

Advancing the Science for Aviation and Climate

# ACACIA

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## 1 Introduction

Indirect aviation aerosol effects and other aviation non-CO<sub>2</sub> effects are still subject to significant knowledge gaps in the scientific understanding of aviation-induced climate impacts. This report summarizes the results of ACACIA Task 3.1. The overall aim of Task 3.1 is to identify gaps in field observations regarding the climate effects of aviation with focus on airborne in situ measurements. Task 3.1 is based on a comprehensive literature review of publications from past campaigns, and includes an assessment of the current state of knowledge. Lacking data with respect to contrail and contrail cirrus properties are identified, and recommendations for future measurements are given.

The report is structured as follows: chapter 2 describes the literature review, and gives an overview of past measurement campaigns. Chapter 3 establishes a gap matrix revealing missing parameters required to improve the current understanding of the impact of aviation on clouds. The assessment of this gap matrix is described in chapter 0. The report closes with a summary of work performed in Task 3.1.

## 2 Literature review on past measurement campaigns

In order to identify scientific gaps in the understanding of indirect aviation aerosol effects, existing data and results from past measurement campaigns were reviewed. The focus of the literature research was confined to in situ aircraft measurements of contrails and contrail cirrus. In total, publications of 20 contrail campaigns between 1971 and 2018, and 7 selected campaigns measuring diverse cloud types and/or mineral dust have been investigated. Note, we focussed on a selection of recent field experiments studying natural clouds and mineral dust which we considered as representative. However, there is more literature available investigating the properties of natural clouds and mineral dust.

The first airborne contrail measurement campaign took place 50 years ago. Since then, airborne contrail field experiments evolved not only due to a growing understanding of physical processes but also because of ongoing instrument development. The contrail life cycle, beginning with a linear contrail and ending with the development of contrail cirrus, involves spatial dispersion from tens of meters to thousands of kilometres on a time scale from seconds to days. Typically, contrails are studied as long as they are linear, since contrail cirrus are often not clearly distinguishable from natural cirrus and therefore are difficult to be sampled (Kärcher, 2018). In situ sampling of slightly-aged contrails (10-20min) tends to be difficult as well since the contrast to its environment is low in most cases making an optical recognition challenging (Schröder et al., 2000). Schumann et al. (2017) has published a comprehensive review of past contrail measurement campaigns in which contrails were measured ranging in age from seconds to about 12 hours. Due to the missing measurements of aged contrails and contrail cirrus, the temporal range covered by past contrail measurements represents only a small part of the total life cycle of contrail cirrus (Burkhardt et al., 2010). Remote sensing instruments are able to cover the missing part of the total life cycle of contrail cirrus up to a certain extend. However, remote sensing cannot provide direct measurements of microphysical contrail properties such as particle concentration, size, and shape. In the early days, the in situ measurements of contrails focused on the size and concentration of ice crystals of contrails and their ice water content (IWC). Through technical developments, measurements of other, especially smaller particle sizes ( $< \sim 10\mu\text{m}$ ), and other parameters such as chemical composition of the plumes became possible with time. The measurement of chemical composition and trace gases enables the differentiation of an aircraft pollution plume from other pollution and clouds.

There are several ways to sample contrails. A very common measurement strategy for contrail and exhaust plume sampling is to directly follow another aircraft with known fuel consumption

like a research aircraft or commercial airplanes. Seven out of the 20 field campaigns including for example SULFUR (Busen and Schumann, 1995), CONCERT (Voigt et al., 2010), and ACCESS-2 (Moore et al., 2017) studied in this literature review conducted the direct chasing technique. Another sampling strategy is the investigation of the exhaust plume/contrail of the research aircraft itself which was done, for example, during CRYSTAL-FACE (Gao et al., 2006), and COSIC (Jones et al., 2012). Contrails have also been sampled during dedicated cirrus cloud campaigns like ML-CIRRUS (Voigt et al., 2017) and Coalesc (Osborne et al., 2014).

