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We use the EMAC (ECHAM/MESSy Atmospheric Chemistry) global climate-chemistry model coupled to the aerosol module MADE (Modal Aerosol Dynamics model for Europe, adapted for global applications) to simulate the impact of aviation emissions on global atmospheric aerosol and climate in 2030. Emissions of short-lived gas and aerosol species follow the four Representative Concentration Pathways (RCPs) designed in support of the Fifth Assessment Report of the Intergovernmental Panel on Climate Change. We compare our findings with the results of a previous study with the same model configuration focusing on year 2000 emissions. We also characterize the aviation results in the context of the other transport sectors presented in a companion paper. In spite of a relevant increase in aviation traffic volume and resulting emissions of aerosol (black carbon) and aerosol precursor species (nitrogen oxides and sulfur dioxide), the aviation effect on particle mass concentration in 2030 remains quite negligible (on the order of a few ng m^{-3}), about one order of magnitude less than the increase in concentration due to other emission sources. Due to the relatively small size of the aviation-induced aerosol, however, the increase in particle number concentration is significant in all scenarios (about 1000 cm^{-3}), mostly affecting the northern mid-latitudes at typical flight altitudes (7–12 km). This largely contributes to the overall change in particle number concentration between 2000 and 2030, which results also in significant climate effects due to aerosol-cloud interactions. Aviation is the only transport sector for which a larger impact on the Earth's radiation budget is simulated in the future: The aviation-induced RF in 2030 is more than doubled with respect to the year 2000 value of -15 mW m^{-2} , with a maximum value of -63 mW m^{-2} simulated for RCP2.6.

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1 Introduction

Civil aviation is the fastest growing transport mode. Lee et al. (2009) reported a growth of air traffic (in terms of revenue-per-kilometer) of 38 % between 2000 and 2007, while several future scenarios (Kahn Ribeiro et al., 2007) project an increase of CO₂ emissions from aviation of about a factor of 2 between 2010 and 2030, with an even faster increase up to 2050. Although this sector accounts for a small fraction of the global CO₂ emissions from fossil fuels (2.6 % in the year 2004, Lee et al., 2010), it has a substantial impact on climate due to a wide range of non-CO₂ effects including ozone formation and methane destruction via NO_x emissions, direct and indirect aerosol effects from sulfate and black carbon (BC), the formation of contrail and contrail-cirrus clouds as well as the perturbation of natural cirrus clouds due to BC (see Sausen et al., 2005; Lee et al., 2010, and references therein). In addition to impacts on the climate, emissions of particulate matter from aircraft and related activities at and in the vicinity of airports can have detrimental effects on air quality and related impacts on human health (Herndon et al., 2004; Schürmann et al., 2007; Herndon et al., 2008). The study by Barrett et al. (2010) has also found significant impacts on air quality from aircraft emissions at cruise level, but these results have been questioned by Lee et al. (2013a).

In this work, we analyse the aviation impact on aerosol and climate for different future scenarios. We focus on the year 2030 in the four Representative Concentration Pathways (RCPs, Moss et al., 2010; van Vuuren et al., 2011a). These scenarios were developed in support of the Fifth Assessment Report of the Intergovernmental Panel on Climate Change (IPCC AR5). The results presented here complement those of Righi et al. (2015, hereafter R15), focusing on land transport and shipping. These two papers together represent a follow-up study of Righi et al. (2013, hereafter R13). In R13, we considered year 2000 emissions and performed several sets of model simulations to estimate transport impacts on atmospheric aerosol, to quantify the uncertainty in the effects on particle number concentrations related to the assumed particle size

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distribution of emitted particles, and to explore the non-linearities in the aerosol response to the perturbations induced by transport emissions. Concerning aviation, the R13 study revealed that aircraft emissions perturb the aerosol distribution in the upper troposphere, particularly at northern mid-latitudes, and have significant impacts also near the surface, mainly due to activities nearby airports. The aviation-induced impact on the Earth's radiation budget was found to be quite uncertain, strongly dependent on the assumed size distribution of particles emitted by aircraft. R13 estimated a radiative forcing in the range of -70 to 2.4 mWm^{-2} . The by far largest part of this effect was attributed to low clouds, lying much below the typical emission altitude, in line with the results by Gettelman and Chen (2013) and, more recently, Kapadia et al. (2015).

