Text S3. Description of the CHLOE Depth Profile Age Model
CHLOE is based on the cosmogenic nuclide production equations presented by Gosse and Phillips [2001]. The high-energy cosmic-ray flux is calculated based on standard exponential attenuation with mass depth and the spallation production rate is proportional to that flux. The model then uses this flux distribution as the source term for the calculation of the epithermal and thermal neutron fluxes by means of the diffusion equations derived by Phillips et al. (2001). The spatial distributions of low-energy neutron fluxes are used to calculate the $^{3}6$Cl production by epithermal and thermal neutron absorption. Production parameters given in Phillips et al. [2001] and Stone et al. [1998] are used in the model. CHLOE uses the average bulk chemical composition of the soil profile to compute the depth distribution of the low-energy neutron fluxes. A fast neutron attenuation length of 170 g cm^{-2} was used to model all three profiles. The computed neutron fluxes and the Cl concentrations measured at each depth sampled are then used to calculate the ^{36}Cl production rate. In addition to production by the nucleonic component of the cosmic radiation, CHLOE also computes production rates due to primary and secondary effects of the cosmic-ray muon flux, using approaches analogous to those described above. The program also computes the concentration of nucleogenic ^{36}Cl (i.e., produced as a secondary result of radioactive decay reactions within the minerals) in the sample and subtracts it from the measured total ^{36}Cl to obtain the cosmogenic ^{36}Cl concentration.

CHLOE produces calculations of ^{36}Cl concentrations at the sampled depths, subject to variation of three adjustable parameters: the profile inheritance (t_p), the profile deposition age (t_d), and the rate of surface aggradation/erosion (ϵ). Given reasonable independent constraints on these variables, the model output was fairly sensitive to the first two, but relatively insensitive to the last one. The fitting of calculated ^{36}Cl concentrations to data was therefore restricted to aggradation/erosion rates limited between upper and lower bounds estimated for each site based on particle-size measurements and geological observations, as described above. However, the χ^2 variation shows a relatively high degree of sensitivity to the exposure age and a low sensitivity to the erosion rate. The result of this pattern of insensitivity was that the variation in ϵ played a significant role in estimation of the uncertainty of the best-fit deposition age, but only a small role in estimating its actual value.

CHLOE simulates erosion using classical cosmogenic-nuclide formulations [e.g., Gosse and Phillips, 2001], which assume that erosion is from the surface. Analogous equations are not commonly used for the case of aggradation because there is no fixed relation between the cosmogenic nuclide concentration of sediment deposited on top of a geological unit and that of the material in the unit. However, for the material we are analyzing this is not an issue, because the aggradation resulted from atmospheric deposition and materials that were deposited (mainly silt and calcium carbonate) are separated from the parent material being analyzed by sieving and acid leaching. We therefore analyzed none of the material accumulated due to atmospheric deposition and its cosmogenic nuclide content is thus not a factor in interpreting the data. However, some inconsistency remains because the classical equations, when used for aggradation, treat the process entirely as deposition on the surface. This is true for part of the actual atmospheric deposition (the Av horizon), but not for the pedogenic calcium carbonate that precipitates and silt that infiltrates the profile. This may to some extent affect the outcome of the curve-fitting analysis, but probably not to an extent that is large compared to other uncertainties.

The profile age and uncertainty were calculated by means of χ^2 fitting [Bevington and Robinson, 2003] of the ^{36}Cl concentration data from the various depths to the ^{36}Cl distribution modeled by CHLOE. The sum of chi-squared function (χ^2) was calculated for each age-erosion pair as follows:

$$\chi^2 = \sum_{i=1}^{n} \frac{(O_i - M_i)^2}{S_i^2}$$

where O$_i$ is the observed, normalized, ^{36}Cl concentration at each depth interval, i, and M$_i$ is the modeled value at the same depth. The number of concentration measurements is n. S$_i$ is the standard deviation associated with the ith data point:

