1 **Geochemistry of Woranso-Mille Pliocene Basalts from West-Central Afar, Ethiopia:**

2 **Implications for Mantle Source Characteristics and Rift Evolution**

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20 21 **Abstract**

22
23 The Woranso-Mille (WORMIL) area in the west-central Afar, Ethiopia, contains several
24 Pliocene basalt flows, tuffs, and fossiliferous volcanoclastic beds. We present whole-rock major-
25 and trace-element data including REE, and Sr-Nd-Pb isotope ratios from these basalts to

characterize the geochemistry, constrain petrogenetic processes, and infer mantle sources. Six basalt groups are distinguished stratigraphically and geochemically within the interval from ~3.8 to ~3 Ma. The elemental and isotopic data show intra- and inter-group variations derived primarily from source heterogeneity and polybaric crystallization $\pm$ crustal inputs. The combined Sr-Nd-Pb isotope data indicate the involvement of three main reservoirs: the Afar plume, depleted mantle, and enriched continental lithosphere (mantle $\pm$ crust). Trace element patterns and ratios further indicate the basalts were generated from spinel-dominated shallow melting, consistent with significantly thinned Pliocene lithosphere in western Afar. The on-land continuation of the Aden rift into western Afar during the Pliocene is reexamined in the context of the new geochemistry and age constraints of the WORMIL basalts. The new data reinforce previous interpretations that progressive rifting and transformation of the continental lithosphere to oceanic lithosphere allows for increasing asthenospheric inputs through time as the continental lithosphere is thinned.

Keywords: Afar Rift; Geochemistry; Basalt; Mantle Plume; Lithosphere

1. Introduction

The volcanism and tectonics of the Afar and adjacent region is of interest because it is a key place to understand continental-to-oceanic rift evolution (Makris & Ginzburg, 1987; Hart et al., 1989; Acton et al., 1991; Deniel et al., 1994; Ebinger & Hayward, 1996; Chernet et al., 1998; Wolfenden et al., 2004, 2005; Buck, 2006; Furman et al., 2006; Corti, 2009; Acocella, 2010; Beutel et al., 2010; Bastow & Keir, 2011; Hammond et al., 2011; Bridges et al., 2012; Rooney et

al., 2012; Ferguson et al., 2013). Continental rifting and volcanism in the region started in the Oligocene and continued through the Quaternary. An upwelling mantle plume has long been considered responsible for the genesis of the extensive Ethiopian flood basalts (e.g. Schilling, 1973; Schilling et al., 1992; Marty et al., 1996; Kieffer et al., 2004; Pik et al., 2006). The main phase of volcanism of the flood basalts occurred ~ 30 million years ago and lasted for ~1 Ma (Hofmann et al., 1997). Convecting asthenospheric mantle is also believed to have caused thermal erosion of the subcontinental lithosphere leading to rifting and thinning as well as decompression and partial melting that has sustained volcanism in Afar up to the present (e.g. Deniel et al., 1994; Furman et al., 2006; Rooney, 2010; Rooney et al., 2013). While the Afar mantle plume has been considered primarily responsible for the genesis of the Oligocene Ethiopian flood basalts, the contribution of the LREE-depleted (e.g. Barrat et al., 2003; Daoud et al., 2010) and depleted MORB mantle components (Hart et al., 1989; Deniel et al., 1994; Meshesha and Shinjo, 2008) are known to have increased with time, particularly during Plio-Pleistocene Afar magmatism.

Results from geochemical studies of basalts from the Ethiopian plateau (~30-20 Ma) (Hofmann et al., 1997; Pik et al., 1999; Kieffer et al., 2004; Ayalew & Gibson, 2009; Beccaluva et al., 2009; Ayalew, 2011), the rift escarpment (& associated marginal basins) (~20-5 Ma) (Hart et al., 1989; Rooney et al., 2013), the Main Ethiopian Rift (MER), Afar rift, Djibouti and the Gulf of Aden (≤ 2 Ma) (Schilling, 1973; Vidal et al., 1991; Schilling et al., 1992; Deniel et al., 1994; Trua et al., 1999; Barrat et al., 2003; Daoud et al., 2010; Rooney et al., 2010, 2012; Ayalew et al., 2016) have produced regional magma source characteristics broadly documenting the transition from initial to final stages of continental rifting and rupturing. These studies reveal diverse

1 geochemical compositions resulting largely from interactions between multiple mantle reservoirs,
2 variations in the amount of partial melting and depth at which melting occurred and variations in
3 the depth of primary crystal fractionation $\pm$ assimilation, all of which are related to the degree of
4 lithospheric extension and thinning.

5
6 The western part of the Afar rift, and many of the basins within, contain incised volcano-
7 sedimentary sections of paleontological interest and preserve a wealth of information on hominid
8 origin and evolution (e.g. Renne et al., 1999; Haile-Selassie et al., 2007; Quade et al., 2008;
9 Wynn et al., 2008; Saylor et al., 2016). The Woranso-Mille (WORMIL) area is one of the
10 paleontology and paleoanthropology sites in west-central Afar (Fig. 1; Haile-Selassie et al.,
11 2007) that also contains multiple Pliocene basalt units. The WORMIL Pliocene basalt suite is
12 located between the Miocene basalts of the continental rift shoulder in the west and the
13 Quaternary basalts in the northeast, which are part of the rift axis and have characteristics of a
14 nascent seafloor spreading (e.g. Wright et al., 2006; Rowland et al., 2007; Ebinger et al., 2010).

15
16 Here we report new geochemical and isotopic data on basalts from higher resolution sampling in
17 the northern WORMIL area. The age of the basalt suite has, so far, been considered arbitrarily
18 as part of the 8-6 Ma old Dalha Formation (Varet, 1975; Tefera et al., 1996); and is now
19 constrained to be middle to late Pliocene in age from the $^{40}\text{Ar}/^{39}\text{Ar}$ dating of intercalated tuff
20 units (Deino et al., 2010). The objective is to evaluate the geochemistry of these Pliocene basalt
21 units and compare the results with data from neighboring regions to shed light on the competing
22 roles of lithospheric, mantle plume and depleted asthenospheric sources during magma
23 generation and progressive.

