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Distribution of gold derived from hydrothermal fluids on the modern seafloor and its impact on the gold budget of seawater

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The modern seafloor hydrothermal system plays a significant role in the cycling of energy and mass between the internal and external layers of the oceanic crust and upper mantle. It continues to supply hydrothermal fluids containing three to five orders of magnitude more gold into the ocean than the amount typically present in deep seawater. It has a considerable impact on the distribution and budget of gold with respect to the large geological inventory of the ocean along with other input sources such as river water. The large amount of various types of data compiled for this study reveals that only about 0.3% of the annual hydrothermal flux of gold (2618.3 kg/a) injected into the overlying seawater column as a dissolved phase is eventually trapped in sulfide deposits near vent sites on the seafloor, while about 0.8% is trapped in metalliferous sediments that fall out from the distal nonbuoyant plume. The remaining ~98.9% of gold is delivered into the depths of the global open ocean. The global budget of gold in seawater (about 1.4×10^7 kg), the annual flux of hydrothermal fluids at the seafloor (about 2.6 \times 10³ kg/a), the amount delivered by river water (about 7.2 \times 10^4 kg/a), and significant estuarine removal (15%) allows us to estimate the residence time of gold in the modern ocean to be about 220 years. This value is 70% shorter than that (~1000 years) reported previously. In the future, the use of appropriate artificial means to achieve more efficient precipitation of gold from the hydrothermal system at the seafloor could increase the level of enrichment of gold to obtain gold-rich hydrothermal deposits, yielding greater economic benefits.

KEYWORDS

gold, hydrothermal fluids, sulfides, metalliferous sediments, budget, residence time

1 Introduction

Gold (Au) belongs to the group of precious metals, and it occurs in nature as the monoisotopic element ¹⁹⁷Au, having an abundance of 100%. Owing to the reputation of Au as the most noble of the metals in human history and motivated by hopes of financial gain, many investigators have estimated the content and distribution of Au in the large volume of seawater and have attempted to recover it (Koide et al., 1988; Falkner and Edmond, 1990, and references therein). However, these studies were hampered by inadequately precise analyses and sample-contamination problems. The Au content associated with the vertical profiles of open-ocean seawater were not available until the end of the last century (Falkner and Edmond, 1990). In general, the distribution of Au in the principal layers of the Earth is heterogeneous, with its content in the shallow crust being extremely low compared to that in the deep mantle and core (Palme and O'Neill, 2014). Among the many ways for Au to be exchanged between the inner and outer layers of the oceanic crust and upper mantle, one of the most important is the seafloor hydrothermal system. Hydrothermal fluids predominantly occur along global mid-ocean ridges, volcanic arcs, and back-arc basin spreading centers, where Au is transported from sources deep within the oceanic crust and upper mantle to the seafloor. The Au content in hydrothermal fluids at the modern seafloor is generally three to five orders of magnitude and one to two orders of magnitude higher than those in the overlying seawater and river water, respectively (McHugh, 1988; Falkner and Edmond, 1990; Cidu et al., 1994; Lucas et al., 2015; Gartman et al., 2018; Fuchs et al., 2019). Therefore, hydrothermal processes at the modern seafloor can affect the cycle and budget of Au in the ocean. The average concentration and total quantity of Au delivered to the ocean from various origins must be collected and recalculated as they may reveal the ultimate fate and destination of Au derived from hydrothermal fluids in the ocean as well as the contribution of hydrothermal Au to the Au budget of ocean.

Using extensive analytical datasets that have recently be published for Au in the marine environment, in this work, we attempt to quantify the Au content and distribution in the ocean as well as the associated contribution of the seafloor hydrothermal system, including the annual flux of Au transported by hydrothermal fluids at the seafloor and the proportion of Au derived from the hydrothermal fluids and trapped in seafloor sulfide deposits that have great economic prospects as well as in metalliferous sediments that are difficult to utilize. The distributions of Au in the open ocean, river water, and hydrothermal fluids are assumed to remain approximately constant over timescales less than a million years. We consider additional input sources to the open ocean-such as aeolian dust, cosmic material, aerosols, rain, snow, and groundwater-to be negligible. Finally, using the massbalance equation, we infer the residence time of Au in the deep modern seawater from the total quantity of Au in global seawater, the contributions of Au delivered into the open ocean from seafloor hydrothermal fluids and river water in supplying Au to the open ocean and the proportion of riverine Au trapped in estuaries and on the continental shelves. The results of this study provide significant quantitative insights regarding the trapping efficiency of Au during the formation of hydrothermal sulfide deposits at the seafloor and place constraints on the behavior of Au in the hydrothermal circulation system.

