Absence of hydrothermal oxygen isotope variations in host rocks supports magmatic origin of the giant Grängesberg iron oxide–apatite (IOA) deposit, Central Sweden

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Abstract

The origin of Kiruna-type iron oxide–apatite ores is controversial, and debate presently centres on a ‘magmatic’ versus a ‘hydrothermal’ mode of formation. To complement recent investigations on the Grängesberg iron oxide–apatite ore deposit in the northwestern part of the Palaeoproterozoic Bergslagen ore province in central Sweden, we investigated the oxygen isotope composition of the host rocks of this large iron oxide–apatite ore body. As the metavolcanic and metagranitoid country rocks around the Grängesberg ore body either pre-date or are coeval with ore formation, they would be expected to record an extensive isotopic imprint if the ore body had formed by large-scale hydrothermal processes involving an externally sourced fluid. A direct magmatic formation process, in turn, would have produced localized alteration only, concentrated on the immediate vicinity of the ore body. Here, we test these two hypotheses by assessing the oxygen isotope variations in the host rocks around the main Grängesberg iron oxide–apatite ore body. We analysed oxygen isotopes in quartz from metavolcanic ($n = 17$) and metagranitoid host rocks ($n = 14$) from the vicinity of the ore body, and up to 2 km distance along and across the strike of the ore body. Remarkably, we find no significant variation in $\delta^{18}\text{O}$ values with distance from the ore body, or any deviations in country rock $\delta^{18}\text{O}$ from common magmatic and/or regional values. Only two samples show shifts to values more negative than the common magmatic range, indicating highly localized hydrothermal overprint only. As a large-scale, low-temperature hydrothermal origin of the ore body through voluminous fluid percolation would be expected to have left a distinct imprint on the oxygen isotope values of the country rocks, our results are more consistent with an ortho-magmatic origin for the Grängesberg iron oxide–apatite ore.

Keywords Kiruna type · Iron oxide–apatite ore · Hydrothermal alteration · Host rock · Oxygen isotopes · Grängesberg · Bergslagen · Sweden

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Introduction and approach

The genesis of ‘Kiruna-type’ iron oxide–apatite ores is the focus of a long-standing debate (e.g. Geijer 1910, Parak 1975, Weidner 1982, Naslund 1983, Nyström and Henriquez 1994, Barton and Johnson 1996, Sillitoe and Burrows 2002). Genetic models range from a direct magmatic (ortho-magmatic) mode of origin from Fe–P-rich magmas, via formation from high-temperature magmatic fluids by direct crystallization or liquid immiscibility ($\geq 800^\circ\text{C}$) (cf. Nyström et al. 2008; Jonsson et al. 2013; Weis et al. 2013; Knipping et al. 2015; Bilenker et al. 2016; Tornos et al. 2016, 2017; Hou et al. 2017, 2018; Sun et al. 2019; Troll et al. 2019; Sarlus et al. 2020), to a hydrothermal mode of formation involving transport and precipitation, including replacement-type

reactions, by means of aqueous fluids at moderate to low (sub-magmatic) temperatures (typically ≤ 400 °C) (cf. Parak 1975; Hitzman et al. 1992; Rhodes and Oreskes 1999; Sillitoe and Burrows 2002; Smith et al. 2013; Dare et al. 2014; Westhues et al. 2016, 2017). Whereas a lively debate between the two endmember schools (magmatic vs low-temperature hydrothermal) is ongoing, one aspect that has not yet been fully exploited is the relationship of the Kiruna-type ores with their surrounding host rocks. Using the example of the Grängesberg Mining District in Central Sweden, we investigate if the surrounding country rocks to the main ore body, which comprise (meta-)volcanic and (meta-)plutonic rocks that are possibly coeval with ore formation, record any evidence for large-scale hydrothermal activity. To achieve this, we utilize whole rock and mineral oxygen isotope data to test the possibility of a large-scale pervasive or systematically changing (progressive) hydrothermal overprint in the vicinity of the iron oxide–apatite mineralization. Specifically, hydrothermal iron ore formation has been suggested to encompass large-scale hydrothermal circulation such as seafloor exhalation, sub-seafloor hydrothermal replacement of host rocks, or hydrothermal fluid interaction with an evaporitic source (e.g. Parak 1975; Hitzman et al. 1992; Hitzman 2000; Barton and Johnson 1996; Rhodes and Oreskes 1999; Dare et al. 2014). If a large-scale hydrothermal origin is considered, then fluids from external sources would likely be involved, which would affect the O isotope composition of the immediate host rocks to the ore body and produce an aureole that is distinct from the regional isotope composition (cf. Taylor 1971, 1974; Beaty et al. 1988; Dilles et al. 1992; King et al. 1997). Similarly, in a large-scale hydrothermal aureole, fluids would progressively cool away from the heat source as well as experience progressive fluid–rock interaction, leading to variable isotope signals with distance from the heat source (e.g. Nabelek 1987; Beaty et al. 1988; Ayers et al. 2006; Donoghue et al. 2010; Berg et al. 2018).

