Supplementary Information: Global impacts of aviation on air quality evaluated at high resolution

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Seasonal patterns of exposure and effect of exposure response function

The 74,300 mortalities attributed to aviation emissions in this work is greater than previous estimates, which have ranged from 10,000 (Barrett et al., 2010) to 58,000 (Quadros et al., 2020). As discussed in the main text, this is in part due to our use of the same Turner et al. (2015) epidemiological data for ozone exposure as was used in Quadros et al. (2020).

Compared to the Jerrett et al. (2009) data used by previous assessments (e.g. Eastham and Barrett (2016), Yim et al. (2015)), Turner et al. (2015) relates mortality to the annual mean 8-hour maximum daily average (MDA8) ozone exposure rather than the ozone season 1-hour maximum daily average (MDA1). MDA1 is by definition always equal to or greater than MDA8, but the differences over the US are typically 10-20% (Seltzer et al., 2020). Turner et al. find that respiratory mortality increases by 12% for each 10 ppb increase in annual mean MDA8 ozone, compared to Jerrett et al. who find a 4% increase in respiratory mortality for each 10 ppb increase in ozone season MDA1 ozone. Since ozone levels are higher during ozone season, the distinction between 4 and 12% does not necessarily indicate greater mortality. Malley et al. (2017) found that shifting from the Turner et al. risk estimate to that of Jerrett et al. increased global ozone-attributable mortality by a factor of 2.2 to 2.6, but we find an increase of 3.2 times (assuming the same diseases are affected). This greater fraction is because the peak in ozone exposure resulting from aviation emissions occurs outside of ozone season, as shown in Figure S1. This effect is also illustrated in a video which shows the diurnal variation in surface $PM_{2.5}$ attributable to aviation emissions, on a $0 - 0.6$ µg/m^3 scale and starting with no spin up: https://www.dropbox.com/s/onfkrwliltv6r1w/Surface%20AQ.mp4?dl=0.
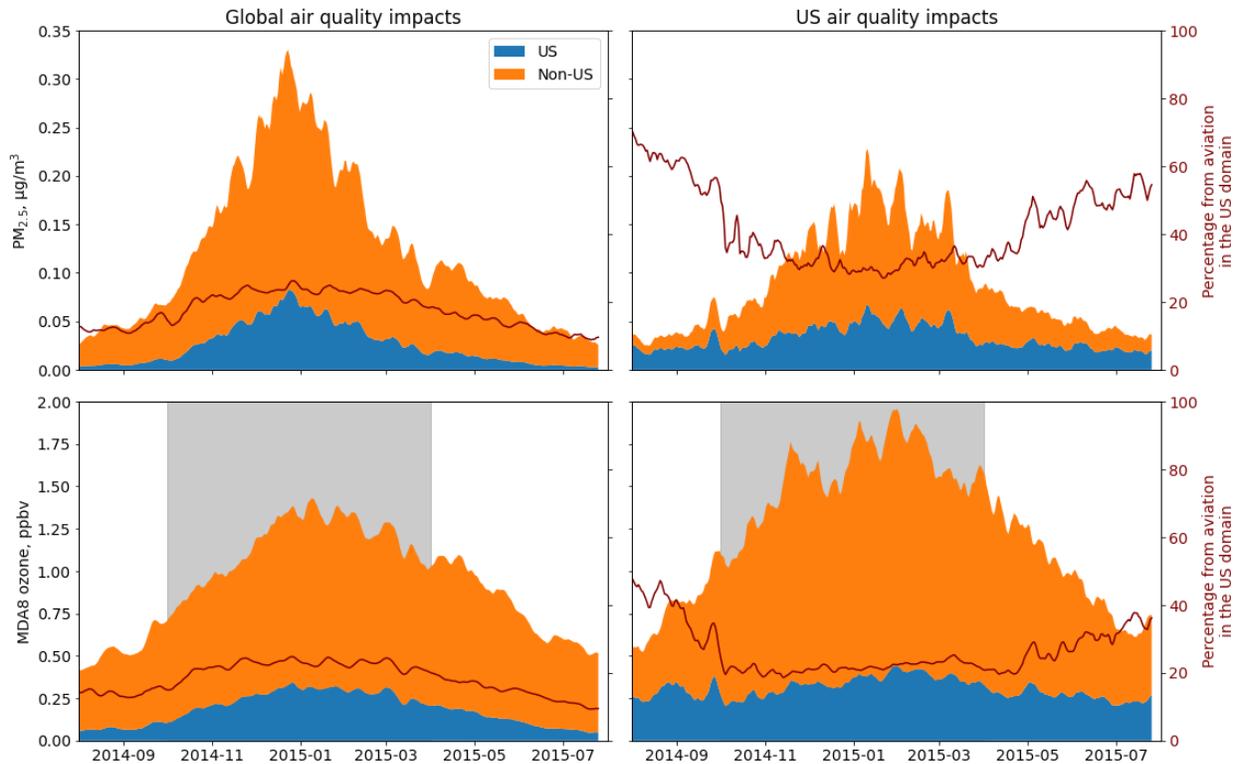
Figure S1.Exposure to air pollutants over the course of one year as a consequence of aviation emissions. Top row: exposure to fine particulate matter (PM$_{2.5}$). Bottom row: exposure to ozone, using the 8-hour maximum daily average (MDA8) metric. Left column: global mean exposure. Right column: exposure in the USA only. The blue shaded area shows impacts resulting from emissions in the US domain only, while the orange shaded area shows impacts resulting from emissions outside of the US. The red line shows the percentage of impacts in the target domain (i.e. global on the left, US-only on the right) resulting from US aviation emissions. The grey shaded area indicates the part of the year which would not be included when calculating the US "ozone season", under the definition used by Jerrett et al. (2009) of April through September.