2. Regional Geologic Setting

2.1 Litho-Stratigraphy

The Afar area consists, primarily, of Miocene to Quaternary volcanic and sedimentary rocks (Zanettin & Justin-Visentin, 1974; Varet, 1975; Barberi & Varet, 1977; Mohr, 1983; Walter et al., 1987; Tefera et al., 1996), of which basalt is by far the most extensive and widespread. In the periphery and escarpment of the Afar rift the units are composed of Neoproterozoic basement rocks, Mesozoic sediments and pre-Miocene/early Miocene trap and flood basalts with minor intercalated silicic units. In the rift floor the lithological units based on Varet (1975), Tefera et al. (1996) and Beyene and Abdelsalam (2005) are: *Mabla Rhyolites* (14-10 Ma), *Dalha Basalts* (8-6 Ma), *Afar Stratoid Series* (4-1 Ma), *Transverse Volcanics* (<4 Ma), *Marginal Silicic Centers* (<4 Ma), *Plio-Quaternary Sediments* and *Quaternary Axial Volcanic Ranges* (Fig. 1).

2.2 Tectonic

The Afar region is formed at the triple junction between the Red Sea, Gulf of Aden and Main Ethiopian rifts as a result of motions between the Nubian, Arabian and Somalian plates (Fig. 1). It is characterized by a thin lithosphere transitional between continental and oceanic regimes. The region began continental rifting starting from ~ 24 Ma to ~4 Ma and transitioned to lithospheric rupture since ~ 4 Ma (Beyene and Abdelsalam, 2005). The Afar lithosphere started to stretch through diffused and localized faults following the westward and subsequent

1 northwestward propagation of the Gulf of Aden rift together with the southward propagation of
2 the Red Sea rift, both overlapping near the triple junction ~20 Ma ago (Audin et al., 2004).
3 Between 10 and 4 Ma continued westward propagation and transfer of extension from the Gulf
4 of Aden extended into the southern Afar concomitant with Dalha basalt emplacement (Audin et
5 al., 2004).

6
7 Extension in the northern MER and its northward propagation to form the triple junction also
8 occurred at around 11 Ma; global plate re-organization at ~7-3 Ma caused by rift propagation
9 resulted in strain transfer from border faults to smaller offset faults (Wolfenden et al., 2004,
10 2005). Moreover between 4 and 2 Ma the along-axis propagation of the Gulf of Aden and Red
11 Sea spreading centers prompted renewed extension and volcanism in the Afar triple junction area
12 (Wolfenden et al., 2004).

13
14 There is a good understanding of the timing between bimodal volcanism and rift propagation in
15 central Afar for the last 4 Ma. Lahitte et al. (2003a, b) reported the role of silicic and basaltic
16 volcanism in relation to the degree of extension and rift propagation. They showed that silicic
17 lavas were the first to erupt in areas of low extensional strain followed by fissural basaltic
18 volcanism as extension increased. The emplacement of magmas under the influence of a plume
19 or of local eruptive centers weakens the lithosphere thereby localizing the deformation zone
20 (Lahitte et al., 2003b).

21
22 Along the western Afar margin the relationship between Mio-Pliocene volcanism and extensional
23 deformation is yet to be well understood. Audin et al. (2004) suggested linkage between the

Dalha basalt volcanism and rift propagation in the Sullu Adu (Guda) area of western Afar but the exact timing of basalt emplacement relative to the localization of deformation into axial rift zones is poorly constrained. The WORMIL area lies at the western flank of the central Afar rift and is ~100 km SW of the prominent Manda Hararo-Tendaho-Gobaad rift and ~150 km west of the present Afar triple junction at Lake Abbe (Fig. 1). The basalt units in WORMIL have previously been mapped as Dalha Formation (e.g. Varet, 1975; Acton et al., 1991; Tefera et al., 1996). Specifically, the extensive basalt unit at Guda plateau was apparently recognized as a horst structure (e.g. Walter, 1980) ascribed to Dalha Formation. The age of this and other basalt units in northern WORMIL has now been constrained to between ~4 Ma and ~3 Ma (Deino et al., 2010; Saylor et al., 2016). Most of the WORMIL basalts are thus younger than the presumed age of the Dalha Formation [8-4 Ma (Audin et al., 2004) or 8-6 Ma (Acton et al., 1991; Beyene and Abdelsalam, 2005)], and belong instead to the Afar Stratoid Series (4-1 Ma).

3. Woranso-Mille Basalts

The basalt sequence in the northern WORMIL area comprises: Mille basalt (MLB), Gugubsi basalt (GGB), Am-Ado –Aralee Issie basalt (AAB), Kerare-Burtele basalt (KBB), Daba Dora basalt (DDB) and Guda basalt (GDB). They are intercalated and interspersed with primary tuffs, reworked volcanoclastic horizons, and fossiliferous clastic sedimentary rocks, which are collectively grouped here as ‘lower’, ‘middle I’, ‘middle II’ and ‘upper’ units (Figs 2 and 3) and are identified primarily by their positions relative to marker tuffs. Neither well defined vent locales nor multiple basalt group stratigraphic sequences are observed, thus extents of individual flows cannot be evaluated. Although, some of the basalt groups (e.g. MLB, KBB, GDB) appear

to represent multiple eruptive pulses (or flows). The WORMIL basalts and the tuffaceous and clastic sedimentary rocks have been cut by several mostly NW-trending normal faults with minor (2-7 m) offsets (Fig 2). The relative stratigraphic position of the basalts with respect to the intercalating tuffaceous and sedimentary units is illustrated in figures 3 and 4. The description of the WORMIL basalt groups is included as supplementary data.

4. Analytical Techniques

Sixtyfive northern WORMIL basalt samples were selected for major and trace element concentration determinations using a PANalytical 2404 X-ray fluorescence (XRF) vacuum spectrometer at Franklin and Marshall College, Lancaster, PA, USA following the techniques outlined in Mertzman (2000). This includes determination of ferrous iron by standard titration methods and total volatile content (LOI). Replicate XRF analyses of standard BHVO-2 are provided in Supplementary Table S1, as are estimates of analytical precision.

