Where now? Reflections on future directions for cosmogenic nuclide research from the CRONUS Projects

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Abstract

The CRONUS-Earth and CRONUS-EU Projects have recently examined the reproducibility of cosmogenic nuclide measurements and the ability of cosmogenic-production models to fit high-quality global calibration data sets. In both cases, although results are adequate for present geochronological needs, they must be improved to meet future demands. We recommend that the cosmogenic-nuclide community embark on an continuing effort to understand the sources of the remaining model-versus-data discrepancy, and that in the meanwhile all cosmogenic ages be benchmarked against reference ages calculated in a consistent fashion. We also recommend that interlaboratory reference materials be routinely analyzed along with samples and blanks, that the results be tracked, and that methods of achieving consistency and precision in sample preparation and analysis be shared within the community. We also recommend additional organization of the community to facilitate the use of intercomparison materials, foster analytical improvements, oversee the evolution of the community age calculator, and synthesize ongoing calibration efforts.

Keywords

cosmogenic nuclide analysis; cosmogenic nuclide calibration; interlaboratory comparison

1. Where now?

The CRONUS-Earth Project (“Cosmic-Ray-Produced Nuclide Systematics on Earth”) was funded by the U.S. National Science Foundation. It was complemented by the CRONUS-EU Project, funded by the European Union. The goals of the CRONUS-Earth Project were to quantify the actual reproducibility of cosmogenic-nuclide analyses on a community basis (as opposed to nominal laboratory uncertainties), to test and improve scaling models for cosmogenic-nuclide production, to provide a common computational basis that incorporates the current state of the art in nuclide systematics, to calibrate five of the commonly employed cosmogenic nuclides in a uniform and systematic way so as to have confidence in the equivalency of ages from any of them, and ultimately to refine the accuracy and precision of cosmogenic-nuclide aged down to the 5% level. The findings of CRONUS-Earth have been synthesized by Phillips et al. (2015). As they describe, the project was successful in most, but not all, of these. The issues identified as the largest impediments to achieving these goals were the lack of agreement between even state-of-the-art geological calibration data and scaling models, analytical irreproducibility, and a need for continued improvements in computational models.

Where does the TCN community go from here? This short article attempts to distill lessons that have been learned through collaborative research and to suggest ways in which the TCN community could act together to improve the quality of its science. We recognize that this effort is likely to be a controversial one. Some members of the community are philosophically opposed to any move toward community-based standards, fearing they will become proscriptive and limit individual investigators’ independence of action. Others may disagree with specific recommendations. We emphasize that we see these recommendations as starting points for community discussion. Unlike engineering professions, where adherence to codes of practice is enforced for reasons of public health and safety, in the scientific arena community practices should be voluntary and flexible. We hope that the points below will stimulate discussion on the best ways of dealing with these issues and will lead to a gradual process of community-wide improvement of practice.

In Section 2.6.4.3, Phillips et al. (2015) evaluated the relative contributions to imprecision and inaccuracy. The three main contributors, in order of importance, were bias in the results of the calibration, analytical imprecision, and limitations of the computational models. Below we discuss these in this order.

1.1 Inadequacy in cosmogenic-nuclide-production models

The CRONUS-Earth Project was able to make unexpectedly rapid progress in developing physically based scaling models through importing a new modeling methodology developed for the purpose of radiation dosimetry for aircraft crews (Lifton et al., 2014; Sato et al., 2008). Evaluation of this model against the CRONUS-Earth primary and secondary data sets shows that it is only a moderate improvement over the old scaling method of Lal (1991), although it definitely is superior to the neutron-monitor based ones. As evaluated by Phillips et al. (2015), this result is likely due to the limited altitude/latitude coverage of the calibration data set, and possibly due to site-specific

geological factors. Were low-latitude/low-elevation sites included in the calibration data set, a more marked improvement over the results of the Lal (1991) scaling would probably be apparent. This outcome suggests that it is not an inadequate understanding of the physics governing cosmogenic-nuclide production that is responsible for the lack of an inadequate statistical fit to the model. The two main factors would seem to be either the small size, and associated particularities, of the modern, high-quality primary data set used for calibration, or that relevant terrestrial phenomena that are not fully understood and incorporated into the models (e.g., (Kim et al., 2007)). These could include effects of long-term atmospheric pressure anomalies or of post-glacial isostatic rebound.