2 Contribution of Au from hydrothermal fluids onto the modern ocean seafloor

An enormous volume of seawater is required to percolate downward through fractured oceanic crust every year to cool newly formed volcanic zones at plate boundaries on the global seafloor. The estimated annual flux of seawater involved in the circulation of hydrothermal fluids at the seafloor is between 7×10^{12} and 1.5×10^{14} kg/a, based on the geochemical mass balance of elements in the ocean (Hannington, 2013). Alternatively, if the temperature of the downwelling seawater reaches 350°C, the annual flux of seawater required to cool the newly formed seafloor crust is 3 $\times 10^{13}$ to 6×10^{13} kg/a (Elderfield and Schultz, 1996; Schultz and Elderfield, 1997). Although the eruption of high-temperature fluids on the modern seafloor is very spectacular, their flux represents a small fraction of the total flux of the hydrothermal fluids circulating in new volcanic zones on the global seafloor. Elderfield and Schultz (1996) and Hannington et al. (2011) have argued that the flux of such high-temperature flows accounts for only about 10% of the total flux of hydrothermal fluids at the seafloor, which is dominated by diffuse flows at much lower temperatures. Subsequent works (Nielsen et al., 2006; German and Seyfried, 2014) have suggested that a proportion of 20% may be more accurate.

Few works have tested and analyzed the Au content in hydrothermal fluids at the modern seafloor due to sampling and measurement difficulties. These results have demonstrated that the Au content differs for high-temperature fluids from different areas of the seafloor (Falkner and Edmond, 1990; Gartman et al., 2018; Fuchs et al., 2019). Toward the end of the 1980s, the first measurements of the Au content were performed for two fluid samples collected at 332°C by two submersible dives at the same hydrothermal vent in the Hanging Gardens hydrothermal field at 21°N in the East Pacific Rise (EPR); the obtained results were considerably different: 7.3×10^{-12} and 4.9×10^{-11} (Falkner and Edmond, 1990). Recent investigations at the Niua South hydrothermal field in the Tonga volcanic arc exhibited Au content ranging from 3.2×10^{-10} to 1.1×10^{-9} , with an average of 5.5×10^{-10} , for nine fluid samples recovered by a remotely operated vehicle from six vents with temperatures of 278°C-325°C (Gartman et al., 2018). This value is similar to the average content of Au (7.0×10^{-10}) obtained from 27 hydrothermal fluid samples with temperatures of 285°C-358°C in the Manus back-arc basin (Fuchs et al., 2019). As the Au content of all these high-temperature fluid samples is much lower than the saturated solubility of Au and because these ascending hydrothermal fluids tend to attain equilibrium with the surrounding rocks (German and Seyfried, 2014; Pokrovski Gleb et al., 2014), small changes in the fluid temperature around 350°C do not considerably affect the Au

content in these fluids. Thus, the average Au content (6.3×10^{-10}) in all the high-temperature fluid samples obtained from the Tonga volcanic arc and the Manus back-arc basin can be considered representative of the global Au content in hydrothermal fluids with temperatures of 350°C derived from volcanic arcs and back-arc basin spreading centers.

Very few high-temperature fluid samples were collected to analyze the Au content within the 21°N hydrothermal field in the EPR. This hydrothermal field formed at a fast-spreading ridge, where intense magmatic activity occurs and hydrothermal fluids with wide ranges of chemical composition and temperature circulate (Falkner and Edmond, 1990; German and Seyfried, 2014). This contrasts significantly with the hydrothermal fields along slow- and ultraslow-spreading mid-ocean ridges, which account for more than half of the global ridge lengths (Hannington et al., 2005; Hannington et al., 2011; German and Seyfried, 2014). Therefore, the measured Au content in the two high-temperature fluid samples from the EPR cannot be used directly to represent the Au content in hydrothermal fluids from the global mid-ocean ridges. In this paper, we assume that the ratio of the Au content in high-temperature hydrothermal fluids from mid-ocean ridges to those from back-arc basins and volcanic arcs is equal to the ratio of the seafloor hydrothermal sulfides formed in these tectonic settings (1.2/9.5; see below). Thus, we obtain the average Au content in seafloor hydrothermal fluids with temperatures of 350°C from the global mid-ocean ridges by multiplying 6.3 \times 10⁻¹⁰ with this ratio. The result is about 7.9 \times 10^{-11} , which is similar to the high value of the Au content in the two fluid samples from the 21°N hydrothermal field in the EPR (Falkner and Edmond, 1990).

