DO AIRCRAFT BLACK CARBON EMISSIONS AFFECT CIRRUS CLOUDS ON THE GLOBAL SCALE ?

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Abstract

Potential modifications of cirrus clouds caused by aircraft-generated black carbon (BC) soot particles were investigated with a global atmospheric model. Many details of the role of BC in cirrus cloud formation are currently not well known. Therefore, hypothetical scenarios based on various assumptions on the ice nucleation efficiency of background and aircraft-induced BC particles were considered. Using these scenarios, the sensitivity of ice cloud microphysics to aviation-induced BC is studied. The model results suggest that cloud modifications induced by aircraft BC particles could change the ice crystal number concentration at northern midlatitudes significantly (10–40% changes of annual mean zonal averages at main flight altitudes), provided that such BC particles serve as efficient ice nuclei. The sign of the effect depends on the specific assumptions on aerosol-induced ice nucleation. These results demonstrate that significant cirrus cloud modifications by BC from aircraft and related climatic impacts cannot be excluded.

1 Introduction

Very fine particles in the typical size range of about 1 nm to 10 µm, so-called aerosols, are omnipresent in the Earth's atmosphere. Atmospheric aerosols, such as liquid sulfate or solid black carbon (BC) soot or mineral dust particles, can be important agents in cirrus cloud formation. Cirrus clouds can be formed by freezing of liquid aerosols (so-called homogeneous nucleation) or via ice nucleation induced by solid aerosols (so-called heterogeneous nucleation). By these mechanisms, anthropogenic aerosols can cause modifications of cirrus cloud properties and related climatic effects. The altitudes of midlatitude cirrus cloud occurrence coincide with the main cruise altitudes of commercial aircraft. This implies a potential of aviation-induced aerosols to affect cirrus cloud formation.

Several potential impacts of aviation on the atmosphere were discussed by the Intergovernmental Panel on Climate Change [1]. The impacts of sulfate or BC particles from aviation on cirrus clouds were among the most uncertain processes. Global simulations of cirrus formation via homogeneous freezing revealed that the impact of aircraft-induced sulfate particles on cirrus properties is likely negligible [2]. On the other hand, process model and observational studies suggest a potential impact of aircraft-emitted BC particles on cirrus clouds.

Process model studies [3] reveal that BC particles can modify cirrus clouds via heterogeneous nucleation if their number concentration is sufficiently high and if they nucleate ice considerably more efficient than liquid particles. Global model calculations [4] suggest that significant large-scale BC particle number concentrations occur in the upper troposphere and lowermost stratosphere (UTLS, main aircraft flight altitudes), with a significant contribution of aviation-induced BC. Several laboratory studies provided information about the ice nucleation efficiency of commercial (Degussa) BC particles and BC generated in the laboratory. However the freezing ability of atmospheric BC particles, including aircraft-generated BC, remains uncertain.

In a previous study [5] we considered hypothetical scenarios of the freezing ability of potential nuclei for heterogeneous ice nucleation (so-called ice nuclei or IN), particularly BC and mineral dust particles, and showed by means of global simulations that heterogeneous ice nucleation has the potential to perturb cirrus clouds globally. However, aircraft-induced particles were neglected in these investigations. The present study aims at evaluating the sensitivity of the simulated global cirrus cloud properties to aircraft-induced BC perturbations. The question is addressed whether BC from aviation has the potential to change the microphysical properties of cirrus clouds globally. Therefore, global simulations were performed considering also the effects of aircraft-generated BC. These estimates represent the first global evaluation of such ice cloud modifications. A more detailed description of the results presented here can be found in a previous publication [6].

2 Methodology

The global atmospheric general circulation model (GCM) ECHAM4 [7] is applied in T30 spectral horizontal resolution which corresponds to approximately 3.8° x 3.8° geographical grid size. The vertical grid has 19 layers, ranging from the Earth's surface up to 10 hPa (about 30 km altitude). All model runs were performed for a 10-year time period following a 15-month model spin-up.

The mass-based aerosol module applied here treats the atmospheric cycles of sulfate aerosols, carbonaceous particles (organic and black carbon, separated into hydrophobic and hydrophilic particles), sea-salt, and mineral dust aerosols. The aerosol particle number concentrations are derived from the aerosol mass concentrations with prescribed lognormal size distributions. For the calculation of BC particle number concentrations, including those from aviation, we refer to our previous publications [4, 6]. The cloud module [2, 3, 5] predicts cloud liquid water, cloud ice, as well as cloud droplet and ice crystal number concentrations. It includes a parameterization of homogeneous freezing. Heterogeneous ice nucleation is considered assuming that BC and mineral dust particles are transformed into ice crystals when the relative humidity exceeds 130%, i.e. distinctly below the homogeneous freezing threshold.

