Project Title: **CLOUD Initial Training Network**
Project Acronym: **CLOUD-TRAIN**

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Beneficiaries: Goethe University Frankfurt (DE), CERN (UN/CH), PSI (CH), Helsinki University (FI), Leeds University (UK), Manchester University (UK), IONICON (AT), TROPOS (DE), KIT (DE), TOFWERK (CH)
Associated Partners: TSI GmbH (DE), Aerodyne Research Inc. (US), Airmodus Ltd. (FI), Lisbon University (PT), CalTech (US), DMT (US)

Summary of the project objectives:

The scientific objective of CLOUD-TRAIN is to investigate, with the CLOUD (Cosmics Leave OUtdoor Droplets) facility at CERN, the potential influences of galactic cosmic rays (GCRs) on aerosols and clouds by modifying the abundance and properties of ions, vapours, aerosols, cloud condensation nuclei (CCN), and ice nuclei (IN). CLOUD-TRAIN assesses the significance of a possible "solar indirect" contribution to climate change. Furthermore, CLOUD focuses on the fundamental understanding of aerosol and cloud processes, with or without the influence of ions. It investigates the role of aerosol nucleation for clouds and climate in general. The role of anthropogenic influences through variation of the key nucleating substances, such as sulphuric acid and nitrogen-containing bases are investigated. As atmospheric budgets of SO2, ammonia and amines are presently dominated by anthropogenic sources, a strong anthropogenic influence on aerosol nucleation and growth processes is expected. These influences have to be separated from naturally occurring variations. By applying a suite of highly advanced instrumentation at the CLOUD facility the following key scientific objectives are investigated: a) new experimental results on nucleation and growth in the presence of multi-component mixtures of vapours of atmospheric relevance, b) new experimental results on ice particle formation and cloud microphysics, and c) new input for global climate models.

Highlight results:

The research within CLOUD-TRAIN led to a number of new findings of highest importance for our understanding of aerosols and climate. The CLOUD-TRAIN experiments discovered an efficient aerosol formation mechanism for pure biogenic particles in the absence of sulfuric acid. Highly oxidized molecules (HOMs) produced from the ozonolysis of alpha-pinene, a monoterpene stemming from plant emissions, are the nucleating agents. A substantial influence of ions on the nucleation of these pure biogenic aerosol particles was found (Kirkby et al., Nature, 2016). These experimental results were used as input for the global aerosol model GLOMAP. The model shows that the pure biogenic particle formation mechanism produces substantial amounts of additional particles during pre-industrial times when anthropogenic sulfur emissions were negligible. This reduces the difference in cloud condensation nuclei (CCN) between present-day and pre-industrial conditions compared to previous assessments. The radiative forcing due to the cloud albedo effect reduces from -0.82 to -0.60 W m-2 (Gordon et al., PNAS, 2016).
The growth rates of the freshly nucleated particles from pure biogenic nucleation at the 1-10 nm size range were also measured. Low volatility HOMs drive the growth of these nanoparticles. The growth is explained in detail by a model of HOM volatility (Tröstl et al., Nature, 2016).
Field measurements performed at the Jungfraujoch in the Swiss alps revealed that pure biogenic nucleation from HOMs indeed occurs in the free troposphere (Bianchi et al., Science, 2016).
The detailed nucleation mechanism for the interaction of dimethylamine and sulfuric acid was studied, proving a kinetic nucleation pathway, determining the nucleation rates at atmospheric conditions, and observing the time-resolved evolution of neutral clusters during the nucleation (Almeida et al., Nature, 2013; Kürten et al., PNAS, 2014).

Based on the extensive data sets of nucleation rates measured in CLOUD, including sulfuric acid, ammonia, organic compounds and ions, a global model of aerosol formation was set up. The role of the different production mechanisms is assessed. Our results show that ionization of the atmosphere by cosmic rays accounts for nearly one-third of all particles formed. However, the simulations show that small changes in cosmic rays do not affect aerosols enough to influence today's climate significantly (Dunne et al., Science, 2016).

Training of researchers:
A joint research programme addressed the scientific objectives of CLOUD-TRAIN and offered a comprehensive research training programme for the fellows. The work was divided into inter-linked individual projects around four sets of joint experiments at the CERN CLOUD facility. The experimental runs focused on aerosol nucleation and growth studies, liquid cloud chemistry studies, and cloud ice phase studies. The research programme was complemented by numerous training activities in the form of summer schools and training workshops.

