Advancing the Science for Aviation and Climate

ACACIA

Deliverable 3.2

Title: Favourable atmospheric situations for aviation-induced effects

Lead partner: ETHZ

Project no.	875036
Instrument	Research and Innovation Action (RIA)
Thematic Priority:	H2020-MG-2018-2019-2020
Start date of project:	1 January 2020
Duration:	42 months
Date of report:	12 May 2022
Document authors:	Nadja Omanovic, Colin Tully, Ulrike Lohmann
Classification:	PU (Public)
File name:	ACACIA-Deliverable D3.2.pdf

TABLE OF CONTENTS

1 Summary

Aircraft emissions increased in the last 30 years and are expected to continue growing in the coming decades. Yet, our understanding of their impact on cloud formation and the subsequent effect on radiative forcing is still poorly understood. Here, we show, through a series of sensitivity studies, how on the one hand soot particles impact cirrus cloud formation, and on the other hand how the radiative and microphysical response changes once we remove aircraft emissions. Natural cirrus clouds are known to exert a warming impact on our climate. Allowing soot particles to enable ice crystal formation in our model leads to a warming of 0.018 ± 0.004 Wm^{-2}, highlighting the warming potential of additional emissions through aerosol-cloud interactions. Enabling soot to act as an ice nucleating particle increases heterogeneous nucleation, while reducing homogeneous nucleation. This confirms our knowledge of the competition between different ice nucleation modes in cirrus clouds. The impact of aviation is only weakly notable in slight changes of the prevalence of the nucleation modes. It appears that the aircraft emissions have a cooling impact on our climate, as removing them from our simulation leads to a net radiative forcing of -0.011 ± 0.004 Wm^{-2}. Given that previous studies reached similar still ambiguous results, we anticipate our findings to be a next step in evaluating the impact of aviation-induced clouds and their impact on our climate. Our findings can provide a useful basis for formulating mitigation strategies in the form of air traffic management or the usage of alternative fuels.

2 Introduction

When the COVID-19 pandemic hit the world in 2020, the aviation industry experienced an unprecedented decline in traffic, with AIRBUS transporting 2’700 million less people compared to 2019 (AIRBUS GMF, 2021). However, within one year the industry slowly started to recover and increase its traffic volume again. Current market forecasts indicate a steady growth over the next years with a two-year lag compared to growth forecasts from 2019 (AIRBUS GMF, 2021). The quick recovery during the pandemic and after previous crises (e.g., the 2008 financial crisis) shows the resilience of the industry (AIRBUS GMF, 2019). Today, air traffic constitutes 2.6 % of the annual global CO_2 emissions (Staples et al., 2018), and contributes 4 % to the total anthropogenic radiative forcing (Kärcher, 2018), and hence to the warming of the climate. In view of the continued growth of this sector in the coming decades, the need to understand the climate impacts of aviation, especially the indirect effects via cloud formation and modification, is becoming more crucial. Identifying and quantifying the aviation-induced effects on clouds may introduce new mitigation strategies next to reductions in CO_2 emissions from aviation to achieve negative emissions required for fulfilling the climate goals of the 2015 Paris Climate Agreement (Lee, 2018; IPCC, 2021). However, these indirect effects are still highly uncertain (Grewe et al., 2017), especially for aircraft-induced clouds (AIC), such as cirrus and contrail cirrus (Kärcher, 2018), which are the focus of this study.

Clouds have either a warming or cooling impact on the atmosphere that depends on their microphysical properties, altitude, thickness, and the surface albedo. Their net radiative forcing is the sum of two effects: the albedo and greenhouse effects. The former describes the ability of a cloud to reflect incoming solar radiation, and thus determines the magnitude of shortwave radiation that reaches Earth’s surface. The latter effect describes the trapping of longwave radiation emitted by the surface and/or the lower lying atmosphere, reducing the radiative transmission to space (Lohmann et al., 2016). A large albedo effect leads to a cooling as less shortwave radiation enters the atmosphere, while a large greenhouse effect results in a warming as the longwave radiation is kept in the atmosphere. Natural cirrus clouds exert a large greenhouse effect through the absorption of upwelling longwave radiation by numerous small ice crystals (ICs), (Kärcher, 2018). As these clouds are colder than the surface and the underlying atmosphere, they re-emit longwave radiation at a much lower magnitude. Simultaneously, they are transparent to incoming solar radiation giving them a low albedo effect. As a net response, natural cirrus exert a positive radiative effect, i.e., a warming effect, on climate (Lohmann et al., 2016). It is still an open question whether AIC possess similar microphysical properties and thus behave in a similar way to natural cirrus (Heymsfield et al., 2010).

Recent studies remain ambiguous on the sign of the radiative forcing, i.e., if AIC induce a warming or cooling effect on climate (Zhou and Penner, 2014; Penner et al., 2018; McGraw et al., 2020; Lee et al., 2020). One factor that introduces wide uncertainty is the formation of ice crystal via a nucleation process. Nucleation describes the formation and growth of a thermodynamically stable cluster within a metastable parent phase. For ice crystals, this is the emergence of the ice phase (stable phase) within a liquid or vapor phase (metastable phase), (Lohmann et al., 2016). There are two modes by which ice can form by nucleation: homogeneous and heterogeneous nucleation. The former refers to the freezing of aqueous solution droplets or cloud droplets at temperatures below -38 °C and at high supersaturations with respect to ice. The latter process occurs on the surface of an ice nucleating particle (INP), which lowers the energy barrier for ice formation (Cziczo et al., 2013; Kanji et al., 2017). Thus, heterogeneous nucleation can occur at higher temperatures and lower ice supersaturations than homogeneous nucleation. A variety of aerosols can act as INPs, including, most prominently, dust particles (Kanji et al., 2017) and, as especially relevant for aircraft emissions, soot particles (Mahrt et al., 2018; Mahrt et al., 2020; Lee et al., 2020; Lohmann et al., 2020). Introducing additional INPs (e.g., via aircraft emissions) in a low-aerosol environment such as at high altitudes can lead to the inhibition of homogeneous nucleation as the INPs consume the water vapor for ice formation, and thus reduce the supersaturation and inhibit the onset of homogeneous nucleation (Zhou and Penner, 2014; Jensen et al., 2016). Hence, a direct competition between the two nucleation processes exists that strongly governs the microphysical properties, and thus radiative properties, of cirrus clouds (Jensen et al., 2016; Kärcher, 2018).

