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Interactive comment on “Comment on: “Possible source of ancient carbon in phytolith concentrates from harvested grasses” by G. M. Santos et al. (2012)” by L. A. Sullivan and J. F. Parr

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Referee report from Martin J. Hodson

This comment concerns a disagreement between two groups of researchers concerning the analysis of phytoliths for ^{14}C , and then the interpretation of the results of such analyses. The main problem seems to be that phytoliths that are extracted from modern plants are giving C^{14} ages that are considerably older than would be expected, and indeed are older than the bulk carbon analysed from the whole plant. Clearly this creates huge problems in using ^{14}C from phytoliths to date soil samples, even if contamination can be overcome. Santos et al. (2012) put it like this: "These intriguing

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14C results raised the following questions: (a) can a chemical extraction procedure that does not use solvents or plastic devices (possible sources of older exogenous C) affect the final 14C results?; (b) can root uptake be a source of old C that may be recalcitrant to the extraction procedures, and therefore remain on the phytolith concentrates?; and (c) what are the sources of phytC itself: carbon fixed solely via photosynthesis, or taken up by roots, or both?"

The main problem Sullivan and Parr have with Santos et al. (2012) is that they used only two data points out of twelve that Sullivan and Parr (2008) presented in order to substantiate their hypothesis. The two samples that were used came from mature leaves and recently fallen leaves, and showed the "typical" result, with much older dates than would be expected. However, the other ten litter samples that were analysed (but not considered by Santos et al., 2012) were also modern samples, and only 2/3 years old, but these gave much closer to modern dates. One has to agree with Sullivan and Parr that selective use of data in this way to justify a hypothesis is not a good idea. We definitely have a difficult issue here, but we need to look at all the data to resolve it. Parr and Sullivan also raise the problem of over-extracting phytoliths, considering that it might be as big a problem as under-extracting which Santos et al. (2012) criticised. I am sure they have a valid point here. I suspect that eventually we will need a standard method for extracting phytoliths for this kind of work as it seems likely that a lot of the problems stem from the multiplicity of methods being used!

Personally, I do not think that the transport of old carbon from the soil, which then gets preferentially laid down into phytoliths is particularly likely, and I tend to agree with Sullivan and Parr. This mechanism would require the small amount of C that is transported in the xylem to somehow get targeted on cells that are forming phytoliths, and to get included in these structures while the huge amounts of carbon that are absorbed during photosynthesis are excluded. Santos et al. (2012) state that, "Very little is known of the location of any old C within the plant tissues or of the chelation (bonding) of Si to organic compounds in the plant sap and the cells, so it cannot presently be deter-

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mined whether old C is directly involved in phytolith formation." They are, in one sense, correct that little is known here. However, Casey et al. (2003) did look at the xylem sap of wheat plants using NMR spectroscopy. They found only two Si-containing species in the xylem exudate, mono and disilicic acid in a ratio of approximately 7:1, and there was no evidence of organosilicate complexes. This does not rule out the Santos et al. (2012) hypothesis, but it does make it more unlikely, as complexes of "old C" with Si in the xylem could be a method of concentrating this carbon in phytoliths.

Obviously there is a problem here that needs resolution. So what is my guess at what is happening to produce the peculiar results that have been observed for modern phytoliths extracted from plants? I think it may be something to do with the pools of carbon within the plant and within the phytoliths. Phytoliths are of two basic types: those laid down in the lumen; and those deposited onto cell walls. The former will contain mostly lipids and nucleic acids etc., whilst the latter will contain mostly carbohydrate. I worked on the latter many years ago (Hodson et al., 1984), they are much more porous than lumen deposits, and I think they may lose their carbon more easily when in breakdown situations (no data on this point though!). There have been suggestions in the past that fractionation of C isotopes into different organic compounds in plants varies (some are mentioned by Santos et al., 2012). The bulk carbon in many plants will be carbohydrate, and I would hypothesise that when the phytoliths drop into the litter layer that this carbohydrate will be the first to be broken down. I am aware that these observations do not explain all of the results in the literature, but they may give some ideas.

In conclusion, I think Sullivan and Parr were quite right to critique Santos et al. (2012) who seemed to get "carried away" with a hypothesis that does not fit all the facts. But we should not be too critical of Santos et al. as it is a complex problem, and their idea has certainly generated some useful thinking and discussion. I shall look forward to finding out why ^{14}C in phytoliths gives such odd results.

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