To test for the presence or absence of a large-scale hydrothermal influence on the surrounding host rocks, samples from the vicinity of the Grängesberg ore body were systematically collected and analysed for their whole rock and quartz $\delta^{18}\text{O}$ values. Hydrothermal overprinting on igneous rocks usually leads to a change in the oxygen isotope composition of the rock with the direction and magnitude of the change depending on fluid origin, fluid–rock ratio and temperature of the fluid–rock interaction (e.g. Taylor 1971; Rhodes and Oreskes 1999; Götze et al. 2001; Donoghue et al. 2008, 2010; Berg et al. 2018; Kleine et al. 2018). The approach chosen in this study is analogous to those by Taylor (1971, 1974), Beaty et al. (1988), Dilles et al. (1992), King et al. (1997) and Ayers et al. (2006), who employed whole rock and mineral $\delta^{18}\text{O}$ to investigate hydrothermal alteration around various ore deposits and intrusions. These studies demonstrated that spatial changes in isotopic signatures can

be used to identify channelized or pervasive movement of hydrothermal fluids due to a localized or pervasive overprint in the form of, e.g. a zonal or “bullseye” pattern around the ore body, or a more asymmetric pattern if a certain sector or domain was particularly strongly affected (e.g. Taylor 1971, 1974; Nabelek 1987; Beaty et al. 1988; Singleton and Criss 2002; Skelton et al. 2007). In these cases, systematic isotopic gradients have been identified in country rocks over distances ranging from a few hundred metres to several kilometres from the investigated ore bodies or intrusions.

Here, we use quartz grains in addition to whole rock samples, since quartz is usually relatively robust and often resistant to secondary low-temperature alteration processes (cf. Taylor 1968; Larson and Taylor 1986; Dilles et al. 1992; Budd et al. 2017). However, quartz may change its isotope composition by, e.g. infilling of microfractures within individual crystals during hydrothermal alteration (e.g. King et al. 1997). For example, hydrothermal activity around the Kidd Creek VMS deposit affected host rock quartz in the immediate surroundings (~ 350 – 400 m) of the ore body, leading to a 5‰ increase from the originally magmatic $\delta^{18}\text{O}$ values of the quartz. However, a fluid-mediated overprint on the $\delta^{18}\text{O}$ composition related to later intrusions or regional metamorphism cannot be entirely ruled out (Hyslop et al. 2008), but such processes seem not to have had a significant impact on the isotope compositions of the Grängesberg ore itself (Jonsson et al. 2013). In contrast, hydrothermally precipitated quartz and amorphous forms of silica tend to display comparatively high oxygen isotope values (e.g. > 15 ‰, Götze et al. 2001; Kleine et al. 2018), allowing for a clear distinction of magmatic versus hydrothermally derived types of quartz. Assuming that all sampled country rocks to the ore were present at the time of ore formation and that tectono-thermal activity post-dating ore formation had not fully overprinted the O isotope composition, we can now resolve if traceable isotope compositional gradients typical for hydrothermal alteration exist in the surroundings to the Grängesberg ore body (cf. Taylor 1971, 1974; Dilles et al. 1992; King et al. 1997; Donoghue et al. 2008).

The Grängesberg iron oxide–apatite deposit

Grängesberg is located in the northwestern part of the Palaeoproterozoic Bergslagen ore province in south-central Sweden (Allen et al. 1996, 2008; Stephens et al. 2009; Stephens and Jansson 2020). The Grängesberg mine itself was the largest iron ore mine in southern and central Sweden, and was in operation until 1989. It was closed due to a combination of low metal prices at the time and political reasons, despite considerable ore reserves remaining. Efforts towards a re-opening of the mine are currently underway. The Grängesberg iron oxide–apatite ore body itself is dominated by

massive magnetite ore with subordinate haematite. Both oxides occur, however, also as veins and disseminations in the immediate surroundings of the main ore body (Fig. 1; Magnusson 1970). In the massive magnetite ore, there are characteristic bands of fine-grained fluorapatite, with variable amounts of silicate minerals such as amphiboles, biotite, chlorite as well as diverse REE minerals (Jonsson et al. 2013). The main part of the deposit, the so-called ‘Export Field’, consists of approximately 80% magnetite and 20% haematite in terms of oxide ore minerals (e.g. Jonsson et al. 2013). The main mineralization occurs as massive lenses or “logs” that dip between 50° and 75° towards the south-east and can be traced for more than 900 m at the surface, where its width ranges between 50 and 100 m (Johansson 1910; Geijer and Magnusson 1944).