The global plate boundaries have a total strike length of 8.9 \times 10^4 km in the ocean, which includes 6.4×10^4 km for the mid-ocean ridges and 2.5×10^4 km for the volcanic arcs and back-arc basin spreading centers (Bird, 2003; Hannington et al., 2011). Since lowtemperature hydrothermal fluids at the seafloor are considered to be a mixture of the high-temperature vent fluids and the recharging cold seawater (German and Seyfried, 2014), we assume that all the Au transported by the high-temperature hydrothermal fluids is either leached from the surrounding rocks or supplied directly by sub-seafloor magma. In this paper, we adopt the recently recommended annual flux of 5.6 \times 10¹³ kg/a (German and Seyfried, 2014) for hydrothermal fluids with temperatures of 350° C, which is of the same order of magnitude as that used by Hannington 2013. Furthermore, we considered that 20% of the circulating hydrothermal fluids can be directly injected into the water column above the seafloor (Nielsen et al., 2006; German and Seyfried, 2014). We assume that these high-temperature fluids erupt uniformly along the mid-ocean ridges, volcanic arcs, and back-arc basin spreading centers. Accordingly, we calculate the annual flux of Au transported to the global ocean by high-temperature fluids at the seafloor to be about 636.3 kg/a through the mid-ocean ridges and to be about 1982.0 kg/a through the volcanic arcs and back-arc basin spreading centers (Supplementary Table 1).

3 Discussion

3.1 Distribution and quantity of Au in the modern seafloor hydrothermal products

3.1.1 Au in seafloor sulfide deposits

Hannington et al. (2011) estimated that about 6×10^{11} kg of hydrothermal sulfide deposits exist at the seafloor along global midocean ridges, volcanic arcs, and back-arc basin spreading centers. This is similar to the result estimated via Monte Carlo simulations combined with probabilistic numbers for the seafloor sulfide deposits (Singer, 2014). We assume that the global distribution of these sulfide deposits on the mid-ocean ridges, volcanic arcs, and back-arc basin spreading centers is uniform. Based on their respective aforementioned strike lengths, the amounts of sulfide deposits in these tectonic settings are about 4.3 \times 10¹¹ kg and 1.7 \times 10¹¹ kg. The hydrothermal sulfides on the seafloor have heterogeneous Au distributions, i.e., the Au content in these sulfides varies by factors of thousands, but most of the values lie in between 0.1×10^{-6} and 20×10^{-6} (Hannington et al., 1986; Hannington et al., 1993; Hannington et al., 2005; D. Knight et al., 2018; Fuchs et al., 2019). Based on these statistical data, the average Au content in sulfide samples from the seafloor at the mid-ocean ridges (n = 1354) is calculated as 1.2×10^{-6} , while that from the volcanic arcs and back-arc basin spreading centers (n = 415) is 9.5 × 10^{-6} . These values are consistent with the results from several detailed reviews on the mean Au content in hydrothermal sulfides in various tectonic settings on the global seafloor (Herzig and Hannington, 1995; Petersen et al., 2016; Cherkashov, 2017). Finally, we quantify the total Au present in hydrothermal sulfide deposits from new volcanic zones on the global seafloor to be about 2.1×10^6 kg (Supplementary Table 2).

Hannington (2013) hypothesized that all hydrothermal sulfide deposits occur within a 10-km width of the axis of a seafloor spreading center. Thus, at a full-spreading rate of 4 cm/a, it would require 0.25 Ma of continuous spreading to form the current seafloor morphology. In comparison, the hundreds of sulfide samples from different hydrothermal fields on the seafloor range from several years to more than 0.2 Ma in age (Tarasov et al., 2005; Cherkashev et al., 2013; Jamieson, 2013; Jamieson et al., 2013; Cherkashov et al., 2017). The oldest known seafloor sulfide sample, with an age of 0.223 Ma, is located in the Petersburgskoye hydrothermal field at 19°52'N along the Mid-Atlantic Ridge (Cherkashov et al., 2017). An assumed spreading time of 0.25 Ma can therefore develop sulfide deposits with all the known ages of new volcanic zones on the global seafloor. The annual amount of Au accumulated in hydrothermal sulfides on the seafloor is about 8.5 kg, which accounts for about 0.3% of the annual flux of Au derived from hydrothermal fluids at the seafloor (Supplementary Table 2).

The sulfide deposits formed in association with modern hydrothermal mineralization are some of the most economically important ore deposits on the seafloor. However, the percentage of the total quantity of Au trapped in sulfide deposits with respect to

the amount transported by the hydrothermal fluids circulating in the sub-seafloor hydrothermal system is only 0.33%. This value agrees with the results obtained from Hole 1256D of the Ocean Drilling Program and from the Troodos ophiolite in Cyprus reported by Patten et al. (2016; 2017). These works indicate that the trapping efficiency of Au is extremely low under the natural conditions prevailing during modern hydrothermal mineralization on the seafloor. Based on the ongoing extraction of active geothermal fluids in Reykjanes, Iceland, Hardardóttir et al. (2009) and Hannington et al. (2016) demonstrated that the Au content in the geothermal fluids has increased about fourfold in the seven years prior and that it can produce sizable high-grade Au deposits. The continuous extraction of fluids causes pressure reduction, which increases the probability of boiling or phase separation and creates an imbalance between the surrounding rock and the hydrothermal fluids (Pokrovski Gleb et al., 2014; Hannington et al., 2016; Petrella et al., 2020). If this method can be applied effectively to the hydrothermal fields on the modern seafloor, more Au can be precipitated from the colloidal suspensions in the vent fluids, like those in the active Reykjanes geothermal system, and become trapped in the sulfide deposits. In the future, this may be an important artificial methodology for enhancing the economic potential of the hydrothermal sulfide deposits on the seafloor.