Data from observational studies reveal that both heterogeneous and homogeneous nucleation can occur during cirrus cloud formation. Since the ice nucleation efficiencies of various particles are known only poorly, the details of the competition between homogeneous freezing and heterogeneous ice nucleation remain uncertain, which complicates a detailed quantification of aviation-induced cirrus changes. In addition to the occurrence of aerosols, also the cooling of air masses is an important prerequisite for cloud formation. Hence, the competition of homogeneous and heterogeneous nucleation also depends on the updraft speed which controls the cooling rate. This is another source of uncertainty in the present global model simulations since vertical velocity fluctuations relevant for cloud formation frequently occur on subgrid scales.

Due to these uncertainties, we apply a simplified description of the competition [6] considering only heterogeneous nucleation in the case of very high IN number concentrations (>0.5 particles/cm^3) when additional homogeneous freezing is unlikely to occur. In contrast, we assume that ice particles are formed solely by homogeneous freezing at smaller IN concentrations. Hence, heterogeneous ice nucleation on BC from aviation is considered here only if heterogeneous nucleation is likely to control cirrus formation. As discussed previously [6], these simplifications are appropriate for the sensitivity analysis performed here.

To assess the potential of aviation-induced cirrus changes, a high ice nucleation ability of aircraft-generated BC is considered. This is achieved as follows: (i) We assume that all aircraft-emitted BC particles can act as IN. (ii) By ignoring possible aging processes of aircraft BC particle populations [4], we maximize the calculated number concentrations of aircraft-induced BC particles. (iii) We assume that heterogeneous nucleation transforms all available IN into

ice crystals and, therefore, neglect that the number of crystals can be limited by the updraft speed.

Assumptions on heterogeneous ice formation	Analysis strategy	Main result
	Scenario 1	
Heterogeneous nucleation on mineral dust, BC from surface sources, and BC from aircraft	Comparison of simulations in/excluding ice formation of BC from aircraft	Aviation causes significant increase in ice crystal number concentrations
	Scenario 2	
Heterogeneous nucleation on BC from aircraft only; background freezes homogeneously	As Scenario 1	Aviation causes significant decrease in ice crystal number concentrations

Tab. 1. Summary of scenarios analyzed.

We investigate two different scenarios of the freezing efficiency of potential IN generated at the Earth's surface (Table 1). In scenario 1, all mineral dust and hydrophilic BC particles act as IN. A simulation considering heterogeneous nucleation on dust particles, on hydrophilic BC from surface sources, and on BC from aircraft is compared to a corresponding model experiment without aircraft BC. In scenario 2, heterogeneous nucleation can exclusively occur on BC from aviation. This is compared to a reference simulation considering homogeneous nucleation only.

3 Results

The annual mean global distribution of the number concentration of potential IN (mineral dust and hydrophilic BC) simulated for the 250 hPa pressure level (within main flight levels, approximately at 10 km altitude) is shown in Figure 1. In Figure 1a, only particles from surface sources are considered. These particles result from BC emissions due to fossil fuel combustion mainly in North America, Europe, and Southeast Asia as well as from BC emissions

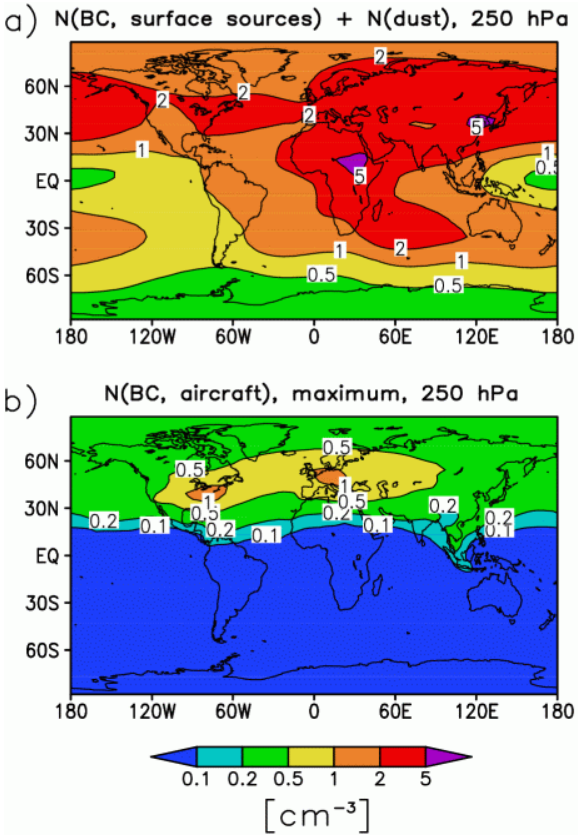

Fig. 1. Annual mean number concentrations [particles/cm^3] of potential IN at 250 hPa (within main aircraft flight level; approximately at 10 km altitude). (a) Mineral dust and hydrophilic BC particles originating from surface sources. (b) BC particles from aircraft (maximum estimate [4]). The Figure is adopted from a previous publication [6].