The ice nucleation ability of soot remains highly uncertain (Kärcher et al., 2007; Marcolli, 2017; Mahrt et al., 2018). Mahrt et al. (2018) showed that soot particles can induce ice formation in cirrus well below the saturation needed for homogeneous freezing of solution droplets. Köhler et al. (2009) and Cziczo et al. (2009) found a decrease in nucleation efficiency once the soot particles were coated with sulphate. However, Mahrt et al. (2020) showed that additional aging of soot particles by sulphuric acid, which is also a constituent of aircraft emissions, leads to an enhanced ice nucleation ability of soot when compared to freshly emitted samples.

Once the aircraft emissions mix with the environmental air and favourable conditions for cirrus clouds exist, so-called condensation trails (contrails) form that may evolve into contrail cirrus, which do not have any line shape anymore and are then indistinguishable from natural cirrus (Kärcher, 2018). The occurrence of these AIC may lead to shifts in the radiative forcing, thus impacting the temperature structure of the atmosphere (Kärcher, 2018), which highlights the need to better understand AIC formation and their effects on the climate.

In this study, we aim to identify regions of cirrus formation and assess the competing and complementing cloud processes, especially to quantify the relevance of the ice nucleation modes on cirrus cloud properties, in the general circulation model (GCM) ECHAM-HAM. A new parameterization for the ice nucleating ability of soot allows us to quantify the effect of soot particles on cirrus formation, with a special focus on aircraft emissions and their impact on cloud processes. With the aviation sector projected to continue its growth over the next years, emissions are expected to increase (Lee et al., 2009; Lee et al., 2010, AIRBUS, 2021). The assessment of non-CO_2 effects of aviation may provide mitigation options that can be readily employed and, thus, contribute to the reduction of climate impacts. Hence, we aim to reduce the scientific uncertainty associated with AIC, their spatial distribution, and their radiative forcing with this study.

3 Data and Methods

We use the aerosol-climate model ECHAM6.3-HAM2.3, which is the sixth generation of the atmospheric circulation model ECHAM coupled to the second-generation aerosol microphysics module, HAM (Stevens et al., 2013; Mauritsen et al., 2019; Neubauer et al., 2019; Tegen et al., 2019). The horizontal resolution is 1.875 x 1.875 ° on a Gaussian grid (T63) with 47 vertical levels (L47) up to 0.01 hPa, which results in a vertical resolution of around 1 km in the upper troposphere at typical cirrus altitudes. The model time step is 7.5 minutes. Monthly mean sea surface temperature, sea ice coverage, and emissions data from the CMIP6 data set are prescribed (McDuffie et al., 2020).

We use an updated microphysics scheme (Dietlicher et al., 2018; Dietlicher et al., 2019) that describes ice under a single category, called the Predicted Particle Properties (P3) by Morrison and Milbrandt (2015). The P3 scheme removes unrealistic conversion rates between ice hydrometeors of different sizes and supersedes the two-moment scheme by Lohmann et al. (2007). In addition, a new parameterization Lohmann et al. (2020) was implemented by into ECHAM-HAM to allow black carbon (soot) to act as an INP in cirrus clouds. They parameterized the number of soot particles suitable for INPs as a function of temperature and ice supersaturation ratio (S_i) based on laboratory measurements (Mahrt et al., 2018, 2020). Their simulations show that the soot INPs are in competition for water vapor with pre-existing ice crystals, mineral dust INPs, and solution droplets. The newly introduced soot INPs are now an additional freezing mode in the cirrus ice nucleation scheme that is coupled to the P3 scheme. We use this new parameterization to determine the impact of aircraft emissions on the competition of the different nucleation modes.

We perform three simulations with 21 ensemble members each using the model setup described above. Each simulation was computed for the years 2000 to 2010 (inclusive), with three months of model spin up from October to December 1999. We were only able to capture 20 ensemble members for our ACOFF simulation for this report due to an unknown computational error." Nudging is applied for all model runs, i.e. the vorticity, divergence and surface pressure of the model are adjusted using reanalysis data to realistically represent the atmosphere at a given time. The results are calculated as annual monthly-weighted means. We test the impact of soot particles by prohibiting soot to act as an INP (REF), allowing it to act as an INP (SOOT), and by excluding all aircraft emissions (ACOFF). The simulations are performed in full nucleation competition between homogeneous, heterogeneous, and pre-existing ice crystals. The different ensemble members allow us to assess the statistical significance of the results.

4 Results

4.1 Radiative forcing

The change in net radiative forcing between two simulations is shown below in Table 1. We first compare the SOOT simulation with the REF simulation, where the only difference in model setup is the inclusion of the soot parameterization by Lohmann et al. (2020). The change in net radiative forcing amounts to 0.018 ± 0.004 Wm^{-2} indicating an additional warming by soot particles and their role in ice nucleation. Allowing soot to act as an INP significantly impacts the radiative forcing budget. In a second step, we compare the SOOT simulation with the ACOFF simulation. Removing all aircraft emissions from the emissions inventory (ACOFF) appears to lead to an additional warming of the climate compared to the SOOT simulation with a net negative forcing of -0.011 ± 0.004 Wm^{-2}. The aircraft emissions and their subsequent effects on cirrus cloud formation result in a cooling of the climate.

