



OPEN Recurrence rates of explosive volcanism in paleo-equatorial Pangaea, and implications for climate near the peak late Paleozoic ice age

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The continental Brousse-Broquiès basin (Massif Central, France) preserves a well-exposed section of upper Carboniferous pyroclastic-volcanic lacustrine strata. We present high-precision geochronology and detailed sedimentology that together define a Kasimovian (305.77–306.10 Ma) volcanic recurrence interval of <10 ky. This is the first quantification of volcanic recurrence from deep (pre-Mesozoic) time, and importantly, represents an unique opportunity to gain insight into the nature of frequent and highly explosive silicic volcanism occurring across eastern equatorial Pangaea during the peak of the late Paleozoic ice age (LPIA). These data emanate from a single, unusually well-exposed and well-constrained site. However, this site is representative of many documented but poorly preserved volcanic centers of coeval or similar ages across west-central Europe. The concentration in both time and space of such highly explosive volcanism in paleoequatorial Pangaea has large implications for the climate system during Earth's penultimate icehouse. The recurrence rates established here support ongoing climate modeling efforts to test the hypothesis that this volcanism imposed a significant cooling effect near and during peak LPIA conditions prior to Permian icehouse collapse. In the wake of enhanced precision in geochronological tools, the integrated approach we apply for quantifying volcanic recurrence rates has broad utility in Earth's deep-time record.

Keywords LPIA, Volcanism, Recurrence rates, Equatorial Pangaea, Massif Central, Eruptive frequency

As part of the long-term carbon cycle, volcanism emits CO₂ to the atmosphere such that, to the first order, times of more widespread and active volcanism on Earth correspond with greenhouse climates, and reduced activity corresponds with icehouse conditions (e.g.,^{1,2}). However, it's also well demonstrated that individual, highly explosive eruptions can trigger short-term global cooling via stratospheric SO₂ injection (e.g.,^{3–6}). Low-latitude volcanism is especially likely to induce global-scale climate perturbations, with aerosol dispersal into both hemispheres (e.g.,^{4,7,8}). Explosive volcanic events are largely unrecognized as climatic forcing agents beyond decadal timescales, owing in part to the lack of age resolution (e.g.,⁹). However, in a recent compilation, Soreghan et al.¹⁰ demonstrated a global peak of explosive silicic volcanism centered in western Europe—eastern equatorial Pangaea—with additional centers in Australia, and suggested this volcanism might help explain the intensification and maintenance of icehouse conditions even as pCO₂ began to rise in the earliest Permian. To rigorously test this idea, our contribution herein enhances the resolution of this compilation by providing the first documentation of the frequency and nature of explosive volcanism in western Europe during this time. We target an exceptionally well-exposed section recording a single eruptive center representative of the nucleus of highly explosive volcanism that characterized eastern equatorial Pangaea in the late Paleozoic. Thus, the recurrence rates established here—the first for any deep-time interval—are especially important for supporting ongoing climate modeling efforts to test the mechanisms by which this volcanism could have imposed a cooling effect leading up to and during the peak of the Earth's penultimate icehouse.

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Geologic setting: orogenic collapse in the variscan system

Widespread calc-alkaline (andesitic-rhyolitic) magmatism accompanied syn-orogenic collapse of the Variscan belt ca. 300 Ma in western Europe (eastern equatorial Pangaea; Fig. 1A; cf.,^{11,12}). Several extensional basins (0–5°N) that formed during this time preserve numerous and relatively complete records of upper Paleozoic sedimentation with intercalated volcanics. The locations of Carboniferous (Pennsylvanian)-Permian calderas remain obscure, but include known or hypothesized locations in Italy, Poland, the Central Alps, France, and Germany (refs. in Supplementary Material 1). The source(s) of dated Permo-Carboniferous calc-alkaline volcanics in basins of the French Massif Central (FMC; Fig. 1B) are also unknown but likely differ from north (e.g., Autun, Aumance, and Decize–La Machine Basins,^{13–15}) to south (e.g., St. Affrique and Lodève Basins,^{16–18}). Speculated volcanic center(s) for southern FMC basins include proximal locations in south-central France (Aveyron,¹⁹) to more distal sources in the central-western Pyrenees^{20,21} and southeastern France (Corsica-Sardinia-Provence²²).

The continental Brousse-Broquiès sub-basin (of St. Affrique basin) in south-central FMC (B-B; Fig. 1B) developed above the flat-and-ramp Espinouse Detachment¹² ca. 305–295 Ma. It contains ~60 m of well-exposed volcanogenic strata in an abandoned quarry near Réquista (Fig. 2A). This volcanoclastic section, undated prior to this work, overlies Cambrian-Ordovician strata and underlies upper Stephanian strata (dated with plant fragments,¹⁹). Through the lens of this exquisitely exposed, high-resolution section, we present detailed sedimentology and high-precision age data that constrain the frequency and nature of explosive volcanism representative of that occurring broadly across Europe near the peak LPIA.

