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1	Os isotopic constraints on crustal contamination in Auckland
2	Volcanic Field basalts, New Zealand
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23 24 25 26 27 28	 Evidence of crustal contamination observed in Auckland Volcanic Field basalts. Highly radiogenic Os isotope ratios coupled with very low Os concentrations indicate a crustal metasedimentary contaminant. Slightly elevated Os isotope ratios with high Os concentrations indicate contamination from sulphide bearing olivines.

29 Abstract

30 The Auckland Volcanic Field (AVF) represents the youngest and northernmost of three 31 subjacent Quaternary intraplate basaltic volcanic fields in the North Island, New Zealand. Previous studies on AVF eruptive products suggested that their major- and trace- element, and 32 33 Sr-, Nd- and Pb-isotopic signatures primarily reflect their derivation from the underlying asthenospheric and lithospheric mantle. All AVF lavas however ascend through a ca. 20-30 km 34 35 thick continental crust, and some do carry crustal xenoliths, posing the question whether or not 36 crustal contamination plays a role in their formation. Here we present new Os and Pb isotopic 37 data, and Os and Re concentrations for 15 rock samples from 7 AVF volcanic centres to 38 investigate mantle and crustal petrogenetic processes. The samples include the most primitive lavas from the field (Mg# 59-69) and span a range of eruption sizes, ages, locations, and 39 geochemical signatures. The data show a large range in Os concentrations (6-579 ppt) and 40 41 ¹⁸⁷Os/¹⁸⁸Os isotope ratios from mantle-like (0.123) to highly radiogenic (0.547). Highly 42 radiogenic Os signatures together with relatively low Os contents in most samples suggest that 43 ascending melts experienced contamination primarily from metasedimentary crustal rocks with high ¹⁸⁷Os/¹⁸⁸Os ratios (e.g., greywacke). We further demonstrate that <1% metasedimentary 44 45 crustal input into the ascending melt can produce the radiogenic Os isotope signatures observed 46 in the AVF data. This low level of crustal contamination has no measurable effect on the 47 corresponding trace element ratios and Sr-Nd-Pb isotopic compositions. In addition, high Os 48 contents (195-578 ppt) at slightly elevated but mantle-like Os isotopic compositions (187Os/188Os = 0.1374-0.1377) in some samples suggest accumulation of xenocrystic olivine-49 50 hosted mantle sulphides from the Permian-Triassic ultramafic Dun Mountain Ophiolite Belt, 51 which traverses the crust beneath the Auckland Volcanic Field. We therefore infer that the AVF 52 Os isotopic compositions and Os contents reflect contamination from varying proportions of heterogeneous crustal components, composed of Waipapa and Murihiku terrane 53 metasediments, and ultramafic rocks of the Dun Mountain Ophiolite Belt. This demonstrates, 54 55 contrary to previous models that primitive lavas from the Auckland Volcanic Field do show 56 evidence for variable interaction with the crust.

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62 **1. Introduction**

63 Monogenetic basaltic volcanic fields are the surface expression of small-scale magmatic 64 systems and are found in a number of different tectonic settings, including extensional systems 65 (e.g., Cascades, USA; Borg et al., 2000), subduction zones (e.g., Wudalianchi, China; Hwang et al., 66 2005), or intraplate settings relating to lithospheric rifting (e.g., Panter et al., 2006) and delamination of the lower lithosphere (e.g., Hoernle et al., 2006). The formation of individual 67 monogenetic volcanic centres has mainly been attributed to isolated, often small-volume, 68 69 batches of magma (<0.1 km³) (e.g., Connor and Conway, 2000; Németh, 2010; Kereszturi et al., 70 2013), which erupt quickly (e.g., Németh, 2010), creating volcanic fields which can encompass 71 tens to hundreds of individual centres (e.g., Condit and Connor, 1996; Conway et al., 1998; 72 Connor and Conway, 2000; Valentine et al., 2005). The fields show a range of surface 73 expressions, dependent on the eruption style and magma-water interaction, including tuff 74 rings, maars, scoria cones, and lava flows (e.g., Allen and Smith, 1994; Németh, 2010; Kereszturi 75 et al., 2014). A link between eruptive volumes of monogenetic basaltic volcanoes, and 76 mineralogical and geochemical composition of the underlying mantle has recently been 77 proposed (McGee et al., 2015) highlighting the importance of understanding the characteristics 78 of different sources contributing to magmatism beneath volcanic fields.

79 Petrological and geochemical studies have shown that most eruptive centres in 80 monogenetic fields are composed of silica- undersaturated basanites, nephelenites and alkaline 81 basalts that are akin to Ocean Island Basalts (OIBs) (e.g., Huang et al., 1997; Cook et al., 2005; 82 Valentine and Gregg, 2008). Nevertheless, eruption products can show considerable 83 geochemical variations both within volcanic fields (e.g., Valentine and Hirano, 2010; Timm et al., 2010; McGee et al., 2013) and within individual centres, the origins of which remain 84 controversial (e.g., Bradshaw and Smith, 1994; Valentine and Gregg, 2008; Needham et al., 85 2011; Brenna et al., 2010, 2011; McGee et al., 2012). Several studies have attributed these 86 87 variations in geochemical and isotopic signatures, both for individual eruptions and field-wide 88 scales, to heterogeneities in the underlying mantle (e.g., Huang et al., 1997; McBride et al., 2001; 89 Cook et al., 2005; McGee et al., 2013), magma modification by lithospheric contamination 90 (mantle and crust) or fractional crystallisation (e.g., Lassiter and Luhr, 2001; Alves et al., 2002; 91 Chesley et al., 2002; Jamais et al., 2008; Timm et al., 2009). Although crustal assimilation and 92 contamination may be facilitated by storage or ponding of magma within the crust (e.g., 93 Bohrson et al., 1997), eruptive products in monogenetic volcanic fields are generally mafic (>8 94 wt.% MgO) thus arguing for limited fractional crystallization and against prolonged crustal 95 magma storage. Therefore, the variations in major, and trace element signatures, and Sr-, Nd-, 96 and Pb-isotopic compositions of rocks from monogenetic volcanic fields are proposed to primarily reflect the composition of the underlying mantle (e.g., Valentine and Perry, 2007;
McGee et al., 2012, 2013, 2015). Nevertheless, even mafic magmas in monogenetic fields are not
primary melts and therefore are likely to have undergone some interaction with the crust
through which they ascend, potentially affecting the physical and chemical properties of the
melts. Thus, it is a prerequisite to distinguish between the geochemical signatures of mantle
heterogeneity versus crustal contamination (e.g., Blondes et al., 2008; Jung et al., 2011).

103 To understand the role of the crust and mantle petrogenesis beneath an archetypal continental monogenetic volcanic field we use the ¹⁸⁷Re-¹⁸⁷Os decay system. This system is 104 105 highly sensitive to crustal contamination due to the large difference in incompatibility between 106 parent (Re) and daughter (Os) elements during partial melting of mantle lithologies. During 107 partial melting Os behaves compatibly in mantle sulphides and thus dominantly remains in the 108 mantle, whereas Re is moderately incompatible and preferentially enters the melt. This 109 contrast in behaviour of Re and Os therefore result in extreme fractionation (e.g., high Re/Os in 110 melt and crust, and low Re/Os in the mantle) which, with time, leads to an overall significant 111 contrast between the ¹⁸⁷Os/¹⁸⁸Os ratios of continental crust (up to ¹⁸⁷Os/¹⁸⁸Os = 5.0; McBride et al., 2001) versus typical mantle sources (ca. 0.12; Meisel et al., 1996). ¹⁸⁷Os/¹⁸⁸Os isotopic values 112 113 in the eruptive products of volcanic systems therefore provide a unique tool to decipher the influences of crustal contamination (e.g., Central European Volcanic Province: Jung et al., 2011; 114 115 Newer Volcanics Province: McBride et al., 2001) and mantle source heterogeneity.

Here we present new Os and Re concentrations, as well as Os and Pb isotope data for 15 mafic samples from the Auckland Volcanic Field, New Zealand. These new data are aimed at identifying the relative importance of mantle heterogeneity, including crustal recycling, and crustal contamination into ascending melts, in order to give new insights into dynamics of melt generation and ascent beneath the city of Auckland.

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122 2. The Auckland Volcanic Field

123 The monogenetic basaltic Auckland Volcanic Field (AVF) is located 400 km west of the 124 currently active arc, the Hikurangi Margin (e.g., Seebeck et al., 2014)(**Fig. 1.A**). The AVF is the 125 northernmost of three intraplate monogenetic volcanic fields that become progressively 126 younger towards the north (Ngatutura and Okete ca. 2.7-1.5 Ma; Briggs et al., 1994, and South 127 Auckland Volcanic Field (SAVF) ca. 1.59-0.51 Ma; Cook et al., 2005) (**Fig. 1.A**).

