Testing the plagioclase discriminator on the GEOROC database to identify porphyry-fertile magmatic systems in Japan

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Abstract: Despite showing favourable geology, there are no known porphyry-type deposits in Japan. This, therefore, provides an ideal porphyry-'barren' case area on which to test the recently developed plagioclase-based discriminator for porphyry-'fertile' calc-alkaline systems. The discriminator was applied to plagioclase data for Japan in the online GEOROC database for convergent margin settings. Of the 3933 data points available, and considered reliable, 91% fall in the 'barren' field which compares with 73% for equivalent data worldwide. 'Fertile' signatures in certain parts of Kyushu and central Honshu should be further investigated, as well as areas where there are magnetite-series magmatic systems with elevated whole-rock Sr/Y, but for which there are currently little or no plagioclase data (SW-, N-central- and NE-Honshu). Why the Japanese arcs appear to show anomalously poor potential for porphyry deposits compared with other arcs worldwide, including Western Luzon, is as yet unclear, but possibly relates to tectonic factors or the nature of the magmatic source, or because melt PH_2O in many of the magmatic systems was too low, as suggested from low average excess Al in plagioclase.

Keywords: Porphyry deposits, plagioclase discriminator, excess aluminium, Japan

1. Introduction

Japan has the world's third largest gross domestic product (GDP), despite having extremely limited mineral resources. The mining sector accounted for <0.2% of GDP between 1994 and 2013 (Wacaster, 2014). Of Japan's average annual refined copper consumption of around 1.5 Mt, none since 2007 has been derived from domestic ore. Its copper and copper ore is therefore imported, at a high cost, or produced from scrap or unclassified materials (2004 to 2014 data, Wacaster, 2014, and references therein). The discovery of large domestic copper deposits would be of major economic significance, if environmental restrictions allow their exploitation.

Copper mining in Japan has traditionally been from Besshi- and Kuroko-type volcanogenic massive sulphide, polymetallic and skarn-type Pb-Zn-Cu deposits which are currently either worked-out or uneconomic. However, most copper mined worldwide (around 75%, Sillitoe, 2010) is from porphyry copper deposits (PCD) which are relatively high tonnage, but often low grade, and are usually amenable to low cost bulk mining techniques. Japan has generally favourable geological conditions for PCD, at least in certain areas, however none have so far been discovered (e.g. Imai, 2004). Indications that they may exist include the presence of a few minor examples of porphyry-style molybdenum mineralisation, and numerous high-sulfidation and Pb-Zn-Cu vein and skarn deposits (Qin & Ishihara, 1998, and references therein). None of the magmatic systems, however, show porphyry-type patterns of alteration and mineralisation, there being only rare examples of potassic alteration (Ishihara, 1980).

The reasons for the lack of PCD in Japan have long been debated. Current thinking is mainly around the ideas that: 1) the presence of Kuroko-type deposits indicates an extensional back arc setting rather than the compressional regime usually required for the development of PCD (Uyeda & Nishiwaki, 1980); 2) extensive Meso-Cenozoic cauldron subsidence/ignimbrite eruptions, particularly in SW Japan, 'dissipated' base metals and sulphur (Sillitoe, 1980); 3) there is a low proportion (<50%) of magnetite-series intrusions and an incomplete zonation of mineralisation due to the narrow width of the Japanese archipelago (Qin & Ishihara, 1998); 4) the upper mantle and/or lower crust was relatively reducing compared with porphyry-'fertile' arcs, as evidenced from a lack of anhydrite phenocrysts in Japanese magmatic systems, which is likely to have favoured copper partitioning into sulphides at depth rather than into residual melts to form PCD (Sato, 2012). Despite having discussed many of these reasons for the lack of PCD in Japan, both Qin and Ishihara (1998) and Hammarstrom *et al.* (2014) concede that they may well be present at depth (>1 km), possibly buried beneath thick sequences of volcanic rocks.

