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Toxic mercury pulses into late Permian terrestrial and marine environments

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ABSTRACT

Large spikes in mercury (Hg) concentration are observed globally at the latest Permian extinction (LPE) horizon that are thought to be related to enhanced volcanic emissions of the Siberian Traps large igneous province (LIP). While forming an effective chemostratigraphic marker, it remains unclear whether such enhanced volcanic Hg emissions could have generated toxic conditions that contributed to extinction processes. To address this, we examined the nature of enhanced Hg emissions from the Siberian Traps LIP and the potential impact it may have had on global ecosystems during the LPE. Model results for a LIP eruption predict that pulses of Hg emissions to the atmosphere would have been orders of magnitude greater than normal background conditions. When deposited into world environments, this would have generated a series of toxic shocks, each lasting >1000 yr. Such repeated Hg loading events would have had severe impact across marine trophic levels, as well as been toxic to terrestrial plant and animal life. Such high Hg loading rates may help explain the co-occurrence of marine and terrestrial extinctions.

INTRODUCTION

Large igneous provinces (LIPs) have shaped evolution of life on the planet. Defined as emplacements of >100,000 km^3 of mainly mafic magma (Ernst and Youbi, 2017), LIPs are temporally associated with all of the major mass extinctions of the Phanerozoic (Bond and Grasby, 2017; Ernst and Youbi, 2017). Even the extinction of dinosaurs, purported to be caused by an asteroid impact, has been suggested to be linked to a LIP event (e.g., Schoene et al., 2019). The exact killing mechanism of a LIP remains controversial, however, and not all LIPs drive mass extinctions (Bond and Grasby, 2017). LIPs can impact global biogeochemical systems through, for example, emissions of greenhouse gases driving global warming and ocean acidification (Svensen et al., 2009) and halogens that deplete the ozone layer (Black et al., 2014), as well as an increased flux of toxic metals to the environment (Grasby et al., 2015). Most volcanically released metals are particle bound and drop out near eruptive centers. Mercury (Hg), however, is a volatile metal with a 0.5–1 yr atmospheric residence time that allows effective interhemispheric mixing and global atmospheric distribution (Grasby et al., 2019). Eventually, 30% of atmospheric Hg is precipitated onto world terrestrial and 70% onto marine environments (Amos et al., 2014). Such Hg fallout from large volcanic eruptions over the past several hundred years is recorded in modern sediment and ice records, as well as in sediments deposited contemporaneously with most LIP events throughout the Phanerozoic (Grasby et al., 2019). In the case of LIPs, Hg spikes in sediments also record additional Hg released from accelerated biomass burning and soil erosion (Grasby et al., 2019; Chu et al., 2020). What remains unclear is whether these Hg spikes form just a simple chemostratigraphic marker, or potentially a signal of volcanic-induced Hg toxicity.

In the methylated form, Hg is a potent neurotoxin associated with behavioral, developmental, nephrological, reproductive, and endocrinological consequences (Eagles-Smith et al., 2018). Modern anthropogenic Hg emissions, which are approximately equal to geogenic sources, are of high concern given the toxicity and bioaccumulation of Hg in the environment. A LIP event could have conceivably increased Hg emissions several times higher than modern anthropogenic emissions levels (Grasby et al., 2015). If so, then volcanic Hg release could have contributed to extinction processes through creation of toxic conditions.

The Hg emission rates of LIPs, and consequently the potential severity of LIP-induced Hg toxicity on global ecosystems, are uncertain. Estimates of Hg emissions are constrained by assumptions on the nature of LIP volcanism itself. To examine this, we focus on one of the largest LIPs in Earth history, the Siberian Traps, with an estimated 2–4 × 10^6 km^3 of magma released (Vasil'ev et al., 2000). The Siberian Traps are linked to the latest Permian extinction (LPE), the largest known mass-extinction event in the Phanerozoic (Erwin, 2006), which was marked by the loss of >90% of marine and 70% of terrestrial species, including the only known mass extinction of insects (Labandeira and Sepkoski, 1993). Spikes in Hg concentrations are widely observed in marine and terrestrial strata deposited at the extinction boundary (e.g., Sanei et al., 2012; Grasby et al., 2019, Chu et al., 2020) and are thought to reflect enhanced Hg emissions by the Siberian Traps. We examine here the potential impact that such enhanced Hg emissions could have had on the global environment.