Additional trace element concentrations (Be, REE, Hf, Ta, Pb, Th, U) for a subset of thirteen samples were determined by Inductively Coupled Plasma Mass Spectrometry (ICP-MS) at Miami University, Oxford, OH, USA. ICP-MS solutions for unknowns and standards were generated via flux fusion involving the following steps; 1) powders were dried at 110°C for 12 hours, 2) 50 mg of dried powder was combined with 75 mg lithium metaborate (LiBO_2) in a pre-fired graphite crucible, 3) mixtures were fused at 950°C for 20 minutes and allowed to cool in air, 4) resulting glass beads were added to 125 mL of 1% high-purity HNO_3 and agitated on a gyratory shaker until total dissolution was achieved. A processing blank (zero standard) was

prepared following the same steps using only 75 mg of LiBO_2 . Within 24 hours of dissolution, approximately 10 mL aliquots of unknown, standard, and blank solutions were analyzed with a Varian 820 ICP-MS equipped with an autosampler. The blank and a matrix-matched suite of 12 calibration standards (CRMs) were analyzed at the beginning of the instrument session. Background adjusted counts were corrected for within-run drift through the use of mass appropriate internal standards (100 ppb of ^{72}Ge , ^{115}In , ^{185}Re , and ^{209}Bi). Replicate ICP-MS analyses of sample BB12 and of standard BHVO-2 are provided in Supplementary Table S1, as are estimates of analytical precision.

Pb, Sr and Nd isotope ratios were measured by Thermal Ionization Mass Spectrometry (TIMS) on a Thermo-Finnigan Triton at Miami University for the same samples analyzed by ICP-MS. Approximately 0.1 gram of sample powder was dissolved in concentrated HF-HNO_3 with digestion steps and Pb, Sr, and Nd separations following conventional procedures outlined by Snyder (2005). Pb isotope mass fractionation corrections used in this study are $^{206}\text{Pb}/^{204}\text{Pb} = 0.098\text{‰/amu}$; $^{207}\text{Pb}/^{204}\text{Pb} = 0.094\text{‰/amu}$; and $^{208}\text{Pb}/^{204}\text{Pb} = 0.10\text{‰/amu}$. The long-term NBS 981 external reproducibilities (2σ) for $^{206}\text{Pb}/^{204}\text{Pb}$, $^{207}\text{Pb}/^{204}\text{Pb}$, and $^{208}\text{Pb}/^{204}\text{Pb}$ are ± 0.015 , ± 0.020 , and ± 0.063 , respectively. Measured $^{87}\text{Sr}/^{86}\text{Sr}$ was corrected for mass fractionation using $^{86}\text{Sr}/^{88}\text{Sr} = 0.1194$. Long-term external reproducibility (2σ) of the NBS 987 Sr standard at Miami University is ± 0.000017 for $^{87}\text{Sr}/^{86}\text{Sr}$ with a running mean of 0.710239 ($n = 174$). $^{143}\text{Nd}/^{144}\text{Nd}$ was corrected for fractionation using $^{146}\text{Nd}/^{144}\text{Nd} = 0.7219$. The long-term external reproducibility (2σ) of the La Jolla Nd standard at Miami University is ± 0.000008 for $^{143}\text{Nd}/^{144}\text{Nd}$ with a running mean of 0.511846 ($n = 119$). The above noted 2-sigma errors are based on long-term reproducibility of standards with individual within-run errors in all cases less

than these values, thus these internal errors are not reported.

5. Results

5.1 Major and Trace Element Geochemistry

Major and trace element data determined by XRF and incompatible element data determined by ICP-MS are presented in Table 1 for representative samples from the six previously defined WORMIL basalt groups. Two spatially associated samples of unknown chronostratigraphic context (BW13 and BMT39) are also included. The full XRF sample suite can be found in Supplementary Data Table 2. The major element oxides plotted in all diagrams are anhydrous normalized values with total Fe expressed as Fe^{+2} (FeO^*).

The WORMIL samples illustrate a range of differentiation ($\text{MgO} \sim 4.8 - 9.1 \text{ wt\%}$ and $\text{SiO}_2 \sim 44 - 51 \text{ wt\%}$; Tables 1 and S2). On the total alkalis versus silica classification diagram the entire suite plots in the subalkaline region and is dominated by basaltic compositions that also demonstrate within- and between-group differences (Fig. 5A). Selected basalts in either geographic or temporal proximity (or both) to the WORMIL suite are shown for comparison (see caption). The relationships between the ratios Nb/Y and Ti/Y were used by Pik et al. (1998) to classify/subdivide the Oligocene Ethiopian flood basalts. This plot also serves to illustrate two important characteristics of the WORMIL basalt suite: (1) within- and between-group geochemical distinctions, and (2) a compositional range extending from very low Nb/Y and Ti/Y, akin to, for example, the LT (low-Ti) flood basalt subgroup, to higher values overlapping

1 with those of spatially associated higher Ti flood and NW Plateau basalts and southern Afar –
2 northern MER basalts.

3
4 Major, minor, and trace elements are compared to MgO in Figures 6 and 7. Collectively, the
5 major element plots of Figure 6 do not illustrate simple suite-wide differentiation trends. Rather,
6 these plots emphasize distinctions between the basalt groups and within-group variations
7 indicative of group- (eruptive episode) specific petrogenetic histories. The CaO, FeO*, TiO₂, and
8 P₂O₅ plots clearly highlight these characteristics. The KBB and ABB groups contain the least
9 differentiated, highest MgO and lowest TiO₂, P₂O₅, and K₂O flows in the suite, while also
10 displaying the most coherent within-group trends. The KBB flows often are plagioclase phyric
11 and possess variable olivine contents ranging to the highest in the WORMIL suite, in contrast to
12 the finer grained olivine and plagioclase-poor AAB flows. The GDB and GGB groups define the
13 most differentiated members of the suite with elevated TiO₂, P₂O₅, and K₂O contents. It should
14 be noted that apparently anomalous elevated CaO and K₂O values in a few of the KBB, MLB,
15 and GDB samples may reflect post-solidification alteration processes, consistent with the
16 presence of secondary clay and carbonate minerals.

