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Evidence for old carbon contamination in ^{14}C wiggle-match age series for the 946 CE eruption of Changbaishan volcano

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Abstract. Volcanic eruptions that are not historically attested are commonly radiocarbon dated by "wiggle matching" sequential ^{14}C measurements of the rings of trees killed by the eruption against an accepted calibration curve. It is generally assumed that carbon laid down in the wood is uncontaminated by ^{14}C-free ("old") carbon, although evidence for contamination is well documented. Often, ill-fitting ring ages are excluded from analysis. The 'Millennium Eruption' of Changbaishan volcano on the China-DPR Korea border offers a valuable case study in wiggle match dating, since several independent groups reported age estimates before the determination and acceptance of a precise eruption year of 946 CE. Some of the discrepancies and incompatibilities between published dates were attributed to old carbon effects. Here, we apply a new methodology to correct for contamination levels of up to 4.5% old carbon to eight wiggle match date series for the Millennium Eruption. Without discarding ring ages, we find agreement indices as high as, or higher than, those for the published dates, and five of the eight date series yielded high-agreement-index eruption dates closer to 946 CE than the published dates. None of the five yield a best result at zero contamination. Differences between the eruption dates reveal a weak association with the direction of the sampled tree from the caldera, but no relationship with distance. Our results suggest that old carbon contamination is possible over a wide area, potentially leading to over-estimation of eruption ages by years, decades or more, cautioning against over-reliance on wiggle-match ages that are not corroborated by other lines of evidence. Our revised protocol that accounts for contamination offers a way forward in the application of wiggle match dating of eruptions and provides a platform for understanding discrepancies that exist when comparing wiggle match series.

1 Introduction

Understanding potential relationships between volcanic eruptions, climate, and human history relies on accurate chronologies (Bűntgen and Oppenheimer, 2020). Each phenomenon has its own dating issues, but even events as singular and noteworthy as major volcanic eruptions can leave only vague and often contradictory records in written histories (Scarth, 2009). Dates for eruptions that are not historically attested by a literate society are rarely known to a decade or century, let alone a calendar year. However, decadal or better accuracy dating of eruptions assumes great significance for events such as the Bronze Age eruption of Thera in the eastern Mediterranean (Friedrich et al., 2006; Pearson et al., 2018; Pearson et al., 2020; Friedrich et al., 2020) or the Tierra Blanca Joven eruption of Ilopango in Central America (Dull et al., 2019; Smith et al., 2020), which both occurred close to a literate society that may have been directly or indirectly affected.

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For the past sixty years, there has been widespread use of radiocarbon to establish eruption dates, with increasing accuracy as the technologies developed, more refined calibration curves promulgated (Hogg et al., 2020; Reimer et al., 2020), and sophistication of statistical analyses (Crema and Bevan, 2020). Wiggle match (WM) dating – the comparison of radiocarbon age series for ring sequences of trees killed by the eruption to patterns in radiocarbon calibration curves – has been applied where possible, as the most likely to give a calendar date (Galimberti et al., 2004). Some eruptions, such as the 'Millennium Eruption' of Changbaishan on the China-DPR Korea border, have been wiggle-match dated many times (e.g., Sun et al., 2014; Xu et al., 2013; Yin et al., 2012). The Thera (Friedrich et al., 2020; Pearson et al., 2018) and Ilopango (Dull et al., 2019; Smith et al., 2020) eruptions have been subject to recent wiggle match analyses. That of Thera, at least, is still controversial, with conflicting views on matters such as applicability of calibration curves, choice of samples, and possibility of contamination of dated samples by extraneous carbon (Cherubini et al., 2014; Manning & Kromer, 2012; Pearson et al., 2018).

Few of these wiggle match analyses have accepted, or even considered the possibility of contamination of the dated wood by ^{14}C-free ("old") geogenic carbon, whose presence could make the measured ages up to several centuries too old (Grootes et al., 1989a, b; Soter, 2011). The assumption – and strongly stated declaration (Manning & Kromer, 2012) – that contamination by old carbon is impossible has been maintained despite evidence for volcano flank degassing of magmatic carbon dioxide over significant areas dating back fifty years (e.g., Aiuppa et al., 2006; Allard et al., 1991; Chatters et al., 1969; Sulerzhitsky, 1971). Outgassing has been recorded as deviations from atmospheric in ^{14}C measurements on leaves (Chatters et al., 1969; Pasquier-Cardan et al., 1999) and in tree ring sequences (Bergfeld et al., 2010; Cook et al., 2001).

Contamination of eruption wiggle-match dates by magmatic ^{14}C-depleted carbon is controversial (Hogg et al., 2019; Holdaway et al., 2018; Holdaway et al., 2019). However, contamination has been suggested at Changbaishan to explain and justify removal of ^{14}C rings with low statistical support in the WM methodology (Sun et al., 2014; Xu et al., 2013). At present, WM analyses are performed without regard to possible contamination of the wood samples by ^{14}C-depleted carbon when they were laid down, assuming tacitly that the contamination term $\varphi = 0$ in the equation (Soter, 2011)

$$\Delta t = -\tau \ln(1-\varphi)$$

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where Δt is the offset to the conventional radiocarbon age (in years), and τ = 8033 years (conventional mean lifetime used in ^{14}C dating, i.e. the half-life of ^{14}C divided by ln(2)). A fraction φ = 1% of old carbon results in an apparent age increment Δt of c. 80 years.

The large (VEI-7) late First Millennium CE eruption of Changbaishan volcano (also known as Tianchi, Baitoushan, Baegdusan, Paektusan, and Mt Paektu) on the China-DPR Korea border (Fig. 1), and source of the widespread Baegdusan-Tomakomai (B-Tm ash) (Yatsuzuka et al., 2010), has been dated by radiocarbon many times, with varying results (Fig. 2). Conventional single sample ages provided eruption dates from 550 to 1150 CE (modal date 1000 CE) (Liu et al., 1998). Extensive WM dating of the eruption using tree-ring sequences that provide *a priori* information on temporal relationships of ^{14}C measurement, are summarised by Xu et al. (2013), Yin et al. (2012), and Oppenheimer et al. (2017), and have yielded calendar dates between 859 and 984 CE (Fig. 2).