The Grängesberg iron ore body is hosted in a suite of metavolcanic rocks with basaltic to rhyolitic composition that belongs to the ca. 1.91–1.87 Ga volcano-sedimentary succession of Bergslagen. This was previously known as the

“leptite-hälleflinta formation” (e.g. Allen et al. 1996). The majority of metavolcanic rocks in Bergslagen are rhyolitic (and to a lesser extent dacitic) and have been affected by regional K- and/or Na-alteration (Allen et al. 2008; Stephens et al. 2009; Stephens and Jansson 2020, and references therein). The immediate host rocks to the Grängesberg ore are dominated by metavolcanic to meta-subvolcanic rocks of intermediate to basic compositions (Jonsson et al. 2013). Moreover, the ore body as well as the host rocks in the Export Field (Fig. 1) are transected by dykes and sills of mafic to more felsic compositions, as well as by a set of significantly younger granitic pegmatite dykes (Geijer and Magnusson 1944; Jonsson et al. 2011, 2013). Notably, haematite-dominated ore occurs mostly in the structural footwall, and in the vicinity of crosscutting pegmatite dykes. Although most of the haematite in the Grängesberg ore body is believed to be of magmatic origin, some of it, especially in the form of veins and disseminations, most likely is a product of later alteration processes (Jonsson et al. 2013). Alteration zones within the host rocks next to, and within the

Fig. 1 **a** Overview map of Fennoscandia showing the Bergslagen province and the Kiruna–Malmberget mining district in Sweden. **b** Geological map over the main ore zone of the Grängesberg Mining District. **c** Vertical section (line X–Y in panel **b**) through the main ore body. The ore zone extends downwards at a 50°–75° dip to the SE.

Black horizontal lines are adits (maps after Jonsson et al. 2013). The immediate country rocks to the Grängesberg ore body are dominated by metavolcanic rocks and numerous minor intrusions such as dykes and sills, while older and younger granites are located further from the ore field

ore body, comprise disseminated and discrete phyllosilicate (biotite, chlorite) and amphibole-rich assemblages (so-called *sköl* assemblages) and these also contain variable amounts of accessory iron oxides and fluorapatite (Frietsch 1982; Jonsson et al. 2011). Besides these assemblages, there are also traces of alkali alteration (K-feldspar, albite) present in the Grängesberg area. This, however, is a regional feature throughout the whole Bergslagen region, not confined to the Grängesberg area (Allen et al. 2008; Stephens et al. 2009; Stephens and Jansson 2020), and hence not directly associated with the ore-forming process here. In fact, country rocks in the vicinity of the ore body preserve a predominantly igneous chemical character as has previously been noted by Persson et al. (2013), e.g. on the basis of their K_2O and Na_2O relationships (see below). Moreover, the massive magnetite ores seem also unaffected by this alkali alteration or by the regional metamorphism. The hydrous alteration assemblages present are, in turn, always spatially related with the ore and are absent beyond the main ore zone and the direct vicinity of the ore body (Jonsson et al. 2013; Persson et al. 2013), implying a genetic relationship between the two and the absence of a pronounced mineralogical aureole to the Grängesberg ore body.

The host rocks and the Grängesberg iron oxide–apatite ores have been affected by at least two phases of ductile deformation during the ca. 1.9–1.8 Ga Svecokarelian orogeny (e.g. Stephens et al. 2009; Högdahl et al. 2013). If a larger-scale hydrothermal “aureole” was generated during ore formation, i.e. prior to deformation and metamorphism, the O isotope pattern around the deposit might be expected to have been affected by these hydrothermal processes (cf. Taylor 1971, 1974; Beaty et al. 1988; Dilles et al. 1992; Ayers et al. 2006). Thus, some pattern of variation in isotopic values around the deposit would be expected, for instance in the form of a folded pattern or as a seemingly random mixture of high and low values due to, e.g. channelized flow and later deformation, despite later overprinting processes. As the main host rock in the hanging wall east of the ore body, a gneissic granitoid with a strong stretching lineation, is possibly younger than the ore itself (Jonsson et al. 2011; Högdahl et al. 2013), a recorded alteration pattern would primarily be expected in the foot wall to the west of the Export Field, whereas the east of the Export Field would be more representative of regional oxygen isotope signatures.

Methods

Samples ($n=23$) of the host rocks to the Grängesberg ore body have been collected from outcrops in the area around the iron oxide–apatite deposit in a suite of traverses up to 2 km away from the main ore body (Figs. 2, 3, 4; Table 1). From these samples, we analysed whole rock ($n=17$) and

quartz separates ($n=14$) for their oxygen isotope compositions (Table 1).