17
18 Selected trace element concentrations and the Ce/Pb ratio are plotted against MgO in Figure 7. In
19 a similar fashion to the major element plots, the trace element plots do not illustrate simple suite-
20 wide differentiation trends. Rather, these plots further emphasize distinctions between the basalt
21 groups and within-group variations indicative of group- (eruptive episode) specific petrogenetic
22 histories. The KBB and AAB groups, which are both intermediate in age, but
23 chronostratigraphically distinct, contain among the highest Ni and lowest Cr, Rb, Sr, Zr, and Nb

flows in the suite, while also displaying the most coherent within-group trends. The youngest (GDB) and second oldest (GGB) basalt possess the lowest Ni, the highest Zr, and Nb, and variable but generally elevated Sr, Cr, Rb, and Ba contents. The distinctive trends and clusters illustrated by the compatible element Cr and the incompatible immobile elements Zr and Nb accentuate the unique and complex fingerprints of the different chronostratigraphic groups. The Ce/Pb versus MgO plot also highlights between-group distinctions while revealing no simple correlation between Ce/Pb and degree of differentiation. Moreover, the WORMIL suite falls within or very close to the average range of Ce/Pb suggested for oceanic basalts (see caption).

Incompatible element characteristics are summarized in Figure 8 and compared to selected regional basalt suites (see caption). The WORMIL samples and the comparison suites are divided into two sets of plots to facilitate clarity; no petrogenetic implication is assigned to this division. Chondrite-normalized REE patterns (Fig. 8A& C) vary systematically according to apparent degree of differentiation from relatively flat with $(La/Yb)_N$ of 1.9 in the higher MgO AAB and KBB basalts to LREE enriched with $(La/Yb)_N$ of 5.6 in the lower MgO GGB and GDB basalts. The MLB and DDB samples plus the two samples of unknown context (BW13 and BMT39) exhibit patterns intermediate to these endmembers. There is a notable absence of pronounced negative Eu anomalies with increasing total REE content for the main basalt groups with only sample BW13 preserving a significant negative anomaly ($Eu/Eu^* = 0.81$). In contrast, the two highest total REE GDB samples (BDD23 and BWK25) preserve slight positive Eu anomalies ($Eu/Eu^* = 1.2-1.3$). The more LREE-enriched basalts show patterns similar to Pliocene-Quaternary southern Afar and northern MER basalts and Oligocene to Miocene higher-

1 Ti flood basalts, shields, and dikes. In contrast, the less LREE-enriched DDB, AAB, and KBB
2 samples are most similar to Oligocene Low-Ti flood basalts

3
4 On the primitive mantle normalized multi-element plots(Fig. 8B& D) there is broad similarity in
5 the trace element patterns for most of the northern WORMIL basalts, yet as in the case of the
6 REE, the patterns clearly distinguish the basalt groups. As noted for Rb, Ba, Zr, and Nb in Figure
7 7, the more evolved basalts (GGB and GDB) possess the highest incompatible element
8 concentrations, the MLB and DDB have intermediate concentrations, and the least evolved
9 basalts (AAB and KBB) are the most depleted. The trace element patterns also illustrate the
10 prevalence of crossing patterns and the somewhat erratic behavior of Ba, Th, Pb, P, and Sr. All
11 of the WORMIL basalts possess Rb and K depletions, a feature observed in other Ethiopian
12 basalt suites (e.g. Ayalew et al., 2016). As with the REE, the full trace element patterns for the
13 GDB group resemble those from Pliocene-Quaternary southern Afar and northern MER basalts.
14 Other specific comparisons are less obvious.

15 16 *5.2 Sr-Nd-Pb Isotope Geochemistry*

17
18 Table 2 summarizes the new WORMIL Sr, Nd and Pb isotope results for the same subset of
19 samples reported in Table 1. No suite-wide correlations exist between $^{87}\text{Sr}/^{86}\text{Sr}$, $^{143}\text{Nd}/^{144}\text{Nd}$, or
20 $^{206}\text{Pb}/^{204}\text{Pb}$ and parent-daughter element ratios or $1/\text{Sr}$, $1/\text{Nd}$, or $1/\text{Pb}$, respectively. The
21 $^{208}\text{Pb}/^{204}\text{Pb}$ ratios display a narrow range of variation from 38.18 to 38.70 and no correlation with
22 Th/Pb , but with a hint of decreasing $^{208}\text{Pb}/^{204}\text{Pb}$ with increasing $1/\text{Pb}$. An exception is AAB
23 sample BAR62, which has an elevated $^{208}\text{Pb}/^{204}\text{Pb} = 40.88$. BAR62 also possesses a high Th/Pb

ratio (2.42) well above the values for the remainder of the suite (0.95 ± 0.22). This sample also displays the least radiogenic Nd isotope ratio of the WORMIL suite.

Figure 9 compares the WORMIL isotope characteristics to those of the previously identified regional basalt suites and to other regional reservoirs (see caption). In Sr-Nd space (Fig. 9A) the KBB, MLB and GGB samples (+ BMT39 and BW13) cluster around the Afar plume endmember composition (Rooney et al. (2012, 2013) that falls at the confluence of the west-central Afar, southern Afar – northern MER, and Gulf of Aden trends. The remaining WORMIL samples project to more isotopically evolved compositions toward endmembers such as EMI, EMII and Pan African Lithosphere (Zindler and Hart, 1986; Hofmann, 2003; Rooney et al., 2012, 2013). The very heterogeneous Pan African crust plus younger spatially associated upper crustal lithologies (Oligo-Miocene and Pliocene rhyolites; Ayalew and Yirgu, 2003; Walter et al., 1987) occupy a large portion of Sr-Nd space within which all of the WORMIL samples fall. The highest $^{87}\text{Sr}/^{86}\text{Sr}$ GDB sample BWK25 also has among the highest Ce/Pb ratios (35) of the suite, and displays elevated levels of P, Sr, Ba, Eu and overall incompatible element enrichment (Fig. 8B). In contrast, the AAB sample BAR62 with the lowest $^{143}\text{Nd}/^{144}\text{Nd}$ and anomalously high $^{208}\text{Pb}/^{204}\text{Pb}$ (~41) also carries a high Ce/Pb ratio (33) yet possesses relatively low P, Sr, Ba, and Eu, and a depleted trace element pattern.

In the Pb-Pb plot (Fig. 9B) all samples fall at the overlap of the regional trends mentioned above. This placement is within the triangular area defined by the three often cited regional endmembers: the Afar plume, the depleted mantle (DM) and the Pan African Lithosphere (PAL or PAL). This placement also falls at the low $^{207}\text{Pb}/^{204}\text{Pb}$ range of representative lower and

upper crustal lithologies. Similarly, in $\text{Sr-}^{206}\text{Pb}/^{204}\text{Pb}$ space (Fig. 9C) all samples fall within the Afar-MER fields except for the previously discussed BWK25 that accentuates a general trend within the dataset toward upper crustal compositions or EMII-PAL type components.