3.1.2 Au in metalliferous sediments on the seafloor

Metalliferous sediments on the seafloor predominantly comprise fine-grained particles that were formed by hydrothermal plumes that settled on the seafloor. The metalliferous sediments represent an important dispersal mechanism for the abundant chemical materials derived from the hydrothermal fluids as they diffuse into the ocean. The accumulation age of the metalliferous component of the first meter of basal sediments in the EPR (corresponding to about 0.1 Ma) multiplied by the accumulation rate of Fe (95.9 mg/cm²/ka) at the same location and by the formation rate of new areas of global oceanic crust, 2.56×10^6 m²/a, and then divided by the average Fe content (32.7%) in the metalliferous sediments (Barrett et al., 1987; Hannington, 2013), yields 7.5×10^8 kg/a, which we adopt in this paper as the total annual flux of metalliferous sediment deposition on the seafloor of the new volcanic zones of the mid-ocean ridges. If the ratio of the deposition flux of metalliferous sediments from midocean ridges to volcanic arcs and back-arc basin spreading centers is consistent with the ratio (4.3/1.7) of their strike lengths, then we estimate 1.04×10^9 kg/a as the deposition flux of metalliferous sediments in new global volcanic zones, which is two to three orders of magnitude higher than that of the sulfide deposits. The hydrothermal fields in the EPR are the modern-day areas that have been investigated most extensively for the Au content of the metalliferous sediments on the seafloor. A detailed statistical study has shown that after removing their carbonate components, the average Au content in metalliferous sediments from the EPR is about 2.03 \times 10⁻⁸ (Gurvich, 2006), which is one order of magnitude higher than that in pelagic sediments not affected by seafloor hydrothermal processes (Li and Schoonmaker, 2014). The annual flux of Au in the global seafloor metalliferous sediments is about 21.2 kg/a when the average Au content in the metalliferous sediments on the global seafloor is the same as in the EPR, which accounts for about 0.8% of the annual flux of Au derived from the seafloor hydrothermal fluids (Supplementary Table 3).

3.1.3 Delivery of remaining Au into the ocean

Subtracting the annual flux of Au in the global seafloor sulfide deposits and metalliferous sediments yields the remaining annual flux of Au of about 2588.6 kg/a, which is about 98.9% of the annual flux of Au derived from hydrothermal fluids at the seafloor. The distribution and quantity of Au in hydrothermal products on the modern seafloor is summarized in Supplementary Figure 1. The Au transported by the rising hydrothermal fluids is distributed to different places once it reaches the seafloor; a small portion of Au is trapped directly in near-vent sulfide deposits, slightly more Au is carried by hydrothermal plumes and falls out into the distal metalliferous sediments, whereas an enormous proportion of Au transported by the hydrothermal fluids from deep within the oceanic crust and upper mantle is finally injected into the overlying column of seawater.

3.2 Global ocean budget of Au and the riverine flux

3.2.1 Total quantity of Au in modern ocean seawater

The modern ocean is a vast body of saline water that covers most of the Earth's surface; it thus becomes a huge natural repository of Au. The concentration of dissolved Au varies with depth in the vertical profile of seawater, and the content of Au in a column of open seawater ranges from 2×10^{-15} to 6×10^{-14} (Koide et al., 1988; Falkner and Edmond, 1990). In addition, the distribution of Au varies globally, with the Au content in open seawater generally being lower than that in coastal and marginal seawater, which may be affected by terrestrial materials and tailing sewage (Large et al., 2015). The total mass of the global seawater is a staggering 1.41×10^{21} kg (Garrison, 2016). If the most common content of Au (1×10^{-14}) obtained from seawater profiles from the Atlantic and Pacific Oceans is taken as the average value for seawater globally (Falkner and Edmond, 1990), which is also similar to the values given in a recent work (Bruland et al., 2014), the total quantity of Au dissolved in seawater globally is about 1.4 $\times 10^7$ kg.

