

VOLCANISM AND CLIMATE RECORDED IN GIANT GRAINS FROM DUST OF THE LATE PALEOZOIC PANTHALASSIC OCEAN

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ABSTRACT: Models indicate that atmospheric transport distances for large eolian particles are limited to tens to perhaps hundreds of kilometers owing to rapid gravitational settling, yet a number of studies of modern–recent sediment have documented the enigmatic transport of so-called “giant” (generally > 63 μm) grains over thousands of kilometers. Additionally, stratospheric injection of tephra by volcanic eruptions has the potential to increase the atmospheric transport distance of volcanically sourced aerosols in particular. Here we report the first finding of wind-transported giant grains in Earth’s pre-Pleistocene record, in upper Carboniferous–Permian shallow-marine carbonates of the Akiyoshi (paleo) atoll, which formed in the remote equatorial Panthalassic ocean. The paleogeographic setting of the Akiyoshi Limestone makes it an ideal system for studying far-traveled continental and volcanic dust. Siliciclastic grains with diameters commonly $\sim 300\ \mu\text{m}$ and up to $\sim 2\ \text{cm}$ occur in this system, transported at least 4,500 km and up to 10,000 km away from the nearest viable source regions. Textural and compositional (both mineralogical and geochemical) data indicate the presence of both continentally sourced eolian grains and volcanically sourced grains of intermediate to felsic compositions, with the relative proportions of volcanic to continental grains peaking in the Moscovian (middle Late Carboniferous). The peak timing of volcanic versus continental grain deposition indicates that large-magnitude volcanic eruptions affected this part of equatorial Panthalassa in the Late Carboniferous, and that Pangean continental aridity increased significantly into the Permian. Compositional data indicate likely source regions west of the Akiyoshi (paleo) atoll, consistent with the common occurrence of westerlies over this paleoequatorial region, thus indicating the operation of monsoonal circulation over the Paleo-Tethys sea.

INTRODUCTION

Mineral aerosols (airborne dust particles) are an important component of the atmosphere, affecting both direct and indirect radiative forcing, as well as ecosystem fertilization and cloud formation (e.g., Boyd et al. 2000; Mahowald and Kiehl 2003; Karydis et al. 2011; Kok et al. 2023). Until recently, most research on dust focused on the < 10 μm fraction, as such particles can remain suspended for long distances, whereas models suggested that coarse particles settle rapidly proximal to their source(s) (Mahowald 2011; Adebisi and Kok 2019). In the last few decades, however, observations from atmospheric sampling, and recent sediment deposits have found evidence for surprisingly long-range transport of so-called “giant” grains (> 63 μm) (e.g., Betzer et al. 1988; Ram and Gayley 1991; Van Malderen et al. 1992; Arimoto et al. 1997; Middleton et al. 2001; Van der Does et al. 2018; Ryder et al. 2019; Varga et al. 2021). Documentation of the prevalence of this phenomenon through time is important, since coarse grains affect climate differently than fines. Coarse

particles can increase absorption of incoming and outgoing solar radiation, influence cloud formation and distribution, and contribute to iron delivery and ballasting, among other effects (Boyd et al. 2000; Nenes et al. 2014; Kok et al. 2017; Adebisi and Kok 2019; Adebisi et al. 2023). Currently, the climate effects of coarse grains are mostly neglected in models owing to their short anticipated residence times.

Additionally, coarse grains in aerosols and eolian dust deposits can act as tracers of atmospheric circulation as well as indicators of the relative moisture of source regions, and—potentially—the prevalence and explosivity of volcanism. Accordingly, giant grains offer a potential trove of paleoclimatic data previously undocumented in the geologic record.

In this study we present the first documented example of extreme, long-range transport of giant grains in Earth’s deep-time record. Our data come from the Carboniferous–Permian Akiyoshi Limestone (Japan), which formed part of an atoll isolated in the vast Panthalassic Ocean (Sano 2006) (Fig. 1). The giant grains preserved here capture transport from both explosive volcanic and arid continental sources. These data shed light on