Each method of contrail sampling has its advantages and disadvantages and, hence, is best suited for different research questions. Measurements directly behind another research aircraft like ACCESS (Moore et al., 2017) are beneficial for investigations of alternative fuels or different fuel compositions and the examination of young contrails with known age. Collecting the contrails of the research aircraft itself has the advantage that only one aircraft is needed and coordination with aircraft authorities is reduced, compared to the chasing of commercial aircraft. The European Research Infrastructure In-service Aircraft for a Global Observing System IAGOS (Petzold et al., 2017) is a prominent example for a long-term, in situ observation program on commercial aircraft. In the framework of IAGOS, commercial aircraft are equipped with a package of instruments measuring the chemical composition, trace gases, aerosol properties and cloud properties. In addition to a good monitoring of the atmospheric composition, natural clouds and contrails intercepting the flight path of the commercial aircraft are sampled.

The literature showed that there is no coherent approach of distinguishing contrails from natural cirrus clouds or for confirming the existence of contrails from campaign to campaign. Also considered size ranges for reporting ice crystal number concentration vary strongly between studies, therefore making comparisons of reported ice crystal number concentrations difficult. Particularly, when comparing contrail data from different years, it is important to consider the improved methods of contrail detection and characterization. In the early contrail campaigns the detection of contrails had to be done without measurements of pollutants or trace gases, mostly because airborne instrumentation for the measurements of these quantities did not exist, and instrumentation was also not capable to detect high particle concentrations as observed in many young contrails leading to underestimation of ice crystal number concentration due to coincidence. Some campaigns such as FIRE-ARM (Poellot et al., 1999) used a combination of FSSP and condensation nuclei (CN) data instead of trace gases to identify contrails. As one of the first contrail campaigns, SUCCESS (Heymsfield et al., 1998) deployed instruments to measure  $\text{NO}$ ,  $\text{NO}_x$  and  $\text{NO}_y$  by chemiluminescent reaction with ozone in 1996. Supporting data from remote sensing, e.g. lidar or satellite observations, can also be used to observe wide-spread contrail cirrus. This approach was already used in the beginning e.g. during SUCCESS.

A large subset of the reviewed literature of this study was also used by Schumann et al. (2017) to establish a library for contrail observations (COLI). The COLI library builds a collection of microphysical and geometric properties from 33 in situ and remote sensing measurement campaigns between 1971 and 2014. In agreement with the findings of this literature review, Schumann et al. (2017) concludes that measurements from different campaigns show remarkable differences. They express the need for new instrumentations exceeding the current limitation of particles measurements with respect to high concentrations and the requirement of overarching data sets not only containing the measurements of different instruments but also information about the limitations of the instruments recording the data and the aircraft producing the contrail.

### 3 Gap matrix

Future field campaigns and laboratory studies are of highest value, if they target quantities necessary to improve the knowledge and to fill the gaps of understanding. In this task, lacking

observations were identified on the basis of a target parameter list defined in ACACIA WP1 which are required to improve the current understanding of the effects of aircraft soot on cirrus clouds and of aircraft-generated volatile particles on liquid water clouds. Since Task 3.1 focuses on the evaluation of the effects and parameters connected to contrail and contrail cirrus clouds, we selected a subset of 8 target quantities from the target parameter list of WP1 considering in total 23 parameters. Columns 1 and 2 in Table 1 summarize the identified target quantities, and the 23 target parameters, respectively.

**Table 1. Target quantities and parameters required to improve the understanding of aviation effects on cirrus clouds as specified in ACACIA WP 1**

Target quantity	Specific parameter	Level of understanding
Soot microphysical and chemical properties	Mass- and number concentration	medium
	Size distribution	medium
	Mixing state	medium
	Specific information on the amount and morphology of very large soot particles (> 200nm)	low
	Maximum sizes and morphology of aircraft generated soot particles	low
Soot optical properties	Soot optical properties (e.g. mass absorption coefficient, single scattering albedo)	low
Indicator(s) for pollution from aircraft	Aircraft-induced soot	medium
	Other pollutants, e.g. NO <sub>x</sub>	medium
	Isotopologues, e.g. of sulphur compounds	low
	Tracers/markers	medium
	Profiles of optical measure of aircraft aerosol (e.g. extinction profiles) for comparison to satellites	low
Mineral dust	Mass- and number concentration	high
	Size distribution	high
	Mixing state	medium
Cirrus properties	Ice crystal number concentration (ICNC)	medium
	Ice water content (IWC)	medium
	Crystal size	medium
	Optical properties (e.g. depolarization ratio)	low
Temperature, pressure, humidity	Temperature	high
	Pressure	high
	RH <sub>ice</sub> in- and outside clouds	high
Dynamical forcing	Vertical velocity	high
	Other indicators for activity of waves and /or turbulence	low

