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Deciphering the chemical stratigraphy with LA-ICP-MS for the "Oldest Ice Challenge"–Where do we go from here?

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The success of the "Oldest Ice Challenge"crucially depends on extracting paleoclimatic information at sufficiently high resolution while avoiding misinterpretation by post-depositional signal alteration. The record of chemical impurities provides an important set of paleoclimate proxies, but its preservation is challenged especially in deep ice e.g., by crystal growth, diffusion and chemical reactions. Ice core impurity analysis with laser ablation inductively coupled plasma mass spectrometry (LA-ICP-MS) has a two-fold value in this context. Combining micro-destructive sampling with micron-scale resolution, LA-ICP-MS can be used for stratigraphic analysis with line profiles along the main core axis and for mapping the spatial impurity distribution via 2D imaging. A combined approach promises to extract stratigraphic signals while considering constraints from post-depositional processes. However, current limitations arise primarily from the need for an improved signal interpretation in concert with a call for further technical innovation. Here we present first results that already highlight a way to fully capitalizing on the high-resolution impurity analysis once the "Oldest Ice"has been retrieved. The 2D chemical images afforded by LA-ICP-MS can provide the "ground truth"for assessing the spatial significance of single line profiles and have already revealed a close association with grain boundaries for impurity species such as Na and Mg. In order to trace the development of this impurity-grain boundary association, we show how the imaging can in principle also be extended to firn samples, potentially opening a door for investigating signal formation with the imaging approach. In some new images recorded from Greenland and Antarctic ice, widely dispersed aggregates of particles become visible for primarily dust-related species such as Al and Ti. Combining image segmentation and elemental ratios allows us to investigate both the localization and composition of terrestrial dust, showing consistency in a preliminary validation against cryo-Raman spectroscopy. Aiming at a more detailed glacio-chemical characterization in the future, we show how the combination of laser ablation with "time of flight"mass spectrometry greatly expands the range of elements retrieved during a single image acquisition, even when analyzing highly pure Antarctic ice. A further technical innovation which is yet to be realized would be a large cryocell with imaging capabilities to avoid the destructive preparation of cm-sized ice strips and to make decimeter-sized images feasible. Proceeding along this roadmap may ultimately provide a new tool for deciphering the deep ice chemical stratigraphy –in the "Oldest Ice"and elsewhere.

Primary author: BOHLEBER, Pascal (Ca'Foscari University of Venice)

Co-authors: STOLL, Nicolas (Alfred Wegener Institute Helmholtz Centre for Polar and Marine Research); DALLMAYR, Remi (Alfred Wegener Institut); LARKMAN, Piers (Ca'Foscari University of Venice); RITTNER, Martin (TOFWERK AG, Thun, Switzerland); ROMAN, Marco (Ca'Foscari University of Venice); WEIKUSAT, Ilka (Alfred-Wegener-Institut Helmholtz-Zentrum für Polar- und Meeresforschung); WILHELMS, Frank (Alfred-Wegener-Institut Helmholtz-Zentrum für Polar- und Meeresforschung); BARBANTE, Carlo (Ca'Foscari University of Venice; CNR-Institute of Polar Sciences (CNR-ISP))

Presenter: BOHLEBER, Pascal (Ca'Foscari University of Venice)

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