We suggest that this problem can be pursued by research in two directions: (1) acquisition of additional data on cosmogenic-nuclide ‘control points’, and (2) use of physics-based models to investigate the sensitivity of cosmogenic-nuclide production to various physical phenomena. With regard to the first, we need additional data on the geographical and temporal distribution of localities that produce problems with the production model(s). The data reviewed above appear to indicate that late-glacial age sites around the North Atlantic and in interior North America are most strongly affected. However, local anomalies in age are the outcome of a global fit to the calibration data set, and thus the sites that exhibit the largest discrepancies are not necessarily those responsible for the misfit. The largest ‘blank spots’ in the latitude/altitude space of the CRONUS calibration data sets (Figure 4 in Phillips et al., 2015) are at low latitude/low elevation and high latitude/high elevation, and these should probably be the highest priorities for future calibration efforts.

Due to the structure of the CRONUS-Earth Project, we could only incorporate data that were available as of the summer of 2010. Since that time several new calibration studies have been published (cited in Phillips et al., 2015), some of them coming out of the CRONUS-EU Project, that could be added to supplement the database discussed above. New studies to add more ‘control points’ should be encouraged, as should viewing them in the context of a global set of controls on cosmogenic-nuclide production, rather than as isolated ‘calibration sites’. In particular, most calibration sites to date have been in the age range 20 to 5 ka; inclusion of older calibration sites might help better constrain the anomalies. We note that as more control points can be added from geographically dispersed localities, the fundamental structure of the calibration may change, shifting the perspective on areas/times that appear to be ‘anomalous’ and those that do not.

With regard to the second direction (use of physics-based models to investigate the sensitivity of cosmogenic-nuclide production to various physical phenomena), models such as LSDn (Lifton et al., 2014) or the model of Argento et al. (2015) can be used to explore what magnitudes of known physical phenomena (e.g., long-term variations in atmospheric pressure) or little-explored phenomena (e.g., variations in water content of the atmosphere) are needed to produce age anomalies similar to those observed. Convergence of measured and physically calculated values will increase confidence that the physical phenomena are understood.

1.1.1 Local calibrations

The inability of a global model to adequately reproduce inferred production rates in some at least quasi-coherent spatial or temporal domain has led to the use of ‘local calibrations’: calibrations based on a set of data limited to a restricted region, and often to a restricted time period. On the one hand, unexplained regional influences do clearly affect nuclide production rates and ad-hoc local calibrations are a necessity for accurate correlations at the global scale. But on the other hand, this approach is fraught with hazards for the discipline. The fundamental strength of cosmogenic-nuclide dating within the broader field of geochronology is that it is based on quantifiable physics. Proposed methods of geochronology that relied on complex and environmentally dependent phenomena (e.g., rock-varnish cation-ratio dating, uranium-trend dating, carbonate accumulation in soils, and fault-scarp morphological evolution) are no longer considered to be quantitative dating tools.

Extensive use of local calibrations carries a risk of loss of confidence in the technique. For example, paleoclimatologists wishing to perform global correlations involving numerous cosmogenic ages would be forced to sort through a large number of local-calibration papers, attempting to evaluate the merit of the calibration data and the technical correctness of the calibration for each one. Lacking the time or expertise to perform this, the tendency will be to discount the validity of the method. Furthermore, local calibrations are presumably restricted in application to specific spatial and temporal domains, but the limits of those domains are seldom quantified.

We recognize that these criticisms are likely to be controversial. Much good science has been done, and will continue to be done, in areas where local calibrations provide the best means of age-calculation. We do not maintain that local calibrations should not be employed, but rather that they should be used with restraint and caution and should be continuously tested. The long-term solution to the systematic issues facing the field lies in elucidating the global physics responsible for patterns in production that are not currently understood, not in a proliferation of local calibrations.

The short-term goal of local chronology and the long-term one of comprehensive global understanding of cosmogenic nuclide production systematics are not necessarily at odds. An increasing number of local calibration points can presumably be mined to place constraints on global production anomalies and point the way to improved global systematics.