One of the youngest dates for the eruption arises from re-calculation of a WM series in Wei et al. (2007) (Fig. 2), which was not, as stated, the result of a standard wiggle match protocol. The date of 1027 CE in Wei et al. (2007) corresponds to the highest peak in the probability distribution for the single sample C, which was taken from rings laid down thirty years before tree death. Therefore, based on their methodology, the eruption actually took place in 1057 CE (Fig. 2). Our WM analysis using OxCal 4.4 (Ramsey, 2009) and the IntCal13 curve (Reimer et al., 2013) as used by Wei et al. (2007) yielded an eruption date probability distribution peaking at 1196 CE (95.4% confidence interval 1156 to 1209 CE).

Thanks to recognition of the cosmogenic ^{14}C event of 774 CE (Büntgen et al., 2018; Miyake et al., 2012) in subfossil trees killed by the Millennium Eruption, and sub-seasonal resolution on the sulphur deposition record in Greenland ice cores, the eruption is now securely dated by to late 946 CE (Oppenheimer et al., 2017; Hakozaki et al., 2018). A historical chronicle from Japan recording ash fallout on 3 November 946 CE hints that the eruption may have occurred within 24 hours prior to that observation, allowing for transport time of the ash cloud (Oppenheimer et al., 2017).

Half of the radiometric ^{14}C dates listed by Sun et al. (2014) in their Table 3 are, however, older than the cosmogenically-tuned date. Such deviations could result from at least three factors: first, an eruption might be preceded by fumarole activity and diffuse degassing, with significant venting of volcanic gases including H_2S, SO_2, and CO_2, leading to the pre-eruption death of vegetation including trees (Farrar et al., 1995; de Jong, 1998); trees that died before the eruption from other causes can also be preserved in pyroclastic deposits.

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Second, pumice fallout and high temperature pyroclastic flows can strip outer rings from trees, with the last rings then not reflecting the real date of the eruption. Conversely, carbonised wood with intact bark may be protected from pumice fallout and not suffer the same severity of impact from pyroclastic flows (Yatsuzuka et al., 2010). Finally, diffusive outgassing of magmatic-hydrothermal carbon could release large quantities of ^{14}C-free CO_2, altering the regional ^{14}C atmospheric titre (Beavan-Athfield et al., 2001; Bruns et al., 1980; D'Arcy et al., 2019; Pasquier-Cardin et al., 1999; Saupé et al., 1980; Sulerzhitzky, 1971; Tortini et al., 2017). Carbon and sulphur can enter leaves and become fixed in the cellulose of the tree (McCarroll and Loader, 2004). Tree ring studies show that both magmatic carbon and sulphur influence the ^{12}C, ^{13}C and ^{14}C proportions within tree rings in volcanic areas (D'Arcy et al., 2019). Additionally, biospheric sources can contribute old carbon (Grootes et al., 1989a; Soter, 2011).

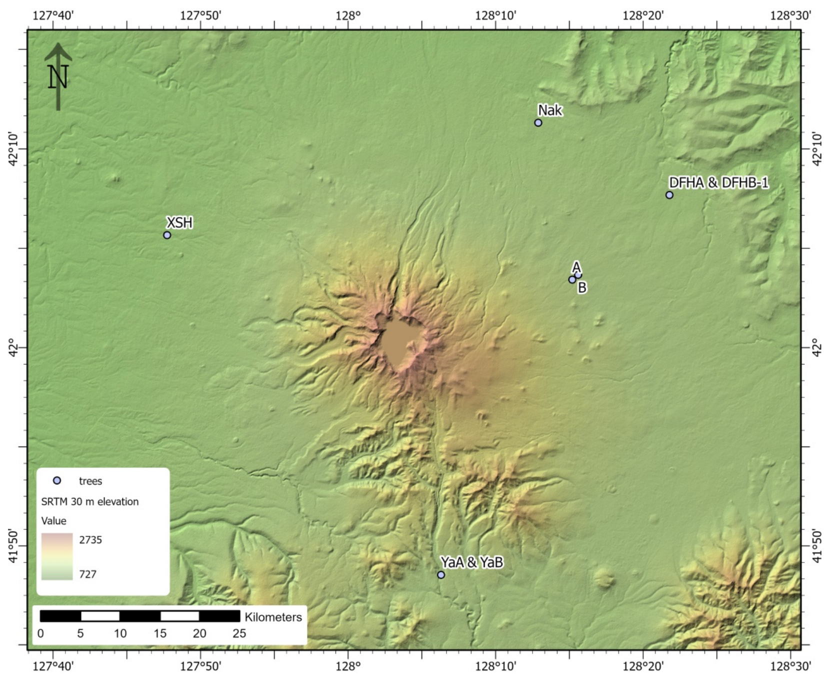

Figure 1: Location of trees with wiggle match date series used in this study. Data for trees from XSH (Xu, et al. 2013); Nak (Nakamura, et al. 2007a); DFHA, DFHB-1, YaA, YAB (Yin, et al. 2012); A,B (Yatsuzuka, et al. 2010).

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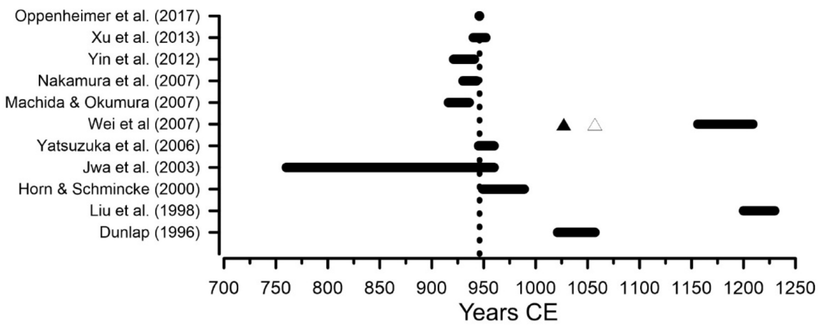

Figure 2: Confidence intervals (95.4%) for wiggle match dates for the Changbaishan Millennium eruption, in relation to the 946 CE date for the eruption (vertical broken line) anchored by the 74 CE cosmogenic event (Oppenheimer et al., 2017). Wiggle match series as summarised by Sun et al. (2014) plus that of Wei et al. (2007). Wiggle match series from Xu et al. (2013) (1), Yin et al. (2012) (4), Yatsuzuka et al. (2010) (2), and Nakamura et al. (2007a) (1) discussed here. Black triangle, published date; open triangle, published date adjusted for ring count to tree periphery, both from Wei et al. (2007).

