



BACCHUS

Impact of Biogenic versus Anthropogenic emissions on Clouds and Climate: towards a Holistic UnderStanding

Collaborative Project

SEVENTH FRAMEWORK PROGRAMME ENV.2013.6.1-2

Atmospheric processes, eco-systems and climate change

Grant agreement no: 603445

Deliverable number:	D1.2
Deliverable name:	Preliminary sets of collected aerosol data and re-analyzed and new cloud and cloud-aerosol-interaction observations and corresponding documentation (description of campaigns and observations)
WP number:	1
Delivery date:	Project month 18 (31/05/2015)
Actual date of submission:	02/06/2015
Dissemination level:	PU
Lead beneficiary:	TROPOS
Responsible scientist/administrator:	Berko Sierau (ETHZ), Maria Christina Facchini (CNR-ISAC), Albert Ansmann (TROPOS)
Estimated effort (PM):	134

Contributor(s):	J. Atkinson (ETHZ), S. Decesari, S. Sandrini, F. Pollini (CNR- ISAC), H. Su (MPIC), D. Rosenfeld (HUJI), J. Schmale (PSI), J. Ditas, C. Pöhlker (MPIC), M. Kanakidou (UoC), M. Vrekoussis (CYI), A. Welti (TROPOS), G. Roberts (CNRM-GAME), J. Ovadnevaite (NUIG), M. Rinaldi, (CNR-ISAC), N. Kivekäs (FMI), H. (UoF), K. Carslaw (ULEEDS)
Estimated effort contributor(s) (PM):	S. Sandrini (CNR-ISAC) 5 PM, S. Decesari (CNR-ISAC) 2 PM; F. Pollini (CNR-ISAC) 3 PM; M.C. Facchini (CNR-ISAC) 4 PM, B. Sierau (ETHZ) 2 PM, J. Atkinson (ETHZ) 9 PM, A. Welti (TROPOS) 5 PM, J. Buehl (TROPOS) 9 PM, J. Schrod (UoF) 9 PM, D. Weber (UoF) 4 PM, A. Kube (UoF) 2.2 PM, J. Kossmann (UoF) 3.4 PM, A. Tofaris (UMAN) 1 PM, P. Connolly (UMAN) 19 PM, J. Ditas (MPIC) 6.75 PM, M. Kanakidou (UoC) 4.3 PM, G. Roberts (CNRM-GAME) 9 PM
Internal reviewer:	U. Lohmann (ETHZ), C. Schnadt Poberaj (ETHZ)

SUMMARY OF RESULTS

Work package 1 (WP1) covers the aerosol and cloud observations performed within the framework of BACCHUS. Focus is on the characterization of aerosol particles to serve as cloud condensation nuclei (CCN) and ice nuclei (IN) based on past and published in situ measurements, as well as those from new observations (e.g., BACCHUS Cyprus 2015 campaign). The goal is to establish a global database or database infrastructure (which does not necessarily contain the data physically) on CCN and IN properties for different natural and anthropogenic aerosol types and mixtures around the world. WP1 also deals with vertically resolved field observations of aerosols and clouds in key climate regions from aerosol/cloud lidars and cloud radars, organized in networks (e.g., ACTRIS, CLOUDNET) or arranged as super sites within large field campaigns (e.g., BAECC), and by including aircraft observations (e.g., ACCACIA, ACRIDICON). Satellite remote sensing with very high horizontal resolution (e.g., the Visible Infrared Imager Radiometer Suite (VIIRS), onboard NASA's Earth-observing satellite NPP) is also used to study aerosol-cloud interaction in key regions of the global climate. In the first step, closure studies, validation efforts, and quality assurance work and re-analysis of past observations are performed and have to be completed by the end of November 2015 (milestone 1).

WP1 is organized in six tasks (Tasks 1.1-1.6):

- Task 1.1: Definition of the natural background aerosol and measurement framework for the IN database
- Task 1.2: Compilation of geographic differences in IN/CCN
- Task 1.3: Quantification of natural (biological and dust) and anthropogenic contributions to IN
- Task 1.4: Quantification of biogenic and anthropogenic contribution to organic aerosol and their ability to act as CCN
- Task 1.5: Obtaining the vertical structure of aerosols and clouds
- Task 1.6: Establishment of a harmonized dataset of aerosol, CCN/IN and cloud microphysical properties

1. Contribution from Task 1.1 "Definition of the natural background aerosol and measurement framework for the IN database" (ETHZ, UOC, CNR-ISAC, NUIG, MPI-C, TROPOS) and task 1.2 "Compilation of geographic differences in IN/CCN" (ETHZ, UHEL, FMI, TROPOS, UMAN, CYI, CNR-ISAC, NUIG, PSI, ULEEDS, UOF)

The design and construction of the "Ice Nucleation (IN)" database infrastructure was completed. From now on, it will be used to collect data on ice nucleating particles (INP). It includes an extensive framework for auxiliary information (e.g., description of campaigns, measurement location, time, particle chemical composition, instrumentation, etc.). The database utilises a web search portal written in PHP (see Figure 1) and data files will be in netCDF format. The database is now open for submissions and will soon be open for access to registered users. Submissions have been solicited from the global community and the first datasets have started to be received. The BACCHUS idea behind the development of the database and its infrastructure has been presented at the 14th Conference on Cloud Physics in Boston, July 2014, as was discussed and agreed on by experts of the IN community. Still, the infrastructure is built to be adaptable for future improvements and extensions.

