

### Marine Aerosol Cloud Interactions, MaCloudInc 2.2

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- Introduction and motivation

MaCloud Inc. (Marine Aerosol –Cloud Interactions) aims to build on recent advances in marine aerosol formation processes, both in terms of secondary and primary formation processes. In addition to coastal iodine-driven nucleation and growth events, it has been recently established that open ocean particle production and growth is quite frequent over the NE Atlantic and appears to be driven by organics. In spite of the potentially major influence these particulates have on marine clouds, these constituents and processes are currently not adequately represented in meteorological and climate models (Forster et al., 2007), so in order to improve predictive capability, a greater bottom-up understanding of these behaviors must be obtained through comprehensive observations in real-world scenarios.

- Scientific objectives

This project aims to better characterize these coastal and open ocean events. Further, aerosol mass spectrometry has revealed unique marine aerosol organic characteristics with effective cloud nucleating properties despite low growth factors. The project will aim to characterize the growth factor and CCN activity of varying organic matter enrichments in sea-spray aerosol. In particular, the project will:

- (1) use a range of on-line aerosol mass spectrometric techniques and off-line HNMR techniques for aerosol chemical characterisation (e.g. AMS, ATOFMS, MOVI-CIMMS, TD-CIMMS);
- (2) use a range of aerosol physics instrumentation for physical characterisation (NAIS/AIS, PH-CPC, nano SMPS, SMPS) with a focus on cluster formation as a function of I<sub>2</sub> and H<sub>2</sub>SO<sub>4</sub>;
- (3) use a range of mass spectrometer techniques for gaseous characterisation (O<sub>3</sub>, VOC, OH, H<sub>2</sub>SO<sub>4</sub>, MSA, I<sub>2</sub>, AMSs API-TOF, PSM, and PTR-MS)
- (4) use a range of hygroscopic uptake and CCN instruments both for air and laboratory studies into secondary and primary organic aerosol. (HTDMA, VHTDMA, CCN, Bubble tanks).

Aims and objectives:

- (1) to source apportion marine aerosol
- (2) quantify formation and evolution characteristics
- (3) quantify hygroscopic and CCN properties
- (4) quantify marine aerosol impacts on cloud microphysics.

The 2011 campaign will characterise the high biological activity period impacts on aerosol formation and aerosol-cloud interactions and follows the winter 2010 low biological activity period of measurements in 2010 which was part supported by EUSAAR.

- Reason for choosing station

Mace Head is the best location for this work for a number of reasons. It has been shown to be one of the best stations for the measurement of Northern Atlantic marine air, as it is frequently influenced by

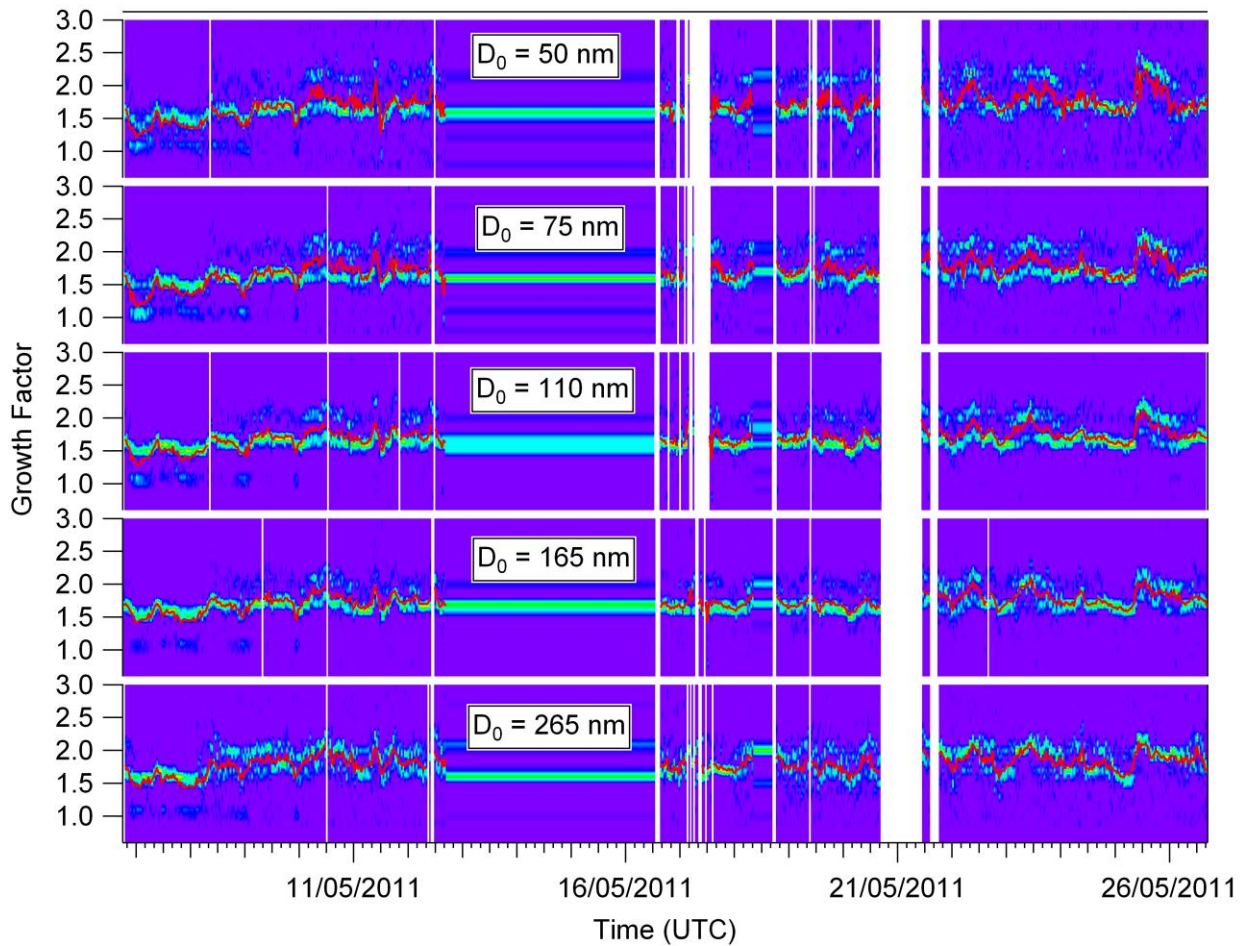
airmasses from the west that have not been influenced by European emissions. The site is very well characterized, with a wealth of previous measurements (from both long-term and intensive campaigns). The measurements were also very well supported by the permanently deployed instrumentation at the site, which provided meteorological, remote sensing and other in situ aerosol composition measurements that would strongly complement the measurements proposed. It is also useful that the PI has worked at the site on a number of occasions in the past, so is already familiar with the facilities.

