equally apply to alternative jet fuels that are sulfur-free. Other components of this analysis − including net climate impacts and additional production costs − would differ.

ASSOCIATED CONTENT

Supporting Information

Further discussion, analyses, and results. This material is available free of charge via the Internet at http://pubs.acs.org.

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Notes

The authors declare no competing financial interest.

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REFERENCES

(1) Lee, D. S.; Pitari, G.; Grewe, V.; Gierens, K.; Penner, J. E.; Petzold, A.; Prather, M. J.; Schumann, U.; Bais, A.; Berntsen, T.; Iachetti, D.; Lim, L. L.; Sausen, R. Transport impacts on atmosphere and climate: Aviation. *Atmos. Environ.* **2010**, *44* (37), 4678−4734.

(2) Barrett, S. R. H.; Britter, R. E.; Waitz, I. A. Global mortality attributable to aircraft cruise emissions. *Environ. Sci. Technol.* **2010**, *44* (19), 7736−7742.

(3) Woody, M.; Baek, B. H.; Adelman, Z.; Omary, M.; Lam, Y. F.; West, J. J.; Arunachalam, S. An assessment of Aviation's contribution to current and future fine particulate matter in the United States. *Atmos. Environ.* **2011**, *45*, 3424−3433.

(4) Ratliff, G.; Sequeira, C.; Waitz, I.; Ohsfeldt, M.; Thrasher, T.; Graham, G.; Thompson, T.; Graham, M.; Thompson, T. *Aircraft Impacts on Local and Regional Air Quality in the United States.* PARTNER report (Report No. PARTNER-COE-2009-002).

(5) Brunelle-Yeung, E.; Masek, T.; Rojo, J. J.; Levy, J. I.; Arunachalam, S.; Miller, S. M.; Barrett, S. R. H.; Kuhn, S. R.; Waitz, I. A. *Methods for assessing the impact of aviation environmental policies on public health. To appear in Transport Policy* **2011**.

(6) The Boeing Company. *Boeing Current Market Outlook 2011−2030*; Boeing, U.S., 2011.

(7) Levy, J. I.; Woody, M.; Baek, B. H.; Shankar, U.; Arunachalam, S. Current and future particulate matter-related mortality risks in the United States from aviation emissions during landing and takeoff. *Risk Anal.* **2011**, in press. DOI: 10.1111/j.1539-6924.2011.01660.x.

(8) Federal Aviation Administration. *Destination 2025: Executive Summary*. FAA, 2011. http://www.faa.gov/about/plans_reports/media/Destination2025.pdf (accessed September 16, 2011).

(9) Hileman, J. I.; Stratton, R. W.; Donohoo, P. E. Energy content and alternative jet fuel viability. *J. Propul. Power* **2010**, *26* (6), 1184−1195.

(10) ASTM International, West Conshohocken, PA. *Standard Specification for Aviation Turbine Fuels, Std. ASTM D1655-09*, 2007. DOI:10.1520/D1655-09.

(11) UK Ministry of Defence. *Defence Standard 91−91, Issue 7 (18 February 2011), Turbine Fuel, Kerosine Type, Jet A-1, NATO Code: F-35, Joint Service Designation: AVTUR*, 2011.

(12) United States Environmental Protection Agency. *Control of Air Pollution From New Motor Vehicles; Revisions to Motor Vehicle Diesel Fuel Sulfur Transition Provisions; and Technical Amendments to the Highway Diesel, Nonroad Diesel, and Tier 2 Gasoline Programs: Direct final rule*; EPA, U.S., 2005.

(13) Wilkerson, J. T.; Jacobson, M. Z.; Malwitz, A.; Balasubramanian, S.; Wayson, R.; Fleming, G.; Naiman, A. D.; Lele, S. K. Analysis of emission data from global commercial aviation: 2004 and 2006. *Atmos. Chem. Phys.* **2010**, *10*, 6391−6408.

(14) Stettler, M. E. J.; Eastham, S.; Barrett, S. R. H. Air Quality and Public Health Impacts of UK Airports. Part I: Emissions. *Atmos. Environ.* **2011**, In press, DOI: 10.1016/j.atmosenv.2011.07.012.