Volcaniclastic sedimentation in Brousse-Broquiès basin

Sedimentology

Detailed sedimentological investigations include high-resolution digital outcrop imaging and a cm-scale measured section (Fig. 2), facies analysis of volcanoclastic rocks (using the scheme of White and Houghton²³), and petrography (Figs. 3 and 4). Whole rock geochemical and clay mineralogy data complement this work (Supplementary Material 2–3). We define three distinct facies:

Bedded chert

Planar beds (3–60 cm thick) of white-gray microcrystalline quartz (Fig. 3A) internally display abundant soft sediment deformation (SSD) including recumbent to chaotic folds and slumps, fold- and dish structures, and conical fluid-escape structures (Fig. 3E–G). Fine (<2 mm) tuff and lapilli fragments define discrete layers and

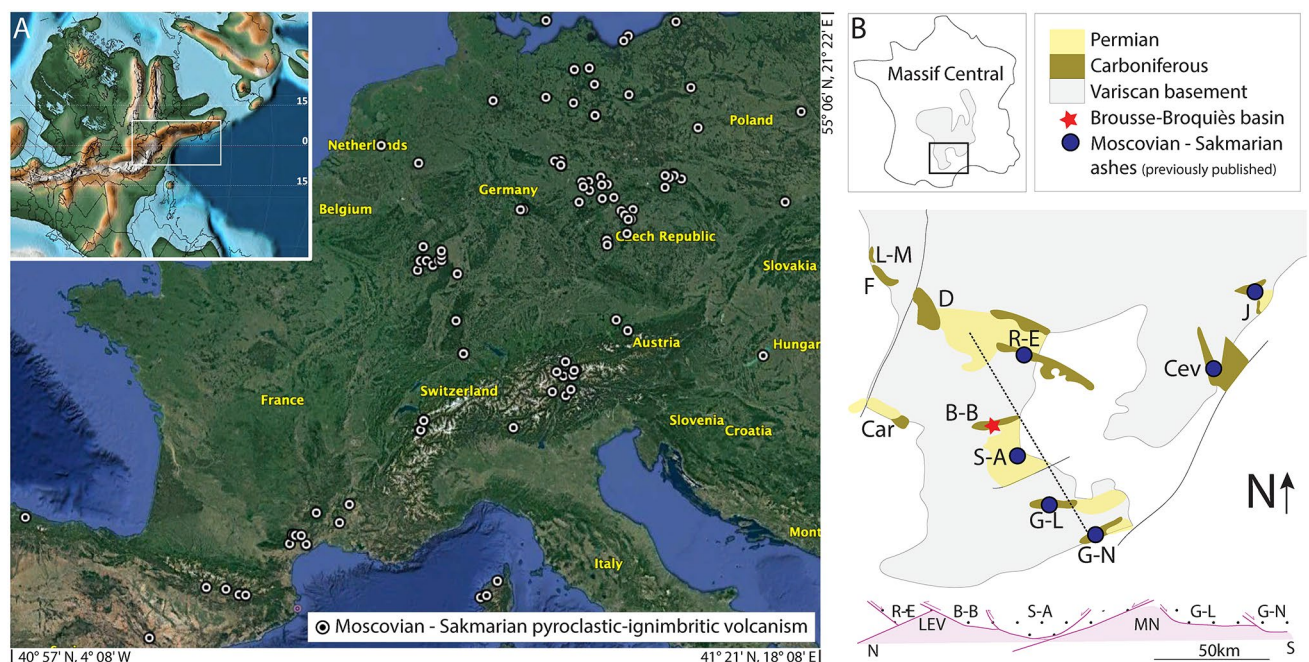


Fig. 1. (A) Distribution of known Moscovian-Sakmarian (310–290 Ma) volcanic deposits in Western Europe (dataset from Soreghan et al., 2019); modern coordinates projected on GoogleEarth. Inset: Early Permian paleogeography from C.R. Scotese PaleoAtlas. (B) Structural map of the southern FMC (drafted in Adobe Illustrator Version 27.6.1, adapted from Timmerman et al., 2004) showing distribution of dated Moscovian-Sakmarian volcanics within upper Carboniferous basins of the southern FMC: Lacapelle-Merival (L-M), Figeac (F), Carmaux (Car), Decazeville (D), Bertholene-Lassours (R-E), St. Affrique (S-A), Graissessac-Lodève (G-L), Gabian-Neffies (G-N), Cévennes-Ales (Cev), and Jauzac (J). Red star on Brousse-Broquiès (B-B) basin (study area). Supplementary Material 1 summarizes age and reference data of dated volcanics.