	► Background			Searc	h the dat	abase Change passwor	d Activity logs Log out (Contact the dat	tabase admini:	strator			
Consortium Consorti	▶ Work packages Name	Туре	Start	End	Institute	PI/data owner	URL	T min/max	RH min/max	Location	Altitude	Downloads	Link
Events 21:002/2000 Subjects Subjects <t< th=""><th>CLACE2013</th><th>Conc</th><th>20130124</th><th>20130227</th><th>ETH Zurich</th><th>γvonne.boose@env.ethz.ch Zamin Kanji,</th><th></th><th>241.00/247.00 K</th><th></th><th>Jungfraujoch</th><th>Surface</th><th>0</th><th></th></t<>	CLACE2013	Conc	20130124	20130227	ETH Zurich	γvonne.boose@env.ethz.ch Zamin Kanji,		241.00/247.00 K		Jungfraujoch	Surface	0	
Mark Laboration CLACE2D21 Conc 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112 2020112	CLACE2014	Conc	20140124	20140216		yvonne.boose@env.ethz.ch Zamin Kanji,		241.00/247.00 K	% ice 92.00/103.00	Jungfraujoch	Surface	0	
Web 0022 Web 0025 Apple All search fields are optional. They instance of the tables are optional. They instance of t	CLACE2012	Conc	20120112	20120127		Zamin Kanji,		241.00 K		Jungfraujoch	Surface	0	
Project Name Meeter Project Institute Project Leader Project Lead	News - 3 March 2015: ChirMB-BACCHUS campaign started News - 06 February 2015: Press release Meteo France News - 13 January 2015:	Welcome to the BACCHUS INDB search page. All search fields are optional. For selection boxes, multiple options can be selected by holding control and clicking with the mouse. If 'Any' is selected at the same time as other options, 'Any' will be used and those other selections ignored. Find observations of ice nuclei From until Compositions From Until From Until From Until From Until From Until From Until From Until From Until From Until From Until From Until From Until From Until From Until From Until From Until From Until From Until From Until From Unti											
Any Any Short term Ship Long term Sip p Sempling conditions Instrument name N Mode Collection method Deposition Priter Diposition No Diposition No Diposition No Diposition No Diposition	First BACCHUS Annual Meeting	me		Pro	oject Institu	ute Proje		Project DOI		Project U	RL		
Any Any Short term Ship Long term Sip p Sempling conditions Instrument name N Mode Collection method Deposition Priter Diposition No Diposition No Diposition No Diposition No Diposition					-								
Airborne Airborne Winter Long term Siging Summer Sampling conditions Instrument name IN Mode Collection method p/T conditions Any Any Any Any Anbient Deposition Orgonalisation Particle cutoff Ambient aerosol also observed? Inlet dried Inlet info Lower: Any Yes No No Upper: No No No No Location Alticle IN Conc. value Temp range RH range Minc Matrument, Mac Observations; Minc Mac Minc mac Minc Mac Observations; Minc mac Max Minc Mac Minc Mac Minc Mac Minc Minc Mac Minc Mac Minc Minc <	Project Ler	ngth		Pr	oject Envir	onment	P	latform		Project Sea	ason		
Any Any Deposition Filter Condensation Prive Impactor Dry Impactor Start Particle cutoff Ambient serosol also observed? Inlet dried Inlet info Lower: Any Verify Yes No No Location Altude Inic: Max: Min: Max: Observations: Min: Min: Max: Observations: Min: Min: Max: Observations: Min: Min: Max: Incervate and south as Prise use decimal degrees and entervest and south as Please use decimal degrees and entervest and south as Please use select tox or text boxes,	Short ter						, :	Airborne Ship		Winter Spring			
Particle cutoff Ambient aerosol also observed? Inlet dried Inlet info Lower: Anny Yes Anny Upper: No No No Location Atitude IN Conc. value Temp range RH range Lat: Select region: Instrument: Instrument: Instrument: Instrument: Minc maxc Surface Observations: Observations: Observations: Observations: Observations: Minc Maxc Minc Minc Minc Please use decimal degrees and enter west and south as OR Min: Max: IN Composition Bins RH Liq; Instrument: Minc	Sampling c	Sampling conditions Instrument name					Any Deposition Condensation			ıt			
Lower: Any Yes Upper: Yes No Location Atitude IN Conc. value Temp range RH range Lat: Select region: Instrument: Instrument: RH ice: Min: maxc Any Min: Maxc Observations: Uor Crosses the 180 longnitude Boundary Layer Min: Maxc Min: Maxc Inc Please use decimal degrees and enter west and south as OR Min: Max: Min: <		Search											
Lat:	Lower:	Lower:			Any (es	osol also observed?	1	Any Yes		_			
point location use minimums not both. Ubservations:	Lat: Minc Lon: minc Crosse line. Please use enter wes negative v point vec	ma esthe decin t and values	x: -180 longnit mal degree south as . To search	sele An Su Bo Lo sand OR s an Plea	ect region: y urface bundary L wer Trop Min: hPa ase use sele	ayer Max: NIC common National	ment: Max: rvations: Max:	Instrument: Min: M Observations: Min: M		RH Ice: Instrume Min: Observat Min: RH Liq: Instrume Min: Observat	nt: Max: Max: Max: nt: Max: ions:		
Search							Search						