Diffuse release of geogenic carbon dioxide through soils and pyroclastic deposits is a widespread phenomenon on volcanoes with active magmatic-hydrothermal systems (Allard et al., 1991; Sun et al., 2018; Williams-Jones et al., 2000; Zhang et al., 2015a), diluting regional atmospheric ^{14}C, and raising atmospheric CO_2 abundance, particularly within forest canopies (Grootes et al., 1989a, 1989b; Soter, 2011). Both factors would affect the ^{14}C content of dated wood, and may explain the 'old' WM dates for the Millennium Eruption.

The factors leading to WM series giving eruption dates significantly younger than the cosmogenically attested date (Fig. 2) are more difficult to understand. The most likely might be that "young carbon", such as younger soil humic acids, was bonded so tightly to the dated wood samples that the standard pre-treatments failed to remove them, or that recent exposure allowed bacterial infection and introduction of "young" carbon via chitin not removed during cellulose extraction (Krüger et al., 2014).

The secure dating of the Millennium Eruption to a calendar year allows an examination of the accuracy of the various WM date series applied so far to the eruption and the potential contribution of "old" carbon from biospheric sources or infinite age carbon from magmatic sources. Changbaishan is known to generate diffuse emission of magmatic carbon (Zhang et al., 2015a; Zhang et al., 2015b), and, soil gas concentration of CO_2 on the western slopes of Changbaishan have reportedly reached 500-1000 ppm (Zhang et al., 2015a), indicating the potential for bias in reported radiocarbon dates of the Millennium Eruption. The magma body extends beyond the edifice (Kyong-Song et al., 2016; Kim et al., 2017; Hammond et al., 2020), particularly to the north, which could fuel diffuse degassing at some distance from the summit.

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Here, we test the hypothesis that non-equilibrium, magmatic carbon or biogenic carbon, or both, incorporated in wood can affect WM ^{14}C age series. We do this by identifying alternative high agreement index fits for each of three WM series for the Millennium Eruption at low (0–5%) levels of constant (over the life of the tree) old carbon contamination, the low end of the range identified by Sulerzhitzky (1971) for north-eastern Asia. In doing so, we present a radiocarbon methodology that (i) allows for the possibility of contamination, and (ii) is systematic and robust in its treatment of age measurements with low agreement indices. It therefore avoids the Procrustean method of rejecting WM ages in an effort to force a fit between the WM series and the chosen (φ = 0%) section on the calibration curve.

2 Material and methods

For our analyses we used the WM series for trees YaA and YAB (Yin et al., 2012) and XSH (Xu et al., 2013) as exemplars. We calculated tree death dates from these WM series using the OxCal4.3.2 (Ramsey, 2009; Ramsey, 2017), D-sequence option (Ramsey et al., 2001) and the IntCal13 calibration curve (Reimer et al., 2013) for comparison with the original analyses. For each of these exemplars, we investigated the presence of alternative fits with high Oxford agreement indices (A, A_{comb}, A_{model}, $A_{overall}$, as below) (Ramsey, 1995, 2001; Ramsey et al., 2004) for the WM date series, allowing non-zero values of φ. We used the Oxford agreement indices as an alternative to outlier analysis.. These are: A, individual agreement indices, which are useful for identifying which samples do not agree with the model (values should be > 60%); A_{comb}, which tests to see if distributions can be combined (the acceptable threshold depends on the number of ages *n* in the wiggle series, i.e. $1/\sqrt{(2n)}$; A_{model}, tests if the model is likely as a whole, given the data (the value should be > 60%). Finally, $A_{overall}$, individual agreement index, is the product of the individual agreement indices (the value should be >60%) –. We also investigated the effects on predicted eruption dates of removing "low A age" samples, i.e. the ages of ring sequences with non-significant values (<60%) of the A agreement indices in OxCal4.3.2 (with φ = 0-4.5%) as done by, for example, Xu et al. (2013).

Again for direct comparison, we followed Xu et al. (2013) in including a model incorporating a regional offset of ± 10 years, using the Delta_R option in OxCal4.3.2. For the purposes of this initial study, contamination was simulated by adding increments in Δt individually, but calibration algorithms could be modified to generate the alternative fits and parameters for any desired range of the φ term.

We repeated the analyses, but without removing "low A age" samples (as above), for WM sequences for trees DFHA and DFHB-1 (Yin, et al. 2012), trees A and B (Yatsuzuka et al., 2010) and the tree analysed by

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Nakamura et al. (2007a). As a further measure of the integrity of each WM fit, we logged the number of ring ages that did not reach the critical A value of 60% (for $A_{overall}$ and A_{model}) equivalent to a χ^2 test at 95% level for a combination of normal distributions) and A_{comb} ($1/\sqrt{n}$).

To obtain an eruption date estimate using all the WM data, we repeated the analyses for all trees using the IntCal20 calibration curve (Reimer et al., 2020) to provide the most recent WM estimates for the eruption date. Finally, we summed the probability distributions for the dates corresponding to the peak A index values.

Locations of these trees relative to the Changbaishan caldera when they were sampled are shown in Fig. 1. As some of the trees may have been transported significant distances from their growth positions entrained in pyroclastic currents or mudflows, we reviewed the published descriptions of the logs as found. Tree XSH (Xu et al. 2013) was likely preserved *in situ* as it was lying horizontally, with only the upper surface slightly carbonised; the lower surface was intact, with bark attached. Two of the four trees sampled by Yin et al. (2012), those at the Yengshan site, were buried together, one broken off but in growth position, and the other horizontal: both had "perfect bark" and were unlikely to have been transported any distance. Both trees (DFHA, DFHB-1) from the Dongfanghong site were removed from an array of logs exposed in a section through the ignimbrite and could have been transported some distance.

