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Domingos de Azevedo Quadros, F.; Snellen, M.; Dedoussi, I.C.

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Flávio D A Quadros , Mirjam Snellen  and Irene C Dedoussi 

Section Aircraft Noise and Climate Effects, Faculty of Aerospace Engineering, Delft University of Technology, Delft, The Netherlands

E-mail: i.c.dedoussi@tudelft.nl

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Abstract

Emissions from civil aviation degrade air quality, and have been estimated to lead to ~16 000 premature deaths annually. Previous studies have indicated that aviation emissions in different regions have varying corresponding air quality and human health impacts. Given the global nature of aviation activity and its forecasted regionally heterogeneous growth, this phenomenon poses challenges in aviation air quality decision making. In this study, we quantify the differences in the regional air quality responses to aviation emissions, and analyze their drivers. Specifically, we use the GEOS-Chem atmospheric chemistry-transport model to quantify the regional fine particulate matter (PM_{2.5}) and ozone sensitivity to aviation emissions over Asia, Europe, and North America for 2005. Simulations with perturbed regional aviation emissions are used to isolate health impacts of increases in aviation emissions originating from and occurring in different regions. Health impacts are evaluated as premature mortality attributed to both landing and takeoff and cruise emissions. We find that the sensitivity of PM_{2.5} global population exposure to full-flight emissions over Europe is 57% and 65% higher than those to emissions over Asia and North America, respectively. Additionally, the sensitivity of ozone global population exposure to aviation emissions over Europe is larger than to emissions over Asia (32%) and North America (36%). As a result, a unit of fuel burn mass over Europe results in 45% and 50% higher global health impacts than a unit of fuel burn mass over Asia and North America, respectively. Overall, we find that 73% and 88% of health impacts from aviation emissions over Europe and North America, respectively, occur outside the region of emission. These results suggest that inter-regional effects and differences in regional response to emissions should be taken into account when considering policies to mitigate air quality impacts from aviation, given the projected spatially heterogeneous growth in air transportation.

1. Introduction

Emissions from the combustion associated with aircraft, in addition to impacting the climate (Lee *et al* 2010), are also a known contributor to the degradation of air quality (Ashok *et al* 2014, Masiol and Harrison 2014). Among other deleterious consequences such as worsened visibility (Delucchi *et al* 2002), decreased crop yields (Ainsworth 2017), and damage to wildlife and vegetation (Ashmore 2005), this additional air pollution in the form of fine particulate matter (PM_{2.5}) and ozone (O₃) is associated with adverse effects on human health (Turner *et al* 2016, Burnett *et al* 2018). Yim *et al* (2015) estimated

that the civil aviation activity in the year 2005 was associated with 16 000 (90% CI: 8300–24 000) premature deaths globally, with 87% of that amount due to increased surface concentrations of PM_{2.5} and 13% due to increased ozone surface concentrations. Societal costs associated with the air quality impacts of aviation have been found to be comparable to those related to climate and noise impacts (Lee *et al* 2010, Wolfe *et al* 2014, Grobler *et al* 2019).

Long-term forecasts estimate a compound annual growth rate of 4.3% in global air traffic between 2015 and 2035 (International Civil Aviation Organization 2018). This expected growth is not spatially uniform, with the intra-regional 2015–35 annual rates ranging

from 2.6% for North America, 2.7% for Europe, to 6.7% in Central Southwest Asia (International Civil Aviation Organization 2018). Despite improvements in aircraft technology and air traffic management, emissions from international aviation are projected to continue to grow through 2050 (International Civil Aviation Organization 2019).

Policywise, measures regarding air quality are often guided by regional, national or local standards defining acceptable levels of concentration of various pollutants, and these standards may drive planning decisions concerning aviation (International Civil Aviation Organization 2016). The International Civil Aviation Organization (ICAO) engine emission standards, applicable to turbojet and turbofan engines, determine maximum emission indices (defined as mass of emission species per mass of fuel burned) allowed for a standard landing and takeoff (LTO) cycle (International Civil Aviation Organization 2017). LTO is defined as the operations within 3000 ft above ground. These engine standards were created in 1981 and through revisions over the years continue to tighten the limits on the emission of oxides of nitrogen (NO_x), unburnt hydrocarbons (HC), carbon monoxide (CO), and non-volatile particulate matter. This regulatory framework, focused on LTO and local air quality standards, is consistent with the framing of aviation's air quality impacts as a local air quality issue. This is exemplified in Europe by the exclusion of non-LTO emissions in the National Emission Ceilings Directive for the accounting related to the emission targets up to the year 2030 (European Parliament and the Council of the European Union 2016).

However, there is increasing scientific evidence highlighting the contribution of non-LTO emissions and cross-border pollution transport to the air quality impacts of aviation (Cameron *et al* 2017). Tarrasón *et al* (2004), using an atmospheric chemistry-transport model (CTM) focused on Europe, identified the LTO NO_x impact on surface air quality as an order of magnitude smaller than that of non-LTO. They attributed this to the larger share of non-LTO emissions (95% of all aviation emissions), atmospheric vertical transport and the high efficiency of NO_x ozone production in the free troposphere. They also found that about half of the impacts of full-flight (i.e. LTO plus non-LTO) NO_x emissions on air quality in Europe were associated with emissions outside the region. Barrett *et al* (2010), employing a global CTM, found that 99% of population-weighted aircraft-attributable $\text{PM}_{2.5}$ is secondary sulfate–ammonium–nitrate aerosol (SO_4^{2-} – NH_4^+ – NO_3^-) formed primarily from NO_x , about 90% of which is emitted during non-LTO phases of flight (Olsen *et al* 2013, Simone *et al* 2013). Søvde *et al* (2014), using an ensemble of five atmospheric models, found consistent increase in tropospheric ozone from aviation NO_x cruise emissions.

Aerosol precursors emitted at cruise levels impact the surface both through vertical transport and by accelerating the oxidation of the precursors at the surface as they form aerosol, establishing an intercontinental mechanism of $\text{PM}_{2.5}$ impact (Leibensperger *et al* 2011). Yim *et al* (2015) combined global, regional and local atmospheric models and found that 75% of the global premature deaths due to aviation-attributable $\text{PM}_{2.5}$ and ozone were attributed to non-LTO emissions. Koo *et al* (2013) used the adjoint of a CTM, finding that non-LTO emissions were responsible for 60%–90% of full-flight impacts in different regions. They also found a significant cross-regional component to impacts, with 95% of the resulting premature mortalities from full-flight emissions over the US occurring outside the country, and 64% of impacts from emissions over Europe occurring outside the region.