#### *Aircraft soot properties*

The first target quantity is connected to soot particles. Soot particles and therefore also aircraft-generated soot particles may act as potential ice nucleating particle (INP) (DeMott, 1990). To properly represent their influence in global models, measurements of microphysical and optical properties are necessary. A semi-direct effect caused by soot particles absorbing solar radiation can cause cloud dissipation, and thus cause the atmosphere to be more permeable to solar

radiation (Ackerman et al., 2000). For the investigation of semi-direct soot effects, soot optical properties are critical. In particular, information on large soot particles (> 200nm) is necessary. Mahrt et al. (2018) confirmed the irrelevance of sub 100nm soot particles for the ice crystal formation at  $T < 233$  K. For soot particles exceeding the size of 100nm, they were able to prove the ability of soot particles to contribute to ice formation. However, not only size, but also chemical properties of soot particles determine their ability to form ice (Pruppacher and Klett, 1997; Koehler et al., 2009). In general, soot particles generated by aircraft are smaller than atmospheric background soot particles (Moore et al., 2017), e.g. from biomass burning, but the maximum size of aircraft-generated soot is still an open science question. Up to now, there is no established method for differentiating aircraft-generated soot from other soot sources available. In order to determine the effects and to quantify the impact of soot from air traffic, novel methods for distinguishing aircraft-induced soot from other soot is necessary.

#### *Pollutants as tracers and markers*

Aircraft exhaust consists of a variety of different pollutants, e.g.  $\text{NO}_x$ ,  $\text{SO}_x$ , CO, making in situ characterization of these quantities crucial tracers or markers to identify contrails and aircraft exhaust plumes.

#### *Mineral dust*

As described above, soot may act as potential INP. For mineral dust particles many laboratory studies (Pruppacher and Klett, 1997; DeMott, 2002; Hoose and Möhler, 2012) and field measurement (Sassen et al., 2003; DeMott et al., 2003a, b) campaigns verified a very good ice nucleating ability. However, freezing properties of mineral dust particles differ depending on the chemical composition (Niedermeier et al., 2011). Soot and mineral dust present in the same airmass compete for available condensable water vapour. A better characterization of the aerosol mixing state and, hence, the freezing properties of both soot and mineral dust will lead to an improved understanding of ice nucleation processes.

#### *Cirrus properties*

A clear separation of natural cirrus from contrails, aged contrails, and especially contrail cirrus becomes more difficult with increasing age of the contrail. Different formation processes of natural cirrus (Krämer et al., 2016) and contrails potentially show different cirrus properties (ice crystal number concentration, ice water content, crystal size and optical properties). A precise characterisation of the cirrus properties will help to distinguish natural cirrus from contrails and improve the prediction of climate effects of contrails and contrail cirrus.

#### *Meteorological parameters*

Meteorological parameters like humidity, temperature, pressure and vertical velocity control the freezing process of particles. These parameters are among the standard parameters and are measured in nearly every campaign.

To establish the gap matrix, it was evaluated which observational data are available for each campaign, and missing parameters were identified through the analysis. The gap matrix is visualized in Table 2. Note: for better readability, an excel file of the gap matrix is attached to this report. The 27 campaigns are listed as rows of the matrix and all 23 parameters of the 8 target quantities are specified as single columns. In addition to the acronym of a campaign also the year of the measurements, and the main references are listed. The campaigns are sorted due to their main objective. The first 20 rows list campaigns investigating contrails. The following four rows summarize campaigns focused on various cloud types. The remaining three entries show a selection of mineral dust closure experiments involving in situ and remote sensing observations for mineral dust characterization at various distances from the dust source. Parameters measured during the different campaigns are visualized by green coloured cells. Additional information on the instrumentation observing a specified parameter is written inside the green matrix cells. Red cells indicate that data for a specific parameter are not available for a particular campaign.