Since the 1990s, there have been major advances in our understanding of PCD formation, but few new investigations in Japan. The major exception to this is a study by Imai (2004) who showed that magmatic rocks from certain areas of Japan show similarities to those which host PCD in other parts of the western Pacific, specifically in the chemistry of apatite, an increasingly studied PCD indicator mineral.

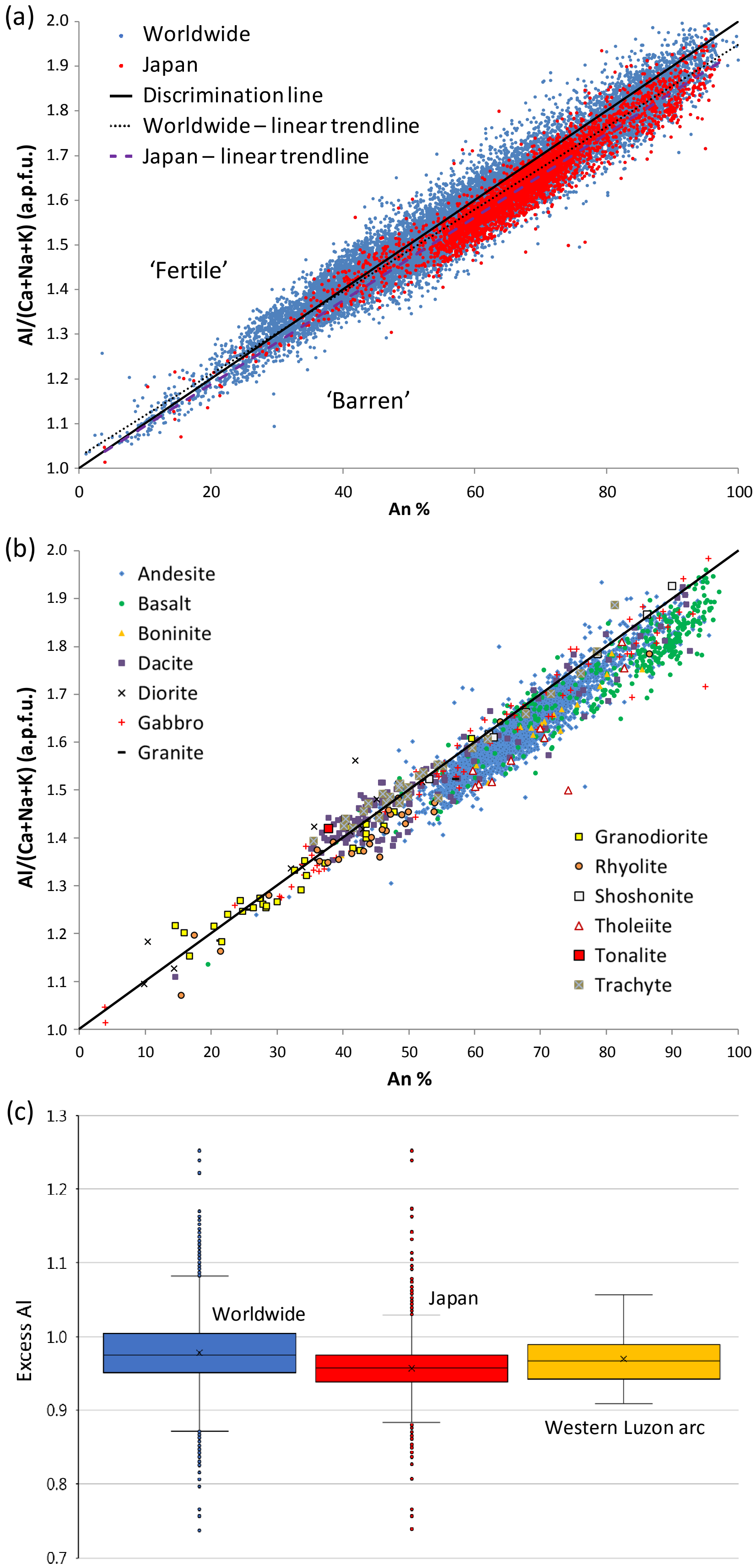

Fig. 1a) Discrimination diagram for plagioclase data from Japan (n = 3933) and worldwide (n = 18477) from the GEOROC database, 'convergent margin' dataset. Solid black line discriminates 'fertile' (plagioclase with excess Al above line) from 'barren' magmatic systems (Williamson et al., 2016); b) Discrimination diagram as in a) but for different rock types (based on 'ROCK NAME' in the GEOROC database) from Japan; c) Box and whiskers plot for plagioclase from the GEOROC database for 'convergent margin' settings: Blue is data worldwide (n = 18477); red is data from Japan (n = 3933); yellow is for the Western Luzon arc (n = 108). The line through the centre of the coloured 'box' is the median, and the cross is the mean.

Williamson *et al.* (2016) recently proposed that the chemistry of plagioclase could be a guide to the prospectivity of calc-alkaline magmatic suites for PCD. This paper aims to test the plagioclase discriminator proposed by Williamson *et al.* (2016) on data for Japan from the GEOROC database to determine if the lack of PCD is reflected in the chemistry of plagioclase, or whether some regions might be highlighted as potentially prospective using the discriminator as a 'first pass' exploration indicator. The plagioclase discriminator is based on the diagram in Figure 1a of An% against Al/(Ca+Na+K), both parameters calculated on the basis of atoms per formula unit (a.p.f.u.). Points lying above the discrimination line plot in the porphyry-'fertile' field as they contain excess Al (Al*), where Al* = (Al/(Ca+Na+K)-1)/0.01An>1 (Williamson *et al.