3.2.2 Annual riverine flux of Au into the ocean

Rivers around the globe also majorly determine the quantity and chemical composition of a column of seawater as they annually discharge about 3.6×10^{16} kg of freshwater into the oceans (Milliman and Farnsworth, 2013). Although the distribution of Au within river water samples is heterogeneous, most of the Au content ranges from 1×10^{-12} to 5×10^{-12} , with an average of 2×10^{-12} , based on analyses of dozens of river-water samples recovered from North America,

Europe, and Australia (McHugh, 1988; Cidu et al., 1994; Lucas et al., 2015). The content of Au in some rivers and in the groundwater around Au mines that were contaminated by mine sewage is one to two orders of magnitude higher than that in normal river water (McHugh, 1988; Lucas et al., 2014; Large et al., 2015; Myagkaya et al., 2016), but the flux of such sewage into the rivers is too small to affect the Au content in river water globally. Consequently, in this paper we adopt 2×10^{-12} to be the average Au content in river water globally, which yields an annual flux of Au transported by river water of about 7.2×10^4 kg/a (Supplementary Figure 2).

Nekrasov et al. (1996); Leybourne et al. (2000), and Large et al. (2015) have demonstrated that Au is present in a higher proportion in river water as suspended particles or in a colloidal state than as the predominant dissolved species, which consist of a mixture of colloids, nanoparticles, aqueous clusters, or Au absorbed onto detrital clays and Au-organic complexes in the ocean. Therefore, a significant portion of the Au delivered to the ocean is more likely to be deposited in estuaries and on continental shelves than be extended into the open ocean. This conclusion is in agreement with the work from the Swan River estuary in Western Australia, which shows that the content of Au in the estuary water ranges from $1.1 \times$ 10^{-11} to 3.7×10^{-11} and that it is about one order of magnitude higher than that in the upriver water (Lucas et al., 2015). Assuming that the percentage of Au removed by the estuaries and continental shelves is the same as that of Os, which is also an immobile precious metal, and that about 15% of the total riverine Os is deposited in estuaries and on continental shelves (Turekian et al., 2007), we observe that the annual flux of Au transported by river water is reduced to about 6.1×10^4 kg/a (Supplementary Table 4).

3.3 Impact of Au derived from seafloor hydrothermal fluids on the Au budget of the global ocean

3.3.1 Residence time of Au in modern openocean seawater

The input of aeolian dust, cosmic material, aerosols, rain, snow, and groundwater can be considered to provide additional fluxes of Au into the ocean, in excess of those derived from river water and hydrothermal fluids. However, some of these sources have negligible Au content, some have very small input fluxes, and others are difficult to dissolve in seawater and eventually settle onto the seafloor. Thus, these input sources barely affect the input flux of Au into modern seawater (Falkner and Edmond, 1990; Love and Brownlee, 1993; Oxburgh, 2001; Jickells et al., 2005; Lucas et al., 2014). The residence time (T) of Au in the ocean is defined at steady state by its mass balance. The equation is T = I/F, where I (about 1.4×10^7 kg) is the inventory of the Au in the global seawater, and F is the net riverine and hydrothermal fluid input fluxes (about 6.4×10³ kg) to the ocean. Therefore, from existing datasets of hydrothermal fluids at the seafloor and river water flowing into the open ocean, we estimate the residence time of Au in the ocean to be about 220 years, 70% lower than that estimated in a previous work (Falkner and Edmond, 1990). This residence time is much shorter than the turnover time of the deep ocean (Bruland et al., 2014), and it indicates that Au has the obvious behavioral characteristics of a non-conservative element in the ocean.

3.3.2 Seafloor hydrothermal eruptions and the formation of Au-rich deposits in geological history

The net impact of Au associated with the venting of fluids at the seafloor is similar to the relationship with river water and the ocean because the Au has markedly higher concentrations in the vent fluids and river water than in the seawater. This implies that sediment deposition and authigenic mineral formation in the marine environment can remove a large amount of dissolved Au from the seawater, significantly and continuously altering the Au budget of the global ocean. However, the contribution of Au delivered to the ocean from hydrothermal fluids at the modern seafloor is less than that delivered by river water. Nevertheless, larger episodic volcanic eruptions at the seafloor may have transported larger fluxes of hydrothermal fluids and, consequently, a higher average Au content into the ocean earlier in the Earth's history, i.e., during the Mesoarchean, the Neoarchean, and the end of the Proterozoic Eras, during which the average content of Au in seawater was several times higher than the present values (Large et al., 2015). The input of Au delivered into the ocean by hydrothermal fluids at the seafloor during these times has gained tremendous importance because it not only affected the total quantity and economic prospects of the hydrothermal sulfide deposits at the seafloor but also may have reduced the residence time of Au in the global seawater significantly.

4 Conclusions

The annual flux of Au derived from hydrothermal fluids at the modern seafloor is about 2618.3 kg/a. About 0.3% of this (8.5 kg/a) is first trapped in near-vent hydrothermal sulfide deposits, and another ~0.8% (21.2 kg/a) diffuses with hydrothermal plumes and falls out into the distal metalliferous sediments. The remaining ~98.9% (2588.6 kg/a) is eventually delivered to the seawater column.