Besides being involved in the aerosol formation pathway, the ozone produced from NO_x is also harmful to human health by itself, with Eastham and Barrett (2016) estimating 8600 premature deaths yearly due to (full-flight) aviation-attributable ozone. Ozone produced at cruise altitudes has longer atmospheric lifetimes than the NO_x directly emitted near the ground, further enabling global-scale effects (Gauss *et al* 2006).

Intercontinental air pollution effects is a subject that has also been researched outside the context of aviation. The Task Force Hemispheric Transport of Air Pollution collaboration produced studies based on an ensemble of models looking at various aspects of this issue including ozone and primary $\text{PM}_{2.5}$ (United Nations 2010, Anenberg *et al* 2014). Zhang *et al* (2017) estimated the transboundary $\text{PM}_{2.5}$ impacts occurring through atmospheric transport and dispersion as well as through shifts in emissions due to international trade. These studies show that even when considering other (non-aviation) anthropogenic emissions, which unlike aviation emissions are not located at high altitude, the inter-regional effects can be significant—being associated with 20% to more than 50% of ozone-related premature mortalities and 2% to 5% of particulate matter mortalities (United Nations 2010).

Given the long-distance nature of aviation's air quality impacts and since there is non-uniformity globally in the background atmospheric composition, population density, baseline disease incidence rates, the same amount of emissions can result in different levels of impact depending on their source location. Population density has a strong effect of adding weight to Eastern Asia in terms of total premature mortality (Koo *et al* 2013, Eastham and Barrett 2016). As discussed by Yim *et al* (2015) and Grobler *et al* (2019), when country-specific valuations of mortality based on economic metrics are used, more weight is put on the air quality in North America and Europe.

Local atmospheric conditions, largely driven by emissions from other sectors as well as meteorology, also play a role in how sensitive a region's air quality can be to aviation emissions. Woody *et al* (2011) explain higher levels of PM_{2.5} formation from NO_x in the US by considering the abundance of ammonia available to react with nitrates. However this has not been assessed for regions besides the US, and for pollutants beyond PM_{2.5}. In contrast to previous studies that looked at the magnitude of global health impacts of aviation emissions (Yim *et al* 2015, Eastham and Barrett 2016), we focus here on the sensitivity of impacts to aviation emissions perturbations, and the resulting differences across regions. We quantify the regional air quality responses to aviation emissions, and isolate, for the first time, the extent to which different factors (emissions within and outside of the region, population density, epidemiological characteristics, and background atmospheric processes) drive these responses. We simulate the regional air quality sensitivity to aviation emissions by performing multiple simulations to isolate the impacts of increases in aviation emissions over Asia, Europe and North America. The long-range air quality impacts of aviation emissions are quantified for each source-receptor regional pair. Differences in the regional sensitivity of health impacts are calculated in stages allowing for decoupling of the effects of population density and human health impact functions. Spatial variations observed in these sensitivities to aviation emissions are then discussed in regards to their implications for air quality decision making for aviation.

2. Methods

The regional sensitivities of air quality impacts to aviation emissions are estimated using the GEOS-Chem atmospheric CTM. Specifically, we simulate a set of scenarios where aviation emissions are increased alternatively in one of three continental regions shown in figure 1, and quantify the corresponding pollutant concentrations, population exposure, and human health impacts.

2.1. Aviation emissions and test scenarios

Full-flight aviation emissions are obtained from the AEIC model (Simone *et al* 2013), which builds estimates from 2005 civil aviation flight schedule data from OAG Aviation. Full-flight emissions are defined here as LTO emissions and non-LTO emissions (the latter primarily consisting of cruise level emissions but also emissions during the climb and descent above 3000 ft). The aviation emissions inventory used is obtained by running AEIC with the BADA 3.15 aircraft performance model (EUROCONTROL Experimental Centre 2019), and considering the LTO cycle described by Stettler *et al* (2011). Emissions for NO_x,

CO and HC are calculated from ICAO engine certification data. Black carbon emissions are estimated using the SCOPE11 method for LTO (Agarwal *et al* 2019), and using a constant emission index of 30 mg kg⁻¹ fuel for the non-LTO phases of flight, consistent with the values of 25–35 mg kg⁻¹ adopted in other studies (Eyers *et al* 2004, Wilkerson *et al* 2010, Yim *et al* 2015). The speciation of the emission variables is the same used by default in GEOS-Chem, based on Barrett *et al* (2010).

The global sum of aviation emissions for the full year is 180 Tg of fuel burn, 16.4 Tg of which is used during LTO (9.1%). Emission indices for each emitted species and type of engine and a breakdown between domestic and international flight emissions are listed in the supporting information (SI, section S1 (available online at stacks.iop.org/ERL/15/105013/mmedia)). International flights are responsible for 57% of fuel burn mass (inter-EU flights are not considered international).

The inventory is built at the spatial resolutions of the simulation grids (section 2.2) and with a monthly temporal resolution. Fuel burn (and corresponding emissions) is lower during winter, with individual months having a global average fuel burn between -4.5% (January) and +5.6% (July) of the yearly average (SI section S1).

Four different aviation scenarios are evaluated: the baseline case using the estimated 2005 aviation emissions, and three cases where all aviation emissions released within one of three regions are positively perturbed (multiplied by a constant). The regions, shown in figure 1, represent Asia, Europe, and North America (denoted as AS, EU, and NA). The perturbations are tapered off toward the edges of the regions for a smooth transition to the unperturbed areas. A 1.1 multiplier was chosen for Europe as a compromise between having a strong enough model response above noise levels and a low enough perturbation that corresponds to marginal increases representative of a few years worth of growth in emissions (3.6 years at 2.7% annual growth in the case of Europe). Different multipliers are applied for each region such that the increase of full-flight fuel burn mass per area is the same in all three cases, after accounting for differences in total areas and baseline aviation emissions between the regions (table 1). Within each case, the same multiplier is applied to all emission species, at all altitude levels and constantly through time.