*, 2016). Points below the discrimination line plot in the 'barren' field, where host rocks are unlikely to have an associated PCD. In a modification to Williamson *et al.* (2016), the most representative method for calculating excess Al is Al* = (Al/(Ca+Na+K))-(0.01*An), which gives equal weighting to the level of excess Al across the An% range. The presence of excess Al has been suggested to be due to minor (<3 mol.%) incorporation of $AlAl_3SiO_8$, with co-substitution of $[\ ]Si_4O_8$ and H_2O (Kyono & Kimata, 2001). This is likely to necessitate high melt PH_2O (Williamson *et al.*, 2016) which is consistent with the requirement for high water contents in magmas forming PCD (Richards, 2011).

2. Geological setting

The tectonic setting of Japan is extremely complex with many unresolved questions. The following brief description is largely from a recent review by Van Horne *et al.* (2017). The building blocks of what we now know as Japan were originally part of a continental arc which was rifted off the eastern edge of the Eurasian continent in the mid-Miocene. Since then, these blocks have lain on the Eurasian plate below which the Pacific and Philippine Sea plates are being subducted towards the WNW. The three plates meet at a triple junction in the Pacific Ocean, approximately 250 km southeast of Tokyo.

The pre-Neogene bedrock of Japan consists of a series of arc-parallel subduction-related accretionary complexes, ophiolites and granitic intrusions, including gneisses with Proterozoic to mid-Paleozoic protoliths. Prior to rifting from the Eurasian continent, basement rocks were intruded by Cretaceous to younger granitoids (Sasaki & Ishihara, 1979). In the NE, the granitoids vary from tonalites to monzogranites of the magnetite-series. Cretaceous granitic intrusions in the Kitakami belt, which are dominantly I-type, host skarn Cu-Fe deposits (Imai, 2004). In SW Japan, the granitoids are mostly ilmenite-series granodiorites to monzogranites, although they grade into magnetite-series granites towards the north (Fig. 2a).