128 The AVF consists of 53 individual centres (**Fig. 1.B**; Hayward et al., 2011) that 129 collectively cover ca. 360 km² (**Fig. 1**; Allen and Smith, 1994; Kermode, 1992). The centres 130 include one or more explosion craters, tuff rings (some now occupied by lakes), scoria cones 131 and lava flows. The individual centres are interpreted to have formed through single eruptions of small magma batches (e.g., Allen and Smith, 1994), except for Rangitoto, the youngest and 132 133 largest volcano where two eruption episodes have been identified (Rangitoto 1 [553±7 cal. yrs. 134 BP] and Rangitoto 2 [504±5 cal. yrs. BP]; Needham et al., 2011). The total volume of the AVF 135 field is estimated at 1.7 km3 dense rock equivalent (DREtot), ca. 41% (0.7 km3) of which is 136 represented by Rangitoto (Kereszturi et al., 2013). Basaltic volcanism commenced at ca. 190 ka 137 (Lindsay et al., 2011) and shows some distinct changes in eruption frequency over time (e.g., Molloy et al., 2009; Hopkins et al., 2015), however the ages of many of the individual eruptive 138 139 centres are currently poorly constrained (Lindsay et al., 2011).

140 The crust underlying the AVF is 20-30 km thick, and composed of Waipapa and the 141 Murihiku terranes (Fig. 1.C) (e.g., Kermode 1992; Eccles et al., 2005; Horspool et al., 2006; 142 Mortimer et al., 2014), which are overlain by up to 1-2 km of Miocene Waitemata terrane 143 sediments. The western Waipapa and eastern Murihiku terranes mainly consist of late Triassic 144 to late Jurassic low-grade meta-sediments, separated by the Dun Mountain terrane. This terrane 145 represents a Permian to late Cretaceous oceanic arc ophiolite obduction event at the eastern Gondwana margin (e.g., Kimbrough et al., 1992) and mainly consists of ultramafic rocks, 146 including dunites, lherzolites, harzburgites and werhlites (cf. McCoy-West et al., 2013). This 147 148 terrane is reflected in a distinct positive magnetic anomaly known as the Junction Magnetic Anomaly (JMA), which can be traced continuously through the North and South islands of New 149 Zealand (Hatherton and Sibson, 1970). The JMA traverses the Auckland area as a narrow (ca. 2-150 151 5 km), linear series of positive magnetic anomalies interpreted to be eastward-dipping 152 serpentinised shear zones, extending throughout the 20-30 km thick crust beneath the AVF 153 (Eccles et al., 2005). Direct evidence of the Dun Mountain Ophiolite Belt (DMOB) beneath the 154 AVF occurs in the form of serpentinite xenoliths within volcanic rocks in tuff rings from Pupuke, St. Heliers and Taylors Hill volcanoes (e.g., Searle, 1959; Bryner, 1991; Jones, 2007; Spörli et al., 155 156 2015). The basement rocks that make up the region of the study area have been extensively 157 investigated (e.g., Bryner, 1991; Kermode, 1992; Eccles et al., 2005; McCoy-West et al., 2013), allowing potential sources of crustal contamination to be well characterised. 158

The Auckland Volcanic Field has been the focus of previous geochemical and petrological studies (Huang et al., 1997; Smith et al., 2008; Needham et al., 2011; McGee et al., 2011, 2012, 2013, 2015). Low degrees of partial melting (≤6%), a limited proportion of fractional crystallisation, in addition to a limited range of Sr-, Nd-, and Pb-isotopic compositions of the AVF lavas were interpreted to reflect the mantle origin of the AVF lavas. Huang et al. (1997) furthermore attributed the more radiogenic ²⁰⁶Pb/²⁰⁴Pb isotopic compositions (>19.2) of 165 the AVF rocks to the presence of 'young' HIMU signature formed in ≤ 0.2 Ga in the underlying 166 mantle (cf. Thirlwall. 1997). Alternatively, McGee et al. (2013) suggested that AVF lavas with more radiogenic ${}^{206}Pb/{}^{204}Pb$ isotopic compositions, coupled with low SiO₂ (<48 wt.%) and 167 Nb/U, but high CaO/Al₂O₃, Na₂O and K₂O, and elevation Ce/Pb, Nb/Ce and U/Pb represent melts 168 169 derived from carbonated garnet peridotite domains hosted in a depleted mantle-like peridotitic 170 asthenosphere. The more siliceous AVF alkali basalts (SiO₂ \ge 48 wt.%) were in contrast 171 attributed to interaction of ascending lithospheric melts with (or direct melting of) an EMII-type lithospheric mantle, previously metasomatised by subduction-related fluids. These 172 173 interpretations of the AVF major and trace element, and Sr-, Nd-, and Pb- isotopic systematics 174 rely on the assumption that the AVF eruption products are directly representative of their 175 mantle sources, as proposed by McGee et al. (2013), and are not affected by contamination. 176 However, a number of studies have noted xenolithic materials (e.g., crustal schistose and non-177 schistose fragments, meta-igneous rocks, and abundant large olivines) within some AVF centres, for example at St Heliers, Taylors Hill, and Mangere Mt (Spörli et al., 2015), providing direct 178 179 evidence for the interaction of ascending magmas with the underlying crustal and mantle 180 lithologies (e.g., Bryner, 1991). Therefore, traditionally used major and trace element contents 181 and ratios, and Sr-, Nd-, and Pb-isotopes may not be sensitive enough to distinguish between 182 crustal contamination and mantle heterogeneity. We address this problem here using the more 183 sensitive Os isotope system.

184

185 3. Methods

186 3.1. Sample selection

187 Specific samples were chosen for this study in order to complement existing data and to 188 cover not only a range of geochemical compositions of the field (c.f. McGee et al., 2011, 2012, 189 2013, 2015) but also a range in ages (Needham et al., 2011; Lindsay et al., 2011), locations, 190 eruptive volumes and types (Kereszturi et al., 2013, 2014). Based on previous analyses, fifteen 191 primitive samples were chosen (Mg# =59-69) from Rangitoto, Mt Wellington, Purchas Hill, 192 Three Kings, Wiri, Puketutu, and Pupuke. New Pb isotope data are added by this study for 7 193 samples, and new major and trace element data are added for 5 samples (see Table 1 for 194 classification). For all fifteen samples new Re and Os concentrations, and Os isotope ratios are 195 determined. Generally the selected samples contain minor olivine phenocrysts ($\leq 3 \text{ mm across}$) 196 in a plagioclase, pyroxene, and olivine-bearing groundmass, except for samples from Pupuke, 197 which contain abundant large olivines ≥ 5 mm across.

198

199 3.2. Analytical techniques

200 Sample preparation for chemical and isotopic analyses were conducted at Victoria 201 University of Wellington, New Zealand (VUW). Samples were chipped using a Rocklabs Boyd 202 crusher to <15 mm, then reduced to powder in an agate ring mill. Major elements were analysed 203 by XRF analysis at the Open University, Milton Keynes, United Kingdom (UK), on an ARL® 8420+ 204 dual goniometer spectrometer. Powdered samples were fused with lithium metaborate and 205 analysed following methods of Ramsey et al. (1995). Whin Sill dolerite was run as an internal standard with associated accuracy of <1% except for Na₂O (2.37%) and P₂O₅ (1.59%) and 206 207 precision of <2%. For trace element concentrations 50 mg of sample powder was digested in 208 hot concentrated HF + HNO3 for 4 days, then dried to incipient dryness and taken up in 209 concentrated HCl. Following this, samples were converted back into HNO₃ and left for 3 days in 210 hot 1M HNO₃ to form the final analytical solution. Centrifuged sample dilutions were measured 211 on an Agilent 7500CS ICP-MS at Victoria University, Wellington, using BHVO-2 as a primary 212 standard, and BRCR-2 as a secondary internal standard. All results and standard values are 213 reported in the supplementary material (SM) (SM.1 and SM.2). Precision on BCR-2 (n=15 from 214 five digestions) was <6.5% 2sd% except for Nb, Cs, and Ba (≤8 %) and Ta, Pb, and Nb (≤20.5 %) 215 and values were all within <6 % of the reference value, except for Cu, Cs, and Ta. All standard 216 values are outlined in the supplementary material.

217 Pb isotope samples were prepared and purified in an ultra-clean chemical separation 218 laboratory at VUW. Powdered sample was leached in ultrapure 6M HCl for 1 hour at 120°C, 219 rinsed with MilliQ water, and digested in ultrapure conc. HNO3 + ultrapure conc. HF, then turned 220 into solution with 0.8M HBr. The solution was centrifuged, loaded onto 5 mm columns equipped 221 with AG1-X8 resin, and Pb was extracted in a double-pass using 6M HCl. Pb isotopic 222 compositions were analysed using a Neptune MC-ICP-MS at Durham University, UK, results and 223 errors are reported in Table 1. International standard NBS-981 was used to monitor machine 224 drift, with internal precisions (2SE) of ²⁰⁶Pb/²⁰⁴Pb <±0.0012, ²⁰⁷Pb/²⁰⁴Pb <±0.0013, ²⁰⁸Pb/²⁰⁴Pb 225 <±0.0044. All data are normalised to NBS-981 standard values reported by Baker et al. (2004) (206Pb/204Pb =16.9416, 207Pb/204Pb = 15.4998, 208Pb/204Pb = 36.7249), all standard 226 227 measurements can be found in supplementary material (SM2.3).

