Magmatic origin of giant ‘Kiruna-type’ apatite-iron-oxide ores in Central Sweden

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Iron is the most important metal for modern industry and Sweden is by far the largest iron-producer in Europe, yet the genesis of Sweden’s main iron-source, the ‘Kiruna-type’ apatite-iron-oxide ores, remains enigmatic. We show that magnetites from the largest central Swedish ‘Kiruna-type’ deposit at Grängesberg have δ18O values between 0.4 and 3.7‰, while the 1.90–1.88 Ga meta-volcanic host rocks have δ18O values between 4.9 and 9‰. Over 90% of the magnetite data are consistent with direct precipitation from intermediate to felsic magmas or magmatic fluids at high-temperature (δ18Omag > +0.9‰, i.e. ortho-magmatic). A smaller group of magnetites (δ18Omag ≤ +0.9‰), in turn, equilibrated with high-δ18O, likely meteoric, hydrothermal fluids at low temperatures. The central Swedish ‘Kiruna-type’ ores thus formed dominantly through magmatic iron-oxide precipitation within a larger volcanic superstructure, while local hydrothermal activity resulted from low-temperature fluid circulation in the shallower parts of this system.

Sweden is by far the biggest producer of iron in Europe1,2 and iron ore has traditionally been sourced from two principal regions, the Kiruna-Malmberget province in northern Sweden and the Bergslagen region in central Sweden3 (Fig. 1). The apatite-iron-oxide ores of the Grängesberg Mining District (GMD) represents the largest iron ore accumulation in the classic Bergslagen ore province4, with a past production of 156 Mt of ore, averaging 60% Fe and 0.81% P. Besides iron, phosphates such as fluorapatite and REE-silicates constitute an economically significant reserve of REE5. The apatite-iron-oxide deposits of the GMD and its northern continuation are classified to be of ‘Kiruna-type’, whereas thousands of smaller, but still significant, iron-oxide deposits of banded-iron and skarn-iron types also occur in the Bergslagen province6,7. The GMD deposits themselves, are thus traditionally grouped together with those of the Kiruna-Malmberget region6,8 (i.e., the type locality of the internationally renowned but genetically ambiguous ‘Kiruna-type’ ores). The origin of this ore-type has been a matter of substantial debate for over 100 years, and several fundamentally different modes of formation have been suggested. These include direct magmatic segregation or crystallization, magmatic hydrothermal replacement, and hydrothermal precipitation in the sense of iron-oxide-copper-gold (IOCG-type) deposits9–15. Here we address the primary question, using oxygen isotopes, whether the GMD iron oxides formed through direct magmatic precipitation from either a magma or from high-temperature magmatic fluids (≥ 800°C), likely leading to intrusive-style, massive iron ores, or whether they formed through precipitation from aqueous hydrothermal fluids at lower temperatures (e.g., ≤ 400°C), that would likely form disseminations and vein-type magnetites.

Host rocks to the GMD ores comprise mainly intermediate to felsic meta-volcanic rocks that formed between ~1.90 and 1.88 Ga in a subduction- or back-arc-related tectonic setting. Both the apatite-iron-oxide ores and the meta-volcanic host rocks have been affected by later ductile deformation and the 1.85 to 1.80 Ga Svecokarelian low- to medium-grade metamorphism16. The exact timing of ore formation in the GMD is not known, but a major regionally-deformed granite body (> 1.85 Ga) occurs directly east of the main Grängesberg ore field and post-dates the ores (Fig. 1), because it carries characteristic metre-sized xenoliths of apatite-iron-oxide mineralization. In most parts of the Bergslagen ore province, the host rocks to the ores are SiO2-rich dacites to rhyolites. In contrast, the host rocks to the GMD ores exhibit more intermediate compositions with a dominance of
Figure 1 | (A). Overview map of Fennoscandia with the Grängesberg mining district (GMD), the Bergslagen province, and the Kiruna-Malmberget mining district indicated. (B). Geological map over the main ore zone in the GMD. (C). Vertical section (line X–Y in panel B) through the main ore body of the GMD. The ore zone extends downwards at a 70–80° dip to the SE. Black horizontal lines are adits. Modified from Geological Survey of Sweden (SGU) datasets and A. Hallberg, SGU.