Post-Cretaceous magmatism occurred within a number of separate arcs (Imai, 2004): the SW Kuril arc (eastern Hokkaido), NE Japan arc (SW Hokkaido through NE Honshu to central Honshu), Izu-Bonin arc, Kyushu-Palau ridge, SW Japan arc (northern Kyushu) and northern Ryukyu arc (southern Kyushu). The intrusive and extrusive magmatic rocks within these arcs have variable compositions and intrusion/eruptive styles (see summary in Imai, 2004).

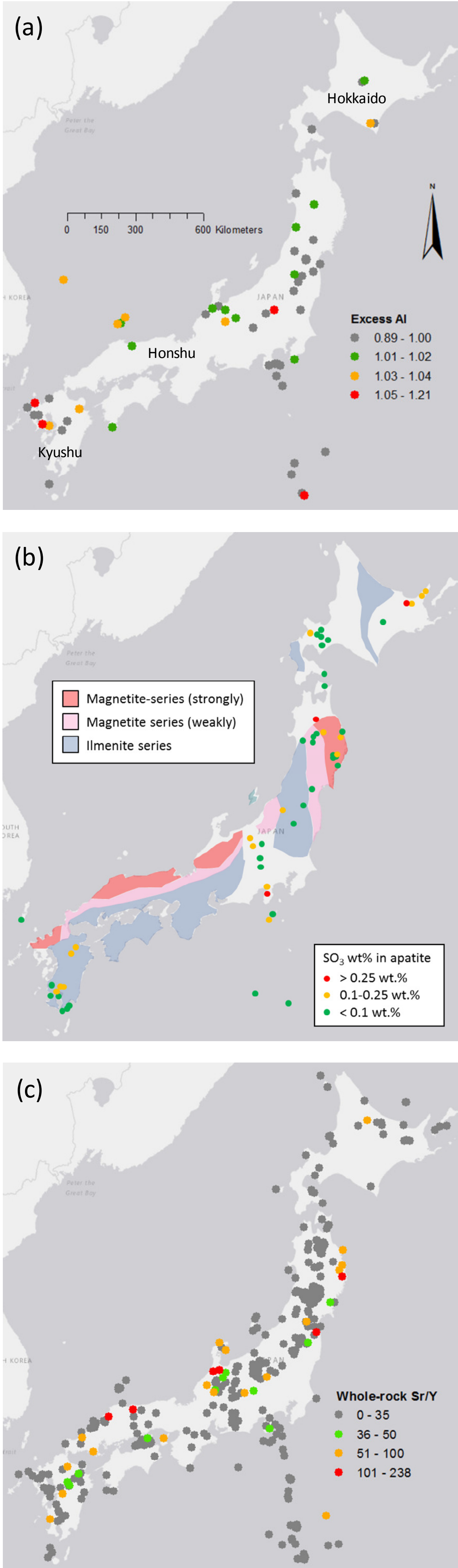

Fig. 2a) Spatial distribution of excess Al in plagioclase from Japan (3rd quartile, n>4). Data is from the GEOROC database for ‘convergent margin’ settings; b) SO_3 wt.% in apatite (data from Imai, 2004, average for each location); background map is for magnetite- and ilmenite-series terranes from Sasaki and Ishihara (1979); c) Map of whole-rock Sr/Y for data from the GEOROC database for ‘convergent margin’ settings (n = 2127). Background map (from OpenStreetMap) is in a projected (Mercator) coordinate system.

According to Sato (2012), mineralisation in Japan can be classified into four types, based on tectonic history: (i) Paleozoic–Mesozoic stratiform Cu and Mn deposits within accretionary complexes; (ii) Cretaceous–Paleogene deposits related to felsic magmatism in a continental margin arc environment, which has provided a major source of Sn, W, Mo, Cu, Pb, Zn and Fe; (iii) Miocene epigenetic and syngenetic deposits related to felsic magmatism during back-arc opening; and (iv) late Miocene–Quaternary island-arc- related volcanogenic deposits. Maps showing the distribution of mineralisation within Japan can be found in Qin and Ishihara (1998) and Sato (2012).