Os isotope compositions and Re and Os contents were determined at Geotop, Université du Québec à Montréal, Canada, following the method of Meisel et al. (2003). For these analyses 0.8 g aliquots of whole rock powder were spiked with a known enriched tracer solution of ¹⁹⁰Os/¹⁸⁵Re, and digested in Teflon-sealed quartz tubes with 3 ml 6M HCl and 3 ml conc. HNO₃ at 300 °C and 100 bars in a high-pressure asher unit (HPA-S, Anton-Parr). Following this, Os was 233 extracted using the Paris Br₂ technique (Birck et al., 1997). 2 ml of chilled Br₂ was added to the 234 digested sample and left on a hot plate at 90 °C for 2 hours. This scavenges the already oxidised Os (OsO4) from the aqueous solution into the liquid Br2, leaving Re (and PGEs) within the 235 aqueous solution. 20 drops of HBr was added to the isolated liquid Br2 (including Os) to reduce 236 Os from volatile Os⁸⁺ (OsO₄) to non-volatile Os⁴⁺ (OsBr₆²⁻), and then evaporated down. A final 237 238 step of micro-distillation using Cr^{VI} containing H₂SO₄ was used to purify the Os, and make sure it 239 was quantitatively separated from isobaric Re (after Birck et al., 1997). Samples were then measured using a Triton TIMS in negative-ion mode for Os (Creaser et al., 1991), and sector 240 241 field (SF)-ICP-MS for Re. Os and Re concentrations were calculated by isotope dilution from 242 known spike solution values. Os blanks for total procedure are 0.3 pg, and 7 pg for Re, these 243 value are subtracted from the sample totals in data processing along with oxygen interface and 244 sample-spike unmixing corrections. Results and errors for Os and Re analysis are reported in 245 Table 1. The widely used standard Durham Romil Osmium solution (DROsS) was used with an internal precision (2SE) for ¹⁸⁷Os/¹⁸⁸Os of average measurements of <±0.00012 (Luguet et al., 246 247 2008; Nowell et al., 2008, see supplementary details for measurements (SM2.4)), and for Re 248 standard NIST SRM 3143 was used, with internal precision (2SE) for ¹⁸⁷Re/¹⁸⁵Re of average 249 measurements of $<\pm 0.05$ (see SM.2.4). Duplicate analyses were undertaken in two ways, on 250 aliquots of one sample solution post-digestion to test consistency of digestion techniques, and of 251 sample powders to check the internal consistency of samples.

252

253 4. Results

254 4.1. Major and trace elements

255 Volcanic rocks from the AVF range in composition from sub-alkaline silica undersaturated basanites to alkali basalts (e.g., SiO₂ = 39.8-48.8 wt.%, Fig. 2.) following the rock 256 257 classification of LeMaitre (2002). In general, the data for the AVF samples form a broad negative 258 trend with MgO vs. SiO₂ and Al₂O₃ and positive trends with CaO Fe₂O₃^t, TiO₂, and P₂O₅ indicative 259 of olivine and pyroxene fractionation (Fig. 2.). The exceptions are, two samples from Rangitoto, 260 which have lower Fe_2O_3t , TiO_2 , CaO and P_2O_5 values and higher SiO_2 and Al_2O_3 values at a given content of MgO, and two samples from Pupuke, which have lower values for CaO, TiO2 and 261 262 $Fe_2O_3^t$, but higher values of MgO than the other AVF rocks.

All AVF samples used in this study show primitive mantle-normalised trace element distributions broadly similar to ocean island basalt (OIB) (normalisation values after McDonough and Sun, 1995), with typical positive Nb-Ta, and negative K anomalies (**Fig. 3.**). As

previously identified by McGee et al. (2013), AVF samples with low SiO_2 contents have higher 266 267 Nb and Ta relative to the large ion lithophile elements (LILE; Rb, Ba, K) (e.g., Nb/Ba \geq 0.2), and higher Light Rare Earth Element (LREE; La, Gd, Nb) concentrations. Furthermore, these samples 268 269 (those with low SiO₂ content) have high LREE relative to their heavy rare earth elements (HREE; Yb), resulting in high LREE/HREE ratios (e.g., $(La/Yb)_N > 20)$, and elevated ratios of highly to 270 271 moderately incompatible elements, such as Th/Yb \geq 2. In comparison to the low SiO₂, two of the 272 Rangitoto samples with high SiO₂ values have lower Nb and Ta concentrations relative to LILE (e.g., Nb/Ba \leq 0.2) and lower LREE but similar to slightly higher HREE (e.g., (La/Yb)_N <20). 273 These high SiO₂ Rangitoto samples also have higher Th relative to HREE (e.g., Th/Yb <1), lower 274 275 Ce/Pb (<20), yet higher K/Nb (>300) and Zr/Nb (>7). These Rangitoto samples also show a 276 positive Sr-anomaly (Fig. 3), which is less prominent in all other samples; conversely all other 277 samples show a prominent negative K-anomaly (Fig. 3), which is not observed within the 278 Rangitoto samples.

279 These findings are consistent with the previous work of McGee et al. (2013, 2015), and 280 are also comparable to samples from the South Auckland Volcanic Field (SAVF; Cook et al., 281 2005). Cook et al. (2005) noted higher Nb, (La/Yb)_N and Th/HREE values for samples with low-282 SiO_2 (their Group B) and low Nb, $(La/Yb)_N$ and Th/HREE for samples with high-SiO₂ (their Group A) and contrasting incompatible trace element ratios (e.g., K/Nb, Zr/Nb and Ce/Pb). 283 284 These observations are also consistent with those of Hoernle et al. (2006) and Timm et al. 285 (2009, 2010) who divided their data from New Zealand intraplate volcanic centres and fields 286 into 'low-SiO₂' and 'high-SiO₂' groups, showing similar major and trace element geochemical 287 compositions to the AVF low- and high-SiO₂ samples. For the purpose of this study we have 288 chosen not to group our data due to the limited number of samples, but highlight the similarity 289 in geochemical signatures for the AVF samples.

290

291 4.2. Pb isotopes (combined with published Sr-, Nd-, and Pb-isotopes)

292 Pb isotopic data for the AVF rocks are reported in Table 1. Seven new Pb isotope 293 determinations have been generated to supplement the data of McGee et al. (2013). In general the AVF Pb isotopic compositions show limited variation, with $^{206}Pb/^{204}Pb$ = 19.064-19.383, 294 ²⁰⁷Pb/²⁰⁴Pb = 15.576-15.615, and ²⁰⁸Pb/²⁰⁴Pb = 38.485-38.931. On Pb isotope variation 295 296 diagrams the AVF data plot between the mid ocean ridge basalt (MORB), HIMU (high μ = high time integrated U/Pb) and enriched mantle (EM I or II) end-members (following the 297 298 classification of Zindler and Hart, 1986; Fig. 4. A&B.). Purchas Hill sample AU44711 shows the highest 206Pb/204Pb (19.383) and high 208Pb/204Pb (38.929), but 207Pb/204Pb similar to rocks 299

300 from other AVF centres (15.613). Conversely, the two Rangitoto samples show the lowest $^{206}Pb/^{204}Pb$ (19.064), low $^{208}Pb/^{204}Pb$ (38.485), and some of the highest $^{207}Pb/^{204}Pb$ values 301 302 (15.615) of all AVF volcanic rocks (Fig. 4.). Most other AVF samples plot between the Purchas 303 Hill and Rangitoto samples, with the possible exception of the samples from Wiri Mt, which have the lowest 207Pb/204Pb (15.576-15.579) at intermediate 206Pb/204Pb (19.219-19.225) and 304 305 ²⁰⁸Pb/²⁰⁴Pb (38.791-38.795). These Pb isotope compositions observed in AVF rocks generally overlap with those from the SAVF (²⁰⁶Pb/²⁰⁴Pb = 18.959-19.332; ²⁰⁷Pb/²⁰⁴Pb = 15.579-15.624, 306 307 and ²⁰⁸Pb/²⁰⁴Pb = 38.752-38.953: data from Cook et al. (2005), values are re-normalised to 308 standard SRM-981 values reported in Baker et al. (2004), to maintain consistency with values 309 from McGee et al. (2013), and this study; Fig. 4.).