Whole rocks were analysed with a conventional vacuum extraction line at the University of Cape Town, South Africa. About 10 mg of sample was dried overnight at 50 °C and then loaded into Ni tubes. The samples were then degassed at 200 °C for 2 h under vacuum, then reacted with ClF_3 for 4–6 h at 550 °C. The liberated oxygen was passed over a hot carbon rod and converted to CO_2 which was then collected in glass tubes. The in-house standard MQ was run in duplicate with each batch of eight samples and used to convert the raw data to the SMOW scale; MQ has a $\delta^{18}O$ value of 10.1‰ calibrated against NBS-28, assuming a value of 9.64‰ (Coplen et al. 1983). Quartz was analysed by the laser fluorination method described by Harris and Vogeli (2010), whereby they were reacted with 10 kPa of BrF_5 and the purified O_2 was collected onto a 5 Å molecular sieve in a glass storage bottle. Oxygen isotope analysis was carried out using a Finnigan DeltaXP dual inlet gas source mass spectrometer. The in-house Monastery garnet (MON GT) standard was analysed in duplicate with each batch of ten samples. The MON GT was calibrated against UWG-2 garnet standard from Valley et al. (1995) and has a $\delta^{18}O=5.38‰$ (Harris and Vogeli 2010), which assumes $\delta^{18}O=5.80‰$ for the UWG-2. All raw data were normalized to a MON GT value of 5.38‰. All oxygen data were recorded in the usual δ notation relative to SMOW where $\delta^{18}O=(R_{\text{sample}}/R_{\text{standard}}-1)\times 1000$, and R =the measured ratio $^{18}O/^{16}O$. Based on the long-term duplication of both in-house standards, the $\delta^{18}O$ values have 2σ errors of 0.2‰. Detailed information on sample preparation is given in Harris et al. (2015).

Results

The country rock samples comprise intermediate- to high-silica metavolcanic and metaplutonic rocks and thin section images of representative rock samples are provided in Fig. 3. All samples and analytical data are reported in Table 1. The assemblages are not particularly altered for most parts and show predominantly igneous mineralogical and chemical signatures (Figs. 2, 3, 4), although some metamorphic overprint and regional alkali alteration are detected in some samples. With respect to oxygen isotope systematics, the Grängesberg metavolcanic and metaplutonic host rocks record $\delta^{18}O$ values exclusively between +5.8 and +8.6‰, which essentially overlaps with the range of $\delta^{18}O$ values for ‘normal’ intermediate arc magmas (cf. Taylor 1968; Bindemann 2008; Budd et al. 2017, Fig. 5; Table 2). Quartz from meta-granites and meta-dacites in the vicinity around the ore body exhibits $\delta^{18}O$ values from +6.0 to +10.3‰, with the majority

Fig. 2 Field photographs of major rock types in the Grängesberg area (see also Table 1). **a** Coarse porphyritic, granitic gneiss with mafic enclaves. **b** Fine and even-grained felsic metavolcanic rock with a pronounced east-dipping foliation. **c** Feldspar-porphyritic, phyllosilicate altered felsic metavolcanic rock. **d** Very fine-grained banded and

laminated felsic volcanic rock with skarn layers (green and brown). **e** Plagioclase-porphyritic, massive, felsic volcanic rock that is associated with amphibolites. **f** Mafic (metabasic) amphibolite with plagioclase porphyroclasts

showing values around + 8 and + 9‰ ($n = 9$), which is also within the range of igneous quartz reported in the literature and considerably below the values typical for hydrothermal quartz (cf. Taylor 1968; Larson and Taylor 1986; Blatt 1987; Chiba et al. 1989; Ghent and Valley 1998; Götze et al. 2001; Bindeman 2008; Jourdan et al. 2009; Budd et al. 2017; Kleine et al. 2018; Fig. 4). Equilibrium magma compositions have been calculated from the $\delta^{18}\text{O}$ values in quartz by applying the appropriate fractionation factors between quartz and andesite, dacite and rhyolite melts at the corresponding temperatures (1000 °C, 900 °C and 800 °C); ($\Delta_{\text{quartz-andesite}} = +0.91\text{‰}$,

$\Delta_{\text{quartz-dacite}} = +0.84\text{‰}$, $\Delta_{\text{quartz-rhyolite}} = +0.77\text{‰}$, Zhao and Zheng 2003). Grängesberg host rock quartz samples with $\delta^{18}\text{O} \geq +6.7\text{‰}$ ($n = 11$) yield equilibrium magma isotope compositions between $\delta^{18}\text{O} = +6.1$ and +9.5‰ (Table 2), with the majority of calculated magma isotope values falling between +7.0 and +8.5‰ ($n = 11$). This range corresponds to the measured whole rock data from the area (Table 1) and to values characteristic for intermediate- to high-silica igneous rocks in general (cf. Bindeman 2008). The calculation of equilibrium magmas for the spectrum of relevant rock types (magma compositions) that occur in the Grängesberg area