3. Materials and Methods

Datasets for feldspar from Japan were downloaded from the GEOROC database (http://georoc.mpch-mainz.gwdg.de/georoc, state: 01/01/2017). Data for 'convergent margin' settings only were used. For quality control, data were filtered for oxide wt.% totals of between 98.5 and 101.5 and then for data-points with K_2O<1.2 and FeO<1.2 wt.% to exclude K-feldspar and reduce the likelihood of including data affected by alteration (phyllic or propylitic). Any data from samples labelled as altered in any way were omitted. Oxide wt.% values (SiO_2, Al_2O_3, CaO, Na_2O, K_2O, FeO and MgO) were converted to a.p.f.u. based on 8 oxygen atoms for calculation of excess Al and An%, the latter from (Ca/(Ca+Na+K))x100. The data were then filtered for total cations (a.p.f.u.) of between 4.98 and 5.02, to exclude poor-quality data. Records for plagioclase from inappropriate rock types were removed, i.e. where 'ROCK NAME' in the GEOROC database is for a rock unlikely to host a PCD (e.g. dunite), or sedimentary rocks, xenoliths, highly alkaline rocks etc. For a full list of records used in this study, and excluded rock types, see online Annex A.

For whole-rock geochemical data, used to plot the Sr/Y map in Figure 2c and V/Sc in online Annex B, the GEOROC database was first filtered for data from Japan and 'convergent margin' settings. The data were then filtered, as much as possible, according to the criteria ('Selection criteria for compiled analyses') listed in Loucks (2014). Records included in the study have a loss on ignition <3.5 wt.%, oxide totals between 96.5 and 101.5 wt.%, no obvious chemical evidence for plagioclase cumulates (i.e. Eu/Eu* < 1.3, and < 20 wt.% Al_2O_3), SiO_2 wt.% between 58 and 70 wt.%, and do not have strongly alkaline compositions. For a list of records used in this study, and excluded rock types, see online Annex B.

The spatial distribution of excess Al in plagioclase and whole-rock Sr/Y and V/Sc from different magmatic systems was assessed by plotting values onto a map of Japan using ArcMap 10.2.2 (Fig. 2a, b, online Annex B). The values plotted for excess Al in plagioclase are for the 3rd quartile (n>4) of data points from each unique spatial location. The aim of this approach was to reduce the likelihood of bias in the map produced by the inclusion of outliers that might mask other data points from the same locality. Data points are shown in grey where excess Al is <1, i.e. containing no excess Al, and then in a colour scale up to the maximum value of 1.21. The whole-rock Sr/Y data in Figure 2c and V/Sc data in online Annex B are for individual data, with the highest value data points plotted on top of those with lower values.

4. Results

Data for plagioclase from Japan ('convergent margin' settings) are plotted in Figure 1a and 1b on the An% vs Al/(Ca+Na+K) discrimination diagram of Williamson *et al.*

(2016). The majority, 91% (3566/3933), of data-points fall in the 'barren' field, i.e. containing no excess Al. This compares with 73% (13400/18477) for plagioclase from convergent margin settings worldwide. The particularly low values for excess Al in Japan are clearly shown on the box and whiskers plot in Figure 1c. In Figure 1b, where data points for plagioclase from different rock types are shown, trachytes and some dacites, granodiorites and andesites range into the 'fertile' field.

The map of Japan in Figure 2a shows the spatial distribution of excess Al plotted as 3rd quartile values (n>4). One of the obvious features of the map is the relatively small number of sample localities which remain after those with less than 5 data points have been excluded. Many of the values plotted show a 'barren' signature, i.e. with Al*<1. The distribution of higher values is sporadic, although, arguably, there are groupings in northwest Kyushu and central Honshu.