(15) Van Donkelaar, A.; Martin, R. V.; Leaitch, W. R.; MacDonald, A. M.; Walker, T. W.; Streets, D. G.; Zhang, Q.; Dunlea, E. J.; Jimenez, J. L.; Dibb, J. E.; Huey, L. G.; Weber, R.; Andreae, M. O. Analysis of aircraft and satellite measurements from the Intercontinental Chemical Transport Experiment (INTEX-B) to quantify long-range transport of East Asian sulfur to Canada. *Atmos. Chemi. Phys.* **2008**, *8* (11), 2999−3014.

(16) Bey, I.; Jacob, D. J.; Yantosca, R. M.; Logan, J. A.; Field, B.; Fiore, A. M.; Li, Q.; Liu, H.; Mickley, L. J.; Schultz, M. Global modeling of tropospheric chemistry with assimilated meteorology: Model description and evaluation. *J. Geophys. Res.* **2001**, *106* (23), 23,073−23,096.

(17) Byun, D. W.; Schere, K. L. Review of the Governing equations, Computational Alogrithms, and Other Components of the Models-3 Community Multiscale Air Quality (CMAQ) Modeling System. *Appl. Mech. Rev.* **2006**, *59* (2), 51−77.

(18) Chipperfield, M. P. New Version of the TOMCAT/SLIMCAT Off-Line Chemical Transport Model: Intercomparison of Strato-

spheric Tracer Experiments *Q. J. R. Meteorol. Soc.* **2006**, *132*, 1179–1203, doi:10.1256/qj.05.51, .

(19) Mahashabde, A.; Wolfe, P.; Ashok, A.; Dorbian, C.; He, Q.; Fan, A.; Lukachko, S.; Mozdzanowska, A.; Wollersheim, C.; Barrett, S. R. H.; Locke, M.; Waitz, I. A. Assessing the environmental impacts of aircraft noise and emissions. *Prog. Aerosp. Sci.* **2011**, *47* (1), 0376–0421.

(20) US Environmental Protection Agency. *The Benefits and Costs of the Clean Air Act: 1990 to 2020*; Office of Air and Radiation: Washington, DC, 2011.

(21) Pope, C. A.; Burnett, R. T.; Thun, M. J.; Calle, E. E.; Krewski, D.; Ito, K.; Thurston, G. D. Lung cancer, cardiopulmonary mortality, and long-term exposure to fine particulate air pollution. *JAMA* **2002**, *287* (9), 1132–1141.

(22) Laden, F.; Schwartz, J.; Speizer, F. E.; Dockery, D. W. Reduction in fine particulate air pollution and mortality: Extended follow-up of the Harvard Six Cities study. *Am. J. Respir. Crit. Care Med.* **2006**, *173* (6), 667–672.

(23) United States Environmental Protection Agency. *Expanded Expert Judgment Assessment of the Concentration-Response Relationship Between $PM_{2.5}$ Exposure and Mortality: Final Report*; EPA, U.S., 2006.

(24) Roman, H. A.; Walker, K. D.; Walsh, T. L.; Conner, L.; Richmond, H. M.; Hubbell, B. J.; Kinney, P. L. Expert judgment assessment of the mortality impact of changes in ambient fine particulate matter in the U.S. *Environ. Sci. Technol.* **2008**, *42* (7), 2268–2274.

(25) Cooke, R. M.; Wilson, A. M.; Tuomisto, J. T.; Morales, O.; Tainio, M.; Evans, J. S. A probabilistic characterization of the relationship between fine particulate matter and mortality: Elicitation of european experts. *Environ. Sci. Technol.* **2007**, *41* (18), 6598–6605.

(26) Ostro, B. *Outdoor air pollution: Assessing the environmental burden of disease at national and local levels. Environmental Burden of Disease Series, No. 5*; World Health Organization: 2004.

(27) Hammitt, J. K.; Robinson, L. A. The Income Elasticity of the Value per Statistical Life: Transferring Estimates between High and Low Income Populations. *J. Benefit-Cost Anal.* **2011**, *2*(1).

(28) Wang, Y.; Logan, J. A.; Jacob, D. J. Global simulation of tropospheric O3-NOx-hydrocarbon chemistry, 2. Model evaluation and global ozone budget. *J. Geophys. Res.* **1998**, *103/D9*, 10,727–10,756.