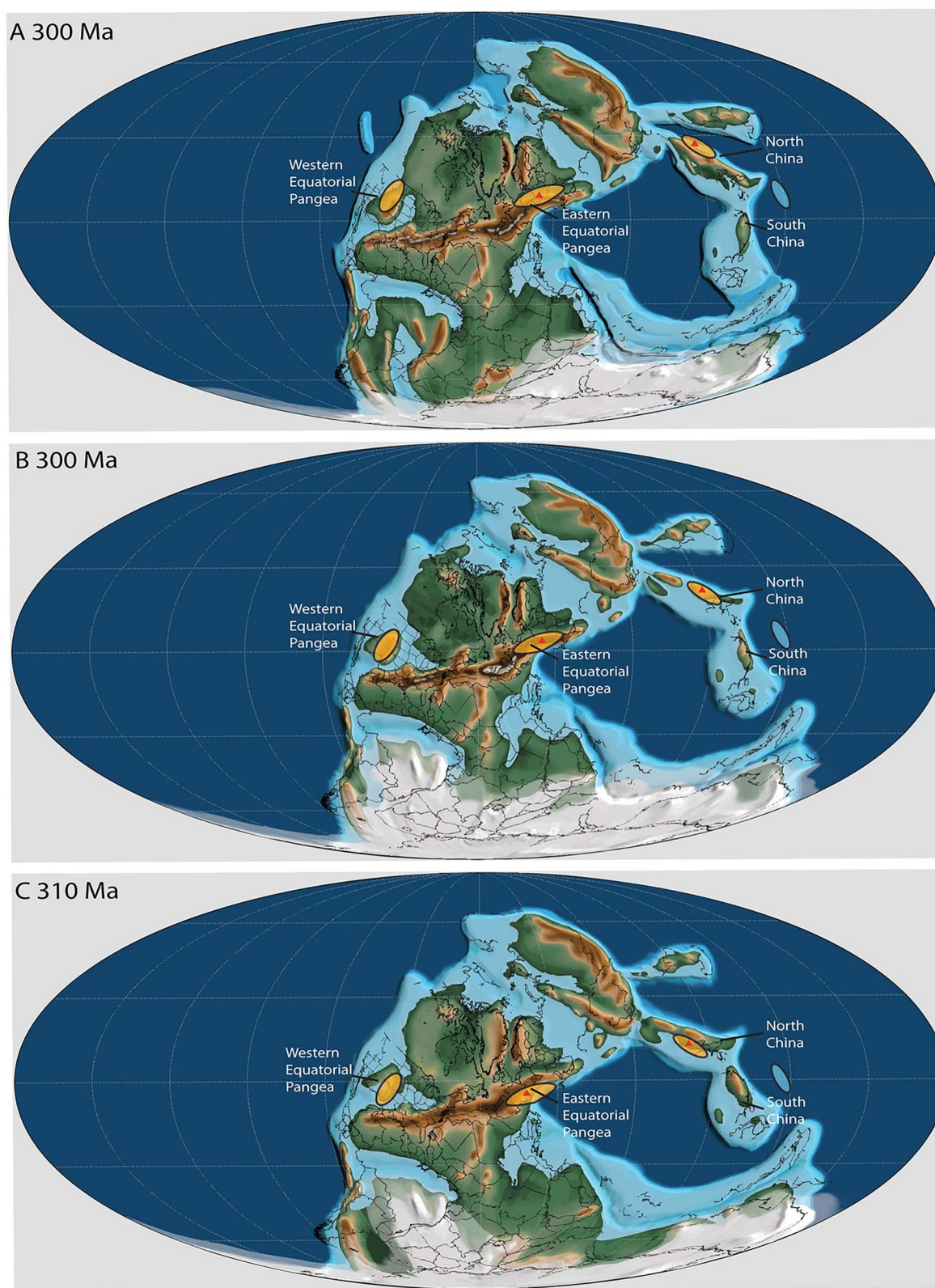


FIG. 1.—Paleogeographic reconstructions for: **A)** 290 Ma, **B)** 300 Ma, and **C)** 310 Ma modified from Scotese (2021) to illustrate locations of regions discussed in the text. Blue ellipses, Akiyoshi atoll; red triangles, volcanic sources; yellow ellipses, sources of continental dust.

volcanic events and climatic conditions during the study interval and demonstrate the ubiquity and utility of the atmospheric transport of giant grains, even in Earth's deep-time record.

GEOLOGIC BACKGROUND

Depositional and Tectonic Setting of the Akiyoshi Terrane

The Akiyoshi Terrane of southwest Japan (Figs. 1, 2) is an accretionary complex comprising oceanic and trench-fill deposits of Carboniferous to Permian age (Kanmera and Nishi 1983; Kanmera et al. 1990; Sano and Kanmera 1991). The Akiyoshi Limestone rests on a basaltic seamount that formed in the equatorial Panthalassic ocean (Fujiwara 1968; Sano and Kanmera 1988). The Akiyoshi Limestone, a shallow marine unit that accumulated atop the atoll, together with basinal and deep marine chert, formed in an oceanic setting from middle Viséan (~ 340 Ma) to late Guadalupian (~ 260 Ma) time (Sano and Kanmera 1988; Sano and Kanmera 1991). The Akiyoshi atoll neared and was ultimately obducted onto the Japan margin in the late Permian (Kanmera et al. 1990; Sano and Kanmera 1991; Kasuya et al. 2012). Owing to its tectonic and paleogeographic setting far removed from any continental or intermediate-felsic volcanic sources, any siliciclastic material of felsic-intermediate composition found in the Akiyoshi Limestone is allochthonous and thus must have been delivered to the atoll via wind transport. This is the basis for using the Akiyoshi Limestone as a paleo-dust trap.

Post-Paleozoic remagnetization of the Akiyoshi Limestone precludes precise paleolatitudinal reconstructions (Moriga et al. 1988), but recent analyses of the analogous (coeval paleoatoll terrane) limestone in Kyushu, south of the Akiyoshi Terrane, yielded a weak primary magnetic remanence indicating a paleolatitude of ~ 12° S (Kirschvink et al. 2015), consistent with Kasuya et al.'s (2012) placement of the Akiyoshi Limestone at a position of ~ 15° N (~ 260 Ma). The Gplates reconstruction by Domeier and Torsvik (2014) indicates generally northward plate motions through the time of deposition of the Akiyoshi Limestone, implying that the Akiyoshi atoll was positioned at latitudes ~ 10° N or lower from Middle Pennsylvanian through early Permian time. Using two plausible end-member assumptions for plate velocities of 5–10 cm/yr, chosen because larger oceanic plates tend to display higher spreading rates related to enhanced slab pull of oceanic lithosphere (Muller et al. 2008), the Akiyoshi Terrane likely migrated ~ 1,250 km to 2,500 km over the ~ 25 My encompassed between the (early) Moscovian and (late) Artinskian stages.

Paleoclimatic Setting

The Late Paleozoic Ice Age (LPIA) was a time of intense global cooling beginning in the late Devonian (~ 360 Ma) and persisting as late as middle-late Permian time (~ 280–260 Ma), with glaciation varying in intensity and spatial distribution throughout (e.g., Isbell et al. 2003; Fielding et al. 2008; Montañez and Poulsen 2013; Soreghan et al. 2019). Widespread evidence for continental glaciation that grounded to sea level exists across many of the Gondwanan continents of the southern paleo-pole (e.g., Isbell et al. 2003; Fielding et al. 2008a); additionally, controversial evidence exists for upland glaciation in western and eastern equatorial Pangaea (e.g., Becq-Giraudon et al. 1996; Soreghan et al. 2008, 2014; Pfeifer et al. 2021). As measured by the extent of diamictite distribution, glaciation peaked in the Asselian (earliest Permian; ~ 299 Ma) and largely collapsed by the Artinskian (~ 290 Ma; e.g., Montañez and Poulsen 2013; Soreghan et al. 2019); widespread exposure of epeiric systems near the time of the Carboniferous–Permian (~ 299 Ma) boundary suggests that the glacial maximum potentially occurred at this time (Koch and Frank 2011), although fusulinid data suggest a maximum in latest Asselian (Davydov et al. 2014). Far-field (low-latitude) records of the LPIA include carbonate-dominated cyclothem in Laurussia, recording glacial–interglacial eustatic variations (e.g., Heckel