5. Discussion

The majority (91%) of plagioclase compositions from Japan fall in the 'barren' field of the discrimination diagram of Williamson *et al.* (2016). This is consistent with Japan having no known PCD (e.g. Imai, 2004) and suggests low prospectivity for PCD on an arc scale. The proportion of 'barren' data is higher than for the Western Luzon arc (78%, 84/108, see Fig. 1c) which does contain PCD, despite there being relatively little plagioclase data for areas of Western Luzon where PCD are present. That 73% of plagioclase in the 'worldwide' dataset also fall in the 'barren' field is likely to reflect the rare occurrence of PCD, particularly large deposits, in calc-alkaline magmatic systems (Richards, 2016).

It was considered possible that the largely 'barren' signature in plagioclase from Japan may be due, at least in part, to the relatively high An% of the plagioclase from Japan (av. = 67) compared with that worldwide (av. = 62). This is unlikely, however, as the trendlines on Figure 1a for data from Japan and worldwide are parallel (Fig. 1a). Interestingly, there is a small increase in the proportion of points in the 'fertile' field with decreasing An%, for data from both Japan and worldwide (Fig. 1a). For Japan, trachyte, dacite and granodiorite rock compositions, amongst others, range into the 'fertile' field (Fig. 1b). This is consistent with the general trend of intermediate to felsic magmas becoming more water rich as they evolve and that high PH_2O is a prerequisite for PCD formation (Richards, 2011) which results in excess Al in plagioclase (Williamson *et al.*, 2016).

Despite most data for Japan lying in the 'barren' field, there are a number of discrete locations with excess Al (third quartile, n>4) in NW Kyushu and central Honshu. Some of these lie within magnetite-series terranes which would be most favourable for PCD (Fig. 2b). To further assess the spatial distribution of excess Al in plagioclase it has been compared with another mineral 'fertility' indicator, SO_3 wt.% in apatite from Late Cenozoic arc magmatic rocks, Cretaceous granitoids in the Kitakami belt and Miocene ilmenite-series granitic rocks from southern Kyushu (Imai, 2004). Elevated SO_3 (>0.25 wt.%) in apatite is characteristic of hydrous magmatism producing PCD, as demonstrated in the Western Luzon arc (Imai, 2004). Unfortunately, the lack of correspondence between the locations of data for excess Al and SO_3 in apatite precludes a direct comparison.

Other indicators of Cu-ore forming arc magmas are whole-rock Sr/Y and V/Sc (Loucks, 2014). The most prospective magmas are thought to have Sr/Y values above 35 and V/Sc above 10, these parameters calculated for rock compositions with 58 to 70 wt.% SiO_2 (Loucks, 2014). For the whole-rock dataset from Japan, only 8.8% (188/2127) have Sr/Y > 35, and only 2.1% (11/522) V/Sc >10. Data for Sr/Y from the GEOROCK database, 'convergent margin' setting, are shown on a map of Japan in Figure 2c. There are particularly high values for whole-rock Sr/Y in SW, N-central and NE Honshu. Despite there being relatively little data for V/Sc for many areas, it shows elevated values (>10) in central Honshu and NW Kyushu (see online Annex B). Comparing the maps for Sr/Y and V/Sc with Figure 2b (Sasaki & Ishihara, 1979), there is a general correspondence between elevated values for these parameters and areas dominated by magnetite-series (strongly) magmatic systems. These areas would appear to be most prospective for PCD and should therefore be targeted for further investigations using mineral, including plagioclase, and other indicators.

6. Conclusions

In general, from a broad assessment of excess Al in plagioclase (as a 'first pass' exploration tool), the arc segments within Japan appear to have relatively low prospectivity for PCD. Why this is the case compared with other arc segments in the western Pacific Rim (e.g. the Western Luzon arc), is intriguing, particularly whether it is controlled by tectonic factors, the nature of the magmatic source or crustal processes. There are, however, discrete high points for excess Al in plagioclase, SO_3 in apatite and whole-rock Sr/Y within the Japanese archipelago which need further investigation. It is possible, as suggested by Qin and Ishihara (1998) and Hammarstrom *et al.* (2014), that PCD may be present but are too deep (>1 km) to have been discovered.

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