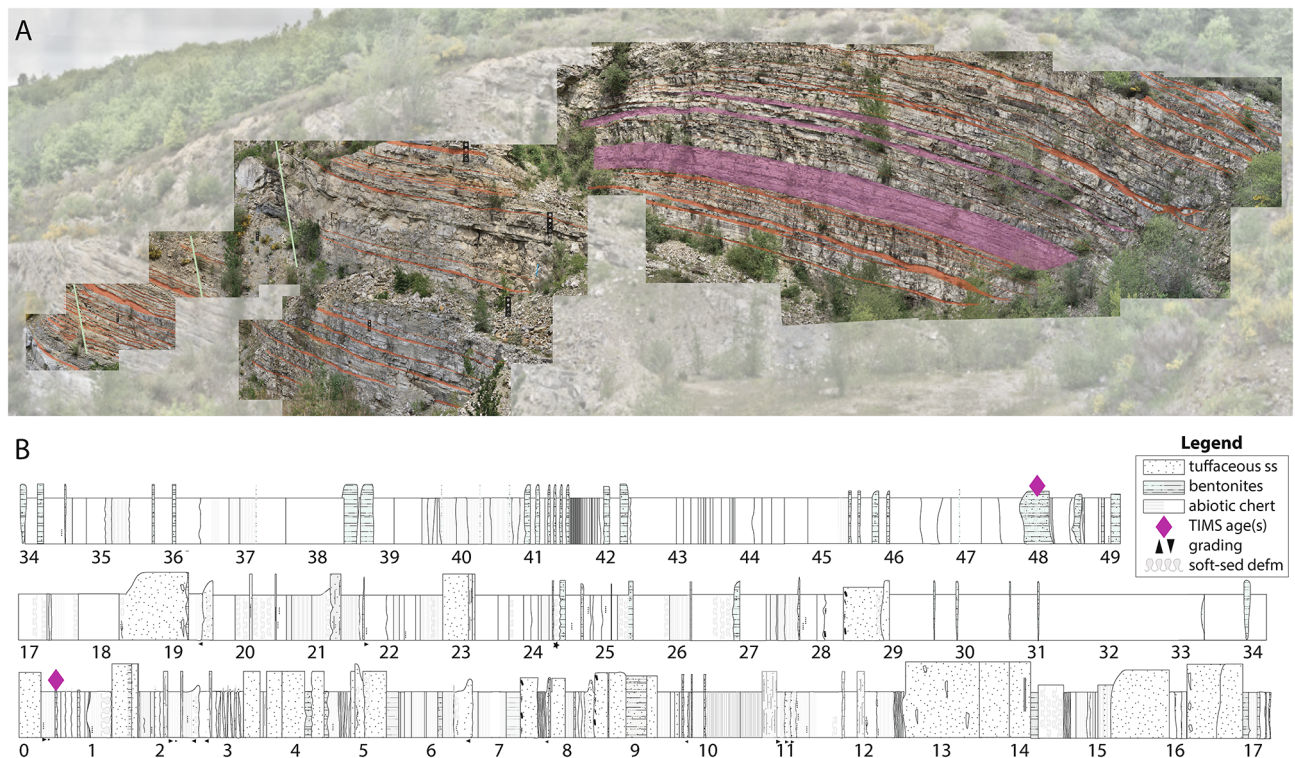


Fig. 2. Stratigraphy of Brousse-Broquiès basin. (A) Gigapan image (<https://gigapan.com/gigapans/216124>) with highlighted bentonites (red = singular bentonite; purple = densely interbedded bentonite/chert). (B) Detailed stratigraphic log with sample locations of new ages.

micrograded sequences (Figs. 3C and 4C–E), and ripples, hummocky cross stratification, and coarse (cm-scale) lapilli fragments are commonly preserved on bedding planes (Fig. 3H–I).

This unit is interpreted as bedded chert formed by chemical precipitation of silica in a lacustrine setting. Lacking evidence of biogenic silica precipitation, we interpret the origin of the chert as abiogenic—rare in modern lakes (e.g., Lake Magadi), but recognized in deep-time volcanic lakes (cf., Permian Bolzano Volcanic Complex, Italy,²⁴). Tuff and lapilli that range in size from mm-scale grains that define internal laminations (Fig. 3C) to ~2–3 cm fragments preserved along bedding planes (Fig. 3H–I) represent ash fall deposits. We infer the SSD (convolute bedding and dewatering structures) to record seismically induced liquefaction (cf.,^{25,26}) associated with volcanic eruptions. Pinch-swell geometries and laterally discontinuous layers (mid-section; Fig. 2A) similarly record seismicity, or post-depositional Na-silicate dehydration²⁴.

Bentonite

A total of ~70 massive (unstratified) tabular beds (1–30 cm thick) of green-gray fissile (Fig. 3A–B), clay-rich, fine-grained tuff occur with abrupt basal and upper contacts (Fig. 2). These units are composed primarily of altered glass, potassium feldspar, angular quartz, and muscovite (Fig. 4A–B), and have silica-rich compositions that plot in the volcano-tectonic fields of high-K calc-alkaline trachyte/trachy-andesites. R1-type illite-smectite (I/S) mixed-layer clay minerals (Supplementary Material 3 characterize the clay fraction (<2 μm) mineralogy of these units.