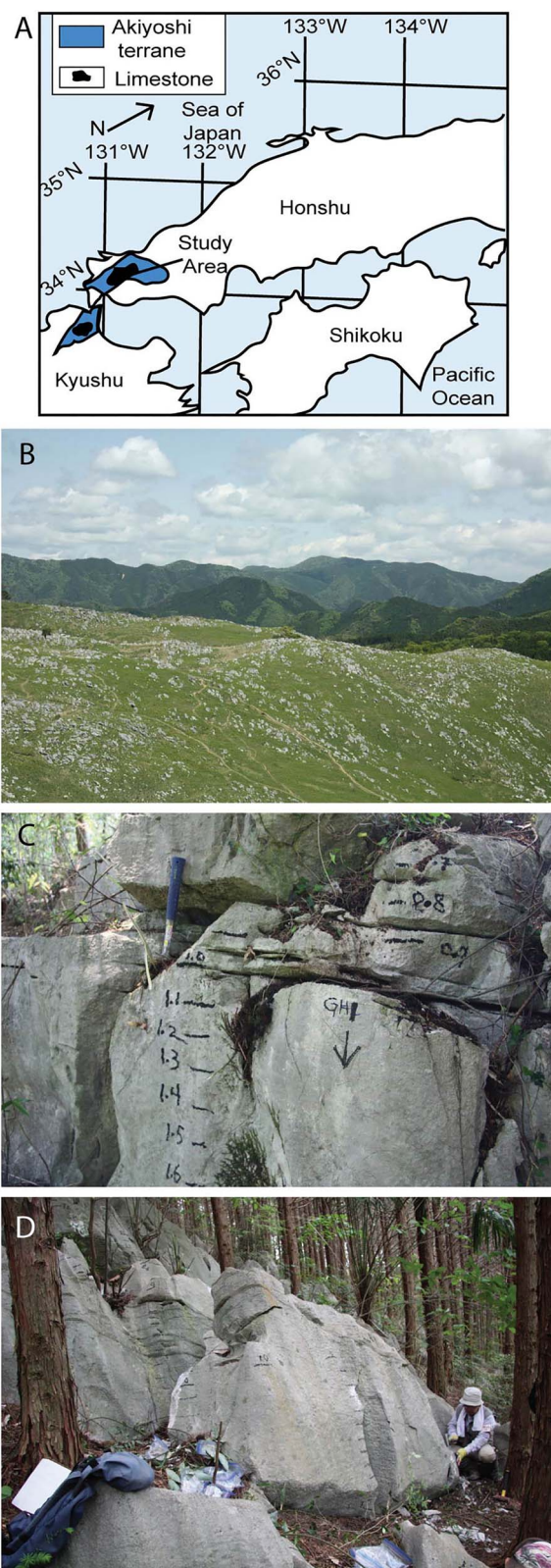


FIG. 2.—A) Modern location of the Akiyoshi Terrane in southwest Japan. B) Image of the Akiyoshidai Quasi-National Park in the foreground with the Akiyoshi Limestone sampling area in the forested region in the background. C, D) Studied exposures within wooded area. Note overturned structure of section.

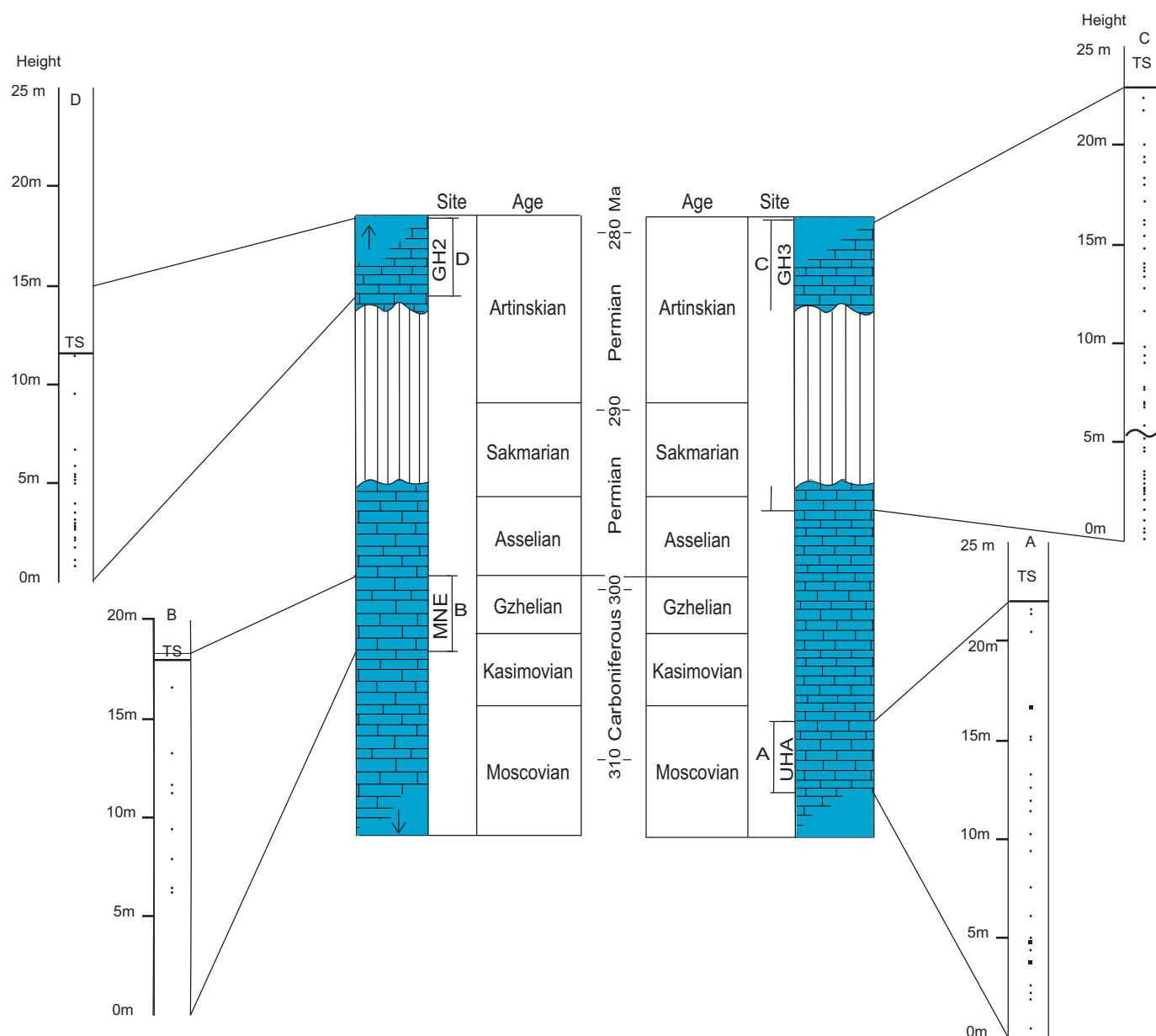


FIG. 3.—Stratigraphic framework of the study section. Note that each section was dated using fusulinid biostratigraphy (see text). Light blue columns on left and right denote the Akiyoshi Limestone, with A, B, C, and D brackets indicating our specific study intervals (UHA, MNE, GH3, and UHA are the field names for these study intervals). Enlargements of columns A, B, C, and D show thicknesses of study intervals, annotated with giant grains (black dots) and giant lithics (black squares). Note the disconformity in C, discussed in text. TS indicates top of section.

1993; Bishop et al. 2010). Shallow-marine carbonate strata of the oceanic Akiyoshi Limestone similarly record sea-level variations inferred to reflect glacioeustasy (Nakazawa and Ueno 2004; Sano 2006).

METHODS

Several discrete intervals (10–20 m) of the Akiyoshi Limestone representing the mid-Moscovian (21 m), latest Kasimovian–early Gzhelian (13 m), latest Asselian (3.5 m)–earliest Sakmarian (12 m – 20 m), and late Artinskian times (70 m in total) were logged and sampled at 20 cm increments (Fig. 3; see Patterson 2011 and Qi 2016). A disconformity discovered (revealed by biostratigraphic analyses) in the upper Asselian–lower Sakmarian section resulted in a truncated Asselian interval of only 3.5 m. Fusulinoidean

biostratigraphy (by Dr. V. Davydov for Moscovian–Asselian strata and K. Ueno for Sakmarian–Artinskian) provided age constraints. Supplemental Table S1 lists sampling locations, and Figure 3 illustrates schematized stratigraphic logs of the study intervals. Here, we focus specifically on grains with long axes generally larger than silt size ($> 63 \mu\text{m}$) recovered both in previous work (Patterson 2011; Qi 2016) and in newly-processed sections. For both previously-collected and new sections, thin-section analyses guided facies and stratigraphic analyses, as well as fusulinid determinations.