**Table 2. Gap Matrix compares the necessary parameters that improve the understanding of air traffic effects in cirrus clouds with already measured parameters from different measurement campaigns. Note: for better readability, an excel file of the gap matrix is attached to this report.**

Target quantity	Soot				Soot optical properties				Indicators for pollution from aircraft				Mineral dust		Cirrus properties			Humidity		Temperature pressure		Dynamical forcing		
Specific parameters	Mass- and number concentration	Size distribution	Mixing State	Specific information on the amount and morphology of very large soot particles (> 200nm)	Maximum sizes and morphology of aircraft generated soot particles	Soot optical properties (e.g. mass absorption coefficient, single scattering albedo)	Aircraft induced soot	Other pollutants (NOx)	Isotopologues, e.g. of sulphur compounds	Tracers / markers	Profiles of optical measure of aircraft aerosol (e.g. extinction profiles) for comparison to satellites	Mass- and number concentration	Size distribution	Mixing state	Ice crystal number concentration (ICNC)	Ice water content (IWC)	Crystal Size	Optical properties (e.g. depolarization ratio)	RHice inside clouds	RHice outside clouds	Temperature	Pressure	Vertical velocity	Other indicators for activity of waves and/or turbulence
<b>Condensation Trails</b>																								
Campaign	Year	Publication																						
PMS	1971	Knollenberg (1972)																						
CSAE	1989	Baumgardner and Cooper (1994)																						
ICE-1989	1989	Raschke et al. (1990)																						
FIRE/ARM	1989																							
SULFUR	1991	Poellot et al. (1999)																						
		Busen and Schumann (1995)																						
		Schumann et al. (1996)																						
		Petzold et al. (1997)																						
		Schröder et al. (1999)																						
AIRCONTRAIL	1994	Schröder et al. (1999)																						
SUCCESS	1996	Heymsfield et al. (1998), Tan et al. (1998)																						
FIRE-SUCCESS	1996	Baumgardner and Gandrud (1998)																						
CRYSTAL-FACE	2002	Gao et al. (2005)																						
PAZI-2	2005	Fevre et al. (2009)																						
SCOUT-O3	2005	Vaughan et al. (2008)																						
CR-AVE	2006	Flores et al.																						
CIRRUS-III	2006	Schäuble et al. (2009)																						
CONCERT	2008	Voigt et al. (2010)																						
COSIC	2009	Jones et al. (2012)																						
CONCERT2011	2011	Kaufmann et al. (2014)																						



The data presented in the gap matrix were taken from the following publications: Knollenberg (1972), Raschke et al. (1990), Poellot et al. (1999), Busen and Schumann (1995), Schumann (1996), Petzold et al. (1997), Schröder et al. (2000), Heymsfield et al. (1998), Tan et al. (1998), Baumgardner and Gandrud (1998), Gao et al. (2006), Febvre et al. (2009), Vaughan et al. (2008), Flores et al. (2006), Schauble et al. (2009), Voigt et al. (2010), Jones et al. (2012), Kaufmann et al. (2014), Moore et al. (2017), Voigt et al. (2017), Weger et al. (2018), Kleine et al. (2018), Bräuer et al. (2021), Voigt et al. (2021), Osborne et al. (2014), Klingebiel et al. (2015), Wendisch et al. (2016), Herenz et al. (2018), Heintzenberg (2009), Weinzierl et al. (2009), Ansmann et al. (2011), Weinzierl et al. (2011), Weinzierl et al. (2017).

#### 4 Assessment of gap matrix

Our analysis of the gap matrix investigates whether the 23 target parameters had been measured during past field experiments, and evaluates the data availability and level of scientific understanding of the target parameters. The scientific understanding of the parameters is rated as high, medium or low based on the following criteria: understanding of parameters is rated as high when a significant amount of data from past measurement campaigns is available and the properties of this parameter are well defined. For medium-rated parameters, measurements are available, but open question still exist for these parameters. Parameters with the rating “low” have not been measured during past campaigns and no data is available. Column three of Table 1 reports the rating of the specified parameter, which was evaluated during the assessment of the gap matrix.

The results of the literature review for soot microphysical and chemical properties, mass- and number concentration, size distribution and mixing state can be rated with a medium level of understanding since measurements are provided from recent campaigns like ACCESS and ML-CIRRUS. However, the amount and morphology of very large soot particles with diameters larger than 200nm as well as the maximum size of aircraft-generated soot particles is very unknown. According to the literature review the separation of aircraft-generated soot from other sources of soot remains open scientific question.