As is typical in fine-grained volcanic rocks, assessing the degree of reworking is challenging²⁷. Nevertheless, compositions dominated by altered glass, potassium feldspar, angular quartz, I/S R1 mixed-layer clay minerals (cf.,²⁸) are consistent with a pyroclastic origin. This facies is interpreted as devitrified pyroclastic tuff wherein smectite has converted to mixed-layer illite-smectite. The presence of muscovite indicates some mixing or inheritance of detrital material, likely via reworking after deposition. Bedding geometries are consistent with sublacustrine deposition as fallout from eruption clouds (cf.,²⁹) rather than surge or basal-surge deposits which tend to be thicker and more stratified³⁰.

Tuffaceous silt-sandstone

Moderately to poorly sorted, fine-medium tuffaceous sandstone occurs, composed of detrital (subrounded quartz grains) and/or reworked material including volcanic lithics (Fig. 4F) and chert fragments up to ~5 cm (Fig. 3D), as well as organics (plant- and carbonized wood fragments typically at bed bases). The contacts between these massive beds (5 cm–2 m thick) and other units are typically abrupt (locally undulous) but locally gradational (with chert).

This facies is interpreted as inter-to-syn-eruptive deposits of predominantly epiclastic material, reworked from lake margins. Epiclastic deposits can signal a hiatus in pyroclastic contributions (cf.,³¹), and in this setting