310 Previously published Sr and Nd isotope data for the same samples analysed in this study show a restricted range from 87 Sr/ 86 Sr = 0.702710 to 0.703125 and from 143 Nd/ 144 Nd = 311 312 0.512939 to 0.512956 (McGee et al., 2013; Fig 4. C&D., reported in supplementary material 313 SM.1.). Although no obvious trend has been identified between Sr and Nd isotopic compositions, 314 some of the AVF data (McGee et al., 2013) show similar trends to those observed by Cook et al. (2005) in the SAVF samples. Some samples have distinctly higher ⁸⁷Sr/⁸⁶Sr (ca. 0.7032) than the 315 samples from the other centres at similar ¹⁴³Nd/¹⁴⁴Nd values (ca. 0.5129) (Fig. 4.C&D.). 316 Similarly there is no clear correlation observed between ²⁰⁶Pb/²⁰⁴Pb and ¹⁴³Nd/¹⁴⁴Nd, although 317 318 ²⁰⁶Pb/²⁰⁴Pb and ⁸⁷Sr/⁸⁶Sr show a poorly defined negative trend (Cook et al., 2005; McGee et al., 319 2013; Fig. 4.C.). Similar to most AVF samples, the SAVF 'Group B' type (Cook et al., 2005) are characterised by lower ${}^{87}Sr/{}^{86}Sr$ values (≤ 0.7028) at similar ${}^{143}Nd/{}^{144}Nd$ values (0.51296-320 321 0.51298), coupled with higher ²⁰⁶Pb/²⁰⁴Pb (19.210-19.332), higher ²⁰⁸Pb/²⁰⁴Pb (38.862-38.935) 322 and lower ²⁰⁷Pb/²⁰⁴Pb values (15.579-15.600). In comparison, SAVF 'Group A' type (Cook et al., 323 2005), has higher ⁸⁷Sr/⁸⁶Sr values (≥0.7029) and similar ¹⁴³Nd/¹⁴⁴Nd values (0.51294-0.51298), coupled with lower ²⁰⁶Pb/²⁰⁴Pb (18.959-19.286), lower ²⁰⁸Pb/²⁰⁴Pb (38.752-38.905), and higher 324 325 ²⁰⁷Pb/²⁰⁴Pb values (15.597-15.624) similar to samples from Rangitoto (See Fig. 4.).

326

327 4.3. Re, Os, and ¹⁸⁷Os/¹⁸⁸Os

¹⁸⁷Os/¹⁸⁸Os values and Re and Os concentrations are given in **Table 1**. The AVF samples
show a wide range in Os and Re concentrations (Os = 5.8-578 ppt, Re = 25.9-411.9 ppt; **Fig. 5.**),
but concentrations fall in the range of contents reported from other OIB-type lavas (Os = 1-600
ppt: Schiano et al., 2001; Re = 100-642 ppt: Hauri and Hart, 1997). Similarly, the AVF volcanic
rocks have a significant range in Os isotope compositions from mantle-like ¹⁸⁷Os/¹⁸⁸Os of 0.1230
(cf. Meisel et al., 2000) to radiogenic values up to ¹⁸⁷Os/¹⁸⁸Os = 0.5470 (**Fig. 6**.). The majority of

samples (n = 19; including duplicates) have ¹⁸⁷Os/¹⁸⁸Os ≥0.15, higher than the range inferred for
mantle-derived magmas (Lassiter and Hauri, 1998; Widom et al., 1999; Rasoazanamparany et
al., 2015). In general no obvious correlations between Os and Re concentrations, ¹⁸⁷Os/¹⁸⁸Os
values and MgO, Ni, Cu or Zr concentrations or trace element ratios are observed. The
exceptions are samples from Pupuke, which have high MgO, Ni, and Os concentrations (Fig. 5.),
but relatively unradiogenic ¹⁸⁷Os/¹⁸⁸Os values (Table 1.).

340 The overall range in ¹⁸⁷Os/¹⁸⁸Os values in the AVF rocks (0.1230 to 0.5470) is larger 341 than those observed in most OIB-like basalts (ca. 0.11 to 0.14; Reisberg et al., 1993; Hauri and 342 Hart, 1993; Roy-Barman and Allègre, 1995; Marcantonio et al., 1995; Widom and Shirey, 1996; Lassiter and Hauri, 1998; Schiano et al., 2001; Day et al., 2010), and overlap with those in alkali 343 344 basaltic and tholeiitic lavas from other continental intraplate volcanic fields. These include Newer Volcanic Province, SE Australia ($^{187}Os/^{188}Os = 0.1342-0.4456$; McBride et al., 2001); 345 346 Central European Volcanic Province, Germany (187Os/188Os = 0.1487-0.7526; Jung et al., 2011) 347 and the East African Rift System ($^{187}Os/^{188}Os = 0.1239-0.4366$; Nelson et al., 2012) (Fig 6.). 348 Samples with more radiogenic Os isotopic composition generally contain less Os (e.g., Rangitoto sample AU59309: ¹⁸⁷Os/¹⁸⁸Os = 0.5470, Os = 5.8 ppt), and those with lower ¹⁸⁷Os/¹⁸⁸Os contain 349 more Os (e.g., Wiri Mt. sample AU43931: ¹⁸⁷Os/¹⁸⁸Os = 0.1283, Os = 194 ppt). In addition, low Os 350 concentration samples (<40 ppt) generally have more variable Os isotope signatures (0.1623-351 352 0.5470), becoming less variable (0.1230-0.1374) with increasing Os contents (>100 ppt). On an 353 ¹⁸⁷Os/¹⁸⁸Os versus Os diagram (Fig. 6.) the AVF data generally plot between crustal and mantle end-member fields, similar to other Pacific OIB fields (Fig. 6.) 354

355 Duplicate analyses of the same sample using the same digestion, and same sample 356 quantities, show highly reproducible results (e.g., Purchas Hill and Wiri samples; Table 1), 357 however, duplicate analyses of the same sample using different digestions show results which 358 plot along a positive trend between high Os with low ¹⁸⁷Os/¹⁸⁸Os, and low Os with high 359 ¹⁸⁷Os/¹⁸⁸Os)(Fig. 6.). These include samples from AVF centres Rangitoto, Three Kings, Purchas 360 Hill, Mt Wellington, and Puketutu. This effect has previously been observed (e.g., Allègre and 361 Luck, 1980; Potts, 1987; Alves et al., 2002) and termed the 'nugget effect'. This suggests that the 362 variable ¹⁸⁷Os/¹⁸⁸Os in an individual digestion is caused by sampling differing amounts of 363 mineral-hosted sulphides, or oxide micro-inclusions, with either inherited crustal, or mantle 364 ¹⁸⁷Os/¹⁸⁸Os signatures in a single powdered sample. The high reproducibility of Os contents and 365 ¹⁸⁷Os/¹⁸⁸Os for a split of the same digestion (c.f. **Table 1**) demonstrates effective digestion of the 366 sample, and argues against the variability in the results from different digestions of the same 367 sample being due to incomplete sample dissolution. Single samples are likely to contain

sulphide populations that have different Os contents and ¹⁸⁷Os/¹⁸⁸Os values, which are variably
 sampled by duplicate sample splits.

370

371 5. Discussion

372 5.1. The effects of mantle source heterogeneity and fractional crystallisation on Os 373 signatures

374 The petrogenesis of the AVF basalts has been the subject of a number of geochemical and isotopic studies detailed in section 2 (Huang et al., 1997; Needham et al., 2011; McGee et 375 376 al., 2011, 2012, 2013, 2015). The isotopic data, with specific emphasis on Pb isotope ratios, have 377 been used previously (Huang et al., 1997; McGee et al., 2013, 2015) to identify and fingerprint 378 three heterogeneous mantle sources, which are variably mixed to produce the isotopic and 379 geochemical variability observed within the AVF eruptive products. New Pb isotope data added 380 by this study (Fig. 4 A&B.) are consistent with those published for the AVF (McGee et al., 2013) and the SAVF (Cook et al., 2005), however, new Os isotope data obtained by this study show no 381 382 correlation to the Pb-Sr-Nd isotope systems (Fig. 7), suggesting that different processes control 383 these isotopic signatures within the AVF melts. This hypothesis is further supported by the lack 384 of correlation between Os and Re contents and the major and trace element concentrations of 385 the AVF rocks (Fig. 5.). In addition, no correlation is observed between the key trace element 386 ratios, and trace element anomalies (K and Sr) that have previously been used to determine the 387 mantle source signatures of the AVF rocks (e.g., La/Yb ratios; McGee et al., 2013, 2015). The 388 different behaviours of the major and trace element concentrations and isotope systems 389 therefore question whether or not a heterogeneous mantle source alone is responsible for the AVF Os signatures. 390

The generally low and variable Os concentrations, and lack of correlation between Os 391 392 and Re and the other major and trace element concentrations could be explained through 393 partial melting and fractional crystallisation. However, the concentrations of Os and Re in most 394 AVF samples show no obvious correlation between ¹⁸⁷Os/¹⁸⁸Os and MgO or, Ni, Cu or Zr (Fig 5.), 395 suggesting that fractional crystallisation of the major phases found within the AVF rocks 396 (olivine and pyroxene ±plagioclase) has an insignificant influence on the Os and Re budgets. 397 However, Platinum Group Elements (PGE), including Os, in addition to being siderophile are also chalcophile, and thus are relatively concentrated in sulphide minerals or PGE-rich metal 398 399 alloys (e.g., $D_{0s} \sim 10^4$ between sulphides and silicate melt; Roy-Barman et al., 1998; Jamais et al., 2008; Park et al., 2013; McCoy-West et al., 2015). Very minimal amounts of sulphide 400

