

## Abschließender Sachstandsbericht Leibniz-Wettbewerb

Titel: Marine biologische Produktion, organische Aerosolpartikel und maritime Wolken: Eine Prozesskette (MarParCloud) Antragsnummer: K197/2015

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Federführendes Leibniz-Institut: TROPOS

Projektleiter/in: Dr. Manuela van Pinxteren

#### Hinweise zur Erstellung

Der Sachstandsbericht ist bis zu sechs Monate nach Ende der Projektlaufzeit einzureichen und soll einen Umfang von 5 Seiten nicht überschreiten.

Die Sachstandsberichte sollen Informationen zum Projektverlauf hinsichtlich der Meilensteine und Ziele des Vorhabens selbst und hinsichtlich der übergeordneten Leibniz-Ziele des Leibniz-Wettbewerbs für den Senatsausschuss Wettbewerbsverfahren (SAW) beinhalten. Der SAW nimmt die Berichte zur Kenntnis und ggf. auf ihrer Grundlage Stellung zum Vorhabenverlauf gegenüber der Projektleiterin oder dem Projektleiter des Vorhabens.

Bitte tragen Sie ergänzend zum Sachstandsbericht in die Maske im elektronischen Antragssystem ein Executive Summary (max. 400 Wörter) ein sowie Informationen zu Ergebnissen und Erfolgen (Publikationsliste, Wissenstransfer, etc.), Chancengleichheit und Internationalisierung, und Vernetzung.

Der Sachstandsbericht kann auf Deutsch oder Englisch abgefasst werden.

Folgende Vorgaben hinsichtlich Textgestaltung und Seiteneinrichtung der Sachstandsberichte sollen berücksichtigt werden:

- Seitenränder: Der rechte, linke und obere Seitenrand betragen 2,5 cm, der untere Seitenrand 2 cm.
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- Tabellen: für eingefügte Tabellen, Graphiken usw. gelten die Angaben analog.

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# 1. Zielerreichung und Umsetzung der Meilensteine

The project MarParCloud aimed at achieving a better understanding of the biological production of organic matter (OM) in the oceans, its export into marine aerosol particles and finally its ability to act as ice and cloud condensation nuclei (INP and CCN).

Within MarParCloud, a combination of analytical method developments together with several dedicated lab and field studies and modelling approaches was carried out. The core of the project was a joint field campaign at the Cape Verde islands in autumn 2017.

Altogether, within MarParCloud we could successfully elucidate mechanisms of OM enrichment in the SML and showed the proof of concept of the connection between organic matter emission from the ocean to the atmosphere and up to the cloud level. The marine contributions to cloud effects (i.e. INP and CCN number concentrations), however, were found to be rather limited. These interesting findings underline the complexity of this topic and the different sets of analysis that are required and specifically show the need for differentiating between the aerosol number and aerosol mass (planned in further in-depth studies). The modelling activities experienced several unexpected challenges that are currently being solved and will allow implementing the organic measurements in a new source function for the oceanic emission of OM.

The technological developments within the project, the exchange of sampling strategies, the joint analysis, and the intensive data discussion led to the success of the project. We strongly benefited from the close collaboration within the MarParCloud group that was established in our regular meetings and in the joint activities and create a fruitful network between the MarParCloud partners.

The success of the project is illustrated in the manifold publications in peer-reviewed journals and the opening of a Special Issue in Copernicus journals dedicated to MarParCloud and within the three Ph.D works, among them one already defended the thesis and two are close to finishing.

## 2. Aktivitäten und Hindernisse

The main activities in MarParCloud concentrated on the development and applications of analytical methods to determine important OM classes, INP concentrations and bacteria (WP1-WP4). WP2 designed and created a bubble tower dedicated to the study of a class of sugarlike compounds (transparent exopolymeric particles - TEP) in the water column under manifold controlled conditions, which very successfully resulted in the elucidation of mechanisms of TEP enrichments in the surface ocean. WP3b constructed a plunging waterfall tank that was employed during laboratory experiments and during the field campaign at the CVAO and the MarParCat, a robust catamaran for SML and bulk water sampling that could be applied at the harsh conditions at the Cape Verde islands. WP2b provides a broad suite of auxiliary measurements (such as nutrient and particulate organic matter concentrations in seawater at the Cape Verde campaign). All WPs benefited from these infrastructures and measurements. Modelling approaches concentrated on the development of a new source function that included organic material in freshly emitted sea spray aerosol in a theoretical approach, based on Langmuir-Adsorption and the sea spray source function from the literature. This source function showed that the organic fraction of the simulated sea spray aerosols been is highly dominated by lipids followed by proteins that agreed well with the observations. Moreover, WP5 provided theoretical calculations of atmospheric boundary conditions for the Cape Verde field campaign.