None of the descriptions precluded the possibility that a tree had been killed by root suffocation by high soil CO_2 levels (Gerlach et al., 2001; Rogie et al., 2001; Lewicki et al., 2007) or by the effects of other volcanic gases (de Jong 1998) in the period preceding the eruption. Notwithstanding this possibility, Yin et al. (2012) concluded that the Hengshan "…trees were burned to death *in situ*".

To summarise, we modified the standard protocol for analysis and interpretation of WM age series (Fig. 3A), taking into account the potential for old carbon contamination of the wood samples ($\varphi \neq 0$), and the importance of independent correlative dating (Fig. 3B).

We plotted the mean tree death (assumed to be coeval with the eruption) dates and the probability distributions of those dates for each WM series at $\varphi = 0\%$ and at the φ value corresponding to the first peak in A_{comb} with $\varphi > 0\%$. For each of the three exemplar trees, we plotted A_{comb} values for wiggle fits corresponding to values of φ between 0 and 4.5%, and dates of tree death versus those A_{comb} values, with and without regional offset modelling, using the means (with standard deviation) of regional offsets (Fig. 4).

For all trees considered, the differences between eruption dates corresponding to $\varphi = 0\%$ and φ at the first A_{comb} peak thereafter were assessed by plotting the mean eruption date and the date probability for each series in relation to both the normalised A_{comb} value for the series and the date for the eruption anchored by the

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cosmogenic event (Fig. 5). A_{comb} values were normalised as quotients against the actual critical value because A_{comb} varies with sample size.

Possible geographic effects on tree WM sequence ages were assessed by plotting the φ = 0% and first A_{comb} peak dates (as differences from the date anchored by the cosmogenic event) against distance and bearing from the centre of the caldera (Fig. 6).

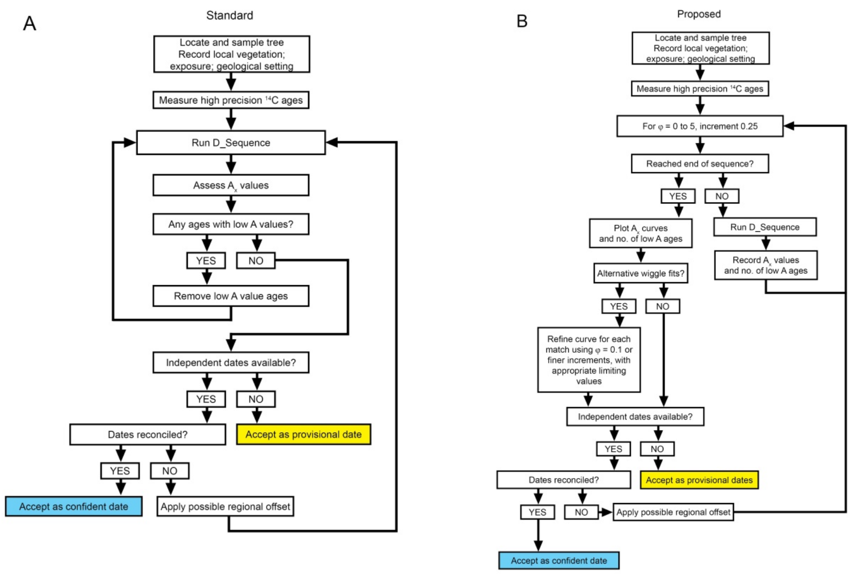

Figure 3: Decision trees for analysis of wiggle match radiocarbon age series assuming (A) no contamination by old carbon (φ = 0), and (B), allowing low levels of old carbon (geologic and/or biospheric carbon) (φ ≠0). Based on use of OxCal 4.4 D-Sequence option (Ramsey 2009).

3 Results

The WM series from tree YaA yielded multiple significant A_{comb} peak fits at φ values of 0-4% (Fig. 4A-C). In the absence of external evidence for the eruption date provided by the cosmogenic event in 774 CE, any of the highest agreement index peaks, including that at φ = 1% (eruption c. 1025 CE, Fig. 4D-F) for which the A_{comb} value was higher than at φ = 0%, could be argued to be the eruption date. Even improving the fit for φ = 0% by removing the low A ring ages did not give an A_{comb} value for the fit at φ = 0% as high as that for φ ≈ 1% (Fig. 4A).

For tree YAB, there were two peaks in A_{comb}, with the higher (with all low A ring dates removed) again at c. φ = 1% (Fig. 4B). However, with all ring dates included, the highest peak was at φ = 0.25%, with a marginally better fit than for φ = 0% (Fig. 4B).

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Tree XSH yielded a single, strong A_{comb} peak at φ = 0.25% (Fig. 4C), corresponding to an eruption date probability encompassing 946 CE (Fig. 4B).

Five of the WM series, each including all ring ages, showed an improved fit (in terms of proximity to the cosmogenically tuned date with φ ≠ 0% (Fig. 5). WM series for the first post φ = 0% A_{comb} peaks for trees A (Yatsuzuka et al., 2010), YaA and YAB (Yin et al., 2012) were much later, in the 11th century CE, where they agreed with the (incorrect) date range reported by Dunlap (1996)(Fig. 2). If these had been the only WM dates, they would have been accepted, if only provisionally. None yielded an eruption date as young as that proposed by Liu et al. (1998).

Repeating the analyses using the IntCal20 curve (Reimer et al., 2020) in OxCal 4.4 yielded similar results to the IntCal13 analyses, with all but one tree (XSH) having multiple fits at different levels of contamination (Fig. 7; Supplementary Figures 1-8). None of the trees, including the XSH tree, had A index peaks at φ = 0%, the closest being, again, tree XSH (Fig. 7H) (at φ = 0.25 %). The XSH tree WM series also had a near rectilinear relationship between A indices and the number of low A rings, in a near perfect WM result.