For all samples, detrital silicate minerals, interpreted as atmospheric dust, were extracted following the procedure in Sur et al. (2010). Briefly, samples were cleaned of external debris by washing with 1N HCl, rinsed with distilled water, then dried, crushed to the size of small pebbles, and rinsed again. Approximately 200–300 g of the resultant gravel was

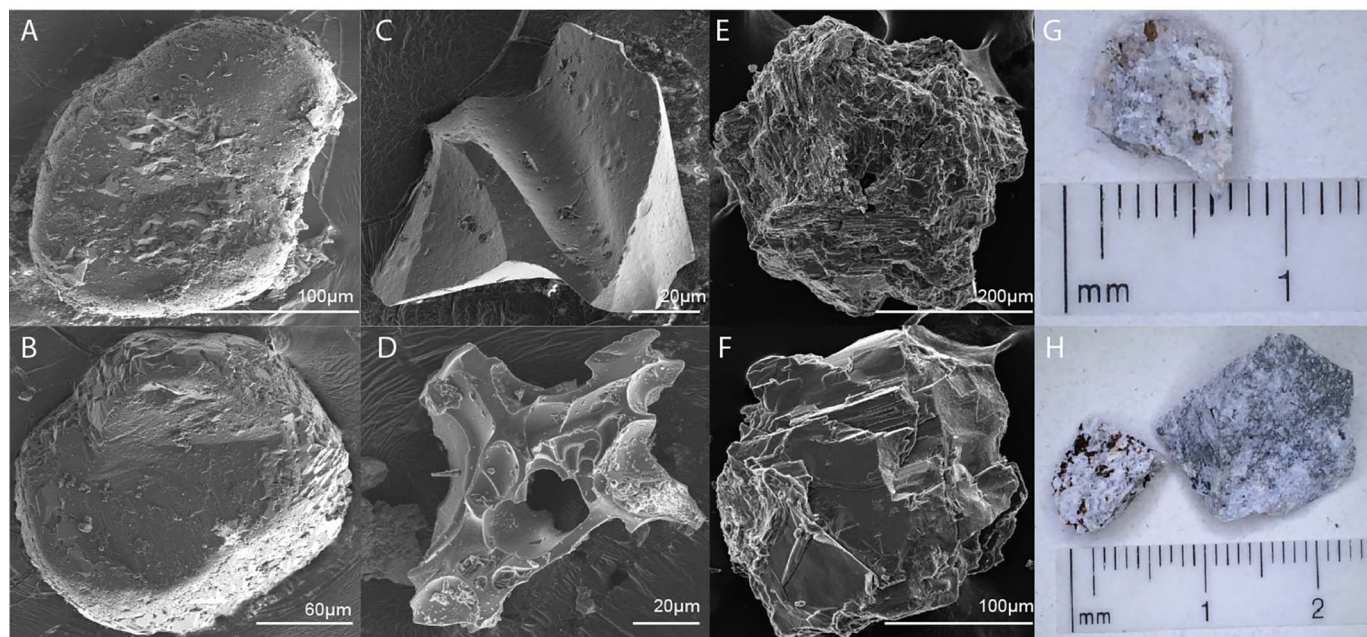


FIG. 4.—Photomicrographs (SEM) and photographs of the giant grain types. **A, B** Well-rounded quartz grains (continental origin). **C, D** Glass of rhyolitic composition (volcanic). **E, F** Lithic fragments (volcanic origin). **G, H** Giant lithic fragments (volcanic origin).

weighed and then subjected to dissolution in 2N HCl at 50°C. The insoluble residue was then rinsed and centrifuged several times with distilled water, freeze dried, weighed, and then combusted at 550°C for 24 hours to remove organic matter and oxidize any pyrite. Subsequently, iron oxides were removed using citrate-bicarbonate-dithionite (CBD) followed by rinsing and freeze drying to obtain the final mass of the silicate mineral fraction (SMF).

A selection of the largest grains ($n = 1\text{--}30$ per sample), as well as a random sampling (up to 70) of additional grains were analyzed with respect to their shape, surface textures, and compositions with backscattered-electron (BSE) imaging using a scanning electron microscope (SEM) and an electron microprobe (EPMA). Randomization was done by numbering the grains and using a random-number generator for grain selection. For samples yielding < 10 grains generally larger than $\sim 50\text{ }\mu\text{m}$, all grains were mounted for analysis. Grain dimensions (short and long axes), qualitative shape (angularity), and elemental composition (for mineralogy) were determined using backscattered electron (BSE) and energy dispersive X-ray spectroscopy (EDXS) modes. Authigenic quartz grains, identifiable by their prismatic shape, were included in the random sampling, but this was done to measure the ratio of authigenic to detrital grains in each section. Authigenic grains were not considered during the rest of this study. Figure 4 illustrates photomicrographs of representative types of grains observed in this study.

Radiogenic isotope data ($^{87}\text{Sr}/^{86}\text{Sr}$ and $^{143}\text{Nd}/^{144}\text{Nd}$) were obtained on three large (cm-size) lithic clasts of igneous origin (which all occurred in the Moscovian section) and several samples of co-associated dust in the $< 180\text{ }\mu\text{m}$ fraction to supplement the mineralogical data in assessing sources and possible provenance regions. Nd-Sr isotope ratios were measured at the University of Michigan by thermal-ionization mass spectrometry on a Triton Plus TIMS instrument (Thermo Fisher Scientific) equipped with eight Faraday collectors. Total Sr and Nd process blanks were $\sim 50\text{ pg}$ and $\sim 100\text{ pg}$, respectively, requiring no corrections to data. All Nd-Sr isotope measurements were conducted in static mode with isotopic standards JNdi-1 (Nd) and NBS 987 (Sr) following methods of Aciego et al. (2009) and Aarons et al. (2013). Samples ($\sim 50\text{ mg}$) were fully digested using double-distilled HF-HNO_3 , HClO_4 , and HCl acids in screw-top teflon beakers on computer-controlled hotplates under class 100 clean lab conditions, and Sr and Nd separations performed on miniaturized chromatographic ion-exchange columns with

Eichrom element-specific resins, following procedures of Gleason et al. (2002), Aciego et al. (2009), and Aarons et al. (2013) for high-precision isotope-ratio analysis.

RESULTS

Of 283 limestone samples processed, 80 ($\sim 28\%$) yielded grains exceeding silt size. Most of the giant grains retrieved from the Akiyoshi Limestone fall within the sand fraction, up to 2 mm in diameter (very coarse sand); however, nine igneous lithic fragments from three discrete horizons in the Moscovian interval (levels 3.9, 4.9, and 16.7 m as measured from the base of the section) exceeded sand size, measuring $\sim 2\text{ mm--}2\text{ cm}$ diameter in long dimension (Fig. 4). Excluding these nine lithic fragments, the average giant grain exhibits a long axis of $264\text{ }\mu\text{m}$ and a short axis of $209\text{ }\mu\text{m}$. Table 1 summarizes average and maximum sizes for each time slice. Supplemental Data Table S2 summarizes, for each grain analyzed, the long-axis and short-axis lengths, mineralogy, grain shape, and long-axis and short-axis ratios, and provides an assessment of origin (volcanic versus continental), as well as auxiliary notes.