1.1.2 Benchmarking

Until a comprehensive global understanding of variation in production rates is achieved, one measure that can help to address the problems is benchmarking. By this, we mean reporting ages relative to a generally accepted ‘benchmark’ as well as to any local calibration or alternative scaling that may be employed. This will help geoscientists seeking geochronological control to put the locally calibrated ages in context, and in particular, to immediately see whether the particular local calibration results in a minor or a radical adjustment of ages relative to the global model.

We suggest using ages calculated according to the LSDn scaling method (Lifton et al., 2014) and with the CRONUS-Earth production parameters as the benchmark. We suggest the LSDn scaling, not because it is more accurate than other methods, but because it has the strongest physical basis and thus is most likely to be developed into a

model that can explain current discrepancies. Consistent use of this benchmark, regardless of the scaling method or calibration preferred by the particular investigator in the particular study, will go a long way toward transparency in the presentation of results and accessibility to non-expert users of the chronology.

1.2 Improvements to analytical methodology

Section 2.1 in Phillips et al. (2015) has reviewed both formal laboratory intercomparison studies and field-data intercomparisons arising from the CRONUS-Earth and CRONUS-EU projects. These have shown that stated laboratory analytical uncertainties uniformly underestimate the variability of the intercomparison data sets, as quantified by the coefficient of variation. The magnitude by which the empirical coefficients of variation exceeded the average reported analytical uncertainties (i.e., the empirical coefficient of variation divided by the average reported analytical uncertainty, expressed as a percentage) ranged from about 15% for ^{10}Be and ^{26}Al measurements on the CRONUS 'N' material to over 300% for ^{14}C on the CRONUS 'A' and ^{21}Ne on the CREU-1 materials. There is no uniform explanation for the 'excess spread' of these data sets. For ^3He , ^{21}Ne , and ^{36}Cl the principal cause is apparently interlaboratory bias, for ^{14}C it is underestimation of within-laboratory spread, and for ^{10}Be and ^{26}Al it is an approximately equal mixture of the two. How can (1) the actual dispersion of analytical data be reduced, and (2) reported laboratory analytical uncertainties be brought in line with the actual reproducibility of the analytical data?

Fortunately, unlike for the case of scaling bias, straightforward, if laborious, solutions are available. The most important one is for all sample-processing laboratories to routinely process recognized intercomparison materials (ICM's) and report the results. Just as blanks are routinely prepared with batches of samples, ICM's should be processed at regular intervals, say every 10 or 20 samples. The results should be reported to a central service and updated regularly on a website, as well as being formally published at intervals. Such a program would enable a much more authoritative assessment of the true state of analytical reproducibility for each nuclide than has been possible through the CRONUS-Earth and EU projects. Perhaps more importantly, it would enable individual laboratories to assess their analytical reproducibility and bias against objective metrics such as the global average for the nuclide or the average of the state-of-the-art laboratories (i.e., those having the most reproducible results). This would hopefully spur laboratories whose performance is lagging to revise procedures in a way that would increase their accuracy and precision. The radiocarbon community, for example, has for many years conducted a systematic intercomparison effort (Scott et al., 2010).

One important, although certainly not unexpected, outcome of the various CRONUS intercomparisons is that the degree of analytical reproducibility is dependent on the nuclide concentrations of the ICM's. Through the two CRONUS projects a variety of new ICM materials have been made available. However, in most cases the range of concentrations is limited to one, or at most two, levels. None of the currently available materials are within the lower range of commonly analyzed field samples, yet this is the range where variability is largest and quantification of uncertainty is the most critical. Production of ICM's at high, intermediate, and low concentration ranges should be a future goal for the cosmogenic-nuclide community.

In an ideal world, intercomparison statistics would be transparent, in other words, each sample-preparation laboratory would make its intercomparison results public. Such transparency would encourage laboratories to be realistic in claims for dating accuracy and precision, would provide a track record that would engender confidence in such claims, and would contribute to encouraging laboratories to improve their precision and accuracy. Similarly, AMS laboratories now assign analytical uncertainties based on sophisticated statistical models, but seldom back up the validity of their uncertainty estimates with publically available empirical data. It would be highly desirable for AMS laboratories to embark on a “double-standardization” program in which they routinely measured sub-standards at various levels against their master standards and routinely made the results public. This would hopefully have the same effects as listed above for the sample-preparation laboratories. In both the case of individual laboratory sample preparation incorporating frequent use of ICM’s and that of AMS laboratories employing double standardization, funding agencies must be willing to support the added material and labor costs.