From the total quantity of Au in seawater globally $(1.4 \times 10^7 \text{ kg})$ and from the input fluxes of Au derived from hydrothermal fluids at the seafloor (2588.6 kg/a) and river water (6.1×10^4 kg/a), we estimate the residence time of Au in the modern open ocean to be about 220 years, which is about 70% lower than the previous value and considerably shorter than the turnover time of the deep ocean. In the future, more efficient mechanisms to precipitate Au from the seafloor hydrothermal system, such as the use of appropriate artificial means like continuous extraction from hydrothermal fluids or breaking the buffer balance between the fluids and the surrounding rock, may be used to increase the enrichment of Au in seafloor hydrothermal systems to form Au-rich deposits and obtain increased economic benefits.

Data availability statement

The original contributions presented in the study are included in the article/Supplementary Material. Further inquiries can be directed to the corresponding author.

Author contributions

WH and JLu contributed to conception, methodology and design of the study. WH and JLi organized the data and performed the statistical analysis. WH wrote the first draft of the manuscript. FH, PL, and RC made revisions of the manuscript. All the authors contributed to the article and approved the submitted version.

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References

Barrett, T. J., Taylor, P. N., and Lugoqski, J. (1987). Metalliferous sediments from DSDP leg 92: the East pacific rise transect. *Geochimica Cosmochimica Acta* 51 (9), 2241–2253. doi: 10.1016/0016-7037(87)90278-X

Bird, P. (2003). An updated digital model of plate boundaries. *Geochemistry Geophysics Geosystems* 4 (3) 1-52. doi: 10.1029/2001GC000252

Bruland, K. W., Middag, R., and Lohan, M. C. (2014). "8.2 - controls of trace metals in seawater," in *Treatise on geochemistry (Second edition)*. Eds. H. D. Holland and K. K. Turekian (Oxford: Elsevier), 19–51. doi: 10.1016/B978-0-08-095975-7.00602-1

Cherkashev, G. A., Ivanov, V. N., Bel'tenev, V. I., Lazareva, L. I., Rozhdestvenskaya, I. I., Samovarov, M. L., et al. (2013). Massive sulfide ores of the northern equatorial mid-Atlantic ridge. *Oceanology* 53 (5), 607–619. doi: 10.1134/S0001437013050032

Cherkashov, G. (2017). "Seafloor massive sulfide deposits: distribution and prospecting," in *Deep-Sea mining: resource potential, technical and environmental considerations.* Ed. R. Sharma (Cham: Springer International Publishing), 143-164).

Cherkashov, G., Kuznetsov, V., Kuksa, K., Tabuns, E., Maksimov, F., and Bel'tenev, V. (2017). Sulfide geochronology along the northern equatorial mid-Atlantic ridge. *Ore Geology Rev.* 87, 147–154. doi: 10.1016/j.oregeorev.2016.10.015

Cidu, R., Fanfani, L., Shand, P., Edmunds, W. M., Van't dack, L., and Gijbels, R. (1994). Determination of gold at the ultratrace level in natural waters. *Analytica Chimica Acta* 296 (3), 295–304. doi: 10.1016/0003-2670(94)80249-1

D. Knight, R., Roberts, S., and Webber, A. P. (2018). The influence of spreading rate, basement composition, fluid chemistry and chimney morphology on the formation of gold-rich SMS deposits at slow and ultraslow mid-ocean ridges. *Mineralium Deposita* 53 (1), 143–152. doi: 10.1007/s00126-017-0762-4

Elderfield, H., and Schultz, A. (1996). Mid-ocean ridge hydrothermal fluxes and the chemical composition of the ocean. *Annu. Rev. Earth Planetary Sci.* 24 (1), 191–224. doi: 10.1146/annurev.earth.24.1.191

Falkner, K., and Edmond, J. M. (1990). Gold in seawater. *Earth Planetary Sci. Lett.* 98 (2), 208–221. doi: 10.1016/0012-821X(90)90060-B

Fuchs, S., Hannington, M. D., and Petersen, S. (2019). Divining gold in seafloor polymetallic massive sulfide systems. *Mineralium Deposita* 54 (6), 789-820. doi: 10.1007/s00126-019-00895-3

Garrison, T. S. (2016). Oceanography: an invitation to marine science (Toronto, Canada: nelson education).

Gartman, A., Hannington, M., Jamieson, J. W., Peterkin, B., Garbe-Schönberg, D., Findlay, A. J., et al. (2018). Boiling-induced formation of colloidal gold in black smoker hydrothermal fluids. *Geology* 46 (1), 39–42. doi: 10.1130/G39492.1 %J Geology

German, C. R., and Seyfried, W. E. (2014). "8.7 - hydrothermal processes," in *Treatise on geochemistry (Second edition)*. Eds. H. D. Holland and K. K. Turekian (Oxford: Elsevier), 191–233. doi: 10.1016/B978-0-08-095975-7.00607-0

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Supplementary material

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Gurvich, E. G. (2006). Metalliferous sediments of the world ocean: fundamental theory of deep-sea hydrothermal sedimentation (Berlin: Springer).