Fig. 3 Photomicrographs of major rock types in the vicinity of the Grängesberg iron oxide–apatite ore deposit. **a** Representative granitic gneiss. **b** Biotite-bearing, foliated fine-grained felsic metavolcanic rock. **c** Phyllosilicate-rich felsic metavolcanic rock, in this example with muscovite and phlogopite. **d** Layered and laminated

felsic metavolcanic rock with alternating fine-grained and very fine-grained layers. **e, f** Plagioclase–porphyric metavolcanic rock associated with amphibolite and larger plagioclase phenocrysts. **a–e** are in crossed polarised light, **f** is in plane-polarised light. The scale bar is 1000 µm

shows that little variation of magmatic values is in fact expected in the region, strengthening the coherence of our result of a magmatic origin of the majority of analysed quartz grains. A small number of quartz grains, however, have $\delta^{18}\text{O} \leq +6.7\text{‰}$ ($n=3$) and are not consistent with magmatic values. This may be because the quartz in these samples was disturbed by later infilling of microfractures

during medium- to high-temperature ($> 400\text{ °C}$) hydrothermal processes or that the separated quartz contained vein or amygdale quartz that had formed at relatively high temperatures from localized hydrothermal processes (cf. King et al. 1997; Zheng et al. 1999; Donoghue et al. 2010; Kleine et al. 2018).

Fig. 4 Alkali oxide variation diagram (K_2O+Na_2O vs $100 \times K_2O/(K_2O+Na_2O)$; Hughes 1973) for country rock samples to the Grängesberg ore body as determined by Persson et al. (2013). The country rocks show predominantly unaltered igneous character, although some samples are affected by contact metamorphic processes and some others show the effects of regional alkali alteration (see text for details)

Discussion

The $\delta^{18}O$ values for pristine intermediate to felsic volcanic rocks, such as arc-derived andesites and dacites, are typically between +6.0 and $\sim +8.0\%$ and are higher than primitive magmas derived from the depleted upper mantle ($5.7 \pm 0.3\%$), due to a combination of fractional crystallization and crustal contamination (Hoefs 1997; Eiler 2001; Bindeman 2008). During hydrothermal alteration of volcanic rocks, circulating water will lead to isotopic exchange with the rock (e.g. Taylor 1974). Since the infiltrating water in most hydrothermal systems will have had lower $\delta^{18}O$ values than the rock, water–rock interaction will lower the $\delta^{18}O$ in the rock, provided that the fractionation factor between rock and water is low enough (cf. Taylor 1971, 1974; Hoefs 1997; Faure and Mensing 2005; Donoghue et al. 2010; Kleine et al. 2018). In contrast, an increase in $\delta^{18}O$ of the rocks above magmatic values is usually caused by fluid rock interaction at low temperatures ($< 300^\circ C$), most often through the breakdown of original minerals and precipitation of secondary minerals (e.g. transformation of feldspar to clay minerals) that tend to have high $\delta^{18}O$, thus causing

Table 1 Country rock sample set Grängesberg iron oxide–apatite deposit, Central Sweden