The numerical experiments in this work have been conducted using the EMAC-MADE global aerosol model (see R13 and references therein), which is able to track both aerosol mass and number perturbations, and to simulate the aerosol-cloud and aerosol-radiation interactions, hence allowing the estimation of aerosol radiative forcing effects. In addition to the global effects, we also analyse specific regions, where aviation emissions are expected to change significantly compared to 2000, as a consequence of changes in the transport patterns. Since we focus on the relative contribution of aviation to global aerosol and climate, changes in the background concentrations due to the effect of the anthropogenic emissions from other sectors are shown as well. Finally, we note that in the present study only the changes in the emissions of short-lived species (aerosol and precursor gases) are considered, whereas the investigation of the impact of a changing climate on aerosol distribution is not accounted for (we refer to Pye et al., 2009; Kloster et al., 2009; Megaritis et al., 2013, for a discussion on this topic). For this reason, we drive the model using year 2000 conditions for long-lived species (CO_2 and methane) and radiatively active gases (other than water vapour), and use meteorological data for the period 1996–2005 to nudge the model dynamics. The analysis in this paper focuses on the same period.

The future aviation impacts were investigated by Unger et al. (2013), who considered RCP4.5 for the background combined with several aviation emission scenarios.

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They found that cooling effects induced by short-lived compounds can neutralize or even overcome the CO₂ warming, depending on the time horizon under consideration. That study, however, did not take into account the RF due to aerosol-cloud interactions, which we found to be significant for year 2000 emissions, when considering low-level clouds, and potentially much larger than the CO₂ forcing (R13). Olivié et al. (2012), found a positive climate impact of non-CO₂ compounds from aviation using the previous IPCC SRES A1B scenario (Nakicenovic et al., 2000) up to 2100. Their model accounts for both direct and indirect effects of aerosols, but the indirect effect is limited to the sulfate component, which may lead to an underestimation of the total indirect effect. Our study, therefore, represents a step forward since it includes a more complete representation of the aerosol indirect effect, including the recently-reported aviation effect on low clouds, in the framework of the most recent RCP scenarios. Including the four RCPs in our experiments also allows us to provide a more complete analysis of the future development of aviation impacts on aerosol and climate.

This paper is organized as follows: A brief overview of the EMAC-MADE model system and its setup, including the considered emissions is provided in Sect. 2. The impact of aviation on the atmospheric aerosol distribution and aerosol burdens is discussed in Sect. 3, while Sect. 4 presents the corresponding aerosol-induced climate impacts. The main conclusions are summarized in Sect. 5.

2 Model setup, emission inventories and model simulations

The simulations of this work are performed with the ECHAM MESSy Atmospheric Chemistry (EMAC) model, coupled with the Modular Aerosol Dynamics submodel for Europe (MADE), adapted for global applications. We refer to R13 and R15 for a more detailed description of the model system and its configuration, here we only summarize its main features. MADE describes the aerosol population by means of three log-normal aerosol modes (Aitken, accumulation, and coarse) and eight aerosol species: Black carbon (BC), particulate organic matter (POM), nitrate (NO₃), ammonium (NH₄),

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sulfate (SO₄), mineral dust, sea-salt and aerosol water. To reduce the computational burden, the chemistry setup is based on a simplified mechanism, including basic tropospheric background chemistry (NO_x-HO_x-CH₄-CO-O₃ chemistry) and the sulfur cycle. The model is able to simulate the competition for ammonium between the sulfate and nitrate formation processes, which is particularly important in the upper troposphere, as we will show in Sect. 3. All model experiments are performed with a T42L19 resolution (corresponding to 2.8° × 2.8° in the horizontal) and 19 vertical layer up to 10 hPa, and covering a period of 10 years. The model dynamics (temperature, winds, and logarithm of surface pressure) is nudged using the data from the European Centre for Medium-range Weather Forecast (ECMWF). This minimizes the dynamical differences between the different experiments and allows to extract a significant signal even with a relatively limited amount of simulated years.

The model's ability to reproduce the vertical aerosol distribution was evaluated by Aquila et al. (2011), using observational data from several aircraft campaigns over the globe. They concluded that the aerosol representation in the UTLS (Upper-Troposphere Lower-Stratosphere) by EMAC-MADE is reasonably good.

We have applied the emission datasets developed by Lamarque et al. (2010) in support of the IPCC and the four Representative Concentration Pathways for the future projection in 2030 (RCP; Moss et al., 2010; van Vuuren et al., 2011a). As extensively discussed in R15, the RCPs are defined based on the projected value of the global anthropogenic radiative forcing in 2100 and includes four projections with increasing climate impact: RCP2.6, RCP4.5, RCP6.0, and RCP8.5. The RCP are climate-policy scenarios, but are quite limited in terms of air-quality projections, as they do not cover the full range of air pollution mitigation policies currently available (Chuwah et al., 2013). As pointed out by R15 and several other studies (e.g., Takemura, 2012), they often show an opposite behaviour between long-lived and short-lived species.