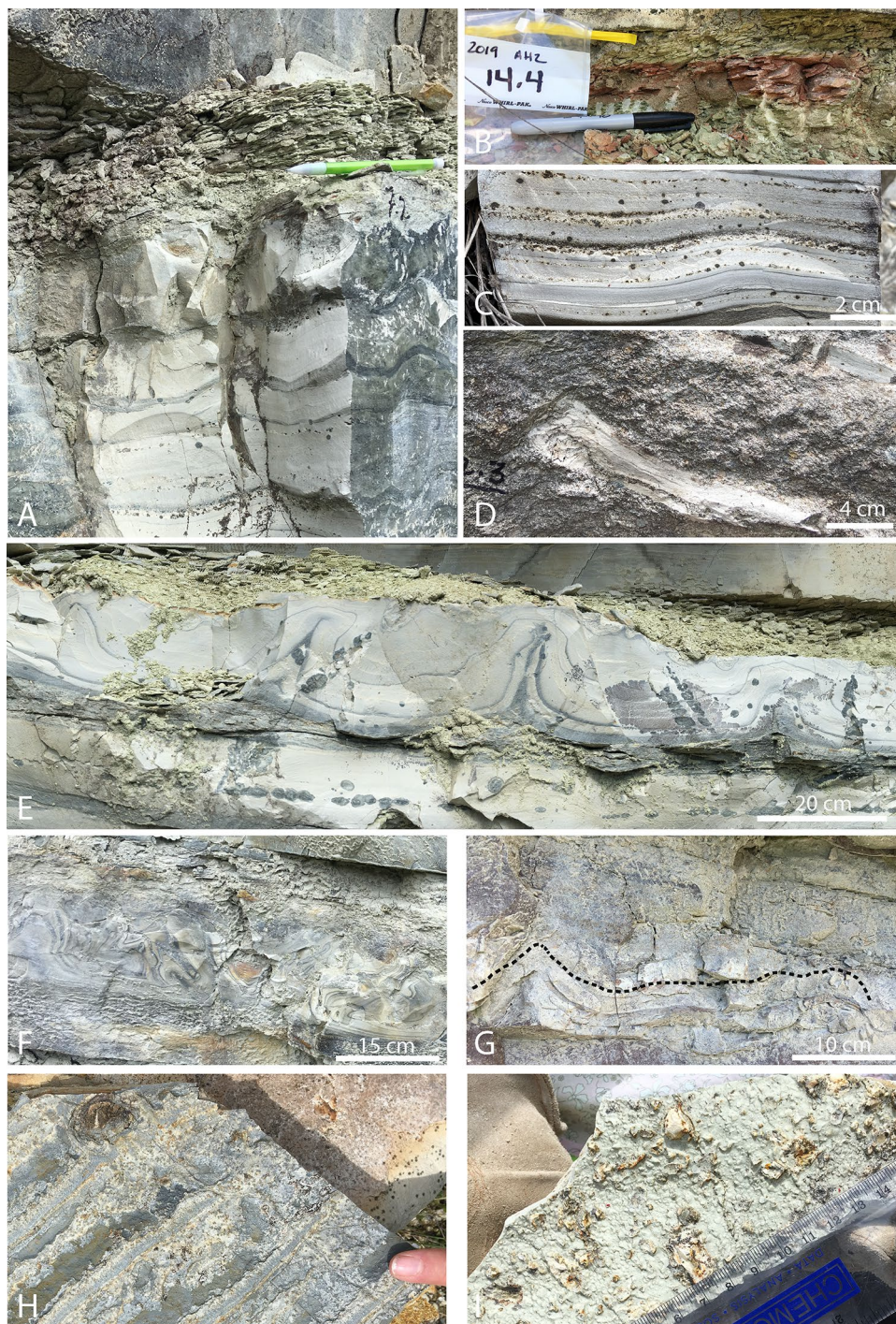


Fig. 3. Photos of the primary facies in Brousse-Broquiès basin including abiogenic chert (A,C), fissile bentonite (A,B), and tuffaceous silt-sandstone (D). Chert beds internally display abundant micrograding of fine (< 2 mm) pumice/lapilli (C), convolute bedding (F), and abundant soft sediment deformation (E,G). Ripples, hummocky cross stratification, and coarse (0.5–3 cm) lapilli are preserved on bedding planes (H–I). Tuffaceous sandstones (D) have rip-up clasts of chert and volcanic fragments.

may represent mass flows induced by slope failure (cf.,³²). Near the base of the section, load casts on bed bases and inverse grading (up from chert) are consistent with low-density turbidites (cf.,³³) or a waning of the eruptive source during deposition.

Geochronology

High resolution U–Pb ages of zircons from two bentonites were obtained using the chemical abrasion isotope dilution thermal ionization mass spectrometry (CA-ID-TIMS) method. The youngest analyzed zircon grains

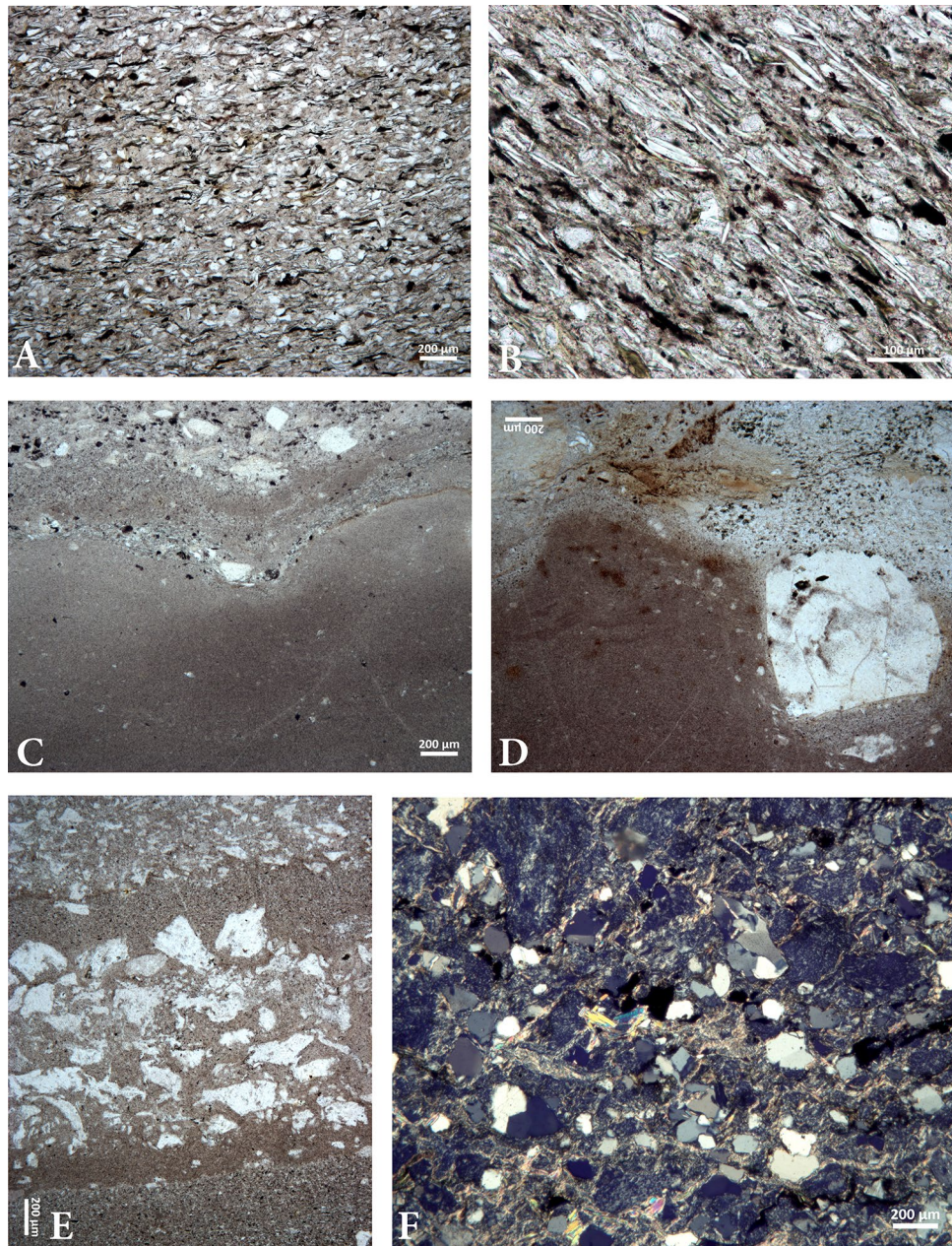


Fig. 4. Thin section photomicrographs of bentonite (A–B), micrograding of fine pumice/lapilli (white) in abiotic chert (gray) (C–E), and tuffaceous sandstone (F).

