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Lead isotopes in New England (USA) volcanogenic massive sulfide deposits: implications for metal sources and pre-accretionary tectonostratigraphic terranes

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Abstract

Lead isotope values for volcanogenic massive sulfide (VMS) deposits provide important insights into metal sources and the nature of pre-accretionary tectonostratigraphic terranes and underlying basements. Deposits of this type in New England formed in diverse tectonic settings including volcanic arcs and backarcs, a supra–subduction zone arc, a rifted forearc foreland basin, and a rifted continental margin. Following VMS mineralization on or near the seafloor, components of the tectonostratigraphic assemblages—volcanic ± sedimentary rocks, coeval intrusions, sulfide deposits, and underlying basements—were diachronously accreted to the Laurentian margin during the Paleozoic. Lead isotope data for galena show relatively large ranges for $^{206}Pb/^{204}Pb$, $^{207}Pb/^{204}Pb$, and $^{208}Pb/^{204}Pb$. Evaluation of potential lead sources, using for comparison Pb-isotope data from modern and ancient settings, suggests that principal sources include the mantle, volcanic ± sedimentary rocks, and deeper basement rocks. Integration of the Pb-isotope values with published data such as Nd isotopes for the volcanic rocks and from deep seismic reflection profiles points to the involvement of several basements, including those of Grenvillian, Ganderian, Avalonian, and West African (and (or) Amazonian) affinity. Clustering of Pb-isotope data for VMS deposits within individual Cambrian and Ordovician volcanic and volcanosedimentary settings, delineated by differences in $^{206}Pb/^{204}Pb$ and μ ($^{238}U/^{204}Pb$) values, are consistent with lead derivation from at least four and possibly five different tectonostratigraphic assemblages with isotopically distinct basements. Collectively, our Pb-isotope data for New England VMS deposits provide a novel window into the nature of subarc basement rocks during pre-accretionary sulfide mineralization outboard of Laurentia during early Paleozoic time.

Key words: New England, Pb-isotopes, metal sources, volcanogenic massive sulfides, tectonostratigraphic terranes, Proterozoic basements

Introduction

Studies of tectonostratigraphic terranes have provided fundamental insights into Earth processes for more than four decades (e.g., Coney et al. 1980; Hsü et al. 1990; Nance and Murphy 1994; Shelley and Bossière 2000; Hibbard et al. 2006; van Staal et al. 2009; Papanikolaou 2013; Golozubov and Simanenko 2021). Tectonostratigraphic terranes are regionally extensive, fault-bounded geologic blocks characterized by a distinctive stratigraphic sequence or rock assemblage that differs significantly from those of adjacent or proximal blocks. Within the New England Appalachians, constraints on the distribution and origin of tectonostratigraphic terranes have come from diverse types of data, including (1) differences in lithology and metamorphic grade (e.g., Zen 1983; Fyffe et al. 1988; Keppie 1989; Wintsch et al. 1992; Pollock 1993; Spear et al. 2008), (2) faunal assemblages (e.g., Skehan et al. 1978; Neuman 1984; Pollock et al. 1994; Poole and Neuman 2003), (3) chemical and isotopic compositions of volcanic rocks and granitic plutons (e.g., Ayuso 1986; Dorais and Paige 2000; Moench and Aleinikoff 2003; Tomascak et al. 2005; Aleinikoff et al. 2007; Potter et al. 2008; Fu et al. 2014), (4) regional gravity and magnetics (e.g., Zen 1983; Lyons et al. 1996; Li et al. 2018), (5) deep seismic reflection data (e.g., Unger et al. 1987; Spencer et al. 1989; Luo et al. 2021, 2022), (6) paleomagnetic signatures (e.g., Kent and Opdyke 1980; Potts et al. 1995; Waldron et al. 2022), (7) U–Pb ages of igneous and detrital zircons (e.g., Tucker et al. 2001; McWilliams et al. 2010; Macdonald et al. 2014; Karabinos et al. 2017; Reusch et al. 2018; Waldron et al. 2019; Walsh et al. 2021; Kuiper et al. 2022), and (8) Lu–Hf data for detrital and magmatic zircons (e.g., Pollock et al. 2022). Despite this wealth of information, however, major questions remain with respect to certain aspects of the terranes, including locations of current boundaries and pre-accretionary settings. In particular, a long-standing challenge in New England has been delineating the main Iapetus suture zone that separates peri-Laurentian from peri-Ganderian terranes (Moench and Aleinikoff 2003; Aleinikoff et al. 2007; Dorais et al. 2012*a*; Macdonald et al. 2014; Karabinos et al. 2017; Valley et al. 2020; van Staal et al. 2021*b*; Waldron et al. 2022). This study, expanded from preliminary reports (LeHuray and Slack 1985; Swinden et al. 1988; Slack et al. 1991), presents new Pb-isotope data for galena and other sulfide minerals (pyrrhotite and chalcopyrite) from stratabound volcanogenic massive sulfide (VMS) deposits in New England that yield insights into the sources of lead and constraints on basement rocks underlying Paleozoic volcanosedimentary sequences in the region.

VMS deposits are important global sources of Zn, Cu, Pb, Ag, and Au. Based on extensive studies of ancient and modern deposits, mineralization is widely attributed to formation on and beneath the seafloor from moderate-to high-temperature (200–350 °C) hydrothermal solutions, in which solid and fluid components come from a variety of sources including volcanic and sedimentary rocks, magmas, and seawater (Franklin et al. 2005; Galley et al. 2007; Hannington 2014). Base and precious metals, including lead, are interpreted to derive via the hydrothermal leaching of underlying volcanic ± sedimentary rocks and in some cases deeper basement rocks (Doe and Zartman 1979; Sundblad and Stephens 1983; Bjørlykke et al. 1993; Ayuso et al. 2007; Tessalina et al. 2016; Gill et al. 2019). The formation of VMS deposits typically occurs in extensional regimes within mid-ocean ridges (MORs), volcanic arcs, volcanic back arcs, and intracontinental rifts. Various rift-related processes and associated shallow-level magmatism provide, respectively, major plumbing systems and the sources of heat (and possibly metals) responsible for mineralization. Lead isotope signatures of VMS deposits represent an integrated composition of all sources that contributed lead to the mineralizing hydrothermal fluids, with Pb-isotope heterogeneity reflecting the extent of isotopic variations among the rocks and sediments in the source areas. Importantly, Pb-isotope signatures of VMS deposits record pre-accretionary and in some cases syn-accretionary tectonic settings (Slack and Swinden 1988; Potra and Macfarlane 2014; Goldfarb et al. 2021) and are generally unaffected by post-ore processes such as metamorphism (e.g., Yu et al. 2020). A key exception involves tectonic or igneous remobilization of sulfides, including galena, into veins or fault zones, as such processes may in some cases change Pb-isotope compositions if isotopically different lead is introduced into the vein/fault system (Brevart et al. 1982; Wagner and Schneider 2002; Zhong and Li 2016).

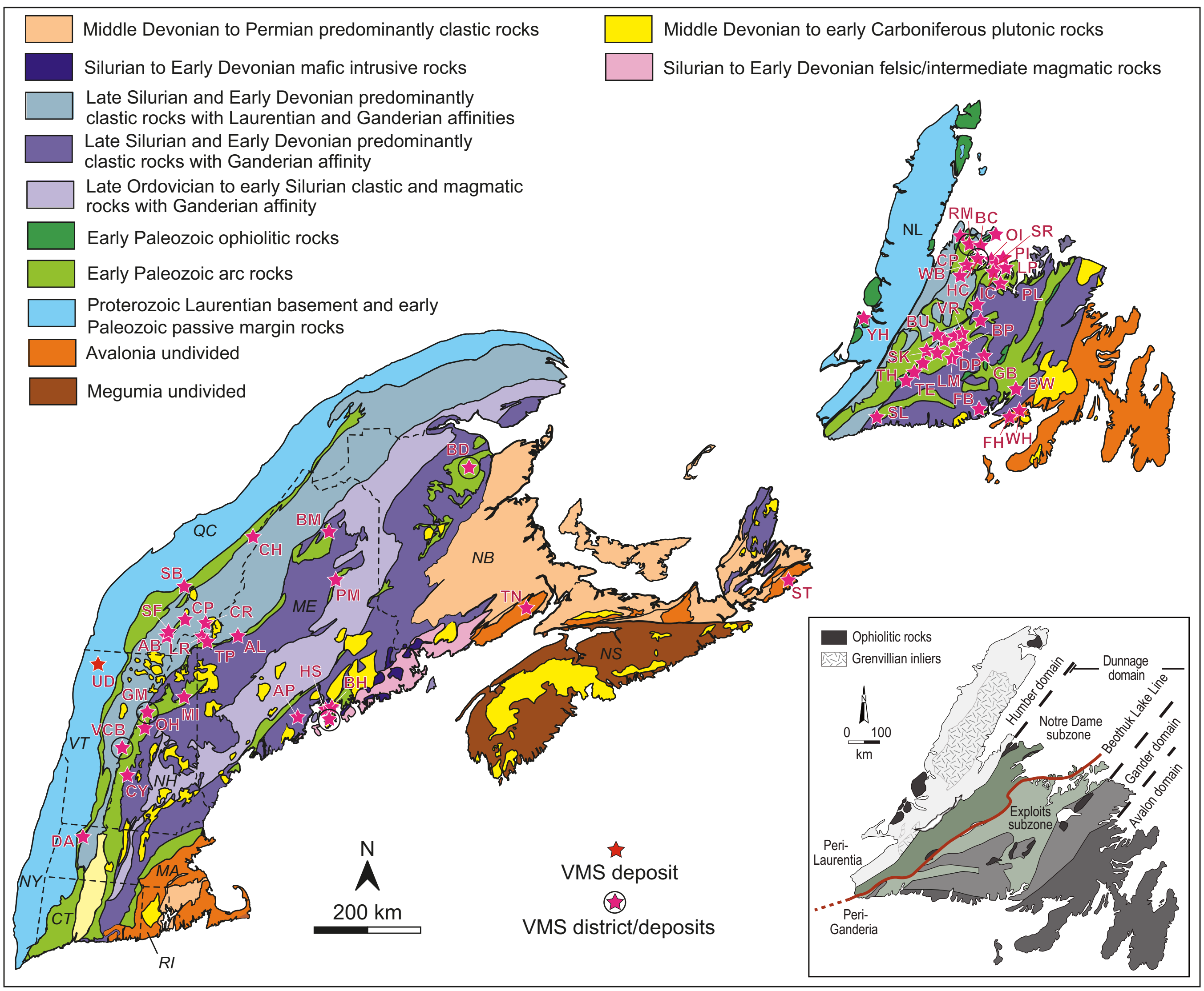
The lithotectonic terminology of the Appalachian orogen used in this paper is revised from the original tectonostratigraphic zonal divisions of Williams (1979) and Williams et al. (1988). These zonal divisions were replaced by realms and domains by Hibbard et al. (2007) and locally further subdivided in the various recognized constituents by van Staal and Barr (2012). The original Humber Zone of Williams (1979) is referred to as the Humber margin of the Laurentian realm. The Notre Dame subzone of the Dunnage Zone became the peri-Laurentian arc–backarc tract (Zagorevski et al. 2008) of the oceanic Dunnage domain, whereas the Exploits subzone became the peri-Ganderian arc–backarc tract (Valverde-Vaquero et al. 2006; Zagorevski et al. 2010) of the oceanic Dunnage domain. The Gander Zone and Proterozoic rocks of the Hermitage flexure together are referred to as the Gander margin, which combined with the associated peri-Ganderian arc–backarc tract of the Dunnage domain represent Ganderia (van Staal et al. 2021*b*). The Avalon Zone is referred to as the Avalonia domain, or Avalonia for short (van Staal et al. 2021*a*). Avalonia covers the same rocks as the original Avalon Zone of Williams (1979), except for small parts of Atlantic Canada and coastal Maine, which are now grouped with Ganderia (van Staal et al. 2021*b* and references therein). *Terrane*, as defined above, refers to regionally extensive blocks with characteristics that differ from those of adjacent blocks. *Assemblage* is used here for volcanic and volcanosedimentary belts with similar or different basements that may have travelled independently during plate tectonic processes, as distinguished from *terrane assemblage* as defined by Waldron et al. (2022) for volcanic and volcanosedimentary units that arrived at the Laurentian margin together with or without an identical basement.

Regional geological setting

The bedrock geology of the northern Appalachian orogen records the opening and subsequent closing of an early Paleozoic ocean termed Iapetus (Wilson 1966; Harland and Gayer 1972). This orogen preserves the Neoproterozoic to early Paleozoic continental margins of the ocean, Gondwana and Baltica to the east and Laurentia to the west, respectively, and a complex series of Cambrian to Devonian oceanic volcanic and sedimentary arc/back arc sequences, including ophiolite complexes in Newfoundland, New Brunswick, southeastern Québec, and western Maine (e.g., Robinson et al. 1998; Zagorevski et al. 2015; Tremblay and Pinet 2016). These oceanic tracts and ophiolites formed in the Iapetus and later Rheic oceans and were accreted to the Laurentian margin in a series of early Paleozoic orogenies culminating with the arrival of Gondwana and final closure of these oceans in the Carboniferous–Permian (Nance et al. 2010; van Staal et al. 2012; Walsh et al. 2021; Waldron et al. 2022). Intrusive rocks were emplaced episodically throughout the history of the orogen, and successor basins developed on the newly cratonized continental crust. Figure 1 is a generalized lithotectonic map of the northern part of the orogen showing the locations of economically or geologically important VMS deposits and occurrences.