Of the 80 limestone samples yielding giant grains, grain abundance varies from 3 to > 250 , with no systematic relationship to depositional facies. When normalized by volume of limestone processed (Table 1), the 3.5 m representing the upper Asselian section yielded $\sim 2\text{--}3$ times the concentration (116 kg^{-1}) of giant grains relative to the upper Kasimovian–lower Gzhelian (34 kg^{-1}) and Moscovian (45 kg^{-1}) intervals, similar to the concentration of giant grains in the Artinskian (96 kg^{-1}). However, the disconformity that affects the Asselian interval could have skewed the results from this artificially truncated section, so the data from this section are considered to be low confidence. In other words, this disconformity captures a prolonged interval of time and thus the apparent concentration of allochthonous material here might be an artifact of time condensation.

Grain shape and composition vary as well (Supplemental Data Table S2). Grain shapes range from well-rounded to very angular, with rounded grains commonly exhibiting surface pitting. Rounded grains are most commonly quartz, or subordinately orthoclase and plagioclase. Subangular-angular grains common in all study intervals include quartz, orthoclase, plagioclase,

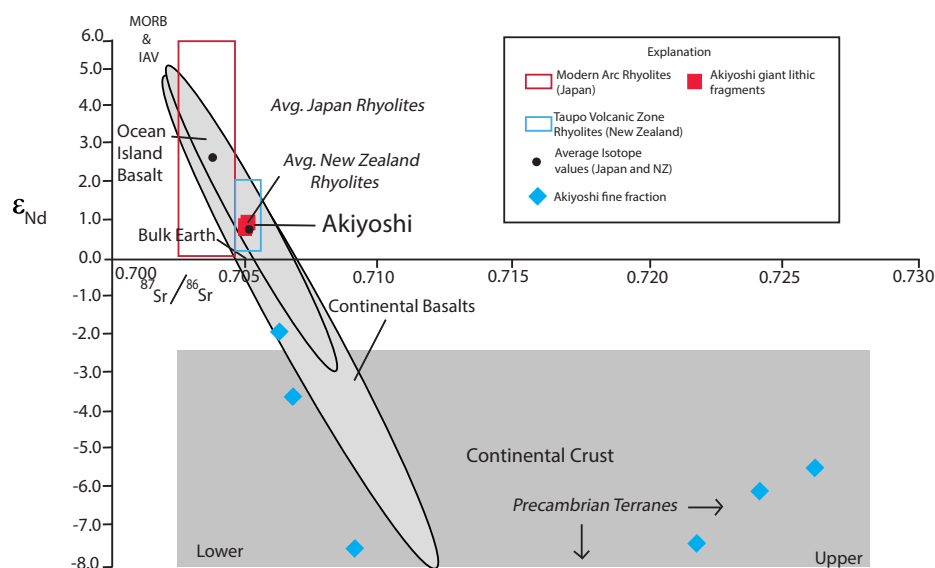


FIG. 5.— ϵ_{Nd} vs $^{87}\text{Sr}/^{86}\text{Sr}$ (measured values) for three Akiyoshi giant lithic fragments (e.g., Fig. 4G, H) compared to modern arc rhyolites (Japan and New Zealand) in Nd-Sr isotope space. Generalized fields for mid-ocean ridge basalt (MORB), oceanic island (OIB), island arc (IAV), and continental basalts define the crust–mantle Nd-Sr isotope array (White 2023) for reference. Akiyoshi lithic grains (red squares) have measured ϵ_{Nd} values of +1, overlapping with Late Cenozoic Japan Rhyolites (Takanashi et al. 2012) and New Zealand Rhyolites of the Taupo Volcanic Belt (McCulloch et al. 1994) used as proxies for modern volcanic arc rhyolites (black circles, average values for Nd-Sr ranges as indicated by rectangles). Akiyoshi fine-grained “dust” extracts (blue diamonds) have Nd-Sr isotope signatures typical of dust producing regions in older continental terranes (e.g., Aarons et al. 2013).

and igneous rock fragments. Rock fragments exhibit varying compositions (quartz+sanidine, quartz+orthoclase+albite, orthoclase+plagioclase+hornblende, etc.) ranging from intermediate to felsic. A special class of angular grains occurs, characterized by a vesicular texture, very high silica contents (up to 80%), and conchoidal fracturing (Fig. 4C, D). The vesicles, bubble walls, conchoidally fractured surfaces, and lack of visible minerals mark these as particles of volcanic glass, likely of rhyolitic composition based on the high silica content, close alignment of Al to summed Na+K+Ca, and minor but significant FeO (~0.5–1.5%) (Supplemental Figure S3a–S3c). Sanidine and local accessory phases (e.g., zircon) occur uniquely in the Artinskian section. Of the nine lithic fragments that exceed the sand-size fraction, all contain a predominantly albitic matrix, with amphibole and quartz, with lesser amounts of minerals comprising titanite, potassium feldspar, ilmenite, biotite, hematite, and chlorite. These compositions are most closely affiliated with intermediate to felsic volcanism; however, the size of these grains, flow textures inhibiting observation of grain boundaries, and the similarity between albite and quartz in backscattered-electron imaging preclude naming these rock types with absolute confidence. Nevertheless, the available data indicate andesitic to rhyolitic compositions. EDS data for all samples analyzed in 2020 and 2021 are included as Supplemental Data (S4).

Pseudomorphs of giant grains that consist of clay minerals with small inclusions of quartz, albite, potassium feldspar, mica, and Cr and Ti oxides also occur. The clay-matrix chemistry is dominated by silica with lesser quantities of Al, Fe, K, and Na. This matrix chemistry matches or is very similar to the compositions of the volcanic glass particles that occur throughout the studied sections, suggesting that

these pseudomorphs are fragments of altered glass (Supplemental Data S3a and S3b).

The three giant Moscovian lithic fragments display $^{87}\text{Sr}/^{86}\text{Sr}$ values ranging from 0.705136 to 0.705260 and ϵ_{Nd} values ranging from +0.9 to +1.1, typical of many modern volcanic arc settings (Fig. 5). Back-calculated ϵ_{Nd_i} values for 300 Ma range from +3.8 to +4.0, consistent with island arc or juvenile continental arc sources (White 2023). Six samples of the Moscovian and Gzhelian finer fraction exhibit more radiogenic $^{87}\text{Sr}/^{86}\text{Sr}$ values of 0.706405 to 0.726455, with less radiogenic ϵ_{Nd} values of –1.9 to –7.5, consistent with source regions containing components of older continental crust (Figs. 5, 6). All $^{87}\text{Sr}/^{86}\text{Sr}$ analytical data are tabulated and provided in Supplemental Table S5.

DISCUSSION

Grain Origins

The rounded shapes and commonly pitted surface textures of the rounded grains are consistent with a previous history of eolian saltation (Smith et al. 2018). In contrast, the occurrence of angular grains comprising high-silica volcanic glass, sanidine, orthoclase, quartz, (minimal) anorthite, and large lithic fragments of andesitic to rhyolitic compositions indicate an igneous (volcanic) source that ranges from intermediate to felsic, respectively. Hence, the shapes and compositions of the giant grains indicate two origins: 1) eolian transport from continental sources for the rounded grains (mostly quartz, subordinate feldspar), and 2) eolian fallout from volcanic sources for the angular grains of igneous composition.