from bentonites at the base and top of the section define Kasimovian Maximum Likelihood Ages (MLAs;³⁴) of 306.10 ± 0.07 Ma ($n=4$) and 305.77 ± 0.08 ($n=3$), respectively, that are resolvable outside the analytical uncertainties, and are < 400 ky apart (Table 1). These ages bracket the duration of this well-exposed series (Fig. 2) and define the timing of significant eruptions in the region (southern FMC; Fig. 1B). The prismatic/euhedral shapes of (youngest) volcanic zircons are diagnostic; older detrital or xenocrystic zircons (Supplementary Material 4) are likely inherited from Ordovician metarhyolite and orthogneiss in the Lézou massif and Albigeois nappes north of the basin.

Brousse-Broquiés volcanism

The sedimentological characteristics of the volcanoclastic strata in Brousse-Broquiés basin are consistent with deposition in a moderate-to-deep closed freshwater lake that experienced minimal water-depth fluctuation, comparable to time-equivalent volcanoclastic deposits in the Aragón-Béarn Basin, western Pyrenees (cf.,²⁰). Interbedded tuffaceous silt-sandstone amongst predominantly fine (chert and bentonite) units, and rare bedding plane HCS, is consistent with gravity flow deposits in a relatively deep lake. The broad lateral extent (~500 m) of chert and bentonite beds, as well as sedimentary structures common in the chert facies (laminations, graded

	Pb(c)	Pb	U	Th	Ratios								Ages (Ma)								corr
					206 Pb	208 Pb	206 Pb	err	207 Pb	err	207 Pb	err	206 Pb	err	207 Pb	err	207 Pb	err			
Sample	(pg)	Pb(c)	(pg)	U	204 Pb	206 Pb	238 U	(2σ%)	235 U	(2σ%)	206 Pb	(2σ%)	238 U	(2σ)	235 U	(2σ)	206 Pb	(2σ)	coef		
TOP																					
z2	0.18	29.1	109.8	0.21	1890.5	0.065	0.048557	.(11)	0.35368	.(89)	0.05285	.(87)	305.65	0.32	307.5	2.4	321	20	0.26		
z4	0.81	9.4	152.5	0.46	582.7	0.145	0.048583	.(19)	0.35726	2.(00)	0.05336	1.(94)	305.82	0.55	310.2	5.3	343	44	0.33		
z1	0.16	18.1	63.3	0.08	1224.3	0.025	0.048606	.(14)	0.35883	1.(15)	0.05357	1.(10)	305.95	0.42	311.3	3.1	352	25	0.41		
												MLA	305.77	0.08							
												WM	305.78	0.23	MSWD=	0.51					
												YSG	305.65	0.32							
BASE																					
z1	0.20	7.7	31.6	0.31	500.2	0.097	0.048620	– .(34)	0.34850	– 2.(71)	0.05201	– 2.(61)	306.00	1.00	303.6	7.1	285	60	0.37		
z17	0.22	13.0	62.6	0.07	886.7	0.021	0.048594	– .(18)	0.34892	– 1.(42)	0.05210	– 1.(37)	305.89	0.55	303.9	3.7	289.0	31.0	0.31		
z2	0.24	5.7	28.4	0.26	378.1	0.081	0.048464	– .(38)	0.34552	– 4.(32)	0.05173	– 4.(18)	305.10	1.10	301.0	11.0	272.0	96.0	0.41		
z4	0.20	58.1	228.3	0.53	3440.5	0.167	0.048640	.(08)	0.35360	.(42)	0.05275	.(41)	306.17	0.23	307.4	1.1	317.0	9.4	0.20		
												MLA	306.10	0.07							
												WM	306.09	0.20	MSWD=	0.23					
												YSG	305.09	1.15							

Table 1. Summary of geochronological data including raw CA-ID-TIMS age data and calculated MDAs (3 methods) from the top (AH3-3.5) and base (AH-0.4) of section. MLA (Maximum Likelihood Age) is a statistical approach to calculating MDA (Vermeesch, 2021); WM (Weighted Mean) is the weighted mean of the youngest three or four zircon grains; YSG (Youngest Single Grain) is the Pb206/U238 age of the youngest single grain. Significant values are in (bold/italics).

and convolute bedding) record subaqueous deposition with common disturbances (e.g., seismicity). Formation of abiogenic silica typically implies semiaridity and high silica flux (cf.,²⁴) that is, for example, consistent with eolian contributions of highly reactive volcanic ash. The 70 discrete bentonites mark high (and persistent) pyroclastic influx (Fig. 2). However, alternations among bentonites, chert, and epiclastic units reflect episodic deposition (cf.,³⁵), where syn-eruptive fine bentonite marks sudden (event) deposition into a low-energy lake with background sedimentation of abiotic chert and coarser epiclastics shed from lake margins. The transitions from thicker (3–30 cm), more widely spaced bentonites in the lowermost and uppermost sections to thinner (1–5 cm) and more abundant bentonites in the mid-section potentially records variability between higher-magnitude, less-frequent events (lower and upper) to smaller, more-frequent events (mid).