Early attempts to understand the varied and complex geological relationships in the northern Appalachians led to the recognition of five principal tectonostratigraphic zones, based on stratigraphic and structural contrasts between Cambrian–Ordovician and older rocks (Williams 1979; Williams and Hatcher 1983). These zones were first proposed

Fig. 1. Generalized lithotectonic map of the northern Appalachian orogen showing locations of volcanogenic massive sulfide (VMS) deposits. Geology modified from Hibbard et al. (2006), van Staal et al. (2021*b*), and Kuiper et al. (2022); inset map modified after Lode et al. (2017). VMS deposits from Emmons (1910); Gair and Slack (1979, 1980), Sangster (1980); Swinden and Thorpe (1984), McCutcheon et al. (1993), Gauthier et al. (1994), Sears and Wilton (1996), Moench et al. (1999), Piercey (2007), Mosier et al. (2009), and Gill et al. (2016). Key to deposit labels: Newfoundland: BC, Betts Cove; BP, Burnt Pond (includes Moose Pond and South Moose Pond); BU, Buchans district (includes 12 deposits); BW, Barasway de Cerf; CP, Catchers Pond; DP, Duck Pond (includes Tally Pond and Boundary); FB, Facheux Bay; FH, Frenchman Head; GB, Great Burnt Lake; HC, Handcamp; IC, Indian Cove (includes Seal Bay); LM, Lemarchant; LP, Lockport; OI, Oil Islands; PI, Pilleys Island (includes Bull Road); PL, Point Leamington; RM, Rambler and Ming; SK, Skidder; SL, Strickland; SR, Shamrock; TE, Tucks Hill East; TH, Tucks Hill (includes Side of the Hill); VR, Victoria; WB, Whalesback (includes Little Bay); WH, Winter Hill; YH, York Harbour. New Brunswick: BD, Bathurst district (includes 25 deposits); TN, Teahan. Nova Scotia: ST, Sterling. Québec: AB, Albert; CH, Champagne; CP, Cupra; CR, Clinton River; SB, Solbec; SF, Suffield. Maine: AL, Alder Pond; AP, Appleton; BH, Black Hawk; BM, Bald Mountain; HS, Harborside; LR, Ledge Ridge; PB, Penobscot Bay deposits (Hercules, Eggemoggin, Deer Isle, and North Castine); PM, Pickett Mountain (formerly Mount Chase); TP, Thrasher Peaks. New Hampshire: CY, Croydon; GM, Gardner Mountain (includes Gardner Mountain, Paddock, and Royce); MI, Milan; OH, Ore Hill. Vermont: UD, Udall; VCB, Vermont copper belt (includes Elizabeth, Ely, Pike Hill, and Gove). Massachusetts: DA, Davis. Some deposits are small prospects that have not been explored or delineated. Note that Proterozoic basement rocks of Laurentian and Ganderian affinity are not shown (see Fig. 2). In inset map, Beothuk Lake Line is equivalent to Red Indian Line that is no longer used (see text).



in the Newfoundland sector of the orogen where structural and metamorphic complications are less intense and were quickly expanded to include the entire orogen as well as the Caledonides of northern Europe (e.g., van Staal et al. 1998; Waldron et al. 2022). The zonal subdivision in Newfoundland is shown in the inset to Fig. 1. Tectonostratigraphic Zones were subsequently redefined as Domains by Hibbard et al. (2007) and below we follow this terminology.

From west to east, the tectonostratigraphic domains comprise the (1) Humber domain: Cambrian to Ordovician passive-margin sedimentary rocks deposited on Grenvillian-age (1.3–0.95 Ga) basement at the Laurentian continental margin; (2) Dunnage domain: Cambrian to Silurian oceanic metavolcanic and metasedimentary rocks deposited in Iapetus. The Dunnage domain is subdivided based on contrasting stratigraphy, structure, fauna, and isotopic and geophysical signatures into a western Notre Dame peri-Laurentian arc–backarc tract and an eastern Exploits peri-Ganderian arc–backarc tract, separated by a major structural boundary termed the Beothuk Lake Line (BLL) that formerly was termed the Red Indian Line (RIL) (Williams et al. 1988), which is not used here because the Newfoundland and Labrador Government in 2021 officially changed the name of Red Indian Lake to Beothuk Lake (www.assembly.nl.ca/HouseBusiness/Bills/ga50session1/bill2112.htm); (3) Gander domain: Cambrian to Devonian metasedimentary and lesser metavolcanic rocks formed at and near the Ganderian continental margin of Iapetus; (4) Avalon domain: Neoproterozoic to Devonian metavolcanic and metasedimentary rocks that, within its Proterozoic history, may represent an orogenic cycle that preceded Iapetus; and (5) Meguma domain: Neoproterozoic (?) to Devonian metasedimentary and minor metavolcanic rocks. Mesoproterozoic basement rocks of Laurentian affinity occur as inliers in parts of the Humber and Gander domains.

All tectonostratigraphic domains except the Meguma domain are represented in the bedrock geology of New England (Hibbard et al. 2006; Pollock et al. 2012). Major tectonic boundaries such as the Baie Verte–Brompton Line (BVBL) between the Humber and Dunnage domains and the BLL, which separates peri-Laurentian rocks of the Notre Dame peri-Laurentian arc–backarc tract from rocks of the Exploits peri-Ganderian arc–backarc tract (Fig. 2), can be traced throughout the northern Appalachians, including Maine, New Hampshire, Vermont, Massachusetts, and Connecticut (e.g., van Staal et al. 1998; Aleinikoff et al. 2007; Dorais et al. 2012*a*; Valley et al. 2020; van Staal et al. 2021*b*; Waldron et al. 2022). The Liberty–Orrington Line in southern Maine and New Brunswick is a major suture between Cambrian–Ordovician metamorphic rocks of the Miramichi and Liberty Orrington belts to the north and Lower Silurian rocks of the Fredericton Trough to the south (van Staal et al. 2009 and references therein). A simplified Paleozoic geological history of the region began with successive accretion of multiple Cambrian to Early Devonian volcanic arcs to the Precambrian Laurentian margin of North America, with concurrent and later (Late Devonian to Carboniferous) deposition of sedimentary rocks in forearc, marginal, and successor basins (e.g., van Staal et al. 2009; Hibbard et al. 2010; Wilson et al. 2017). In New England, pre-Devonian strata are dominated by metamorphosed volcanic and sedimentary rocks, intruded locally by Ordovician to Cretaceous plutons chiefly of granitic or gabbroic composition (e.g., Robinson et al. 1998). Regional metamorphic grade increases from subgreenschist (prehnite–pumpellyite facies) in northern Maine southward through greenschist and amphibolite facies, to sillimanite + K-feldspar facies, including the development of migmatites in southwestern Maine and areas to the south, including in New Hampshire, Massachusetts, and Connecticut.

Globally, lead isotope ratios of ore deposits have been widely used to constrain tectonostratigraphic units and crustal boundaries (Brevart et al. 1982; LeHuray 1982, 1986, 1989; LeHuray and Slack 1985; LeHuray et al. 1987; Williams et al. 1988; Macfarlane et al. 1990; Bjørlykke et al. 1993; James and Henry 1993; Keppie and Ortega-Gutierrez 1995; Tosdal et al. 1999; Mortensen et al. 2008; Huston et al. 2014; Blichert-Toft et al. 2016; Champion and Huston 2016). We follow the approach of these and other workers in using Pb isotopes of VMS deposits to evaluate lead sources and the tectonostratigraphic settings of New England terranes during the early Paleozoic. Compiled for comparison are Pb-isotope values for 185 samples from 68 VMS deposits in the Canadian Appalachians (Table S1). Importantly, previous studies have documented a paleotectonic provincialism of Pb-isotope values in VMS deposits of Newfoundland and demonstrated that such data reflect fundamentally different lead sources from opposite sides of the Iapetus and Rheic oceans (e.g., Swinden and Thorpe 1984; Piercey 2007).

VMS deposits

VMS deposits occur throughout the northern Appalachian orogen and range in age from Neoproterozoic to Early Devonian (Fig. 1). In Newfoundland and New Brunswick, the VMS deposits occur mainly in Cambrian and Ordovician volcanic and sedimentary sequences of the Dunnage domain. By far the largest number of deposits is in rocks associated with Cambrian to Silurian volcanic arc and backarc sequences, known from both peri-Laurentian and peri-Ganderian settings. A small number of relatively small deposits is also present in Neoproterozoic volcanic sequences of the Avalon domain (e.g., Winter Hill, Newfoundland; Stirling, Nova Scotia; and Teahan, New Brunswick) that predate the opening of Iapetus.

Table 1 lists data for the principal VMS deposits and occurrences of New England for which the ages of host rocks range from Middle Cambrian to Early Devonian (Gair and Slack 1979, 1980). These metal accumulations occur over a large region of the Dunnage and Gander domains in Massachusetts, Vermont, New Hampshire, and Maine. From west to east (Figs. 1 and 2), these are (1) the Upper Silurian–Lower Devonian Connecticut Valley–Gaspé trough (CVGT) composed of clastic and carbonate metasedimentary rocks with very minor tholeiitic metabasalt that accumulated in a retroarc foreland basin (Slack 1994; Tremblay and Pinet 2005; Rankin et al. 2007; McWilliams et al. 2010; Perrot et al. 2018); (2) the composite Cambrian (?) to Ordovician Shelburne Falls–Bronson Hill belts that comprise siliciclastic metasedimentary rocks and overlying calc–alkaline and tholeiitic metavolcanic rocks of arc affinity (Aleinikoff 1977; Hollocher 1993; Kim and Jacobi 1996; Moench and Aleinikoff 2003; Gerbi et al. 2006; Karabinos et al. 2017; Valley et al. 2020); (3) the Ironbound Mountain (Silurian) and Second Lake (Lower Devonian) volcanosedimentary belts that formed in rifted arc settings (Moench et al. 1999); (4) the Jim Pond volcanic belt de-

Fig. 2. Simplified lithotectonic map of New England, southeastern Québec, and western New Brunswick showing principal volcanic and volcanosedimentary arcs and volcanogenic massive sulfide (VMS) deposits. Note that CVGT (Connecticut Valley–Gaspé Trough) contains Upper Silurian–Lower Devonian cover strata that formed above both Grenvillian and peri-Laurentian rock assemblages (western New England and northern New Hampshire) and above Gondwanan and peri-Ganderian rock assemblages (southeastern Québec and northwestern Maine). Gray region of peri-Ganderian rock assemblages is considered part of the Exploits peri-Ganderian arc–backarc tract (see text). Geology modified from Karabinos et al. (2017), Valley et al. (2020), West et al. (2021), van Staal et al. (2021*b*), and Kuiper et al. (2022); Neoproterozoic basement rocks of Ganderian affinity after Dorais et al. (2012*b*); Ironbound Mountain and Second Lake volcanic belts (after Moench et al. 1999) include correlative Clinton River belt in southeastern Québec. Avalonia includes St. Croix terrane of southern Maine and southern New Brunswick, and New River–Annidale and Brookville terranes of southern New Brunswick. Shelburne Falls arc locally includes metasedimentary rocks of Moretown terrane, mainly in Connecticut. Post-Acadian stratified rocks and igneous plutons are not shown. Boundaries of most major tectonostratigraphic terranes and regional faults after van Staal et al. (2021*b*) and Kuiper et al. (2022). Abbreviations: BLL, Beothuk Lake Line (=Red Indian Line); BVBL, Baie Verte–Brompton Line; CD, Clinton dome; CMT, Central Maine Trough; LD, Lyme dome; LOL, Liberty–Orrington Line; MGC, Massabesic Gneiss Complex; PD, Pelham dome; WD, Willimantic dome. Dashed east–west line for BLL in northern Vermont from van Staal et al. (2021*b*); BLL and BVBL merge in southern Vermont and are mainly coincident from there to the south in western Massachusetts and western Connecticut (Valley et al. 2020). Heavy dotted lines mark boundaries of CVGT and related strata. Deposit labels as in Fig. 1.