due to biomass burning and wind-driven mineral dust emissions occurring mainly at low latitudes in the northern hemisphere (NH) and in the southern hemisphere (SH). The annual mean number concentrations of these potential IN range between about 0.5 and 5 particles/cm^3 at 250 hPa. BC generally contributes to more than 50% (more than 70% at mid- and high latitudes) to these particle numbers. Observational data reveal that only a fraction of the available BC and dust particles serve as IN. Hence, the background (no aviation) IN number concentrations considered in scenario 1 are larger than the highest IN concentrations observed in the at-

mosphere and are therefore considered as upper limit background IN concentrations in the sensitivity analysis performed here. As the counterpart to scenario 1, we completely neglect IN from surface sources in scenario 2, but assume that the background aerosol freezes homogeneously.

Figure 1b highlights maximum number concentrations of aircraft-induced BC particles simulated for 250 hPa by neglecting possible reductions in the particle number-to-mass ratio due to aging of the fresh exhaust [4]. Since the exhaust particle aging is currently not well understood, this estimate is used here to evaluate the maximum potential of aircraft-induced IN perturbations (see also Section 2). These maximum concentrations of aircraft-generated BC particles range between about 0.2 and 1 particles/cm^3 at NH midlatitudes, where most of the commercial aircraft operate. Taking into account the number concentrations obtained for surface-derived IN (Figure 1a), this results in a relative aircraft-induced increase in the potential IN number concentration at 250 hPa of more than 20% in large parts of the NH. The largest increase at 250 hPa ranges up to 40%. Due to air mass transport, significant increases occur also far from the major emission regions. Even when a maximum estimate of the loss of aircraft BC particle number due to exhaust aging is considered [4] (not shown), the aviation-induced increase frequently exceeds 10% and amounts up to 20% in the most frequented flight areas. Hence, the aviation-induced BC perturbations simulated here pave the way for the possibility of related cirrus changes.

Figures 2a and 2d show annual averages of the zonal mean ice crystal number concentration (ICNC) simulated in the reference experiments (no heterogeneous ice nucleation on BC from aircraft) of scenarios 1 and 2, respectively. In both simulations, the typical annual mean (including cloud-free periods) ICNC range between 0.1 and 0.4 crystals/cm^3 at NH midlatitudes. The corresponding annual mean ice cloud occurrence ranges from 20 to 40% and shows no significant differences between the two experiments (not shown). In the NH, the availability of potential IN mostly is large enough (>0.5 particles/cm^3) [6] that cirrus cloud formation is controlled by heterogeneous nucleation in scenario 1 (Figure 2a) resulting in larger ICNC compared to the reference experiment of scenario 2 (Figure 2d) where cirrus clouds are formed by homogeneous nucleation.

Figure 2 also highlights the relative aviation impact on the annual mean ICNC simulated for the two scenarios. In scenario 1 (Figures 2b and 2c), mineral dust and BC particles serve as IN, and the addition of BC particles from aircraft leads to an increase in ICNC (see also Table 1). Significant increases in the range of 10–60% are simulated. The effect is restricted to NH mid- and high latitudes around the main flight levels.

In scenario 2, heterogeneous nucleation can occur on aviation-induced BC only. The impact of heterogeneous nucleation in this scenario is to compete with homogeneous nucleation, provided that sufficient aircraft BC particles are available. Figures 2e and 2f reveal that aviation causes a reduction of ICNC under these assumptions (see also Table 1) because the number concentration of aircraft-generated BC particles is lower than the number of aerosols frozen homogeneously in the reference experiment (ice nucleation on BC from aviation neglected) and the number of ice crystals formed from aircraft BC is large enough to prevent homogeneous freezing. The reduction occurs at NH midlatitudes and ranges between 10 and 60%, in terms of annual means. It is largest in areas of strong orographic forcing, where large updrafts lead to high ICNC resulting from homogeneous nucleation in the reference case.

In both scenarios, the aviation impact is significant over large areas of the NH. The aircraft-induced changes in ICNC are accompanied by weak changes in the ice crystal sizes (not shown). No significant changes in annual mean ice water content and cloud occurrence were simulated.