We also evaluate the effect of thresholding on the total ozone impact. Turner et al. (2015) evaluated the effect of a threshold at 35 ppbv ozone, below which ozone exposure is assumed to have no effect. We find that the application of the threshold reduces ozone impacts by 8.8% in the US and 12% globally. Since Turner et al. (2015) did not include this threshold in their central estimate of ozone impacts, we do not include it in our central assessment.

Finally, in this study we apply the Global Estimate of Mortality Model (GEMM) to estimate mortality attributable to PM$_{2.5}$ exposure resulting from aviation emissions. Compared to results generated using five other relative risk data sources (see Table S1), we find that the GEMM-based value is the lowest. Using the Hoek et al. (2013) meta-analysis data employed by Eastham and Barrett (2016) would increase PM$_{2.5}$-related mortality by 16%, whereas using the Krewski et al. (2009) data from the extended analysis of the American Cancer Society cohort – employed, for example, by Dedoussi et al. (2020) in evaluating mortality due to cross-state air pollution – would cause a 68% increase.

Table S1. Central estimates of mortality from aviation-attributable PM$_{2.5}$ and ozone exposure when using different relative risk sources. Differences relative to the source used for the main text (in bold italics) are shown as percentages. MDA1 and MDA8 refer to the maximum daily value using a 1- or 8-hour average respectively. *NCD + LRI: Non-communicable disease and lower respiratory infections.

Risk data source	Exposure factor	Threshold	Cause of death	Global mortalities estimated
GEMM (Burnett et al., 2018)	*Annual mean PM$_{2.5}$*	*-*	*NCD + LRI**	*21,200 (-)*
Hoek et al. (2013) meta-analysis	Annual mean PM$_{2.5}$	-	Cardiovascular	24,700 (+16%)
Vodonos et al. (2018)	Annual mean PM$_{2.5}$	-	All-cause	40,400 (+91%)
Chen and Hoek (2020) meta-analysis	Annual mean PM$_{2.5}$	-	All-cause	45,700 (+120%)
EPA expert judgement (2010)	Annual mean PM$_{2.5}$	-	All-cause	64,500 (+200%)
ACS cohort (Krewski et al., 2009)	Annual mean PM$_{2.5}$	-	All-cause	35,600 (+68%)
Turner et al. (2015)	*Annual mean MDA8 ozone*	*-*	*Respiratory disease*	*53,100 (-)*
Turner et al. (2015)	Annual mean MDA8 ozone	35 ppbv	Respiratory disease	46,900 (-12%)
Jerrett et al. (2009)	Ozone season MDA1 ozone	-	Respiratory disease	16,800 (-68%)
Jerrett et al. (2009)	Ozone season MDA1 ozone	56 ppbv	Respiratory disease	9,540 (-84%)
Jerrett et al. (2009)	Ozone season MDA1 ozone	-	COPD + asthma	10,700 (-80%)
Jerrett et al. (2009)	Ozone season MDA1 ozone	56 ppbv	COPD + asthma	6,890 (-87%)

Non-aviation emissions inventories

In addition to the aviation emissions described in the main text, we simulate a suite of non-aviation emissions in GEOS-Chem. These are listed in Table S2. All listed emissions were identical between simulations. In addition to those listed, the ParaNO$_x$ extension is used to calculate the effect of in-plume chemical processing for ship emissions (Holmes et al., 2014; Vinken et al., 2011). Where both a regional and global source are available the regional inventory overwrites the global one. For example, both MIX and CEDS provide estimates of electricity generation emissions, but MIX applies to Asia only while CEDS is global. We therefore use MIX in Asia, and CEDS elsewhere (where not otherwise overwritten by other regional inventories)..

Table S2. Non-aviation emissions used in GEOS-Chem.