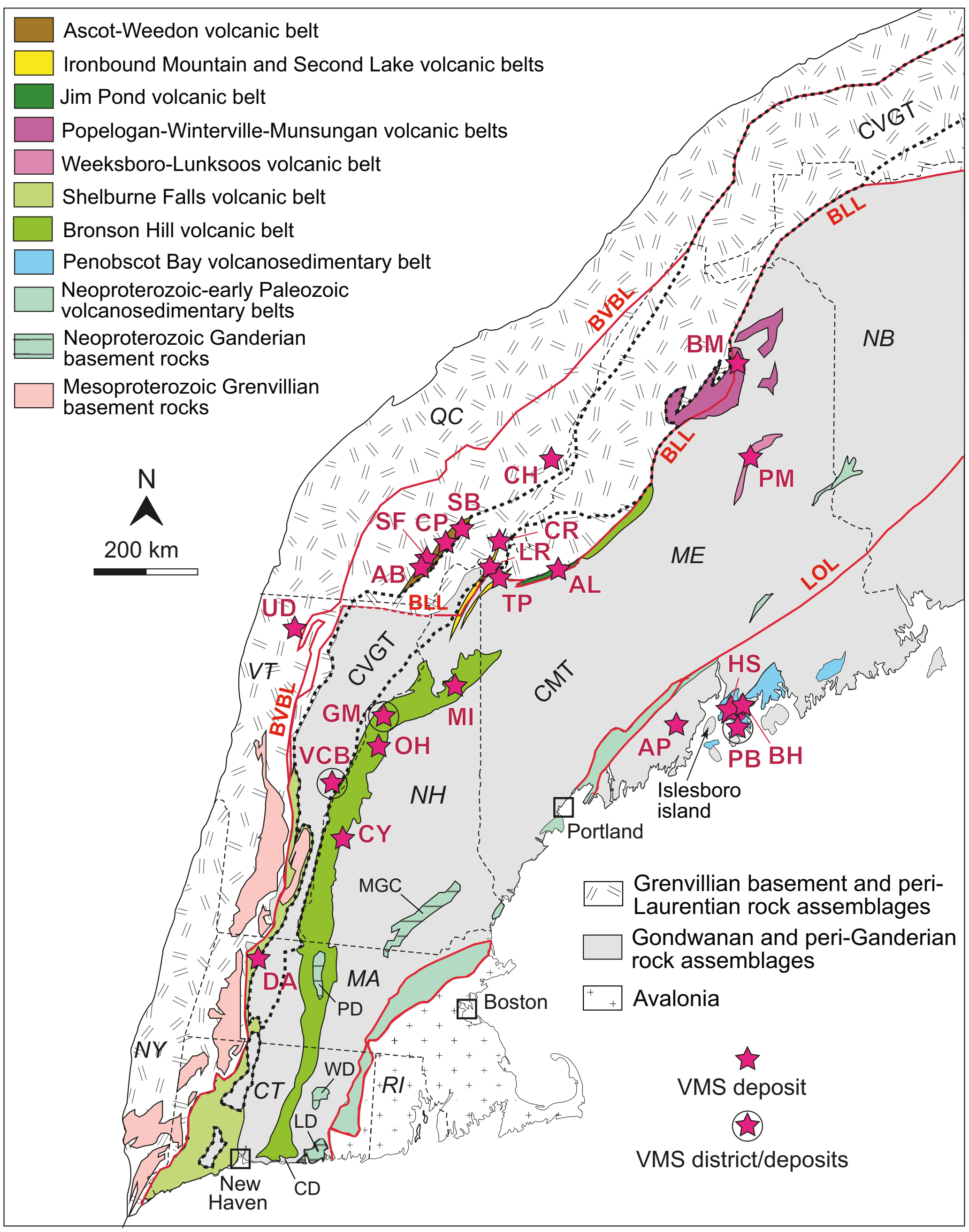


Table 1. Geological and Pb-isotope data for New England volcanogenic massive sulfide deposits.

Sample No.	Deposit name[1]	Metal signature	Deposit size	Deposit age[2]	Type[3]	Belt[4]	References[5]	Analyzed material	$^{206}Pb/^{204}Pb$	$^{207}Pb/^{204}Pb$	$^{208}Pb/^{204}Pb$	μ	Data source[6]
MC#1	Pickett Mountain (ME)[7]	Zn–Pb–Cu–Ag–Au	6.3 Mt	Middle Ordovician	BM	WL	1, 2	Galena	18.244	15.660	38.168	9.99	1
MC#2	Pickett Mountain (ME)[7]	Zn–Pb–Cu–Ag–Au	6.3 Mt	Middle Ordovician	BM	WL	1, 2	Galena	18.244	15.659	38.177	9.99	1
JS-80-61	Thrasher Peaks (ME)	Zn–Cu–Ag	~2.0 Mt	Early Devonian	BF	BH	3, 4, 5	Galena	17.970	15.547	37.949	9.56	1
LRx-1-1550	Ledge Ridge (ME)	Zn–Cu–Pb–Ag–Au	4.0 Mt	Silurian	M	BH	4, 5	Whole ore	17.960	15.515	37.757	9.42	1
AP-1	Alder Pond (ME)	Zn–Cu–Pb–Ag	3.4 Mt	Early Ordovician	BM	JP	5, 6	Galena	17.659	15.391	37.419	8.94	1
JS-80-18 A	Bald Mountain (ME)	Cu–Zn–Ag–Au	30 Mt	Middle Ordovician	BM	PL	7, 8, 9, 10	Galena	18.148	15.608	38.022	9.78	1
JS-80-18	Bald Mountain (ME)	Cu–Zn–Ag–Au	30 Mt	Middle Ordovician	BM	PL	7, 8, 9, 10	Galena	18.144	15.600	37.987	9.75	1
BM23-360	Bald Mountain (ME)	Cu–Zn–Ag–Au	30 Mt	Middle Ordovician	BM	PL	7, 8, 9, 10	Galena	18.154	15.601	37.998	9.75	2
BM17-299.5	Bald Mountain (ME)	Cu–Zn–Ag–Au	30 Mt	Middle Ordovician	BM	PL	7, 8, 9, 10	Galena	18.113	15.574	37.894	9.64	2
BM105-291.3	Bald Mountain (ME)	Cu–Zn–Ag–Au	30 Mt	Middle Ordovician	BM	PL	7, 8, 9, 10	Galena	18.098	15.571	37.895	9.63	2
BM114-420.7	Bald Mountain (ME)	Cu–Zn–Ag–Au	30 Mt	Middle Ordovician	BM	PL	7, 8, 9, 10	Galena	18.126	15.587	37.935	9.70	2
CL27-267.2	Bald Mountain (ME)	Cu–Zn–Ag–Au	30 Mt	Middle Ordovician	BM	PL	7, 8, 9, 10	Galena	18.125	15.578	37.901	9.65	2
BM21-389	Bald Mountain (ME)	Cu–Zn–Ag–Au	30 Mt	Middle Ordovician	BM	PL	7, 8, 9, 10	Pyrrhotite residue	18.153	15.645	38.121	9.95	2
C5-353.5-l	Bald Mountain (ME)	Cu–Zn–Ag–Au	30 Mt	Middle Ordovician	BM	PL	7, 8, 9, 10	Pyrrhotite leach	18.182	15.620	37.989	9.83	2
C5-353.5-r	Bald Mountain (ME)	Cu–Zn–Ag–Au	30 Mt	Middle Ordovician	BM	PL	7, 8, 9, 10	Pyrrhotite residue	18.172	15.601	37.930	9.75	2
9737	Harborside (ME)[8]	Zn–Cu–Pb–Ag	0.7 Mt	Middle Cambrian	BF	PB	11, 12	Galena	18.029	15.591	37.844	9.74	1
Herc-1	Hercules (ME)	Cu–Zn–Pb–Ag	<0.1 Mt	Middle Cambrian	BF	PB	13, 14	Galena	18.079	15.609	37.919	9.81	1
JS-85-6A	Eggemoggin (ME)[9]	Cu–Zn–Pb–Ag	<0.1 Mt	Middle Cambrian	BF	PB	13, 14	Galena	18.315	15.633	38.136	9.85	1
JS-85-7A	Deer Isle (ME)	Zn–Pb–Cu–Ag–Au	<0.1 Mt	Middle Cambrian	BF	PB	13, 14	Galena	18.099	15.602	37.877	9.77	1
JS-85-5A	North Castine (ME)	Zn–Cu–Pb–Ag–Au	<0.1 Mt	Middle Cambrian	BF	PB	13, 14	Galena	18.058	15.600	37.856	9.77	1
BH-1	Black Hawk (ME)[10]	Zn–Cu–Pb–Ag	0.9 Mt	Middle Cambrian	BS	PB	12, 15, 16	Galena	18.079	15.616	37.958	9.84	3
JS-80-47	Appleton (ME)	Cu	<0.1 Mt	Middle Cambrian	BS	PB	17, 18	Galena	18.073	15.650	38.456	10.00	1
JS-79-22	Milan (NH)	Zn–Cu–Pb–Ag–Au	<0.1 Mt	Middle Ordovician	BF	BH	5, 13	Galena	18.229	15.648	38.121	9.94	1
NE-4	Ore Hill (NH)	Zn–Pb–Cu–Ag	<0.1 Mt	Middle Ordovician	BM	BH	5, 19	Galena	18.077	15.556	37.767	9.57	1
NE-3	Royce (NH)	Zn–Pb–Ag	<0.1 Mt	Middle Ordovician	BS	BH	5, 20	Galena	18.187	15.601	37.982	9.74	1
JS-79-15	Croydon (NH)	Cu–Zn–Pb–Ag	<0.1 Mt	Middle Ordovician	BS	BH	5, 20	Galena	18.190	15.587	37.929	9.68	1
80GM-37	Gardner Mountain (NH)	Cu–Zn–Pb–Ag	<0.1 Mt	Early Devonian	BS	BH	5, 20, 21	Galena	18.226	15.632	38.114	9.87	1
JS-80-6	Paddock (NH)	Cu–Zn–Pb–Ag	<0.1 Mt	Early Devonian	BS	BH	5, 20, 21	Galena	18.414	15.673	38.306	10.00	1
G2-2	Gove (VT)	Cu–Zn	<0.1 Mt	Late Silurian	MS	CVGT	22	Whole ore	19.755	15.643	37.943	11.10	1
PH-9A	Pike Hill (VT)	Cu–Zn	0.1 Mt	Late Silurian	MS	CVGT	22	Galena	17.965	15.582	37.753	9.72	1
PH-2	Pike Hill (VT)	Cu–Zn	0.1 Mt	Late Silurian	MS	CVGT	22	Chalcopyrite	20.064	15.590	37.654	11.50	1
CP-17	Ely (VT)	Cu–Zn	0.5 Mt	Early Devonian	MS	CVGT	22, 23	Whole ore HCL leach	18.788	15.549	37.630	9.85	1

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Table 1. (*concluded*).

Sample No.	Deposit name[1]	Metal signature	Deposit size	Deposit age[2]	Type[3]	Belt[4]	References[5]	Analyzed material	$^{206}Pb/^{204}Pb$	$^{207}Pb/^{204}Pb$	$^{208}Pb/^{204}Pb$	μ	Data source[6]
CP-12	Ely (VT)	Cu–Zn	0.5 Mt	Early Devonian	MS	CVGT	22, 23	Pyrrhotite	18.988	15.538	37.559	10.11	1
EZ-39	Elizabeth (VT)	Cu–Zn	2.9 Mt	Early Devonian	MS	CVGT	22, 24, 25	Galena	17.858	15.474	37.403	9.26	1
EZ-539-1	Elizabeth (VT)	Cu–Zn	2.9 Mt	Early Devonian	MS	CVGT	22, 24, 25	Whole ore	19.507	15.606	37.921	10.78	1
RG-80-105	Davis (MA)	Cu–Zn	0.5 Mt	Early Ordovician	BM	BH	26	Galena	18.202	15.608	38.009	9.77	1

[1]State abbreviations: MA, Massachusetts; ME, Maine; NH, New Hampshire; VT, Vermont.
[2]Ages of deposits inferred from U–Pb dates on igneous zircons from enclosing metavolcanic rocks or on detrital zircons from enclosing metasedimentary rocks (see text).
[3]Deposit type abbreviations (classification of Barrie and Hannington 1999): BF, bimodal felsic; BM, bimodal mafic; BS, bimodal siliciclastic; M, mafic; MS, mafic–siliciclastic.
[4]Volcanic and volcanosedimentary belt abbreviations: BH, Bronson Hill; CVGT, Connecticut Valley–Gaspé trough; IM–SL, Ironbound Mountain–Second Lake; JP, Jim Pond; PB, Penobscot Bay; PL, Popelogan; SF, Shelburne Falls; WL, Weeksboro–Lunksoos.
[5]Key to numbered references: 1, Scully (1993); 2, McCormick (2021); 3, Fournier (1970); 4, Eisenberg (1982); 5, Moench et al. (1999); 6, Neil (2005); 7, Foose et al. (2003); 8, Slack et al. (2003); 9, Schulz and Ayuso (2003); 10, Ayuso et al. (2003); 11, Bouley (1978); 12, Marvinney and Berry (2015); 13, Emmons (1910); 14, Young (1962); 15, LaPierre (1977); 16, Yates and Howd (1988); 17, U.S. Geological Survey (2022); 18, Cheney (1967); 19, Secord and Brown (1986); 20, Prager (1971); 21, Margeson (1982); 22, White and Eric (1944); 23, Offield et al. (1993); 24, Howard (1969); 25, Slack et al. (2001); 26, Slack et al. (1983).
[6]Sources of lead isotope data: 1, this study; 2, Ayuso et al. (2003); 3, Doe and Zartman (1979).
[7]Formerly named the Mount Chase deposit.
[8]Also known as the Callahan deposit.
[9]Also known as the Emerson deposit.
[10]Also known as the Kerramerican or Blue Hill deposit.

posited in an arc as part of a supra–subduction zone sequence (Coish and Rogers 1987; Moench and Aleinikoff 2003; Gerbi et al. 2006); (5) the Ordovician Popelogan belt consisting in Maine of tholeiitic metavolcanic rocks and minor metasedimentary rocks that formed in an intraoceanic arc (Schulz and Ayuso 2003; van Staal et al. 2016; cf. Wang et al. 2022*a*); (6) the Ordovician Weeksboro–Lunksoos belt of tholeiitic metavolcanic rocks having arc and backarc geochemical signatures (Schulz and Ayuso 2003; McCormick 2021); and (7) the Cambrian Penobscot Bay belt of tholeiitic metavolcanic rocks and siliceous metasedimentary rocks that formed on a rifted continental margin (Schulz et al. 2008) in the hinterland behind the extensional Penobscot arc (van Staal et al. 2021*b*). Included in this last category, west of Penobscot Bay, is the small Appleton deposit hosted in amphibolite of the Lower Ordovician Penobscot Formation (cf. Fyffe et al. 1988; Reusch et al. 2018).