In the real world, there is often substantial resistance to such transparency measures. It is certainly a goal that the cosmogenic-nuclide community can commit itself to moving toward, if it wishes to. The strongest push in that direction will probably be for leading laboratories to voluntarily release their own track records. As more laboratories move in the same direction, it will create an impetus for transparency.

In order to be effective in improving processing and analytical reproducibility and accuracy, any organized intercomparison program must be partnered with a systematic effort to identify the aspects of sample preparation and analysis that are contributing to poor reproducibility. Although this can to some extent be addressed by systematic experiments laid out in journal publications, it is probably most effectively accomplished by means of websites and on-line list serves, wiki’s, and chat rooms. To be accessible and useful, the outcomes of these experiments and discussions must be available in a systematic and accessible fashion. Many professions with societal responsibilities have done this by means of ‘best practices’ manuals. Although formal and fixed manuals of this type may not be the best solution for the cosmogenic nuclide community, an editable ‘Wikipedia’-type of online manual with provision for annotation and discussion points might be a useful way of making accessible the community efforts at improving accuracy and precision of cosmogenic nuclide measurements without being proscriptive.

Finally, we address the difficult issue of reporting analytical uncertainties. Currently, cosmogenic geochronologists use analytical uncertainties that are derived only from laboratory counting statistics, beam currents, and associated within-laboratory data. (As noted above, even these uncertainty estimates are seldom backed up with empirical data.) These analytical-laboratory uncertainty estimates do not take into account any sources of variability that might be introduced during sample processing and thus, not surprisingly, significantly underestimate the actual variability of the analytical data (section 2.1). This, in turn, results in underestimation of the uncertainty associated with cosmogenic-nuclide ages. The CRONUS synthesis effort (Phillips et al., 2015) has accommodated this fact by subsuming the “excess” analytical uncertainty into the production-rate uncertainty. This procedure should, on average, yield realistic age uncertainties, but it has the effect of penalizing laboratories that make the effort to be more precise than average and still grants unrealistically small age uncertainties to those

that are more imprecise than average! (But note that in most cases the increment of the age uncertainty added by the analytical uncertainty is still fairly small compared to that arising from calibration-site bias.)

In the ideal world alluded to above, this issue could be addressed by each sample-preparation laboratory adding an appropriate sample-preparation uncertainty based on its publically available ICM track record. This approach would constitute a major step toward verifiability of claims for dating precision and would create confidence in cosmogenic geochronology within the earth sciences. An intermediate step might be to remove the analytical uncertainty from the production-rate term and agree to add it as a uniform increment to all analytical measurements, unless the laboratory could, through a publically available track record, demonstrate that a smaller uncertainty was justified. Regardless of the path attempted, we recommend that the cosmogenic community move in this direction.

1.3 Improvements to computational models

The CRONUScalc model was constructed to represent all significant effects on cosmogenic-nuclide production that could be quantified at the time the code was written. Nevertheless, future advances in scientific understanding will need to be included in revised versions of the model. Periodic new versions will be issued to keep the calculator abreast of current developments.

Several fairly straightforward additions or improvements to the calculator are planned in the near future. The most significant of these is to add the capability to use ^{21}Ne data to calculate surface-exposure ages. Another is that, at present, CRONUScalc computes ^3He ages based only on high-energy neutron reactions. The capability to also incorporate low-energy neutron reactions (in most cases a minor contribution to the total ^3He production) will be added soon. In order to help users better understand the sources of uncertainty for calculated ages, the contribution to total uncertainty from each uncertainty source (e.g. propagated analytical uncertainty or production-rate uncertainty) will be reported in addition to the total uncertainty. A routine to flag ^{36}Cl samples in which a large proportion of the production is from low-energy neutron absorption by Cl, which substantially increases the calculated age uncertainty, will be implemented. Finally, improvements are underway to improve the computational efficiency and reduce calculation times.