6. Discussion

6.1 Process and source effects

The Afar rift setting is an excellent location to assess the contributions from depleted and enriched mantle reservoirs, particularly given that previous work has demonstrated that while certainly not absent, the crustal influences on basaltic magma evolution are generally secondary to the nature of the ultimate magma sources being tapped (e.g., Hart et al, 1989; Pik et al., 1999; Rooney et al., 2013, 2014; Ayalew et al., 2016). Moreover, even some of the more differentiated MER and Afar mafic lavas that likely evolved through combined shallow crystallization and mixing with locally derived rhyolitic or liquids can be interpreted as good isotopic representations of the mantle reservoirs involved (Hart et al., 1989).

The lack of systematic suite-wide major and trace element variations leads us to view individual, chronostratigraphically identifiable basalt groups as reflective of magmatic systems or events with similar but unique petrogenetic histories. That the observed between-group variability cannot simply be explained by crystallization dominated differentiation from any of the analyzed materials is well illustrated by the overlapping MgO contents of groups differing in TiO_2 by more than 0.5 wt% (Fig. 6) and in Cr by ~200 ppm (Fig. 7), by the lack of sub-parallel

1 enrichment of the REE from the least differentiated to the most differentiated groups (Fig. 8A &
2 C), and the irregular and crossing trace element patterns (Figure 8 B & D). Clear within-group
3 elemental variations are noted in many of the binary plots of Figures 6 and 7 and in the trace
4 element plots of Figure 8. For example, the flat to decreasing concentrations of Al_2O_3 , Ni and Cr
5 and flat to increasing concentrations of CaO, P_2O_5 , and Sr with decreasing MgO are consistent
6 with at least some of the within-group variation resulting from fractionation of olivine $\pm$
7 plagioclase. Also, olivine- and plagioclase-dominated crystallization can explain the observed
8 major and trace element trends within the AAB group, yet the rather low Cr contents even at
9 high MgO require earlier differentiation from a parental liquid not represented. This interpretation
10 also applies to the GDB, KBB, and main MLB groups. Such a higher pressure, likely lower
11 crustal clinopyroxene-involved crystallization stage is consistent with conclusions drawn from
12 other studies of Ethiopian basalts, including many illustrated for comparison in our figures (e.g.,
13 Pik et al., 1988, Rooney et al., 2014, 2017). Basalts of the KBB and MLB groups exhibit
14 internal variations that likely reflect addition or subtraction of at least olivine, differentiation that
15 may be taking place in a shallow reservoir or during magma ascent or surface flow.

16
17 The distinct trends between Cr and MgO illustrated in Figure 7, one of correlated decreasing Cr
18 and MgO and one of relatively constant Cr with decreasing MgO, emphasize our interpretation
19 that the WORMIL suite was derived from multiple, more primitive (higher Mg#, Ni, Cr) parental
20 magmas, none of which are represented. Further, an important but variable role for clinopyroxene
21 is required, possibly as a prominent component of deep crustal fractionation processes. The
22 clinopyroxene signature may also reflect bulk magma source heterogeneity owing to the

1 presence of mafic (pyroxenitic) lithologies. Such mafic lithologies are documented by xenoliths
2 from the northwestern Ethiopian Plateau (e.g., Beccaluva et al., 2011).

3

4 Open system processes, whether involving heterogeneous bulk crustal components or silicic
5 melts alone or coupled with crystallization, certainly could, in part, contribute to the observed
6 WORMIL trace element pattern (Fig. 8) and isotopic (Fig 9) diversity. Additionally, the trends in
7 Nb/U and Nb/La versus Ce/Pb space (Fig. 10) pointing toward characteristics displayed by
8 representative Low-Ti Ethiopian flood basalts, which previously have been suggested to have
9 incorporated lower $\pm$ upper crustal materials during crystallization (e.g., Pik et al., 1999; Kieffer
10 et al., 2004; Ayalew and Gibson, 2009), indicate that at least some of the WORMIL samples may
11 have experienced similar petrogenetic histories. If such processes are involved, they are only
12 cryptically preserved since no clear modal evidence is observed. The highest $^{87}\text{Sr}/^{86}\text{Sr}$ GDB
13 sample BWK25 and the lowest $^{143}\text{Nd}/^{144}\text{Nd}$ AAB sample BAR62 seem likely candidates for
14 crustal addition, yet key trace element characteristics do not support this (Fig. 10). Moreover, the
15 lack of decoupling between WORMIL basalt major and trace element characteristics (Fig. 6, 7,
16 8), and the lack of any simple relationship between isotope variation and degree of
17 differentiation argues against crustal assimilation as the primary control on the observed isotopic
18 diversity. This suggests that source characteristics likely provide the primary control.

19

20 Prior to looking more closely at the isotopic constraints for variable sources, we first
21 qualitatively assess the likely melting regime involved in WORMIL basalt petrogenesis. Figure
22 11 is a $(\text{Tb}/\text{Yb})_{\text{N}} - (\text{La}/\text{Sm})_{\text{N}}$ plot of the WORMIL basalts and comparative basalts using only those
23 samples with >6 wt% MgO to eliminate at least some of the possible effects of upper-level

