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Interactive comment on "Determination limits for cosmogenic ^{10}Be and their importance for geomorphic applications" *by* Sara Savi et al.

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While every AMS laboratory by necessity has a protocol for dealing with low-level backgrounds and uncertainties, and most AMS laboratories have published their background correction procedures in the technical literature, the cosmogenic nuclide field in general does not have a comprehensive paper on the treatment of very low level samples. I was hoping that this manuscript would fill that void. However, it almost completely ignores a substantial body of literature on blank subtraction, in particular for Poisson processes, and takes an oversimplified approach to backgrounds and blank subtraction. I do not think that it advances the field in a significant and widely useful way.

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Discussion paper

As explained, for example, by Elmore et al. (1984) and many others [e.g., Donahue et al. 1990; Wacker et al., 2010; ...], the blank or background in an AMS measurement depends on (1) contamination during sample preparation; (2) contamination in the ion source, commonly referred to as 'cross-talk'; and (3) tails of other ionic species or isobaric interferences in the detector, commonly referred to as 'interference'. In addition, the blank can also include contamination in the cathode material (Middleton et al., 1994), and the metal binder mixed with the sample, here subsumed under sample preparation. These effects are listed in the supplementary information section S5 but should rightfully belong in the main text due to their importance.

Treatment of the background depends on the source of the counts. If the counts are due to consistent laboratory contamination from reagents, then it is appropriate to treat the blank as a fixed number of atoms added during sample preparation. In this case, the number of atoms added to the sample during preparation is calculated and then subtracted from the total number of atoms determined for that sample, as is done in this manuscript. If the background is due instead to cross-talk, then the blank should more appropriately be subtracted as a count rate. This is because the background 10Be ions are being evaporated from various surfaces inside the source at a constant rate, independent of sample beam current. Finally, if the background is due to interference in the detector (most likely from boron, unless B interference is eliminated by a gas-filled-magnet or by post-stripping), then there is a correction factor applied that is proportional to the interference count rate, determined empirically for each laboratory and most often for each run. Each AMS laboratory should evaluate the sources of background and provide this information to the user whether they make the 'machine' background subtractions or not.

Relevant to machine backgrounds, Savi et al. mistakenly state that a cross-talk value of 0.1 permil implies that this source of background can be ignored. Unfortunately this is not usually the case for low-level blanks. AMS standards are typically much higher ratio than the samples, so that the standardization can be made with high precision.

This manuscript did not state where the measurements were made, but the Savi et al. (2016) paper from which the data were derived states that they were made at the University of Cologne against standards with 10Be/9Be ratios of 5.35 x 10-13, 6.36 x 10-12, and 2.71 x 10-11. Dewald et al. (2013) indicate a beam current of 5 μA for beryllium and a transmission of 33% at the Cologne AMS. That would correspond to roughly 5 counts per second (cps) at the detector for the lowest standard, and about 250 cps for the highest standard. A cross-talk value of 1 x 10-4 would then correspond to a count rate of one count per 30 minutes for the lowest standard and about 1-2 cps for the highest standard. Is this really negligible? Savi et al. don't state their beam currents for blanks and samples, but blank 10Be/9Be ratios range from about 1E-16 to 1E-15. If the blanks and samples have the same current as the standards (which they usually don't), then a blank of 1E-16 would correspond to about 1 count per 16 minutes. At this level, even if only the lowest standard were previously run, cross-talk would account for around 1/3 of all counts. If higher standards were used instead, or if higher ratio samples were previously run in the source, then cross-talk could completely dominate the measured AMS background. It is unclear how much of the variation in the blank, then, is due to laboratory cleanliness versus cross-talk in the AMS source for low-level blanks without additional information such as machine blanks that were run concurrently. Certainly if the higher ratio samples (e.g. 53-57) were run adjacent to a blank they would contribute on the order of 1 count per few minutes, or up to 5-10 counts due to cross-talk, depending on the run time.