Main joint activities within MarParCloud were the field campaigns (MILAN 2017, Cape Verde 2017, research cruise EMB184, 2018) and joint tank studies (tank experiments in Bremen 2016, Wilhelmshaven 2017, Cape Verde 2017, and during the research cruise EMB184, 2018). A strong internal exchange was realised within our annual meetings that took place at the different institutes of the MarParCloud partners as well as within additional campaign preparation and data discussion meetings.

Problems and changes:

Some work packages underwent slight modifications to the original work plan. Generally, more field work was performed during the project than originally planned, because we obtained the possibility to organize and take part in additional campaigns that allowed to achieve a broader, more robust data set. The main MarParCloud event, the field campaign at the Cape Verde islands, was intensified as we decided that a broader set of instruments would be beneficial especially for atmospheric measurements. These measurements provided additional information on the marine boundary layer and more detailed analysis of the aerosol particles at higher altitude, on a mountain, and finally helped to better study our original research questions and stronger elucidate the process chain of organic matter from the ocean up to clouds. Moreover, external partners participated in the field campaign leading to additional measurements and data analysis that were of benefit to the MarParCloud consortium (listed in detail in the overview paper of van Pinxteren et al., 2019).

In WP1, the method development for amino acids took more time than planned, which was attributed to the low concentrations in the ambient samples, matrix effects, and changes in the analytical system (a new instrument was installed during the project time). The focus of the chemical characterization of OM in WP1 was slightly shifted from sugars to lipids and protein analysis, because the method development and measurements for marine sugars was performed in a thematic similar project that focussed on the Arctic atmosphere. We will profit from these methods developed in the Arctic project because they are currently applied to the MarParCloud samples and provide a broader chemical characterisation of the samples. For WP2, technical problems within the building of the bubble towers and additional campaigns and field work resulted in some delay of experiments with the bubble towers. The solar simulator envisaged for the photochemical experiments during the field campaign at the Cape Verdes was broken and the repair of this system could not be carried out within the financial resources of MarParCloud. Therefore, experiments concerning photochemical OM reactions (that were planned in WP3b) could not be carried out. In addition, a malfunction of the liquid nitrogen generator at the Centre of marine sciences of the Cape Verde prevented the trace gas sampling during the main sampling campaign. For these reasons the original working plan of WP3b was revised in close cooperation with the PIs and we focussed our resources on the assessment of the origin of organic matter in ambient aerosols at the Cape Verde using lipid biomarker and their carbon isotopes. This allowed to quantify the marine derived fraction of lipids in the aerosols and thus contributed to a core question of MarParCloud. WP4 fulfilled all the objectives, however due to a thematic broadening of the campaign, higher than planned finances, time and personal efforts were devoted to the project. Due to the complex, challenging and time-consuming analyses of OM composition in SML and aerosol samples, the originally planned study of microbial turnover of relevant OM classes via targeted approaches (RNA-SIP) in WP3a was replaced by concentrating on studying the water-air transfer of bacteria by 16S Illumina sequencing. Radioactive tracers were replaced by microsensors approaches to determine respiration and production rates by dark and light incubations, respectively. Despite these modifications, WP3a provided important insights into the interplay of OM and microorganisms in SML and aerosol samples, namely the effect of bubble bursting and the relationship between bacterial abundance and ice nucleating activity. Additionally, during cruise EMB184, WP3a collaborated with external partners from Stockholm University to implement a bio-aerosol sampler into marine habitats, from which we are confident to gain further insights of how the microbial community in the surface water influences the (bio-optical) properties of aerosol particles. Regarding modelling (WP5) several delays occurred because the first measurement results could not be used for the validation of the new source function. Several unexpected errors in the simulation set-ups or non-working new implemented model code led to further delays. Several parameters in the model need to be newly adjusted and this is currently ongoing. In parallel to solving these issues the implementation of the organic field measurements in a new source function of the oceanic emission of OM is currently ongoing. Altogether, despite these changes and modifications, the aims of MarParCloud in terms of measurements and data analysis were very successfully achieved, but the modelling part is still work in progress.

3. Ergebnisse und Erfolge

#### Publications and Presentations:

16 publications on the MarParCloud results have been published or are submitted by now in peer-reviewed journals (more are currently in preparation). We opened a joint Special Issue within the Copernicus journals "Atmospheric Chemistry and Physics" (ACP) and "Ocean Science" (OS) with the title. "Marine organic matter: from biological production in the ocean to organic aerosol particles and marine clouds". It aggregates papers resulting directly from the MarParCloud project and closely related projects. Seven manuscripts from the MarParCloud consortium have been submitted by now to the SI. In addition, eight further manuscripts have been published in/submitted to other peer-reviewed journals (see accompanying form). Within these publications, there are two aggregating overview papers led by the MarParCloud PIs that give an introduction and overview to two of the joint field campaigns, the MILAN campaign by *Stolle et al.* (2019, BAMS) and the Cape Verde campaign by *van Pinxteren et al.* (2019, ACPD). These overview papers as well as the single, more specific results-focusing papers include contributions from the different MarParCloud WPs and therefore show the strong linkage and collaborations within the MarParCloud teams and disciplines.