1 differentiation. This diagram is useful to qualitatively assess both the relative depth of melting
2 (Tb/Yb variations) and the relative degree of partial melting and/or source enrichment
3 (La/Sm)(e.g. Rooney, 2010). Given the compelling evidence for clinopyroxene either as a deep
4 fractionating phase or residual mantle source phase in WORMIL petrogenesis, it should be noted
5 that this would lead to only minor variations in Tb/Yb (e.g., Rooney, 2010), but could lead to a
6 an increase in La/Sm. This said, representatives from the low-Cr KBB and ABB groups and a
7 low-Cr DDB sample define the low La/Sm and Tb/Yb portion of the WORMIL array, whereas
8 low-Cr MLB and GDB samples plot nearly to the other end of this array. Thus while there is no
9 correlation with our index of clinopyroxene involvement, there is a general correlation between
10 the two trace element ratios suggesting that the covariation may in part be controlled by the
11 degree of source enrichment and/or degree of melting plus slight variations in depth of the
12 melting column, both of which can imply the involvement of variable source lithologies. Most of
13 the WORMIL samples plot within or just to higher Tb/Yb than the space defined by averages of
14 D-MORB, N-MORB, and E-MORB, similar to some of the west-central Afar lavas, and trending
15 toward fields for southern Afar and northern MER basalts and average OIB. Ignoring for a
16 moment the GDB group, there otherwise is a general trend to more shallow melting (lower
17 Tb/Yb) through time. The youngest GDB basalts defy this trend, but it is noted that this set
18 includes samples with the highest La/Yb, anomalous P enrichments, high Ce/Pb and Nb/U,
19 constant $^{143}\text{Nd}/^{144}\text{Nd}$ below the main WORMIL array (Fig. 6, 8, 10), and the sample (BWK-25)
20 with the most radiogenic Sr (Fig. 9). Together these characteristics could point to melting of an
21 enriched mantle reservoir containing apatite and/or assimilation of cumulate materials from prior
22 magmatic fractionation events(e.g., Rooney et al., 2007). While some level of mantle source
23 heterogeneity may be implied from Figure 11, the relationships illustrated suggest that WORMIL

parental liquids were derived from mantle sources that did not retain garnet in the residue and thus likely originate from, and certainly last equilibrated within, the spinel stability field (<80-100 km depth; Ayalew and Wilson, 2009). Using the crust-mantle structure interpretations presented by Bastow et al. (2011), this suggests that WORMIL magma generation is taking place in the upper-most asthenosphere, possibly extending into the lower-most subcontinental lithospheric mantle.

Having argued that a primary control on the WORMIL geochemistry is inherited from the mantle source(s), we have employed strategies similar to those described in Rooney et al. (2012) to explore this notion. We consider the dominant (and minimum number of) isotopic endmembers contributing to WORMIL diversity to be: 1) DM – depleted mantle of the convecting asthenosphere, 2) Afar Plume mantle (AP), and 3) Pan African Lithosphere (PAL)(Fig. 9), and adopt the specific Sr-Nd-Pb isotope characteristics and daughter element concentrations for these endmembers proposed by Schilling et al., (1992) and Rooney et al. (2012). Following the approach of Hanan et al. (1986) we first evaluate the WORMIL isotope data in ternary Pb isotope space and have converted the ternary relative abundances of ^{206}Pb , ^{207}Pb , and ^{208}Pb into x-y coordinates. The resulting diagram is presented in Figure 12 (see caption for details). Figure 12A illustrates the placement of the WORMIL suite and other selected regional suites in the context of the DM, AP, and PAL endmembers. While the other suites shown tend to form diagonal arrays, the WORMIL suite defines a cluster with a couple of outlying points. Since two-component mixing on this diagram will yield a linear relationship between endmembers, it follows that robust linear regressions through the various datasets can identify potential endmember components. Figure 12B plots calculated regressions through basalts erupted from

the following specific volcanic fields or spatially restricted areas: WCA, west-central Afar (Hart et al., 1989); southern Afar eruptive centers (Ayalew et al., 2016); and from axial and marginal northern MER (Rooney et al., 2012; $\text{MgO} > 7 \text{ wt\%}$) fault zones (WFB, Wonji Fault Belt Series; DZ, Debre Zeyit Series; BTJ; Butajira Series. Regressions for the BTJ, DZ and WFB suites yield r^2 values of 0.99, 0.95, and 0.74, respectively. The regressions for the WCA and S. Afar suites are essentially identical and yield r^2 values of 0.74 and 0.96, respectively. All of these regressions project to the proposed composition of the Afar plume endmember (Rooney et al. 2012) and to points along the mixing line between DM and PAL that reflect different ratios of these endmembers. The addition of the WCA and S. Afar suites to the previously defined BTJ, DZ, and WFB trends and their collective projections through the Afar plume component further supports the designation of this endmember and the “ordered, two-stage mixing process” forwarded by Rooney et al. (2012) whereby depleted mantle and lithospheric reservoirs blend to varying degrees, with these heterogeneous hybrids later mixing with Afar plume material. The WORMIL basalt suite does not define a linear relationship even when only those samples with $> 7 \text{ wt\% MgO}$ are considered, although the majority of the samples are bracketed by the pseudo-mixing arrays of the other suites. We take this to reflect that while the three endmember model portrayed broadly applies to the WORMIL basalts, complexities due to heterogeneities in the lithospheric component and its role in their petrogenesis leads to the observed scatter.

To further explore the three endmember mixing model we have incorporated the Sr and Nd systems and employed the algorithms described by Schilling et al. (1992) to calculate the proportion of the three model endmembers (DM, AP, PAL) needed to describe the combined Sr-Nd-Pb isotope signatures of each WORMIL sample. Four separate calculations using the isotope

pairs ^{207}Pb - ^{206}Pb , ^{208}Pb - ^{206}Pb Sr- ^{206}Pb , and Nd- ^{206}Pb were performed per sample and when all are in good agreement, a simple average is taken to represent the best estimate of the endmember contributions (see Supplementary information for details). The calculated suite-wide ranges in the contributions from the three endmembers are: DM 65-73%, AP 16-28%, and PAL 7-12%. These averages are well within the ranges calculated for the other suites described above and illustrated in Figure 12. The modeling suggests that the greatest contributions from the AP component (28%) and least contribution from the PAL component (7%) are in the oldest basalts (MLB). Progressing up section, the GGB and KBB basalts share identical endmember contributions (DM 69%; AP 22%, PAL 8%). The greatest contribution from the PAL is preserved in the youngest DDB (12%) and GDB (10%) basalts. To a first approximation we take these results to indicate that the WORMIL basalt suite is being sourced from the same fundamental isotope reservoirs as young basalts from the present day axial zones of the central to northern MER and the Afar. While adequately modeled as three endmembers, these reservoirs represent convecting depleted mantle $\pm$ heterogeneous constituents of foundered Pan African lithospheric mantle, deeper upwelling material associated with the Afar plume, enriched lithospheric mantle, and lower to upper crustal lithologies.