The aerosol submodel MADE used for this study simulates both aerosol mass and number, and therefore requires number emissions to be provided as input. These have been calculated from mass emissions under specific assumptions on the size distri-

5 bution of emitted particles. In R13, we analysed several sets of parameters to test the effect of such assumptions on the simulated impacts of transport on aerosol distributions and climate. To reduce the computational burden, in the present study we have performed experiments assuming only one set of parameters to describe the size distribution of emitted particles. Namely, we have used the parameters as for the reference case in R13. Compared to the other cases assumed in R13, this is a middle-of-the-road choice, based on measured values by Petzold et al. (1999) in the engine exhaust of a B737-300 aircraft, and combined with an assumption for fuel sulfur content ($0.8 \text{ g(SO}_2\text{) kg}_{\text{fuel}}^{-1}$) suggested in the assessment by Lee et al. (2010).

10 All the RCPs project a steady increase in aviation emissions of short-lived compounds between 2000 and 2030. This is significantly different from the other two transport sectors discussed in R15, for which a decrease in emissions of aerosol and aerosol precursors was found in most regions for all scenarios: This is a consequence of air pollution control measures (land transport) and sulfate reduction policies (shipping), which lead to emission reductions despite the steady growth in traffic volumes of these two sectors. For large commercial aircraft, on the contrary, no significant changes in technology are foreseen for aircraft engines in the near future, as no viable alternative to jet engines has been identified (Sims et al., 2014). Therefore the growing emissions are essentially a direct consequence of increasing air traffic volumes. The development and the implementation of new technologies in the aviation sector is more difficult than for road traffic and shipping, given the much higher safety standard required by aircraft, the relatively long life-time of the commercial fleet (about three decades), and the necessity to keep costs low (Kahn Ribeiro et al., 2007). Current efforts to minimise the climate impact of aviation are focusing on the improvements of fuel efficiency, modification of aircraft routes (Grewe et al., 2014a) and introduction of low-sulfur fuels (Unger, 2011; Bock, 2014).

25 As there are relevant regional differences in the distribution of air traffic, we have analysed the emission changes between 2000 and 2030 on the global scale and in three different regions of intense air traffic: The flight routes connecting the USA and

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This approach could have some limitations due to non-linearities in the response of the system to the emission perturbation. For the aviation sector, however, R13 found that the impact of the non-linearities is small. The aviation effects calculated for the year 2030 are related to the effects in the year 2000, as simulated by R13, in terms of the changes D in aviation-induced surface-level concentrations between 2000 and 2030 for the different scenarios. In analogy to R15, this is given by the difference:

$$D_{\text{RCP}}^{\text{AIRC}} = \Delta_{\text{RCP}}^{\text{AIRC}} - \Delta_{2000}^{\text{AIRC}}. \quad (2)$$

To provide a more complete view on the changes in aviation-induced aerosols, we relate this quantity to the total changes in concentration (i.e., from all sources):

$$D_{\text{RCP}}^{\text{ALL}} = \text{REF}_{\text{RCP}} - \text{REF}_{2000}, \quad (3)$$

and to the changes in the concentrations induced by other (non-aviation) sources:

$$D_{\text{RCP}}^{\text{OTHER}} = \text{NOAIRC}_{\text{RCP}} - \text{NOAIRC}_{2000}. \quad (4)$$

Note that, of course, $D_{\text{RCP}}^{\text{ALL}} = D_{\text{RCP}}^{\text{AIRC}} + D_{\text{RCP}}^{\text{OTHER}}$.

3 Aviation impacts on aerosol in 2030

The aviation impact on BC, SO₄, NO₃ and particle number concentrations in fine mode (i.e., $\lesssim 1 \mu\text{m}$, sum of the Aitken and accumulation mode) are plotted in Figs. 3–6, respectively. We consider zonally averaged fields from the surface to the UTLS. As a reference for comparison, the first row depicts the year 2000 results from R13 showing the concentrations induced by all sources (left), by aviation only (middle) and by other (non-aviation) sources (right). R13 showed that the aviation impact on particle mass concentration is quite negligible, being around 0.1 ng m^{-3} for BC, $2\text{--}5 \text{ ng m}^{-3}$ for SO₄, and $2\text{--}3 \text{ ng m}^{-3}$ for NO₃. The relevance of these impacts, however, depends also on the

simulated background aerosol concentrations in the UTLS, which for BC is known to be biased high in most global models (Schwarz et al., 2013) and, to a lesser extent, in EMAC-MADE (Aquila et al., 2011). The aviation impact is much larger on particle number concentration, in particular in the northern mid-latitudes between 200 and 300 hPa, contributing about 30–40 % of total concentration in this region. This is due to the relatively small size of particle emitted by aviation (around 25 nm in our simulations).