(29) Jacob, D. J.; Crawford, J. H.; Kleb, M. M.; Connors, V. S.; Bendura, R. J.; Raper, J. L.; Sachse, G. W.; Gille, J. C.; Emmons, L.; Heald, C. L. The Transport and Chemical Evolution over the Pacific (TRACE-P) aircraft mission: design, execution, and first results. *J. Geophys. Res.* **2003**, *108*, 9000 DOI: 10.1029/2002JD003276.

(30) Lal, D. M.; Malhotra, P. K.; Peters, B. On the production of radioisotopes in the atmosphere by cosmic radiation and their application to meteorology. *J. Atmos. Terr. Phys.* **1958**, *12* (4), 306–328.

(31) Brost, R. A.; Feichter, J.; Heimann, M. 3-dimensional simulation of Be-7 in a global climate model. *J. Geophys. Res.* **1991**, *96* (D12), 22423–22445.

(32) Koch, D. M.; Jacob, D. J.; Graustein, W. C. Vertical transport of tropospheric aerosols as indicated by Be-7 and Pb-210 in a chemical tracer model. *J. Geophys. Res.* **1996**, *101* (D13), 18651–18666.

(33) Liu, H.; Jacob, D. J.; Bey, I.; Yantosca, R. M. Constraints from ^{210}Pb and ^{7}Be on wet deposition and transporting a global threee-dimensional chemical tracer model driven by assimilated meteorological fields. *J. Geophys. Res.* **2001**, *106* (D11), 12109–12128.

(34) Allen, D. J.; Dibb, J. E.; Ridley, B.; Pickering, K. E.; Talbot, R. W. An estimate of the stratospheric contribution to springtime tropospheric ozone maxima using TOPSE measurements and beryllium-7 simulations. *J. Geophys. Res.* **2003**, *108* (D4), 8355.

(35) Liu, H.; Considine, D. B.; Horowitz, L. W.; Crawford, J. H.; Strahan, S. E.; Damon, M. R.; Rodriguez, J. M.; Xu, X.; Carouge, C.; Yantosca, R. M. Using beryllium-7 to assess cross-tropopause transport in global models. *J. Geophys. Res.* **2010**, submitted for publication.

(36) Usoskin, I. G.; Kovaltsov, G. A. Production of cosmogenic Be-7 isotope in the atmosphere: Full 3-D modeling. *J. Geophys. Res.* **2008**, *113* (D12), D12107.

(37) Unger, N. Global climate impact of civil aviation for standard and desulfurized jet fuel. *Geophys. Res. Lett.* **2011**, *38* (20).

(38) McCormick, M. P.; Thomason, L. W; Trepte, C. R. Atmospheric effects of the Mt. Pinatubo eruption. *Nature* **1995**, *373*, 399–404.

(39) QinetiQ *Reduction of sulphur limits in aviation fuel standards (SULPHUR)*; Report QinetiQ/09/01835 Issue 1.1; Farnborough, 2010. http://www.easa.eu.int/rulemaking/docs/research/EASA_SULPHUR_Project_11-01-2010.pdf (accessed September 16, 2011).

(40) Carbon Dioxide Information Analysis Center. *The Environmental Measurements Laboratory's Stratospheric Radionuclide Database (RANDAB)*. http://cdiac.ornl.gov/epubs/db/db1019/db1019.html (accessed September 16, 2011).

(41) National Urban Security Technology Laboratory. *Surface Air Sampling Program (SASP)*. http://www.eml.st.dhs.gov/databases/ (accessed Oct 17, 2010).

(42) Dutkiewicz, V. A.; Husain, L. Stratospheric and tropospheric components of Be-7 in surface air. *J. Geophys. Res.* **1985**, *90* (ND3), 5783–5788.

(43) Usoskin, I. G.; Alanko-Huotari, K.; Kovaltsov, G. A.; Mursula, K. Heliospheric modulation of cosmic rays: Monthly reconstruction for 1951–2004. *J. Geophys. Res.* **2005**, *110* (A12), A12108.

(44) Brown, W. *Changing Trends in the Refining Industry*. www.eia.gov/oiaf/aeo/otheranalysis/aeo_2006analysispapers/tri.html (accessed September 16, 2011).