METHODS

The Siberian Traps LIP is thought to have erupted the majority of its volume over ~300 k.y., and then continued at a reduced level for at least another 500 k.y. (Burgess et al., 2017). If extruded evenly over that period, lava

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production rates would have been similar to modern global basalt production at mid-ocean ridges (~3 km^3/yr) (Pavlov et al., 2019) and Hg flux would not be significantly higher than at present. However, such a constant eruption style is unlikely. Volcanic eruptions are a stochastic process, with extrusion of lava being distributed as a series of sequential eruptions, which can be modeled by a multifractal time series (Godano and Civetta, 1996; Gusev, 2014). Such a stochastic eruption process is supported for LIP events by high-resolution radioisotope age dates of the Deccan Traps (Schoene et al., 2019) that show a fractal-like pattern. Such a process is best described by a binomial multiplicative cascade (Godano and Civetta, 1996; Gusev, 2014), and we thus modeled the Siberian Traps eruptive history (and consequently the Hg flux) as a multifractal time series (Kirichenko et al., 2019) (see the Supplemental Material[1]). The total Hg mass released by the Siberian Traps, estimated to be ~4.0×10^8 Mg based on known relationships to SO_2 emissions (Grasby et al., 2015), was distributed as a function of eruption volume generated in our model, such that the temporal variations of Hg emissions to the atmosphere are directly associated with the modeled eruption rate. Given uncertainty in the eruptive history of the Siberian traps, we modeled a series of random total eruption periods (40 k.y., 80 k.y., 160 k.y., 320 k.y., 640 k.y.) that straddle the estimated 300 k.y. main eruptive phase (Burgess et al., 2017). For each eruption period, 100 model runs were generated to produce a statistical range of potential results.

Results from our atmosphere emissions model were then fed into a seven-reservoir box model of the global mercury cycle based on that of Amos et al. (2014). This model distributes the atmospheric emissions through a series of mass transfers between global terrestrial and marine Hg reservoirs (Fig. 1) as represented by a system of coupled first-order differential equations (Supplemental Material). The model tracks the movement of Hg between reservoirs over time, until eventually the volcanic Hg released to the atmosphere is transferred into long-term geologic storage in marine sediments.

RESULTS

Our model generates a fractal distribution for the eruption of the Siberian Traps, with a resultant volcanic Hg flux associated with each eruption period. The atmospheric Hg emissions are plotted as excess to normal background geogenic emissions in Figure 2. Here Hg release is characterized by a series of pulsed emissions to the atmosphere throughout the LIP event, rather than a constant increased emission rate. There would have been Hg sources additional to what we model from only volcanic emissions, such as that related to organic-matter combustion through heating by Siberian Traps sills in the Tunguska sedimentary basin (Svensen et al., 2009; Grasby et al., 2011), as well as Hg release from forest fires and soil erosion related to landscape disturbance by the Siberian Traps (Grasby et al., 2019). However, mass-flux estimates suggest that these additional sources, while representing additional Hg loading to the environment, are less-important contributions of Hg than volcanic emissions (Sanei et al., 2012). As such, our results represent a minimum estimation of the total Hg flux during a LIP eruption.

The Hg released to the atmosphere by the Siberian Traps is transferred to the terrestrial and marine environment by atmospheric fallout and river transport in a series of pulsed Hg fluxes to the land and then the upper ocean that mimic the pattern for atmospheric injection in Figure 2. Our model also shows that between average eruption episodes, there is sufficient time for effective transfer of Hg from the upper ocean to long-term geologic sequestration (Fig. 3A) as a form of natural ocean remediation. This transfer of Hg pulses to the sedimentary reservoir is characterized by a series of spikes in Hg concentration, consistent with Hg spikes observed in sediments deposited coevally with the Siberian Trap eruptions (Sanei et al., 2012; Grasby et al., 2019) as well as other LIP events during the Phanerozoic (Grasby et al., 2019). However, the effective transfer of volcanic Hg pulses to long-term sequestration in ocean sediments would be on the time scale of ~10^3 yr (Fig. 3B). While this represents relatively short-duration transient Hg loading on a geological time scale, for global ecosystems this would have been experienced as a series of highly elevated Hg levels over millennial time scales.