Sample	Description	Coordinates RT90		Quartz $\delta^{18}O$ (‰)	2σ	Whole rock $\delta^{18}O$ (‰)	2σ
		East	North				
VRT-GMD29	Reddish-grey metagranite, foliated	1,454,958	6,662,272	7.9	± 0.2	–	–
VRT-GMD 31	Greyish-pink metagranite, foliated	1,456,585	6,661,782	6.7	± 0.2	–	–
VRT-GMD40	Greyish metagranite, foliated	1,456,305	6,661,990	9.3	± 0.2	–	–
VRT-GMD45	Greyish-pink biotite-rich metagranite, foliated	1,454,534	6,661,130	8.3	± 0.2	–	–
VRT-GMD48	Grey biotite-rich granitic gneiss, foliated	1,454,386	6,660,546	8.4	± 0.2	7.9	± 0.2
VRT-GMD49	Dark-grey granitic gneiss, foliated	1,455,395	6,661,477	9.2	± 0.2	7.8	± 0.2
VRT-GMD52	Grey-pinkish metagranite, foliated	1,456,209	6,662,922	8.7	± 0.2	6.9	± 0.2
VRT-GMD53	Pale pink-coloured metavolcanic rock, foliated	1,454,816	6,662,790	–	–	8.4	± 0.2
VRT-GMD56	Grey-pinkish granite, isotropic	1,455,284	6,661,802	–	–	7.4	± 0.2
KPN-090007A	Grey-reddish intermediary metavolcanic rock, foliated	1,454,596	6,663,236	–	–	5.8	± 0.2
KPN-090026–4	Greyish-pink felsic metavolcanic rock, foliated	1,453,806	6,662,229	9.1	± 0.2	6.8	± 0.2
KPN-090033A	Greyish-pink intermediary metavolcanic rock, foliated	1,453,794	6,662,943	10.3	± 0.2	8.6	± 0.2
KPN-090042A	Greyish-pink felsic metavolcanic rock, foliated	1,453,884	6,663,419	6	± 0.2	7.5	± 0.2
KHO-09003	Reddish-grey felsic metavolcanic rock with rounded magnetite, foliated	1,454,400	6,663,551	–	–	7.7	± 0.2
KHO-09005	White Na-altered felsic metavolcanic rock with magnetite disseminations, foliated	1,454,562	6,663,500	7	± 0.2	5.8	± 0.2
KHO-090010	Grey felsic metavolcanic rock, foliated	1,454,196	6,661,292	–	–	6.8	± 0.2
KHO-090011A	Grey fine-grained felsic metavolcanic rock, foliated	1,454,420	6,661,821	–	–	6.8	± 0.2
KHO-090012	Pale grey, fine-grained felsic metavolcanic rock, foliated	1,454,571	6,661,728	6.7	± 0.2	7.0	± 0.2
KHO-090013	Grey biotite-rich intermediary metavolcanic rock, foliated	1,454,467	6,661,972	–	–	7.7	± 0.2
KHO-090013G	Greyish-green amphibolite, metaigneous, foliated	1,454,473	6,661,971	–	–	6.0	± 0.2
KHO-090017	Pink fine-grained felsic metavolcanic rock, foliated	1,453,588	6,660,757	8.8	± 0.2	–	–
KHO-09127b	Grey felsic metavolcanic rock, foliated	1,454,632	6,662,214	–	–	8.3	± 0.2
KHO-09129A	Fine-grained granitoid metavolcanic rock	1,453,535	6,662,113	8.6	± 0.2	–	–

Fig. 5 **a** Distribution of surface samples used in this study around the Grängesberg ore body (the red ellipse) and their respective oxygen isotope composition. The area presented in Fig. 1b is indicated by the black box. **b** The positions of two traverses (T1 in E–W and T2

in N–S orientation) are indicated by yellow bars across the Grängesberg area, comprising the samples that are shown in Fig. 4. Notably, no major variation beyond the regular magmatic values is apparent (Source of base map: Google Earth)

Table 2 Results of the equilibrium magma re-calculation from quartz grains in country rocks to the Grängesberg ore body

Sample	Quartz $\delta^{18}\text{O}$ (‰)	Whole rock $\delta^{18}\text{O}$ (‰)	2σ	Re-calculated andesite $\delta^{18}\text{O}$ (‰)	Re-calculated dacite $\delta^{18}\text{O}$ (‰)	Re-calculated rhyolite $\delta^{18}\text{O}$ (‰)
VRT-GMD29	7.9	–	± 0.2	7.0	7.1	7.1
VRT-GMD 31	6.7	–	± 0.2	5.8	5.8	5.9
VRT-GMD40	9.3	–	± 0.2	8.4	8.4	8.5
VRT-GMD45	8.3	–	± 0.2	7.4	7.4	7.5
VRT-GMD48	8.4	7.9	± 0.2	7.5	7.6	7.6
VRT-GMD49	9.2	7.8	± 0.2	8.3	8.4	8.5
VRT-GMD52	8.7	6.9	± 0.2	7.8	7.9	8.0
VRT-GMD53	–	8.4	± 0.2	–	–	–
VRT-GMD56	–	7.4	± 0.2	–	–	–
KPN-090007	–	5.8	± 0.2	–	–	–
KPN-090026–4	9.1	6.8	± 0.2	8.2	8.3	8.3
KPN-090033A	10.3	8.6	± 0.2	9.4	9.5	9.5
KPN-090042A	6.0	7.5	± 0.2	5.1	5.2	5.2
KHO-09003	–	7.7	± 0.2	–	–	–
KHO-09005	7.0	5.8	± 0.2	6.1	6.2	6.2
KHO-090010	–	6.8	± 0.2	–	–	–
KHO-090011A	–	6.8	± 0.2	–	–	–
KHO-090012	6.7	7.0	± 0.2	5.8	5.9	5.9
KHO-090013	–	7.7	± 0.2	–	–	–
KHO-090013G	–	6.0	± 0.2	–	–	–
KHO-090017	8.8	–	± 0.2	7.9	8.0	8.0
KHO-09127b	–	8.3	± 0.2	–	–	–
KHO-09129A	8.6	–	± 0.2	7.6	7.7	7.8