The other rows of Figs. 3–6 show the changes between 2000 and 2030 in the concentrations induced by all sources (left), by aviation only (middle) and by other (non-aviation) sources (right). For BC (Fig. 3), SO₄ (Fig. 4) and NO₃ (Fig. 5), the contribution of the aviation sector to the mass concentration changes remains small, as it is clear from the comparison of the left (all sources) and the right (non-aviation sources) columns. The only noticeable feature is the increase in aviation-induced BC concentration for RCP2.6 (Fig. 3, second row). This contributes to increase the overall BC concentration in the tropopause region of the Northern Hemisphere (left panel), which would be otherwise characterized by a decrease (right panel). In the other scenarios, the pattern of aviation-induced BC is similar but the perturbations are clearly smaller than in RCP2.6. In these scenarios, changes are relevant only in the upper-troposphere. RCP2.6 is the only scenario showing a significantly increasing impact of aviation on BC concentration close to surface, with mean values of 0.1–0.3 ng m⁻³ around 30° N. This is more than a factor of 3 larger than the year 2000 impact, and is even more important given that the impact of other sources is getting smaller over the same time period. This could be an issue for air-pollution control in the vicinity of major airports. However, due to the reasons discussed in Sect. 2, it should be questioned whether the assumptions of high aviation emission shares in RCP2.6 are realistic. Another interesting aspect is the negative value for aerosol nitrate in Fig. 5. This is a typical effect of the NH₃-limited environment in the UTLS (Unger et al., 2013, R13), and is due to the competition between nitrate and sulfate for the available ammonium.

The increase in mean aviation-induced particle number concentration (Fig. 6), on the other hand, is quite strong in all scenarios, with values of the order of 1000 cm⁻³ in the

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and the shortwave radiation in this calculation and analyse all-sky and clear-sky fluxes separately. The clear-sky flux is determined online by the model by neglecting clouds in the radiative flux calculations. Comparing the all-sky and the clear-sky effects, an estimate of the aviation-induced cloud radiative forcing can be inferred.

5 The results for the year 2000 (R13, reference case) and for the four RCPs in 2030 are presented in Fig. 8. In R13, we conducted several sensitivity simulations to quantify the uncertainties in the RF related to the assumption on the size distribution of emitted particles. As mentioned in R15 and in Sect. 2, this analysis is not repeated here in order to reduce the computational burden. Only the reference case of R13 is simulated for the
10 RCP scenarios in 2030. Nevertheless, we estimated the RF uncertainty for the 2030 results by simply rescaling them according to the uncertainty range calculated for 2000. These ranges are shown as open boxes in Fig. 8. This rescaling assumes that the same uncertainty can be applied to 2000 and 2030. On the one hand, this is reasonable since no fundamental changes in the aviation engine technology are expected in this time
15 period. On the other hand, however, there are some limitations. The upper limit of this estimate was found to be non-significant in R13 therefore even the sign of the RF in this case is uncertain, while the lower limit could be overestimated by this rescaling. Non-linearities in the system response to such large emission perturbation might reduce the actual lower limit of the RF estimate for 2030 presented here. Addressing these issues
20 will require additional experiments and shall be the focus of future analyses.

Given the results of R15 for land transport and shipping, aviation is the only transport sector for which an increasing impact of aerosol on the radiation budget is simulated. RCP2.6 is the scenario with the largest increase, shifting the all-sky forcing from -15 mW m^{-2} in 2000 to -63 mW m^{-2} in 2030, while values between -30 and -40 mW m^{-2} are calculated for the other scenarios. The total forcing is mostly driven by
25 cloud effects and the large increase with respect to 2000 can be explained by the large increase in aviation sulfate emissions (Fig. 2). Considering the uncertainties associated with the size distribution, extremely large values can be estimated, up to -285 mW m^{-2} for RCP2.6, if the NUC (i.e., nucleation) size distribution of R13 is assumed, charac-

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ties depending on the assumptions on emitted particle size distribution and fuel sulfur content, which even affect the sign of the resulting RF. Compared to the estimates for the year 2000, aviation-induced aerosol radiative forcing is about two (RCP6.0) to four times (RCP2.6) larger.

