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Evaluation of aeolian dust records obtained from Polar Ice Cores

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Abstract

When an ice core sample is analysed for its aeolian dust content, it is melted and the particles detected are suspended in water. Consequently, dust measurement techniques employed in the ice core community differ from those used for in-situ studies of airborne dust.

Methods commonly used to classify insolubles suspended in a liquid are either based on the particles' interaction with light or on the detection of resistive pulses by means of Coulter counting. Data sets obtained with Coulter counters are widely accepted as references and other techniques are judged against their ability to reproduce these.

Unfortunately, optically acquired ice core dust records were found to differ. By analyzing two sections of the NEEM dust record, two different evaluation procedures are discussed before a third protocol is proposed. It is found that relative changes in the archived dust load can be reproduced, while the simultaneous attainment of absolute concentrations or changes in the grain size frequency histograms in high resolution remains difficult.

Keywords: Detection techniques; ice cores; continuous flow analysis.

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1. Introduction

Dust differs from other paleoatmospheric aerosols archived in ice cores, because it carries information in its absolute levels as well as in its size distribution (Pye, 1987, p.129). Therefore, not only the dust concentration, but also the mode of a mono-modal lognormal distribution fitted to the deposit's fine particle fraction is frequently reported (e.g., Petit et al., 1981).

Analysis of insolubles suspended in a liquid is common in the fields of oceanography or microbiology (e.g., Yentsch and Yentsch, 2008; Zhang et al., 2009), where the methods employed are either resistive pulse measurements (i.e., Coulter counting, CC) or the detection of electromagnetic radiation not attenuated or scattered by particulate matter.

The size range of the deposited dust grains prohibits the use of light scattering techniques (Kettner, 2013), but CCs and laser-based attenuation sensors (LS) have been used to measure samples from the recently drilled NEEM ice core (NEEM members, 2013). CCs are widely appreciated not only for their sensitivity, but also for their reproducibility both within the ice core community (e.g., Steffensen, 1997) and beyond (e.g., Zhang et al., 2009).

Below, a comparison between a data set produced by CC and three data series acquired with different LSs is presented. The optical sensors were connected to a Continuous Flow Analysis system (CFA, Kaufmann et al., 2008), which simultaneously collected decontaminated aliquots. These were refrozen and analysed using a CC at the Laboratoire de Glaciologie et Géophysique de l'Environnement (LGGE) in Grenoble. LSs were provided by the German Alfred-Wegener-Institut (AWI), the Danish Centre for Ice and Climate's (CIC) and the Bernese Institut für Klima- und Umweltphysik (KUP).

Inconsistencies between the time series recorded with optical or volumetric detectors have been reported before (Ruth et al., 2003), as have deviations of records obtained with different LSs (Bigler et al., 2011). To mediate these differences, two data evaluation protocols have been proposed (Ruth et al., 2003; Lambert et al., 2012).

These are applied to the NEEM dust record at 1859.0-1871.1 m and 2016.3-2028.4 m depth (dating from 48.6-50.0 and 74.4-76.3 ka before 2000 AD, resp.), before a third correction method is suggested (Kettner, 2013). All methods' strengths and weaknesses are discussed before an outlook is presented how already obtained high resolution dust data could be treated in the future.

2. Dust detection techniques

Bader et al. (1965) presented the first measurements performed on individual dust grains from ice core samples. These were executed using the principle patented by Wallace Coulter (1956). In a Coulter counter (CC), an aperture connects two otherwise electronically isolated, electrolyte-filled chambers. On either side of the aperture, electrodes are placed. During a particle's passage through the orifice a fraction of the electrolyte is displaced, resulting in an increase of the electrical resistance between the two electrodes. This in turn is a direct measurement of the particle's volume and commonly recorded in 256 bins ranging from equivalent sphere diameters in the submicron range to more than 20 μm.

With the introduction of Continuous Flow Analysis (CFA, Sigg et al., 1994) to ice core science, the resolution of paleoclimatic aerosol records could increase significantly. In CFA, a slab of ice is continuously melted and the sample stream is directed into an array of detection lines. Commercially available CCs, however, do not allow for continuous sample feeding.