Of the random sampling of giant grains analyzed, the ratio of volcanic to continentally derived grains averages 61:1 (ignoring the 5:1 ratio displayed

TABLE 1.—Details of dust data by study area.

Age	Number of Horizons Sampled	Number of Grains	Number of Grains Analyzed	Avg. Starting Mass (g)	Avg. Grain Concentration (grains/kg)	Largest Grain Long Axis (μm)	Avg. Long Axis (μm)	Number of Volcanic Grains per kg of Limestone	Number of Continental Grains per kg of Limestone	Volcanic: Continental Grains
Art.	100	2,626	966	272	96	1064	208	89	7	16:1
Ass.	21	606	121	300	116	1250	220	87	29	5:1
Gzh.	49	176	91	107	34	2500	367	32	2	34:1
Mos.	113	1150	135	228	45	20,000	260	44	1	134:1

Note: Time abbreviations used above are as follows: Mos., Moscovian (315–307 Ma); Gzh., Gzhelian (304–299 Ma); Ass., Asselian (299–294 Ma); Art., Artinskian (290–283 Ma).

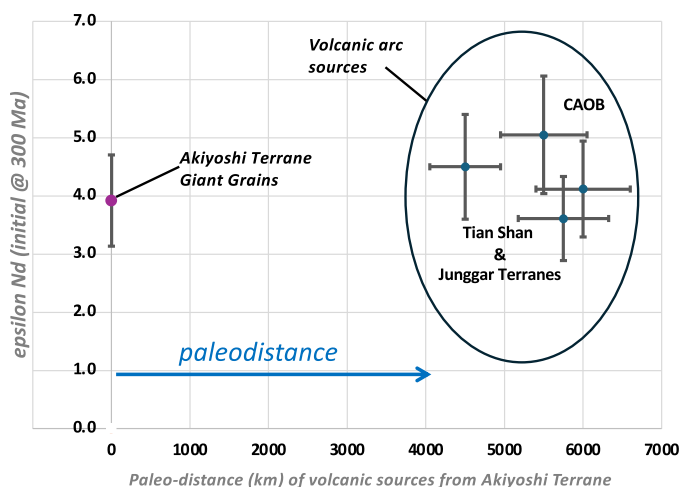


FIG. 6.—Age-corrected (~ 300 Ma) initial epsilon Nd for 3 Akiyoshi giant lithics, with comparisons to potential circum-Panthalassan volcanic arc sources (see refs). $^{143}\text{Nd}/^{144}\text{Nd}$ ratios for Akiyoshi Giant lithics were age-corrected to initial epsilon Nd values (ϵ_{Nd}) at $T = 300$ Ma and plotted at 0 km. These are compared to Late Carboniferous volcanic sources of eastern Pangaea (Nd isotope values for Junggar, Tian Shan, and Central Asian Orogenic (CAOB). Terranes are from various references, with 1 sd of Nd isotopic ranges shown. Estimated paleo-distances in km upwind from Akiyoshi Terrane have large uncertainties (see text). The three Akiyoshi giant lithics have average initial epsilon Nd (ϵ_{Nd}) = +4 with Nd model ages (Tdm, depleted mantle) of ~ 800 Ma (± 50 Ma) compatible with a mixed (mantle + crust) isotopic signature typical of continental arcs. Nd model ages and initial ratios were calculated using Sm/Nd ratios from unpublished REE data (estimated paleo-distances are poorly constrained but assumed to be > 4000 km; see text for references).

in the low-confidence Asselian) with ratios of 134:1, 34:1, and 16:1 for the Moscovian, Kasimovian–Gzhelian, and Artinskian, respectively (Table 1).

Provenance

Continental Provenance.—Rounded quartz and feldspar grains likely reflect deflation from arid to semi-arid regions capable of dust emission, and presumably in relatively low latitudes, given the equatorial setting of the Akiyoshi atoll. Candidate dust-emission regions should exhibit proxy evidence for arid or semiarid conditions (e.g., widespread evaporite, and/or arid–semiarid paleosols), and negative evidence for perhumid conditions (e.g., coal). Consequently, these grains are most likely sourced from a continental setting.

The nearest continental regions capable of potentially sourcing the dust are the North China and Tarim (NC&T) and South China (SC) blocks $\sim 1,500$ km (SC) to $\sim 4,500$ km (NC&T) distant, and west of the Akiyoshi atoll (Fig. 1). The South China block hosted peat (coal) and marine carbonate deposition from Moscovian through Asselian (~ 315 – 294 Ma) times (e.g., Peng et al. 1999; Boucot et al. 2013), precluding this area as a viable region for dust emission until the Artinskian (~ 290 Ma). While North China and Tarim also record coal deposition throughout the late Paleozoic (Boucot et al. 2013), evidence of fluctuating arid–humid or semiarid to humid climate at and above $\sim 30^\circ$ N occurs in the form of evaporites intercalated with mudstone, sandstone, and conglomerate in inferred floodplain strata of the Ahne coalfield, central North China, and the Fengchen Formation, Junggar Basin (Boucot et al. 2013; Wang et al. 2020; Li et al. 2021). Nodular gypsum in discrete horizons of the Ahne coalfield suggests intermittent aridity in North China during the Late Carboniferous–early Permian; however, a deltaic–alluvial–plain setting is typically not conducive to production of the widespread grain pitting resulting from eolian saltation (Li et al. 2021). Given the paleolatitude of North China and Tarim (20 – 45° N), this source would require transport from the extratropical region, and westerly

to northwesterly (monsoonal) atmospheric circulation over the Paleo-Tethys. The latter scenario is a persistent feature in model simulations (Heavens et al. 2012, 2015) and appears to be decoupled from the better-known Pangean megamonsoon (Kutzbach and Gallimore 1989; Heavens et al. 2012; Shields and Kiehl 2018). Alternatively, possible continental source regions also lie farther west of the continental margin of eastern equatorial Pangaea (Fig. 1), in (modern) Eastern Europe and north-central Africa; however, excepting parts of North Africa, these regions were perhumid through the Pennsylvanian, becoming more arid in the Permian (Schneider et al. 2006; Tabor and Poulsen 2008; Boucot et al. 2013; Michel et al. 2015). Additionally, emission from eastern equatorial Pangaea would require $> 10,000$ km of westward transport. Middle Pennsylvanian–lower Permian strata of western equatorial Pangaea record widespread evidence for the requisite aridity, including eolian strata (Kessler et al. 2001; Boucot et al. 2013), and this region lies upwind of the Akiyoshi atoll if zonal circulation prevailed, but implies $\sim 20,000$ km of atmospheric transport (Fig. 1).

Volcanic Provenance.—Volcanic components reflect derivation from sites of intermediate to felsic volcanism, and situated at latitudes $< 45^\circ$ in order to enable atmospheric transport to equatorial regions such as the Akiyoshi Terrane. Potential modern analog arc systems with comparable compositions include the Taupo Volcanic Zone (New Zealand) which displays a close affinity to a back-arc basin transitioning from oceanic to continental crust (McCulloch et al. 1994), the Japanese island-arc system (Hokkaido, Japan; Takanashi et al. 2012), and the Sierra La Primavera continental-margin arc system in Mexico (Mahood and Halliday 1988).