Besides peer-reviewed publications, the MarParCloud team published in scientific newsletters (e.g. Robinson et al., 2019 contributed to the newsletter in "Surface ocean lower atmosphere study". Issue 12. 2019.). Several press releases about the project itself and the related campaigns were published and can be found on the websites of the institutes. All MarParCloud participants, and especially the three Ph.D students, attended manifold national and international scientific conferences and presented their work in oral and poster contributions (15 presentations in total). MarParCloud participants were invited to different institutes to present the project in invited talks (M. van Pinxteren: SOLAS workshop, Dec. 2018, M. van Pinxteren, Lyell Centre, Sept. 2018, N. Triesch: Rudjer Boskovic Institute, May 2018).

#### Qualifications:

PhD thesis:

- Tiera-Brandy Robinson (University of Oldenburg): 01.06.2016 05.09.2019: Transparent exopolymer particles, from the ocean to the air and back again, an aggregates journey
- Xianda Gong (TROPOS): 01.10.2016 01.03.2019 (planned): Cloud Condensation Nuclei and Ice Nuclei Particles Over Tropical and Subtropical Regions in the Northern Hemisphere
- Nadja Triesch (TROPOS): 01.06.2016 01.06.2020 (planned): Organic Carbon in the marine Atmosphere: Sources, Transfer and Implications

Master and Bachelor thesis:

- Franziska Radach (MSc, University of Rostock, 2017)
- Melanie Ziemisch (BSc, University of Oldenburg, 2018)

List of third party funding within the time of the project:

- TROPOS: DFG TR 172: Arctic Amplification (AC)3 phase II, sub-project B04 (DFG 2020-2023)
- ICBM: JPI Ocean: Microplastic FACTS (BMBF, 2020-2022)
- ICBM: MarTERA: Marine Traffic Emission (MATE) (BMBF 2020-2022)

#### Development of technologies:

Three main technology products were developed within MarParCloud that were successfully applied during the project and will of value for further research/campaigns. These are: (1) a bubble tower dedicated to the study of TEP in the water column under manifold controlled conditions (WP2a); (2) a plunging waterfall tank that was employed during laboratory experiments and during the field campaign at the CVAO (WP3b), and (3) the MarParCat, a robust catamaran for SML and bulk water sampling that could be applied at the harsh conditions at the Cape Verde islands (WP3b).

## 4. Chancengleichheit

The MarParCloud group was gender-wise very balanced and consisted of a young, innovative team. The project was led by a female PI and the work packages were led by two female and five male scientists. The Ph.D positions were internationally announced and the group finally comprised two female Ph.D students (one from Germany, one from the U.S.) and one male Ph.D student from China. The MarParCloud consortium hold regular project meetings (at least one big project meeting each year among the participating institute) for the general planning of the project, the reporting about developments, problems and changes as well as the discussing of logistic issues for the field campaigns. The last meeting at ICBM was dedicated to a synthesis of the results and publication plans. All WPs participated in the joint lab and field campaigns. All MarParCloud scientists and especially the Ph.D students attended national and international scientific conferences and presented their results obtained within MarParCloud. Besides, the Ph.D students (and partly the PIs) attended advanced training modules offered by the universities and obtained software trainings (matlab, R). Furthermore, they were all involved in network PhD meetings, summer schools and had (partly) the possibility for research stays at other institutes to receive trainings on specific instruments (N. Triesch attended a 3 weeks research stay at the Rudjer Boskovic Institute). All these activities and especially the long times spent together during the campaigns and provided the possibility to share results and expertise and to build scientific networks.

## 5. Qualitätssicherung

Within the MarParCloud consortium, a vivid information flux and regular meetings among the participants, especially in the course of the planning the joint campaigns were given.

Institute wise, each WP performed regular internal meetings with the Ph.D student /Post Doc and the Pl/supervisors and supervising committees to discuss the work plans, the progress of the work, difficulties and the scientific results. The Ph.D students received a broad suite of trainings and courses to enhance their personal and scientific skills (more details in the previous chapter). In the course of the Research Academy Leipzig, the PhD student of WP1 attended a course on good scientific practise. Within the supervision at the universities and the research centres, good scientific practice was realised. The results of MarParCloud were uploaded on open access platforms like the AWI database PANGAEA (5 data sets are submitted and published, more are in preparation). The EMB 184 station data are stored in the IOW database and the 16S rRNA-gene sequence data will be made available at the European Nucleotide Archive (ENA) by end of 2020. The number of publications in peer-reviewed, mostly open access journals, whereby six publications were first-authored by a Ph.D student, demonstrated the quality of the obtained knowledge and results.