Fig. 1: Web search portal of the IN database

Name	ne Country		Main task	Misc data	Institution
Antarctic Summer Campaign	Antarctica	06-08 2013/14	CCN	> +	TROPOS
RACEPAC	Canada	02-04 2014	CCN	>>	TROPOS
Mace Head	Ireland	2009-2014	CCN	> +	NUIG
PEGASO	Southern Ocean	01/02 2014	CCN	>	NUIG
ACRIDICON- CHUVA	Brazil	09 2014	CCN	3 +	MPIC
ATTO site/ACRIDION	Brazil	2014	CCN	>	MPIC
ACTRIS re- analysis*	9 stations (see 2.1)	2012 to 2014	CCN	3→	PSI
PuCE	Finland	2006-2015	CCN-like	>	FMI
Pallas	Finland	2005-2015	CCN-like		FMI
San Pietro Capo Fiume	Italy	02 2014	CCN	3+	CNR-ISAC
ChArMEx/ENVI- Med/BACCHUS	Cyprus	03/04 2015	IN, CCN	>	CYI, ETHZ, CNRS-CNRM
CLACE 2014	Switzerland	01 2014	IN, CCN	3+	ETHZ, PSI, UOF
JFJ-GAW I	Switzerland	08 2014	IN	>	ETHZ
JFJ-GAW II	Switzerland	01/02 2015	IN		ETHZ
JFJ GAW III	Switzerland	05/06 2015	IN		ETHZ
CALIMA I	Tenerife, Spain	07/08 2013	IN, CCN	>	ETHZ
CALIMA II	Tenerife, Spain	07/08 2014	IN, CCN	> +	ETHZ
San Pietro Capo Fiume	Italy	02/05 2014	IN	>	CNR-ISAC
Amazonian Tall Tower Observatory	Brasil	since 09 2014	IN	9 1	UOF
Obs. de la Martinique	France	since 09 2014	IN	>	UOF
Zeppelin Observatory	Norway	since 09 2014	IN	3+	UOF
Taunus Observatory	Germany	since 09 2014	IN	3 +	UOF

Tab. 1: New and reanalyzed INP and CCN data collected during the BACCHUS timeframe until M18 (*PSI contributed with a re-analysis of ACTRIS CCN data to target specific task 1.2 questions, i.e. the geographic differences in CCN).

Various groups contributed to D1.2 of BACCHUS by collecting aerosol and cloud microphysical data, and information on Cloud Condensation Nuclei (CCN) and Ice Nucleating Particle (INP) properties as part of aerosol-cloud interaction observations. Herein, aerosol data was rather obtained from "auxiliary measurements" and is not emphasized in this report of task 1.1 and 1.2.

Task 1.1 and 1.2 contributed to D1.2 with a substantial amount of new INP and new and reanalysed CCN data as listed in Tab. 1. The INP data will be integrated into the IN database framework, the CCN data into the ACTRIS framework which is then transferred into the GASSP project, as agreed on during the BACCHUS 2015 annual meeting.

All datasets – and especially those of INP and CCN – will be evaluated and interpreted in a way to understand geographical differences in IN, CCN, and their properties. In combination with the contributions of tasks 1.3 - 1.6 to D1.2, and the study of *UNIVLEEDS* towards the understanding of the natural background aerosol, the basis is laid out to analyze aerosol-cloud interactions based on in-situ and remote sensing observations. Major finding towards this goal were reported by *PSI*, *MPI-C*, *TROPOS*, and *UHEL*, *FMI*, *UMAN*, *ETHZ*, *UOF*, *CNRS-CNRM*, *CYI*, *CNR-ISAC*, *NUIG* and some of them are summarized in the following (general contributions are listed in Table 1).

9 data sets have been collected so far by *PSI*. Those include mainly data from ACTRIS stations (Birkenes, Norway; Cabauw, Netherlands; Hyytiälä, Finland; Jungfraujoch, Switzerland; Mace Head, Ireland; Melpitz, Germany; Puy de Dôme, France), as well as from Finokalia, Greece, and Barrow, Alaska. Further data sets from Vavihill, Sweden; London, UK; Paris, France, and the Amazonian rain forest, Brazil, are likely to be incorporated. Most data sets cover the full, or episodes of, the 3-year period from 2012 to 2014, except for the Barrow data set which is from 2007/2008. The data include CCN number concentrations of mono- (4 stations) and poly-disperse (all other) measurements at several super saturations between 0.1 and 1.45 %. In addition, size distribution data is available for all locations. Data on the chemical composition of submicron aerosol is provided for 7 locations.

The current analyses on this data set include the geographical and seasonal comparison of CCN number concentrations at several super saturations, CCN activation ratios, the critical diameters at several supersaturations, the characteristics of the size distributions and aerosol chemical composition, as well as the hygroscopicity parameter kappa. In addition, chemical closure studies have been performed for 4 sites. Figure 2 shows preliminary results. There are clear differences in the annual cycles of the CCN concentrations at SS = 0.2 % at the six stations, shown in panel a. MEL and CAB show concentration maxima in spring and summer, while HYY exhibits maxima in winter and summer. The two mountain stations JFJ and PdD show maxima in summer and the Arctic station BAR features the typical high latitude annual cycle with a maximum during winter (Arctic Haze) and a minimum in summer. Panel b presents the fitted activation ratio curves for several stations based on the particle number concentrations between 10.2 and 800 nm mobility diameter. A critical parameter for the activation of particles is the aerosol size distribution which is represented by the geometric mean diameter in panel c. Again, there are clear differences in the sizes and annual cycles. While at some stations the variability in the size distribution reflects the annual cycle of CCN concentrations, e.g. JFJ, BAR, this is not the case for others, e.g. MEL, CAB. This suggests that at some locations the size distribution is the most important driver of CCN concentrations, and at others additional parameters such as the chemical composition play a role, especially when particles are smaller. The annual cycle of the hygroscopicity parameter kappa, panel d, is similar for all shown stations with the most and least pronounced amplitude in HYY and CAB,