6.2 Progressive Lithospheric Thinning and Rifting

The contributions from the reservoirs outlined above can be assessed in terms of the degree of lithospheric extension and thinning in Afar. The Afar lithosphere represents a transition from a continental rift to continental break-up (e.g. Makris & Ginzburg, 1987; Bastow & Keir, 2011; Hammond et al., 2011). As continental rifting progresses the lithosphere stretches and thins

1 allowing asthenospheric mantle material to ascend and melt due to decompression. In the early
2 Pliocene the lithosphere in the WORMIL area must have been stretched and thinned enough to
3 allow spinel-dominated shallow depth melting as suggested by the relatively low $(\text{Tb/Yb})_N$ ratios
4 illustrated in Figure 11. The apparent decrease in plume component and increase in lithospheric +
5 depleted mantle components through time within the WORMIL suite supports a shallowing of
6 the melting column and heightened interactions between melts from the depleted mantle and
7 melts from enriched lithosphere as thinning and stretching continues to erode the lithosphere.
8 These conclusions are consistent with the source model postulated for MER and Afar volcanism
9 by Hart et al. (1989) that through time the reservoirs of the continental mantle
10 lithosphere are consumed and replaced by depleted asthenospheric mantle as rifting progresses
11 and new proto-oceanic lithosphere is formed.

12
13 The relationship between Mio-Pliocene extensional deformation and volcanism in the WORMIL/
14 western Afar has not so far been well documented. Audin et al. (2004) suggested linkage
15 between the volcanism of the Dalha Formation and the localization of deformation with the
16 formation of an axial rift in the western Afar. The northwestward propagation of the Aden rift
17 into the western Afar during the Pliocene is now reassessed in the context of the newly
18 constrained ages of the WORMIL basalts and the new insights gained from their combined
19 elemental and isotopic data. The lithosphere in the WORMIL area was thinned/extended,
20 accompanied by the formation of an axial rift and shallow decompression melting that led to the
21 eruption of the WORMIL basalts between ~ 4 Ma to ≤ 3 Ma. The significant lithospheric thinning
22 and rifting and subsequent emplacement of the extensive Guda basalt unit, previously presumed
23 to be in 8-6 Ma, in fact took place at a much later time. In this regard, Wolfenden et al. (2004,

2005) suggested that changes in plate configuration within the Afar depression caused by the along-axis propagation of the Gulf of Aden and Red Sea spreading centers had induced lithospheric extension by 7-3 Ma. They also described that extensional strain migrated eastward from the border faults to smaller faults within the same period. These assertions are consistent with the timing of the lithospheric thinning and localization of extension in the western Afar margin and ensuing WORMIL basalt emplacement in the middle to late Pliocene.

7. Conclusions

Six basalt groups are distinguished and constrained in age to between ~3.8 and ~3.0 Ma in the Woranso-Mille area of Afar. The basalt groups show variable concentrations of incompatible trace elements and minor oxides but no consistent, simple suite-wide major or trace element trend(s) exists. Rather, the groups generally are chemically distinct with some displaying within-group linear trends derived by olivine±plagioclase addition or removal in shallow chambers or at the surface. Most of the basalt groups require a deeper phase of clinopyroxene-involved crystallization that likely was accompanied by limited assimilation of lower crustal lithologies. Moreover, multiple parental magmas are inferred.

Trace element patterns and ratios in combination with isotope data emphasize that the WORMIL basalts overlap in characteristics with other basalt suites from the Afar, MER, and adjacent plateau regions. While crustal contributions to this magmatism are present, they are in most cases seen to overprint the primary isotopic signatures imparted from the mantle. In this context the combined WORMIL Sr-Nd-Pb isotope data are most consistent with the involvement of three

1 dominantmantle reservoirs - the Afar plume, depleted mantle (convecting asthenosphere), and
2 enriched lithospheric mantle - such that between-group basalt isotopic variations are attributed
3 primarily to mixing of the Afar plume source materials with variable mixtures of the depleted
4 asthenospheric and enriched lithospheric source materials. We further suggest that the basalts
5 were generated from spinel-dominated shallow melting, requiring significantly thinned Pliocene
6 lithosphere in western Afar.

7
8 Thus, propagation of the Aden rift into western Afar is responsible for middle to late Pliocene
9 lithospheric thinning and localization of extension in the western Afar margin and subsequent
10 WORMIL basalt emplacement. The new data reinforce previous interpretations that progressive
11 rifting and transformation of the continental lithosphere to oceanic lithosphere creates
12 opportunity for increasing asthenospheric inputs through time as the continental lithosphere is
13 thinned.

14
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Figure Captions

Figure 1. Regional Geology of the Afar Depression (modified after Varet, 1975; Tefera et al, 1996; Beyene and Abdelsalam, 2005). MH-TD-GD= Manda Hararo-Tendaho-Gobaad rift, MI-AS-GH= Manda Inakir-Asal-Ghoubett rift, SA-G= Sullu Adu-Guda area, DBV= Dabbahu volcano; red lines are fracture lineaments.

Figure 2. Geologic map of northern Woranso-Mille area, west-central Afar, Ethiopia. DDB/KBB= Daba Dora basalt/Kerare-Burtele basalt, AAB/GGB= Am Ado-Aralee Issie basalt/Gugubsi basalt. Locality names: AMA= Am-Ado, ARI= Aralee Issie, BDB= Bodole Bora, BRT= Burtele, GUB= Gugubsi, KER= Kerare, KSD= Korsi Dora, LHG= Lahaysule Gera, MKM= Makah Mera, MSD= Mesgid Dora.

Figure 3. Lithostratigraphic succession for northern WORMIL area with mapped sedimentary and volcanic units. Black horizontal bars show chronostratigraphic position of named tuffs and the constraints they place on the relative age, not duration, of basalts. AAT= Am-Ado Tuff; AKT= Araskimiro Tuff; BT= Burtele Tuff; DDT= Daba Dora Tuff; KT= Kilaytoli Tuff; MDT= Mesgid Dora Tuff; MLT= Mille Tuff; SHT= Sidi Hakoma Tuff; WT= Waki Tuff. The $^{40}\text{Ar}/^{39}\text{Ar}$ ages are calculated using a reference age for the Fish Canyon Tuff of 28.198 ± 0.044 Ma (as reported in Kuiper et al., 2008). The age for SHT and WT are $\pm 1\sigma$ errors, the other ages are $\pm 2\sigma$.