Making the problem more confusing, there may very well be idiosyncrasies and statistical overestimates of the blank at various laboratories. I know from personal experience that at PRIME Lab and at LLNL a zero blank is never reported. If no counts are detected, then at the end of the run a single count is added artificially and assigned an uncertainty of 100%. This is a conservative approach that overestimates the true value of the blank. (Four of the blanks in this paper have 100% uncertainty, suggestive of a single count. Most have between two and twenty counts in the detector, based on the stated uncertainties.) In addition, the machine blank may easily be variable from run-

to-run at the AMS facility, depending on variables such as cross-talk (which changes over time during a run), the AMS tune, and the defined region of interest in a dE/dx detector.

The most important oversimplification in this paper, and the one that is the most concerning, is the use of Gaussian error analysis. An AMS measurement divides nuclear counts in a detector by the beam current measured in a Faraday cup. The number of counts is a Poisson process governed by both time and beam current, and *must* be treated using Poisson statistics. This is especially critical for blanks and low-level samples, where Poisson statistics deviate strongly from Gaussian (e.g., Currie, 1972). There is in fact a robust literature on low-level Poisson statistics and measurement backgrounds for radioactivity counting, particularly related to health physics. I would argue that if you want to distinguish a low sample from a blank, then the correct approach would be to compare either the counts per Coulomb or counts per time using net Poisson statistics, with the difference being whether you consider the background to come from cross-talk or from intrinsic 10Be in the blank. Methods should follow, for example, Potter and Strzelczyk (2008; 2011), Alvarez (2013), or others dealing with low numbers of counts and variable counting times. Alternatively, a Bayesian approach modified from examples such as Mathews and Gerts (2008) and references therein could be used. The point is that there is an entire literature on blank subtraction and detection limits using Poisson statistics that is almost completely ignored in this manuscript.

Admittedly, for high blanks and samples there are more than enough counts in the detector to justify the use of Gaussian uncertainty analysis, and some AMS laboratories follow this sort of background subtraction (e.g., Nadeau and Grootes, 2013). However, as the authors show, the distribution of blanks is better described by a binomial distribution and a confidence interval or likelihood approach would be preferred. In the end, the treatment of high blanks and high ratio samples, though, is just not as interesting a problem as the treatment of low-level samples and blanks, it is one that is more easily

resolved, and it has already been dealt with in numerous technical AMS papers. The real advance that could be made in this paper would be to show how to properly deal with low-level samples that have measurements near the blank.

I appreciate the discussion in the manuscript about whether to choose a smaller number of blanks corresponding to chemistry batches or a longer-term average blank value. The philosophical approach is very different, depending on what one believes the source of the blank to be. I tend to favor more of a time series approach when evaluating my own blanks relative to known 'machine' blanks or unprocessed carrier material. If my blank is substantially higher than the machine blank then I know that my samples have been contaminated in the lab. If the unprocessed carrier blank is high, then that indicates a problem with the AMS measurement such as cross-talk. The blanks reported here do show some patterns over time, with significantly higher values starting at number 36. Is that due to laboratory contamination or AMS conditions? It is not at all clear that the later blanks should be combined statistically with the earlier blanks. Before doing so there should be some sort of time series analysis to show that there is no drift or trend.

Some smaller issues arise in the paper that should be dealt with in any revision or re-submission. (1) AMS is drastically over-simplified in the description in section 2.2. (2) There seems to be some confusion about the presence or absence of beryllium in minerals. Beryllium is almost universally absent in quartz, but variably present in most other minerals at the ppm level.

In summary, I would very much like to see a paper that goes through how to deal with low-level measurements in a thorough manner, and makes strong recommendations based on robust statistical arguments about the proper way to make background corrections. This paper presents several different options but never really gets into low-count statistics and sweeps several important issues under the rug. While it does address interesting problems of detection limits, I don't see that it brings the field forward in a general way beyond essentially normal practice.

-Darryl Granger

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