## 6. Zusätzliche eigene Ressourcen

In-kind contributions arose mainly for the fieldwork and especially from the main campaign at the Cape Verdes in autumn 2017, were several further measurements were added to enhance the suite of information to be obtained about the aerosol particles and the meteorological conditions at the Cape Verdes. WP1 and4 added off-line aerosol sampling and in-situ measurements on the mountain top for additional aerosol characterization at a higher altitude and also profiling of the atmosphere with an helikite (WP4) to retrieve meteorological information about the marine boundary layer. Additionally, the contract for the two TROPOS Ph.D students working on the project have been prolonged for 6 more months after the end of the project to guarantee that they can defend the thesis while still in Germany. Further, scientific and laboratory personal was needed to support the analyses and data interpretation of the complex campaigns.

### Person months in detail (for each WP):

WP1-TROPOS: 9 (scientific), 6 (non-scientific); WP2a-ICBM: 4.5 (scientific); WP3a-IOW: 4 (scientific), 6 (non-scientific); WP3b-ZMT: 3 (scientific), 4 (non-scientific); WP4-TROPOS: 12 (scientific), 6 (non-scientific); WP5-TROPOS: 6 (scientific)

### Consumables:

WP1-TROPOS: 15.000 Euros for consumables for analytical measurements for surrogate parameters from the field campaign

WP2a-ICBM: 6 000 Euros for the operation of the catamaran during field campaigns WP3a-IOW: 4.000 Euros for general lab equipment (pipettes, chemicals, tubes) WP3b-ZMT: 2.000 Euros for consumables for analytical isotopic measurements

WP4-TROPOS: 12 000 Euro for participation at measurement campaigns in Bremen and at the Cape Verdes, 10 000 Euros for consumables for the off-line analysis of INP

In addition, the IOW (WP 2b) contributed to the ship time for the EMB 184 with 10.500 Euro per day of operation, that led to a total contribution of 157.500 EURO.

Sum: 49.000 + 157.500 (ship time) Euros

## 7. Strukturen und Kooperation

All MarParCloud participants worked closely together. This was mandatory for the joint campaigns within the project were high interactions and good information flows were the basic for successful actions. With our annual meetings, additional Skype conferences for the preparation of the campaigns and intense data discussion session, MarParCloud was truly a joint effort. Moreover, manifold collaborations to partners outside the MarParCloud consortium were established during the project time (listed in detail in the accompanying form). Several collaborations arose from the joint field campaign at the Cape Verde islands, because beyond the MarParCloud consortium, other international universities and research centres were highly interested in this topic and campaign and participated either by joining the campaign and doing measurements or by support within the data analysis and interpretation. This is visible in the author list of the MarParCloud overview paper listing 50 co-authors from 17 different institutes (see overview paper: van Pinxteren et al, 2019. Further intense collaborations were established during the MILAN campaign in Wilhelmshaven (see overview paper: Stolle et al, 2019), and the research cruise EMB184. These scientists brought additional expertise needed for these multi-disciplinary campaigns, such as in the fields of physical oceanography, photochemistry, gas exchange, meteorology, etc. Moreover, we were able to present the MarParCloud project and the Leibniz-Society to an international audience of interested colleagues and institutes. The collaborations, which started during MarParCloud, will certainly be of high value for future publications, projects and cooperations.

### 8. Ausblick

Within MarParCloud, mechanisms for organic matter enrichment in the surface ocean could be elucidated, however, knowledge on other phenomenon that increase for example the enrichment of TEP in the SML are still elusive. We were able to qualitatively show the link between the ocean and the atmosphere, as ocean-derived organic compounds and bacterial taxa, which are well known to thrive in the SML, were highly enriched in submicron aerosol particles and organic matter was strongly concentrated in cloud water. However, oceanic contributions to CCN and INP were small. These interesting findings require further in depth studies differentiating between the aerosol number and aerosol mass. A better analysis of the sources of ambient aerosol particles is needed. To this end, the here applied lipid biomarker analysis and their carbon isotopes have been shown to be a powerful tool for addressing the origin of organic matter in ambient aerosols that will become very useful in future studies. The concept of enrichment of oceanic species in aerosol particles and cloud water should be further analysed. Finally, to quantify the importance of the ocean on the atmosphere, further advanced modelling studies are needed to elucidate to what extend is seawater and the SML a source of organic matter on aerosol particles.

A more general question for ocean atmosphere interaction studies regards how these two dynamic compartments can be linked as they response on different time scales due to various drivers. For the field campaign at the Cape Verdes, we could conclude a likely connection between compounds in the ocean and in the atmosphere, however this is a challenging topic and needs to be addressed more specifically in future studies.