In order to observe the different effects of LTO and non-LTO emissions, a further three simulations are performed, where only the LTO emissions in each region are increased, using the same multipliers as before. These scenarios are simulated only in the perturbed region instead of using the nested grid approach, since the inter-regional

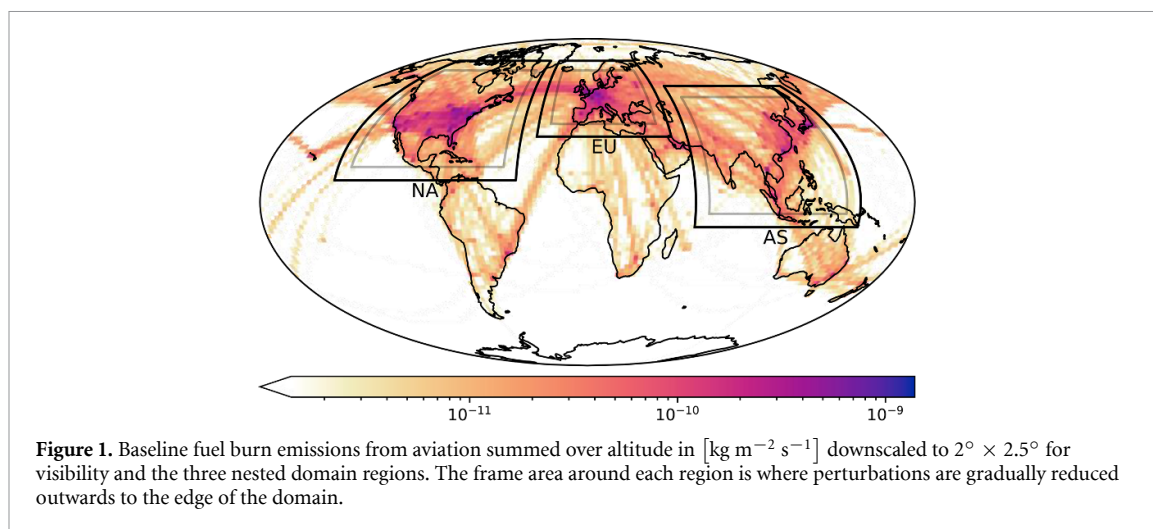


Table 1. Overview of aviation emissions in each region evaluated.

Region	Area [% of world]	Yearly fuel burn [% of world]	Perturbation multiplier
World	$5.11 \cdot 10^{14} \text{ m}^2$	180 Tg	N/A
AS	10.65%	21.6%	1.267
EU	3.81%	20.6%	1.100
NA	8.96%	34.3%	1.141

effects of LTO emissions are expected to be small (Koo *et al* 2013).

2.2. Atmospheric modeling

The atmosphere is modeled using the GEOS-Chem model version 12.6.1 (The International GEOS-Chem User Community 2019) with the Unified Tropospheric-Stratospheric Chemistry Extension (UCX) module (Eastham *et al* 2014), and meteorological data from the MERRA-2 reanalysis product from NASA/GMAO. The default aerosol simulation and $\text{PM}_{2.5}$ parametrization is used (SI section S2.3), including the secondary organic aerosols simple parametrization model from Kim *et al* (2015). The main emissions inventories used are the Community Emissions Data System anthropogenic emissions as the global default (Hoesly *et al* 2018), the U.S. Environmental Protection Agency's NEI2005, the APEI (van Donkelaar *et al* 2008), the DICE-Africa (Marais and Wiedinmyer 2016), the MIX-Asia (Li *et al* 2014), and parts of EDGAR v4.3 (Crippa *et al* 2018). Other emission modules used are listed in the SI (section S2.1).

For each scenario analyzed, four model runs are performed: a global run at $4^\circ \times 5^\circ$ resolution (latitude \times longitude) and, using boundary conditions from the global run, three nested regional grids at $0.5^\circ \times 0.625^\circ$ resolution, coinciding with the emissions perturbations (figure 1). All grids have 72 vertical levels, going from the surface to a 0.01 hPa pressure level. Global impacts are calculated using the results of the nested simulations where available (the three regions of interest) and the results of the coarse

global simulations elsewhere. The effects of the grid resolution are discussed further in the SI (section S2.6).

The initial state for all runs is obtained by a 21 month spin-up of the global coarse resolution model. Then each individual run (global and regional) has 3 months of spin-up followed by 12 months. The latter is considered in the analysis. In total, four global simulations and five nested simulations in each region are performed. The simulation period considered represents the year 2005 for meteorology, aviation and non-aviation emissions. An overview of the resulting background ground level $\text{PM}_{2.5}$ concentrations and ozone mixing ratios in the simulations, as well as a comparison with air quality monitoring records, are presented in the SI (section S3.1).

To evaluate the effect of background atmospheric composition on air quality sensitivity to aviation, an additional set of coarse global simulations is also performed using the 2005 aviation emissions inventory with meteorology and other emissions representing 2013.

The concept of Gas Ratio defined by Ansari and Pandis (1998) as the ratio between free ammonia and total nitrates is used in the analysis of model outputs to identify atmospheric conditions favorable to $\text{PM}_{2.5}$ formation. This ratio is calculated as

$$GR = \frac{[\text{NH}_3] + [\text{NH}_4^+] - 2[\text{SO}_4^{2-}]}{[\text{HNO}_3] + [\text{NO}_3^-]} \quad (1)$$

GR less than 1 indicates that competition between sulfates and nitrates for limited ammonia restrict $PM_{2.5}$ formation as NO_x is increased.

Another metric used in the analysis is ground level formaldehyde to reactive nitrogen (NO_y) ratio (FNR, alternatively defined as the ratio to NO_x or NO_2). This metric uses formaldehyde as a proxy for volatile organic compounds (VOC) to identify regions where ozone production is limited by VOC and where it is limited by NO_x (Sillman 1995). Lower values of FNR are expected where ozone formation is NO_x saturated, and increases in NO_x might result in a net decrease of ozone concentration, due to competing chemical reactions.

The linearity of model's response to the perturbation was evaluated by performing additional simulations with EU emissions multiplied by 1.3 instead of 1.1. This larger perturbation resulted in $PM_{2.5}$ and ozone sensitivities to emissions that were 0.3% and 1.1% lower, respectively, than those obtained with the smaller perturbation (SI section S2.5).