Therefore, Ruth et al. (2003) introduced an optical flow-through detector for microparticles to CFA, that was developed in collaboration with Klotz GmbH (Bad Liebenzell, Germany). In their attenuation-based laser sensor (LS), particles are identified when they partially shield a 1.5×250 µm laser beam. Decreases in the detected intensity are binned according to minimum intensity levels in usually 32 channels.

Absolute sizes are allocated to the bin edges by the manufacturer after linear interpolation between signals caused by latex beads of various sizes prior to the dust measurements. Depending on the manufacturer's calibration, the LS's dynamic range is between 1 µm and 10 or 15 µm.

3. Attenuation sensor corrections

When the size of insolubles suspended in water is of interest, CC measurements often serve as reference for data collected employing optical techniques (e.g., Ruth et al., 2003; Shapiro, 2004). Publishing the first LS-acquired data set, Ruth et al. (2003) already reported a mismatch between size distributions obtained with their LS and a CC.

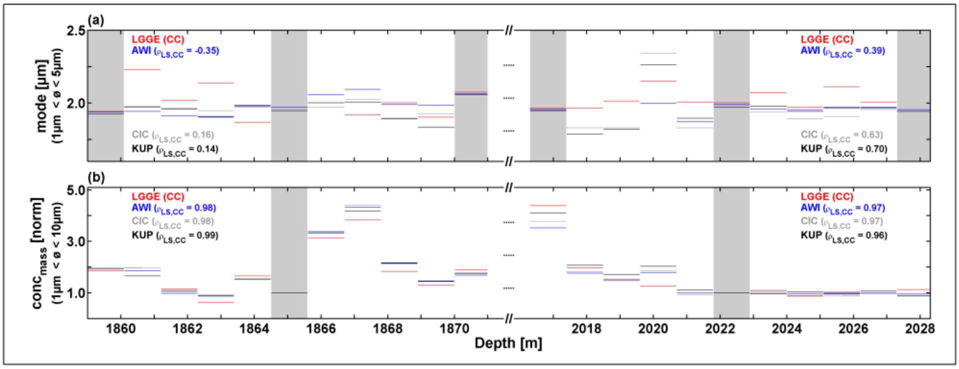

Fig. 1. (a) Mode and (b) normalised concentration - derived from various LSs after evaluating the data using Ruth et al. data treatment - and their correlation to a time series obtained with the LGGE's CC. Grey intervals in (a) indicate intervals in which the LS data has been mapped to the CC-obtained size distribution, while in (b) intervals to which the concentrations are normalised are highlighted.

In order to match the size distributions' modes, Ruth et al. used CC measurements of selected depth intervals to alter the LS's factory-set bin boundaries. Assuming the one at 1 µm to be correctly set, new values for the larger bin edges were determined by the edge of the largest CC bin that, in a cumulative distribution, had less counts than the LS bin in question. Bin edges for depth intervals not directly linked to CC data were obtained by linear interpolation. As measurements with LSs and CCs might result in unequal number concentrations (particles ml^{-1}, see also Fig. 2a), the possibility for an 'efficiency correction' is provided. This is done by scaling the counts in a selected LS bin to the output of the CC in the corresponding size interval.

Results obtained applying this protocol to LS-collected data are compared to LGGE's CC record and presented in Fig. 1. A look at the correlation of the LS data sets with the CC time series in Fig. 1b reveals that relative variations in dust concentration can be nicely captured by any of the three LSs. Relative changes in the modes of the lognormal distribution in the LS data, on the other hand, need to be interpreted with care - also after the bin size correction. It becomes clear that the values found for the respective modes are dependent on the choice of intervals which are forced to resemble the CC data as much as possible. This highlights the underlying problem of mediating a systematic error with numerically different corrections for each measurement.