Proximity to the Akiyoshi atoll, volcanic compositions, and Nd and Sr isotope data were assessed against published data on active volcanism during the study intervals (compiled in Soreghan et al. 2019) to determine the most likely volcanic centers(s) that sourced the Akiyoshi grains, resulting in two options: eastern equatorial Pangaea and North China (Fig. 1). While the most proximal volcanism was that accompanying the accretion of the Akiyoshi atoll onto proto-Japan in the late Permian, Akiyoshi-associated volcanism dates to the early Permian (Minato et al. 1962), so cannot account for the evidence for volcanism observed in the Upper Carboniferous part of the section—including the giant lithic fragments. In contrast, the volcanism in the Tianshan, Junggar, Tarim, and Mongolian regions of the North China block and the Central Asian Orogenic Belt during the Moscovian–Artinskian study interval spans mafic–intermediate–felsic compositions (e.g., Su et al. 2012; Lai et al. 2014; Chen et al. 2020). Felsic material of Moscovian age from the Chinese regions displays $^{87}\text{Sr}/^{86}\text{Sr}$ isotope values of 0.702272–0.705580, and initial ϵ_{Nd} values of 2.8–7.4 (Su et al. 2012; Tong et al. 2018), overlapping values from the large lithic fragments recovered from the Moscovian section ($^{87}\text{Sr}/^{86}\text{Sr}$ of 0.705136–0.705260 and ϵ_{Nd} of 3.8–4.0; Figs. 5, 6). Abundant records of felsic (dacitic to rhyolitic) volcanism spanning all study intervals occur in eastern equatorial Pangaea (western-central Europe; e.g., Breitreuz and Kennedy 1999; Capuzzo and Busy 2000; Koniger et al. 2002; Breitreuz et al. 2007; Awdankiewicz et al. 2013). Available $^{87}\text{Sr}/^{86}\text{Sr}$ values for such volcanism ranges from 0.7100 to 0.7128, compared to the Akiyoshi giant lithic fragments with values of ~ 0.705 (Romer et al. 2001; Koniger et al. 2002) and ϵ_{Nd} values ranging from -6.7 to -7.0 , falling far outside the range of ϵ_{Nd} values ($+3.8$ to $+4.0$) of the Akiyoshi giant lithic fragments (Romer et al. 2001) (Fig. 5). However, these values do overlap with those of the finer fraction from the Akiyoshi dust, meaning that eastern equatorial Pangean volcanism cannot be excluded as a potential source for this fraction. The most parsimonious interpretation, accounting for both compositional and age data, suggests that the volcanism between the northern margin of the North China Block and the southern margin of Tarim, and in the Central Asian Orogenic Belt, likely sourced the giant lithic fragments to the Akiyoshi Terrane but could not have been the sole source of all volcanic material. The wide range of both $^{87}\text{Sr}/^{86}\text{Sr}$ and ϵ_{Nd} suggests a mixing of multiple distal sources, and

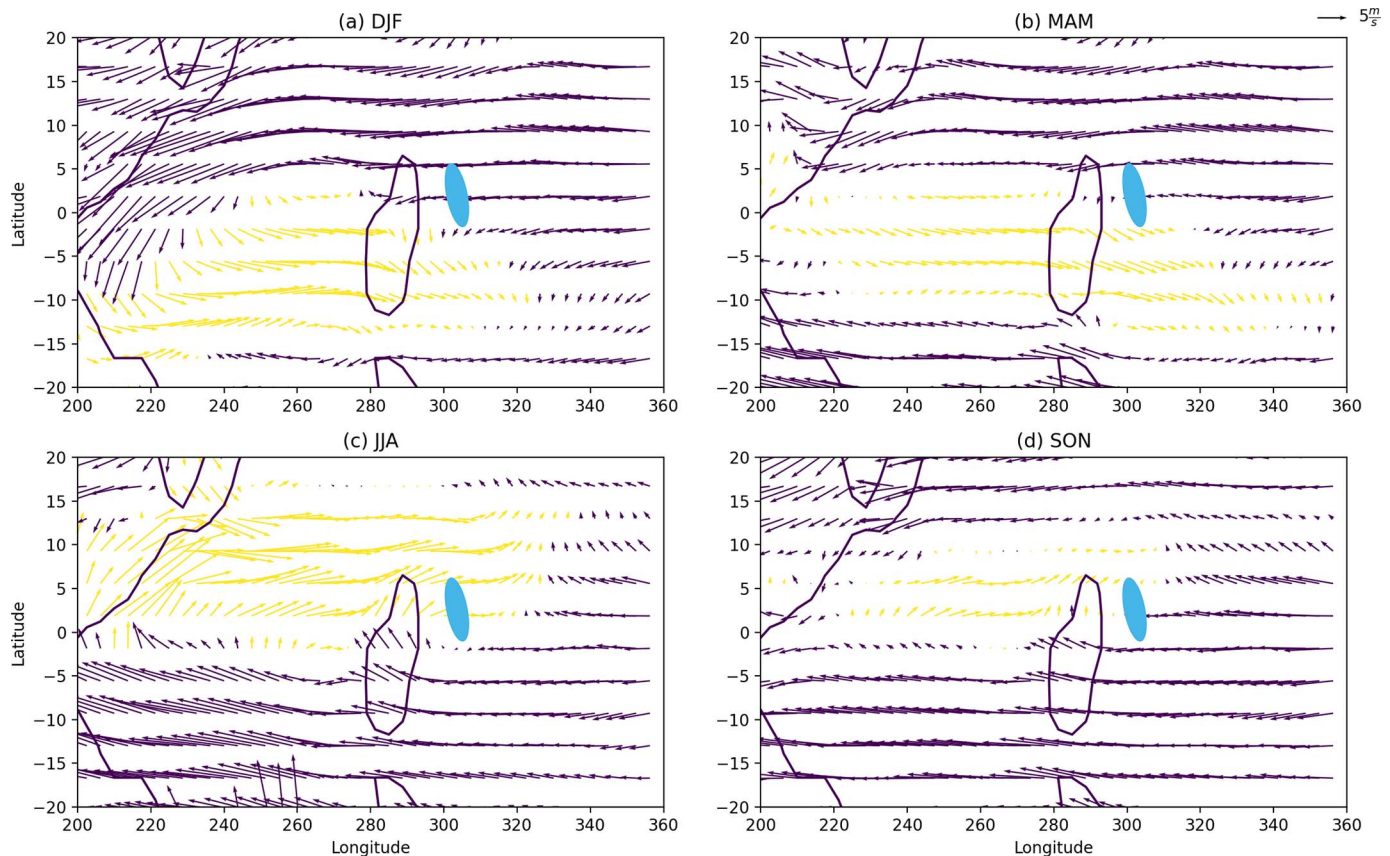


FIG. 7.—Mean winds at 850 hPa (~ 1.2 km altitude) for the four labeled seasons (DJF, December, January, February, etc.) in the icehouse,glaciation,polar experiment of Heavens et al. (2015). To emphasize the monsoonal wind reversals, easterly winds are colored purple and westerly winds are colored yellow. Data are available from Heavens (2018).

furthermore implies the operation of both northwesterly (China block) and westerly (eastern equatorial Pangaea) circulation.