Table 1: Change in net radiative forcing (ΔF_{net}) given in Wm^{-2} between the simulations SOOT and REF, and SOOT and ACOFF. The uncertainty corresponds to the 95 % confidence interval based on the assumptions of two independent samples and a t-distribution.

	ΔF_{net} [Wm^{-2}]
SOOT – REF	0.018 ± 0.004
SOOT – ACOFF	-0.011 ± 0.004

Comparing our results to research conducted so far, we note that, e.g., Zhou and Penner (2014) conducted a series of sensitivity tests by varying the background concentration of dust and sulphate and showed the magnitude and sign of the radiative forcing can be altered. While the soot parameterization by Lohmann et al. (2020) assumes the aging of soot particles with sulphate leads a higher INP activity, Zhou and Penner (2014) restrict the ability of soot particles to form ice as soon as they are coated by three monolayers of sulphate. As discussed in the introduction, the INP potential of soot particles needs further investigations in the laboratory under conditions as close as possible to reality. McGraw et al. (2020) concluded that the inclusion of aircraft emissions and soot as an INP leads to a negative radiative forcing of -0.11 Wm^{-2}, which is an order of magnitude larger than what we find with our model. We note here that the study was conducted with a different model (CESM2.0.1) for only 5 years of simulations, and aircraft emissions were only considered in one sensitivity test, excluding it from all other tests. In contrast to Zhou and Penner (2014) and Lohmann et al. (2020), they parameterize the nucleation ability of soot particles according to Ullrich et al. (2017), which only considers freshly emitted soot particles as INPs.

It is important to note, that ECHAM-HAM does not differentiate contrails and contrail cirrus, but only shows the impact of soot particles on cirrus cloud formation in general. Hence, when comparing to literature values for contrails and contrail cirrus, we note this difference. However, we take the opportunity here to discuss current research and put our results into broader perspective. Kärcher (2018) computed a radiative forcing of 0.02 Wm^{-2} to 0.15 Wm^{-2} for contrail cirrus using modelling and observational data. Bock and

Burkhardt (2016) found a value of 0.056 Wm^{-2} based on one year model simulation. They emphasise the large uncertainties introduced by radiative transfer calculations. A recent study by Lee et al. (2021) showed that the radiative forcing increased from 2011 to 2018 reaching a value of 0.111 ± 0.078 Wm^{-2}. While our SOOT simulation agrees reasonably well with these studies, we find that aircraft emissions have a small cooling impact on a global scale.

4.2 Nucleation modes

Next, we examined the impact of soot particles on the two nucleation modes: homogeneous and heterogeneous nucleation. Figures 1, 2, and 3 show the difference in nucleation modes, and microphysical properties for the two simulations SOOT and REF, while Figure 4 shows the differences in nucleation modes on a global scale for the simulations SOOT and ACOFF.

In Figure 1 we present ICNC source anomalies at 200 hPa as this is a representative level for cirrus clouds and coincides with aircraft cruise altitude. Introducing soot as an INP into the model results in a change from homogeneous nucleation (ICNC_HOM) to heterogeneous nucleation (ICNC_HET). The increase in ICNC_HET is dominated by soot particles forming ice as seen in Figure 1d). Note, the sum of the anomalies in Figure 1c) and 1d) amount to the total ICNC_HET anomaly (Figure 1b). Here the competition between the two nucleation modes is evident, in line with previous studies (e.g., Spichtinger and Gierens, 2009; Jensen et al., 2016). The introduction of more INPs leads to the increased competition for the consumption of the same amount of water vapor. As already mentioned in the introduction, INPs require lower S_i, and thus can initiate ice crystal formation earlier by nucleating first and continuing to reduce the S_i as water vapor is consumed. In addition, pre-existing ice crystals lead to further reduction in S_i through vapor deposition (Zhou and Penner, 2014). Therefore, the required S_i for homogeneous nucleation may not be reached as often as in REF and thus total ice crystal number concentration originating from homogeneous nucleation may be partly reduced. Note, the regions with increases in heterogeneous nucleation correspond to regions with decreases in homogeneous nucleation, further highlighting the competition of these two nucleation modes. Further investigating the response in ICNC from soot particles (Figure 1d), we note that the strongest signals appear over continents reasonably close to their sources, such as industry, biomass burning, and aircraft emissions (Bond et al., 2013). Especially in the northern hemisphere the two prominent regions are North America and China. In the southern hemisphere the main source for soot particles is biomass burning that can be transported via large-scale circulation towards the higher latitudes. However, the transport towards the poles in ECHAM may be too efficient as noted by Schacht et al. (2019) and lead to an overestimation of the soot impact on ice crystal formation. In addition to the competition between the two nucleation modes, we can see a competition between dust and soot particles. It appears that the formation of ice crystals on dust particles is partly inhibited in the tropical regions and the southern hemisphere. It may be that the soot particles become dominant in these regions and consume all the available water vapor for ice nucleation, and thus prohibit dust particles to act as INPs. In the northern hemisphere we see an opposite response, however, these changes do not appear to be significant. These changes we observed on the 200 hPa level in Figure 1 are also visible in an annual zonal mean (Figure 2). The increase in ICNC_HET is, thus, not only limited to the 200 hPa level, but extends over the entire upper troposphere and across all latitudes, where heterogeneous nucleation may occur.