- Method and experimental set-up

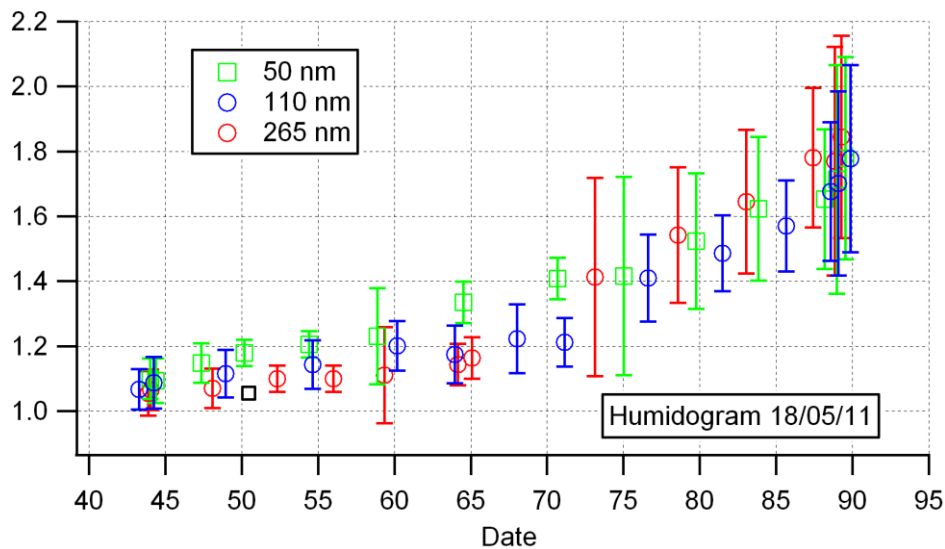
The University of Manchester (UoM) deployed a hygroscopicity tandem differential mobility analyser (HTDMA) (Cubison et al., 2005) and monodisperse cloud condensation nuclei counter (CCN) (Irwin et al., 2010; Good et al., 2010). The purpose of these was to measure the water vapor interactions of particles in the sub- and supersaturated regimes respectively. The HTDMA is similar to other instruments deployed by NUI Galway and Claremont Ferrand, but it brought the ability to be able to perform programmable humidograms (studying water uptake as a function of relative humidity rather than a fixed humidity) and adjust the equilibration times in the instrument. The CCN allowed the study of the activated fraction of particles (compared to a condensation particle counter) as a function of both supersaturation and particle dry size. Inversion methods developed at UoM are employed in the analysis of data from both instruments (Gysel et al., 2009). The instruments were located in a trailer owned by UoM and sampled air through a cyclone to remove larger particles. The aerosol was dried using a membrane drier prior to analysis by both instruments. The instruments were also made coincident with measurements aboard the FAAM BAe-146 research aircraft, which conducted in situ measurements of cloud microphysics above the site.