Fractionation factors for equilibrium calculations were taken from Zhao and Zheng (2003)

$$1000\ln\alpha_{(\text{mt-andesite})} = +0.91\text{‰}$$

$$1000\ln\alpha_{(\text{mt-dacite})} = +0.84\text{‰}$$

$$1000\ln\alpha_{(\text{mt-rhyolite})} = +0.77\text{‰}$$

the opposite effect to medium to high-temperature hydrothermal overprinting (cf. Criss and Taylor 1986; Rhodes and Oreskes 1999; Faure and Mensing 2005; Donoghue et al. 2008, 2010; Berg et al. 2018). Therefore, if a hydrothermal origin (e.g. 300–500 °C) for the Grängesberg ore body would apply, as suggested for other ore deposits including some iron oxide–apatite ores (e.g. Taylor 1974; Parak 1975; Smith et al. 2013; Dare et al. 2014; Westhues et al. 2016, 2017), a widespread, systematic and characteristic low $\delta^{18}\text{O}$ alteration pattern in the host rocks in the vicinity of the ore would be expected as a result (cf. Taylor 1974; Beatty et al. 1988; Singleton and Criss 2002). The magnitude of these effects would depend on the $\delta^{18}\text{O}$ value of the input surface water, and the water/rock ratio, but the effect would be concentric variation of $\delta^{18}\text{O}$ around the focus of hydrothermal activity, especially since the palaeoenvironment is interpreted as shallow marine back-arc-setting, an at least in part open system with fluid influx of marine and/or meteoric isotopic compositions would be envisaged. Tornos et al. (2016) find that hydrothermal alteration at the El

Laco volcano in Chile involved fluids at temperatures around 900 °C, which would suggest a dominantly magmatic fluid ($\delta^{18}\text{O}$ usually 5–10‰; Hoefs 1997). Even if such a fluid is considered, hydrothermal fluids circulating in the host rock volume around the Grängesberg ore would experience distal changes in relation to the host rocks, due to progressive cooling of the magmatic fluids away from the ore body and consequent changes in fractionation behaviour. Moreover, progressive interaction between the percolating fluids and the host rocks would lead to a changing fluid composition and hence a notable isotope variation away from the ore body, thus, temperature changes and progressive fluid–rock interaction ought to leave a detectable variation in the aureole surrounding a large hydrothermal ore deposit (e.g. Taylor 1974; Nabelek 1987; Berg et al. 2018). In contrast, low-temperature hydrothermal fluids that circulate through various host rocks (e.g. emanating from a medium temperature hydrothermal ore deposit) may be representing the distal effects of a high-temperature hydrothermal system, and will lead to an increase in whole rock $\delta^{18}\text{O}$ of the host rocks

due to a reversal of fractionation factors (e.g. Taylor 1974; Donoghue et al. 2008; Berg et al. 2018). In such a case, an increase in $\delta^{18}\text{O}$ values at distance from the ore body would be expected if fluid circulation was an important process. In the Grängesberg case, we consider host rocks in the tectono-stratigraphic hanging wall to the east of the Grängesberg ore body to be possibly younger than the ore and thus to possibly reflect a pristine original composition or regional pattern unrelated to the ore formation, while a potential offset pattern would be expected to the west of the Export Field in case hydrothermal transport and deposition of metals was the main ore-forming process. There are, however, no distinct difference between the western and eastern part in the $\delta^{18}\text{O}$ distribution around the ore body and the $\delta^{18}\text{O}$ values do not show any systematic patterns for either the whole rock or quartz data with regard to geographical distribution (Figs. 4, 5). Indeed, there is very little variation beyond a regular range of essentially magmatic values (+6.0 to $\sim +8.0\text{‰}$; Fig. 5) and, furthermore, there is no real variation from this scheme in whole rock $\delta^{18}\text{O}$ data presented by Jonsson et al. (2013) for host rocks sampled from drill cores (from 570 and 650 m depth) through the Grängesberg ore zone. These drill core samples also range from +5 to +9‰, with the majority of values between +6 to +7‰, which are thus consistent with the predominantly magmatic isotope composition derived from our surface survey. Furthermore, we note that calculated equilibrium magma compositions for ore magnetite from Grängesberg also yield calculated equilibrium magma $\delta^{18}\text{O}$ values of between +6 and +8‰, show, moreover, magmatic Fe isotope values (Jonsson et al. 2013; Troll et al. 2019), and thus align with the magmatic range of the host rock data. These values are also consistent with the theoretical equilibrium magma compositions for our quartz samples, which show calculated $\delta^{18}\text{O}$ magma values between +6.1 and +9.5‰, even when applying fractionation factors for the whole spectrum of intermediate to felsic magmas available in the entire Grängesberg region (Fig. 5, Table 2). The results are thus consistent with a dominantly magmatic mode of origin for the country rocks and the Grängesberg iron ore deposit.