Figure 3 shows the change in zonal mean for effective ice crystal radius (Figure 3a), ice water content (IWC, Figure 3b), total ICNC in-cloud (Figure 3c), and the vertical velocity (Figure 3d) for the two simulations SOOT and REF. In the extratropical regions, we note a decrease in effective radius, which is consistent with our understanding of the competition of the nucleation modes. If more INPs are introduced into an environment with the same water vapor content, ice growth can be limited. Thus, additional soot INPs lead to a reduction in radius (Jensen et al., 2016). At the same time, we have an increase in IWC and slight increase in total ICNC. Hence, we have more numerous smaller ice crystals that remain longer in the cloud as they do not sediment as readily as before and lead to a higher IWC (Lohmann et al., 2008). The increase in vertical velocity is due to the release of latent heat upon formation of ice crystals on dust and soot particles. This may lead to a regeneration of S_i and, thus, allowing further formation and growth of ice crystals.

Figure 4 shows the ICNC for the different nucleation modes on 200 hPa as in Figure 1 but for the difference between the SOOT and ACOFF simulations. While we find a strong response in ICNC for the SOOT – REF anomaly, the signal for SOOT – ACOFF is much weaker and not significant. We only see significant changes in the North Atlantic for the ICNC formed on soot particles (Figure 4d). Hence, less soot particles for ice nucleation are available, which is due to completely turning off aircraft emissions in our emissions inventory. This outcome was expected and given that the largest activity of air traffic is in the North Atlantic, the result agrees well with aircraft flight tracks. We do observe an increase in ICNC_HOM over the east coast of North America, which might be due to enhanced vertical motions in the SOOT simulation from a larger amount of latent heat release of ice crystal formation. However, the responses in homogeneously nucleated ICNC, total heterogeneously nucleated ICNC and ICNC nucleated on dust particles does not show any significant changes, and thus remain inconclusive. Bickel et al. (2020) even had to substantially increase the aviation density to produce a significant result (factor of 12). We do not observe any significant zonal changes for the nucleation modes and ice crystal radius, ice water content, total ICNC, and vertical velocity (analogous to Figure 2 and 3; not shown).

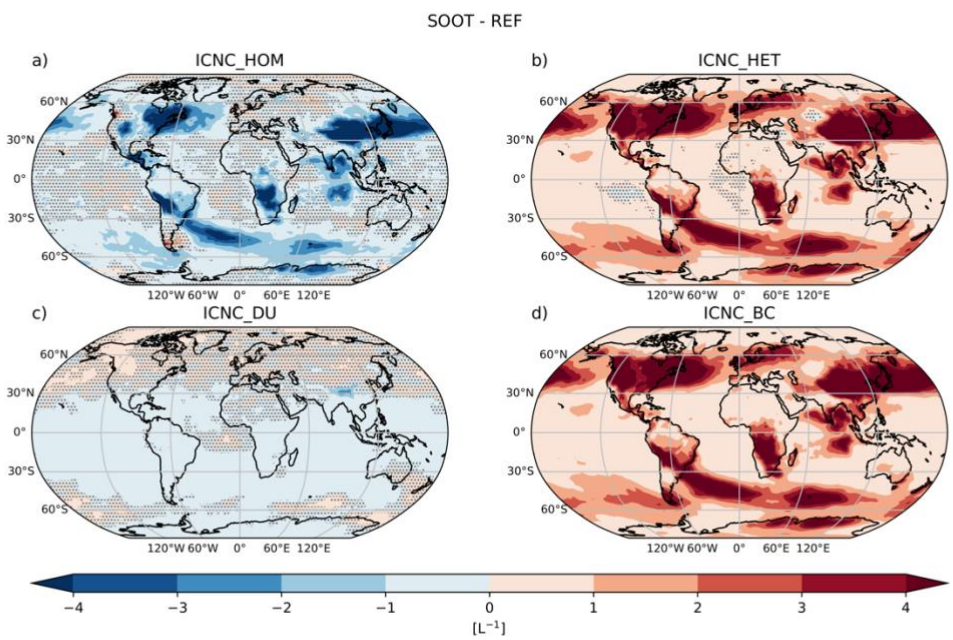

Figure 1: Ice crystal number concentration in L^{-1} on 200 hPa for the difference between the simulations SOOT and REF (SOOT - REF). Shown are homogeneously nucleated ice crystal number concentration (ICNC_HOM, a), heterogeneously nucleated ice crystal number concentration (ICNC_HET, b), heterogeneously nucleated ice crystal number concentration on dust (ICNC_DU, c), and heterogeneously nucleated ice crystal number concentration on black carbon (i.e. soot) (ICNC_BC, d). The grey hatched areas show the data points with insignificant differences at a significance level of 5 %.