Hannington, M. D. (2013). The role of black smokers in the Cu mass balance of the oceanic crust. *Earth Planetary Sci. Lett.* 374, 215–226. doi: 10.1016/j.epsl.2013.06.004

Hannington, M. D., De Ronde, C. E. J., Petersen, S., Hedenquist, J. W., Thompson, J. F. H., Goldfarb, R. J., et al. (2005). "Sea-Floor tectonics and submarine hydrothermal systems," in *Geochemistry, Mineralogy and Genesis of Gold Deposits* (Society of Economic Geologists). https://oceanrep.geomar.de/id/eprint/6271/

Hannington, M., Harðardóttir, V., Garbe-Schönberg, D., and Brown, K. L. (2016). Gold enrichment in active geothermal systems by accumulating colloidal suspensions. *Nat. Geosci.* 9 (4), 299–302. doi: 10.1038/ngeo2661

Hannington, M. D., Herzig, P. M., and Scott, S. D. (1993). "Auriferous hydrothermal precipitates on the modern seafloor," in *Gold metallogeny and exploration*. Ed. R. P. Foster (Dordrecht: Springer Netherlands), 249–282.

Hannington, M., Jamieson, J., Monecke, T., Petersen, S., and Beaulieu, S. (2011). The abundance of seafloor massive sulfide deposits. *Geology* 39 (12), 1155–1158. doi: 10.1130/G32468.1 %J Geology

Hannington, M. D., Peter, J. M., and Scott, S. D. (1986). Gold in sea-floor polymetallic sulfide deposits. *Economic Geology* 81 (8), 1867–1883. doi: 10.2113/ gsecongeo.81.8.1867 %J Economic Geology

Hardardóttir, V., Brown, K. L., Fridriksson, T., Hedenquist, J. W., Hannington, M. D., and Thorhallsson, S. (2009). Metals in deep liquid of the reykjanes geothermal system, southwest Iceland: implications for the composition of seafloor black smoker fluids. *Geology* 37 (12), 1103–1106. doi: 10.1130/G30229A.1 %J Geology

Herzig, P. M., and Hannington, M. D. (1995). Polymetallic massive sulfides at the modern seafloor a review. *Ore Geology Rev.* 10 (2), 95–115. doi: 10.1016/0169-1368(95) 00009-7

Jamieson, J. W. (2013). Size, age, distribution and mass accumulation rates of seafloor hydrothermal sulfide deposits (University of Ottawa (Canada: Doctoral degree).

Jamieson, J. W., Hannington, M. D., Clague, D. A., Kelley, D. S., Delaney, J. R., Holden, J. F., et al. (2013). Sulfide geochronology along the endeavour segment of the Juan de fuca ridge. *Geochemistry Geophysics Geosystems* 14 (7), 2084–2099. doi: 10.1002/ggge.20133

Jickells, T. D., An, Z. S., Andersen, K. K., Baker, A. R., Bergametti, G., Brooks, N., et al. (2005). Global iron connections between desert dust, ocean biogeochemistry, and climate. *Science* 308 (5718), 67–71. doi: 10.1126/science.1105959

Koide, M., Hodge, V., Goldberg, E. D., and Bertine, K. (1988). Gold in seawater: a conservative view. *Appl. Geochemistry* 3 (3), 237–241. doi: 10.1016/0883-2927(88) 90103-5

Large, R. R., Gregory, D. D., Steadman, J. A., Tomkins, A. G., Lounejeva, E., Danyushevsky, L. V., et al. (2015). Gold in the oceans through time. *Earth Planetary Sci. Lett.* 428, 139–150. doi: 10.1016/j.epsl.2015.07.026

Leybourne, M. I., Goodfellow, W. D., Boyle, D. R., and Hall, G. E. M. (2000). Form and distribution of gold mobilized into surface waters and sediments from a gossan tailings pile, Murray brook massive sulphide deposit, new Brunswick, Canada. *Appl. Geochemistry* 15 (5), 629–646. doi: 10.1016/S0883-2927(99)00068-2

Li, Y. H., and Schoonmaker, J. E. (2014). "9.1 - chemical composition and mineralogy of marine sediments," in *Treatise on geochemistry (Second edition)*. Eds. H. D. Holland and K. K. Turekian (Oxford: Elsevier), 1–32. doi: 10.1016/B978-0-08-095975-7.00701-4

Love, S. G., and Brownlee, D. E. (1993). A direct measurement of the terrestrial mass accretion rate of cosmic dust. *Science* 262 (5133), 550–553. doi: 10.1126/ science.262.5133.550

Lucas, A. R., Reid, N., Salmon, S. U., and Rate, A. W. (2014). Quantitative assessment of the distribution of dissolved au, as and Sb in groundwater using the diffusive gradients in thin films technique. *Environ. Sci. Technol.* 48 (20), 12141–12149. doi: 10.1021/es502468d