5 Together with Righi et al. (2013) and Righi et al. (2015), this paper closes a series of three studies on the global impact of land transport, shipping and aviation on atmospheric aerosol and climate in the year 2000 and in the RCP scenarios in 2030. The present study reveals that aviation is the only transport sector for which an increasing impact is simulated in the future. This is essentially due to continuously growing traffic
10 volumes without the implementation of significant technological improvements to reduce the emissions, as it is happening for the other sectors (e.g., emission control on vehicles engines and fuel regulations in shipping). In this set of studies, we also found that transport-induced aerosol particles can efficiently perturb low-level warm clouds, resulting in a strong cooling effect on the Earth's radiation budget, often comparable
15 or stronger than the warming effect of other compounds, like CO₂ and ozone. Future policies addressing the aviation sector should therefore focus on reducing its climate impact. Recent studies suggested a promising approach, based on optimised aircraft routes to reduce climate impacts (Grewe et al., 2014a, b). The focus so far has been on CO₂ and nitrogen oxides, but it shall be extended to include aerosol effects on clouds,
20 especially warm clouds as the results of our study suggest. Another possibility to reduced the aviation impact is the replacement of conventional (fossil) fuel by alternative fuels and biofuels. This will change the amount of emissions, with potential effects on contrail formation and evolution, as well as indirect cloud effects. In light of the results presented in this work, the role of sulfur content of aviation fuels will be of particular
25 relevance. Hence, detailed knowledge on the combustion process, emissions, and related climate impact of biofuels is required.

The estimate of the global impact of the transport sectors using global aerosol-climate models is still affected by several uncertainties. Some of them have been addressed in our series of papers: We showed, for example, that the parameters describ-

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ing the size distribution of emitted particles can be critical and can significantly influence the resulting estimates, especially concerning transport-induced aerosol number concentrations and, as a direct consequence, aerosol-cloud effects and RF calculations. The parameterisation of sub-grid scale processes is a general issue in global models, due to their coarse spatial resolution. In an extensive parametric study, Lee et al. (2013b) have shown that the sub-grid production of a few per cent mass of sulfate particles in plumes is much more important for the uncertainty on cloud condensation nuclei calculation than the SO₂ emissions themselves. This is consistent with our findings for the aviation sector, where a much larger climate impact is simulated when an extra nucleation mode for sulfate particles is considered. We also showed that sulfur content of aviation fuels can play a relevant role in the resulting climate impacts. We estimated that using basic assumptions and two extreme cases, but in the future it will be important to achieve better constraints, also including geographically-dependent sulfur emission factors, since the aviation fuel type often depends on the departure airport.

Finally, we recall that the model version applied for these studies considers a simplified representation of aerosol effects on ice clouds. Only homogeneous freezing of supercooled liquid aerosol is considered, but heterogeneous nucleation processes, such as soot-induced ice formation, are neglected. In the future, we shall improve this aspect of the model, including heterogeneous nucleation via various processes. This will also require an improved representation of aerosol in the UTLS, which is essential for a correct representation of the aerosol concentration and hence a more precise estimate of the impact of aviation emissions.

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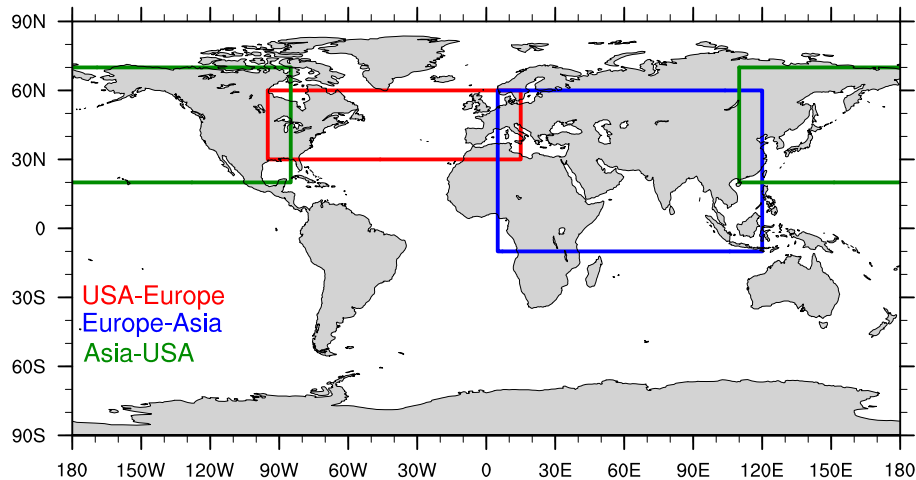


Figure 1. The regions selected for the analysis of the aviation impacts.