Figure 4. Exposed sections of basalt-tuff units in various localities at northern WORMIL: (A) Gugubsi, (B) NE of Am-Ado, (C) Kerare, (D) Lahaysule Gera. MLT= Mille Tuff, SHT= Sidi

1 Hakoma Tuff.

2
3 Figure 5.(A) TAS (Total Alkali-Silica) classification diagram (Le Bas et al., 1986) for WORMIL
4 basalts. Data plotted are anhydrous normalized using total Fe as FeO. The divide between
5 alkaline and sub-alkaline rocks (thick dashed line) is from Irvine and Baragar
6 (1971). Abbreviations for rock names are P, microbasalt; B/T, basanite/tephrite; Tb, trachybasalt;
7 Bta, basaltic trachyandesite; Ta, trachyandesite; A, andesite. Fields for Plio-Pleistocene mafic
8 lavas of the west-central Afar (WCA; Hart et al., 1989) and for Oligocene to late Miocene mafic
9 lavas and dikes from the northwestern plateau region most proximal to the study area (NWP; Pik
10 et al., 1998; Kieffer et al., 2004; Rooney et al., 2013) are shown for comparison. (B) Nb/Y versus
11 Ti/Y plot adapted from Pik et al. (1998). Fields for Pliocene (Akaki Basalt) to Quaternary mafic
12 lavas from the southern Afar and northern Main Ethiopian Rift (Rooney et al., 2007; Rooney et
13 al., 2014; Ayalew et al., 2016) and from the same northwestern plateau suite cited above, but
14 split into two subgroups, NWP-a (< 10 Ma dikes and Low-Ti basalts) and NWP-b (High-Ti dikes
15 and basalts), are shown for comparison.

16
17 Figure 6. Variation diagrams of major oxides with MgO for WORMIL basalts.

18
19 Figure 7. Variation diagrams of selected trace elements and the Ce/Pb ratio with MgO for
20 WORMIL basalts. Values with errors in the Ce/Pb plot reflect cited ranges for non-EM oceanic
21 basalts (25 ± 5) and convecting mantle-derived basalts (29 ± 9) (Hofmann, 2003, 2006).

Figure 8. Chondrite normalized REE (A& C) and primitive mantle normalized incompatible element (B& D) plots for WORMIL basalts. Normalization values are from Sun and McDonough (1989). The WORMIL suite and comparative compositions are split for clarity. Comparison suites as in Figure 5.

Figure 9. Sr-Nd-Pb isotope data. (A) $^{143}\text{Nd}/^{144}\text{Nd}$ versus $^{87}\text{Sr}/^{86}\text{Sr}$, (B) $^{207}\text{Pb}/^{204}\text{Pb}$ versus $^{206}\text{Pb}/^{204}\text{Pb}$, (C) $^{87}\text{Sr}/^{86}\text{Sr}$ versus $^{206}\text{Pb}/^{204}\text{Pb}$. Error bars for isotope ratios are within the symbol size. Fields for the isotopic compositions of comparison suites as described for Figure 5 with the addition of the Gulf of Aden and Tadjoura (Schilling et al., 1992; Deniel et al., 1994) and representative lower and upper crustal lithologies (Ayalew and Yirgu, 2003; Lucassen et al., 2011; Hart et al., 1989 and references therein). Additional components plotted as follows; HIMU, EMI, and EMII mantle endmembers (Hofmann, 2003; Willbold and Stracke, 2006) and proposed Ethiopian isotope endmembers DM (depleted mantle), Afar Plume, and Pan African (PA) Lithosphere from Schilling et al. (1992) and Rooney et al. (2012, 2013).

Figure 10. Trace element ratio and isotope variations with Ce/Pb ratio. (A) Nb/U, (B) Nb/La, (C) $^{143}\text{Nd}/^{144}\text{Nd}$. Fields for magmatic suites and crustal lithologies as described for figures 1 and 9. Yellow box with C represents the compositional space occupied by averages for lower, middle and upper continental crust (Rudnick and Gao, 2014). The shaded fields labeled RCL include representative crustal lithologies described for Figure 9. The gray shaded field labeled OB is the compositional space occupied by averages for D-MORB, N-MORB, E-MORB and OIB from Sun and McDonough (1989) and Gale et al. (2013).

Figure 11. $(\text{Tb/Yb})_N$ versus $(\text{La/Sm})_N$ ratios for WORMIL and selected magmatic suite samples with $\text{MgO} > 6 \text{ wt\%}$ and oceanic basalt types previously described. Chondrite normalization factors from Sun and McDonough (1989). Dashed line represents approximate division between melts derived from mantle lithologies dominated by garnet as the aluminous phase versus those dominated by spinel.

Figure 12. Pb isotope relationships and primary reservoir contributions. Plots are binary representations of ternary (triangular) Pb isotope space (after Hanan et al., 1986) with apices of $^{206}\text{Pb}/^{204}\text{Pb}$ (left -A), $^{207}\text{Pb}/^{204}\text{Pb}$ (top-B), and $^{208}\text{Pb}/^{204}\text{Pb}$ (right-C). In the binary plots, the y-axis units are the proportion of ^{207}Pb (B) and the x-axis units are a function of the ^{206}Pb - ^{208}Pb abundances (D) calculated as $D = C + B \div 2$. For example, the point labeled DM with ternary Pb coordinates (relative abundances) of 0.252 ^{206}Pb , 0.221 ^{207}Pb , and 0.527 ^{208}Pb plots as $x=0.638$ and $y=0.221$. (A) WORMIL ternary Pb isotope characteristics compared to previously defined magmatic suites, crustal lithologies, and mantle/reservoir endmembers. (B) WORMIL suite compared to pseudo-binary mixing arrays calculated as linear regressions following the approach described by Rooney et al. (2012) for basalts from the northern MER (Butajira, BTJ; Debre Zeyit, DZ; Wonji Fault Belt, WFB; Rooney et al., 2007, 2012), the west-central Afar (WCA; Hart et al., 1989) and the southern Afar (Ayalew et al., 2016).

Table 1. Representative northern WORMIL basalt major and trace element geochemistry.

Table 2. Sr-Nd-Pb isotope ratios for northern WORMIL basalts.

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