Aircraft-induced soot and other pollutants have been measured and reported during several former campaigns and can be rated with a medium level of understanding. Several of the reviewed campaigns measured gaseous nitrogen ( $\text{NO}$ ,  $\text{NO}_x$ ,  $\text{NO}_y$ ), which are typically used to distinguish contrails from natural cirrus clouds. Other gases such as chemical aged nitrogen ( $\text{HONO}$ ,  $\text{HNO}_3$ ), sulfuric dioxide ( $\text{SO}_2$ ), carbon dioxide ( $\text{CO}_2$ ) and ozone ( $\text{O}_3$ ) are measured to record changes due to chemical reactions and to assess the impact of aviation on the earth’s radiative budget. Isotopologues, like sulphur compounds, which also can be used as an indicator for aircraft pollution have not been measured so far, and hence, only a very low level of understanding exists. In situ measurements of profiles of optical measures of aircraft aerosols (e.g. extinction profiles), which can be used for comparisons to satellite measurements, are not reported in the publications of former campaigns. The literature review reveals also a low level of understanding connected to in situ measurements of soot optical properties like mass absorption coefficient and single scattering albedo.

Mineral dust properties like mass and number concentrations, size distribution and mixing state have been evaluated as medium to high with respect to microphysical and optical mineral dust properties. However, there are still scientific open questions related to the ice nucleation ability of mineral dust and the magnitude of its role as ice nuclei in the atmosphere.

Cirrus properties like the ice crystal number concentration, ice water content and crystal size have been measured in numerous campaigns starting with the first campaign in 1971. Especially with significant improvements of instrumentation, the quality of these measurements enhanced.

However, there are still major open scientific questions connected to these parameters, which leads to a rating of a medium level of understanding. Optical properties, like in situ measurements of depolarization ratio, of cirrus cloud particles are only rarely reported in the literature about past contrail and contrail cirrus measurements. Temperature, pressure, relative humidity, and vertical wind velocity measurements can be considered as standard observations which were available for most of the reviewed publications. For this reason, the scientific understanding of these four parameters was rated as high. However, other indicators for wave activity and/or turbulence is still poor.

## 5 Conclusion

Task 3.1 of the ACACIA project evaluated the current scientific understanding of contrail and contrail cirrus properties by reviewing published results from former in situ aircraft field campaigns. In the framework of this task, literature of contrail and contrail cirrus in situ aircraft field campaigns was comprehensively reviewed. Additionally, a selection of literature on airborne in situ observations of various cloud types and mineral dust was examined. In total, literature from 27 different in situ aircraft field campaigns was considered, and the availability and understanding of 23 relevant in situ parameters necessary for closing knowledge gaps in the scientific understanding of aviation-induced effects on contrails and contrail cirrus clouds as defined in WP 1 was evaluated. The results of the review were presented in a gap matrix including a rating of the scientific understanding for each of the 23 parameters. The rating process considered the number of field campaigns measuring a specific parameter, as well as major open science questions related to a specific parameter. Based on this evaluation, the level of understanding of each parameter was rated high, medium or low.

A high level of understanding was found for temperature, pressure, humidity and vertical velocity observations in contrails and contrail cirrus. Medium to high understanding was found for mass- and number concentration, and size distribution of mineral dust. 10 parameters as specified in Table 1 have a medium level of understanding, either because the specified parameter was only measured during a few campaigns or because major scientific questions are still open. The scientific understanding for seven parameters including, for example, the maximum size and morphology of aircraft generated soot particles, soot optical properties, cirrus optical properties, indicators for activity of waves and /or turbulence and others (see Table 1 for details) was rated as low.

Parameters with medium and low level of scientific understanding are recommended to be investigated in future in situ aircraft field experiments. More precisely, more measurements are needed for microphysical, chemical and optical properties of soot. Especially the amount and morphology of large soot and the maximum size of aircraft-induced soot particles need to be investigated due to the low level of understanding. Investigations of the influence of fuel mixtures and synthetic fuels (e.g. done in ACCESS) on clouds and climate will be of great importance to ensure climate-friendly/climate-neutral means of air transport in the future. Additional examination of the parameters listed under “indicators for pollution from aircraft” in Table 1 are required to close existing knowledge gaps related to contrails and contrail cirrus. Even though a large amount of data on cirrus cloud particle properties is available, high quality data from sophisticated cloud probe instruments is only available for the most recent campaigns. Continued observations with state-of-the-art instruments is necessary to enhance the understanding of processes, relationships and properties of clouds and contrails.