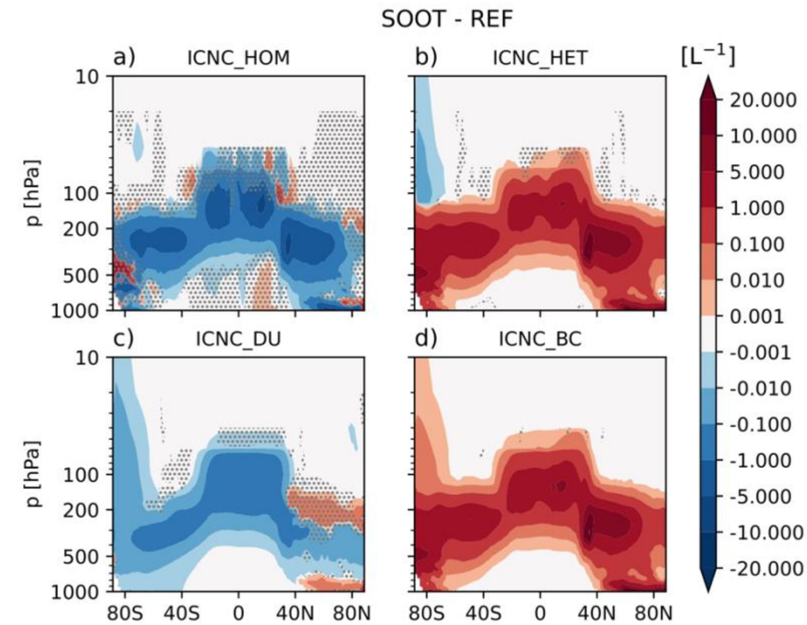

Figure 2: Annual zonal means of ice crystal number concentrations in L^{-1} for the difference between the two simulations SOOT and REF (SOOT - REF). Shown are homogeneously nucleated ice crystal number concentration (ICNC_HOM, a), heterogeneously nucleated ice crystal number concentration (ICNC_HET, b), heterogeneously nucleated ice crystal number concentration on dust (ICNC_DU, c), and heterogeneously nucleated ice crystal number concentration on black carbon (i.e. soot) (ICNC_BC, d). The grey hatched areas show the data points with insignificant differences at a significance level of 5 %.

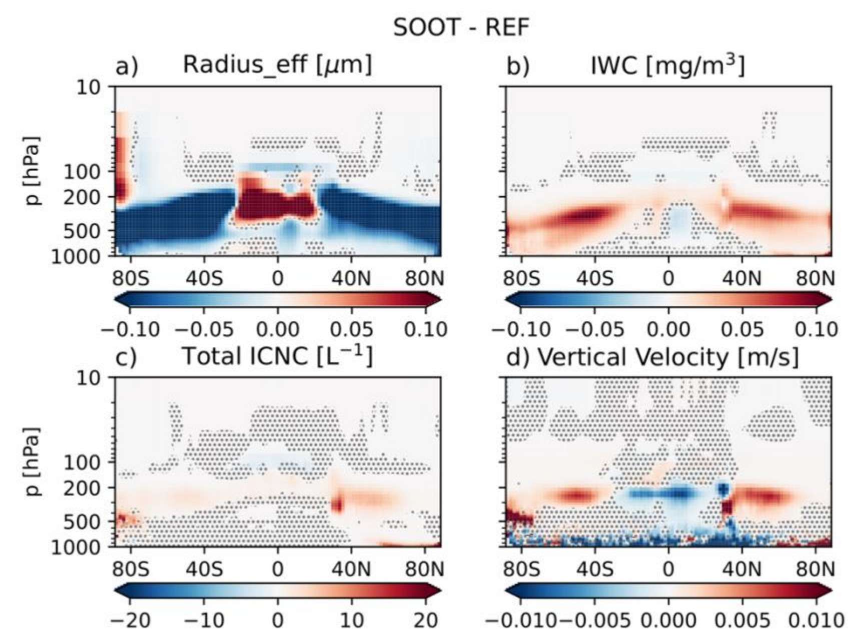

Figure 3: Annual zonal mean of the effective ice crystal radius in μm (a), ice water content in mg/m^3 (b), total in-cloud ICNC in L^{-1} (c), and the vertical velocity in m/s (d) for the differences between the simulations SOOT - REF. The grey hatched areas show the data points with insignificant differences at a significance level of 5 %.

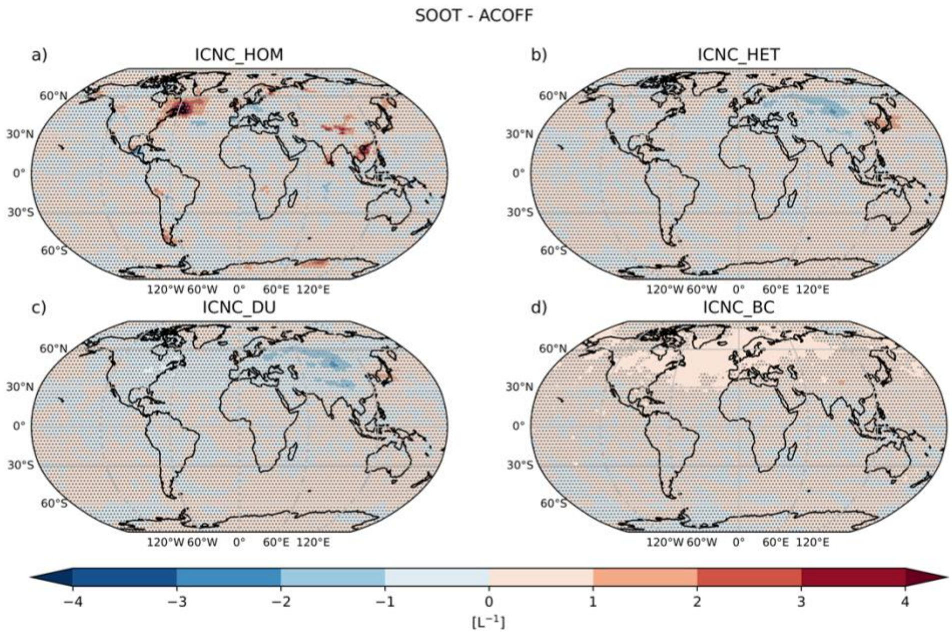

Figure 4: Ice crystal number concentration in L^{-1} on 200 hPa for the difference between the simulations SOOT and ACOFF (SOOT - ACOFF). Shown are homogeneously nucleated ice crystal number concentration (ICNC_HOM, a), heterogeneously nucleated ice crystal number concentration (ICNC_HET, b), heterogeneously nucleated ice crystal number concentration on dust (ICNC_DU, c), and heterogeneously nucleated ice crystal number concentration on black carbon (i.e. soot) (ICNC_BC, d). The grey hatched areas show the data points with insignificant differences at a significance level of 5 %.