401 fractionation will therefore preferentially remove Os from a melt, without a strong co-variance 402 with other elements. This is likely to contribute to the low Os concentrations that are seen in 403 the majority of AVF samples. Conversely, Re is less compatible to moderately incompatible in 404 sulphides and thus is not preferentially incorporated into them. Sulphides formed through 405 fractional crystallisation are either retained in residual minerals in the mantle (e.g., olivine; 406 Lorand et al., 2010) or segregate during magma transport and emplacement (e.g., Bézos et al., 407 2005). The PGE concentrations in the melt therefore depend on the degree of partial melting, as 408 only high degrees of partial melting (ca. ≥20%) will completely exhaust Base Metal Sulphides 409 (BMSs) from the mantle source, resulting in a PGE-enriched melt (e.g., Rehkämper et al., 1999; 410 Lorand et al., 2010; Dale et al., 2012). Accordingly, lower-degree partial melts will result in 411 lower and more variable HSE contents due to incomplete breakdown of sulphide in the mantle 412 source (e.g., Day et al., 2010). As low degrees of partial melting have been suggested to occur 413 beneath the AVF (≤6 %; McGee et al., 2013) variable and generally low Os contents are 414 therefore expected in the AVF melts. However, the absence of co-variance between the above-415 mentioned elements and Os suggest that partial melting and silicate + sulphide phase 416 fractionation cannot alone cause the variability in Os contents observed in the AVF rocks. Some 417 of the AVF samples have high Os contents together with high MgO and Ni contents (Fig. 5.), 418 suggestive of the accumulation of xenocrystic olivine (±pyroxene)-hosted sulphides into the 419 AVF melts during ascent (e.g., Alard et al., 2002). However, PGEs reveal an even more complex 420 behaviour depending on fO_2 , fS_2 , temperature and pressure prevailing during mantle melting, 421 crystal fractionation and during melt ascent and eruption. Regardless of the behavioural 422 complexity of BMSs, due to their low melting temperatures and low viscosities, they are soluble 423 in basaltic melts and will therefore be partially incorporated into the ascending magmas (Alard 424 et al., 2002). It is therefore likely that ascending melt will become contaminated with mantle or 425 crustal sulphides if the magma passes through an appropriate sulphide-bearing source. We 426 therefore propose that the Os systematics of the AVF are not controlled by mantle source 427 heterogeneity or fractional crystallisation, and instead are most likely controlled by 428 contamination and assimilation processes. This possibility is further investigated below in 429 regards to the Os isotopes and Os concentrations of the AVF basalts.

430

431 **5.2. Contamination and assimilation**

432 On an ¹⁸⁷Os/¹⁸⁸Os versus Os diagram (Fig. 6) most data from the AVF samples plot on a
433 curved array between mantle-like unradiogenic ¹⁸⁷Os/¹⁸⁸Os with elevated Os concentrations,
434 and radiogenic ¹⁸⁷Os/¹⁸⁸Os with low Os concentrations. This array is indicative of a mixing end

member (into a mantle-like source) with an elevated Os isotope ratio but a low Os content (Fig. 436
6.). However, ¹⁸⁷Os/¹⁸⁸Os vs. Sr-, Nd-, or Pb- isotopic systems (Fig. 4.) show no indication of a source for this signature, with no obvious trends towards either a combined mantle (e.g., young HIMU, EMII-type pelagic sediments, or carbonatite) or continental crustal signature (Fig. 7.).
The source of this mixing end member therefore remains unclear, and so potential contributors to contamination or assimilation in the mantle and crust are discussed below.

441 **5.2.1. Mantle Source**

442 Typical primitive mantle values for 187 Os / 188 Os are ≤ 0.1290 (Meisel et al., 1996), with 443 variable Os concentrations between 1-600 ppt (Schiano et al., 2001). However, in subduction zones the Os isotopic signatures of lithospheric mantle can be increased through the 444 445 incorporation of highly radiogenic oceanic crust (e.g., Alves et al., 1999; Borg et al., 2000; Dale et 446 al., 2007; Suzuki et al., 2011). In addition, fluids derived from the subducting plate will cause 447 metasomatism of the subcontinental lithosphere, leading to more oxidising conditions in the 448 mantle by increasing the fO_2 and fS_2 . Mantle hosted sulphides, which are more stable under 449 reducing conditions, will destabilise and oxidise to sulphates (e.g., Carroll and Rutherford, 450 1985), releasing sulphide-bonded metals, which include high concentrations of Os, and PGEs 451 (e.g., Jugo, 2009; Suzuki et al., 2011). Subduction-metasomatised lithosphere can therefore host a more radiogenic Os isotope signature than the ambient mantle, and has the potential to 452 453 contain higher concentrations of Os in partial melts. However, because Os is compatible with residual mantle sulphides during low degree partial melting, any erupted lavas will likely have 454 455 elevated radiogenic isotope signature and a low Os concentration (e.g., Widom et al., 1999).

456 The lithosphere beneath the AVF was exposed to multiple episodes of subduction at 457 least from the Cretaceous (e.g., Mortimer et al., 2006) and from Oligocene to Miocene (30 to ca. 458 20 Ma: Seebeck et al., 2014). In addition, studies on peridotitic xenoliths from the South Island 459 volcanic groups (e.g., Scott et al., 2014; McCoy-West et al., 2015) present evidence for 460 subduction-related carbonatitic metasomatism. Carbonatites typically have low Os concentrations and elevated Os isotope ratios (Widom et al., 1999; Escrig et al., 2005), and 461 therefore could represent a mantle end-member causing the radiogenic signature seen for the 462 AVF samples. However, with the exception of the high La/Yb ratio for the Purchas Hill samples, 463 the AVF samples do not show the geochemical signatures (e.g., low Ti/Eu or low wt.% Al₂O₃) 464 465 that have been attributed to carbonatite metasomatism (e.g., Scott et al., 2014). In addition, the 466 ²⁰⁶Pb/²⁰⁴Pb and ⁸⁷Sr/⁸⁶Sr isotope values in the AVF rocks are lower than the HIMU-type South 467 Island xenoliths (206Pb/204Pb >20). Using 187Os/188Os values from Widom et al. (1999) (e.g., 468 ¹⁸⁷Os/¹⁸⁸Os = 0.6), and Os concentration from Escrig et al. (2005) (e.g., Os = 15 ppt) modelling input of carbonatite into the system cannot readily explain the complete spread in Os (ppt)
versus ¹⁸⁷Os/¹⁸⁸Os data spread of the AVF samples used in this study (Fig. 8.).

Alternative models to explain the radiogenic ¹⁸⁷Os/¹⁸⁸Os geochemical composition 471 472 involve the presence of eclogitic or pyroxenitic or carbonated peridotite domains in the 473 asthenospheric or lithospheric mantle (e.g., Hoernle et al., 2006; Sprung et al., 2007; Timm et al., 474 2009; McCoy-West et al., 2010; McGee et al., 2013, 2015). This poses the question whether or 475 not partial melting of different mantle domains (peridotite vs. pyroxenite \pm CO₂ beneath the AVF, as originally proposed by McGee et al. (2013), may lead to the radiogenic Os isotope 476 477 signature of the AVF samples. In general neither the AVF Os isotope ratios, nor the Os 478 concentrations correlate with the typical geochemical tracers for carbonated peridotite, 479 carbonatite metasomatism (e.g., low wt.% SiO2 and low MgO#, or elevated CaO/Al2O3, Ce/Pb, 480 Nb/Ce; Scott et al., 2014; McGee et al., 2015) or pyroxenite (e.g., elevated Al₂O₃ coupled with 481 radiogenic ¹⁸⁷Os/¹⁸⁸Os; Marchesi et al., 2014). This is in agreement with a recent study by 482 McCoy-West et al. (2015) concluding that because the PGE budget is primarily controlled by 483 residual sulphides, carbonatitic metasomatism beneath Zealandia (the wider micro-continent 484 on which New Zealand sits) does not affect the PGE budget of the subcontinental peridotites. 485 These results also support previous conclusions by Handler et al. (1997) who also showed that for wherlite and apatite-bearing peridotites, interaction with carbonatitic melt has no effect on 486 487 the Os isotope composition or concentrations of Os or Re.