It is difficult to estimate the magnitude of this volcanism without knowing the location of the caldera. Volcanic center(s) could include the Central Pyrenees, northern Black Forest (Germany), or southeastern France, as speculated for other documented 310–290 Ma calc-alkaline volcanics in the southern FMC (cf.,^{18,28}); however, some observations from the Brousse-Broquiès sedimentological record indicate a more proximal location (cf.,¹⁹). The spatial distribution and sedimentological characteristics of tephra deposited around a volcanic center can aid in reconstructing eruptive magnitude (cf.,^{36,37}). For example, decompacted tuff thicknesses of ~5–10 cm (but up to 100 cm) are measured ~50 km windward of recent Plinian volcanic eruptions (e.g., Mt. St. Helens, Pinatubo, and Taupo,^{38,39}); and ~2 cm-thick tuffs have also been found up to ~1500 km from Plinian volcanic centers (e.g., Toba,⁴⁰). However, it is difficult to evaluate distance from source based on ash bed thickness at any one location. A more reliable proxy for eruptive intensity is tephra grain size, albeit tephra fine with distance; using theoretical clast dispersal curves, a maximum clast size of ~3 cm occurs ~ < 60 km from the source^{37,41}. Finally, most occurrences (~80%) of seismically-induced liquefaction occur within 30 km of earthquake foci and require Richter magnitudes > ~6^{25,42}. Taken together, the presence of thick bentonites (up to 25–30 cm, compacted), coarse lapilli (up to 3 cm; Fig. 3H–I), and seismites (Fig. 3E–G) in the Brousse-Broquiès basin is consistent with volcanic source(s) more proximal (<50 km) than other speculated sources for calc-alkaline volcanics in the southern FMC, yet there is no (outcrop) evidence for upper Paleozoic calderas here. Most likely, a volcanic center is either buried beneath the Permian ca. 295 Ma Saint-Affrique basin and/or the Mesozoic section east of Saint-Affrique (Causse du Larzac) or is not preserved owing to prevalent late Paleozoic erosion.

Carboniferous volcanic recurrence

The ~400 ky of deposition recorded by the upper and lower bentonites that bound the study section, and the ~70 bentonites within it (Fig. 2), together define a volcanic recurrence interval of <10 ky, about two times the frequency of eruptions during the Miocene ignimbrite flare up in the Cascades (cf.,⁴³). To estimate the frequency of events with consideration for uncertainty within the bentonite count and uncertainty (error) in the bounding ages, we employ a Monte Carlo simulation using 60 (+/– 10) events from 306.100 +/- 0.070 to 305.770 +/- 0.080. This simulation provides a mean event frequency of 2.095 events/10 ky with a 95% confidence interval of 0.96–4.87 (Fig. 5). However, the Brousse-Broquiès basin records only one center of silicic magmatism of many occurring broadly throughout western-central Europe (as well as Australia) during this time (Fig. 1 and refs. above). Geochemical and mineralogical compositions (Supplementary Material 2) of Brousse-Broquiès