5 Conclusions

The aviation industry will continue to grow in the coming decades despite global crises. Introducing aircraft emissions into the atmosphere is known to have an impact on our climate. However, we do not fully understand their true effect due to difficulties in gaining observations of their interactions with ambient air. In the recent years, this topic gained more attention shedding light on the radiative impact of aircraft emissions. Nevertheless, we still face unknowns regarding occurrence-frequency of aircraft-induced clouds and their microphysical properties. While the number of studies investigating this specific topic increased, discrepancies between the findings remain, rendering an unambiguous assessment difficult. This study aims to reduce this ambiguity by employing the global circulation model ECHAM-HAM with a new a state-of-the-art ice microphysics scheme.

Based on a global analysis of soot particles acting as INPs (Mahrt et al., 2018; 2020; Lohmann et al. 2020) in ECHAM-HAM, we quantify the radiative forcing for three different simulations: REF, SOOT, and ACOFF. We take a closer look at the competition between the two nucleation modes acting in cirrus clouds, homogeneous and heterogeneous nucleation, with the latter comprising nucleation on mineral dust and soot particles.

We find that introducing soot particles as INPs (SOOT) leads to an increased radiative forcing on a global scale compared to the reference simulation without ice active soot particles, which is in line with previous studies (e.g., Zhou and Penner, 2014; Kärcher 2018). The additional INPs lead to a shift from homogeneous nucleation to heterogeneous nucleation, with the dominant mode being the ice nucleation on soot particles. This change is most pronounced in regions close to emissions sources for soot. This is mainly due to the additional INPs consuming the available water vapor, and thus prohibiting the high ice supersaturations from occurring, which are required for homogeneous nucleation. In addition to the direct competition between the two nucleation modes, we note decreases in ice crystal number concentration (ICNC) formed on dust particles. This indicates an additional competition between dust and soot particles, that compete for the available water vapor to form ice crystals. These differences are not only noticeable on a specific pressure level, but also across the upper troposphere as shown in the zonal mean (Figure 2). We clearly increase the number of ice crystals formed by heterogeneous nucleation due to the addition of soot particles. At the same time, we observe changes in microphysical properties, such as ice crystal effective radius, ice water content, and total ICNC. Adding INPs leads to a slight increase in ice water content, while the resulting ice crystals are smaller due to enhanced competition for water vapor, which limits their size. This leads to a slight increase in total ICNC.

Removing aircraft emissions (ACOFF) does not result in a significant response as we found for the simulation SOOT. It even appears that aircraft emissions lead to a cooling effect as the net radiative forcing anomaly relative to the SOOT simulation is negative. While this change is significant, it is rather weak being of the order of mW^{-2}. Comparable results were found by McGraw et al. (2020) with a negative forcing an order larger than our result. While we do see less ICNC formed on soot particles for ACOFF, all other changes remain insignificant. This shows that the impact of aircraft emissions on a global scale remain difficult to capture.

In conclusion, this study highlights the impact of soot particles on ice crystal formation on a global scale. To unambiguously quantify the specific impact of aircraft emissions on cirrus cloud formation, a more detailed analysis needs to be conducted. One such examination may comprise a regional analysis of the North Atlantic flight corridor, where we already observed significant changes in ICNC on soot particles. Understanding these effects in an improved manner may result in new measures for reducing the climate impact of aircraft-induced clouds by air traffic management or alternative fuel compositions.

6 References

AIRBUS, 2019. Global market forecast: Cities, airports & aircrafts 2019-2038. Technical Report. Available from: https://www.airbus.com/content/dam/corporate-topics/strategy/global-market-for ecast/GMF-2019-2038-Airbus-Commercial-Aircraft-book.pdf [Accessed April 26, 2022].

AIRBUS, 2021. Airbus Global market forecast 2021-2040. Available from: https://www.airbus.com/sites/g/files/jlcbta136/files/2021-11/Airbus-Global-Market-Forecast-2021-2040.pdf [Accessed April 26, 2022].

Bickel, M., Ponater, M., Bock, L., Burkhardt, U., and Reineke, S., 2020. Estimating the effective radiative forcing of contrail cirrus. Journal of Climate, 33(5), pp.1991–2005. doi: 10.1175/JCLI-D-19-0467.1.

Bock, L. and Burkhardt, U., 2016. Reassessing properties and radiative forcing of contrail cirrus using a climate model. Journal of Geophysical Research: Atmospheres, 121(16), pp.9717–9736. doi: 10.1002 /2016JD025112.

Bond, T.C., Doherty, S.J., Fahey, D.W., Forster, P.M., Berntsen, T., DeAngelo, B.J., Flanner, M.G., Ghan, S., Kärcher, B., Koch, D., Kinne, S., Kondo, Y., Quinn, P.K., Sarofim, M.C., Schultz, M.G., Schulz, M., Venkataraman, C., Zhang, H., Zhang, S., Bellouin, N., Guttikunda, S.K., Hopke, P.K., Jacobson, M.Z., Kaiser, J.W., Klimont, Z., Lohmann, U., Schwarz, J.P., Shindell, D., Storelvmo, T., Warren, S.G., and Zender, C.S., 2013. Bounding the role of black carbon in the climate system: A scientific assessment. Journal of Geophysical Research: Atmospheres, 118(11), pp.5380–5552. doi: 10.1002/jgrd.50171.

Cziczo, D.J., Froyd, K.D., Hoose, C., Jensen, E.J., Diao, M., Zondlo, M.A., Smith, J.B., Twohy, C.H., and Murphy, D.M., 2013. Clarifying the dominant sources and mechanisms of cirrus cloud formation. Science, 340(6138), pp.1320–1324. doi: 10.1126/science.1234145.