Metal signatures of the New England VMS deposits vary from Cu, to Cu–Zn, to Cu–Zn–Pb, to Zn–Cu–Pb, and to Zn–Pb–Cu with or without minor Ag and (or) Au. Most of the deposits considered here have small tonnages in the range of 0.1–3 Mt; Pickett Mountain is larger (6.3 Mt; Wolfden Resources Corporation 2021) and Bald Mountain is much larger (30.0 Mt; Slack et al. 2003). Total base metal contents of the deposits are typically 3–6 wt.%. Local metamorphic grades are chiefly greenschist or lower amphibolite, but include subgreenschist in the Popelogan belt of northern Maine (Bald Mountain deposit) and middle to upper amphibolite in the CVGT of eastern Vermont (Elizabeth, Ely, and related deposits). Deformation is commonly expressed by one or more penetrative foliations, open to isoclinal folds, and by high angle and in some cases thrust faults and shear zones.

Analytical methods

Sulfide samples discussed in this paper were collected and analyzed in the 1980s using methods available at that time. All samples consist of massive or semi-massive (>50 vol.%) sulfide minerals. Most samples were processed at the then-named Lamont–Doherty Geological Observatory in Palisades, New York, using methods described in LeHuray et al. (1988). Sulfide samples were ultrasonically cleaned in ultraclean H_2O, dried, crushed in an ultrasonically cleaned stainless-steel mortar, and ultrasonically cleaned again. Fractions of the samples were dissolved in HCl + HNO_3. Lead was extracted using brominated anion exchange resins and further purified by electrodeposition using the silica gel–phosphoric acid technique, and then analyzed by a VG Micromass 30 mass spectrometer. Reported Pb-isotope ratios are averages of at least two runs on a single dissolution, corrected for mass fractionation using National Bureau of Standards (NBS) Standard Reference Material (SRM) 981 (Catanzaro et al. 1968), and are accurate to better than ±0.1%.

Other sulfide samples except one were analyzed at the U.S. Geological Survey in Denver, Colorado, using methods described in LeHuray (1982). The samples were ultrasonically cleaned and then crushed in a clean Plattner mortar. Further cleaning was in very dilute, warm HC1 + HNO_3 followed by thorough rinsing in triply distilled water. Leached fractions were obtained by reacting samples overnight in warm 6.2 N HC1. The leachate was decanted and saved; the residue was rinsed thoroughly and dissolved in HNO_3 + HF. Lead was extracted on brominated anion exchange columns, the eluant was converted to nitrate, and lead was further purified by electrodeposition as the oxide on a platinum anode at ca. 2 V. All analyses were done using the surface emission (silica gel) technique on a Nier-type 12-in, 60° mass spectrometer and are mass fractionation-corrected using NBS SRM 981, which was run at regular intervals. Reported Pb-isotope ratios are averages of at least two complete runs per dissolution and are within 0.1% of absolute.

One galena sample (AP-1) was analyzed at the U.S. Geological Survey in Reston, Virginia. The hand-picked galena separate was purified by standard procedures and measured in static mode with a multicollector, automated TI-Box Spectromat mass spectrometer. Lead isotopic ratios were measured to a precision of ~0.1% at 2σ and corrected for mass fractionation by comparison with ratios measured on NBS SRM 981.

Results

Table 1 presents Pb-isotope data for 36 samples from 23 VMS deposits and occurrences in New England as analyzed in this study (all previously unpublished) and reported in earlier publications. Additional information and metadata are in Slack et al. (2023). Overall, values of $^{206}Pb/^{204}Pb$, $^{207}Pb/^{204}Pb$, and $^{208}Pb/^{204}Pb$ range from 17.659 to 20.064, 15.391 to 15.673, and 37.403 to 38.456, respectively. Among all analyzed sulfides, data for galena show smaller a range for $^{206}Pb/^{204}Pb$ (17.659–18.414). The least radiogenic sample is from the Alder Pond deposit in west-central Maine ($^{206}Pb/^{204}Pb$ = 17.659, $^{207}Pb/^{204}Pb$ = 15.391, and $^{208}Pb/^{204}Pb$ = 37.419). A second relatively unradiogenic sample, from the Vermont copper belt (VCB) in the CVGT, has a $^{206}Pb/^{204}Pb$ value of 17.858 and $^{207}Pb/^{204}Pb$ value of 15.474. The most radiogenic $^{206}Pb/^{204}Pb$ values (19.507–20.064) were determined on pyrrhotite, chalcopyrite, and bulk sulfide from the Elizabeth, Pike Hill, and Gove deposits in the VCB; data for two galena samples from the Elizabeth and Pike Hill deposits in this belt are not anomalously low or high relative to those of the other New England VMS deposits. One galena sample each from the Bronson Hill volcanic belt and the Penobscot Bay volcanosedimentary belt also have $^{206}Pb/^{204}Pb$ values higher than all other data reported herein, and based on field and textural relationships are interpreted as epigenetic veins. Results for these two radiogenic galena-rich samples, and for the five very radiogenic galena-absent samples (pyrrhotite + chalcopyrite ± sphalerite) from the VCB, are not considered further in this study. Whole-ore samples from the Gove and Elizabeth deposits lack visible or microscopic galena and also are not considered. In contrast, the whole-ore sample from the Ledge Ridge deposit contains abundant galena, and data for this sample are included in the discussions below. Lead isotope values for pyrite from the Bald Mountain deposit (Ayuso et al. 2003) are not included here, because during diagenesis and metamorphism, this Pb-poor mineral (higher U/Pb and Th/Pb than galena) does not behave as a closed system. As a result, this process typically yields evolved Pb-isotope ratios (mainly high $^{206}Pb/^{204}Pb$) that do not represent the primary values that existed during VMS mineralization (e.g., Ayuso et al. 2014). We suggest that a similar argument also applies to the whole-ore (galena-absent) samples from the Gove and Elizabeth deposits.

Figures 3A and 3B are plots of the $^{207}Pb/^{204}Pb$ versus $^{206}Pb/^{204}Pb$ and $^{208}Pb/^{204}Pb$ versus $^{206}Pb/^{204}Pb$ data. Shown for comparison are Pb-isotope ratios for VMS deposits of the Canadian Appalachians in Québec, New Brunswick, Newfoundland, and Nova Scotia. Most of the New England data plot broadly in four main clusters that largely coincide with fields that outline galena Pb-isotope data for different tectonostratigraphic terranes and subterranes in the Canadian Appalachians. In detail, the following similarities are noted: (1) data from the Popelogan, Weeksboro–Lunksoos, and Shelburne Falls–Bronson Hill volcanic belts in northern Maine, northern New Hampshire, and western Massachusetts (Fig. 2) plot within or near the data fields for the Exploits peri-Ganderian arc–backarc tract of the Dunnage domain in Newfoundland (all samples including those from the Lemarchant VMS deposit), and for the Tetagouche part of the Dunnage domain in New Brunswick (all from the Bathurst district); (2) data for the two samples from the Ironbound Mountain and Second Lake volcanic belts in western Maine are relatively unradiogenic, and plot close to values for deposits of the peri-Laurentian Notre Dame arc–backarc tract in Newfoundland and the Ascot–Weedon volcanic belt in southeastern Québec; (3) the Alder Pond deposit, west-central Maine, is less radiogenic in $^{207}Pb/^{204}Pb$ and $^{208}Pb/^{204}Pb$ than all of the VMS deposits in the Notre Dame and Exploits arc–backarc tracts, excluding results for the strongly deformed Great Burnt Lake deposit (Hinchey and Sandeman 2022), which has anomalous values that are considered suspect as they represent analyses of pyrite, not galena, and thus may not be original Pb-isotope signatures (cf. Ayuso et al. 2014); (4) samples from the Penobscot Bay volcanic belt in coastal Maine are isotopically distinct and mostly are less radiogenic than those of the Exploits peri-Ganderian arc–backarc tract; and (5) data for the unradiogenic galena samples from the VCB plot near the field for the peri-Laurentian Notre Dame arc–backarc tract of the Dunnage domain.

Calculated values of μ ($^{238}U/^{204}Pb$) for the galena samples from the different VMS deposits vary from 8.94 to 10.0 (Table 1). Average μ values for the Ordovician terranes change from west to east, as follows: (1) Jim Pond volcanic belt, 8.94 $\pm$ 0.00; (2) Ironbound Mountain and Second Lake, 9.49 $\pm$ 0.10; (3) Shelburne Falls–Bronson Hill volcanic belts, 9.80 $\pm$ 0.15; (4) Popelogan volcanic belt, 9.70 $\pm$ 0.06; and (5) Weeksboro–Lunksoos volcanic belt, 9.99 $\pm$ 0.00. Average μ values for deposits in the Cambrian Penobscot Bay volcanic belt and the Silurian–Devonian VCB are 9.83 $\pm$ 0.09 and 9.49 $\pm$ 0.23, respectively. Calculated model ages (after Stacey and Kramers 1975) also range widely, from 280 to 514 Ma, and with few exceptions are significantly younger than the age of the host volcanic rocks as determined by U–Pb zircon geochronology. These model ages do not indicate homogeneous lead sources and probably do not convey meaningful age information, and thus are not discussed further.

Discussion

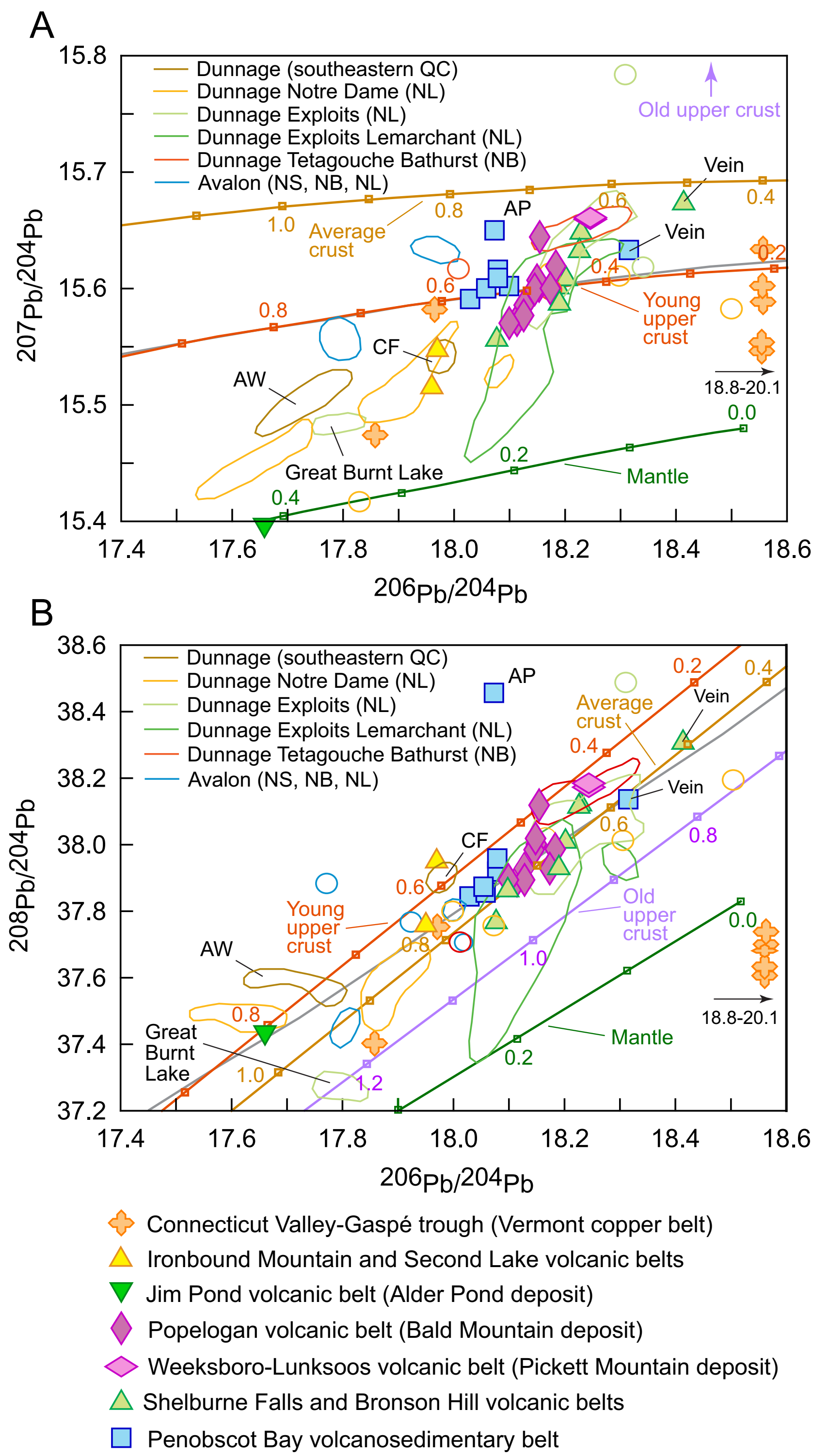
General associations

Among New England VMS deposits, no correlations are apparent between Pb-isotope values ($^{206}Pb/^{204}Pb$, $^{207}Pb/^{204}Pb$, $^{208}Pb/^{204}Pb$, and μ) and parameters such as deposit size, grade, or metal signature. For example, the large (30 Mt) Bald Mountain Cu–Zn–Ag–Au deposit is isotopically very similar to the very small (<0.1 Mt) Eggemoggin Cu–Zn–Pb–Ag deposit. Second, values for the relatively high-grade Ledge Ridge Zn–Cu–Pb–Ag–Au deposit are broadly like those of the low-grade Pike Hill Cu–Zn deposit. In addition, the Milan Zn–Cu–Pb–Ag–Au deposit has Pb-isotope values similar to the Davis Cu–Zn deposit. Moreover, large deposits are not especially radiogenic as shown by data for the Bald Mountain deposit (30 Mt) that has lower uranogenic and thorogenic values relative to the Pickett Mountain (6 Mt) deposit (Table 1, Fig. 3). We also note that no secular evolution is apparent with respect to metal signatures, given that Early or Middle Ordovician deposits can be dominated by Cu and Zn (Bald Mountain and Thrasher Peaks) or have abundant Pb and Ag in addition to Cu and Zn (Pickett Mountain and Milan), and that Early Devonian deposits (Elizabeth and Ely) are dominantly Cu-rich, whereas one Silurian deposit (Ledge Ridge) is polymetallic with a Zn–Cu–Pb–Ag signature. Tonnages, Pb-isotope values, and metal contents of the New England VMS deposits are thus likely controlled by the size and duration of the hydrothermal system, the presence or absence of caprocks that facilitate subseafloor zone refining, and the source(s) of metals, i.e., footwall rocks from which the metals were leached, with abundant felsic volcanic rocks promoting the formation of polymetallic, Pb-rich deposits (Franklin et al. 2005; Galley et al. 2007; Piercey 2015).