Combining the results from all the WM analyses yielded a highest summed probability distribution for the dates corresponding to the A index peaks that centred on the cosmogenically attested date (Fig. 8). Both the alternative WM dates had much lower probabilities in comparison.

Although eruption dates for the eight WM series appeared to decline with distance from the caldera (Fig. 6A), the relationship was not significant (P_{uncorr} = 0.238; $P_{permutation}$ = 0.2409). All the trees were within 30 km of the caldera, within the 50 km at which "younging" of radiocarbon ages was observed for the Taupo First Millennium eruption (Holdaway et al., 2018, 2019). In contrast, eruption dates corresponding to A_{comb} peaks with φ ≠ 0 approached the CTD closely (Fig. 6B). All were excellent estimators of the cosmogenically affirmed ("actual") date (Fig. 5B, 6B), and there was no relationship to their distance from the caldera.

Two of the three trees whose φ ≠ 0 A_{comb} peak eruption dates were c. 100 years younger than the actual date were almost due south of the caldera and close to each other (Fig. 1, 6). The third was east of the vent, close to a tree whose eruption date was closer to the actual date at the first φ ≠ 0 A_{comb} peak (Fig. 6C, 6D). The trees with better agreement with the actual date were all either WNW (tree XSH) or ENE of the caldera (Fig. 6D).

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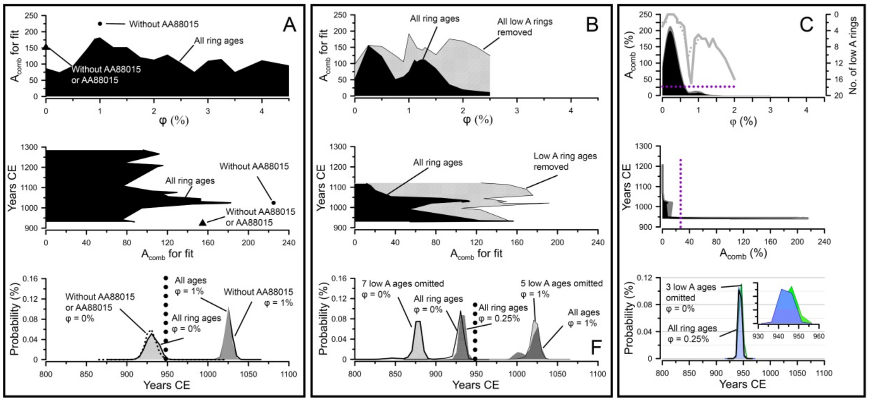

Figure 4: A_{comb} values for wiggle match radiocarbon age series for trees (A) YaA and (B) YAB (Yin et al., 2012), and (C) for tree XSH (Xu et al., 2013) for φ = 0-4.5%, with A_{comb} values for series with low A value ages removed as in the original publications, and the number of low A rings for each φ value. In each panel: upper, A_{comb} values for wiggle match fit at φ = 0-4.5%; middle, mean calendar eruption dates corresponding to A_{comb} values for the range of φ values; bottom, probability distributions for eruption dates corresponding to conditions as shown.

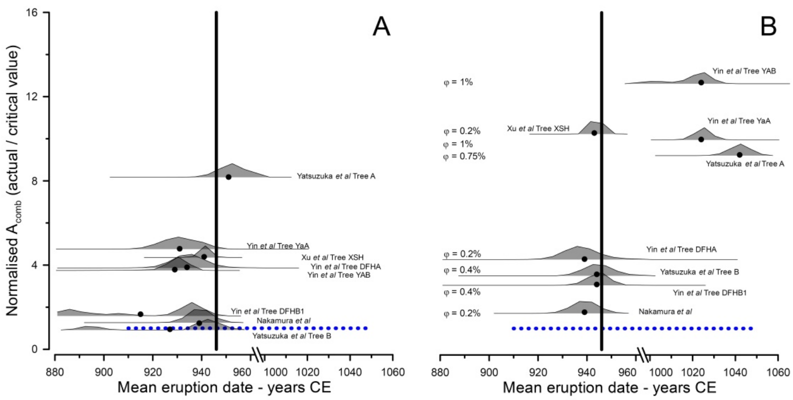

Figure 5: Comparison of eruption date probability distributions for (A) φ = 0%, and (B) φ values for first post-0% peak agreement wiggle match fit. Solid circles, mean eruption date; dotted blue line, critical value of A_{comb} for each sample size (normalised); vertical line, eruption date validated by cosmic ray spike record in the XSH tree.

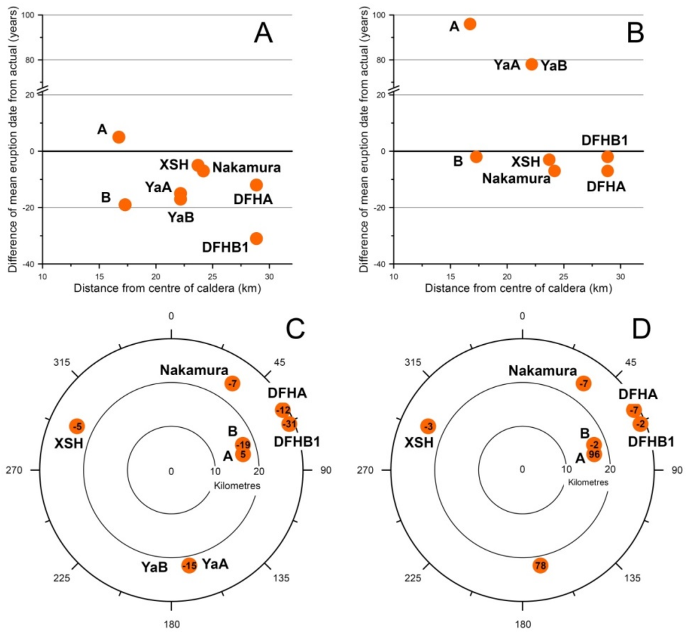

Figure 6: Spatial relationships of wiggle match trees to the caldera. **A**, **B**, difference between wiggle match date and CTD versus distance from caldera: **A**, φ = 0%; **B**, first A_{comb} spike, φ ≠ 0%. **C**, **D**, as for A and B but arrayed as bearing and distance: **C**, φ = 0%; **D**, first A_{comb} spike, φ ≠ 0%.