Figure 2 | Chondrite-normalised REE diagram of host rocks and iron-oxide ores from drill cores 717 and 690. Chondrite values for normalisation after Sun & McDonough. The iron-oxide ores and the moderately altered host rocks share similar REE concentrations and patterns with the regional bedrock and recent subduction zone volcanic rocks. The hydrous altered host rocks (marked with hatched pattern), in turn, are enriched in REE relative to moderately altered host rocks and recent subduction zone volcanic rocks.
andesites and dacites. The major ore body itself consists of several lenses that occur along a planar NNE-trending zone with a moderate to steep eastward dip. The principal orientation of the major ore body is stratiform, i.e. it is parallel to the internal contacts within the meta-volcanic host rocks that themselves are sub-parallel to the main tectonic trend in the region. Magnetite occurs either in massive ore lenses or as veins, bands or disseminations (hereafter referred to as VeDi-magnetites). Within the central part of the GMD ore field, a number of dykes and sills exhibit cross-cutting relationships to rock units of similar composition, on which basis we interpret the

| Sample | Meters along drillcore (m) | Sample description | Whole Rock $\delta^{18}$O (%) | Quartz $\delta^{18}$O (%) | Magnetite $\delta^{18}$O (%) |
|--------|---------------------------|--------------------|-------------------------------|-------------------------|---------------------------|
| Drillcore 690 |
| KES090003 | 7.8 | Volcanic host rock (meta-dacite) | 6.2 | 7.4 | — |
| KES090004 | 11.5 | Volcanic host rock with hydrous alteration assemblage | 7.0 | — | — |
| KES090007 | 23.6 | Volcanic host rock (meta-dacite) | 5.2 | — | — |
| KES090008A | 23.6 | Massive magnetite ore | — | — | 1.9 |
| KES090009 | 29.5 | Massive magnetite ore | 6.0 | — | 2.2 |
| KES090011 | 31.4 | Massive magnetite ore | — | — | 2.8 |
| KES090012 | 26.1 | Massive magnetite ore | — | — | 1.2 |
| KES090014A | 32.2 | Volcanic host rock (meta-rhyolite) | 9.0 | — | — |
| KES090020 | 55.5 | Massive magnetite ore | — | — | 1.1 |
| KES090024 | 63.7 | Massive magnetite ore | — | — | 1.0 |
| KES090027 | 78.7 | Massive magnetite ore | — | — | 1.2 |
| KES090030 | 88.8 | Massive magnetite ore | — | — | 1.8 |
| KES090037 | 120.4 | Massive magnetite ore | — | — | 1.4 |
| KES090039 | 122.4 | Volcanic host rock with hydrous alteration assemblage | — | — | 3.7 |
| KES090040 | 123.7 | Volcanic host rock with hydrous alteration assemblage | 5.5 | — | — |
| KES090044 | 141.1 | Volcanic host rock (meta-andesite) | 6.1 | — | —0.4 |
| KES090045 | 139.5 | Volcanic host rock with hydrous alteration assemblage | 6.5 | — | — |

Grängesberg oxygen isotope data, including sample position along the drillcore profile

| Sample | Meters along drillcore (m) | Sample description | Whole Rock $\delta^{18}$O (%) | Quartz $\delta^{18}$O (%) | Magnetite $\delta^{18}$O (%) |
|--------|---------------------------|--------------------|-------------------------------|-------------------------|---------------------------|
| Drillcore 717 |
| KES090048 | 33.8 | Volcanic host rock with hydrous alteration assemblage | 6.9 | 8.0 | — |
| KES090054 | 55.9 | Volcanic host rock with hydrous alteration assemblage | 7.1 | — | — |
| KES090056 | 59.8 | Volcanic host rock (meta-dacite) | 6.9 | — | — |
| KES090059 | 84.3 | Volcanic host rock (meta-dacite) | 6.6 | — | — |
| KES090061 | 117.3 | Volcanic host rock with hydrous alteration assemblage | 6.2 | — | — |
| KES090065 | 147.9 | Volcanic host rock with magnetite veins | 4.9 | 5.8 | 1.3 |