1.4 Avenues to growth of the cosmogenic nuclide community

Ultimately, improvements to the accuracy and precision of cosmogenic ages and increases in the confidence reposed in cosmogenic dating by the earth sciences rest in large part on actions taken by cosmogenic geochronologists as a community. Although improvements to analytical methodologies may be initiated by individual scientists, movement in the direction of systematic quantification of accuracy and precision and analytical transparency can only be accomplished at the community level. Furthermore, future changes to the CRONUScalc calculator should probably be based on some form of consensus of the community.

Efforts that could be undertaken as a community include the following:

- (1) *Establishment of a systematic analytical intercomparison effort.* At least some form of ICM is now available for all of the major nuclides. However, in most cases the ICM's are available only at one concentration level, but three concentration levels (high, intermediate, low) are really needed. If the goal is achieved of all laboratories analyzing appropriate ICM's on a regular basis, the amounts manufactured to date will probably last only a few years. A concerted effort to manufacture ICM's in large amounts at three concentrations is probably needed if systematic intercomparison is going to be an avenue to analytical-uncertainty quantification and improvement.
- (2) *Setting up an ICM reporting site.* The amounts of data that would be generated by a systematic intercomparison effort would be large. To be useful, they would need to be able to be rapidly compared with ICM data from other laboratories and to be included in statistical evaluations in a real-time fashion. This could be accomplished by a dedicated website. The website could be set up to upload new data, post it, and include it in the ongoing statistical evaluation. Some persons would have to be responsible for website setup, maintenance, and quality control, as well as writing and posting periodic evaluations. If complete transparency were to be achieved, the website would serve as the basis for documenting the basis for individual laboratory analytical uncertainties.
- (3) *'Best-practices' website.* If systematic intercomparison is to serve as the basis for analytical improvements, exchange of information on analytical technique will be necessary. As discussed above, some form of on-line forum is probably the best venue for such exchanges. At some point, distillation of the discussion into a readable, living, best-practices document will be necessary. This will have to be overseen by persons with considerable expertise in the sample preparation procedure being summarized.
- (4) *Calculator oversight.* CRONUScalc is currently housed by the EarthChem website (<http://www.earthchem.org/overview>). "EarthChem is a community-driven effort to facilitate the preservation, discovery, access and visualization of data generated in the geosciences, with particular emphasis on geochemical, geochronological, and petrological data." It is supported by the U.S. National Science Foundation and is internationally accessible. It would appear to be a suitable long-term site for the calculator. However, changes will undoubtedly need to be made to the calculator as knowledge advances. It would be best if this were by some form of consensus of the cosmogenic community rather than at the purview of a small group of investigators. This might be achieved by appointment of an international advisory committee.
- (5) *Ongoing calibration efforts.* Setting up the calculator simultaneously for ^{10}Be , ^{26}Al , ^{36}Cl , ^3He , and ^{14}C , reaching consensus on calibration data sets, performing quality control, and completing the calibration was something of a heroic effort for the CRONUS-Earth Project. However, future calibration efforts can build upon the foundation established and the future calibration activities should have the capability to be much quicker and more flexible. As discussed above, if the limitations of the current scaling models are to be overcome, an active and ongoing investigation utilizing new data sets as they become available will be desirable. If such results are to be adopted rapidly by the community, some

measure of community consensus regarding what data sets to use will be desirable. This, again, might be performed by an elected advisory committee.

Scientific presentations on applications of radiocarbon and methodological developments are presented at many meetings, but most internal business of the radiocarbon community is conducted at the triennial Radiocarbon Conferences. The cosmogenic-nuclide community might adopt a similar model. The most obvious venue for a community meeting is the also-triennial International Conference on Accelerator Mass Spectrometry. In recent years this has often been preceded or followed by a workshop on cosmogenic nuclides. The main disadvantage is the relatively narrow focus of the parent meeting. Other options with broader appeal might include the annual Goldschmidt Conferences, the Radiocarbon Conference, or the American Geophysical Union Annual Meeting.

The combined CRONUS-Earth and CRONUS-EU projects have given the cosmogenic-nuclide community a foundation in both theory and empirical data that it previously lacked. In order to build on this foundation, a new level of coordination and organization will be required. The nature and goals of this new community-based effort can, and should, be debated, but the effort will be necessary if cosmogenic-nuclide is to advance to a mature field of geochronology.

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