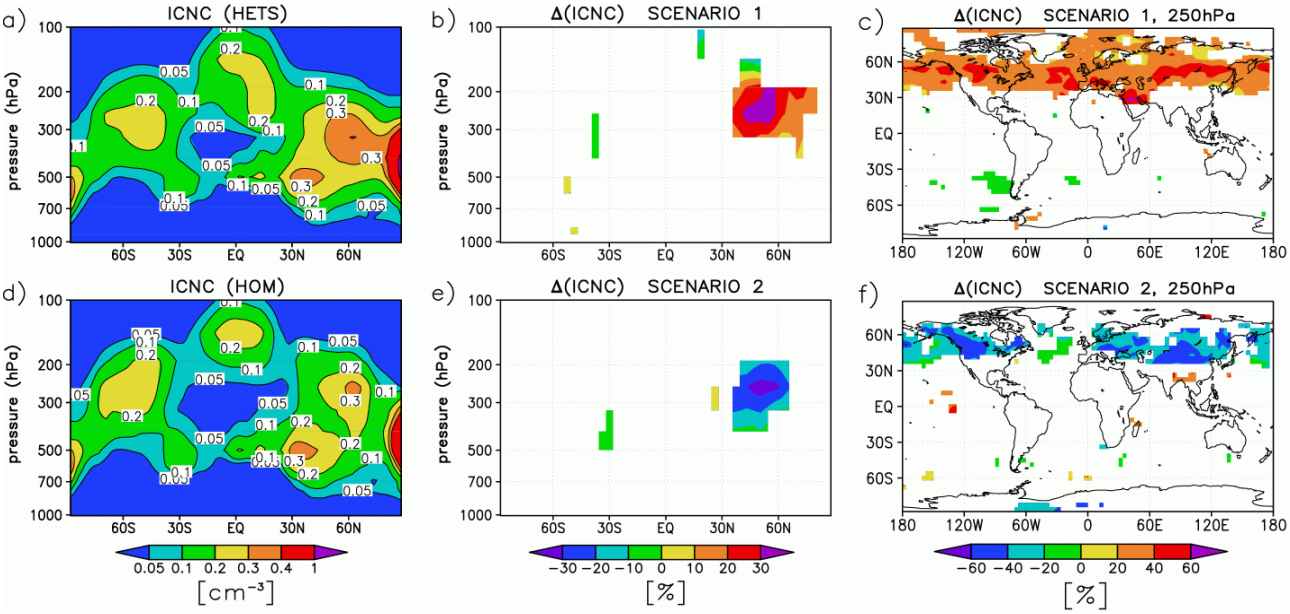

Fig. 2. Annual averages of the zonal (constant latitude) mean ice crystal number concentration (ICNC) in the reference experiments (no heterogeneous ice nucleation on BC from aircraft) of (a) scenario 1 (simulation HETS) and (d) scenario 2 (simulation HOM). The ICNC values were averaged over cloudy and cloud-free periods. Only clouds with an ice water content larger than 1 mg/kg(air) and a grid box fractional cloud cover larger than 1% were considered. Also shown are corresponding vertical (zonal mean) and horizontal (250 hPa) distributions of the relative change of the annual mean ICNC induced by heterogeneous ice nucleation on BC particles from aviation, simulated in (b, c) scenario 1 and (e, f) scenario 2. Only changes significant at the 95% level of the student t-test are shown. Note the different scaling of the contour levels chosen for the vertical (b, e) and horizontal (c, f) distributions of Δ(ICNC). The Figure is adopted from a previous publication [6].

4. Conclusions

The simulations presented here open the possibility of a significant impact of aircraft BC emissions on the number concentration of potential IN (BC and mineral dust particles) in the UTLS. Large-scale increases in the annual mean number concentration of potential IN in the range of 10–40% at flight altitude were simulated for large parts of the northern hemisphere. Provided that BC particles from aviation serve as efficient IN nucleating ice below the homogeneous freezing threshold humidity, significant aviation-induced perturbations of the ice crystal number concentration were simulated. These perturbations occur mainly at northern midlatitudes around the main flight altitudes. In the annual mean, they amount to 10–60% of the ICNC simulated neglecting the aviation impact. Aviation causes an increase or decrease in the ice crystal number concentration depending on whether we assume that 'background' (no aviation impact) cirrus cloud formation is dominated by heterogeneous (scenario 1) or homogeneous (scenario 2) nucleation, respectively. Both scenarios demonstrate that, based on the current knowledge, significant cirrus modifications by BC from aircraft cannot be excluded. In light of in-situ measurements of the IN number concentration performed in the UTLS, scenario 2 appears to be more likely than scenario 1. However, further investigations are required to corroborate this.

The uncertainties of the freezing properties of atmospheric IN and the subgrid-scale fluctuations of the vertical wind speed in the UTLS are currently too large to accurately quantify the aviation impacts on ice cloud properties. Further in-situ observations and laboratory measurements are urgently required to better constrain

global model scenarios. Corresponding improvements in the representation of ice clouds in GCMs should also be achieved [5]. Besides investigations of the effects of aircraft BC aerosols on cirrus, a better quantification of the impact of spreading, persistent contrails on cloud cover is also required for a complete assessment. If aircraft BC emissions were as effective in modifying the ICNC of cirrus clouds as suggested by this study, the related impact on the Earth's radiation budget should be determined.

Acknowledgments

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