488 A number of studies (e.g., Kumar et al., 1996; Saal et al., 2001; Pearson and Nowell, 489 2004; Luguet et al., 2008) present evidence that mantle pyroxenites and eclogites can contain secondary metasomatised sulphides with highly radiogenic ¹⁸⁷Os/¹⁸⁸Os of >2, coupled with high 490 491 concentrations of Os. If the AVF melts accumulated or interacted with such secondary sulphide 492 of metasomatic origin, one would expect a correlation between high Os concentration and high 493 ¹⁸⁷Os/¹⁸⁸Os, the contrary to what is generally observed. Only three of the AVF samples analysed 494 (from Pupuke, and Wiri centres) do show slightly elevated ¹⁸⁷Os/¹⁸⁸Os of 0.1374-0.1377 coupled 495 with high Os concentrations (>200 ppt; Fig. 6.), potentially indicative of a mantle-derived origin. 496 Given that the latest lithospheric metasomatism event occurred 20-30 Ma ago, and based on the decay of ¹⁸⁷Re to ¹⁸⁷Os, the Os isotopic ratio of the present peridotitic lithospheric mantle would 497 498 be ca. ¹⁸⁷Os/¹⁸⁸Os = 0.1473-0.1524 (assuming ¹⁸⁷Re/¹⁸⁸Os = 50 in recycled crust: Widom et al., 1999; and an original ambient mantle value of ¹⁸⁷Os/¹⁸⁸Os = 0.1231-0.1283, from this study). 499 This ¹⁸⁷Os/¹⁸⁸Os ratio is however significantly higher than the values recorded in lithospheric 500 mantle xenoliths from Zealandia (including dunite and wherlite samples from Ngatutura Point: 501 502 ¹⁸⁷Os/¹⁸⁸Os = 0.1282±0.0012 ca. 80 km south of Auckland; McCoy-West et al., 2013). Although 503 the ¹⁸⁷Os/¹⁸⁸Os and Os content is likely to be variable beneath North Island, New Zealand, the 504 generally lower peridotite mantle-like ¹⁸⁷Os/¹⁸⁸Os values in Zealandia xenoliths suggests that 505 the slightly elevated ¹⁸⁷Os/¹⁸⁸Os at high Os contents observed in the Pupuke and Wiri Mt 506 samples are derived from a different source, which is discussed below.

507

508 **5.2.2. Crustal source**

509 The trace element signature for the Waipapa terrane crustal rock is plotted on Figure 3 510 (data from Price et al., 2015), highlighting the difference in concentrations between trace 511 elements such as Nb and Ta. Due to the different geochemical signatures of crustal and mantle 512 rocks, previous studies have suggested that certain trace element ratios can be indicative of 513 crustal contamination, typically showing high La/Ta (>22) and La/Nb (>1.5) ratios (e.g., Abdel-Fattah et al., 2004) and low Nb/U (<37) ratios (e.g., Hofmann et al., 1986). All AVF samples fall 514 515 outside of these ranges; exhibiting low La/Ta ratios (8.56 to 13.4) and low La/Nb ratios (0.58 to 0.75), coupled with high Nb/U signatures (42.61 to 53.36), with the exception of Rangitoto 516 517 sample AU59309, which shows a slightly lower Nb/U value (35.75) at low La/Ta and La/Nb 518 values. This, coupled with the lack of correlation between the major and trace element 519 signatures and the Os, Re concentrations and Os isotope signatures, indicates that the major and 520 trace element geochemistry of the AVF samples are not affected by crustal contamination.

521 Radiogenic isotope systems can however be more sensitive to crustal input in 522 comparison to the major and trace element compositions (e.g., Wilson, 1989). Sr-, Nd-, and Pbisotope values for the Waipapa terrane (Price et al., 2015) are plotted on Fig 4 in relation to the 523 524 AVF samples. Binary mixing of the two signatures (Waipapa and EMII) into an AVF mantle-like 525 source suggest that a $\leq 10\%$ input from Waipapa terrane rocks and $\leq 5\%$ input from an EMII-like source could be used to explain the spread in the Pb-isotope values (Fig. 4 A&B). However, this 526 527 input would produce a much larger range in Sr- and Nd- isotope signatures, which is not 528 observed within the AVF samples (Fig. 4C&D). This suggests that these isotope systems are also 529 not controlled by crustal contamination and show decoupled signatures to the Os isotopic 530 system (Fig. 7.). Therefore the contamination highlighted by the Os signatures must be minimal 531 in order to not show any impacts on the trace elements or Sr-, Nd- or Pb-isotopes.

For the Pupuke samples (as discussed previously) that show slightly elevated high Os contents, an alternative source potentially causing the contamination with sulphide-bearing olivine is ultramafic rocks of the Dun Mountain Ophiolite Belt (DMOB) (primarily serpentinised dunite and harzburgite) (e.g., Coombs et al., 1976; Sivell and McCulloch, 2000; Eccles et al., 2005). Parts of the DMOB cross directly beneath Pupuke at shallow levels (\geq 1.5 km depth, **Fig 1.C.**) making these rocks a likely melt contaminant. Although 538 no DMOB rocks were analysed for Os isotopes or Os concentrations, O'Driscoll et al. (2012) 539 report Os contents and isotope ratios from the Shetland ophiolite complex. The latter, although older (429 Ma), has similarities to the DMOB in formation (obduction at an arc collision zone), 540 and lithology (serpentinised dunite and harzburgite). 187Os/188Os isotope values reported for 541 542 rocks from the Shetland ophiolite complex range from 0.1204 to 0.1502 with Os concentrations 543 of 300-8000 ppt (O'Driscoll et al., 2012). Assuming a mean value of ¹⁸⁷Os/¹⁸⁸Os = 0.1353 and Os 544 = 4150 ppt, (within range of the Shetland rocks) simple binary mixing modelling (Fig. 8.) requires <10% input from the ultramafic rocks into our proposed ambient mantle values (of 545 187Os/188Os = 0.1231-0.1283 and Os = 50-200 ppt) to explain the 187Os/188Os value and Os 546 content observed in the Pupuke samples. However only ca. 5% olivine accumulation is required 547 548 to explain the higher MgO, Ni and Cr contents of these samples (Fig. 2.), suggesting that either the Os concentration in the contaminant needs to be ≥6000 ppt (still within the range reported 549 550 by O'Driscoll et al., 2012) or Os-bearing sulphides or metal alloys will dissolve more efficiently 551 than silicates and therefore more readily contaminate the ascending melts. Regardless, we 552 therefore attribute samples with elevated ¹⁸⁷Os/¹⁸⁸Os and high Os contents to the accumulation 553 of, or interaction with, the DMOB ultramafic rocks containing ancient subduction 554 metasomatised domains.

The interaction between the DMOB and ascending melts cannot however explain the 555 highly radiogenic Os isotope signatures (187Os/188Os = 0.1623-0.5470) and low Os 556 557 concentrations (<50 ppt) in most other AVF samples. These highly radiogenic ¹⁸⁷Os/¹⁸⁸Os isotope ratios exceed all values reported from the lithospheric mantle beneath New Zealand 558 559 (<0.133: cf. McCoy-West et al., 2015; Liu et al., 2015) and carbonatitic sources (<0.6: Widom et 560 al., 1999) as previously discussed. Continental metasediments typically show highly radiogenic 561 187Os/188Os values (0.165-2.323: Saal et al., 1998) and low Os contents (20 to 100 ppt: Saal et al., 562 1998; Widom et al., 1999), suggesting that crustal derived rocks are most likely acting as a contaminating agent to explain the high ¹⁸⁷Os/¹⁸⁸Os and low Os concentration in most AVF lavas. 563

564

565 5.3. AFC modelling

566In order to quantify the amount of fractional crystallisation and crustal contamination567acquired by the AVF samples, we used the combined assimilation fractional crystallisation568model of DePaolo (1981) (Fig. 8.). Potential crustal contaminants have to be a known basement569lithology, with a highly radiogenic Os isotope signature and low Os concentration. In addition,570the eruptive products must have undergone low levels of fractional crystallisation (<6%; McGee</td>571et al., 2013), and the mixing proportions between contaminant and melt have to be low enough

572 in order to have little effect on trace element values or the Sr-Nd-Pb isotope systems (e.g., Fig.4, 573 discussed in section 5.2.2.). Field studies mapping the basement terranes and crustal xenoliths in lavas indicate that, in addition to DMOB rocks, both Waipapa and Murihiku terrane 574 greywacke metasediments are found beneath the AVF (Fig. 1.B: e.g., Kermode, 1992). Although 575 576 no Os isotopic analyses exist for these terranes specifically, similar greywacke metasediments 577 have been studied in Australia (Saal et al., 1998) and India (Wimpenny et al., 2007), and generally have highly radiogenic ¹⁸⁷Os/¹⁸⁸Os of 1.2832 to 5.1968 and low Os concentrations of 578 579 40-100 ppt (Wimpenny et al., 2007). Os values similar to those exhibited by Wiri Mt AU43931 is chosen as representative of the mantle melt as it plots (along with Rangitoto sample AU59309) 580 581 within the OIB field for Os concentrations, Os isotopic and Pb isotopic values (Fig. 6. and 7.), 582 but does not have the analytical error that is associated with the Rangitoto sample (Table 1).

