

Do aircraft black carbon emissions affect cirrus clouds on the global scale?

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[1] Potential cirrus modifications caused by aircraft-produced black carbon (BC) particles via heterogeneous ice nucleation were studied with a general circulation model. Since the role of BC in cirrus cloud formation is currently not well known, 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 perturbations 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, based on the current knowledge, significant cirrus modifications by BC from aircraft cannot be excluded. **Citation:** Hendricks, J., B. Kärcher, U. Lohmann, and M. Ponater (2005), Do aircraft black carbon emissions affect cirrus clouds on the global scale?, *Geophys. Res. Lett.*, 32, L12814, doi:10.1029/2005GL022740.

1. Introduction

[2] Several potential impacts of aviation on the atmosphere were discussed by *Intergovernmental Panel on Climate Change* [1999], concluding that the indirect effects of aircraft particle emissions, i.e., the impacts of sulfate or black carbon (BC) particles from aviation on cirrus clouds, were among the most uncertain processes. Based on global simulations of cirrus formation via homogeneous freezing, *Lohmann and Kärcher* [2002] concluded that the impact of aircraft-induced sulfate particles on cirrus properties is likely negligible. On the other hand, process model and observational studies suggest a potential impact of aircraft-emitted BC particles on cirrus [*Jensen and Toon*, 1997; *Ström and Ohlsson*, 1998].

[3] Process model studies by *Kärcher and Lohmann* [2003] reveal that black carbon particles can modify cirrus clouds via heterogeneous nucleation if their number concentration is sufficiently high and if they nucleate ice sufficiently below the homogeneous nucleation thresholds, at relative humidities over ice (RHI) below about 140%. Global model calculations by *Hendricks et al.* [2004] suggest that significant large-scale BC particle number concentrations occur in the upper troposphere and lower-

most stratosphere (UTLS), with a significant contribution of aviation-induced BC. Laboratory studies of ice nucleation by commercial (Degussa) BC particles and BC generated in the laboratory give a range of nucleation threshold RHI from 110% up to water saturation [*DeMott et al.*, 1999; *Möhler et al.*, 2005]. However the freezing ability of atmospheric BC particles, including aircraft-generated BC, remains uncertain.

[4] *Lohmann et al.* [2004] considered hypothetical scenarios of the freezing ability of potential atmospheric ice nuclei (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. The present study aims at evaluating the sensitivity of the simulated global cirrus cloud properties to aircraft-induced perturbations of the BC number concentration. Therefore, global simulations were performed similar to *Lohmann et al.* [2004] but supplemented by aircraft-generated BC to investigate its potential to perturb cirrus clouds. These estimates represent the first global evaluation of such ice cloud modifications.

2. Model and Methodology

[5] The general circulation model (GCM) ECHAM4 [*Roeckner et al.*, 1996] is applied in T30 spectral horizontal resolution which corresponds to approximately $3.8^\circ \times 3.8^\circ$ grid size. The vertical grid has 19 layers, ranging from the surface up to 10 hPa. All model runs were performed for a 10-year time period following a 15-month model spin-up.

[6] 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 [*Feichter et al.*, 1996; *Lohmann et al.*, 1999]. The aerosol particle number concentrations are derived from the aerosol mass concentrations with prescribed lognormal size distributions, assuming external mixtures of the different aerosol compounds. For the calculation of BC particle number concentrations, including those from aviation, we refer to *Hendricks et al.* [2004]. The cloud module predicts cloud liquid water, cloud ice, as well as cloud droplet and ice crystal number concentrations. It includes a parameterization of homogeneous freezing [*Lohmann and Kärcher*, 2002]. Heterogeneous ice nucleation can optionally be considered as described by *Lohmann et al.* [2004], assuming that BC and mineral dust particles are transformed into ice crystals at RHI exceeding 130%, i.e., distinctly below the homogeneous freezing threshold.

[7] Ice nuclei seem only to modify, not control, cirrus cloud formation and properties [*Haag et al.*, 2003;

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Table 1. Summary of Model Runs and Main Results

Run	Assumed IN	Type of Run	Main Results of Scenario
<i>Scenario 1</i>			
HETS	BC from surface sources and mineral dust	Ref ^a	Difference
HETAS	as HETS, in addition BC from aircraft	Sens ^b	HETAS–HETS: $\Delta\overline{\text{ICNC}}^c > 0$ $\Delta\overline{\text{IWC}}^c, \Delta\overline{f}^c \sim 0$
<i>Scenario 2</i>			
HOM	no IN, homogeneous nucleation only	Ref ^a	Difference
HETA	BC from aircraft only	Sens ^b	HETA–HOM: $\Delta\overline{\text{ICNC}}^c < 0$ $\Delta\overline{\text{IWC}}^c, \Delta\overline{f}^c \sim 0$

^aReference simulation without aircraft BC.

^bSensitivity experiment including aircraft impact.