Lucas, A. R., Salmon, S. U., Rate, A. W., Larsen, S., and Kilminster, K. (2015). Spatial and temporal distribution of au and other trace elements in an estuary using the diffusive gradients in thin films technique and grab sampling. *Geochimica Cosmochimica Acta* 171, 156–173. doi: 10.1016/j.gca.2015.08.025

McHugh, J. B. (1988). Concentration of gold in natural waters. J. Geochemical Explor. 30 (1), 85–94. doi: 10.1016/0375-6742(88)90051-9

Milliman, J. D., and Farnsworth, K. L. (2013). River discharge to the coastal ocean: a global synthesis (Cambridge, United Kingdom: Cambridge University Press).

Myagkaya, I. N., Lazareva, E. V., Gustaytis, M. A., and Zhmodik, S. M. (2016). Gold and silver in a system of sulfide tailings. part 1: migration in water flow. *J. Geochemical Explor.* 160, 16–30. doi: 10.1016/j.gexplo.2015.10.004

Nekrasov, I. Y., Rao, P. M., and Majithia, M. (1996). Geochemistry, mineralogy and genesis of gold deposits (London: Routledge).

Nielsen, S. G., Rehkämper, M., Teagle, D. A. H., Butterfield, D. A., Alt, J. C., and Halliday, A. N. (2006). Hydrothermal fluid fluxes calculated from the isotopic mass balance of thallium in the ocean crust. *Earth Planetary Sci. Lett.* 251 (1), 120–133. doi: 10.1016/j.epsl.2006.09.002

Oxburgh, R. (2001). Residence time of osmium in the oceans. *Geochemistry Geophysics Geosystems* 2 (6), 1-17. doi: 10.1029/2000GC000104

Palme, H., and O'Neill, H. S. C. (2014). "3.1 - cosmochemical estimates of mantle composition," in *Treatise on geochemistry (Second edition)*. Eds. H. D. Holland and K. K. Turekian (Oxford: Elsevier), 1–39. doi: 10.1016/B978-0-08-095975-7.00201-1

Patten, C. G. C., Pitcairn, I. K., and Teagle, D. A. H. (2017). Hydrothermal mobilisation of au and other metals in supra-subduction oceanic crust: insights from the troodos ophiolite. *Ore Geology Rev.* 86, 487–508. doi: 10.1016/j.oregeorev.2017.02.019

Patten, C. G. C., Pitcairn, I. K., Teagle, D. A. H., and Harris, M. (2016). Mobility of au and related elements during the hydrothermal alteration of the oceanic crust: implications for the sources of metals in VMS deposits. *Mineralium Deposita* 51 (2), 179–200. doi: 10.1007/s00126-015-0598-8

Petersen, S., Krätschell, A., Augustin, N., Jamieson, J., Hein, J. R., and Hannington, M. D. (2016). News from the seabed – geological characteristics and resource potential of deep-sea mineral resources. *Mar. Policy* 70, 175–187. doi: 10.1016/j.marpol.2016.03.012

Petrella, L., Thébaud, N., Fougerouse, D., Evans, K., Quadir, Z., and Laflamme, C. (2020). Colloidal gold transport: a key to high-grade gold mineralization? *Mineralium Deposita* 55 (7), 1247–1254. doi: 10.1007/s00126-020-00965-x

Pokrovski Gleb, S., Akinfiev Nikolay, N., Borisova Anastassia, Y., Zotov Alexandre, V., and Kouzmanov, K. (2014). Gold speciation and transport in geological fluids: insights from experiments and physical-chemical modelling. *Geological Society London Special Publications* 402 (1), 9–70. doi: 10.1144/SP402.4

Schultz, A., and Elderfield, H. (1997). Controls on the physics and chemistry of seafloor hydrothermal circulation. *Philos. Trans. R. Soc. London A: Mathematical Phys. Eng. Sci.* 355 (1723), 387–425. doi: 10.1098/rsta.1997.0014

Singer, D. A. (2014). Base and precious metal resources in seafloor massive sulfide deposits. Ore Geology Rev. 59, 66–72. doi: 10.1016/j.oregeorev.2013.11.008

Tarasov, V. G., Gebruk, A. V., Mironov, A. N., and Moskalev, L. I. (2005). Deep-sea and shallow-water hydrothermal vent communities: two different phenomena? *Chem. Geology* 224 (1), 5–39. doi: 10.1016/j.chemgeo.2005.07.021

Turekian, K. K., Sharma, M., and Gordon, G. W. (2007). The behavior of natural and anthropogenic osmium in the Hudson river-long island sound estuarine system. *Geochimica Cosmochimica Acta* 71 (17), 4135–4140. doi: 10.1016/j.gca.2007.05.020