Figure 2: a) Annual cycle of CCN number concentrations at a supersaturation of 2 %; b) fitted activation ratio curves (y = a*ln(SS%)+b); c) annual cycle of the submicron aerosol geometric mean diameter; d) annual cycle of kappa. BAR = Barrow, CAB = Cabauw, HYY = Hyytiälä, JFJ = Jungfraujoch; MEL = Melpitz, PdD = Puy de Dôme.

respectively. Not shown are critical diameters. For the stations in panel a, at $SS = 1 \% D_{crit}$ ranges between 30 and 40 nm, for SS = 0.5 % between 45 and 60, and SS = 0.2 % between 85 and 120 nm.

The next steps include further data quality checks in collaboration with the data providers, as well as a more thorough investigation of different environmental conditions (e.g., planetary boundary layer vs free troposphere, marine vs boreal influence, local pollution vs long-range transport) and their influence on CCN number concentrations. Emphasis will be put on the role of particle size distributions, and closure studies based on the chemical composition of the aerosol population.

High latitude CCN measurements have been performed in the Arctic and Antarctic regions within the framework of two different campaigns. The Radiation-Aerosol-Cloud Experiment in the Arctic Circle (RACEPAC) took place in North Canada (Inuvik and Tuktoyaktuk) during spring 2014. Measurements were also made at the Princess Elisabeth Antarctica Research Station during the Antarctic summer 2013/2014 (cf. Fig. 3). In both cases the total CCN number concentration was measured and used to estimate the hygroscopicity parameter κ activation behavior of sub-micron arctic aerosol particles.

In the northern sub-Arctic region the total particle background concentration varied between 100 and 500 particles per cm³. κ -values for the sub-arctic aerosol in North Canada were estimated to be between 0.1 and 0.5. This large variability can be explained by the varying origin of the air masses. The total particle concentration in the Antarctic is comparable to the background concentrations measured in Canada. Furthermore, single particle nucleation events generating up to 4000 particles per cm³ were detected. Since the CCN number concentration for the highest measured supersaturation of 0.7% (not shown in Fig.

3) is not affected during these events, a large particle number of diameter below 35 nm must be present. The κ -values of the Antarctic were found to have a mean of 0.76 and show lower variation. The κ -values of both data sets were found to agree well with global simulations.



Figure 3: 50-days period of CCN measurements at Princess Elisabeth Antarctica Research Station. First row shows total particle number concentration (black) and the concentration of cloud condensation nuclei at a supersaturation of 0.1% and 0.3%. Calculated κ -values are reported in the second row.

A first "BACCHUS field experiment" was carried out: the joint ChArMEx/ENVI-Med/BACCHUS campaign that took place in March 2015 at the remote site of Agia Marina Xyliatou in Cyprus. Herein, a large set of instruments has been deployed in parallel to IN (ETHZ) and CCN (CNRS-CNRM) observations by CYI. All instruments have been working properly, sampling and characterizing air masses from various origins ranging from African/Saudi Arabia dust to European and Middle East pollution. Nucleation has been investigated by instruments operated by CNRS-LaMP. Size distribution of CN and CCN were performed by CNRS-LaMP and CYI. Optical properties of PM10 (light scattering coeff., extinction and absorption) were performed by CNRS-LSCE, CNRS-LA, and CYI. The chemical composition of non-refractory PM1 was performed with a monitoring mass spectrometer and was completed by 24h filter sampling in PM1, PM2.5, PM10 for the chemical determination of ions, carbon (EC, OC), and trace metals. Extra filters (covering the daytime IN measurements performed by ETHZ) will be analyzed for metals and tracers of fungal spores (Mannitol, Arabitol). PM2.5 and PM10 were monitored continuously by TEOM. All the data of this campaign will be posted on the French MISTRALS website (http://mistrals.sedoo.fr/) and the ACTRIS database (http://actris.nilu.no/ - see below).

Optical/chemical mass closure studies and source apportionment (from on-line PM1 and off-line PM10) will be performed to identify which are the major sources contributing to PM and Optical properties of aerosols and will be compared with remote sensing observations (supphotometer, lidar) (CYI).

In parallel with the intensive BACCHUS field campaign at Cyprus, intensive surface observations were performed at Finokalia atmospheric monitoring station, on Crete. These include 24h filter collection for chemical composition analysis of PM10 (ions, OC/EC, trace metals), continuous aerosol chemical composition measurements, size distribution, CCN (instrument from Georgia Tech. Prof. Nenes),

nephelometer and aethalometer observations, In addition, concentrations of selected volatile organic compounds have been measured. Air masses from various origins ranging from air masses reaching the site after being transported over the West Mediterranean Sea to those affected by African dust, European / Middle East pollution have been sampled. These observations will be used to derive optical and chemical mass closure. Data analysis will also enable source apportionment to identify which are the major sources contributing to PM and optical properties of aerosols. The results will also be compared with remote sensing observations (sunphotometer).