$$S_i = S_{i,36} + S_{inheritance} + S_{other}$$

where S$_{i,36}$ is the standard deviation from the ^{36}Cl analytical measurement, S$_{inheritance}$ is the contribution to the standard deviation from variability in the inherited ^{36}Cl concentration, and Sother is the contribution from other sources of variability, principally analytical uncertainties in the chemical analyses, bulk densities, and other parameters, combined with uncertainties in the ^{36}Cl production parameters. S$_{i,36}$ was taken directly from the AMS analyses. S$_{inheritance}$ was estimated based on a depth profile of ^{36}Cl concentration measured on a lacustrine beach deposit of known age (from radiocarbon chronology) formed by Lake Lahontan [Kurth, 2003].

χ^2 fitting of the profile fitting yields an age uncertainty that is 3 percent larger than theoretically calculated, assuming all other sources of variation were accounted for adequately. This enhancement of the ?2 can presumably be attributed to unaccounted-for variability of the inherited component. Sother was estimated based on an empirical comparison of ^{36}Cl ages with independently-constrained ages for 30 surface samples [Phillips et al., 2001] and was assigned a value of 6 percent. For each profile we also report the reduced sum of χ^2 (${\chi_\nu}^2$), which is the sum of χ^2, as given above, divided by n, the number of samples in the profile. The magnitude of ${\chi_\nu}^2$ is a measure of the goodness of fit of the data to the model. In general, for laboratory systems in which the model can be assured to provide a complete description, a ${\chi_\nu}^2$ of less than one is considered a satisfactory fit [Bevington and Robinson, 2003].

When dealing with environmental measurements for which the model may be incomplete, somewhat larger ${\chi_\nu}^2$ are often considered acceptable.

The array of sum of χ^2 values was contoured. The best age estimate corresponds to the minimum value of the sum of χ^2 (in the t_d versus t_{inh} parameter space). One-standard-deviation uncertainty bounds were estimated from the maximum and minimum age limits of the ${\chi^2}_{min}+\Delta{\chi_\nu}^2$ contour in the age-erosion parameter space. ${\chi^2}_{min}$ is the minimum value of the calculated sum of χ^2 within the parameter space and $\Delta{\chi_\nu}^2$ is the critical value of the change in sum of χ^2 for a specified level of confidence and number of fitted parameters (ν) (Table 4) [e.g., Davis, 2002]. For our problem, the appropriate level of confidence is 68.3 percent (corresponding to one standard deviation uncertainty) and two fitted parameters (t_d and t_{inh}), giving a $\Delta{\chi_\nu}^2$ of 2.30. The approach to uncertainty estimation described above is comprehensive (it includes potential systematic as well as random sources of uncertainty) and it includes an explicit calculation of model accuracy, based on the fit of multiple samples within a single profile, as opposed to a single cosmogenic nuclide age determination for which model error can only be estimated. We believe that the overall uncertainty bounds calculated using this approach are conservative and are likely to overestimate, rather than underestimate, the actual uncertainties.

References

Bevington, P.R., and D.K. Robinson (2003), Data reduction and error analysis for the physical sciences, 320 pp., McGraw-Hill, Boston.

Davis, J.C. (2002), Statistics and data analysis in geology, 638 pp., John Wiley and Sons, New York.

Gosse, J.C., and F.M. Phillips (2001), Terrestrial in situ cosmogenic nuclides: theory and application, Quaternary Science Reviews, 20, 1475-1560.

Kurth, G. (2003), Cosmogenic nuclide dating of old, high pluvial shorelines in the western Great Basin, M.S. thesis, XX pp., New Mexico Institute of Mining and Technology, Socorro.

Phillips, F.M., W.D. Stone, and J.T. Fabryka-Martin (2001), An improved approach to calculating low-energy cosmic-ray neutron fluxes near the land/atmosphere interface, Chemical Geology, 175, 689-701.

Stone J. O. H., J.M. Evans, L.K. Fifield, G.L. Allan, and R.G. Cresswell (1998) Cosmogenic chlorine-36 production in calcite by muons, Geochimica et Cosmochimica Acta, 62, 433-454.