Principal lead sources

The first-order source of lead in VMS deposits is widely considered to be footwall rocks leached by the deeply convecting hydrothermal fluids responsible for sulfide mineralization (e.g., Franklin et al. 2005; Hannington 2014). However, the pathways by which the lead is incorporated into these rocks may be complex. Within VMS deposits, lead is derived mainly from felsic volcanic rocks and clastic sedimentary rocks, and deposits underlain by significant proportions of felsic volcanics $\pm$ continental crust are characterized by high lead contents (e.g., Barrie and Hannington 1999; Galley et al. 2007). Studies of modern VMS deposits have shown that those in MOR settings contain lead derived predominantly from the mantle, based on very similar Pb-isotope compositions of the VMS sulfides and underlying MOR basalts (LeHuray et al. 1988; Godfrey et al. 1994). In contrast, data for deposits on sediment-covered ridges reflect sources from both basalt and terrigenous sediment (LeHuray et al. 1988; Stuart et al. 1999), whereas arc-type VMS deposits typically display a range of compositions that may reflect multiple lead sources, including mantle, subducted slab (basalt $\pm$ pelagic sediments), local footwall volcanic $\pm$ sedimentary rocks, and in some cases deeper continental basement (Arribas 1993; Halbach et al. 1997; Chiaradia and Fontboté 2001). In such an environment,

Fig. 3. Lead isotope plots of data for New England volcanogenic massive sulfide (VMS) deposits (Table 1) compared to fields for deposits in the Canadian Appalachians (Table S1). (A) $^{206}Pb/^{204}Pb$ versus $^{207}Pb/^{204}Pb$ plot. (B) $^{206}Pb/^{204}Pb$ versus $^{208}Pb/^{204}Pb$ plot. Growth curves after Lode et al. (2017) as modeled from data of Kramers and Tolstikhin (1997); ages are in Ma. In (A), growth curve for old upper crust is off scale (above), at $^{207}Pb/^{204}Pb$ values of 15.83–15.86. Grey curve is average crustal evolution curve of Stacey and Kramers (1975). Deposit abbreviation: AP, Appleton. Geologic abbreviations: AW, Ascot–Weedon volcanic belt; CF, Clinton–Frontenac volcanosedimentary belt. Province abbreviations: NB, New Brunswick; NL, Newfoundland; NS, Nova Scotia; QC, Québec. Note that five samples from the Vermont copper belt having very high $^{206}Pb/^{204}Pb$ ratios lack galena (see text). No data are shown for the Udall mine in northern Vermont (Figs. 1 and 2), because only pyrite was available for sampling.



most of the principal lead sources—subducted sediments delivered to the arc volcanics via partial melting, slab/sediment dehydration, and hydrothermal activity, continental crust incorporated in the arc basement, and pelagic sediments—contain lead derived originally from subjacent continental basement, and the isotopic composition of lead in the VMS deposits will thus reflect this source. Radiogenic lead sources (predominantly basement rocks) are implicated for galena in some VMS deposits, and not the host volcanic rocks, because the latter in many cases have higher $^{206}Pb/^{204}Pb$ and lower $^{207}Pb/^{204}Pb$ and $^{208}Pb/^{204}Pb$ values (e.g., Ayuso et al. 2003, 2007; Tessalina et al. 2016), including for whole-rock data recalculated to the U–Pb zircon age of the enclosing volcanic strata as done for this study (not shown) using data for galena from the Harborside deposit (Table 1) and host Castine Volcanics (Schulz et al. 2008). Seawater can be a volumetrically very minor source of lead based on data for some modern VMS deposits (LeHuray et al. 1988; Godfrey et al. 1994), but is not quantitatively important. Lead isotope data may also serve as proxies for the sources of other metals commonly enriched in VMS deposits—both modern and ancient—such as Cu, Zn, Ag, and Au.

Insights into lead sources of New England VMS deposits are provided by comparing data for these deposits with the mantle and crustal growth curves of Kramers and Tolstikhin (1997). Figure 3A shows that all samples have $^{207}Pb/^{204}Pb$ values between the mantle curve and the average crustal curve; the distribution of most data suggests that lead was derived from varying proportions of young upper crust, the mantle, and old upper crust. In this context, young upper crust refers to predominantly felsic rocks <2.0 Ga with lower U and Th contents relative to old upper crust (Kramers and Tolstikhin 1997). Both samples from the Ironbound Mountain and Second Lake volcanic belts (Thrasher Peak and Ledge Ridge deposits, respectively) and one sample each from the Jim Pond Formation (Alder Pond deposit), CVGT (Elizabeth deposit), and the Popelogan volcanic belt (Bald Mountain deposit) have greater mantle components relative to the other New England samples, based on relatively low uranogenic ($^{207}Pb/^{204}Pb$) values and on low μ values (<9.74) below the average crustal Pb evolution curve that records a juvenile lead source from the mantle or mantle-derived basalts (Doe and Zartman 1979). Lead isotope data for the Shelburne Falls–Bronson Hill, Popelogan, and Penobscot Bay belts straddle the curve for young upper crust, the former two including samples with both less- and more-radiogenic $^{207}Pb/^{204}Pb$ values.

Sources of thorogenic lead ($^{208}Pb/^{204}Pb$) are constrained by relationships of the VMS data to the growth curves shown in Fig. 3B. Included are curves for the mantle, average crust, young upper crust, and old upper crust, the last characterized by higher Th and U contents relative to other primary terrestrial reservoirs (see Kramers and Tolstikhin 1997). Average crust integrates data for upper and lower continental crust. This plot suggests that, based on relatively high $^{208}Pb/^{204}Pb$ values, young upper crust was the predominant reservoir for lead contained in most of the New England VMS deposits (Shelburne Falls–Bronson Hill, Popelogan, and Penobscot Bay volcanic belts). However, results for one sample from the Elizabeth deposit are less radiogenic and fall near the growth curve for old upper crust; this is the same sample that plots to less-radiogenic values of uranogenic lead (Fig. 3A).

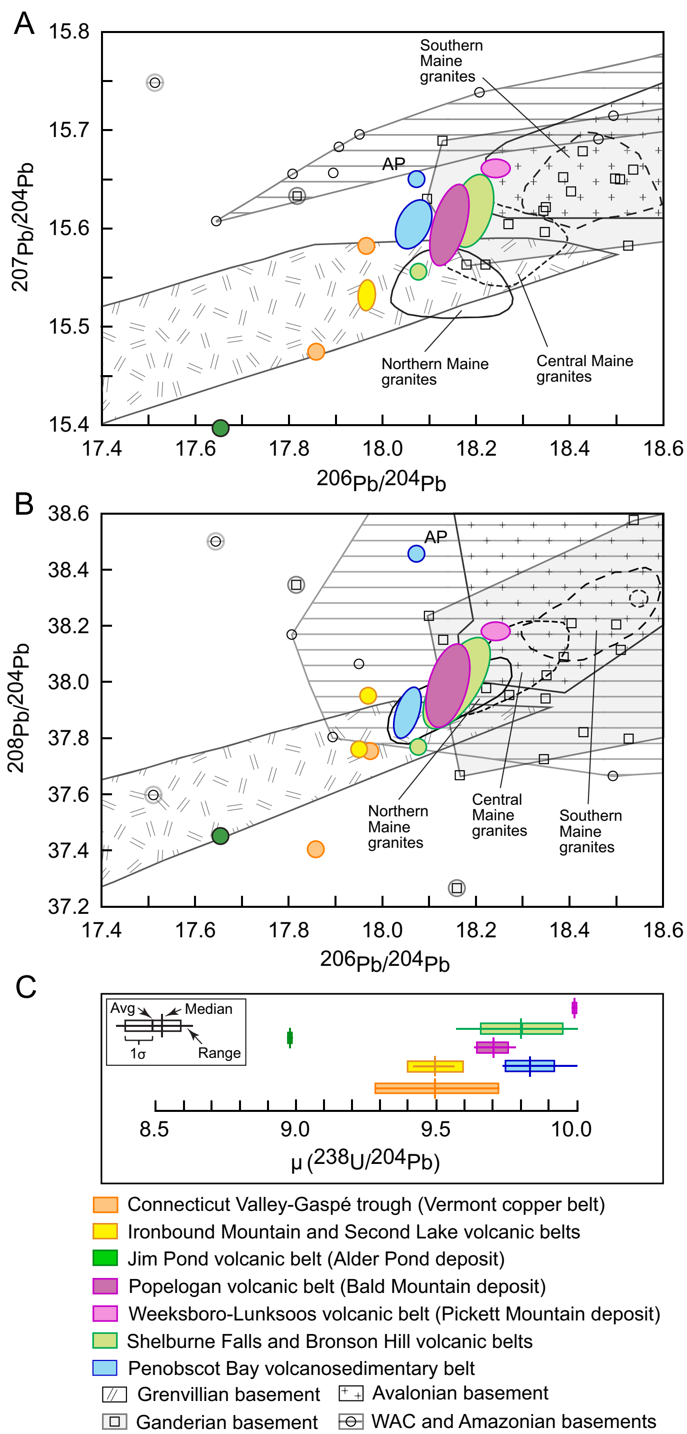
Radiogenic endmembers

Figures 4A and 4B show grouped Pb-isotope analyses for individual volcanic and volcanosedimentary belts (Fig. 2). The data for the Penobscot Bay, Shelburne Falls–Bronson Hill, and Popelogan belts define steep elongate trends in both uranogenic and thorogenic Pb-isotope space. Results for the other belts are scattered, mostly at less radiogenic values. Such steep trends in Pb-isotope data are characteristic of VMS deposits worldwide, both modern and ancient, and are generally understood to reflect mixed sources of lead with little or no later re-equilibration with whole-rock lead (e.g., Doe and Zartman 1979; Doe 1982; Doe et al. 1985; Bjørlykke et al. 1993; Fouquet and Marcoux 1995; Halbach et al. 1997; Ayuso et al. 2003, 2007; Kim et al. 2006; Huston et al. 2014; Tessalina et al. 2016; Gill et al. 2019; Yu et al. 2020). Whereas the least radiogenic endmember is generally ascribed to a mantle source, identification of the radiogenic endmember is often elusive, being attributed in most cases either to pelagic sediment or continental crust. Contributions from these two potential radiogenic endmembers, and others, are evaluated below.

Continental crust

Basement of continental crust that existed beneath the volcanic arcs or rifted sedimentary basins during VMS formation represents a key potential radiogenic endmember source. Based on Pb-isotope data, such basement rocks have been invoked for numerous ancient VMS deposits, such as in the Miocene Kuroko district in Japan and the Cretaceous Troodos ophiolite in Cyprus (Doe and Zartman 1979; Doe 1982). Importantly, a variety of geochemical and geochronological evidence indicates that continental crust underlies some modern intraoceanic volcanic arcs, such as the Vanuatu, Solomon Islands, and Izu–Bonin–Mariana arcs (Buys et al. 2014; Tapster et al. 2014; Tani et al. 2015). By analogy, similar crust may have formed the deep substrate beneath the volcanic arcs discussed in this study. One line of evidence for hidden subarc basement rocks comes from inheritance in igneous zircons (e.g., Zagorevski et al. 2007, 2008). For example, in western Maine, Moench et al. (2000) reported that rhyolite of the Ordovician Jim Pond Formation contains one xenocrystic zircon grain with a Grenvillian SHRIMP U–Pb age of ~1075 Ma. For the Weeksboro–Lunksoos volcanic belt in eastern Maine, Ayuso et al. (2003) listed 9 of 40 anomalously old (522–1384 Ma) SHRIMP U–Pb ages for igneous zircons from an Ordovician syn-volcanic dacite dome near the Pickett Mountain VMS deposit; one 898 Ma grain was found by McCormick (2021) using LA-ICPMS U–Pb dating of a footwall rhyolite unit at the deposit; this Neoproterozoic age is consistent with a source from either peri-Ganderian or Avalonian basements. One Neoproterozoic zircon was found by Valley et al. (2020) in a trondhjemite from the Bronson Hill arc, but previous U–Pb zircon studies of igneous rocks from

Fig. 4. Comparison of Pb-isotope data for New England volcanogenic massive sulfide (VMS) deposits with fields for granites in Maine and potential continental basements. (A) $^{206}Pb/^{204}Pb$ versus $^{207}Pb/^{204}Pb$ plot. (B) $^{206}Pb/^{204}Pb$ versus $^{208}Pb/^{204}Pb$ plot (C) μ ($^{238}U/^{204}Pb$) plot. Results for vein-type and radiogenic galena-poor samples are not shown. Granite data (feldspar separates) from Ayuso and Bevier (1991). Isotopic fields (all corrected to 450 Ma) for Grenvillian basement from Ayuso et al. (1996) and references therein; for Ganderian basement (data points shown by small open squares) from Aleinikoff et al. (2007) and Dorais et al. (2012*b*); for Avalonian basement from Ayuso et al. (1996), Pe-Piper and Piper (1998), and Aleinikoff et al. (2007); and for West African craton (WAC) and Amazonian craton basements (data points shown by small open circles) from Stewart et al. (2001), Tohver et al. (2004), de Souza et al. (2016), and Fernandes and Juliani (2019). Due to a lack of published whole-rock Pb-isotope data for WAC, age-corrected values for Paleoproterozoic basement rocks on Islesboro Island are used as proxies (cf. Reusch et al. 2018; van Staal et al. 2021*b*); data for Islesboro Island and Amazonian craton basements encompass the same general field. Projections of basement fields to more radiogenic $^{206}Pb/^{204}Pb$ and $^{208}Pb/^{204}Pb$ values beyond the scales of these plots are based on data in references cited above. In (C), median values are identical, or nearly so, to averages and are not shown except for the Shelburne Falls and Bronson Hill volcanic belts.



this arc did not report any inheritance (Tucker and Robinson 1990; Moench and Aleinikoff 2003). No unequivocal evidence of inheritance has been described for zircons from volcanic rocks of the Popelogan arc in Maine or the Penobscot Bay belt (Ayuso et al. 2003; Schulz et al. 2008).