Work performed:
One technical and four scientific CLOUD campaigns were conducted at CERN:

- CLOUD7 (Oct – Dec 2012): nucleation of dimethylamine and sulfuric acid.
- CLOUD8 (Oct – Dec 2013): nucleation of alpha-pinene oxidized species, homogeneous freezing of water and solution particles, ice particle morphologies during ice formation, sublimation and re-growth, uptake and aqueous-phase processing of SO2 and organics at different temperatures, glassy organic aerosols.
- CLOUD9 (Sep – Nov 2014): measurements of aqueous phase chemistry (chemical processing of sulfur dioxide and isoprene in liquid clouds), ice microphysics, glassy (highly viscous) secondary organic aerosol and their cloud nucleating properties, and ion production and loss mechanisms.
- CLOUD10T (Apr – Jun 2015): new instruments for CLOUD10 run were tested and calibrated.
- CLOUD10 (Sep – Dec 2015): simulate aerosol particle nucleation and growth as observed in the Hyytiälä boreal forest field station under daytime and night-time condition.

In between the experimental phases at CERN, the fellows performed the integrated data analysis, performed instrument developments, participated in field experiments and research activities at their home institutions, did secondments, participated in the training events and conferences, prepared subsequent CLOUD campaigns and wrote publications.

Training activities:

- three summer schools were conducted: Braunfels, Ger, 2013; Hyytiälä, Fin, 2014; Cascais, P, 2015.
- Mass Spectrometry Workshop, Wuppertal, Germany, 2016 (organized by TOFWERK).
- Media Training Workshop, CERN, Switzerland, 2015 (organized by CERN).
- Personal Career Development Plans for each young researcher were realized including training of complementary skills.
- all fellows of CLOUD-TRAIN regularly gave oral presentations at Data Workshops, Collaboration Meetings, and Conferences.
- all fellows published research papers as first authors or are currently in the final stages of preparation of first author papers. All fellows participated in numerous peer-reviewed publications as co-authors.

All research and training objectives were successfully fulfilled. All milestones were reached as planned and in time.

Potential impact and use:
Overall, the CLOUD-TRAIN results clearly lead to an improved understanding of the role of aerosols and clouds in the pre-industrial, present-day and future climate system. Improved scientific understanding of climate forms a decisive impact for taking far-reaching decisions by politics, industry and society concerning our actions with respect to avoiding and mitigating climate change.

Over the past years, CLOUD established as the world-leading facility for atmospheric aerosol nucleation research. Methods for measuring nucleation and nano-particle growth rates were substantially improved over the course of the project. Experimental procedures were advanced to allow collection of comprehensive data sets of highest quality. New procedures for data quality assessment were implemented. Crucial parameters are measured redundantly. Data analysis procedures were cross-checked by independent analysis teams using complementary approaches. CLOUD established new methods of how to conduct aerosol nucleation experiments at conditions representative for the atmosphere that are now setting new standards in the field. The ITN researchers gained extensive experimental know-how and analytical skills. They are well trained to become an important part of the next generation of leading researchers in atmospheric and aerosol science.

In close cooperation of the private and the academic sector during CLOUD-ITN, instruments as well as software solutions for data analysis were substantially improved. The close exchange helped the industry partners to improve their products and services and to gain a direct insight of the instrumental needs of the researchers.

To date, several of the ESRs have successfully completed their PhDs, most are close to finishing it. All have successfully found follow-up positions.

The results of the CLOUD-TRAIN experiments were published in more than 30 peer-reviewed scientific papers including several publications in Nature, Science and PNAS, (http://www.cloud-train.eu/CLOUD-TRAIN_publications.html). Further publications are in preparation. The experimental results for nucleation and growth rates for various chemical systems were parameterized and implemented into a global aerosol and cloud model to assess the role of these processes on the global scale. A unique set of experimental data is now available to the atmospheric modelling community for implementation in models of various scales. The CLOUD results were presented to the research community at international conferences. Several press statements were released along with the high-impact publications, resulting in numerous newspaper articles, broad coverage on the web and other media. ITN fellows gave presentations at many public events.