Dietlicher, R., Neubauer, D., and Lohmann, U., 2018. Prognostic parameterization of cloud ice with a single category in the aerosol-climate model ECHAM(v6.3.0)-HAM(v2.3). Geoscientific Model Devel- opment, 11(4), pp.1557–1576. doi: 10.5194/gmd-11-1557-2018.

Dietlicher, R., Neubauer, D., and Lohmann, U., 2019. Elucidating ice formation pathways in the aerosol– climate model ECHAM6-HAM2. Atmospheric Chemistry and Physics, 19(14), pp.9061–9080. doi: 10.5194/acp-19-9061-2019.

Grewe, V., Dahlmann, K., Flink, J., Frömming, C., Ghosh, R., Gierens, K., Heller, R., Hendricks, J., Jöckel, P., Kaufmann, S., Kölker, K., Linke, F., Luchkova, T., Lührs, B., Van Manen, J., Matthes, S., Minikin, A., Niklaß, M., Plohr, M., Righi, M., Rosanka, S., Schmitt, A., Schumann, U., Terekhov, I., Unterstrasser, S., Vazquez-Navarro, M., Voigt, C., Wicke, K., Yamashita, H., Zahn, A., and Ziereis, H., 2017. Mitigating the climate impact from aviation: Achievements and results of the DLR WeCare Project. Aerospace, 4(3). doi: 10.3390/aerospace4030034.

Heymsfield, A., Baumgardner, D., DeMott, P., Forster, P., Gierens, K., and Kärcher, B., 2010. Contrail microphysics. Bulletin of the American Meteorological Society, 91(4), pp.465–472. doi: 10.1175/200 9BAMS2839.1.

Jensen, E.J., Ueyama, R., Pfister, L., Bui, T.V., Lawson, R.P., Woods, S., Thornberry, T., Rollins, A.W., Diskin, G.S., DiGangi, J.P., and Avery, M.A., 2016. On the susceptibility of cold tropical cirrus to ice nuclei abundance. Journal of the Atmospheric Sciences, 73(6), pp.2445–2464. doi: 10.1175/JAS- D-15-0274.1.

Kanji, Z.A., Ladino, L.A., Wex, H., Boose, Y., Burkert-Kohn, M., Cziczo, D.J., and Krämer, M., 2017. Overview of ice nucleating particles. Meteorological Monographs, 58, 1.1–1.33. doi: 10.1175/AMSMONO- GRAPHS-D-16-0006.1.

Kärcher, B., Möhler, O., DeMott, P.J., Pechtl, S., and Yu, F., 2007. Insights into the role of soot aerosols in cirrus cloud formation. Atmospheric Chemistry and Physics, 7(16), pp.4203–4227. doi: 10.5194 /acp-7-4203-2007.

Kärcher, B. and Yu, F., 2009. Role of aircraft soot emissions in contrail formation. Geophysical Research Letters, 36(L01804), pp.1–5. doi: 10.1029/2008GL036649.

Kärcher, B., 2018. Formation and radiative forcing of contrail cirrus. Nature Communications, 9(1), p.1824. doi: 10.1038/s41467-018-04068-0.

Lee, D., Fahey, D., Forster, P., Newton, P., Wit, R., Lim, L., Owen, B., and Sausen, R., 2009. Aviation and global climate change in the 21st century. Atmospheric Environment, 43, pp.3520–3537. doi: 10.1016/j.atmosenv.2009.04.024.

Lee, D., Pitari, G., Grewe, V., Gierens, K., Penner, J., Petzold, A., Prather, M., Schumann, U., Bais, A., Berntsen, T., Iachetti, D., Lim, L., and Sausen, R., 2010. Transport impacts on atmosphere and climate: Aviation. Atmospheric Environment, 44(37), pp.4678–4734. doi: 10.1016/j.atmosenv.200 9.06.005.

Lee, D., 2018. International aviation and the paris agreemenet temperature goals. Technical Report. Available from: https://assets.publishing.service.gov.uk/government/uploads/system/uploads/attachment_data/file/813343/international-aviation-paris-agreement.pdf [Accessed May 09, 2022].

Lee, D., Fahey, D., Skowron, A., Allen, M., Burkhardt, U., Chen, Q., Doherty, S., Freeman, S., Forster, P., Fuglestvedt, J., Gettelman, A., De Le ́on, R., Lim, L., Lund, M., Millar, R., Owen, B., Penner, J., Pitari, G., Prather, M., Sausen, R., and Wilcox, L., 2021. The contribution of global aviation to anthropogenic climate forcing for 2000 to 2018. Atmospheric Environment, 244, p.117834. doi: 10.1016/j.atmosenv.2020.117834.

Lohmann, U., Stier, P., Hoose, C., Ferrachat, S., Kloster, S., Roeckner, E., and Zhang, J.: Cloud microphysics and aerosol indirect effects in the global climate model ECHAM5-HAM, Atmos. Chem. Phys., 7, 3425–3446, https://doi.org/10.5194/acp-7-3425-2007, 2007.

Lohmann, U., Lüönd, F., and Mahrt, F., 2016. An introduction to clouds: From the microscale to climate. Cambride University Press. 399pp. doi: 10.1017/CBO9781139087513.

Lohmann, U., Friebel, F., Kanji, Z.A., Mensah, A.A., and Neubauer, D., 2020. Future warming exacer- bated by aged-soot effect on cloud formation. Nature Geosciences, 13, pp.674–680. doi: 10.1038/s4 1561-020-0631-0.

Mahrt, F., Marcolli, C., David, R.O., Grönquist, P., Barthazy Meier, E.J., Lohmann, U., and Kanji, Z.A., 2018. Ice nucleation abilities of soot particles determined with the horizontal ice nucleation chamber. Atmospheric Chemistry and Physics, 18(18), pp.13363–13392. doi: 10.5194/acp-18-13363-2018.