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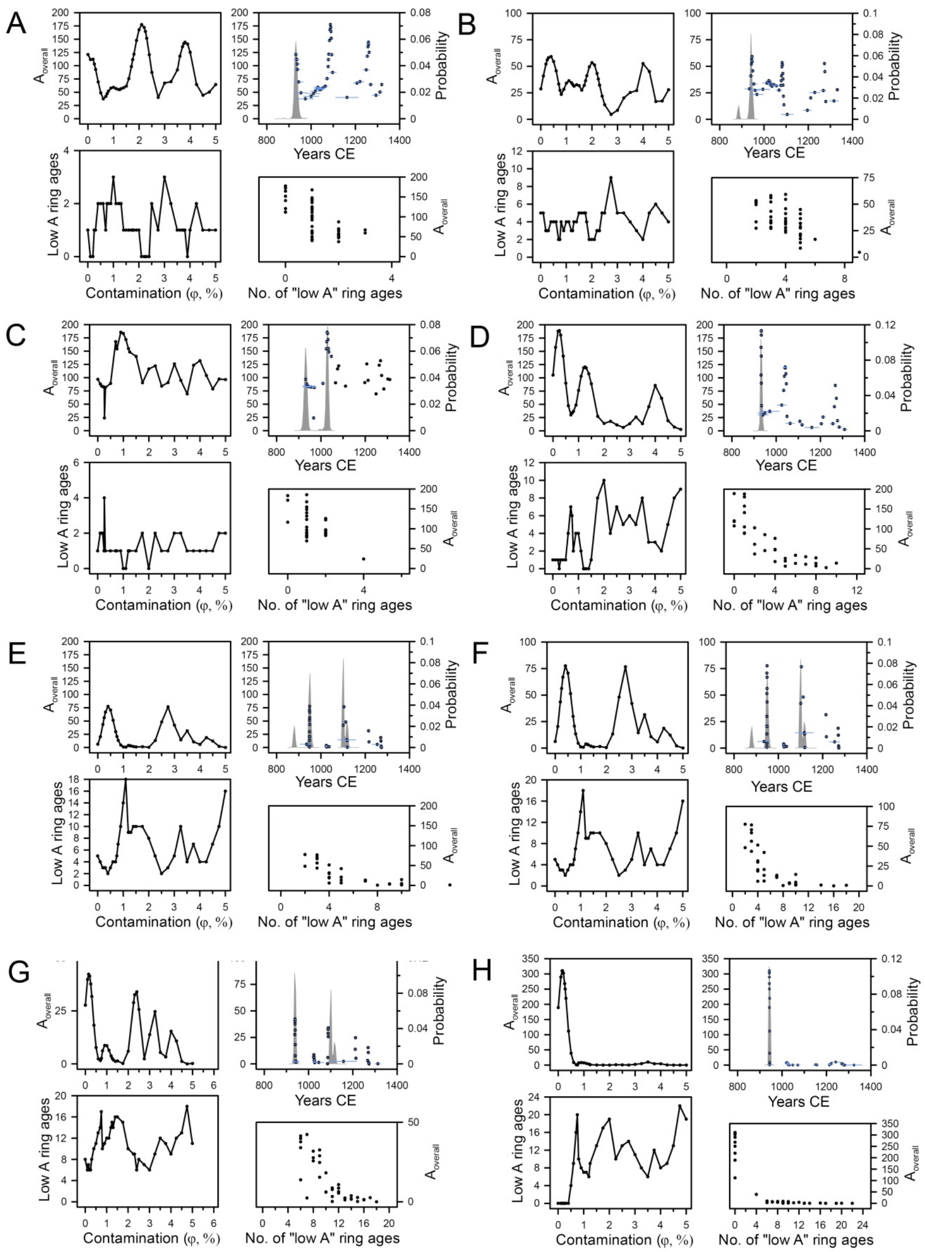

Figure 7: Relationships between levels of contamination in wiggle match wood samples and Agreement Indices (here $A_{overall}$) for the wiggle match series dating the Changbaishan eruption. **A**, Tree DFHA: **B**, Tree DFHB1; **C**, Tree YaA; **D**, Tree YAB; **E**, Tree A; **F**, Tree B; **G**, Tree Nakamura; **H**, Tree XSH.

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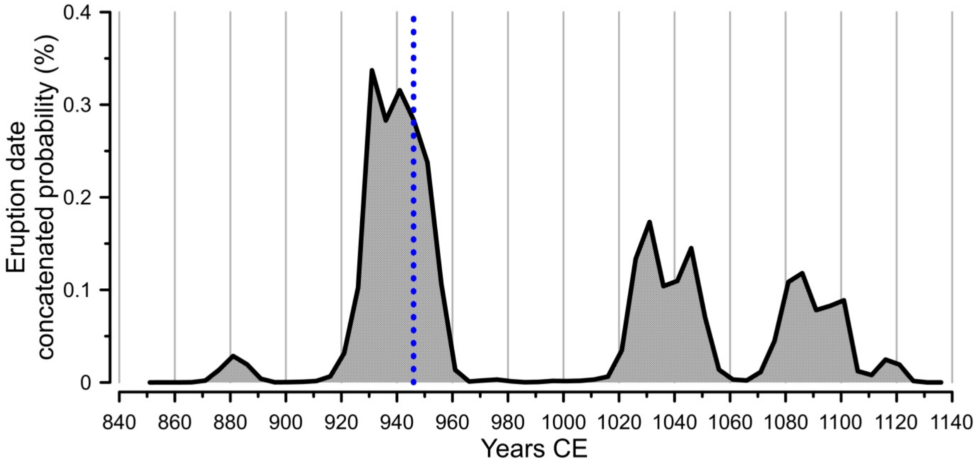

Figure 8: Summed probabilities for eruption dates corresponding to A index peaks of wiggle matches for trees in Fig. 7-14, using the IntCal20 calibration curve. The higher probability distribution around the eruption date of 946 CE allows the dates for other wiggle match peaks (1030 - 1050 CE and 1080 - 1100 CE) to be discarded. Blue dotted line, date of the Changbaishan Millennium Eruption referenced to the 774 CE cosmogenic event.