583 Modelling suggests that ≤1% bulk assimilation of greywacke metasediments with Os = 584 44.5 ppt and ¹⁸⁷Os/¹⁸⁸Os = 1.2832 (Saal et al., 1998) into a mantle melt (¹⁸⁷Os/¹⁸⁸Os = 0.1283; Os 585 = 194 ppt; Wiri Mt value from this study), coupled with \leq 5% fractional crystallisation, can 586 reproduce the range of observed radiogenic Os isotope signatures (and low Os contents) in the AVF samples (Fig 8.). Variations in mantle source signature between $^{187}Os/^{188}Os = 0.1$ and 0.15, 587 588 and Os content 50 to 200 ppt (to simulate heterogeneity in the mantle) have little impact on the percentage contribution from the contaminant, which remains at $\leq 1\%$ for these ranges (details 589 590 can be found in the supplementary material). Such low percentages of crustal contamination 591 have little to no effect on the Sr-, Nd-, or Pb-isotope signatures as modelled in Figure 4.

592

593 5.4. Implications for magma origin, generation and ascent

594 The AVF 187Os/188Os and Os contents fall into the range of 187Os/188Os and Os contents 595 from other intraplate volcanic fields (e.g., Newer Volcanic Province; Central European Volcanic 596 Province) and oceanic islands (e.g., Canary Islands; Cook Austral Islands, Comores; Hawaiian 597 Islands, Fig. 6.). Although the formation of large oceanic island volcanoes, such as the Canary or 598 Hawaiian Islands has mainly been attributed to the presence of relatively stationary thermal 599 anomalies in the mantle (e.g., Morgan, 1971; Bennett et al., 1996; Montelli et al., 2006), the 600 origin of small-scale intraplate volcanism is much less clear. Models to explain the origin of 601 small continental intraplate volcanoes or volcanic fields include local lithospheric extension 602 (e.g., Weaver and Smith, 1989; McCoy-West et al., 2013), lithospheric delamination (e.g., Jull and 603 Kelemen, 2001; Elkins-Tanton, 2007), or edge driven convection (e.g., King, 2006). The 604 processes driving melt generation beneath the Auckland and other Quaternary Volcanic Fields 605 on North Island, New Zealand is a matter of current debate, but there is a general consensus that

extension-related magmatism is involved (Weaver and Smith, 1989; Smith et al., 1993; Huang et
al., 1997; Cook et al., 2005; Needham et al., 2011).

608 As a consequence of long-lasting exposure of the lithospheric mantle to subduction 609 metasomatism (through the influx of fluids during the Mesozoic), and plume-related 610 magmatism during the Cretaceous, Huang et al. (1997) and Cook et al. (2005) argue that both 611 the 'young' HIMU and EMII-type signatures observed in the volcanic field geochemistry 612 originate in a partially melted zone in the lithospheric mantle. Conversely, McGee et al. (2013, 2015) suggest that the 'young' HIMU-type signature is linked to carbonated domains within the 613 614 upper asthenosphere. Between 70-90 km depth beneath the Auckland region seismic 615 experiments revealed a low velocity zone, which is interpreted to represent regions of partial melt within the mantle caused by crustal extension following rollback of Pacific Plate 616 617 subducting beneath the North Island of New Zealand (Horspool et al., 2006). Beneath South 618 Island New Zealand, the base of the lithosphere has been estimated to lie at ca. 100 km (e.g., 619 Molnar et al., 1999), which would place this proposed partially melted zone within the 620 lithospheric mantle. However, because this lithospheric thickness of ca. 100 km has been 621 determined on the Pacific Plate (Molnar et al., 1999), there is some uncertainty as to how 622 representative this thickness is of the Auckland area, which sits on the Indo-Australian Plate 623 (e.g., Stratford and Stern, 2004). Regardless, regional lithospheric extension events may 624 sufficiently thin the lower lithosphere to facilitate regional influx of hot MORB-like 625 asthenospheric mantle and subsequent partial melting of the uppermost asthenospheric and 626 lithospheric mantle via decompression and heating.

627 As previously outlined, the geochemical composition of lavas from Rangitoto is 628 consistent with their derivation from the metasomatised lithosphere (e.g., Needham et al., 2011; 629 McGee et al., 2015). Accordingly the lowest ¹⁸⁷Os/¹⁸⁸Os of 0.123 in lavas from Rangitoto could be 630 representative of the ¹⁸⁷Os/¹⁸⁸Os of the lithospheric mantle beneath the Auckland Volcanic 631 Field. This would be consistent with the unradiogenic ¹⁸⁷Os/¹⁸⁸Os values of peridotite xenoliths 632 from Zealandia (ca. 0.12-0.13; McCoy-West et al., 2013; Liu et al., 2015), however the error 633 associated with this measurement mean constraining an accurate value is problematic. Alternatively, the low-silica lavas from Wiri Mt. show ¹⁸⁷Os/¹⁸⁸Os of 0.128 similar to the Os 634 635 isotopic composition of peridotite xenoliths in the 1.68±0.15 Ma intraplate Ngatutura Volcanics 636 (e.g., Briggs et al., 1994). Based on the major and trace element and Sr-, Nd-, and Pb-isotopic 637 composition in lavas from Wiri, McGee et al. (2013) proposed a predominately garnet-bearing 638 peridotitic source for these lavas (± carbonate; McGee et al., 2015). However, because Wiri is 639 located above the DMOB, we cannot be sure that the ${}^{187}\text{Os}/{}^{188}\text{Os}$ in these lavas are

representative of the present day mantle beneath the AVF, or reflect minor contamination fromthe DMOB.

642 Regardless of mantle melt generation complexities beneath the AVF most of our new Os 643 isotope data are more radiogenic than typical mantle values, suggesting that the ascending melts assimilated radiogenic ¹⁸⁷Os/¹⁸⁸Os isotope signatures from the metasedimentary 644 Murihiku or Waipapa terranes beneath Auckland. The exceptions to this are the olivine-bearing 645 646 samples from Pupuke (and potentially Wiri Mt.), indicating the significant assimilation of sulphide-bearing olivine from the subsurface DMOB (Eccles et al., 2005). Of note is that both 647 648 Pupuke and Wiri Mt. are located near or on faults separating the DMOB from adjacent rocks of the Murihiku Terrane (c.f. Fig. 1.C; Eccles et al., 2005), which suggests olivine accumulation 649 650 beneath the AVF could occur while magma ascends along faults cutting or bounding the 651 ultramafic rocks of the DMOB (Eccles et al., 2005). Generally terrane boundaries represent 652 crustal (and potentially lithospheric) weak-zones, which are commonly associated with shear 653 zones and intense faulting (e.g., Smith and Mosley, 1993). Because all three main Quaternary 654 intraplate volcanic field in the North Island of New Zealand are roughly spatially correlated to 655 the position of the DMOB, and the crustal-scale Taranaki fault (e.g., Giba et al., 2010; Fig. 1.C.), 656 we suggest that feeder melts may exploit the faults and shear-zone along this major terrane boundary to ascend to the surface (Fig. 9.) rapidly rather than forming large crustal magma-657 658 reservoirs. Even though ascent may be rapid the high solubility of sulphides in basaltic silicate 659 melt provides an effective way of contaminating rising melts with crustal sulphides.

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661 6. Conclusions

662 In summary, we show that most of the AVF samples studied show Os isotopic 663 compositions higher than typical mantle values (≥ 0.13) and Os contents lower than 50 ppt. 664 Conversely, a minority of samples (from Pupuke, Wiri and Rangitoto) show unradiogenic Os 665 isotopic ratios coupled with high (>150 ppt) Os concentrations. Neither of these signatures 666 shows any obvious correlation to the tracers of mantle sources, implying that they are not 667 primarily caused by source heterogeneity, nor do they show co-variance with elements 668 dominant in olivine or pyroxene minerals, suggesting that they are not primarily caused by 669 fractional crystallisation. Contrary to previous interpretations our results suggest that AVF 670 melts do interact with continental crust during ascent leading to contamination signatures. 671 From these two signatures two differing sources of crustal contamination are identified; 1) 672 metasediments (e.g., Waipapa and Murihiku Terranes) which contain highly radiogenic Os 673 coupled with minimal Os concentrations and 2) xenocrystic olivine hosted sulphides (from the

674 DMOB) with unradiogenic mantle-like 187 Os/ 188 Os and high Os contents. Less than 1% 675 contamination from the crust coupled with $\leq 5\%$ fractional crystallisation, and $\leq 10\%$ 676 contamination from xenocrystic olivine-bearing sulphide is sufficient to reproduce the Os 677 contents and isotopic compositions observed in the AVF rocks.

The presence of mantle-derived sulphide hosted in olivines in eruptive products from centres located above the DMOB strongly argues for their derivation from crustal levels. We therefore propose that, although there is no evidence for long residence time of melts within the crust, crustal contamination does occur beneath most of the AVF. However, the low amount of crustal contamination is insufficient to affect the primitive geochemical and Sr-, Nd- and Pbisotopic signatures of the AVF products.