The data analysis of the ChArMEx/ENVI-Med/BACCHUS campaign will enable the characterization/definition of background levels/properties of natural aerosol in the region. The data aims to support the GASSP database as described in the paragraph above (UOC). The cloud-related part of campaign is described in more detail later in this report. However, for the first time in-situ ground-based and airborne, as well as remote sensing and satellite observations of IN and aerosol-cloud interactions were carried out simultaneously by members of the BACCHUS consortium. Data analysis is underway. A follow-up campaign is planned for April 2016.

The *UOF* contribution to the experimental field data of ice nucleating particles (INP) builds on the sampling of aerosol particles at remote sites and lab analysis of deposition/condensation mode INP therein by the static diffusion chamber FRIDGE. They have manufactured four sampling devices for automated collection of aerosol samples. Collaborations with institutions in Norway, Brazil, and France were established for the sampling of aerosol in the Arctic, Amazon and Caribbean. Contracts were signed. Samplers were deployed at the sites and sampling commenced in September 2014. An optical particle sizer (TSI 3330) was purchased from the project and set up at Martinique. 160 aerosol samples were collected, shipped to Frankfurt, and analyzed in the lab. The preliminary statistical features of the INP number concentration are given in Figure 4. The sampling routine was interrupted during the months 01-

Site	Environment	Country	City nearby	Coordinates	Altitude	Local	
(Environment)						Partners	
Amazonian	Tropical	Brazil	Manaus	2°8'48"S	133	MPIC, Prof.	
Tall Tower	Rainforest			59°0'18"W		Artaxo, U'	
Observatory						S. Paulo	
Obs. de la	Subtropical	France	Font-Saint-	14°44'05"N	487	Inst. De	
Martinique	Marine		Denis	61°08'48"W		Physique du	
						Globe Paris	
Zeppelin	Arctic	Norway	Ny Alesund	78°54'29"N	475	Norwegian	
Observatory	Marine			11°52'53"E		Polar Inst.	
Taunus	Central	Germany	Frankfurt	50° 13' N,	825	U' of	
Observatory	Europe			8° 27' E		Frankfurt	

Table 2: INP sampling sites



Figure 4: INP Concentration at sampling sites Amazon (AZ, n=29), Martinique (MQ, n=33) and Svalbard (SB, n=28)

04 of 2015 for a reevaluation of the sampling procedure. In May 2015 sampling will be continued. The experimental methods of the project were evaluated in blind tests and against the methods of other major groups who contributed to ice nucleus measurements during the 5^{th} International Ice Nucleation Workshop at Karlsruhe in March 2015. In March 2015 we also contributed to the Mediterranean field campaign at Cyprus by INP measurements at ground level.

Additional ground based INP observational campaigns were conducted *(ETHZ, CYI, CNR-ISAC)*. ChArMEx/ENVI-Med/BACCHUS that was completed in March 2015 in Cyprus, with observations taken on 19 days. The applied instrument was HINC (Horizontal Ice Nucleation Chamber). IN measurements were performed at -30°C, 105 % RH with respect to water for the investigation of condensation freezing, and 93 % RH wrt water (124 % wrt ice) for the investigation of deposition freezing (set points discussed and agreed on during the annual BACCHUS meeting). Planning for a second INP observation campaign in August 2015 in Ireland is under way.

In July 2014, a second campaign was carried out on Tenerife, Spain, called CALIMA II (Cloud Affecting particLes In Mineral dust from the sAhara), the predecessor of CALIMA I of summer 2013. CALIMA aimed at quantifying the effectiveness of mineral dust particles close to a main global dust source to act as ice nuclei. CALIMA was conducted at the Izaña Atmospheric Observatory, located at 2400 m a.s.l. on Tenerife, Canary Islands, west of the African shore. During the measurement period the station was located in the Saharan Air Layer (SAL) most of the time. If not exposed to a dust event, the location is considered typical for being in the free troposphere especially during night times. The campaign lasted for four weeks. Ice nuclei concentrations were measured with the Portable Ice Nucleus Counter (PINC) at temperatures between 232 K and 253 K at relative humidities above and below water saturation. A cloud condensation nuclei (CCN) counter was continuously measuring the polydisperse CCN number concentration at 10 different supersaturations between 0.1 - 1 %. Filter samples were taken to investigate the chemical composition of the dust particles.

Two more IN-related experimental campaigns were carried at a rural site (San Pietro Capo Fiume, 30 km from Bologna) in the period 07-21 February 2014 (winter) and 19-30 May 2014 (spring). In parallel with the second sampling period, filters were sampled on top of the Monte Cimone in the Northern Apennines

at the "O. Vittori" Climate Observatory, to compare ice nuclei concentration simultaneously sampled in the Po Valley and at the top of the Appennine mountains.

The finalized data sets from Cyprus and Tenerife will be the first to be entirely integrated into the BACCHUS IN database.