However, even state-of-the-art instruments have limitations, for example with respect to the measurement of high concentrations of ice crystals which govern the precision and feasibility of measurements of certain parameters. As already expressed by Baumgardner et al. (2017) and

Schumann et al. (2017), complete data sets are necessary reporting not only measured data, but also limitations and size ranges of the instruments as well as information about the aircraft. We support their conclusion, that upper measurement limits of particle concentrations due to coincidence or decreasing precision of optical array probe (OAP) instruments for particles smaller than 50 $\mu$ m hinder an improved understanding. Further development of instruments and improvement of measurement technology exceeding today's limitations is fundamental to precisely characterize required parameters for reliable estimations of climate effects connected to contrails and contrail cirrus.

## 6 Appendix

**Table 3. Abbreviations of instruments**

Abbreviation	Full name
SNOOPY / SP2	Single Particle Soot Photometer
PSAP	Particle and Soot Absorption Photometer
UHSAS	Ultra-High Sensitivity Aerosol Spectrometer
CPC	Condensation Particle Counter
NANOSMPS	Scanning Mobility Particle Sizer
SIOUX	Chemiluminescence Detector
ITCIMS	Ion Trap Chemical Ionization Mass Spectrometer
AENEAS	Atmospheric Nitrogen Oxides Measuring System
DOAS	Differential Optical Absorption Spectrometer
SKY-OPC	In-flight Aerosol Spectrometer
IAGOS	Aerosol Particle Counter
OAP	Optical Array Probe
FSSP-100	Forward Scattering Spectrometer Probe
FSSP-300	Forward Scattering Spectrometer Probe
1D-C	PMS 1-dimensional Particle Size Spectrometer
2D-C	PMS 2-dimensional Particle Size Spectrometer
NIXE-CAPS	Novel Ice eXperiment – Cloud and Aerosol Particle Spectrometer
CIP-GS	Cloud Imaging Probe (grey scale)
CAS-DPOL	Cloud and Aerosol Spectrometer with single particle depolarization
PCASP-100	Passive Cavity Aerosol Spectrometer Probe
CAS	Cloud and Aerosol Spectrometer
PN	Polar Nephelometer
CPI	Cloud Particle Imager
AMS	Aerosol Mass Spectrometer
MASP	Multiangle Aerosol Spectrometer Probe
CDP	Cloud Droplet Probe
OPC	Optical Particle Counter
PIP	Precipitation Imaging Probe
PHIPS	Particle Habit Imaging and Polar Scattering Probe
ALABAMA	Aircraft-based Laser Ablation Aerosol Mass Spectrometer
MASS	Mobility Aerosol Sampling System
MAS	Multiwavelength Aerosol Scatterometer
DMPS	Differential Mobility Particle Sizer
REP	Hallet-type Replicator
CVI	Counterflow Virtual Impactor

Abbreviation	Full name
SID-3	Small Ice Detector 3
CR-2	Buck Instruments Model CR-2 Cryogenic Hygrometer
1011B	General Eastern (GE) Chilled Mirror Hygrometer - Model 1011B
AIMS-H2O	Water Vapour Mass Spectrometer
WARAN	Water Vapour Analyzer
WVSS-II	Water Vapour Sensing System
FLASH	Fluorescence Hygrometer
ACH	Frost-point Hygrometer
FISH	Fast In-Situ Stratospheric Hygrometer
JPL	H2O - Instrument from Jet Propulsion Laboratory
HU	H2O - Instrument from Harvard University
SHARC	Sophisticated Hygrometer for Atmospheric Research Tunable Diode Laser
OJSTER	Tunable Diode Laser Spectrometer
DLH	Diode Laser Hygrometer
HAI	Hygrometer for Atmospheric Investigation Tunable Diode Laser
MMS	Meteorological Measurement System
BAHAMAS	Basic HALO Measurement and Sensor System
UCSE	Basic Meteorology
TDC	Meteorology Probe
MTP	Microwave Temperature Profiler
AIMMS-20	Aircraft-Integrated Meteorological Measurement System

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