^cAnnual means of zonally averaged ice crystal number concentration (ICNC), ice water content (IWC), and ice cloud frequency (f) at northern midlatitude UTLS.

Cziczo *et al.*, 2004], except at enhanced IN concentrations [Gierens, 2003]. 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. The competition also depends on the updraft speed or cooling rate, respectively [DeMott *et al.*, 1997; Kärcher and Lohmann, 2003]. 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 [Lohmann and Kärcher, 2002].

[8] Facing these uncertainties, we apply a simplified description of the competition considering only heterogeneous nucleation in the case of high IN number concentrations ($>0.5 \text{ cm}^{-3}$) 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. In-situ observations suggest that IN concentrations larger than 0.5 cm^{-3} are comparatively rare in the UTLS [Rogers *et al.*, 1998] ($\sim 60\%$ of the measured IN concentrations are smaller than 0.01 cm^{-3} , $\sim 95\%$ smaller than 0.1 cm^{-3}). Such high IN concentrations result in cirrus cloud formation dominated by heterogeneous nucleation for a wide range of atmospheric conditions [Gierens, 2003]. Hence, heterogeneous ice nucleation on BC from aviation is considered here only if heterogeneous nucleation is likely to control cirrus formation. Since the modeled large-scale number concentration of aircraft-generated BC particles frequently exceeds 0.5 cm^{-3} in the main emission areas (Section 3 and Hendricks *et al.* [2004]), this simplification appears to be appropriate for the sensitivity analysis performed here.

[9] 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 [Hendricks *et al.*, 2004], 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.

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

3. Results and Discussion

[11] The annual mean global distribution of the number concentration of potential IN (hydrophilic BC and mineral dust) simulated for the 250 hPa pressure level (within main flight levels) is shown in Figure 1. The results were taken from the HOM simulation. 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 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 cm^{-3} at 250 hPa. BC generally contributes to more than 50% (more than 70% at mid- and high latitudes) to these particle numbers. These number concentrations, considered in scenario 1, are larger than the highest IN concentrations

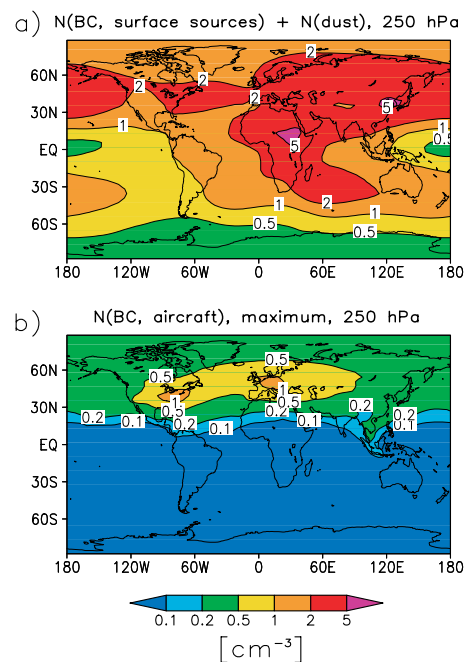


Figure 1. Annual mean number concentrations [cm^{-3}] of potential IN at 250 hPa (main aircraft flight level). (a) Hydrophilic BC and mineral dust particles originating from surface sources. (b) BC particles from aircraft (maximum estimate by Hendricks *et al.* [2004]). The results were taken from the HOM simulation.