TOXIC Hg PULSES

Volcanic and hydrothermal emissions are the primary source of Hg to the environment, with an estimated ~500 Mg/yr under background conditions (Grasby et al., 2019). Mercury then undergoes complex geochemical cycling before ultimately being transferred to the ocean via atmospheric deposition and riverine transport (Amos et al., 2014). On land, Hg has a strong affinity for soil organic matter, and mobile Hg-humic or Hg-fulvic complexes are easily methylated to produce highly toxic and bioavailable methylmercury (MeHg), which is then readily taken up from soil by plant root systems and transferred to edible parts (Natasha et al., 2020). In seawater, Hg typically exists as inorganic Hg (IHg) species in seawater, mainly in particulate (Hg_p) and dissolved (Hg_{dis}) forms (Ullrich et al., 2001). Hg_p readily sinks to the seafloor, whereas Hg_{dis} drawdown occurs through organic-matter binding (Sanei et al., 2012; Grasby et al., 2019). In the water column and surface sediments, IHg can be microbially methylated (forming MeHg) (Hsu-Kim et al., 2013). In contrast to IHg, MeHg is rapidly assimilated by organisms owing to the strong binding affinity with sulfur-containing ligands (i.e., thiol ligands). Therefore, MeHg has a strong capability to bioaccumulate along food chains, reaching, for instance, concentrations 10^7 higher in fish than in seawater (Ullrich et al., 2001).

The environmental risk that excess Hg loading represented to late Permian life lies in the

[1]Supplemental Material. Methods and input parameters for the global mercury model used in this study. Please visit https://doi.org/10.1130/10.1130/GEOL.S.12275729 to access the supplemental material, and contact editing@geosociety.org with any questions. The code used in this study is available from author Xiaojun Liu.

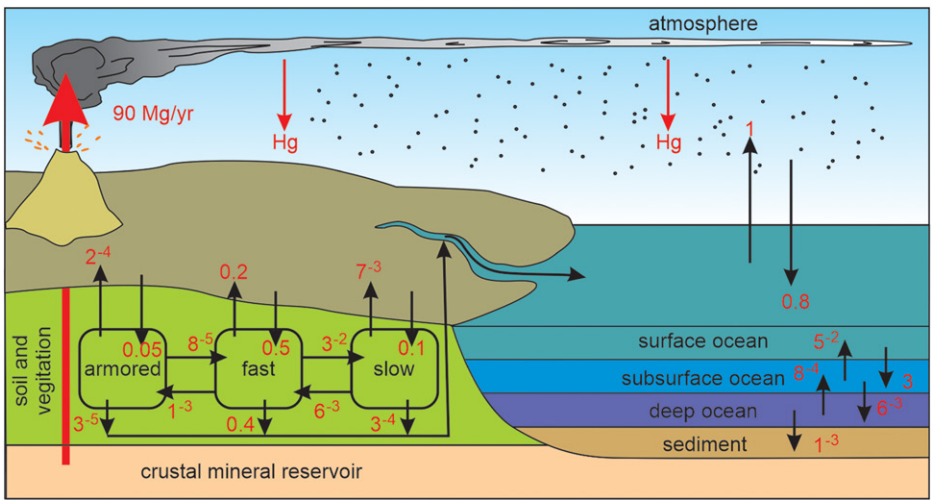

Figure 1. Diagram showing model design, along with Hg flux coefficients (in years) based on Amos et al. (2014). Mercury is cycled between the atmosphere, fast terrestrial environment (vegetation), slow and armored soil reservoirs (representing variable speed of movement of Hg through organic carbon remineralization), and the ocean (surface, subsurface, deep), and ultimately removed by geologic sequestration in marine sediments.