Prevailing atmospheric wind directions and velocities play large roles in controlling tephra fallout (Fischer and Schminke 1984; Woods 1995; Jenkins et al. 2015). Thus, the likely sourcing of volcanic dust that included both large lithic fragments as well as finer dust to the Akiyoshi Terrane during the Moscovian implies a significant component of westerly circulation, reinforcing the hypothesis of monsoonal circulation over the Paleo-Tethyan region. Western equatorial Pangaea is less favored as a potential continental source because it would require prevailing zonal easterlies while the most likely source for the (distinctive) giant lithic fragments requires westerlies, and a significantly longer transport distance.

Although most instances of giant grains documented in the literature represent continental dust sourced from the Sahara and Asia, Ram and Gayley (1991) and Lundberg and McFarlane (2012) documented volcanic grains (up to 300 μm and 190 μm , respectively) at least 1,000 km from their sources, and noted the implication of significant stratospheric injection requiring a high (~ 7) volcanic explosivity index (VEI; Fischer and Schminke 1984). Analogously, the occurrence in the Akiyoshi Limestone of lithics of up to 2 cm diameter that traveled $\sim 4,500$ km requires stratospheric injection, likely together with other mechanisms to extend atmospheric transport distances. Van der Does et al. (2018) noted four potential mechanisms capable of enhancing atmospheric transport of giant grains: 1) high atmospheric windspeeds, reducing the time it takes for a particle to travel a given distance, 2) turbulence resulting in repeated re-lofting of particles, thus extending residence time in the atmosphere, 3) triboelectrification, in which collision of like-charged particles forms a “charged cloud” capable of increasing buoyancy

of particles, 4) tropical-cyclone events, in which particles can be ejected from the top of the storm system to higher atmospheric altitudes.

Implications for Paleoclimate

As documented above, mineralogic, grain-shape, and isotopic evidence indicates influx of both continental and volcanic particles to the Akiyoshi atoll. Both continental grains of up to 200 μm (long axis) transported from North China ($\sim 4,500$ km distant) and/or eastern equatorial Pangaea ($\sim 10,000$ km) require westerly circulation at low latitudes. Large lithics transported from between the northern margin of North China and the southern margin of Tarim ($> 4,500$ km) range in size from 0.5 to 2 cm, while the remaining volcanic grains of up to 1–2 mm (long axis) were likely transported from multiple sources including eastern equatorial Pangaea and the North China–Tarim region — again, both requiring westerly atmospheric circulation. Westerly flow at the equatorial latitude of the Akiyoshi atoll implies the presence of a monsoonal circulation pattern (Riehl 1954). Accepting Heavens et al.’s (2012, 2015) model results of monsoonal circulation over Paleo-Tethys decoupled from that over Pangaea (but sometimes occurring simultaneously), summer westerlies would have been a sustained feature over the Akiyoshi atoll during late Paleozoic time (Fig. 7).

The idea of monsoonal circulation away from coastal waters seems counterintuitive to the experience of monsoons in the Holocene, where monsoonal circulations like the Indian Monsoon, East Asian Monsoon, East African Monsoon, etc., are associated with and driven by strong land–sea temperature contrasts (e.g., Fasullo 2012). Passing over the question of whether surface thermal contrasts are the principal driver of the monsoons (e.g., Geen et al. 2020), it has been known since at least

Chao and Chen (2001) that longitudinal sea-surface temperature gradients can drive monsoonal circulations just as much as longitudinal contrasts in temperature between the ocean and nearby land areas. The more difficult question is how much monsoonal circulation over Paleo-Tethys and the adjacent Panthalassic Ocean is an extension of monsoonal circulation over Pangea. Heavens et al. (2015) argued that they are decoupled. In contrast, Shields and Kiehl (2018), who used the same model as Heavens et al. (2012, 2015) but applied to the latest Permian (251 Ma), interpreted monsoonal circulation over Pangea, Paleo-Tethys, and the adjacent Panthalassic as the same circulation. One significant point to be drawn from Shields and Kiehl (2018) is that removing North China from the paleogeographic boundary conditions compresses the extent of tropical westerlies closer to Pangea.

Assuming minimal variability in the strength or seasonal duration of westerly flow over the Akiyoshi atoll, significant variability in both the concentration and origin (volcanic versus continental) of the grains through time (Table 1) reveals information on trends in explosive volcanism and continental aridity of source regions. The exclusive presence of giant (up to 2 cm) lithic fragments in the Moscovian interval suggests that, while more giant grains accumulated in the Akiyoshi Limestone during the early Permian, highly explosive volcanism was more common in the Middle Pennsylvanian. The apparent twofold increase in volcanically sourced grain concentrations in the Permian relative to the Pennsylvanian thus may indicate more efficient transport of volcanic giant grains. This increase in transport efficiency is most likely a result of the proto-Japanese arcs and the Ailaoshan volcanic belt volcanism, which transitioned to more felsic (and generally more explosive) compositions in the early Permian, corresponding to the predominantly felsic composition of grains extracted from the Akiyoshi Limestone (Minato et al. 1962; Lai et al. 2014). These regions are not viable sources for continental grains, indicating that while potential sources of volcanic grains were closer to the Akiyoshi atoll during the Permian than during the Late Carboniferous, potential sources of aridity remained farther removed. Even compensating for this effect, the threefold to tenfold apparent increase in continentally sourced grains in the Permian implies a 50–400% increase in giant-grain emission at source, suggesting that viable dust-bearing continental sources expanded through time, consistent with widespread continental aridification in low latitudes by Artinskian (early Permian) time (Boucot et al. 2013).

CONCLUSIONS

Data from the Carboniferous–Permian Akiyoshi Limestone establishes the first documentation of long-range atmospheric transport of giant grains in Earth's pre-Quaternary record. The Akiyoshi Limestone trapped detrital siliciclastic particles that commonly range up to ~ 300 µm in diameter and originated from both continental and explosive volcanic sources located 10^3 – 10^4 km from the Akiyoshi atoll. The rare but reproducible occurrence of especially large lithic fragments up to 2 cm in diameter record remarkably explosive volcanism in Moscovian time, and fine material includes preserved vesicular glass. The increasing proportion of continentally derived material with time is consistent with the known progression of aridity of the Pangean supercontinent. Provenance data indicate persistent westerly transport to the equatorial Akiyoshi atoll, suggesting the operation of a Paleo-Tethyan monsoon.

The Akiyoshi Limestone served as a remarkable dust trap in the vast Panthalassic Ocean, capturing snapshots of both instantaneous events (eruptions) and climatic evolution of distant regions. In addition to more typical fine-grained dust in deep time, giant-grain records such as this represent a powerful tool for exploring extremes of volcanism and eolian transport in deep time.

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