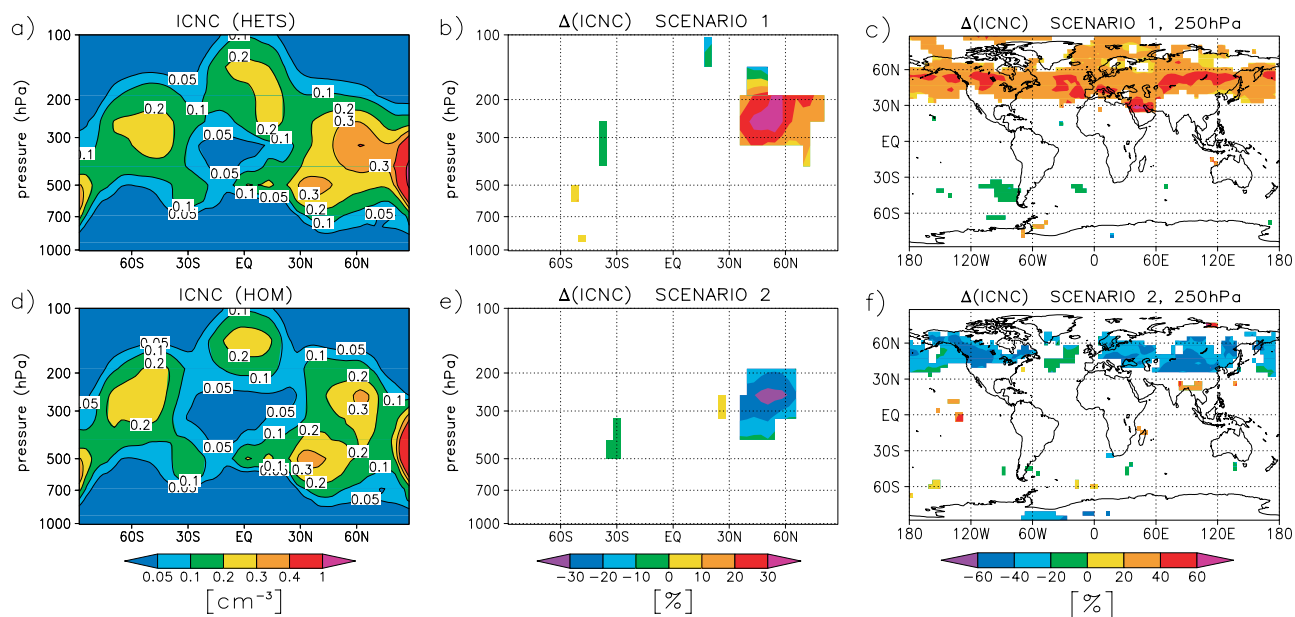


Figure 2. Annual means of the zonally averaged ice crystal number concentration (ICNC) in the (a) HETS and (d) HOM experiments. The ICNC values were averaged over cloudy and cloud-free periods. Only clouds with an ice water content larger than 1 mg/kg(air) and a grid box fractional cloud cover larger 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 (HETAS – HETS) and (e, f) scenario 2 (HETA – HOM). 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 $\Delta(\text{ICNC})$.

observed by *Rogers et al.* [1998] (Section 2) and are therefore considered as upper limit background (no aviation) IN concentrations in the sensitivity analysis performed here. As the counterpart to scenario 1, we completely neglect IN from surface sources in scenario 2.

[12] 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 [*Hendricks et al.*, 2004]. Since the exhaust particle aging is currently not well understood, this estimate is used to evaluate the maximum potential of aircraft-induced IN perturbations (Section 2). The maximum number concentrations range between about 0.2 and 1 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), the maximum estimate 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 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 [*Hendricks et al.*, 2004] (not shown), the aviation-induced increase frequently exceeds 10% and amounts up to 20% in the most frequented flight areas. This appears to be in contradiction to the measurements by *Rogers et al.* [1998] where no significant increase of the IN concentration in aircraft exhaust plumes was detected. However, *Chen et al.* [1998] stressed that this is likely due to the lower detection particle size limit of $\sim 0.1 \mu\text{m}$. Since aircraft-generated BC particles are mostly smaller, aircraft-induced IN perturba-

tions cannot be excluded and the potential perturbations simulated here pave the way for possible aircraft-induced cirrus changes studied below.

[13] Figures 2a and 2d show annual averages of the zonal mean ice crystal number concentration (ICNC) simulated in the reference experiments (HETS and HOM) of scenarios 1 and 2, respectively, without aircraft BC. In both simulations, the typical annual mean (including cloud-free periods) ICNC range between 0.1 and 0.4 cm^{-3} at NH midlatitudes. The corresponding annual mean ice cloud frequencies range from 20 to 40% and show no significant differences between the two experiments (not shown). In the NH, the availability of potential IN mostly is large enough ($>0.5 \text{cm}^{-3}$) that cirrus clouds form by heterogeneous nucleation in the HETS experiment resulting in larger ICNC compared to the HOM simulation.

[14] 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 (difference HETAS–HETS; 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.

[15] In scenario 2, heterogeneous nucleation can occur on aviation-induced BC only (HETA). 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 (difference HETA–HOM; see also Table 1) reveal that aviation causes a reduction of ICNC under these assumptions because the number concentration of aircraft-generated BC particles is lower than the number of aerosols frozen homogeneously in

the HOM experiment 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 HOM case.

[16] 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 frequency were simulated.

4. Conclusions

[17] Our simulations 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 IN nucleating ice below the homogeneous freezing threshold RHI, 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.

[18] 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 [Lohmann *et al.*, 2004]. Besides making headway in investigations of the indirect effect of aircraft BC aerosols on cirrus, a better quantification of the direct impact of spreading, persistent contrails on cloud cover is also required for a complete assessment.

[19] If aircraft BC emissions were as effective in modifying the ICNC of cirrus clouds as suggested by this study, the related radiative forcing should be determined. However, our simulations indicate that, in contrast to the overall indirect effect of anthropogenic aerosols on global climate, the aircraft indirect effect is too small to be identified as the difference between two separate GCM simulations. Too small signal-to-noise ratios would prohibit the quantifica-

tion of the corresponding radiative forcing. Solving this crucial signal detection problem requires a methodical extension of the Stuber *et al.* [2001] approach. This is beyond the scope of the present study but will be subject of future work.

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