Neodymium and Pb-isotope data for volcanic rocks that host the VMS deposits yield additional insights into the nature of underlying basements that existed during arc formation (e.g., Swinden et al. 1997; Ayuso and Schulz 2003; Zagorevski et al. 2007, 2008; Walsh et al. 2021). Volcanic and coeval plutonic rocks of the Hawley Formation (Shelburne Falls arc) in northwestern Massachusetts that host the Davis VMS deposit have εNd_t values of −3.4 to +7.4 (median +5.7; $n = 10$), suggesting a predominant mantle source during arc formation (Bock et al. 1996; Pierce 2013). For the Ammonoosuc Volcanics of the Bronson Hill arc, results show εNd_t values of −3.0 to +3.4 (median +2.1; $n = 7$) that indicate both mantle and crustal components in the magma source (Foland and Allen 1991; Dorais et al. 2012*a*). Data for volcanic rocks of the Popelogan arc in northern Maine show εNd_t values of +0.7 to +6.5 (median +4.7; $n = 29$, excluding one unradiogenic hanging wall argillite) that document arc formation on oceanic crust without evidence of a continental crustal basement (Ayuso and Schulz 2003). In contrast, data for the broadly coeval Weeksboro–Lunksoos belt to the southeast yield εNd_t values of −5.7 to +6.0 (median −2.6; $n = 9$) that suggest a substantial component of continental crust (Ayuso and Schulz 2003). In the Penobscot Bay volcanosedimentary belt, negative and small positive whole-rock εNd_t values for samples of volcanic rocks from the Ellsworth Schist and overlying, and nearly coeval, Castine Volcanics indicate a predominant mantle component in the source area(s) of the parent magmas (total range of εNd_t −0.9 to +8.6, median +7.3, $n = 14$; Schulz et al. 2008). Radiogenic whole-rock Pb-isotope values for metabasalt and metarhyolite indicate a continental crustal source for some of the magmas that formed the New England volcanic arcs, including in the Bronson Hill, Weeksboro–Lunksoos, and Penobscot Bay belts (Ayuso and Schulz 2003; Schulz et al. 2008; Dorais et al. 2012*a*). Overall, for most early Paleozoic volcanic sequences of the New England Appalachians, Nd isotopic evidence suggests varying contributions from both mantle and crustal sources.

Different pre-accretionary basement blocks of continental crust are represented in the northern Appalachians. These include crust of Laurentian, Gondwanan, and possible Baltican affinity. Gondwanan-derived basements (Amazonia and West Africa) are thought to be present beneath Ganderia and Avalonia (e.g., van Staal et al. 2012, 2021*a*), although Avalonia also may be underlain by Baltican-derived basement that was later incorporated into Gondwana (van Staal et al. 2021*b*). Lead isotope data for VMS deposits in the Dunnage domain of Newfoundland (Fig. 3) show a general contrast in $^{206}Pb/^{204}Pb$ values between the Notre Dame and Exploits arc–backarc tracts, which suggests an isotopic provincialism and contrasting basement lead sources for these two tracts (Swinden and Thorpe 1984; Swinden et al. 1988); basement contrasts in this region are also expressed by differences in εNd_i values (Zagorevski et al. 2008). As in Newfoundland, it is well established for New England that Laurentian and peri-Laurentian crust underlies rocks to the west of the BLL as summarized by Zagorevski et al. (2008) and van Staal et al. (2021*b*). However, in western Maine, deep seismic reflection profiles and petrologic data on post-accretionary (Acadian) granitic plutons imply that Laurentian crust extends at least 10 km east of the surface projection of this boundary and that Ganderian crust was thrust over Laurentian crust (Spencer et al. 1989; Stewart et al. 1993; Pressley and Brown 1999; see also Tomascak et al. 2005). In central New Hampshire, petrologic data on Oliverian granitic domes argue for the presence of buried Laurentian crust ca. 100 km east of the BLL (RIL in Dorais et al. 2008). Farther southeast, in Connecticut and Rhode Island, Wintsch et al. (2014) concluded on the basis of U–Pb ages and cataclastic textures of zircon grains that Avalonian crust was wedged between Ganderian cover and basement during the Alleghenian orogeny (see also Ayuso et al. 1996; Whalen et al. 1996). In this same region, high-resolution Ps receiver function imaging and wavelength migration imaging of seismic data suggest westward underthrusting of rifted Grenvillian crust during the Taconic and (or) Acadian orogenies, together with the presence of a deeper, west-dipping slab of likely subduction origin (Luo et al. 2021, 2022). In addition, permissive evidence for tectonic interleaving of crustal basements exists in the Penobscot Bay area of Maine, where seismic reflection profiles show multiple strong reflectors in the deep crust (Luetgert and Mann 1990; Stewart et al. 1993) that could record more than one continental basement slice. Collectively, these data indicate that the present crustal structure of New England is significantly more complex than existed beneath the multiple, individual Paleozoic volcanic arcs that formed in the Rheic Ocean prior to their accretion to Laurentia (cf. Waldron et al. 2022), and that Pb-isotope data for the VMS deposits represent a novel window into the nature of these subarc basements during sulfide mineralization.

Potential influences of the different pre-accretionary continental basements on the Pb-isotope systematics of the VMS deposits are discussed below for each volcanic and volcanosedimentary belt. Note that whole-rock Pb-isotope data for the different basements compiled here are restricted to analyses of Proterozoic rocks and exclude results for Silurian and Devonian granites (whole rocks and feldspar separates) because such data are likely influenced by lead sources within Paleozoic sedimentary rocks as well as juxtaposed basements.

Pelagic sediment and other potential endmembers

Pelagic sediment as used herein refers mainly to carbonate and siliceous ooze and clay, based on studies of modern samples, and excludes metalliferous sediments and samples having major volcaniclastic or terrigenous components. Importantly, magmatic sources of modern arc volcanic rocks typically contain significant fractions of subduction-derived pelagic sediment, as evidenced by a variety of geochemical and radiogenic isotope data (e.g., Keleman et al. 2014; Li et al. 2019; Ishizuka et al. 2020). Lead isotope values compiled here for modern pelagic sediments ($n = 51$; Fig. S1) within and near intraoceanic arcs and in the open ocean, corrected to 450 Ma, are less radiogenic in $^{206}Pb/^{204}Pb$, excepting one sample, than all data fields for the peri-Ganderian Popelogan, Weeksboro–Lunksoos, Shelburne Falls–Bronson Hill, and Penobscot Bay belts, and lack sufficiently high $^{207}Pb/^{204}Pb$ values to represent the projected radiogenic endmember for any of these belts. The pelagic sediment field for thorogenic lead does include data that could in theory represent some of the $^{208}Pb/^{204}Pb$ endmembers. However, because the Pb-isotopic compositions of early Paleozoic pelagic sediments are unknown, we cannot determine whether such sediment was an important radiogenic component in the Pb-isotope systematics of the VMS deposits. Nonetheless, an effect from ancient pelagic sediments is considered likely, via transport within the subducted slab followed by mass transfer into the mantle wedge and thence into the crust, prior to VMS mineralization. This subduction-related process may explain some of the Pb-isotope provincialism described by Swinden and Thorpe (1984) and Swinden et al. (1988) for the Notre Dame and Exploits arc–backarc tracts, in which sediments underlying the VMS deposits of the Notre Dame peri-Laurentian arc–backarc tract were derived at least in part from Laurentia in contrast to sediments beneath Exploits deposits that came mainly from Ganderia.

Two additional potential sources for the relatively high $^{206}Pb/^{204}Pb$ values of some samples are seawater and hydrothermal fluids. Godfrey et al. (1994) determined that up to 20% of the lead in plume particles associated with the modern TAG hydrothermal field on the Mid-Atlantic Ridge is derived from ambient seawater. However, Pb-isotope data for sulfides and vent fluids from other modern seafloor hydrothermal systems do not indicate a significant contribution of lead from seawater (Chen et al. 1986; LeHuray et al. 1988; Fouquet and Marcoux 1995), nor do mass–balance constraints given the significant mass of lead in the deposits relative to the very low content of lead in seawater. Because all of our New England samples consist of massive or semi-massive sulfide without a significant proportion derived from plume fallout, such as Fe–Mn iron formation or other metalliferous sediment, a major seawater source for the radiogenic $^{206}Pb/^{204}Pb$ component of the samples reported in this study is considered unlikely. However, we cannot rule out the involvement of a high U/Th reservoir such as oxygenated, CO_2-rich hydrothermal fluid containing uranyl carbonate ions, e.g., $UO_2(CO_3)_3{}^{4-}$ (Piercey and Kamber 2019), although such a fluid probably would have deposited appreciable primary carbonate that is absent or rare in the New England VMS deposits.

Comparisons with data for granitic plutons

Lead isotope data for uranogenic lead in the VMS deposits (Fig. 4A) show that most samples from Maine are less radiogenic in $^{206}Pb/^{204}Pb$ than K-feldspar data for granites in this region and that $^{207}Pb/^{204}Pb$ values cover a similar range. In contrast, the $^{208}Pb/^{204}Pb$ VMS values (Fig. 4B) overlap those of the Maine granites, especially the field for the northern group. Ayuso (1986) and Ayuso and Bevier (1991) interpreted these fields as representing the principal source of lead for the granites, derived from three different crustal blocks (Grenville, undefined, and Avalon) influenced mainly by radiogenic continental basement rocks, except for plutons in the CVGT that have a significant component derived from the subcontinental mantle. The relatively high $^{206}Pb/^{204}Pb$ values for the granitic K-feldspars were suggested by Ayuso (1986) to possibly reflect radiogenic metasedimentary xenoliths in the granites. A corollary is that deep facies of the intruded thick succession of Upper Silurian and Lower Devonian metasedimentary rocks in the CVGT could have been extensively melted and hence also contributed a more radiogenic component to the granitic magmas (e.g., Tomascak et al. 2005), as well as a younger model age in terms of higher $^{206}Pb/^{204}Pb$ values for this source. It is possible that the range of $^{206}Pb/^{204}Pb$ values for each of the three granite groups records, in part, isotopically different basements that existed beneath the pre-accretionary volcanic arcs prior to granite emplacement. In support of this hypothesis, all of the VMS deposits in the Penobscot Bay area except Appleton have lower $^{206}Pb/^{204}Pb$, $^{207}Pb/^{204}Pb$, and $^{208}Pb/^{204}Pb$ values than granites of the southern group from this region (Figs. 4A and 4B), suggesting that the lead sources for these granites were not exclusively the basements but instead came partly from overlying Siluro–Devonian metasedimentary rocks. A similar interpretation was proposed by Moench and Aleinikoff (2003) based on differences in the Pb-isotope signatures of VMS deposits and Acadian granites within the central group as defined by Ayuso (1986) and Ayuso and Bevier (1991), and by Tomascak et al. (2005) based on Nd- and Pb-isotope data for the Acadian Mooselookmeguntic igneous complex in western Maine.

Implications for pre-accretionary tectonostratigraphic terranes

Results of our study support some previously proposed terrane boundaries in New England but challenge others. The most controversial has been the Iapetus suture zone (BLL), which marks the Early Ordovician boundary between the eastern edge (present coordinates) of peri-Laurentian volcanic arcs and the multiple arcs that formed in peri-Ganderian settings and later accreted to Laurentia beginning with the Early Ordovician closure of the Iapetus Ocean (Waldron et al. 2022, and references therein). The Pb-isotope data for the VMS deposits in New England (Fig. 3) suggest lead derivation from sources within at least three and possibly four different tectonostratigraphic assemblages, as discussed below.

