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The contribution of aviation NO_x emissions to climate change: are we ignoring methodological flaws?

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Overview

The contribution of aviation to anthropogenic climate change results from CO_2 and non-CO_2 emissions. The latter emissions comprise of nitrogen oxides, water vapour, and aerosols as well as contrail and contrail-cirrus effects. A series of updates can be noted in recent studies related to the effects of NO_x-emissions; the inclusion of two physical processes and an updated radiation calculation (see below). However, in our opinion, two further published methodological shortcomings have not been fully considered which leads to a considerable underestimation of the contribution of aviation's NO_x emissions to climate change. First, methane response calculations implicitly assume steady-state instead of an adequate transient development. Second, most studies determine ozone changes are caused by switching off or reducing aviation NO_x emissions, instead of calculating aviation contributions to the ozone. Such methodological simplifications largely underestimate the contribution of the aviation NO_x emissions to climate change by a factor of 6 to 7 and can thereby be considered as flaws. Note that the contribution of an emission to climate change (=‘status report’) and the contribution of a change in emissions to climate change (=‘mitigation option’) require different calculation methods [1, 2]. While for calculating the contribution of emissions to atmospheric compositions (and hence climate change), to which we are referring here, a clear recommendation was made (e.g. [1]), the methodological approach for evaluating mitigation measures might still be ambiguous, but should certainly not ignore the results of contribution calculations [3].

Aviation's contribution to climate change

Current estimates of the contribution of aviation to the near-surface temperature change amount to roughly 5% of the total anthropogenic warming, with an uncertainty range of 2%–14% [4]. The larger part of the warming results from non-CO_2 effects among which the formation of contrails and its transition into contrail-cirrus has been recently widely discussed [5]. Contrail formation depends on the atmospheric conditions (temperature and humidity), aircraft characteristics (overall propulsion efficiency) and fuel characteristics (H_2O-emission index and specific heat content). The hot and moist exhaust mixes with the ambient air and becomes saturated with respect to liquid water, leading to the formation of droplets, which freeze if the temperature is low enough; and they persist if the ambient air is saturated with respect to ice. Water vapour emissions are not only triggering contrails, but also lead to an enhancement of the atmospheric water vapour concentration, which in total contributes only little to the aviation's contribution to climate change. However, for individual flights operated at higher altitudes water vapour emissions may have a larger effect than on average [6]. Aviation particle emissions have a small contribution to the atmospheric particle concentration and hence their direct contribution to climate change is small (e.g. [7]). However, they may largely affect contrail properties and a reduction of the number of soot particles also leads to a reduction of the formed ice particles in the contrail, which reduces their impact on climate [8].

Finally, aviation NO_x emissions play an important role. Besides playing a role in air quality effects [9], they contribute to climate change by formation of ozone and destruction of methane. As both are greenhouse gases, ozone build up adds to global warming, while the destruction of methane reduces global warming. Altogether, the warming effect largely dominates [4, 10–14]. In addition, the methane change has further implications on the atmospheric composition. Methane is a precursor for ozone, so a decrease in

Table 1. Estimates of the contribution of aviation NO_x emissions to the climate change in terms of RF for the year 2005. Starting with the results presented in Lee *et al* (2009) [4] (column 1) and adding additional processes such as PMO and SWV (column 2). PMO is taken with a mean factor of 40% of the methane RF, based on different estimates (29% [21], 58% [22], 23% [17], 47% [16] and 42% [23]); and SWV with a factor of 15% [18, 24]. The revised formula for calculating the methane RF (column 3) can be found in Etminan *et al* (2016) [19]; original in Myhre *et al* (1998) [25] (see also supplementary material for the calculation). Note that the SWV and PMO RF-calculation is not affected by the revision of the RF-methane formula, since the relationship is established with the original one. Instead, the transient calculation of methane changes also affects the feedback on ozone (PMO) and stratospheric water vapour (SWV). Corrections for methane lifetime (column 4) and ozone contribution method (column 5) are applied as explained in this work. The ozone contribution from aviation is taken from Dahlmann *et al* (2011) ([26] see their supplementary material for the time period 2000–2009).

				Correction of flaws	
Radiative forcing of aviation NO_x emission in 2005 in $mW\ m^{-2}$	Lee *et al* 2009	Additional processes (PMO, SWV)	Revised methane RF formula	#1 Methane lifetime	#2 Ozone contribution method
Ozone	26.3	26.3	26.3	26.3	41.2
Methane	−12.5	−12.5	−15.4	−10.0	−10.0
PMO		−5.0	−5.0	−3.3	−3.3
SWV		−1.9	−1.9	−1.2	−1.2
Total NO_x-RF	13.8	6.9	4.0	11.8	26.7

methane due to aviation NO_x emissions leads to a decrease in background ozone, which is called 'primary mode ozone' (PMO) [15–17]. Additionally, less methane enters the stratosphere, where it is decomposed into carbon dioxide and water vapour. Eventually, this reduces the stratospheric water vapour (SWV) concentration and since water vapour is a greenhouse gas, it reduces climate warming [18]. Finally, the formula of calculating the radiative forcing (RF) for methane concentration changes was recently updated, now including the representation of shortwave radiation effects [19]. This leads to a stronger negative methane RF of aviation NO_x emissions. Available literature shows that the estimate of the total NO_x-RF decreased by adding the effects of PMO, SWV, and the revised RF-formula from 13.8 $mW\ m^{-2}$ [4] to 4.0 $mW\ m^{-2}$ for the year 2005 (table 1), leading to the conclusion that besides CO_2 only contrails play a significant role in aviation's contribution to climate change [20].