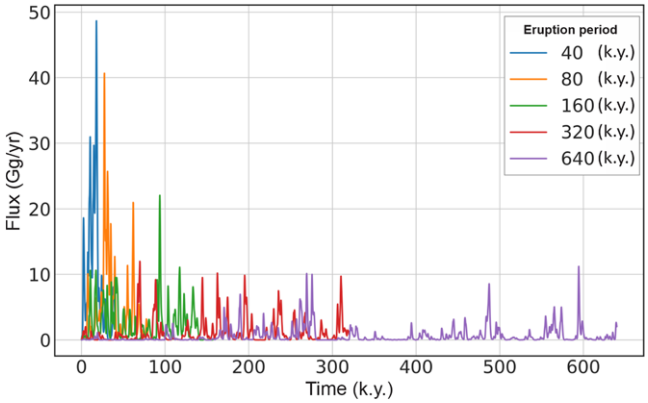

Figure 2. Plot showing calculated volcanic Hg flux rates to the atmosphere for multifractal eruption process of the Siberian Traps, using different assumed main eruption periods ranging from 40 to 640 k.y. Resultant flux to the terrestrial and upper ocean reservoirs mimics this pattern.

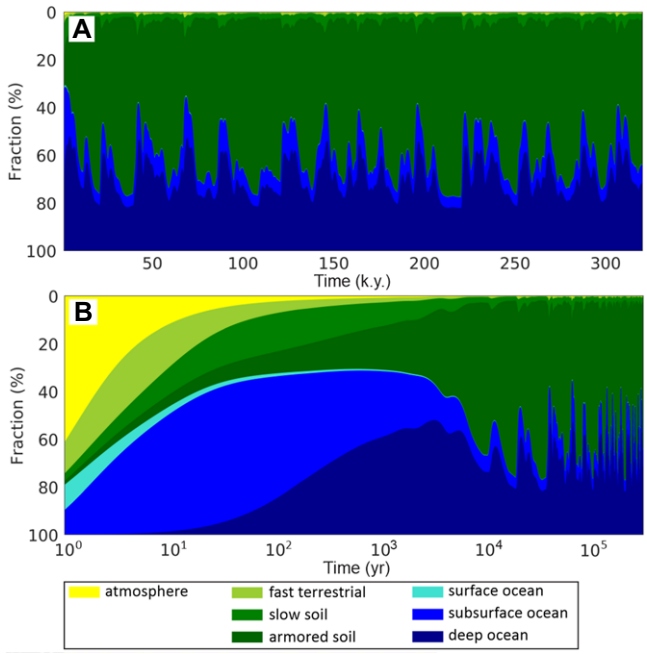

Figure 3. (A) Plot showing distribution of Hg in ocean and sediment systems during a period of large igneous province eruption, illustrating a series of peak Hg loads to surface water that eventually become sequestered to sediments for long-term geologic storage. (B) Same results as in A, but with time plotted at an exponential scale, illustrating that high Hg loads can be sustained in the water column for >10^3 yr.

toxicity of IHg and MeHg. At high trophic levels, terrestrial and marine animals exposed to elevated IHg can suffer a variety of adverse effects, including muscle weakness, memory problems, impacts on hearing, sight, digestive, nervous, and immune systems, as well as lung, kidney, and skin disorders (Bernhoft, 2012; Dietz et al., 2013; Eagles-Smith et al., 2018; Natasha et al., 2020). Similarly, IHg exposure also causes increased incidence of cell morphological abnormalities in plankton, decreasing the biomass of lower marine trophic levels (Beauvais-Flück et al., 2018). In terrestrial environments, high soil levels of IHg cause severe toxicity in plants that compromises ecosystem viability, and decrease plant biomass by causing seed injury, hastening plant senescence, inhibiting photosynthesis, denaturing and inactivating enzymes, and disrupting cell and organelle membrane integrity (e.g., Natasha et al., 2020). Our results suggest that for the shortest eruption period (40 k.y.), IHg levels in the Permian terrestrial and upper ocean systems may have been enriched >450 times above normal during eruption phases (median result of 100 eruption models) (Fig. 4). For the longest modeled eruption scenario (640 k.y.), this may still have been >150 times above normal.