2.3. Health impact assessment

The health impacts are quantified as the increase in premature mortality in people over 30 years of age due to additional exposure to $PM_{2.5}$ and ozone. Human exposure to increases in ground level concentration of pollutants are calculated by applying the LandScan 2005 high resolution global population distribution data, which sums to 6.44 billion people (Bright *et al* 2006). Area-averaged concentration increases are calculated considering either all grid cells or only those with a greater than zero population, which represents predominantly the changes over land (SI section S2.2). Country-specific age distribution (fraction of population over 30 years old) and baseline mortality rates for the relevant disease classes are taken from the 2005 estimates of causes of death from the Global Health Estimates 2015 (GHE) (World Health Organization 2016).

$PM_{2.5}$ health impacts are estimated as the increased mortality from non-communicable diseases (GHE code 600) and lower respiratory infections (GHE code 390) according to the concentration-response function (CRF) defined by Burnett *et al* (2018). This CRF was chosen as it was derived from an ensemble of studies covering a wide range of population characteristics and concentration levels. We use this model (called GEMM) to obtain mortality hazard ratios from the yearly average $PM_{2.5}$ concentrations outputted by the simulations, and the hazard ratios are then applied to the baseline incidence rate of cause-specific mortality to give a total number of excess deaths from concentrations above a counterfactual value. The excess premature mortality attributable to additional aviation emissions is taken as the difference to the baseline emissions scenario. Alternative estimations performed using different CRFs and different mortality endpoints are given in

the SI (section S4). Consistent with other aviation air quality studies (Yim *et al* 2015, Grobler *et al* 2019), we do not consider different toxicities for specific $PM_{2.5}$ constituents, nor the effect of particle number.

Ozone health impacts are estimated as the increased mortality from all respiratory diseases (GHE codes 390, 400, and 1170) according to the CRF established by Turner *et al* (2016), which represents a 1.12 hazard ratio for a 10 ppbv increase in the yearly maximum daily 8-hour average (MDA8) ozone mixing ratio. This CRF is the result of an analysis of long-term ozone effects in a large cohort study in the United States, involving over 12.6 million person-years of follow-up. Alternative estimations with different CRFs and mortality endpoints are given in the SI (section S4).

2.4. Uncertainty estimation

We quantify uncertainty using independent variables associated with atmospheric modelling in terms of $PM_{2.5}$ concentration and ozone mixing ratio changes and with the CRFs used. A multiplicative triangular error distribution $T(0.36, 1, 2)$ is applied to $PM_{2.5}$ and $T(0.5, 1, 1.5)$ to ozone changes, following the same methodology applied to GEOS-Chem results by Grobler *et al* (2019), which was based on an inter-model comparison study (Cameron *et al* 2017). For the $PM_{2.5}$ CRF we consider a normal distribution in the theta parameter of the GEMM model, as reported by Burnett *et al* (2018). For the ozone CRF, we adopt a triangular distribution for the hazard ratio, defined by the central value and the 95% confidence intervals reported by Turner *et al* (2016). The uncertainties are combined using a Monte Carlo approach with 10^5 samples, with 2.5th and 97.5th percentiles reported along with the results from nominal input values. Other sources of error, such as in the baseline disease incidence rates and population distribution are not considered.

3. Results and discussion

The effects of full-flight and LTO-only emissions perturbations on air quality are presented in sections 3.1 and 3.2, respectively. Estimates of the resulting health impacts associated with the air quality degradation are presented in section 3.3. How these sensitivities differ between 2005 and 2013 is presented in section 3.4 and the limitations of this analysis are discussed in section 3.5.

3.1. Full-flight air quality impacts

The global average baseline $PM_{2.5}$ concentrations and ozone mixing ratios through 2005 are shown in figure 2 considering only populated areas, which excludes most bodies of water. The seasonal patterns follow the Northern Hemisphere (NH), where 71% of the area considered is and where average concentrations are higher. $PM_{2.5}$ background levels are higher during

the northern winter, driven by longer atmospheric lifetimes of nitrate precursors, while ozone levels are higher during summer when solar activity is highest.

Figure 2 also shows the marginal increase in global concentrations for each (full-flight) perturbation scenario, normalized by additional fuel burn mass. The largest increases happen during the NH winter months. The aviation-induced increase in $PM_{2.5}$ is composed mainly by ammonium and nitrate (SI section S3.5). The sensitivity of concentrations to aviation emissions over Europe is consistently higher than those to emissions over the other regions throughout the year. The temporal difference in aviation-attributable surface ozone with background levels can be attributed to longer lifetimes in the mid-troposphere during winter while the surface background mixing ratios are governed by the steady-state photochemical seasonal cycles (Eastham and Barrett 2016).

On average, the sensitivity of global ground level $PM_{2.5}$ in populated areas to the EU perturbation is 107% and 64% greater than the sensitivities to the AS and NA perturbations, respectively (figure 3). When changes over non-populated areas are included, the sensitivity to the perturbation in EU is 33% and 40% greater than the sensitivity to perturbations in AS and NA, respectively. The population-weighted averages are 4.4 (AS), 3.3 (EU) and 3.3 (NA) times the non-weighted averages over populated areas. These results reflect both the proximity of aviation emissions near densely populated regions and the air quality sensitivity in these areas. When accounting for differences in the population spatial distribution by using the population-weighted metric, the sensitivity of global $PM_{2.5}$ to aviation emissions over AS is similar to that of emissions over NA (+5%), while the sensitivity to emissions over EU is higher (+57% than AS, +65% than NA).

The fact that the air quality sensitivity to aviation emissions over EU is higher than to emissions elsewhere before population density data or CRFs are applied means that atmospheric conditions for emissions in that region are more favorable to $PM_{2.5}$ formation. Westerly prevailing winds transport cruise emissions from EU to Asia, while cruise emissions over AS and NA get advected first to the Pacific and Atlantic oceans respectively. Additionally, we find that this higher impact of emissions in Europe is associated with higher availability of ammonia to react with NO_x , particularly during winter, compared to the United States coasts, East China and Japan (SI section S3.3). A lower percentage of population in NA (39%) is over areas where the average gas ratio over January was higher than 1.3, than in AS (82%) and EU (62%). The southeast of AS, where gas ratio is largely above 1.3 in both winter and summer, lies at lower latitudes than the majority of cruise emissions in the Northern Hemisphere.

The mean atmospheric circulation patterns have been found to be associated with higher global impacts of aviation emissions above the northern tropic (Barrett *et al* 2010, Koo *et al* 2013). We note that although we identify these factors leading to differences in sensitivity, we do not decouple which mechanism contributes the most.