Mahrt, F., Alpert, P.A., Dou, J., Grönquist, P., Arroyo, P.C., Ammann, M., Lohmann, U., and Kanji, Z.A., 2020. Aging induced changes in ice nucleation activity of combustion aerosol as determined by near edge X-ray absorption fine structure (NEXAFS) spectroscopy. Environ. Sci.: Processes Impacts, 22 (4), pp.895–907. doi: 10.1039/C9EM00525K.

Marcolli, C., 2017. Pre-activation of aerosol particles by ice preserved in pores. Atmospheric Chemistry and Physics, 17(3), pp.1595–1622. doi: 10.5194/acp-17-1595-2017.

Mauritsen, T., Bader, J., Becker, T., Behrens, J., Bittner, M., Brokopf, R., Brovkin, V., Claussen, M., Crueger, T., Esch, M., Fast, I., Fiedler, S., Fl ̈aschner, D., Gayler, V., Giorgetta, M., Goll, D.S., Haak, H., Hagemann, S., Hedemann, C., Hohenegger, C., Ilyina, T., Jahns, T., Jimen ́ez-de-la-Cuesta, D., Jungclaus, J., Kleinen, T., Kloster, S., Kracher, D., Kinne, S., Kleberg, D., Lasslop, G., Kornblueh, L., Marotzke, J., Matei, D., Meraner, K., Mikolajewicz, U., Modali, K., M ̈obis, B., Mu ̈ller, W.A., Nabel, J.E.M.S., Nam, C.C.W., Notz, D., Nyawira, S.-S., Paulsen, H., Peters, K., Pincus, R., Pohlmann, H., Pongratz, J., Popp, M., Raddatz, T.J., Rast, S., Redler, R., Reick, C.H., Rohrschneider, T., Schemann, V., Schmidt, H., Schnur, R., Schulzweida, U., Six, K.D., Stein, L., Stemmler, I., Stevens, B., Storch, J.-S. von, Tian, F., Voigt, A., Vrese, P., Wieners, K.-H., Wilkenskjeld, S., Winkler, A., and Roeckner, E., 2019. Developments in the MPI-M Earth System Model version 1.2 (MPI-ESM1.2) and its response to increasing CO2. Journal of Advances in Modeling Earth Systems, 11(4), pp.998–1038. doi: 10.1029/2018MS001400.

McGraw, Z., Storelvmo, T., Samset, B.H., and Stjern, C.W., 2020. Global radiative impacts of black carbon acting as ice nucleating particles. Geophysical Research Letters, 47(20), e2020GL089056. doi: 10.1029/2020GL089056.

Morrison, H. and Milbrandt, J.A., 2015. Parameterization of cloud microphysics based on the prediction of bulk ice particle properties. Part I: Scheme description and idealized tests. Journal of the Atmospheric Sciences, 72(1), pp.287–311. doi: 10.1175/JAS-D-14-0065.1.

Penner, J.E., Zhou, C., Garnier, A., and Mitchell, D.L., 2018. Anthropogenic aerosol indirect effects in cirrus clouds. Journal of Geophysical Research: Atmospheres, 123(20), pp.11652–11677. doi: 10.102 9/2018JD029204.

Staples, M.D., Malina, R., Suresh, P., Hileman, J.I., and Barrett, S.R., 2018. Aviation CO2 emissions reductions from the use of alternative jet fuels. Energy Policy, 114, pp.342–354. doi: 10.1016/j.en pol.2017.12.0079.

Stevens, B., Giorgetta, M., Esch, M., Mauritsen, T., Crueger, T., Rast, S., Salzmann, M., Schmidt, H., Bader, J., Block, K., Brokopf, R., Fast, I., Kinne, S., Kornblueh, L., Lohmann, U., Pincus, R., Reichler, T., and Roeckner, E., 2013. Atmospheric component of the MPI-M Earth System Model: ECHAM6. Journal of Advances in Modeling Earth Systems, 5(2), pp.146–172. doi: 10.1002/jame.20015.

Stier, P., Feichter, J., Kinne, S., Kloster, S., Vignati, E., Wilson, J., Ganzeveld, L., Tegen, I., Werner, M., Balkanski, Y., Schulz, M., Boucher, O., Minikin, A., and Petzold, A., 2005. The aerosol-climate model ECHAM5-HAM. Atmospheric Chemistry and Physics, 5(4), pp.1125–1156. doi: 10.5194/acp -5-1125-2005.

Tegen, I., Neubauer, D., Ferrachat, S., Siegenthaler-Le Drian, C., Bey, I., Schutgens, N., Stier, P., Watson- Parris, D., Stanelle, T., Schmidt, H., Rast, S., Kokkola, H., Schultz, M., Schroeder, S., Daskalakis, N., Barthel, S., Heinold, B., and Lohmann, U., 2019. The global aerosol–climate model ECHAM6.3– HAM2.3 – Part 1: Aerosol evaluation. Geoscientific Model Development, 12(4), pp.1643–1677. doi: 10.5194/gmd-12-1643-2019.

Zhang, K., D., O., Kazil, J., P., S., S., K., Lohmann, U., S., F., Croft, B., Quaas, J., Wan, H., Rast, S., and J, F., 2012. The global aerosol-climate model ECHAM-HAM, version 2: Sensitivity to improvements in process representations. Atmospheric Chemistry and Physics, 12, pp.8911–8949. doi: 10.5194/ac p-12-8911-2012.

Zhou, C. and Penner, J.E., 2014. Aircraft soot indirect effect on large-scale cirrus clouds: Is the indirect forcing by aircraft soot positive or negative? Journal of Geophysical Research: Atmospheres, 119(19), pp.11303–11320. doi: 10.1002/2014JD021914.