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1095 Figure Captions

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1096Figure 1. Schematic map of the Auckland Volcanic Field (AVF). (A) Location of the AVF within the North1097Island, New Zealand, inset shows the location of New Zealand in relation to the east coast of Australia. (B)1098DEM map and location of the volcanic centres in the AVF, highlighted are those sampled for this study,1099shaded zones show the proposed terranes at 1.5 km depth (from Eccles et al., 2005). The green dashed line1100(A&B) indicates the Dun Mountain Ophiolite Belt identified by the Junction Magnetic Anomaly (JMA) (Eccles1101et al., 2005). (C) Schematic diagram adapted from Kermode (1992) and Eccles et al. (2005) outlining the1102basement geology beneath the AVF: cross section A-A' is shown in diagram A.

1104Figure 2. Diagram panels showing major elements vs. Mg0 (wt.%) for studied samples. Grey symbols1105represent previously published data from the SAVF, with squares denoting Group A and crosses denoting1106Group B samples, as assigned by Cook et al. (2005); and circles showing data from Briggs et al. (1994), which1107were ungrouped. Also shown in the SiO2 vs. MgO panel is the impact of mixing olivine into the samples,1108showing that the addition of 5% Dun Mountain derived olivine (values from Sano and Kimura, 2007) could1109produce the Pupuke sample signatures.1110

1111Figure 3. Diagram showing representative primitive mantle-normalised multi-element compositions to1112present the AVF trace element data range (McGee et al., 2013). The end members are exemplified by Purchas1113Hill and Rangitoto 2. Wiri Mt. samples show intermediate trace element ratios. The grey field marks the1114range from the AVF, black line shows a typical OIB-type signature (OIB values from Sun and McDonough,11151989, and normalisation values from McDonough and Sun, 1995), and blue dashed line show the1116composition of the Waipapa terrane metasediments (from Price et al., 2015).

Figure 4. 206Pb/204Pb vs. (A) 207Pb/204Pb, (B) 208Pb/204Pb, (C) 87Sr/86Sr, and D) 143Nd/144Nd for all samples 1118 1119 from the Auckland Volcanic Field (this study and McGee et al., 2013), and South Auckland Volcanic Field 1120 (Cook et al., 2005). The inset shows the mantle reservoir end members from Zindler and Hart (1986): 1121 Enriched Mantle (EMI and EMII), Pacific MORB Mantle, and HIMU. Values for Northland from Huang et al. 1122 (2000), for Otago from Timm et al. (2010), for Banks Peninsular from Timm et al. (2009), and for Lookout 1123 Volcanics from McCoy-West et al. (2010), for Waipapa metasediments from Price et al. (2015). Modelling 1124 lines show binary mixing of Waipapa metasediments (blue) and lithospheric mantle (EMII; green) into AVF 1125 mantle-like source, with the shaded area showing the potential signatures caused by these inputs. 1126 1127

1128Figure 5. Os and Re concentrations versus Mg0 (in wt.%), Ni, Cu and Zr (all in ppm) contents for samples1129analysed in this study. Symbols are as in Figure 2. Note that only 5 samples were analysed by this study for1130major and trace elements. Previous studies (McGee et al., 2013) did not present Cu data for these samples,1131therefore a reduced number of samples are plotted in Cu vs. Os and Re plots.

1132 1133 Figure 6. 187Os/188Os vs Os (ppt) for all samples from the AVF. Also shown in grey symbols are global OIB 1134 values for Pacific regions: Austral-Cook (Hauri and Hart, 1993; Reisberg et al., 1993; Hanyu et al., 2011); 1135 Samoa (Jackson and Shirey, 2011); Newer Volcanic Province, Australia (NVP; McBride et al., 2001); 1136 Louisville Seamount Chain (Tejada et al., 2015), and for the Atlantic regions: the Canary Islands 1137 (Marcantonio et al., 1995; Widom et al., 1999; Day et al., 2009); St Helena and Comores (both from Reisberg 1138 et al., 1993); Cape Verde Islands (Escrig et al., 2005); Azores (Widom and Shirey, 1996; Larrea et al., 2014); 1139 Central European Volcanic Province (CEVP; Jung et al., 2011). Error bars are shown for one Rangitoto 1140 sample, for all other analyses the errors are smaller than the symbol size. 1141

- 1142Figure 7. Diagram after Day (2013), to show mixing relationships for various potential contaminants in1143(A)206Pb/204Pb, (B) 143Nd/144Nd, and (C) 87Sr/86Sr vs. 187Os/188Os isotope space. Data values for MORB and
 - 1144 OIB from Widom et al. (1999), Hofmann (1997), and Day et al. (2010); pelagic sediments from Roy-Barman

1145 and Allègre (1995) and Eisele et al. (2002); carbonatite from Widom et al. (1999) and Escrig et al. (2005); 1146 young HIMU from Day et al. (2009); and continental crust from Saal et al. (1998) and Widom et al. (1999). 1147 Figure 8. Whole-rock 187Os/188Os vs Os concentration (ppt) for AVF samples with proposed methods of 1148 1149 signature formation. Sulphide assimilation is modelled (green) using binary mixing from AVF mantle values 1150 with values measured for sulphides found in dunite within Shetland Ophiolite complex (187Os/188Os = 0.1353, 1151 Os = 4150 ppt: O'Driscoll et al., 2012). Fractional crystallisation is modelled (blue) using the Rayleigh 1152 equation for samples from 50 to 7 ppt, showing that it requires $\leq 10\%$ fractional crystallisation to reduce the 1153 Os concentrations. Crustal assimilation is modelled (orange) through binary mixing of post 10% fractional 1154 crystallisation of the mantle-derived melt ($^{187}Os/^{188}Os = 0.1283$, Os = 7 ppt) with <10 % crust, with values 1155 reflective of greywacke (187Os/188Os = 1.283, Os = 44.5 ppt: Saal et al., 1998). Assimilation fractional 1156 crystallisation (AFC) is modelled (after DePaolo, 1981) with Dos= 20 (Widom et al., 1999), r (rate of 1157 fractional crystallisation) = 0.95, for mantle values ($^{187}Os/^{188}Os = 0.1283$, Os = 194 ppt (Wiri sample) with 1158 crustal values of greywacke (black line, ¹⁸⁷Os/¹⁸⁸Os = 1.283, Os = 44.5 ppt; Saal et al., 1998), and carbonatite 1159 (grey line, ¹⁸⁷Os/¹⁸⁸Os = 0.6, Os = 15 ppt; from Widom et al., 1999 and Escrig et al., 2005). 1160 1161 Figure 9. Schematic model to illustrate the proposed magma ascent pathways for the AVF eruptions. Melts 1162 are derived from a heterogeneous source, including ambient peridotite mantle containing HIMU-like 1163 carbonated peridotite veins (McGee et al., 2015) at a depth of >80 km, and a subduction metasomatised

1165 carbonated periodate vens (integet et al., 2015) at a depth of 900 km, and a subdated on metasonalised
1164 lithosphere at <80 km depth, all of which give the ascending magma its Sr-, Nd-, Pb-isotopic and major and
1165 trace element signatures (McGee et al., 2013). Minimal fractionation occurs on ascent, efficiently reducing
1166 the 0s concentration from mantle values, followed by preferential assimilation of olivines from the Dun
1167 Mountain Ophiolite Belt (DMOB) as xenocrystic material causing increase in 0s concentration, and finally
1168 minor crustal contamination of magmas causing the radiogenic isotope signatures. A crustal depth of 20-30
1169 km and the positions of the low velocity melt zone region are from tomography by Horspool et al. (2006),
1170 and upper cross section is adapted from Seebeck et al. (2014).

1172 Table Caption

1173 Table 1. Selected geochemical data for AVF centres, * denotes data from lavas previously analysed by McGee

1174 et al. (2013). Suffixes 'i, ii, iii' etc denote duplicate analyses from the same sample but with a different

1175 digestion, (2) denotes duplicate analysis of the same sample and same digestion.

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Supplementary Material

SM1 – All major and trace element, and isotopic values for samples analysed used in this study.

SM2 – All standard data for analysis of major, trace and isotopes.

SM2.1. Major elements by XRF.

SM2.2. Trace elements by solution ICP-MS.

SM2.3. Pb isotope analysis by multi-collector ICP-MS.

SM2.4. Os isotope analysis by TIMS.

- SM2.5. Re isotope analysis by sector field ICP-MS.
- SM3 Modelling parameters for the contamination effects on other isotope systems
 - SM3.1. Values used for modelling
 - SM3.2. Calculations for ²⁰⁶Pb/²⁰⁴Pb vs ⁸⁷Sr/⁸⁶Sr
 - SM3.3. Calculations for ²⁰⁶Pb/²⁰⁴Pb vs ¹⁴³Nd/¹⁴⁴Nd
 - SM3.4. Calculations for ²⁰⁶Pb/²⁰⁴Pb vs ²⁰⁸Pb/²⁰⁴Pb
 - SM3.5. Calculations for ²⁰⁶Pb/²⁰⁴Pb vs ²⁰⁷Pb/²⁰⁴Pb

SM4 – Modelling parameters for AFC calculations

SM4.1. Optimum values used for plotting figure 8.

SM4.2. Variations in mantle source signature.

SM4.3. Variations in r value.