Compared to existing literature, the $PM_{2.5}$ sensitivities to regional aviation emissions perturbations we report here are higher than the sensitivity to global aviation emissions estimated by values presented in Eastham and Barrett (2016) (0.282 population-weighted $\mu g m^{-3} (Tg \text{ of fuel})^{-1}$) and Yim *et al* (2015) (0.033 area-weighted $\mu g m^{-3} (Tg \text{ of fuel})^{-1}$). Besides differences in the models used, grid resolution, $PM_{2.5}$ parameterization, and emission inventories, non-linearity of the response to perturbation can also contribute to the observed differences.

Impacts on ground level ozone are more spatially diffuse, with the population-weighted averages being 1.13 (AS), 1.05 (EU) and 1.03 (NA) times the averages over populated areas (figure 3). Aviation emissions in Europe also lead to higher population-weighted ozone impacts (+32% than AS, +36% than NA). All cases have higher population-weighted increases than the 2.87 pptv $(Tg \text{ of fuel})^{-1}$ estimated for global full-flight emissions by Eastham and Barrett (2016), although they considered the maximum daily 1 h average over ozone season, when aviation contribution is smaller.

While there is stronger association between impacts and the perturbation in the same region, there are still significant cross-regional effects, as shown in figure 4. An increase in emissions over Europe impacts Asia more (+32% for $PM_{2.5}$ and +16% for ozone) than a local increase of the same amount. In terms of total population exposure, Asia receives most of the impacts in all cases. Additionally, most exposure from North American emissions happen outside the region (92% for $PM_{2.5}$ and 88% for ozone).

These significant cross-regional impacts pose regulatory challenges when accounting for aviation's environmental impact on a regional scale. For example, the projected long-term growth in air traffic is spatially heterogeneous, estimated at ~3% annually for EU and NA and ~6% annually for AS (Boeing Commercial Airplanes 2019). Considering the 2005 fuel burn over the EU and AS regions (table 1) and the different air quality sensitivities over fuel burn increases (figure 3), a twice as large rate of increase in emissions over AS compared to EU would result in proportionately lower impacts (34% and 59% higher increases in $PM_{2.5}$ and ozone global exposure, respectively). In the same scenario, the resulting AS population exposure attributable to emissions over EU would be 64% ($PM_{2.5}$) and 55% (ozone) of those due to the more rapidly increasing emissions over AS.

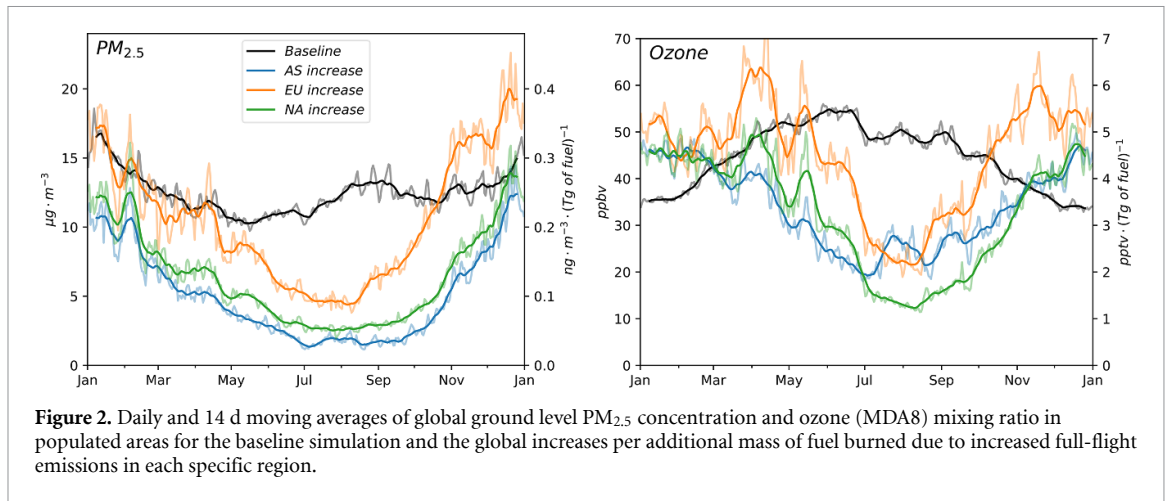


Figure 2. Daily and 14 d moving averages of global ground level $PM_{2.5}$ concentration and ozone (MDA8) mixing ratio in populated areas for the baseline simulation and the global increases per additional mass of fuel burned due to increased full-flight emissions in each specific region.

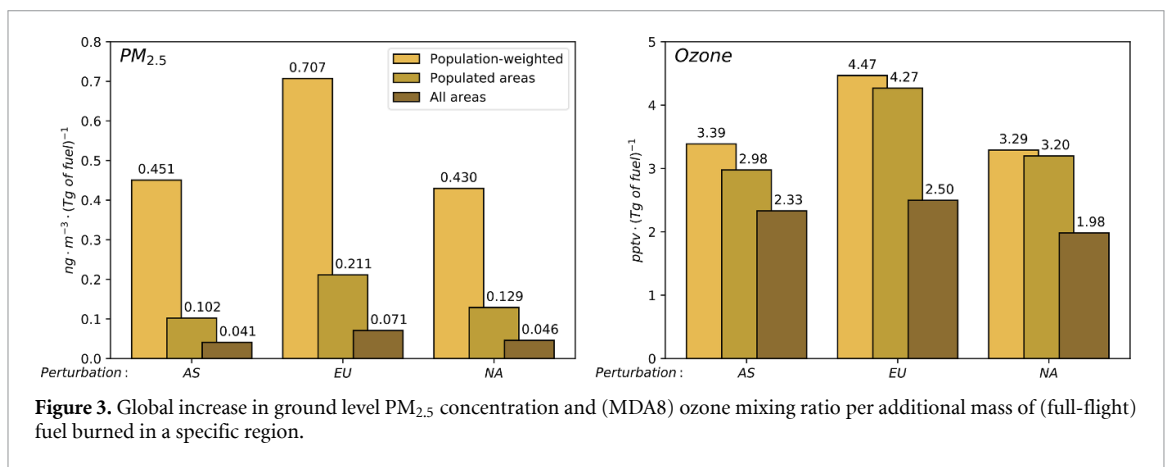


Figure 3. Global increase in ground level $PM_{2.5}$ concentration and (MDA8) ozone mixing ratio per additional mass of (full-flight) fuel burned in a specific region.