- Preliminary results and conclusions

Analysis work is currently ongoing, but initial results have been promising. The growth factors of particles at 90% RH have been retrieved and can be seen to vary, so a variety of different aerosol types were characterized:



The HTDMA was also able to collect humidograms of the ambient aerosol:



The analysis of the CCN data is currently ongoing but initial results show that good data coverage was obtained. Overall, a high-quality dataset was obtained where a number of different aerosol types were encountered

- Outcome and future studies

These results will be the subject of ongoing analysis. Once the inversion and quality assurance of the HTDMA and CCN data are complete, they will be compared with the other data available from the site from other institutes. This will include other equivalent hygroscopicity measurements and measurements of composition from the Aerosol Mass Spectrometer and impactor analyses. On a technical level, consistency

between hygroscopicity measurements will be assessed and where differences exist, we will investigate this in lights of variables such as organic content of the particles and the residence time in the instrument. The measurements of composition will be used to inform a closure study between the measured and modeled values of growth factor and critical supersaturation. The performance of detailed models such as ADDEM (Topping et al., 2005a, b) will be tested, as well as less explicit parametrisations such as kappa-Kohler (Petters and Kreidenweis, 2007) that are better suited for implementation in large-scale models. These assessments will also be compared with similar exercises performed in other environments and geographic areas (Good et al., 2010; Irwin et al., 2010; Allan et al., 2009). These results will be analysed in the context of the data on meteorology, air mass origin and phytoplankton activity provided by NUI Galway, to assess the potential influence of biogenic organic matter, as has been performed in previous studies (O'Dowd et al., 2004). Finally, we will try to form linkages between the aerosol composition and properties measured on the ground with stratocumulus cloud microphysics, measured in situ using FAAM or the remote sensing products available at Mace Head. This will in turn help to validate and inform model treatments of cloud microphysics in these environments. Throughout this, we will work closely with our collaborators at Galway and Claremont Ferrand and will work together to produce publications on these subjects, speculatively aiming to submit to a high-impact journal during the latter half of 2012.

- References

- Allan, J. D., Topping, D. O., Good, N., Irwin, M., Flynn, M., Williams, P. I., Coe, H., Baker, A. R., Martino, M., Niedermeier, N., Wiedensohler, A., Lehmann, S., Müller, K., Herrmann, H., and McFiggans, G.: *Composition and properties of atmospheric particles in the eastern atlantic and impacts on gas phase uptake rates*, *Atmos. Chem. Phys.*, 9, 9299-9314, 2009.
- Cubison, M. J., Coe, H., and Gysel, M.: *A modified hygroscopic tandem dma and a data retrieval method based on optimal estimation*, *J. Aerosol. Sci.*, 36, 846-865, 2005.
- Forster, P., Ramaswamy, V., Artaxo, P., Berntsen, T., Betts, R., Fahey, D. W., Haywood, J., Lean, J., Lowe, D. C., Myhre, G., Nganga, J., Prinn, R., Raga, G., Schulz, M., and Dorland, R. V.: *Changes in atmospheric constituents and in radiative forcing*, in: *Climate change 2007: The physical science basis*, edited by: Solomon, S., Qin, D., Manning, M., Chen, Z., Marquis, M., Averyt, K. B., Tignor, M., and Miller, H. L., Cambridge Univ. Press, Cambridge; New York, 129-234, 2007.
- Good, N., Topping, D. O., Allan, J. D., Flynn, M., Fuentes, E., Irwin, M., Williams, P. I., Coe, H., and McFiggans, G.: *Consistency between parameterisations of aerosol hygroscopicity and ccn activity during the rhamble discovery cruise*, *Atmos. Chem. Phys.*, 10, 3189-3203, 2010.
- Gysel, M., McFiggans, G. B., and Coe, H.: *Inversion of tandem differential mobility analyser (tdma) measurements*, *J. Aerosol. Sci.*, 40, 134-151, DOI 10.1016/j.jaerosci.2008.07.013, 2009.
- Irwin, M., Good, N., Crosier, J., Choularton, T. W., and McFiggans, G.: *Reconciliation of measurements of hygroscopic growth and critical supersaturation of aerosol particles in central germany*, *Atmos. Chem. Phys.*, 10, 11737-11752, 10.5194/acp-10-11737-2010, 2010.
- O'Dowd, C. D., Facchini, M. C., Cavalli, F., Ceburnis, D., Mircea, M., Decesari, S., Fuzzi, S., Yoon, Y. J., and Putaud, J. P.: *Biogenically driven organic contribution to marine aerosol*, *Nature*, 431, 676-680, 2004.
- Petters, M. D., and Kreidenweis, S. M.: *A single parameter representation of hygroscopic growth and cloud condensation nucleus activity*, *Atmos. Chem. Phys.*, 7, 1961-1971, 2007.
- Topping, D. O., McFiggans, G. B., and Coe, H.: *A curved multi-component aerosol hygroscopicity model framework: Part 1 - inorganic compounds*, *Atmos. Chem. Phys.*, 5, 1205-1222, 2005a.
- Topping, D. O., McFiggans, G. B., and Coe, H.: *A curved multi-component aerosol hygroscopicity model framework: Part 2 - including organic compounds*, *Atmos. Chem. Phys.*, 5, 1223-1242, 2005b.