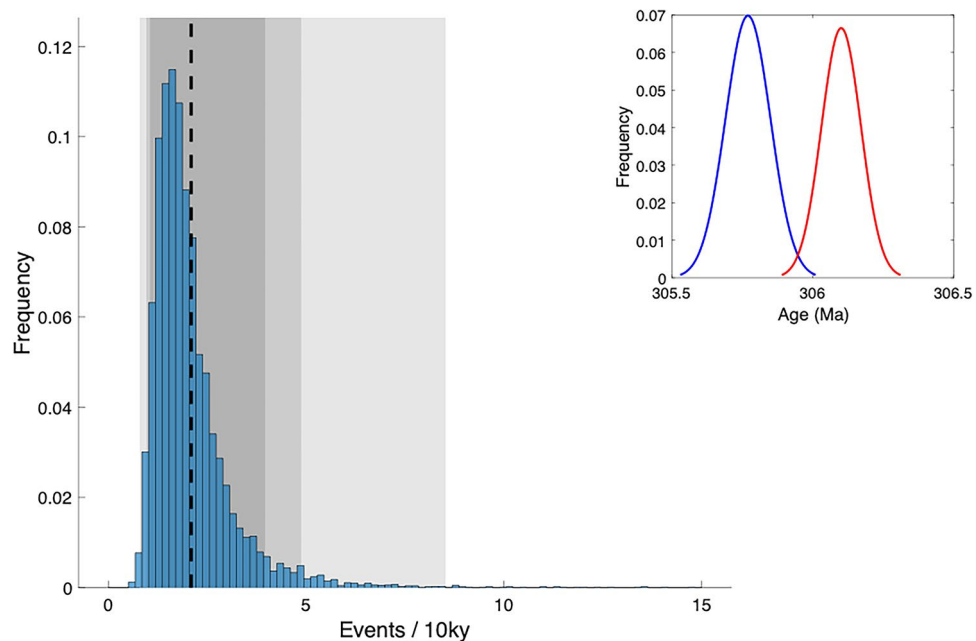


Fig. 5. The left panel shows the frequency of modeled events/10 ky based on 10,000 Monte Carlo simulations. The ages were drawn from a normal distribution centered on the average age with a standard deviation of the age uncertainty (age distributions from the top and bottom of the studied stratigraphic section are shown in the upper right panel). Results (in events/10 ky) show the mean of 2.10 (represented with the black dotted line), median of 1.82 (dashed line), and standard deviation (5.85). Gray shaded regions show the 90% (0.80–8.52), 95% (0.96–4.87) and 99% (1.06–3.96) confidence intervals.

bentonites are comparable to those documented in the Permo-Carboniferous Cinéritique Formation¹⁸; nearby location, proposed upper Carboniferous age) and in known volcanic centers from the Central Pyrenees to the Provence-Corsica-Esterel region^{44–46}. If Brousse-Broquiès-style volcanism is representative in frequency and magnitude of extensional Variscan magmatism occurring throughout the broader orogenic system (Fig. 1, Supplementary Material 1, and refs. above)—an area that likely hosted several volcanic centers—then this implies a sub-centennial eruptive frequency in the region. Such a recurrence rate has been linked to prolonged perturbation of the climate system in both recent (e.g.,⁶) and deep time (e.g.,^{10,47,48}) records, when closely spaced and significant volcanic eruptions compound, amplifying successive climatic perturbations into a persistent forcing. The poor preservation of calderas and difficulty in correlating tephras in deep time confounds accurate estimates of both the magnitude of such volcanism, as well as the number of volcanic centers during this time. Yet, the concentration of late Carboniferous-Permian volcanic products across western-central Europe is well known (Fig. 1; Supplementary Material 1), and their paleo-equatorial position implies pronounced global climate perturbations⁴⁹ owing to the outsized influence of explosive eruptions on radiative forcing at low latitudes.

The new data presented herein will inform future climate modeling efforts to test the hypothesis that this silicic large igneous province (SLIP) of west-central Europe repeatedly perturbed the climate toward either consecutive short-lived, or sustained cold conditions of Earth's penultimate icehouse. In working with records of this age, it is particularly challenging to make critical estimations of volcanic magnitude, including estimations for the rate of degassing or the quantity of volatiles released. Attempts are in progress to estimate the sulfur burden from melt inclusion work, but regardless of the outcome, the tight chronology presented herein will serve as a framework—together with volcanic gas flux and aerosol estimations from a range of modern analogs—to further assess the importance of this volcanism. Following Lee and Dee⁵⁰, an eruptive interval of 10 ky could possibly sustain long-term low $p\text{CO}_2$ levels if carbon sequestration associated with ash fertilization effects could offset—temporarily—the volcanic $p\text{CO}_2$ input. Initially, volcanism near and during peak LPIA could have imposed a negative radiative forcing that exceeded the warming of $p\text{CO}_2$ loading, before eventual $p\text{CO}_2$ buildup tipped the system toward warming, and the ultimate early Permian collapse of the LPIA.

Conclusions

We present the first quantification of near peak-LPIA volcanic recurrence (<10ky) from a reference section of unprecedented exposure and resolution in the Brousse-Broquiès basin (south-central France). This site—capturing a Kasimovian (305.77–306.10 Ma) interval—is one of many contemporaneous and highly explosive volcanic centers that are known but poorly resolved across west-central Europe—paleoequatorial Pangaea. If eruptive recurrence in this site is representative of broad calc-alkaline late to post-Variscan magmatism across low-latitude Pangaea, then the collective impact on the climate system of Earth's most prominent Phanerozoic icehouse was substantial. The late Paleozoic world (starting ~310 Ma) records some of the most abundant and frequent stratospheric injections of sulfate aerosols in Earth's Phanerozoic history, at low latitudes particularly

sensitive for radiative forcing effects. The data presented here help constrain climate simulations to elucidate cooling effects of extreme, low-latitude volcanism—both individual and successive eruptions—near the peak of the LPIA.

Data availability

All data generated or analyzed during this study are included in this published article and its supplementary information files.

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Author contributions

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Declarations

Competing interests

The authors declare no competing interests.

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