Grängesberg oxygen isotope data, including sample position along the drillcore profile

| Sample | Meters along drillcore (m) | Sample description | Whole Rock $\delta^{18}$O (%) | Quartz $\delta^{18}$O (%) | Magnetite $\delta^{18}$O (%) |
|--------|---------------------------|--------------------|-------------------------------|-------------------------|---------------------------|
| Drillcore 575 |
| KES103011 | 89.0 | Massive magnetite ore | — | — | 1.2 |
| KES103016 | 170.0 | Massive magnetite ore | — | — | 1.5 |

Surface Samples

| Sample | Sample description | Whole Rock $\delta^{18}$O (%) | Quartz $\delta^{18}$O (%) | Magnetite $\delta^{18}$O (%) |
|--------|--------------------|-------------------------------|-------------------------|---------------------------|
| KHN010012 | Volcanic host rock (meta-dacite) | 7.0 | 6.7 | — |
| KPN090033B | Volcanic host rock (meta-andesite) | 5.6 | 6.2 | — |
| KPN090026A | Volcanic host rock (meta-dacite) | 6.8 | 9.1 | — |
| KHN00058a | Volcanic host rock (meta-dacite), magnetite-rich | 5.8 | 7.1 | — |
| KPN090042A | Volcanic host rock (meta-dacite) | 7.5 | 6.0 | — |
| KHN0005 | Volcanic host rock (meta-dacite) | 5.8 | 7.0 | — |
| KPN090033A | Volcanic host rock (meta-dacite) | 8.6 | 10.3 | — |
emplacement environment as "shallow sub-volcanic" in character. Hydrous alteration is evident in the direct host rocks to the ore in the form of disseminated and sometimes discrete biotite, chlorite or variably amphibole- and fluorapatite-rich assemblages. Massive magnetite ores seem unaffected by this alteration or by the regional metamorphism. However, the hydrous alteration assemblages are always spatially related with the massive ore and are absent beyond the main ore zone, implying a genetic relationship between the two.

Results

Samples were collected from surface exposures and from three shallow plunging (≤ 20°) drill cores that transect the steeply dipping ore body between −570 and −670 m (Fig. 1). The GMD apatite-iron-oxide ore is either magnetite- or hematite-dominated, and is variably enriched in fluorapatite and accessory silicate phases (amphiboles, biotite, chlorite sensu lato) and REE-minerals. Most ore-types show some degree of banding, defined by aggregates of mostly fine-grained fluorapatite and minor amounts of silicates. The main GMD ore field, locally known as the 'Export Field', is estimated to consist of about 80% magnetite and 20% hematite ore, with a concentration of the latter near the structural footwall.

In addition to fluorapatite, REEs are hosted in associated monazite-(Ce), allanite-(Ce), xenotime-(Y), LREE-bearing epidote, and REE fluorocarbonates. Although the GMD apatite-iron-oxide ores

Figure 3 | Parts (A) and (B) show the oxygen isotope data for two drill cores (Numbers 690 and 717) that traverse the main ore zone at Grängesberg between 570 and 670 m below the surface (see Fig. 1). Shown are the oxygen isotope compositions of the host rocks, quartz separates, massive magnetites and VeDi-magnetites, the latter including magnetite from hematite ore. All oxygen data are reported in standard δ18O-notation relative to SMOW after Hoefs34. The δ18O ranges for the mantle and arc-andesites are after Bindemann18 and Taylor19. Range of igneous magnetites after Taylor35.
are typically enriched in Th, U, La, Ce, Nd, P, Fe, Sm, Tb, Y, Tm and Yb, they are depleted in K, Ba, Sr, Zr and Ti relative to average continental crust. The ores themselves, and particularly theapatite-rich samples and the hydrous host rock alteration assemblages, display negative Eu-anomalies and flat HREE patterns. The hydrous magnetite-rich samples and the hydrous host rock alteration assemblages, continental crust. The ores themselves, and particularly the apatite-rich assemblages associated with the ore show the same REE enrichment patterns as moderately altered host rocks, but are elevated by several orders of magnitude when compared with the meta-volcanic rocks in the region and with typical subduction zone andesites and rhyolites (Fig. 2).