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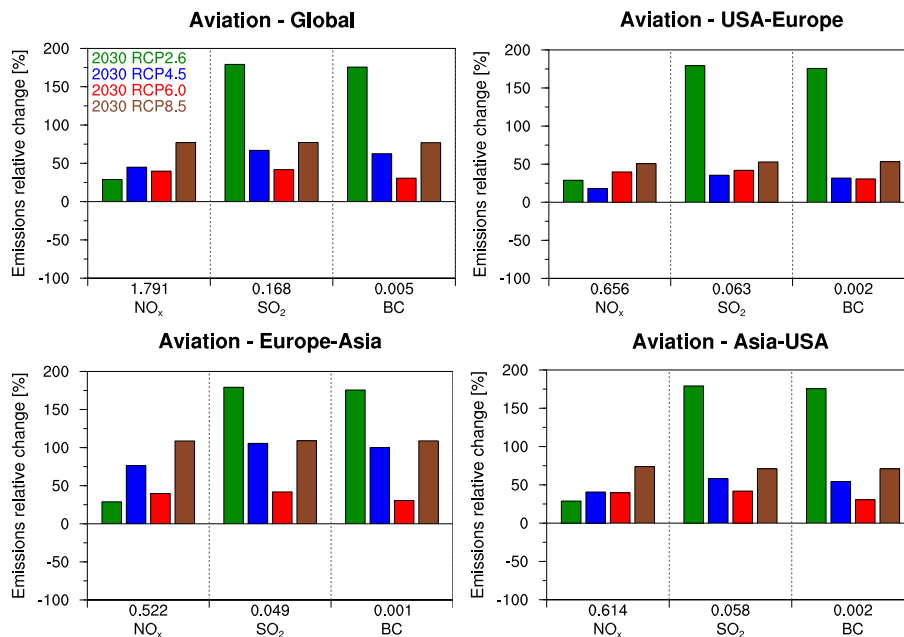


Figure 2. Relative changes in the emissions from aviation in the year 2030 with respect to 2000, for the four RCP scenarios. The changes are calculated globally (top left) and for the regions defined in Fig. 1. Total emissions for the year 2000 are indicated at the bottom of each panel, in units of Tg(species) a⁻¹ for SO₂ and BC, and Tg(NO) a⁻¹ for NO_x.

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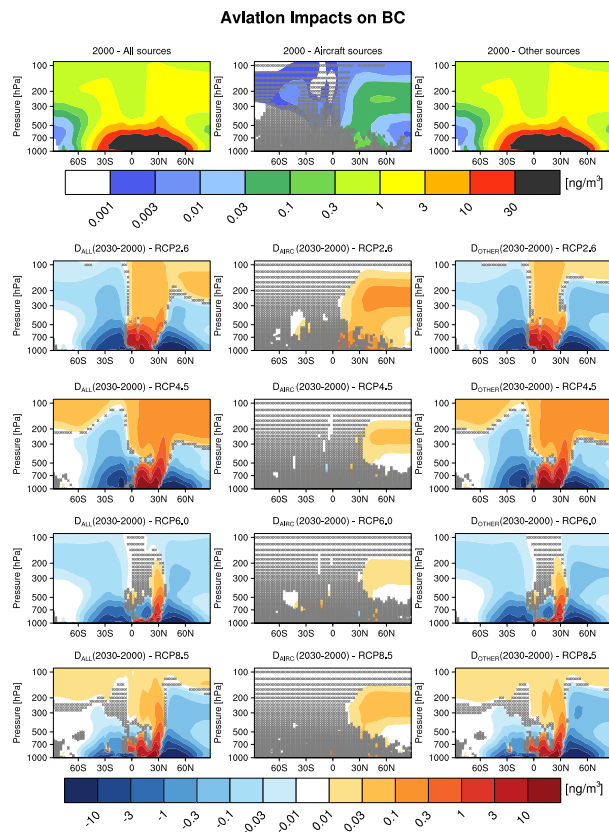


Figure 3. Annual average zonal mean concentrations of BC. The first row shows the results for year 2000 emissions: Total concentration (REF_{2000} , left), the concentration induced by aviation ($\Delta_{2000}^{\text{AIRC}}$, middle) and the concentration induced by other sources (NOAIRC_{2000} , right). The lower four rows show the changes in these quantities between 2000 and 2030 for the four RCPs, as given in Eqs. (2)–(4). Grid points where the difference is not statistically significant according to a univariate t test (5% error probability) are hatched.