4 Discussion

Seven out of the eight wiggle match results using traditional wiggle matching techniques at Changbaishan yielded eruption dates older than the cosmogenically derived date. Our results confirm the possibility that small amounts of infinite age carbon contamination in five of the eight WM radiocarbon age series for the Changbaishan eruption can explain the errors. Constant levels of $\varphi \neq 0$ contamination can improve the A_{comb} value of the WM fit and move the WM eruption date closer to the cosmogenically constrained eruption date, while simultaneously removing the need to discard ages on specific rings or ring series. For the other three trees, the application of constant $\varphi \neq 0$ contamination improved the A values but did not move the wiggle match closer to the known age.

The worsening of the eruption date estimate for tree A (Yatsuzuka et al., 2010) at the $\varphi \neq 0$ peak is predictable as it already yielded a younger date than the known eruption date, therefore removal of contamination would only drive it to even younger ages. Bacterial or fungal infection of the exposed log may have introduced "young" carbon to trees YaA and YaB (Yin et al., 2012), generating low A_{comb} values for ring series close to the accepted date. Contamination by old or young carbon may have varied through time. The application of a constant contamination could not improve the wiggle match fit.

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The Nakamura et al. (2007a) trees, farther away and generally upwind of the caldera showed little change with different levels of contamination, which is consistent with their being exposed to, and photosynthesising , only small amounts of old carbon (Fig. 4). The trees to the ENE of the caldera seemed to have taken up consistent amounts of old carbon, whereas those to the south photosynthesised varying amounts of old carbon before they were killed.

A “significant” wiggle match that assumes $\varphi = 0\%$ is not sufficient evidence to secure an eruption date when there are other potential fits to the calibration curve for the wiggle match series ages which also yield significant A values with $\varphi \neq 0\%$. Where such alternative fits exist, supporting evidence such as a direct dendrochronological date for a cosmic ray event or a geochemically-identified tephra in a securely dated ice core is needed. Indeed, if the first WM ages (Fig. 2) were the only ones available (as they were for some time), the data allowing for just 1% of old carbon contamination would have supported an eruption date in the 11th century CE. For example, an earlier wiggle match date of 938 +8/-5 CE, had been favoured from external evidence from dendrochronology (937–938 CE) and varves (912–972 CE) (Nakamura et al., 2007a) until further wiggle match series were measured (Xu et al., 2013). Finally, the 774 CE cosmogenic event was identified (Miyake et al., 2012) and its signature was identified (Oppenheimer et al., 2017) in the tree analysed by Xu et al. (2013) and its presence corroborated (Hakozaki et al., 2018).

Sulerzhitzky (1971) suggested that contamination by geologic carbon is widespread in radiocarbon-based age measurements for volcanic eruptions in north-eastern Asia. D’Arcy et al. (2019) showed decadal $\delta^{13}C$ shifts of up to 1.9‰ are possible from magmatic contamination associated with the onset of volcanic crises and degassing events. Chatters et al. (1969) recorded levels of geologic carbon contamination in vegetation samples from the Big Island, Hawaii, near and remote from known centres of outgassing. The minimum ‘old’ carbon component in 1967 was 1.5% and 1% in 1968, with maxima (near vents) >90% in 1967 and > 50% in 1968. Excluding values >20%, mean levels of ‘old’ carbon were 5.34% (c. 432 years ‘inbuilt age’) in both years. Their estimates of ‘built-in’ age in vegetation varied from 81 years to 6595 years: one site 32 km downwind from an obvious source (Sulfur Banks) contained 9% geologic carbon, equivalent to a ‘built-in’ age of 758 years (Chatters et al., 1969).

A limitation of our revised methodology is that it assumes a consistent level of contamination throughout the life of the tree. This may not be appropriate for smaller volume, more frequent eruptions (D’Arcy et al., 2019) or with episodic unrest, degassing crises, and hydrological and meteorological modulation of CO_2

flux (Farrar et al., 1995; McGee and Gerlach, 1998; Gerlach et al., 1999; Cook et al., 2001; Rogie et al., 2001; Evans et al., 2002; Lewicki et al., 2007; Cawse-Nicholson et al., 2018). An event on the scale of the Millennium Eruption of Changbaishan may also be preceded by years/decades/centuries of elevated carbon dioxide outgassing. However, more contamination may be experienced in the final few months or years of WM ages before the eruption if it is triggered by rapid reactivation of an already assembled magma body (e.g., Sparks et al., 1977; Pallister et al., 1992; Martin et al., 2008) rather than centuries of elevated carbon dioxide degassing from the accumulation of a large magma body.

Conversely, WM analyses, including those pertaining to the Millennium Eruption, have benefited from advances in radiocarbon analyses, particularly the advent of accelerator mass spectrometry, which significantly reduces measurement errors and allows analysis of much smaller samples, and with the advent of miniaturise AMS systems such as MICADAS® (ETH Zurich), much longer series of age measurements. Longer WM series, of more precise ^{14}C measurements, preferably from logs with clear, unambiguous growth rings, possibly linked to high quality dendrochronological series, raise the probability of an eruption date as accurate as the method permits (e.g., Xu et al., 2013). However, we suggest that dense series (with fewer rings included in each sample) may cause problems until even more date-rich calibration curves than the IntCal20 and SHCal20 become available.

The usual response has been to discard – or downgrade – earlier WM dates as new (ostensibly better) series matches are obtained. However, if a new series includes contamination by old carbon from any source, then it too is flawed, whatever its other advantages. As noted above, where multiple WM trees are available, potentially associated with different levels of contamination, external evidence such as a securely dated cosmic ray event is required to support a given significant wiggle match. For example, an eruption date consistent with 1% contamination could have been accepted for a date matching other evidence if the cosmic ray event had not been identified.