3. Contribution from Task 1.3 "Quantification of natural (biological and dust) and anthropogenic contributions to IN" (MPI-C, ETHZ, CNR-ISAC, UHEL, NUIG, UOC, CYI, FMI, UOF)

On-line measurements of bioaerosol by UVAPS (UltraViolet Aerosol Particle Sizer) and WIBS (Wideband Integrated Bioaerosol Sensor) – two instruments to detect and measure the size of bioaerosols) all over the world are summarized and characterized through this task. We have now established a database to collect measurement data via the MPI-C FTP server (ftp.mpic.de). Currently, bioaerosol information from the following campaigns have been collected: Mainz 2008; Amazon 2008; Hyytiala, Finland 2009; Killarney National Park, Ireland 2010; Karlsruhe, Germany I 2010; MEGAPOLI Study (Paris, France) II 2010; BEACHON Study (Colorado, USA) 2011; Karlsruhe, Germany II 2011; Mainz II 2011; Beijing 2013; Xiamen 2013; Nanjing 2013; ATTO 2014-15. For each campaign, the number concentration and size distribution of total aerosols and bioaerosols are provided. The BACCHUS Bioaerosol Database Protocol is developed based on the general BACCHUS Database Protocol to regulate the data exchange and is briefed as follows: (1) Data will be stored at MPIC, data submission will be coordinated by Alex Huffman from University of Denver and Zhibin Wang from Max Planck Institute for Chemistry. (2) Data providers reserve all rights of their data. (3) Data access is limited to the BACCHUS project and external users, who have signed the data protocol. The data providers must approve any transfer/usage of data stored in the database on a case-by-case basis. Especially, when data are used in publications, joint authorship must be offered and intellectual input must be discussed with the data owner and scientists who have contributed substantially to data processing (such as synthesis or merging of datasets in addition to that carried out by the data provider).

Concurrent INP and bioaerosol measurements will be linked via the BACCHUS IN database framework to have direct information and access to both sets of information. Concurrent CCN and bioaerosol measurements will be linked via the ACTRIS network and thus integrated into GASSP at a later stage as well.

4. Contribution from Task 1.4 "Quantification of the biogenic and anthropogenic contribution to organic aerosol and their ability to act as CCN" (CNR-ISAC, PSI)

The existing long-term dataset analysis of aerosol chemical and CCN measurements provided by the ACTRIS network is being analyzed (also object of study in Task 2.2 - CCN closure experiments). Specifically, seven background stations in Europe are providing the most comprehensive records of CCN and aerosol chemistry (ACSM) measurements. Continuous size-distribution measurements are also available from these sites.



Figure 5: List of ACTRIS stations with long-term simultaneous CCN and ACSM observations. The measurement coverage of ACSM is described in detail at the website: <u>http://www.psi.ch/acsm-stations/overview-full-period</u>.

The analysis of the ACTRIS records is planned in two steps:

- 1. Linking the CCN concentration frequency distributions and supersaturation (SS) spectra to generic proxies of anthropogenic (e.g., black carbon) and natural aerosol components.
- 2. Linking CCN to organic fractions (e.g., biomass burning organic aerosols, etc.).

In this deliverable the analysis for "step 1", based on the datasets acquired at the Jungfraujoch station between 2012 and 2013, is provided. All data were averaged to 1 hour resolution. Polydisperse CCN measurements were available for SS = 0.10, 0.15, 0.20, 0.25, 0.30, 0.35, 0.40, 0.5, 0.7 and 1.0 %. Black carbon (BC) concentrations were derived from aethalometer observations, while chemical species measurements were performed using an ACSM. A total of 6070 hourly observations with valid data for aethalometer and CCN collected between July 2012 and July 2013 have been used. A slightly smaller number (6000) of observations with valid data for CCN and ACSM was extracted for the same time period for this analysis.

Bidimensional frequency distributions for the concentrations of CCN and selected aerosol chemical compounds have been computed for each SS level. Since aerosol concentrations at the Jungfraujoch experience strong variations during the year, a log scale for concentrations was adopted. Examples of the results are shown below for two supersaturation levels:



Figure 6: 2D frequency distributions of CCN (cm⁻³ STP, at 0.4% and 1.0% supersaturation) and BC, organic matter and sulphate concentrations at the Jungfraujoch ACTRIS site. Colors measure the occurrence of observations (hourly data between July 2012 and July 2013) in each bin. All data were provided by PSI (CCN: Julia Schmale; ACSM: Roman Froehlich; BC: Nicolas Bukowiecki).

Clearly, there is a tendency for CCN occurrence at high concentrations when BC and organic matter (OM) levels are also high, while the variations in sulphate concentrations are weakly related to changes in CCN levels. In some cases, a bimodal distribution can be noticed, especially when CCN concentrations are plotted against BC or OM. A complete understanding of the shape of the observed frequency distributions will need to include the analysis of the aerosol size distribution datasets and of the meteorological factors which govern the climatology of the aerosol at the site, i.e. especially the boundary layer air intrusion carrying elevated concentrations of pollutants.

The statistics for CCN concentrations at moderately low (0.4%) and high (1%) supersaturations in three "regimes", based on the BC frequency distribution is reported below. These data can contribute to the definition of the aerosol CCN properties under "polluted" vs. "relatively clean" or "transition" conditions in the European background atmosphere.



Figure 7: Frequency distributions of CCN concentrations (at SS=0.4% and SS=1.0%) for three "pollution regimes" at the Jungfraujoch station. Regimes correspond to ranges in BC concentrations measured at the site. The resulting median concentrations are 42, 101, and 377 cm⁻³ (for the "clean", "intermediate" and "polluted" regimes at 0.4% SS) and 91, 169 and 497 cm⁻³ (for the "clean", "intermediate" and "polluted" regimes at 1% SS).