The quartz phenocrysts ($n = 14$) in the host rocks to the Grängesberg ore have an average $\delta^{18}\text{O}$ value of +8.2‰, a value representative of relatively normal igneous quartz (e.g. Larson and Taylor 1986; Blatt 1987; Ghent and Valley 1998; Bindeman 2008; Budd et al. 2017). Even though quartz is often seen as being relatively resistant to hydrothermal alteration, as it exchanges oxygen isotopes very slowly with hydrothermal fluids, some potential alteration can be seen in the Grängesberg samples. The quartz samples which exhibit $\delta^{18}\text{O}$ values below 7‰ ($n = 3$) were most likely influenced by localized high-temperature fluid–rock interaction (cf. Donoghue et al. 2010; Kleine et al. 2018) that would be expected for a high-temperature magmatic setting, or

Fig. 6 The oxygen isotope data of whole rocks, quartz samples and calculated equilibrium magmas for quartz along the profiles of traverse 1 (a) and traverse 2 (b). Indicated by the red and black bars, respectively are the ranges of $\delta^{18}\text{O}$ values for the host rocks and calculated equilibrium magmas from magnetite derived from drill cores through the main ore body (drill core data from Jonsson et al. 2013). The ore zone is indicated in both panels by the hatched box in each of the traverses. The established ranges for common intermediate magmatic rocks and for magmatic quartz are shown for reference by the orange horizontal areas. Overall, the data fall into the range of established magmatic I-type (igneous) values and deviations beyond this range are not common in this study area

possibly by fluid circulation during regional metamorphism. Our whole rock $\delta^{18}\text{O}$ data are almost exclusively within the range of common igneous rocks (Fig. 5) and only two whole rock samples from the northern part of the Grängesberg area, one intermediate and one felsic metavolcanic rock, exhibit a $\delta^{18}\text{O}$ value of 5.8‰, and are thus at the lower end of the typical range for intermediate magmas of +6 to 8‰ (Fig. 6). One of these two outlier samples is rich in magnetite disseminations, which will lower the $\delta^{18}\text{O}$, while the other appears to record the effect of localized alteration with a

mineral replacement texture visible on inspection. These rare alteration phenomena seen in the wider array of host rocks to the Grängesberg ore body contrast the hydrous alteration assemblages, such as the biotite, chlorite or amphibole-rich assemblages that are present within the Grängesberg ore body and its direct contact zone, and which are by now interpreted to be directly associated with the emplacement of the ore body (Jonsson et al. 2013; Troll et al. 2019). In summary, apart from rare and localized exceptions, our data do not exhibit any major offset of $\delta^{18}\text{O}$ values from the regional average in the wider range of host rocks around the main iron oxide–apatite ore zone at Grängesberg, which implies that a large-scale hydrothermal circulation of external fluid into the host rocks of the Grängesberg ore body was likely not taking place during the ore-forming event. Therefore, the magmatic $\delta^{18}\text{O}$ values recorded in the host rocks are more consistent with an igneous rather than a hydrothermal origin of the main ore body and are in line with the magmatic $\delta^{18}\text{O}$ and $\delta^{56}\text{Fe}$ values from massive magnetite ore samples from drill core material in the main Grängesberg ore zone (Jonsson et al. 2013; Troll et al. 2019).

Conclusions

Quartz and whole rock isotope data from host rocks to the Grängesberg iron oxide–apatite ore do not show any systematic offset relative to normal magmatic values, and no distinct isotopic variation is observed on either side of the main ore zone (the The Export field) at Grängesberg. In particular the footwall rocks of the ores that are older or synchronous with ore formation do not seem to have been affected by any intense hydrothermal overprinting, as would be expected if this very large iron oxide ore body had formed from dominantly hydrothermal processes. A long-time, steady-state hydrothermal system capable of forming this type of large-scale deposit, such as sub-sea-floor replacements or seafloor exhalations, for example, would generate a spatially widespread offset of primary isotope values, which is not the case here. Lower than ‘normal’ magmatic $\delta^{18}\text{O}$ values are observed in a very small number of country rock samples only, and when present, they are from locations relatively close to the ore body. The absence of a large-scale and marked $\delta^{18}\text{O}$ “hydrothermal aureole” to the Grängesberg ore, coupled with the broadly magmatic stable isotope values of the host rocks and the complementary magmatic data from the centre of the Grängesberg ore body, shows that there is a) no significant variation across the area and b) that there is a broadly similar magmatic signal preserved in the ore as well as in the country rocks. The new $\delta^{18}\text{O}$ country rock data from around the Grängesberg ore thus corroborate the

interpretation of a mainly ortho-magmatic origin of the Kiruna-type iron oxide–apatite ore deposit at Grängesberg.

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