To distinguish between magmatic and magmatic-hydrothermal origins of magnetite, which together we term “ortho-magmatic”, is not possible with our data. However, a low-temperature hydrothermal origin (≈400°C) versus a high-T ortho-magmatic origin can be discerned. Oxygen isotope ratios have been measured on 63 samples of meta-volcanic host rocks, hydrothermal alteration-assemblages, quartz mineral separates, as well as magnetite from massive ore, veins, and disseminations, and from hematite-dominated ore. All data are reported in delta notation relative to SMOW (Table 1). The host rock δ18O values, including whole rocks and quartz mineral separates from both outcrops and drill cores, range from +4.9 to +10.3‰, i.e. within the spectrum of most normal igneous rock compositions. The massive iron-ores from the main ore body yield δ18O values, between +0.9 and +3.0‰ [n = 16] (Fig. 3). The δ18O values of all GMD magnetite samples, including vein and disseminated magnetite types [n = 7] as well as magnetite inclusions in hematite ore [n = 2] range from −0.4 to +3.7‰. The origin of the different magnetite types can be assessed using O-isotope fractionation between magmatic and an andesite parent magma at magmatic temperatures (≥900°C) (Δmagnetite–andesite = −4.0‰) or with magmatic water (Δmagnetite–water = −5.2‰ at 800°C)16,17. Magnetites with δ18O ≥ +1.7‰ yield equilibrium magma values between +5.7 and +7.7‰ (Table 2), i.e. within the range of subduction zone andesite magmas (+5.7 to +8.5‰)18,19. Using the fractionation factor for magnetite from a dacite magma (Δmagnetite–dacite = −4.3‰)20, the theoretical magma values would shift upwards by +0.3‰, moving them further into the intermediate magmatic range that is characteristic for continental subduction zone rocks16,17 (Figs. 3, 4, Table 2). These results are compatible with our andesite to dacite host rock data that range between +4.9 and +9.0‰ (Table 1). In turn, magnetite samples with δ18O values ≥ +0.9‰ satisfy equilibrium conditions with high-temperature magmatic waters of +6.1 to +8.9‰ (Table 2), which corresponds to the range of common magmatic waters (6–8‰)16. Thus, all GMD magnetites with δ18O values ≥ +0.9‰ (>90% of our samples) are consistent with an “ortho-magmatic” mode of formation because they satisfy high-temperature magmatic equilibrium conditions (pale pink box in Fig. 4). Notably, this field hosts all GMD massive magnetite ore samples, as well as several VeDi samples. Magnetites with δ18O values below +0.9‰, in turn, cannot be in equilibrium with a regular magma or with a high-temperature magmatic fluid, but must have formed from a high-δ18O hydrothermal fluid at more moderate temperatures. Low-temperature processes increase mineral-fluid fractionation, i.e. fractionation factors become more negative (e.g. Δmagnetite-water = −7.6‰ at T ≤ 400°C)21, implying oxygen isotope exchange at low temperatures for the magnetites ≤ 0.9‰. A high-δ18O hydrothermal fluid from the uppermost range of meteoric fluids22, or a mix of remnant magmatic water and a meteoric influx, can explain the remaining VeDi magnetites (n = 2), while satisfying the low-temperature fractionation conditions (Fig. 4).

Employing a similar approach on host rock quartz, a mineral that is assumed to be resistant to oxygen isotope resetting once formed, should allow us to test the results derived from the magnetite data. Quartzes have δ18O values between +5.8 and +10.3‰.
Magnetite in isotopic equilibrium with rhyolite magma of 10‰ (5.3 ±/− 0.3‰)