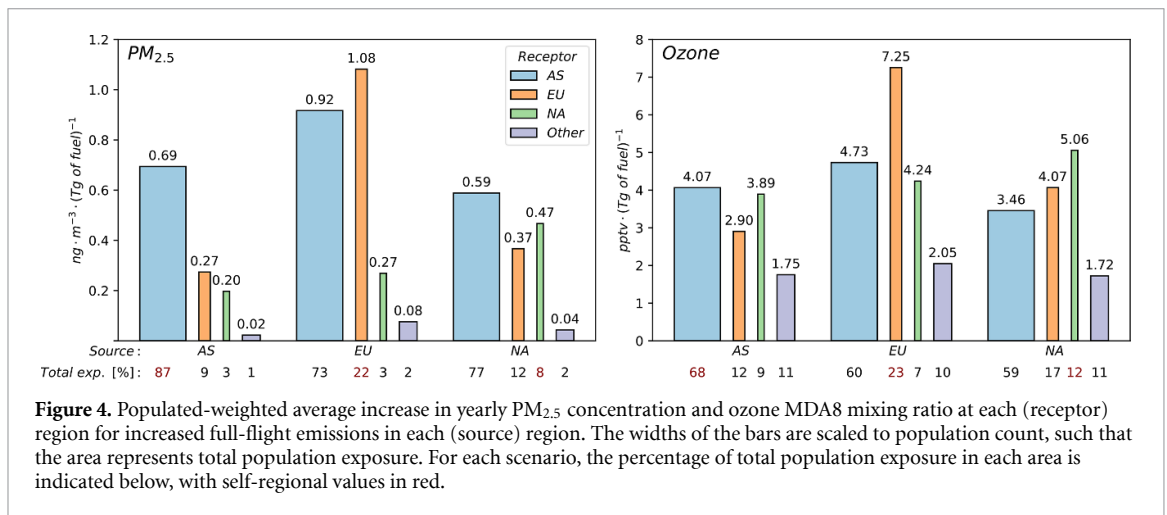


Figure 4. Populated-weighted average increase in yearly $PM_{2.5}$ concentration and ozone MDA8 mixing ratio at each (receptor) region for increased full-flight emissions in each (source) region. The widths of the bars are scaled to population count, such that the area represents total population exposure. For each scenario, the percentage of total population exposure in each area is indicated below, with self-regional values in red.

3.2. LTO air quality impacts

Air quality impacts observed in simulations in which only LTO emissions over a region were perturbed were compared to those in simulations with full-flight perturbations over the same region, in order to differentiate between the LTO and non-LTO contributions to full-flight impacts. We find that for all three regions, the LTO contribution to full-flight population-weighted $PM_{2.5}$ increases varies from

10% to 20% during winter to 40%–50% during summer (figure 5(a)). This is in line with the shorter mid-tropospheric lifetime of nitrate precursors during summer which may reduce the impacts of cruise emissions. Over the full year, LTO-only perturbations result in 17% (AS), 26% (EU) and 29% (NA) of the population-weighted $PM_{2.5}$ within the same region attributed to full-flight emissions over that region. This is higher than the LTO 9%–11% share of fuel

burn, demonstrated by the higher impacts per fuel burn in figure 5(b).

Sensitivity of $PM_{2.5}$ to LTO emissions in EU is higher compared to other regions (figure 5(b)), which is consistent with higher ammonia availability. The percentage of LTO NO_x released over areas with a monthly average $GR > 1.3$ for each region is 57% (AS), 74% (EU), 14% (NA) during January, and 68% (AS), 76% (EU), 21% (NA) during July (SI section S3.3).

LTO emissions cause close to zero net increase in surface level ozone over the year: 3.5% (AS), 0.4% (EU) and 0.1% (NA) of the impacts on the same region from full-flight emissions over the region. Airports are often situated in areas with high NO_x surface level concentrations, where ozone production becomes limited by VOC and additional NO_x emissions can cause a decrease in ozone. Decreases in ozone are associated with the formaldehyde to NO_y ratio, particularly during winter when most of the decreases are observed (SI section S3.4).

Operational strategies such as pushback control and de-rated takeoffs are a possible way of reducing LTO emissions (Ashok *et al* 2017). The seasonal sensitivity trends we report here indicate that the use of such strategies would have a stronger air quality effect during summer at regional levels (figure 5(b)). Similarly, alternative technological mitigation options targeting short-haul flights (which have higher percentage of LTO emissions/impact), such as electric or hybrid flying, would also have stronger effects during the summer, and in addition would result in approximately twice the air quality improvement (in terms of $PM_{2.5}$ concentration from LTO emissions) if they were to be introduced in EU compared to AS and NA.

3.3. Health impacts

Figure 6 presents the sensitivity of regional human health impacts to full-flight emissions per region of perturbation. We find that most of the increase in premature mortalities from additional aviation emissions from any of the three regions happens in Asia, both due to ozone and $PM_{2.5}$. The fact that Asia receives the largest share of health impacts despite not necessarily being the most affected region in terms of air quality from aviation emissions over other regions (figure 4) is due to a larger population count, which leads to higher total population exposure.

The application of country-specific baseline disease mortality rates decreases the share of $PM_{2.5}$ impacts in Asia: in the case of perturbation in AS, for example, AS receives 87% of added global exposure (figure 4) but has 75% of global excess deaths (figure 6). For ozone impacts the opposite occurs, with AS having 68%, 60%, and 59% of global exposure and 75%, 68%, and 67% of deaths resulting from perturbations over AS, EU, and NA, respectively. Country-specific baseline disease mortalities have a net effect from both $PM_{2.5}$ and ozone of higher global

health impacts in all three full-flight perturbation cases. Estimates using different baseline disease mortality rates and different CRFs are given in the SI (section S4).

The sensitivity of total global health impacts to aviation emissions over EU is 45% and 50% higher than the sensitivity to emissions over AS and NA, respectively (table 2). Extrapolating for the total 2005 aviation emissions—estimating the global sensitivity to emissions from other regions (23% of total) from an additional global coarse resolution simulation (SI section S5)—gives a total of 20 300 (95% CI: 9800–40 300) premature deaths due to $PM_{2.5}$ and 38 300 (21 600–57 800) due to ozone. Both these numbers are highly sensitive to the choice of CRF used, however the ratio of impacts from different source regions is mostly independent from the CRF chosen (SI section S4).