5. Contribution from Task 1.5: Obtaining the vertical structure of aerosols and clouds (TROPOS, UHEL, UMAN, CNRS-GAME, HUJI, NUIG, MPI-M, FMI, INRNE, CYI, CUT, ETHZ)

A number of field campaigns have been performed since 2013. The observations are used for BACCHUS validation studies and investigation of aerosol-cloud interaction. Focus of Task 1.5 is on vertical profiling of aerosol layers and mixtures of different origin together with cloud systems in key regions of climate interest (from the tropics to the high latitudes, from polluted to pristine regions, at sea level up to high mountain height levels) by means of in situ measurements (ground-based, aircraft), active remote sensing from ground with lidar and radar, and satellite remote sensing. The observations are performed in the framework of intensive field campaigns and by means of continuously running monitoring networks. During the last 9 months (reporting period), we analyzed previous field campaign data, network data (CLOUDNET), including new CLOUDNET-like observations at the Barbados Cloud Observatory performed by *MPI-M*, Hamburg, with focus on quality assurance, and we concentrated on the validation of satellite retrievals of aerosol and cloud products. The field campaigns provide excellent opportunities for satellite overpasses for validation of VIIRS cloud microphysics retrieval schemes (performed by *HUJI*, Israel). Satellite validation is one of the central activities of Task 1.5 until the end of year 2 of the BACCHUS period.

Below is the list of BACCHUS-related aerosol/cloud field campaigns (note: not funded by BACCHUS):

- BAECC campaign (Hyytiälä, Finland, 2014, in situ, remote sensing, mobile ARM site), coordinated by *UHEL*, Finland,
- HOPE campaigns (Jülich, Melpitz, Germany, 2013, lidar/radar) of the German HD(CP)² project, HOPE-Melpitz coordinated by *TROPOS*, Germany,
- ACCEPT campaign (Cabauw, The Netherlands, 2014, lidar/radar) of the EU ITARS project, jointly coordinated by *TROPOS* and Delft University,
- ACCACIA campaign (Arctic, 2013, aircraft), organized by UMAN
- INUPIAQ of the CLACE-2013 campaign (Jungfraujoch, Switzerland, 2013, in situ), performed by UMAN
- SALTRACE (Barbados, 2013, lidar, aircraft), jointly coordinated by *TROPOS* and DLR, Germany

- Barbados Cloud Observatory long-term monitoring (Barbados, since 2010, lidar/radar), run by *MPI-M*, Hamburg, Germany,
- GoAmazon (Brazil, 2014, HALO aircraft, remote sensing, in situ) coordinated by German institutes (DLR, *MPI-C*, Leipzig University, and others)
- Crete campaign (Greece, 2014, UAV, lidar, in situ), partly funded by BACCHUS, coordinated by *CYI*
- BACCHUS-ENVIMED-ChArMEx campaign (Cyprus, 2015, UAV, lidar, in situ), funded partly by BACCHUS, coordinated by *CYI*, see extended report below.

As a highlight of the reporting period, the BACCHUS Cyprus campaign 2015 from 4 March to 7 April 2015 was successfully conducted (coordination and organization by CYI). At Agia Marina, UAV profiling of aerosol properties (including IN number concentration) was performed (CYI, CNRS-GAME), and IN characterization at ground (ETHZ) was realized. At Limassol (CUT) and at Nicosia (TROPOS) lidar and AERONET photometer observations were performed. The TROPOS equipment was deployed on the roof of one of the CYI buildings. For the first time, detail IN profiling with lidar was conducted. The campaign was organized in collaboration with the French ChArMEx and Envi-Med "CyAr" programs. A dedicated air field has been built at the remote region of Agia Marina Xyliatou where the CYI atmospheric station is located. Flights were performed at altitudes up to 2km (a.s.l) providing observations of the physicochemical aerosol properties within and above the boundary layer capturing, among others, the vertical distribution of dust layers. The CYI UAV (Unmanned Aerial Vehicles or drone) was equipped with a number of instruments for a total payload of 5 kg comprising (a) a meteorological sensor recording temperature and humidity, (b) an micro-aethalometer (AE51) measuring black carbon concentrations, (c) an optical particle counter (Met One 212) measuring the size distribution of particles with size diameter range of 0.3 to 10 μ m and (d) a custom built two-filter holder device to sample Ice Nuclei at 15lt min⁻¹. Parallel to the flights, observations of VOC and aerosol properties (absorption, light scattering, extinction, size distribution, on-line chemical composition, PMx, IN, CCN, CN, etc.) were obtained at the Agia Marina atmospheric station in an effort to perform closure studies.

The National Center for Meteorological Research (*CNRS-GAME*, Toulouse, France) participated in the Cyprus-2015 campaign. The main goal was to complement the ground-based observations of aerosol and cloud condensation nuclei with airborne measurements (UAV) to characterize the vertical distribution of aerosol, radiative fluxes, 3D wind vectors and meteorological state parameters. Payloads were limited to about 500 g. During the campaign, airborne measurements were taken over 4 weeks (5 March to 2 April 2015) with 52 research flights and 38 hours of flight time. Vertical profiles were regularly sampled up to 2100 m above sea level. Often dust layers were observed originating from the Arabian Peninsula and the Sahara Desert. The aerosol profiles generally show a well-mixed boundary layer and compare well with ground-based lidar observations. Flights below and within clouds were also coordinated with satellite overpasses.