Quantifying changes in MeHg levels is more challenging given uncertainties in the modern Hg cycle. However, changes in global marine systems caused by the Siberian Traps would mostly have increased the processes of Hg methylation. Given the first-order relationship between IHg concentration and the rate of MeHg formation (Mason et al., 2012), enhanced volcanic Hg loading alone would increase microbial MeHg production. MeHg production and rates of MeHg incorporation into sea life are also both positively correlated with temperature (Schartup et al., 2019), such that rapid ocean temperature increase driven by CO_2 emissions of the Siberian Traps (Sun et al., 2012) would have greatly increased Hg methylation rates. Similarly, because the highest methylation rates are associated with acidic environments (Ullrich et al., 2001), reduced ocean pH during the LPE event (Beauchamp and Grasby, 2012) would have further enhanced MeHg generation. Anoxic environments are also primary locations for microbial methylation (Sonke et al., 2013); as such, increased anoxia at the LPE (Wignall and Twitchett, 2002) would have greatly expanded environments in which Hg methylation is favored. Given this, we argue that MeHg enrichment factors, relative to normal marine conditions, would have been at least similar to those of IHg, if not greater. Increased MeHg production would have been partially mitigated, however, by transition to euxinic ocean conditions and HgS drawdown (Sanei et al., 2012; Grasby et al., 2019). In modern oceans, MeHg levels are 0.01–0.1 ng/L in regions farthest from anthropogenic sources. If correct, our results suggest that late Permian upper oceans may have had MeHg levels of several to tens of nanograms per liter. Such high MeHg concentrations would have had extreme toxic impacts on the aquatic food chain, above those of IHg alone.

CONCLUSIONS

Our results suggest that the Siberian Trap eruptions would have driven repeated toxic stress on global marine and terrestrial environments during the LPE. These results support suggestions that enhanced volcanic Hg emissions were a contributing factor to extinction processes during that time. Such repeated toxic shock would have represented an

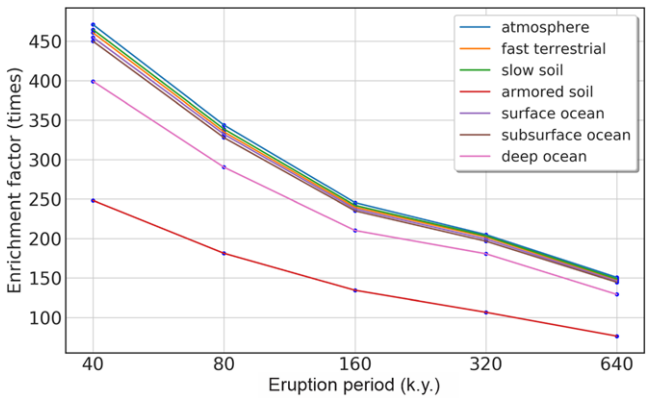

Figure 4. Plot showing median result of 100 eruption models for the maximum enrichment of inorganic Hg in different Hg reservoirs (see Fig. 1) as a function of the modeled eruption period of the Siberian Traps.

additional extinction driver that has not been previously considered, however it may help explain the breadth of extinction at that time—from marine life to insects. For instance, Hg toxicity could explain how even oxic refugia during the LPE (Proemse et al., 2013) did not help to preserve life. Furthermore, Hg toxicity could provide an explanation for the known occurrence of mutated pollen during the LPE (Visscher et al., 2004; Hochuli et al., 2017). Toxic Hg pulses may also explain the closely linked extinction of marine and terrestrial life (Chu et al., 2020). It remains unlikely, though, that Hg contamination was the sole cause for the mass extinction, because decreases in dissolved oxygen and pH known to have occurred during that time are additional critical stressors on marine life. These results suggest that mass-extinction processes that impacted the breadth of life during the late Permian are complex and likely involved multiple environmental stressors that left few survivable niches on the planet.

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