Two methodological flaws

We think that in addition to the two new process-based effects (PMO and SWV) and the revision of the RF-formula, two additional methodological revisions have to be taken into account to avoid two major flaws. These have been already published, but have not adequately entered the scientific discussions: first, the calculation of the aviation methane concentration changes relies on the calculation of the methane lifetime change due to aviation NO_x emissions, commonly expressed in relative lifetime changes (δ). These relative lifetime changes are applied to the methane concentration (C) to obtain the resulting concentration change ($\Delta C = \delta \times C$). This approach implicitly assumes steady-state for the methane response. However, the perturbation lifetime of methane is 12 years, contradicting to the steady-state assumption (see also the supplementary material, available online at stacks.iop.org/ERL/14/121003/mmedia and e.g. [27]). Taking this lifetime change as a transient response [28], which it actually is, reduces the respective methane RF response by 35% [29] and since the PMO and SWV effects are directly related to the methane concentration change this reduction also extends to the estimate of RF due to PMO and SWV (table 1).

The second flaw concerns the method for estimating the contribution of NO_x emissions to the ozone concentration. Most studies compare two simulations, with and without (or reduced) aviation emissions. This is called sensitivity or perturbation approach. Clappier *et al* (2017) [1] (in agreement with other studies, e.g. [2, 30]) have clearly stated that 'when the relationship between emissions and concentrations is nonlinear, sensitivity approaches are not suitable to retrieve source contributions'. To overcome this short-coming, contribution approaches have been introduced in the past [31–33]. And their use reveals that the sensitivity method largely underestimates contributions, e.g. for biomass burning by a factor of two to four [34] and for land transportation by a factor of two [35]. Dahlmann *et al* [26] applied a source contribution method, and the results for aviation in the year 2005 are about a factor of 1.8 larger than those values reported for using the sensitivity approach and hence this methodological discrepancy agrees well with the above mentioned studies [34, 35]. Taking the values from Dahlmann *et al* [26] largely increases the estimate of the contribution of aviation NO_x emissions to climate change from 4 $mW\ m^{-2}$ to 26.7 $mW\ m^{-2}$, i.e. by a factor of 6 to 7 (table 1). Note that the source contribution and perturbation approach leads to identical results, in linear systems, only (Clappier *et al* (2017), Grewe *et al* (2010), Grewe (2013)). NO_x-ozone chemistry, however, is strongly nonlinear, showing an ozone depletion for low NO_x concentration (e.g. tropical oceanic regions) and very high NO_x concentrations (e.g. polluted cities) and peaking positive net-ozone production rates when both groups of ozone precursors, (a) the carbon compounds, such as methane, carbon monoxide and other hydrocarbons and (b) nitrogen oxides are well balanced ([36, 37] Dahlmann *et al* (2011)).

Implications

It is important to stress that this correction of flaws has two major implications: first on the weighting of individual aviation non-CO_2 effects with respect to their impact on climate and second how to assess mitigation options. Concerning the first implication, the aviation CO_2 and NO_x emissions lead to a RF in the year 2005 in the range of 25 to 30 mW m^{-2} ([4] and this work) and contrails to around 50 mW m^{-2} [5, 38]. With respect to the near-surface temperature change, this weighting changes; and the contribution of aviation NO_x emissions to climate change is getting a larger weight, whereas that of contrails is reduced [39]. This larger weight results from two effects: First, the climate sensitivity parameter is larger for aviation ozone changes compared to CO_2 and especially contrail-cirrus. Second, the temperature change is based on the temporal evolution of radiation changes, whereas the RF ignores those. For increasing emission scenarios, as for aviation, short-term effects are thereby stronger weighted. Finally, this leads to the conclusion that all three effects, CO_2, NO_x and contrails have a similar importance with respect to their contribution to climate change [39].

The second implication relates to the question of how to evaluate mitigation options in strongly nonlinear relationships, such as the NO_x-ozone relationship. While Clappier *et al* [1] raised the point that 'source apportionment methods[3] are not appropriate to evaluate the impact of abatement strategies', Grewe *et al* [3] clearly stated that 'the use of the tagging method (see footnote 1) makes the evaluation of mitigation measures more robust', since this evaluation becomes largely independent on other conditions, e.g. the timing of the mitigation option, implementation of other mitigation options, and background concentrations. Note that this agrees with the limitations of the perturbation or sensitivity approach given in Clappier *et al* [1], who stressed the point that 'the calculated impacts will only provide information for the exact conditions'. Hence using the perturbation approach for evaluating the potential of mitigation options makes this assessment vulnerable to any other emission reduction (also in other sectors), whereas using the contribution method results in a much more robust assessment.

As a consequence, assessments of climate mitigation options for aviation are recommended to consider these methodological aspects and to address CO_2 and non-CO_2 effects, including the climate impact from NO_x emissions and contrail formation.

Any data that support the findings of this study are included within the article.

[3] Note that source apportion and tagging methods are largely synonyms for contribution methods.

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