Further activities during the last 9 months have been performed as part of task 1.5. **TROPOS** takes care of the CLOUDNET data analysis with focus on temperature-dependent (i.e., aerosol dependent) heterogeneous ice formation in stratocumulus and altocumulus (stratiform, layered clouds) over Europe and the Barbados tropical station. These clouds are likewise simple from the meteorological point of view (show weak thermodynamic effects compared to convective clouds) and thus show most clearly a potential temperature-dependent aerosol effect on ice nucleation. CLOUDNET is a network of super sites with lidar and/or, ceilometer, cloud radar, and microwave radiometer and performs long-term observations of clouds over Europe. CLOUDNET is coordinated by the EU ACTRIS program. CLOUDNET stations considered in our study are: Mace Head (Ireland), Potenza (Italy), Lindenberg (Germany), and Leipzig (Germany). To contrast the observations in polluted Europe with measurements of pure marine, tropical cloud evolution, we include the Barbados (*MPI-M* Barbados Cloud Observatory) data that have been available since 2010. As a first result, the cloud radars always detect first ice formation already at

temperatures just below 0 °C. This is a surprising preliminary result because laboratory studies show that, except for biological particles, anthropogenic and mineral dust particles become activated as ice nuclei not before temperatures are below -10 °C. However, as a first task, we need to perform extended quality checks regarding all the radar observations, which we have started recently.

NUIG, Ireland, analyzed observations of homogeneous water clouds over Mace Head since 2009. Microphysical cloud properties have been retrieved (work still in progress). A preliminary statistical analysis of polluted and clean clouds is available for the 2009-2013 period with the preliminary results: the effective cloud droplet radius (from radar) is anticorrelated with the aerosol particle extinction coefficient (ceilometer at 300m height). Significantly lower cloud effective radii are found when air masses are approaching from the polluted European continent.

UMAN, GB, analyzed several campaign data sets (e.g., Jungfraujoch campaigns, ACCACIA), with focus on aerosol features (including IN number concentration) and cloud microphysical properties (including ice cyrstal number concentration). Data interpretation is performed in conjunction with advanced modelling (WRF, column modelling, large eddy simulations) to understand cloud processes. This work is ongoing.

FMI, Finland, uses AATSR ADV aerosol and cloud retrieval products, and investigates the impact of aerosols observed around cloud fields on the microphysical properties of these cloud layers. FMI develops a new method in the framework of BACCHUS to properly determine aerosol and cloud properties in the first step before aerosol-cloud interaction is studied. The technique can be globally applied.

HUJI, Israel, concentrates on validation of VIIRS satellite remote sensing of aerosols and clouds, and uses the data of a variety of the above mentioned field campaigns (e.g., BAECC, ACRIDICON). The theoretical basis for closure of CCN(S), updrafts, cloud base drop concentrations, and vertical evolution of cloud microstructure was developed and applied to the first BACCHUS-involved field campaign – GoAmazon. Results are in preparation. Similar closure measurements are also planned in mid-latitude winter in California (CALWATER-2 campaign, 15 January to 7 March 2016). In addition, VIIRS data are already used to investigate aerosol-cloud interactions (e.g., in the pristine southern hemisphere).*MPI-M*, Germany, works on a vertically resolved aerosol model with focus on global sets of CCN and IN for use in climate models. The work is based on global aerosol observations (e.g., AERONET), satellite remote sensing, and atmospheric aerosol modelling.

CUT (in cooperation with TROPOS) developed a new lidar-based IN profiling method (for desert dust). The method can be applied to space lidar observations (CALIPSO) and has thus the potential to provide 3-D global data sets in terms of dust-related IN.*INRNE*, Bulgaria, finally performs continuous aerosol measurements at the high mountain station BEO Moussala (2950 m above sea level).

CHANGES WITH RESPECT TO THE DOW

No major deviation from the DoW have occured.

DISSEMINATION AND UPTAKE

A preliminary set of new BACCHUS field experiment data on aerosol, IN, CCN has been collected, and a reanalysis from previous field campaigns is being implemented for adoption by atmospheric scientists outside of the BACCHUS community.

Main results have also been disseminated through published articles:

- Climate Effects of Aerosol-Cloud Interactions by D. Rosenfeld, S. Sherwood, R. Wood and L. Donner, Science, 2014.
- Occurrence of pristine aerosol environments on a polluted planet by D.S. Hamilton, L.A. Lee, J. Pringle, C.L. Reddington, D.V. Spracklen, and K.S. Carslaw, PNAS, 2014.
- Estimated desert-dust ice nuclei profiles from polarization lidar: Methodology and case studies, by R. E. Mamouri and A. Ansmann, ACP 2015.

and on Open Access Journal "Atmospheric Chemistry and Physics Discussion:

- Vertical profiling of aerosol hygroscopic properties in the planetary boundary layer during the PEGASOS campaigns by B. Rosati et al.
- Organic aerosol evolution and transport observed at Mt. Cimone (2165 m asl), Italy, during the PEGASOS campaign by M. Rinaldi et al.
- Sensitivity estimations for cloud activation in the vicinity of the high alpine research station Jungfraujoch (3580 m asl) by E. Hammer, N. Bukowiecki, B. P. Luo, U. Lohmann, C. Marcolli, E. Weingartner, U. Baltensperger, and C. R. Hoyle

Additionally, an article on CCN field data was submitted to ACPD by Paramonov et al., 2015, based on data collected during the BACCHUS "predecessor" EU-project EUCAARI. The article includes collaborative work of several BACCHUS partners, namely FMI, ETHZ, PSI and UMAN with contributions of many others from the BACCHUS community. This article is an important step towards the understanding of geographic differences of CCN properties as it compares multiple long-term and campaign data sets from in-situ measurements in a global perspective.

• Paramonov, M., et al.: A synthesis of cloud condensation nuclei counter (CCNC) measurements within the EUCAARI network, submitted to Atmos. Chem. Phys. Discuss., 2015.