For comparison, the number of yearly aviation-attributable premature deaths just from $PM_{2.5}$ was estimated by Yim *et al* (2015) as 13 900, considering the CRF of cardiopulmonary diseases and lung cancer from Ostro (2004). Eastham and Barrett (2016) estimated a 9200 increase in cardiovascular mortality due to $PM_{2.5}$ using a CRF from Hoek *et al* (2013). Applying the same CRFs to this study results in 9500 (Ostro 2004) and 13 800 Hoek *et al* (2013) premature deaths from $PM_{2.5}$.

Using a more conservative CRF for ozone (Jerrett *et al* 2009) yields 12 800 premature deaths, and further restricting the mortality endpoints to just chronic obstructive pulmonary disease and asthma (excluding respiratory infections) would lead to 8400 deaths, compared to the 6800 deaths estimated by Eastham and Barrett (2016) using this CRF.

Total ($PM_{2.5}$ and ozone) premature deaths from the LTO-only perturbations are 10.8% (AS), 16.7% (EU), 14.5% (NA) of mortality within the same region caused by local full-flight perturbation. For each region, the local deaths from LTO emissions are 7.8% (AS), 5.1% (EU), and 1.7% (NA) of the global mortality attributed to full-flight emissions over the region. The low contribution of LTO to health impacts results from the large portion of overall impacts caused by ozone and the near zero ozone increase in the LTO-only scenarios, which might not happen to the same extent for the remaining 23% of aviation emissions occurring outside the three regions we focused on.

3.4. Effects of changing non-aviation emissions

The regional sensitivities presented in the previous sections were calculated for 2005, including a 2005 ‘baseline’ atmosphere in terms of other (non-aviation) anthropogenic and biogenic emissions, as well as meteorology. Background (non-aviation) emissions, a key component to different air quality sensitivities, as observed in this study and elsewhere

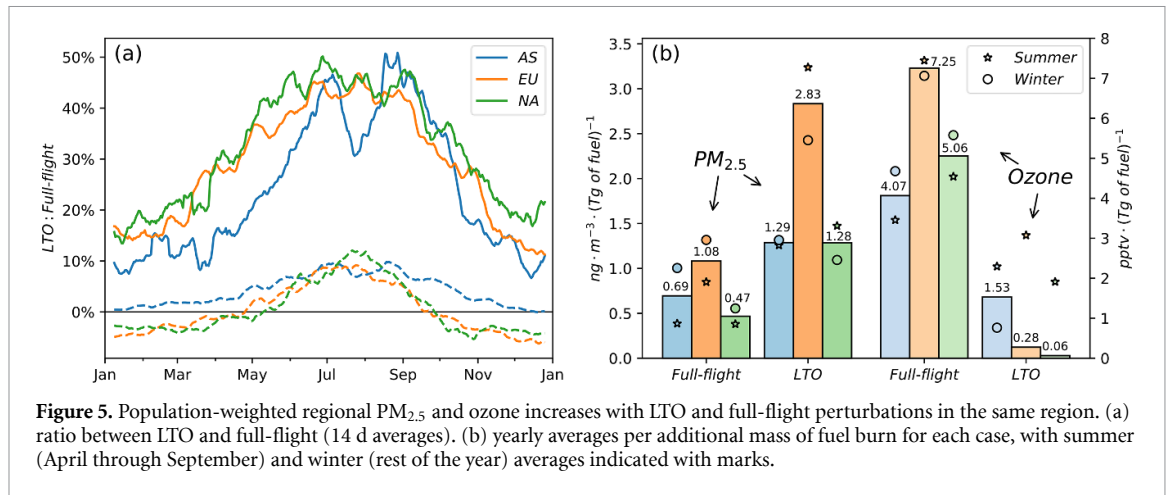


Figure 5. Population-weighted regional $PM_{2.5}$ and ozone increases with LTO and full-flight perturbations in the same region. (a) ratio between LTO and full-flight (14 d averages). (b) yearly averages per additional mass of fuel burn for each case, with summer (April through September) and winter (rest of the year) averages indicated with marks.

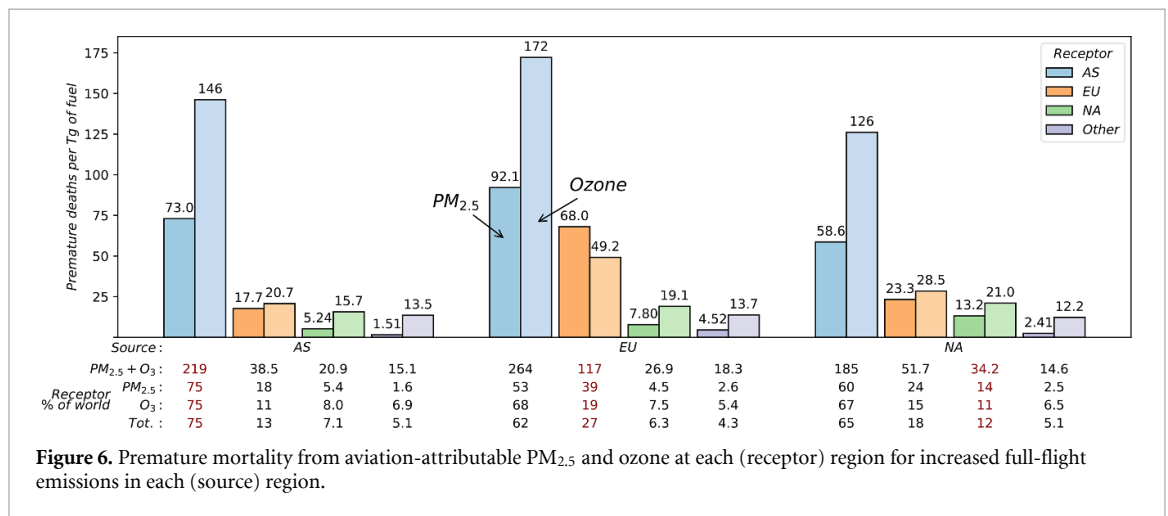


Figure 6. Premature mortality from aviation-attributable $PM_{2.5}$ and ozone at each (receptor) region for increased full-flight emissions in each (source) region.

Table 2. Aviation-attributable premature mortality by region of emission per mass of fuel burned [deaths(Tg of fuel)⁻¹] (95% confidence interval), the fraction of mortality by each pollutant and the fraction of mortality in the same region of perturbation.