Shelburne Falls and Bronson Hill volcanic belts

Both the Shelburne Falls and Bronson Hill belts are built on rocks that have Ganderian detrital zircon signatures (Valley et al. 2020). In the Shelburne Falls belt, volcanic rocks of the Hawley Formation have U–Pb zircon ages of 475.5 ± 0.2 Ma or older (Macdonald et al. 2014), whereas metavolcanic rocks of the Ammonoosuc Volcanics in the Bronson Hill belt range in age from 465 ± 6 to 453 ± 2 Ma (Valley et al. 2020 and references therein). The earliest known magmatic events in these belts are represented by a 493 ± 8 Ma tonalite in the Bronson Hill belt (Rankin et al. 2013) and a 502 ± 4 Ma tonalite in the Shelburne Falls belt (Aleinikoff et al. 2011). However, despite age differences between the two belts, Pb-isotope values of $^{206}Pb/^{204}Pb$, $^{207}Pb/^{204}Pb$, and $^{208}Pb/^{204}Pb$ for the Davis VMS deposit in the Hawley Formation are nearly identical to those for the Royce and Croydon deposits hosted in the Ammonoosuc Volcanics (Table 1). Overall, relatively high $^{207}Pb/^{204}Pb$ and $^{208}Pb/^{204}Pb$ values for VMS deposits in the Shelburne Falls and Bronson Hill belts rule out Grenvillian crust as the radiogenic endmember, and support a Ganderian affinity (Figs. 4A and 4B). We thus agree with the interpretation of Macdonald et al. (2014), based on U–Pb detrital zircon geochronology, that the Hawley Formation has a peri-Gondwanan origin. A possible link of the Shelburne Falls and Bronson Hill volcanic belts to much older crust of the West African craton (WAC) is considered unlikely as the radiogenic endmember, based on the Ganderian detrital zircon signatures of underlying strata (Valley et al. 2020); note that rare occurrences of Paleoproterozoic (1.88 Ga) zircons in the Hawley Formation in the Shelburne Falls belt are detrital and not igneous in origin (Karabinos et al. 2017). We therefore conclude that the Pb-isotope compositions of galena in the VMS deposits of this belt reflect radiogenic endmember sources from Ganderia.

Karabinos et al. (1998, 2017) proposed on the basis of U–Pb zircon ages and geochemistry that the Hawley Formation is part of the Shelburne Falls arc, which accreted to Laurentia prior to the Bronson Hill arc that occurs ~20 to ~50 km to the east (present coordinates). Whereas available U–Pb ages for volcanic rocks of the Hawley Formation are at least 4 m.y. older than those of the Ammonoosuc Volcanics, ages for the inferred comagmatic plutons of the two belts overlap, which Valley et al. (2020) suggested records the evolution of one long-lived composite arc. The similarity of Pb-isotope signatures in VMS deposits of the two belts (which are isotopically like deposits in the Exploits peri-Ganderian arc–backarc tract of Newfoundland) probably reflects a contribution from similar Pb-isotope reservoirs, and thus supports this model for one broadly coeval arc system. One important difference, however, is the presence of boninites in the Hawley Formation (Kim and Jacobi 1996), implying a forearc setting for this unit in contrast to the arc setting inferred for the Shelburne Falls volcanic belt (Karabinos et al. 1998; Moench and Aleinikoff 2003). Whether this difference in tectonic settings is consistent with one arc system remains to be evaluated. Importantly, a northeast-trending deep seismic profile from the Adirondack massif through central Vermont and northern New Hampshire to central Maine suggests that the eastern limit of Grenvillian crust is within the CVGT, west of the Bronson Hill volcanic belt (Hughes and Luetgert 1991). The Nd and Pb isotope data of Dorais et al. (2012*a*) for Ordovician felsic domes (Oliverian Plutonic Suite), which are younger than the volcanic rocks, imply that the Bronson Hill belt and contained Ammonoosuc Volcanics were thrust westward over the Laurentian margin during the Middle Ordovician Taconic orogeny.

CVGT

In east-central Vermont, the CVGT contains several VMS deposits hosted in metasedimentary rocks and minor tholeiitic metabasalt of Late Silurian to Early Devonian age (Slack et al. 2001; cf. Perrot et al. 2018). The stratigraphically higher parts of this tectonostratigraphic terrane contain the VCB deposits, in eastern Vermont and northeastern New Hampshire, and differ from the other terranes described here in having formed during the early stage of the Acadian orogeny, thus representing a syn-accretionary and not a pre-accretionary setting. Restricting our evaluation to analyses of galena, which occurs in one sample each from the Elizabeth and Pike Hill deposits (Table 1), $^{206}Pb/^{204}Pb$ values are relatively unradiogenic and plot near the field for VMS deposits in the Notre Dame peri–Laurentian arc–backarc tract of the Dunnage domain in Newfoundland (Fig. 3). This result suggests that the source of lead in the VCB includes a component of Grenvillian crust, an interpretation consistent with the age-corrected Pb-isotope fields for Grenvillian basement (Figs. 4A and 4B) and with the conventional tectonic setting assigned to the CVGT being a foreland successor basin that formed during extension of eastern Laurentia following the Salinic orogeny (Tremblay and Pinet 2005; McWilliams et al. 2010). Perrot et al. (2018) argued that the distinctive age spectrum of these detrital zircons is permissive for a Laurentian provenance, whereas McWilliams et al. (2010) reported detrital age spectra that suggest mixed signals from both Laurentia and Ganderia. Using petrologic data, Ayuso and Arth (1992) proposed that Grenvillian crust might be a contaminant within the Acadian granites of northeastern Vermont that intrude the CVGT. The Pb-isotope data for the galena-rich samples from the Elizabeth and Pike Hill deposits in the VCB reported here are consistent with a Laurentian crustal lead source based mainly on the relatively unradiogenic $^{206}Pb/^{204}Pb$ values (Figs. 4A and 4B), which are not related to age because these Late Silurian–Early Devonian VMS deposits are among the youngest samples analyzed in our study. This interpretation is supported by the results of deep seismic profiles that suggest Grenvillian crust underlies all of the CVGT (Spencer et al. 1989; Hughes and Luetgert 1991), although it is unknown whether this basement structure existed during Late Silurian–Early Devonian VMS mineralization and prior to Acadian thrusting. Regardless of this uncertainty, the nearly identical Pb-isotope values of the Davis VMS deposit in the Hawley Formation (Shelburne Falls arc) and of the Royce and Croydon deposits (Bronson Hill arc), as discussed above, favor the Macdonald et al. (2014) model in which the BLL is west of the Silurian–Devonian cover sequence in New England, but not in Québec due to earlier oblique collision of peri-Ganderian arcs with an irregular Laurentian margin (van Staal et al. 2021*b*; van Staal and Zagorevski 2023). This regionally extensive tectonic boundary (Figs. 1 and 2) also clearly is west of the Bronson Hill belt, based on whole-rock Nd and Pb isotope data for the Ammonoosuc Volcanics that indicate a peri-Ganderian, not a Grenvillian, source for the parent magmas during the Ordovician formation of this belt (Dorais et al. 2012*a*).

Second Lake and Ironbound Mountain volcanic belts

The Silurian Second Lake and Lower Devonian Ironbound Mountain volcanic belts in westernmost Maine contain two significant VMS deposits at Ledge Ridge and Thrasher Peaks, respectively. Lead isotope data for these two deposits are relatively unradiogenic in terms of $^{206}Pb/^{204}Pb$ and are similar to the field for the Early Devonian VMS deposits of the Clinton–Frontenac belt of southeastern Québec (Fig. 3), which occur in correlative rift–facies metavolcanic and metasedimentary rocks and form part of the CVGT (Chevé 1990; Gauthier et al. 1994; Moench et al. 1999; Tremblay and Pinet 2005). Importantly, the low $^{206}Pb/^{204}Pb$ and $^{207}Pb/^{204}Pb$ values for the Ledge Ridge and Thrasher Peaks deposits, relative to those for nearly all others reported in this study, and the isotopic similarity to values for the Clinton–Frontenac VMS deposits, suggest that the Ledge Ridge and Thrasher Peaks deposits formed in successor basins on Grenvillian crust. This interpretation is supported by the uranogenic values for these two deposits relative to the age-corrected field for Grenvillian crust (Figs. 4A and 4B), and with the results of a deep seismic profile across the Québec–Maine border in this region that implicates a Grenvillian basement beneath the Second Lake and Ironbound Mountain volcanic belts (Spencer et al. 1989).

Jim Pond volcanic belt

The polymetallic Alder Pond deposit in west-central Maine occurs in mafic and felsic metavolcanic rocks of the Jim Pond Formation (Moench et al. 1999; Neil 2005). This formation is interpreted to have formed in an arc setting as part of a supra–subduction zone sequence that was juxtaposed in the Early Devonian on the structurally underlying Boil Mountain ophiolite complex (Gerbi et al. 2006). An Early Ordovician SHRIMP U–Pb zircon age of 484 ± 5 Ma for this formation was reported by Moench et al. (2000). On a regional scale, some previous workers have included the Jim Pond Formation in the Bronson Hill arc, but we discuss it separately here because of an older age for the volcanic rocks and a different tectonic setting. The Pb-isotope composition of the Alder Pond deposit is very unradiogenic, especially with respect to uranogenic lead in plotting slightly below the mantle growth curve (Fig. 3A); data for thorogenic lead plot on the curve for young upper crust (Fig. 3B). The geologic setting and Pb-isotope relationships together suggest that lead in the Alder Pond deposit was derived from both the mantle and a Grenville-type basement (Figs. 4A and 4B), a model consistent with the scenario of Gerbi et al. (2006) in which the Boil Mountain ophiolite and structurally overlying volcanic rocks of the Jim Pond Formation developed on a microcontinent close to the Laurentian margin. Possible support for this model comes from the presence of a $\sim$1075 Ma xenocrystic zircon grain in rhyolite from the Lower Ordovician Jim Pond Formation (Moench et al. 2000). Based on galena Pb-isotope data, similar arguments for the involvement of a Grenvillian lead source can be made for VMS deposits close to the Humber domain, as in the Ascot–Weedon volcanic belt of southeastern Québec (Swinden and Thorpe 1984) where Nd isotope data for felsic tuffs in this belt have εNd_i values as low as −9.9 that record a significant component of evolved continental crust (Samson and Tremblay 1996).

Popelogan volcanic belt

The Popelogan volcanic belt in northern Maine contains the largest VMS deposit in New England at Bald Mountain. This 30-Mt deposit, hosted in an intraoceanic arc sequence of mafic and minor felsic metavolcanic rocks (Schulz and Ayuso 2003; Slack et al. 2003), forms an inlier within the Ordovician Munsungun–Winterville belt of volcanic and sedimentary rocks (471–451 Ma; Wang et al. 2022*b*). A rhyolite tuff from the hanging wall of the Bald Mountain deposit has an early Middle Ordovician U–Pb zircon age of 467 ± 4 Ma (Ayuso et al. 2003). Lead isotope data for Bald Mountain form a cluster having a steep slope with respect to both uranogenic and thorogenic lead (Figs. 4A and 4B). All of these data show higher $^{206}Pb/^{204}Pb$ values than VMS deposits of the Penobscot Bay volcanosedimentary belt; most $^{206}Pb/^{204}Pb$ values are lower than those for deposits of the Shelburne Falls and Bronson Hill volcanic belts. Importantly, the Bald Mountain analyses fall mainly in the field of the Exploits peri–Ganderian arc–backarc tract in Newfoundland, thus supporting previous models for the Popelogan arc having formed in a Ganderian setting east of the BLL (van Staal et al. 2016, 2021*b*). Wang et al. (2022*a*) recently proposed a major relocation of the BLL to the east side of the Munsungun–Winterville belt, on the basis of a Laurentian U–Pb detrital zircon age spectrum obtained for the Cambrian (?) Chase Brook Formation, which is a basal mélange unit that underlies metavolcanic and metasedimentary rocks of the belt. However, metavolcanic and metasedimentary rocks of the Munsungun–Winterville belt comprise multiple thrust sheets (Wang et al. 2022*b*), including lower Middle Ordovician metavolcanic rocks 4 km southeast of the Bald Mountain deposit that overlie calcareous metasiltstone with Upper Ordovician conodont fauna (Harris 1997; Foose et al. 2003; former Middle Ordovician age assignment revised based on Goldman et al. 2023). These data imply the presence of a thrust contact between this metasedimentary unit and the overlying metavolcanic rocks. Moreover, it is unknown whether strata above the Chase Brook mélange are autochthonous on a regional scale and whether the detrital source areas are Grenvillian or Ganderian. Given these constraints and uncertainties, and the presence of Ganderian (Neoproterozoic) inheritance in zircon grains from a dacite in the Popelogan arc in northern New Brunswick (van Staal et al. 2016), we conclude that the BLL lies west of the Bald Mountain deposit but within the Munsungun–Winterville belt, along the eastern contact of the Chase Brook Formation with overlying strata including the early Middle Ordovician volcanic rocks containing the Bald Mountain VMS deposit (Fig. 2; Wang 2021) that formed on Ganderian crust (Schulz and Ayuso 2003). Importantly, the radiogenic $^{207}Pb/^{204}Pb$ endmember for the Bald Mountain deposit strongly suggests a Ganderian and not a Grenvillian source for the contained lead (Fig. 4A), thus arguing against a location for the BLL in northern Maine east of the Munsungun–Winterville belt as proposed by Wang et al. (2022*a*).

Weeksboro–Lunksoos volcanic belt

The Pickett Mountain (formerly Mount Chase) VMS deposit in the Weeksboro–Lunksoos volcanic belt of northeastern Maine is contained within a metavolcanic and metasedimentary arc sequence of early Middle Ordovician age (467 ± 5 Ma; Ayuso et al. 2003; see also Fyffe et al. 2023). Two samples of galena have $^{206}Pb/^{204}Pb$, $^{207}Pb/^{204}Pb$, and $^{208}Pb/^{204}Pb$ values more radiogenic than those of the Bald Mountain deposit. Importantly, results for both samples are within the principal field for VMS deposits of the Bathurst district of New Brunswick in the Exploits peri-Ganderian arc–backarc tract, which is widely viewed as comprising multiple, peri-Ganderian arcs that originated on the margin of the Amazonian craton (van Staal et al. 2012, 2021*b*). Formation of this volcanic belt on Ganderian crust (Waldron et al. 2019; van Staal et al. 2021*b*) is also supported by the presence of Mesoproterozoic ages for some zircon grains in a synvolcanic dacite porphyry near the Pickett Mountain deposit (Ayuso et al. 2003). In addition to the Pb-isotope data, another link to the Exploits peri-Ganderian arc–backarc tract in New Brunswick is provided by the presence of Celtic fauna within Arenig volcanic ash of both the Weeksboro–Lunksoos belt and Tetagouche Group (Poole and Neuman 2003). Note that it is difficult to fingerprint Ganderian basement as the radiogenic endmember for the Weeksboro–Lunksoos belt using Pb-isotope data alone (Figs. 4A and 4B; Ayuso et al. 2003; Dorais et al. 2012*b*; Walsh et al. 2021), thus requiring the integration of other geological and isotopic data.