**Figure 4 | Magnetite δ18O values from GMD compared to other volcanically-hosted iron ore deposits.** For reference, magnetites in equilibrium with MORB (red box), the range for typical 'ortho-magmatic' magnetites after Taylor (pale pink box) and magnetite in equilibrium with an evolved rhyolite with a δ18O of 10‰ (the demarcation between I-type (<10‰) and S-type (>10‰) magmas) are shown. The GMD magnetites plot dominantly above the +0.9‰ demarcation and in the field of 'ortho-magmatic' magnetites after Taylor, and satisfy equilibrium with magma or magmatic fluids at magmatic temperatures (~800–1000°C). A small fraction of the GMD data (n = 2), however, is more consistent with formation from a low-temperature fluid regime. The cut-off point for this is calculated to be +0.9‰ in magnetites, because fractionation factors determine that samples < +0.9‰ cannot be in equilibrium with either a magma or a magmatic fluid at high temperatures (≥800°C). Magnetites with values lower than +0.9‰ are calculated to have been in equilibrium with a high-δ18O (likely meteoric) fluid at temperatures of ≤400°C.

(n = 11, Table 3). Taking Δ_{qtz−magma} = +0.9‰ and +0.8‰, as fractionation between quartz and andesite and quartz and dacite magmas respectively, δ18O melt values of +4.9 to +9.5‰ are derived (n = 22, Table 3). Seven out of 11 quartz δ18O values correspond to calculated magmatic values for andesites and dacites with δ18O ≥ +5.7‰. A calculated fractionation temperature for a co-existing quartz and magnetite pair for the GMD deposit that appears in textural and oxygen-isotope equilibrium yields a temperature of 907 ± 53°C, consistent with temperatures expected for oxide formation from intermediate to evolved (hylous) arc magmas and consistent with phase relations and experimental constraints9,10,13,12. Quartz with δ18O ≤ +6.2‰ (n = 4) would be in equilibrium with a magmatic fluid at temperatures between 800°C and 900°C (Δ_{qtz−water} = +0.3 to −0.1‰), which implies that these quartzes are of ortho-magmatic origin too (following our initial definition).

**Discussion**

The relatively wide range of δ18O in the GMD host rocks (+4.9 to +9.0‰, Table 1) suggests that some host rock and VeDi-samples reflect low- and high-temperature alteration processes (e.g. silicate whole rocks with δ18O > 8.5‰ and < 5.7‰ respectively). Massive magnetite ores, in turn, show textural characteristics and δ18O values that support precipitation from a magma. Those massive magnetites are chemically and mechanically refractory and appear little affected by later overprint (i.e., they were resistant to subsequent exchange with fluids or there were insufficient fluids to exchange with). In addition, the temperature and oxygen isotope data derived for the refractory GMD quartz and magnetite samples fall in the magmatic range also and overlap with those presented in Nystroöm et al. for the iconic El Laco apatite-iron-oxide deposit in Chile (~700–800°C). Discussion is, however, ongoing regarding the genesis of the El Laco deposit, and the possibility of interaction with a high-δ18O hydrothermal fluid exists because the data for most all textural magnetite types at El Laco average around +4‰. These authors argue that a 'natural' spread between “magmatic” and “low-temperature hydrothermal” signals would be expected for a truly “magmatic” formation because a large volcano would logically develop an associated hydrothermal system at shallower levels. Although maybe not observed at El Laco, the GMD data show a small group of low-temperature magnetites, making our proposed dominantly ortho-magmatic formation scenario rather probable. The potential wider significance of this result is further underlined by the fact that our GMD data encompass the available δ18O-data from Kiruna, the largest apatite-iron-oxide deposit in Europe, suggesting very similar modes of formation for the GMD and Kiruna deposits (Fig. 4). On the basis of the data presented, the massive apatite-iron-oxide ores at GMD formed in equilibrium with mafic to felsic magmas at magmatic temperatures (i.e., probably with subduction-type andesites to dacites, which form the host rocks to the ores in the region).
The hydrous alteration assemblages, in turn, have highly variable $\delta^{18}O$ values, but their REE patterns are very similar to that of the ores. In combination with their exclusive occurrence close to the ore zone, this observation suggests a genetic relationship between iron-oxide ores and the REE-enrichment in hydrous alteration assemblages. Medium- to low-temperature hydrothermal alteration associated with ore formation and emplacement is consistent with a scenario of high-temperature oxide-rich magmas intruding an upper crustal volcano-sedimentary pile where they initiate fluid circulation at more moderate temperatures. At GMD, volcaniclastic host rocks are locally extensively brecciated as well as crosscut by dykes and other sheet intrusions, which in combination with our REE and oxygen isotope data supports a shallow-level sub-volcanic environment. The VeDi-type magnetites and hydrous alteration assemblages formed in proximity to the massive ore bodies, and a degree of overlap between these regimes (i.e., high-temperature ortho-magmatic versus low-temperature hydrothermal), is expected for a sub-volcanic mode of formation. Our preferred interpretation is therefore that of a new deformed, originally large and possibly of caldera-type, volcanic complex situated in a subduction zone regime. Ascent, intrusion, and crystallization of oxide-rich melts to shallow levels within the volcanic plumbing system produced dominantly ortho-magmatic magnetite, while low-temperature hydrothermal activity near the surface occurred to some extent. This late hydrothermal activity likely involved exsolved magmatic volatiles as well as external meteoric waters that probably became available to the host system at shallower levels and at progressively lower temperatures. This realisation helps us to better understand the formation of Europe’s most important iron source, the ‘Kiruna-type’ apatite-iron oxide ores of central and northern Sweden.