A systematic correction technique was proposed by Lambert et al. (2012), who established a linear relationship between the logarithms of dust levels detected by either method. The functions mapping the LS data to the CC output - based on measurements of the 11 1.1m long segments in the second interval (2016.3-2028.4m) with the three LSs connected in series - are depicted in Fig. 2a. Differing parameters are a consequence of the disagreement of various LS data series (see also Fig. 4b)

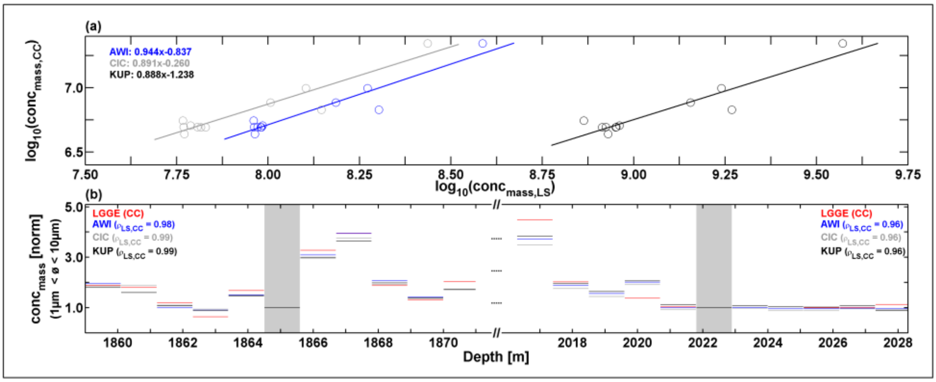

Fig. 2. (a) Functions to map LS-obtained dust levels between 2016.3m and 2028.4m to CC measurements. (b) Correlation of corrected and normalised LS and CC data; shaded intervals mark the levels to which the time series are normalized.

Just as the data manipulation proposed by Ruth et al. (2003), Lambert et al. (2012) can reproduce changes in the dust levels (see Fig. 2b). By design, the evaluation protocol put forward by Lambert et al. will outperform Ruth et al.'s in terms of reproducing CC-obtained absolute dust levels. When no further steps are taken, however, this comes at the expense of loosing the climatic information extractable from changes the dust grains' size distribution.

All methods currently present in the literature depend on additional CC measurements of the same sample. These potentially error prone measurements were simultaneously rendered redundant by Erhardt (2013) and Kettner (2013), who did not strive to reproduce volumetric measurements, but assumed the grain sizes to obey a smooth size distribution. Both followed Lambert et al. (2012) when recognising the need to correct the systematic error systematically and accepted Ruth et al.'s assumption of an erroneous attribution of absolute values to the LS's bin edges.

Erhardt systematically shifted events he found likely to have been attributed to wrong bins to neighbouring ones. Kettner, on the other hand, decided like Ruth et al. (2003) not to keep the bin boundaries as stated by the manufacturer. He only leaves the bin boundary at 1µm and the largest bin's lower limit unchanged. Bin edges in between are determined by minimising the difference between (1) the number of counts in a given bin normalised to its width and (2) the value found by linear interpolation between its neighbouring bins.

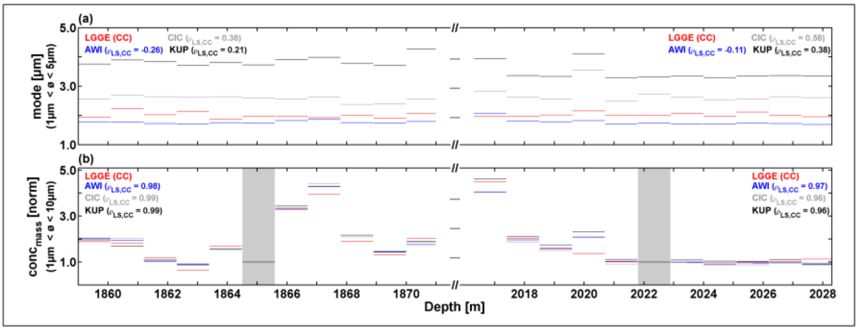

Fig. 3. (a) Mode and (b) concentration after adjustment of bin boundaries following Kettner (2013) and their correlation to the CC-acquired master series. Dust levels in (b) have been normalised with respect to the highlighted core sections.

Fig. 3 shows the LS-obtained results after smoothing the size distributions. The variations in relative dust levels are equally well reproduced as after applying the manipulation techniques published by Ruth et al. (2003) and Lambert et al. (2012). In the interval between 1859.0 and 1871.1m, correlations of the LS- and CC-determined modes are now higher than if corrected following Ruth et al.'s approach. In the deeper core section, however, Ruth et al.'s method reproduces the modes' variation more accurately. Furthermore, the absolute mode values found after applying Kettner (2013) correction, deviate from the independently obtained CC data on average 10, 40 and 100% for the AWI, CIC and KUP data, respectively. This is more than when the bin boundaries are altered using the technique presented in Ruth et al. (2003), where the mean discrepancy is found to be around 5% for any LS.