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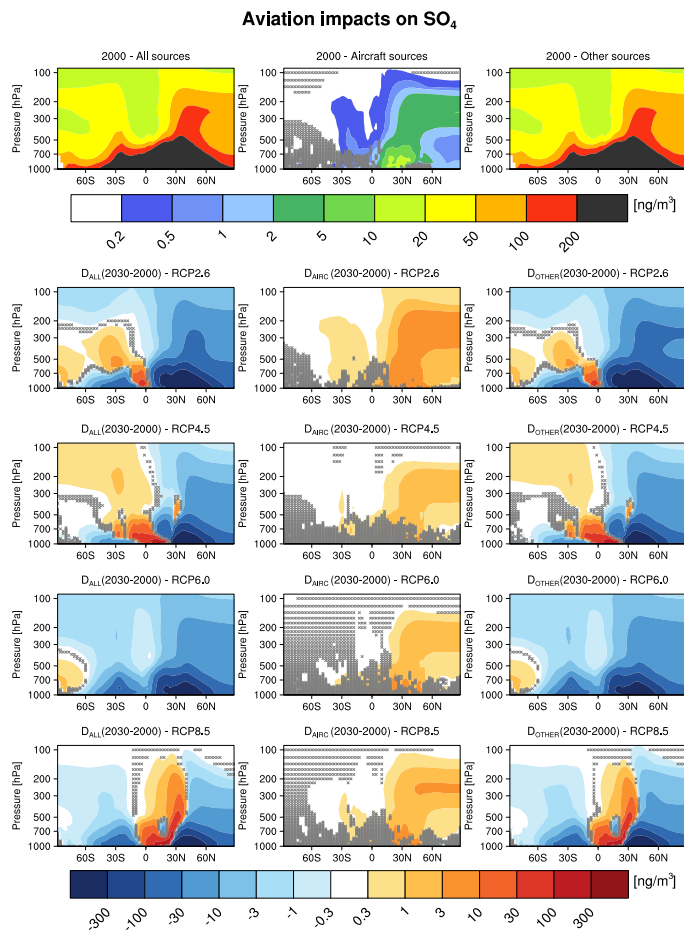


Figure 4. As in Fig. 3, but for aerosol sulfate concentration.

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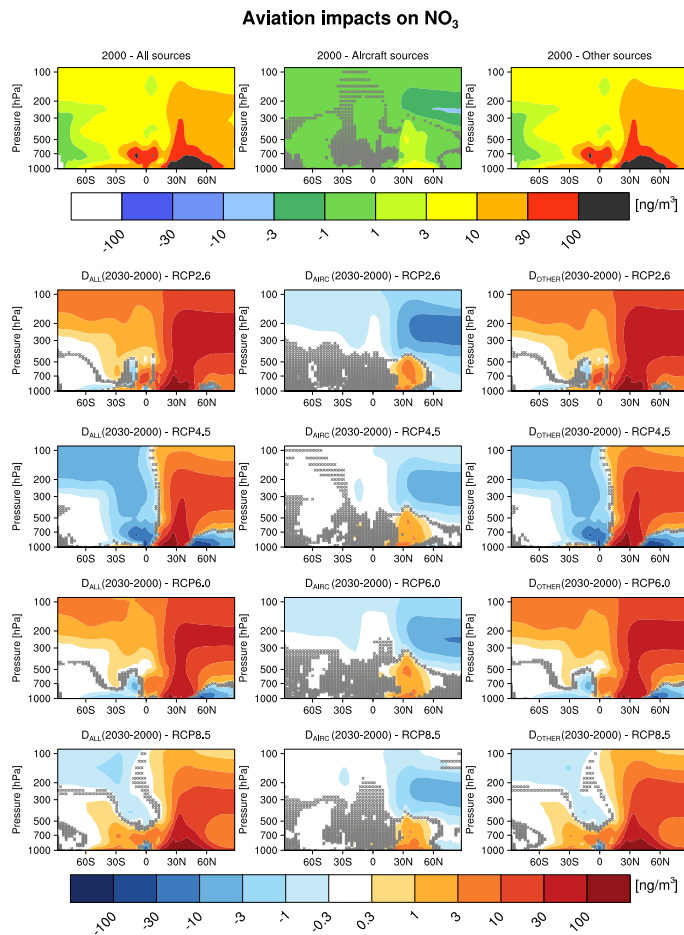


Figure 5. As in Fig. 3, but for aerosol nitrate concentration.

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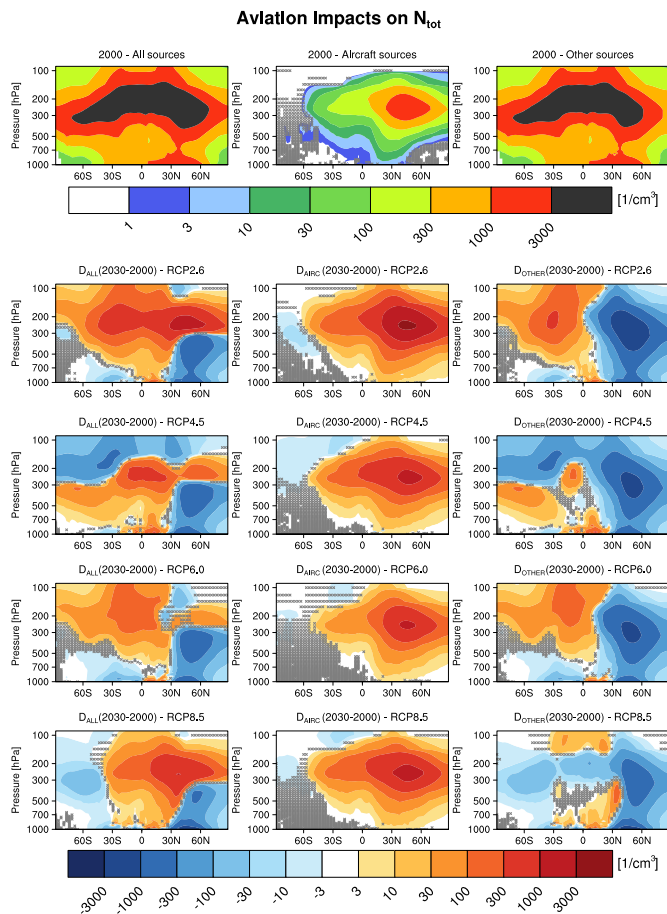