Perturbation region	Mortality increase	$PM_{2.5}$:ozone impacts	Inside:outside
AS	294 (195–433)	33:67	75:25
EU	427 (284–641)	40:60	27:73
NA	285 (190–422)	34:66	12:88

(Woody et al 2011), are also changing over the years (Holt et al 2015, Dedoussi et al 2020). To quantify the effect of the background emissions and meteorology have on the sensitivity to aviation emissions, we also calculate the coarse regional sensitivities using non-aviation emissions and meteorology fields representative of the year 2013.

The sensitivities of global population $PM_{2.5}$ exposure to full-flight emissions are higher in the 2013 scenarios for perturbations in aviation emissions over AS (+8.2%), EU (+6.6%), and NA (+12.3%). The sensitivity of global $PM_{2.5}$ to aviation emissions over EU is still 57%–60% higher than to aviation emissions in the other regions. While intra-regional $PM_{2.5}$ sensitivities changed by ~10%, the cross-regional sensitivity changes ranged from -29% to +16%. The sensitivities of global population ozone

exposure to full-flight emissions change less: +1.5%, -1.4%, and +1.4% for perturbations over AS, EU, and NA, respectively.

Overall, increases of both $PM_{2.5}$ and ozone sensitivities are larger for intra-regional source-receptor pairs (SI section S6), suggesting that the relative importance of LTO emissions increases. This is consistent with the higher GR and FNR observed, however differences in meteorology may also be driving these sensitivity changes, and these are not directly decoupled here. An LTO-only perturbation simulated for the EU domain results in higher air quality impacts for the same amount of added emissions compared to the 2005 scenario (25% more perturbation-induced $PM_{2.5}$ exposure and 6.4% more ozone exposure). The ratio of population $PM_{2.5}$ exposure associated with LTO and

full-flight emissions over EU was 8.8% for 2013 versus 7.5% for 2005.

These results underscore the importance of considering aviation emissions in the context of those from other sectors when estimating aviation's future air quality impacts, as well as the effects of aviation-related mitigation measures in the future, particularly for PM_{2.5} impacts.

3.5. Limitations

While the most recent global aviation emissions inventory available to us is currently for 2005, air traffic has approximately doubled since that time (International Civil Aviation Organization 2018), and the world population increased by over 15%. While this may not necessarily affect the sensitivity values presented here, it is possible that second order air quality sensitivities to aviation emissions become significant for emission increases beyond the ranges evaluated here (SI section S2.5). In addition to the effects of changing non-aviation emissions on the sensitivities presented in section 3.4, air quality impacts from aviation could also be magnified by climate change effects on the atmosphere (Silva *et al* 2017). The effects of long-term evolutions in atmospheric composition and meteorology on the air quality sensitivity to aviation were not investigated in this study.

Water and aerosol from aviation emissions influence cloud formation and radiative properties, with these effects being a significant part of aviation's climate impacts (Lee *et al* 2009). Aerosol interaction with clouds affect aerosol microphysical evolution (condensation, agglomeration, reactivity, photolysis rates, etc) and wet deposition. Since our simulations use prescribed meteorology with these interactions being simulated in a parameterized manner, we do not capture this secondary effect of changes in clouds affecting particulate matter back. In addition, the near-surface aerosol impacts of aviation induced cloudiness could also be considered in future research.

Toxicity of PM_{2.5} constituents was considered to be uniform, i.e. not being specific to their individual chemical composition or particle morphology. There is, however, growing evidence that particulate matter toxicity to humans is dependent on factors besides total PM_{2.5} mass which are not accounted for here (Cassee *et al* 2013, Jonsdottir *et al* 2019). Considering uniform PM_{2.5} toxicity could underestimate the relative importance of LTO emissions, which have higher (ultrafine) black carbon components.

4. Conclusion

Using multiple simulations of a global and regional atmospheric CTM, we isolate the air quality and human health impacts of aviation emissions from different regions. We find significant intercontinental

effects, with 73% and 88% of premature mortality caused by aviation emissions over Europe and North America, respectively, occurring outside those regions. The largest receptor of health impacts from aviation emissions over any of the three regions was Asia, due to principally a larger population. While total health impacts are driven largely by population densities, the air quality impacts of emissions are also driven by atmospheric conditions. Higher PM_{2.5} sensitivities are associated with ammonia availability (GR) and ozone sensitivity to LTO emissions is associated with the formaldehyde to NO_y ratio. The same amount of emissions leads to higher PM_{2.5} and ozone increases, and ultimately cause an average of 45%–50% more health impacts if it is emitted over Europe instead of North America or Asia.

The cross-boundary nature of air quality impacts from aviation indicates that regional full-flight regulations alone in Europe and North America would yield the majority of corresponding air quality benefits outside those regions, while the opposite holds for Asia. Our findings highlight the need to take non-LTO emissions into account when evaluating air quality, as they are associated with 83%–89% of health impacts considering the same region as source-receptor, and 92.2%–98.3% of global health impacts caused by emissions in each region.

The regional differences in the human health impact sensitivities to aviation emissions that we observe underscore the importance of considering aviation emissions in the context of those from other sectors, particularly since background atmospheric composition remains a driver of aviation's regional impacts when we decouple population distribution.

Overall, our findings indicating that the same amount of aviation emissions can have impacts of significantly different magnitudes depending on the emission location suggest that this non-uniformity could be taken into account in policies aiming to minimize total health impacts from aviation more efficiently, considering the projected globally heterogeneous growth of aviation emissions. Finally, our results indicate that when optimizing aircraft design, operations and/or regulatory decisions for minimizing aviation's environmental footprint (air quality, climate, noise), full-flight (LTO and non-LTO) emissions need to be taken into account, as well as their regional distribution.

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Data availability statement

The data that support the findings of this study (atmospheric model outputs used in the calculations and the baseline mortality rates considered) are openly available at the following DOI: [10.4121/uuid:842594f5-6ebc-4150-afbd-7fcaa407aa.db](https://doi.org/10.4121/uuid:842594f5-6ebc-4150-afbd-7fcaa407aa.db).

ORCID iDs

Flávio D A Quadros  <https://orcid.org/0000-0002-0023-7863>

Mirjam Snellen  <https://orcid.org/0000-0002-6579-5655>

Irene C Dedoussi  <https://orcid.org/0000-0002-8966-9469>

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