Penobscot Bay volcanosedimentary belt

VMS deposits of the Penobscot Bay belt of coastal Maine are contained in Middle Cambrian strata that comprise metamorphosed siliceous metasedimentary rocks with minor felsic and mafic metavolcanics of the Ellsworth Schist and volcanic and volcaniclastic rocks of the overlying Castine Volcanics (508.6 ± 0.8 and 503.5 ± 2.5 Ma, respectively; Schulz et al. 2008). Excluding one vein interpreted here as tectonically remobilized sulfide, the Pb-isotope compositions of galena from four of five deposits define a cluster that mostly is distinct from the fields of other New England VMS deposits, being less radiogenic in $^{206}Pb/^{204}Pb$ than the early Middle Ordovician deposits of the Popelogan and Weeksboro–Lunksoos volcanic belts in northern and northeastern Maine, respectively (Figs. 4A and 4B). The small Appleton deposit, in younger Ordovician metabasalt west of Penobscot Bay, is isotopically distinct from Neoproterozoic VMS deposits of Avalonian affinity in Nova Scotia and Newfoundland (Fig. 3), but the very high $^{208}Pb/^{204}Pb$ value is unusual and may reflect the selective introduction of thorogenic lead during post-ore metamorphism. Alternatively, this high $^{208}Pb/^{204}Pb$ value could relate to intense weathering in the lead source (see Zartman and Doe 1981; Zartman and Haines 1988).

Most of the Cambrian Penobscot Bay deposits are more radiogenic in $^{206}Pb/^{204}Pb$ than the Neoproterozoic VMS deposits of the Avalon domain in New Brunswick, Nova Scotia, and Newfoundland (Fig. 3). This contrast supports a model in which Cambrian volcanosedimentary strata of the Penobscot Bay belt formed on Ganderian, not Avalonian basement, consistent with lower $^{206}Pb/^{204}Pb$ values for the Penobscot Bay VMS deposits relative to the field for Avalonian crust (Figs. 4A and 4B). Based on detrital U–Pb zircon data for the Ellsworth Schist (Fyffe et al. 2009) and contrasts in the detrital zircon age spectra of this formation relative to those for other coastal belts of New England and New Brunswick, van Staal et al. (2021*b*) proposed that the basement of the Penobscot Bay region is a WAC succession that includes metasedimentary and minor metaigneous rocks of likely Paleoproterozoic age in the Islesboro block (see Reusch et al. 2018), potentially including some metasedimentary rocks on the mainland to the west in the Rockport area (cf. Osberg and Berry 2020), but no direct evidence exists for the latter possibility. This hypothesis for widespread Paleoproterozoic basement in the Penobscot Bay area is supported by the projection of $^{207}Pb/^{204}Pb$ and $^{208}Pb/^{204}Pb$ values of the local VMS deposits toward the field for likely WAC rocks of the Islesboro block (previously considered Neoproterozoic; Stewart et al. 2001). It is also supported by the lower $^{206}Pb/^{204}Pb$ values of the projected radiogenic endmember for the field of Penobscot Bay VMS deposits relative to the higher values of the projected endmembers for the other principal New England VMS fields (Figs. 4A and 4B). The WAC field includes data for Paleoproterozoic rocks of the Amazonian craton because several studies have proposed that this craton—and not the WAC—was proximal to New England in the late Paleozoic during closure of the Rheic Ocean (e.g., Domeier and Torsvik 2014; van Staal et al. 2021*b*). Although limited, whole-rock Pb-isotope data for the inferred WAC rocks of Islesboro are consistent with results of a deep seismic profile in the Penobscot Bay region. This profile was interpreted by Stewart et al. (1993) to record northward thrusting of the Ellsworth Schist, related early Paleozoic strata (including overlying Castine Volcanics), and rocks of the Islesboro block, over middle Paleozoic metasedimentary rocks of the Fredericton trough and an unexposed basement on the mainland inferred to be Proterozoic (see Stewart et al. 1993). A Paleoproterozoic basement in the Penobscot Bay region, potentially derived from 2.05–1.75 Ga WAC rocks in Morocco (cf. Walsh et al. 2002; Pereira et al. 2015; Letsch et al. 2018; Ikenne et al. 2017), was first proposed by Reusch et al. (2018) for the Islesboro block and since then, this old basement has been recognized in several other parts of coastal New England and the Maritimes (Kuiper et al. 2022; Waldron et al. 2022). If correct, our VMS-based Pb-isotope model implies that WAC basement may extend beyond the Islesboro block and could underlie much of the Penobscot Bay region. Additional support for this interpretation comes from Pb-isotope data for K-feldspars from several granitic plutons in this region (e.g., Vinalhaven and Sedgwick), which have relatively low $^{208}Pb/^{204}Pb$ for given $^{206}Pb/^{204}Pb$ values that approach the growth curve for old upper crust (compare Figs. 3B and 4B).

Implications for subarc basement rocks

The clustering of VMS Pb-isotope data into least three groups of New England volcanic and volcanosedimentary arcs (Figs. 4A and 4B) has implications for the nature of the subarc basement rocks. The steep trends for both uranogenic and thorogenic lead suggest the presence of at least two endmembers for each group. Although these endmembers are unknown, the trends are consistent with an unradiogenic mantle component and a radiogenic component likely dominated by Proterozoic basement, each contributing lead to VMS mineralization. In this context, the key process involves deeply circulating hydrothermal fluids that leach lead from basement rocks, as well as from overlying volcanic ± sedimentary strata of the arc assemblages. Lead sourced in footwall felsic volcanic rocks may derive originally by partial melting of the basement rocks. Basement lead can then become involved in arc and backarc volcanism by the generation of arc magmas during partial melting of deep crustal rocks, and by enrichment of the volcanic pile via mass transfer of lead from the slab by way of the mantle wedge. Variations in the Pb-isotope data clusters are reflected largely by $^{206}Pb/^{204}Pb$ values, which could imply different ages for the radiogenic endmembers, including Proterozoic basements, although this possibility cannot be tested using our analyses because lead in the VMS deposits did not evolve in one homogeneous reservoir (i.e., presence of at least two Pb-isotope endmembers). The inferred deep-seated fluids may carry basement-derived lead that then leached additional lead from footwall volcanic ± sedimentary rocks near the site of sulfide mineralization. Alternatively, the two endmembers may reflect small hydrothermal systems that did not allow sufficient mixing and hence a homogenous Pb-isotope signature.

Because VMS deposits derive their metals mainly from the substrate of the volcanic pile, the isotopic composition of lead in the deposits represents an integrated composition of all of the lead seen by the hydrothermal fluids. The lead in the volcanic pile is most immediately sequestered in felsic igneous rocks, which are generated mainly by partial melting in the deep parts of the crust where the isotopic composition of the lead largely reflects the composition of any crustal substrate. Our study of lead in New England VMS deposits suggests that their Pb-isotope signatures can be used to draw some conclusions about the nature of the pre-accretionary crust and tectonostratigraphic settings of the deposits.

Radiogenic endmembers of the Pb-isotope mixing lines are in all cases interpreted as broadly crustal and reflecting the composition of the crust at various places in the early Paleozoic. The most widespread contributor to VMS deposits in the Ordovician was a relatively radiogenic crust that we interpret as broadly Gondwanan, based on other evidence as outlined above (e.g., Nd isotopes, zircon inheritance, and deep seismic reflection data). Lead from this basement source is found in the Shelburne Falls/Bronson Hill, Popelogan, and Weeksboro–Lunksoos volcanic belts, thus supporting the interpretation that these belts formed in one or more peri-Ganderian arc assemblages that correlate isotopically and tectonostratigraphically with the Exploits peri-Ganderian arc–backarc tract. The different $^{206}Pb/^{204}Pb$ values observed for the Popelogan and Shelburne Falls–Bronson Hill belts—and possibly also the Weeksboro–Lunksoos belt—support previous models in which Ganderia in the New England region (Fig. 2) is a composite terrane comprising multiple volcanic arc assemblages characterized by different Pb-isotope reservoirs and possibly two or more different Proterozoic basements (cf. van Staal et al. 2021*b*; Waldron et al. 2022). Isotopically distinct and less-radiogenic lead is found in a somewhat younger group of VMS deposits within the CVGT and Second Lake/Ironbound Mountain volcanosedimentary sequences. These Pb-isotopic values are similar to those for VMS deposits in the Notre Dame peri-Laurentian arc–backarc tract of Newfoundland, implying that the radiogenic endmember may similarly be Laurentian crust. These deposits are younger than those of the Notre Dame peri-Laurentian arc–backarc tract, and formed in the Late Silurian to Early Devonian, in successor basins following the initial stages of collision of Iapetus volcanic arcs with Laurentia. Although closely associated with sequences of Ganderian affinity, the lead in these deposits may reflect the presence of Laurentian crust in the deep structural footwall, and their present location is not necessarily relevant to the location of the BLL in the Ordovician. The Lower Ordovician Jim Pond deposit in western Maine displays a less radiogenic character and appears to have formed as part of the peri-Laurentian Notre Dame peri-Laurentian arc–backarc tract, based especially on $^{207}Pb/^{204}Pb$ and $^{208}Pb/^{204}Pb$ values that suggest a predominant mantle lead source with a minor contribution derived from Laurentian continental crust (Figs. 4A and 4B).

Variations in calculated values of μ ($^{238}U/^{204}Pb$) for the New England VMS deposits support our model for a regional Pb-isotope provinciality based on the range of $^{206}Pb/^{204}Pb$ ratios. The advantage of μ values is that these represent a single proxy that integrates $^{206}Pb/^{204}Pb$, $^{207}Pb/^{204}Pb$, and model age (Huston et al. 2010). Similar to Nd model ages, μ values for VMS deposits can be used to map and distinguish upper crustal source reservoirs of lead, as done by Champion and Huston (2016) for Archean deposits in Western Australia and eastern Canada. Although ranges overlap in some cases, the different average μ values determined for the Jim Pond volcanic belt (8.94), Ironbound Mountain and Second Lake volcanic belts (9.49), VCB (9.49), Shelburne Falls–Bronson Hill volcanic belts (9.80), Popelogan volcanic belt (9.70), and Weeksboro–Lunksoos volcanic belt (9.99) suggest diverse reservoirs for lead contained in the deposits (Fig. 4C). Identical averages for the deposits of the VCB and Ironbound Mountain and Second Lake volcanic belt are consistent with both belts having formed within Silurian–Devonian cover sequences. The average for the Penobscot Bay volcanic belt (9.83) is similar to that for the Shelburne Falls–Bronson Hill belts (9.80), but geological, geochemical, and tectonic data clearly indicate that these two belts are unrelated. Differences in average μ values among Ordovician VMS deposits in the Jim Pond, Shelburne Falls–Bronson Hill, Popelogan, and Weeksboro–Lunksoos volcanic belts further support our model that $^{206}Pb/^{204}Pb$ variations record the presence in Ganderia of composite subterranes having different Proterozoic basements likely with different ages. Different basements are also implicated between the Popelogan (Bald Mountain deposit) and Weeksboro–Lunksoos (Pickett Mountain deposit) belts, based on contrasting median εNd_t values of +4.7 and −2.6, respectively, and on differences in median μ values (Fig. 4C). Owing to the limited nature of our data, however, we cannot evaluate zoning or gradients in μ or Pb-isotope values that could mark the boundaries between crustal blocks.

Conclusions

Lead isotope compositions of galena from VMS deposits in the New England region provide new insights into tectonostratigraphic assemblages of the arc, backarc, and other volcanic and volcanosedimentary strata that host the deposits, as well as into the identity of underlying basements. These data also contribute to the correlation of tectonostratigraphic terranes along strike within the Appalachian orogen.

Principal sources of lead include the mantle, volcanic ± sedimentary rocks, and basement rocks. Integration of the Pb-isotope values with published data from Nd isotopes of the volcanic rocks and from deep seismic reflection profiles suggests the involvement of several discrete types of basements, including those of Laurentian (Grenvillian) and Gondwanan (Amazonian and West African) affinity. The Pb-isotope data for VMS deposits within individual Cambrian and Ordovician volcanic and volcanosedimentary assemblages delineate four and possibly as many as five clusters, which likely reflect mineralizing processes that homogenized lead from the different metal sources that contributed to each deposit. We propose that within these different clusters, the radiogenic endmembers were isotopically distinct basement rocks and that each tectonostratigraphic assemblage originated in different parts of the Iapetus and Rheic oceans. A similar Pb-isotope approach to discriminating pre-accretionary basements may be fruitful in other orogens that contain VMS deposits within multiple volcanic arc and backarc sequences.

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Data availability

All data are listed in Table 1 and Table S1. Additional data for New England only are in Slack, J.F., LeHuray, A.P., and Ayuso, R.A. 2023. Geological and Pb-isotope data for New England volcanogenic massive sulfide deposits. U.S. Geological Survey Data Release, https://doi.org/10.5066/P91QCII1.

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The authors declare no competing interests.

Supplementary material

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