Figure 6. As in Fig. 3, but for fine particle ($\lesssim 1 \mu\text{m}$, sum of the Aitken and accumulation mode particles) number concentrations.

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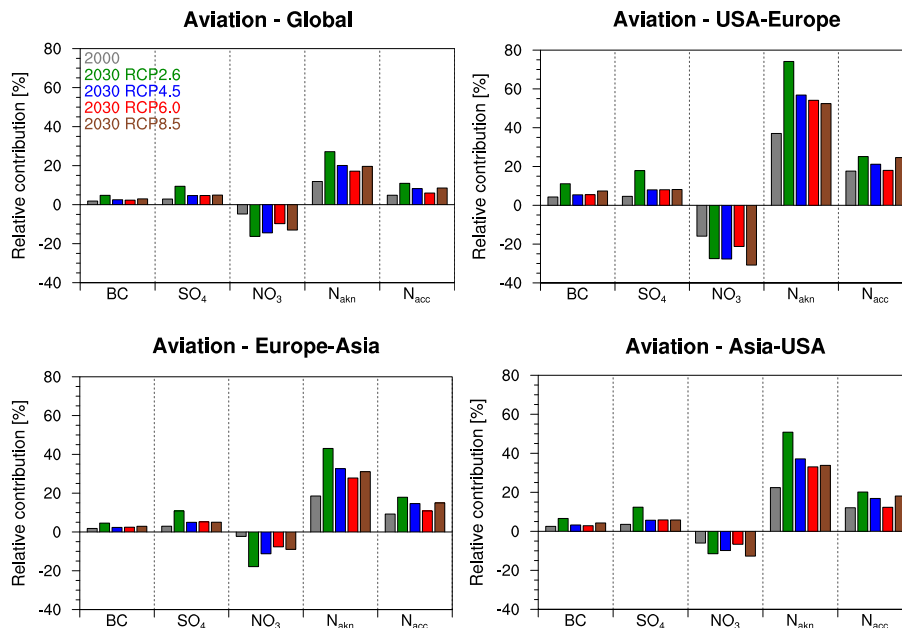


Figure 7. Relative contributions of aviation to the average mass and number burdens of selected aerosol species and different particle size modes (Aitken and accumulation mode), respectively. Results are shown for the year 2000 and for the four RCP scenarios in 2030. The values are integrated in model layers 7 to 9 (~ 8–13 km) globally and in three different regions (defined in Fig. 1).

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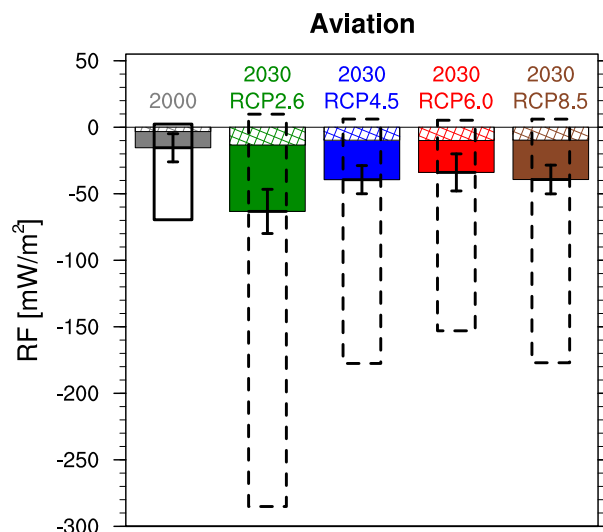


Figure 8. Global mean all-sky RF resulting from aviation emissions in the year 2000 (gray bar) and for the four RCP scenarios in 2030 (colored bars). The hatched part of each bar is the corresponding clear-sky forcing, calculated neglecting the radiative effects of clouds. The whiskers represent the 95 % confidence interval with respect to the interannual variability. The boxes correspond to the uncertainty range derived from the assumption on the size distribution of emitted particles and fuel sulfur content, as calculated by R13 for the year 2000 (solid) and rescaled here to the 2030 values (dashed).

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