### Methods

**REE analysis**. A total of 44 drill core samples including both, moderately to intensely and hydrous altered host rocks (9 and 15 samples, respectively) and sections from the iron oxide ore (20 samples) were analysed for Rare Earth Elements (REE) at Acme Analytical Labs Ltd in Vancouver, Canada. Trace and rare earth elements were analysed by inductively coupled plasma-mass spectrometry (ICP-MS) after preparation by multi-acid digestion. Loss on ignition (i.e. volatile content) was determined by igniting a sample split and then measuring weight loss. Data quality was monitored using a set of certified, internal reference materials. Detailed information on uncertainties and reproducibility can be found at [http://acmealab.com/services/method-descriptions/soil-till-and-sediment/](http://acmealab.com/services/method-descriptions/soil-till-and-sediment/).

**Oxygen Isotopes**. All O-isotope data presented in this paper were produced at the University of Cape Town (UCT). For oxygen isotopes, both conventional and laser fluorination methods were used. Some quartz, and all magnetite, separates were analysed using a conventional silicic line (described in Harris & Ashwal 1982). Approximately 20 mg (magnetite) and 10 mg (quartz) of sample was reacted with Cl$_2$F$_2$, and the liberated O$_2$ converted to CO$_2$ using a hot platinum carbon rod. Magnetite was reacted overnight at 600°C and all other minerals were reacted for 4 hours at 550°C. Some quartz and magnetite separates were analysed using the laser fluorination analytical process described in Harris & Vogeli 1983. Each sample was reacted in the presence of approximately 10 kPa BrF$_3$ and the purified O$_2$ was collected onto a 5Å molecular sieve contained in a glass storage bottle. All isotope ratios were measured off-line using a Finnigan Delta XP mass spectrometer in dual-inlet mode. All data are reported in 6 notation where $\delta^{18}O = (R_{\text{sample}}/R_{\text{standard}} - 1) \times 1000$, and $R$ is the measured ratio (i.e. $^{18}O/^{16}O$ or D/H). Duplicate splits of the quartz standard (NBS28) run with each batch of eight samples were used to convert the raw data to the SMOW scale using the $^{18}O$ value of 5.4% for NBS28 recommended by Coplen et al. 1991. During the course of this work, 8 analyses of NBS28 gave a 2σ error of 0.2. The O-isotope ratios of samples analysed using laser fluorination were measured on O$_2$ gas. Measured values of our internal standard MON GT were used to normalise raw data and correct for drift in the reference gas. The average difference in $^{18}O$ values of duplicates of MON GT analysed during this study was 0.1‰, and corresponds to a 2σ error of 0.2‰. MON GT was recalibrated against the UWG-2 garnet standard of Valley et al. 1993 using the current laser system, and has a revised $^{18}O$ value of 5.4‰, assuming a $^{18}O$ value of 5.8‰ for UWG2.

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Author contributions
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Additional information
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