4. Discussion and future directions

Classical Coulter counters are not compatible with CFA systems, so that all existing high resolution ice core dust records were collected using attenuation-based techniques. Although these LS-obtained records differ with respect to one another and to CC measurements, they can reproduce relative changes in the archived dust levels after correction by any of the methods discussed. Still, dissatisfaction about continuous dust data is mutually shared due to inconsistent results and the LSs incapability to reproduce volumetric Coulter counter (CC) measurements (Fig. 2a and 4b).

The employment of attenuation sensors is problematic, as Ruth et al. (2003) have pointed out, because the geometric shading of light by the insolubles becomes the dominant part of the signal only for particles larger than 7 µm, a multiple of the mean sizes of particles in ice cores (e.g., Petit et al., 1981; Steffensen, 1997). For smaller particles, effects of Mie-scattering (Mie, 1908) cannot be neglected. Optical techniques further have a lower sensitivity resulting in a higher detection limit (Zhang et al., 2009), and they do not directly measure volume - the quantity usually reported. Instead, they yield a signal dependent on the particle's cross section and surface, from which volume needs to be inferred.

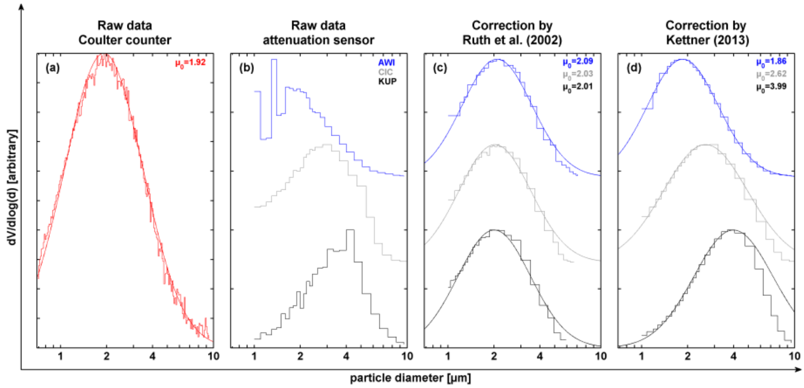

Fig. 4. (a) Size distributions detected with (a) LGGE's CC and (b) multiple LSs in the depth interval (2196.7-2197.8) m. Results after applying the data manipulation protocol put forward by Ruth et al. (2003) and Grindsted's (2013) suggestion are shown in (c) and (d), resp.

None of the data manipulation protocols proposed so far was able to generate agreement between the relative changes in particle size distributions detected with LSs and CCs, respectively. Ruth et al. (2003) proposed a method to force LS-acquired size distributions to resemble CC data over arbitrarily chosen core sections (Fig. 4c). Still, Fig. 1b shows that the obtained values for the size distributions' modes match only in the chosen intervals. The LS data manipulation put forward by Kettner (2013) can yield a higher correlation between LS- and CC-detected size variations, but by design preserves the size distribution's underlying shape. This means that the absolute mode values are bound to differ (Fig. 4b and 4d).

It has to be concluded that (1) highly-resolved particle size records from ice cores are currently not able to reproduce CC-obtained results, (2) further work is required to develop a correction algorithm for presently available records before the extraction of climatic information preserved in the grain size frequency histograms is feasible, and that (3) the replacement of optical methods by continuous resistive pulse measurements for future analysis should be considered.

Potential for future improvement lays in (A) the combination of the two systematic corrections proposed by Kettner (2013) and Lambert et al. (2012), where the former could be used to obtain the distributions variance and the latter to constrain the size distribution's integral. Also the inclusion of Gustav Mie's (1908) work into the correction protocols might be fruitful. Alternatively/additionally in (B) the combination of Erhardt's idea to shift events from one bin to another and Mie-theory. Counts will then not be shifted to neighbouring bins, but to those covering size ranges that Mie-scattering predicts to cause the same signal.

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