Inventory name	Region	Emissions included
APEI (Meng et al., 2019)	Canada	Multiple anthropogenic sectors
NEI 2011 (US EPA, 2015)	North America	Multiple anthropogenic sectors, including ship emissions
MIX v1.1 (Li et al., 2017)	Asia	Multiple anthropogenic sectors
DICE (Marais and Wiedinmyer, 2016)	Africa	Non-industrial anthropogenic emissions
CEDS (Hoesly et al., 2018)	Global	Multiple anthropogenic sectors, including ship emissions
GEIA NH_3 (Bouwman et al., 1997)	Global	Non-anthropogenic ammonia
POET (Granier et al., 2005; Olivier et al., 2003)	Global	Ethanol
C_2H_6 2010 (Tzompa-Sosa et al., 2017)	Global	Ethane from fossil fuel and biofuel
Xiao C_3H_8 (Xiao et al., 2008)	Global	Propane from fossil fuel and biofuel
Liang Bromocarb. (Liang et al., 2010)	Global	Very short lived bromocarbons ($CHBr_3$, CH_2Br_2)
Ordonez Iodocarb. (Ordóñez et al., 2012)	Global	Iodocarbons (CH_3I, CH_2I_2, CH_2ICl, CH_2IBr)
Decaying plants (Millet et al., 2010)	Global	Volatile organic compounds from decaying plants
AFCID (Philip et al., 2017)	Global	Anthropogenic, fugitive construction and industrial dust
Volcano (Carn et al., 2015)	Global	Sulfur dioxide from volcanic degassing and eruption
GFED (Giglio et al., 2013)	Global	Fire emissions from the GFED v4 inventory
Inorg. Iodine (Carpenter et al., 2013; MacDonald et al., 2014; Sherwen et al., 2016)	Global	Marine emissions of inorganic iodine (HOI, I_2)
LightNOx(Murray et al., 2012)	Global	Lightning NO_x

SoilNOx (Hudman et al., 2012)	Global	NO_x from soil and fertilizers	
SeaSalt (Jaeglé et al., 2011)	Global	Sea salt picked up from the ocean surface	
DustDEAD (Zender, 2003)	Global	Entrained mineral dust from the DEAD model	
MEGAN (Guenther et al., 2012)	Global	Biogenic emissions	

Emissions of lightning NO_x, soil NO_x, mineral dust, sea salt, and biogenic emissions are calculated prior to the simulation at a global resolution of 0.5° latitude by 0.625° longitude, with the calculated fluxes for each hour archived and used during all simulations at all resolutions. This approach ensures consistent emissions between simulations at different resolutions. The only exception is emissions of inorganic iodine, which is calculated online based on meteorological fields but is expected to be a negligible contributor to differences.

Disaggregating the effect of resolution on exposure

Increasing model resolution affects both the accuracy of the model calculations and the degree to which pollution and population are "collocated" in the data. To isolate this effect, we calculate total population exposure globally in two ways. First, we calculate exposure by using the original model output. We then perform a second calculation in which we degrade all model output to the coarsest resolution (C24, or ~400 km). The subsequent estimated global mean population exposures for each pollutant are shown in Table S3.

Table S3. Global mean population exposure to each pollutant calculated at the original model resolution ("Raw") or after degrading the model output to C24 resolution ("Degraded"). Results are given as the percentage of the "Raw" C24 calculation.

	$PM_{2.5}$		Soot		Ozone (MDA8)	
	Raw	Degraded	Raw	Degraded	Raw	Degraded
C24 (~400 km)	100%	-	100%	-	100%	-
C90 (~100 km)	133%	116%	114%	90.3%	117%	121%
C180 (~50 km)	135%	116%	122%	85.0%	120%	125%

Comparing the "degraded C180" to "raw C24" results isolates the effect of improved resolution of physical phenomena. For $PM_{2.5}$ and ozone the "degraded C180" exposure is 16% and 25% greater respectively than the "raw C24" exposure. This is less than the combined effect (comparing "raw C180" to "raw C24") for $PM_{2.5}$ (+35%) but greater than that for ozone (+20%). This suggests that the artificial diffusion associated with coarse-resolution simulation reduces the estimated impacts of $PM_{2.5}$ but enhances them for ozone.

For soot, we find that exposure calculated using the "degraded C180" results is 15% lower than the "raw C24" calculation, whereas the "raw C180" results showed greater exposure than the "raw C24" (+22%).

This implies that using a higher-resolution model causes faster removal of soot from the atmosphere, but that this effect is exceeded by the increase in exposure associated with collocation of pollution sources and population. Performing the same calculation using the ~100 km (C90) output data shows changes in surface air quality which are consistent with using the ~50 km results for $PM_{2.5}$ and ozone.

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