The geologic CO_2 environment in mainland north-eastern Asia is unlikely to be unique. A pervasive old carbon contamination in Japan has been attributed to movement of air masses (Nakamura et al., 2013) and degassing of CO_2 from nearby oceanic water (the “island effect”) (Nakamura et al., 2007a), although local geologic sources were also acknowledged (Sakamoto et al., 2003; Yatsuzuka et al., 2010). Indeed, there is now abundant evidence for diffuse emission of carbon dioxide in volcanic terrain (e.g., Bai et al., 2017; Bloomberg et al., 2014; Chatters et al., 1969; Chiodini et al., 2004; Chiodini et al., 2000; Frondini et al., 2008; Hernández Perez et al., 2003; Salazar et al., 2001). Yatsuzuka et al. (2010) not only specifically invoked geologic CO_2 as

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biasing radiocarbon ages within a tree but also suggested that trees can be killed by volcanic gases (SO_2, H_2S) (de Jong, 1998) and CO_2 (Farrar et al., 1995) (perhaps years or even decades?) *before* the emplacement of pyroclastic flows that are routinely taken to indicate the date of tree death. A further complication is that stomata can be blocked by high levels of SO_2, restricting CO_2 entry and potentially masking contamination (D'Arcy et al., 2019).

In extreme situations, trees could be killed standing by high soil CO_2 generated by degassing during volcanic unrest, for example as observed at Mammoth Mountain (Farrar et al., 1995; Gerlach et al., 2001). If those trees were then overwhelmed and transported by pyroclastic currents, their rings would record the date of tree death – and the CO_2 event – perhaps decades or longer before eruption (recognising that only a fraction of episodes of unrest culminate in eruption on these timescales). The issue would not arise for trees sampled from a context such as their being in a preserved forest, which on other evidence was a functioning ecosystem when struck by volcanic shock waves and the pyroclastic flow (Hogg et al., 2012; Clarkson et al., 1988; Clarkson et al., 1992; Clarkson et al., 1995).

Hakozaki et al. (2018) and Oppenheimer et al. (2017) emphasise the importance of external evidence in assessing the accuracy of wiggle match date fits. For the Changbaishan eruption the external evidence of the dendrochronologically dated cosmogenic ^{14}C spike allows both the annual dating of the eruption and the demonstration of old carbon contamination of the wiggle match date series. Alternative fits of wiggle match series at non-zero levels of contamination should be considered in future. It is no longer possible to ignore potentially significant levels of contamination of tree samples by ^{14}C-free magmatic carbon dioxide as well as other terrestrial sources.

We suggest therefore that the present protocol for wiggle match dating of eruptions that assumes $\varphi = 0$ (Fig. 3A) be amended to take into account the possibility (probability?) of old carbon contamination of the wood samples ($\varphi \neq 0$), and the importance of independent correlative dating, as shown in Fig. 3B.

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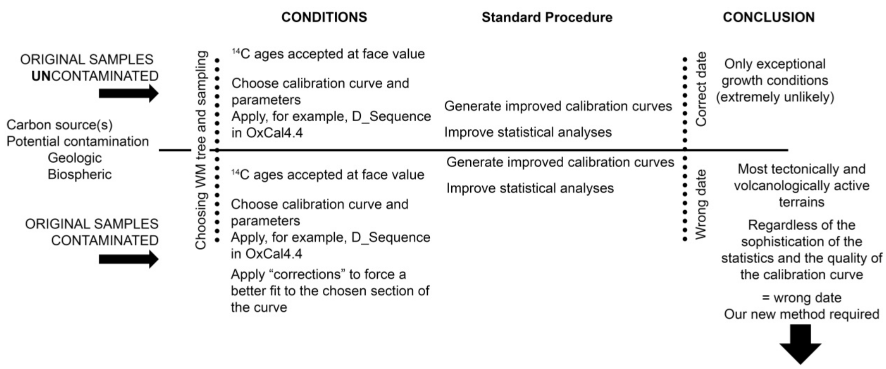

Figure 9: Breakdown of present sources and results for wiggle match dating of eruptions.

5 Conclusions

Our conclusions on the present procedure where the contamination term φ is taken automatically to be 0% are summarised in Fig. 9. It has been known for many years that carbon in vegetation, including trees, can be contaminated by magmatic carbon (Chatters et al., 1969; D'Arcy et al., 2019; Sulerzhitzky, 1971) and this mechanism has been inferred to explain anomalous ring ages and divergence of wiggle match dates at Changbaishan volcano (Xu et al., 2013; Yatsuzuka et al., 2010). We tested a methodology for establishing carbon contamination in a series of wiggle match trees used to date the Millennium eruption of Changbaishan volcano (Fig. 3). We have shown that through systematic wiggle matching with the contamination term φ ≠ 0% allows (1) improvement in the fit agreement A_{comb} parameter of the age series, (2) better agreement of individual wiggle match dates with the cosmogenically constrained eruption date for five out of the eight trees at different locations relative to the caldera, and (3) a reduction in the number of ring age measurements that need to be discarded to achieve a good fit for most wiggle match series. The trees that have systematic contamination signatures could be explained by their proximity to volcano, and downwind location. We note that all trees cannot provide a wiggle match eruption date close to the cosmogenically constrained eruption date even with our new methodology and suggest that this may result from non-constant levels of old carbon contamination, associated with proximity to locally variable CO_2 emissions.

Another cosmogenic event, in 993 CE, has been recognised (Miyake et al., 2014). That event, along with others could be useful for anchoring WM ages for other eruptions (Büntgen et al., 2017, 2018; Oppenheimer et al., 2017) ultimately enabling further testing of our hypothesis. We note that a parallel development is the improved temporal and spatial resolution of radiocarbon calibration curves (e.g., Pearson et al., 2018; Reinig et al., 2019; Friedrich et al., 2020; Reimer, et al. 2020).

6 Code availability

No code was used.

7 Data availability

All data are included in the cited references.

8 Supplement link

9 Author contributions

RNH, BD, and BK conceived the project. RNH performed the analyses and drafted the figures. All co-authors contributed to determining the content and final forms of the figures. RNH prepared the manuscript with contributions from all co-authors.

10 Competing interests

The authors declare that they have no conflict of interest.

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