This paper examines the environmental health impacts of Colonial-era mercury mining in Huancavelica, Peru and the historic use of mercury for silver mining in Potosi, Bolivia. Specifically, this paper evaluates the consequences of mercury and silver mining across a range of impact scales that begin at the level of the individual and expand to include the household, the workplace, the community, the region and the world. These scales are not hierarchical in the sense of suggesting that *individual* impacts are less significant than impacts on a community level, but rather that the *aggregate* impact of exposures and activities increases in moving outward from the individual toward the global scale. Likewise, the impacts considered in this environmental health assessment do not necessarily suggest an evolution or outward expansion of consequences. That is, a household - level impact does not necessarily suggest inevitable progression toward a community - level impact (or wider), but rather serves to highlight that different activities – in the case considered in this paper, related to mining - have different types of impacts that may occur on inherently different scales.

With respect to the concept of evolution or progression, Merson et al. (2012) note that in evaluating environmental health impacts, cross scale effects should be considered that recognize that "*the scale at which an environmental health impact eventually occurs may not be the scale at which the exposure was initiated*." This concept of cross-scale impact introduces an aspect of time to an environmental health assessment; that is, an activity with local-scale effects now, may, for example, create consequences on a regional (or wider) scale in the future, and those future

effects may be in terms of the spread of impacts (such as with the widening distribution of a chemical release), the spread of the perception of impact (such as with an improved understanding over time of the environmental and/or health-related consequences of an activity), or the spread of the consequences of impact (such as with impacts that may occur over generations from exposures that have transcended the time scale of activities and/or the spatial scale of the individuals or communities initially impacted).

The specific issue explored in this paper is the Colonial-era (1550 - 1800) mining of mercury in Huancavelica Peru and, relatedly, its use in the processing of silver ore in Potosi, Bolivia. While mercury has been mined and traded in Peru for millennia (Cooke et al. 2013), it was the Spanish who initiated the first large-scale mine operations in Latin America. As context for their interest in Huancavelica, in 1546 Spain had made almost simultaneous discoveries of silver in Zacatecas, Mexico and Potosi, Bolivia. The discovery of significant silver ore in the New World had quickly highlighted the challenges with contemporary ore extraction techniques and in 1554 this problem was solved by the refinement of the *patio process* – a low temperature processing technique for silver involving amalgamation¹ of silver ore with liquid mercury. Spain at that time had no shortage of mercury, as the Almaden mine in southwest Spain was (and still is) the largest mercury deposit in the world.

¹Amalgamation refers to the binding of silver or gold with liquid (elemental) mercury. In the *patio process*, crushed silver ore is placed in a trough or vessel and mixed with elemental mercury. The mercury binds to the silver in the ore, and because mercury is more dense then the remainder of the ore waste rock, the mercury:silver mixture can be recovered by sluicing or washing the mixture with flowing water. Once the waste rock is removed, the amalgam is heated to drive off the mercury and what is left behind is concentrated and refined silver mineral.

With an abundance of mercury in the Old World, and the discoveries of silver in Zacatecas and Potosi in the New World, Spain began a ~ 250 year period (1550 – 1800) of significant extraction and transport: mercury from Spain to the Americas, and silver from the Americas to Spain. The difficulty inherent in transporting mercury from coastal Mexico - where the Spanish delivered the Old World supply - to the silver mines in Bolivia was relieved by the 1564 discovery of the Santa Barbara mine in Huancavelica. From that point forward, Peruvian mercury was carried to Potosi, and silver was transported back – first to the Pacific Coast of Peru, and then ultimately, by galleon, back to Spain. Overall, throughout this era, ~ 130,000 tons of mercury were transported to the New World from the Almaden mine, with an additional ~65,000 tons mined at Huancavelica (Hylander and Meili 2005; Robins and Hagan 2012). In terms of the scale of this endeavor, it has been estimated that during this period Spain was responsible for ~ 80% of global silver production (Nriagu 1994; Guerrero 2012), and was, in parallel, the dominant global consumer of mercury (Horowitz et al 2014).

While it is no understatement to consider the economic consequences of this mining and transport endeavor as having been a driving force in shaping the modern world, the environmental health impacts have, until recently, been largely unexplored and invisible. What is, was, and has always been known, however, is that mining is dangerous. In Huancavelica, not only were miners subjected to the physical injuries and respiratory illnesses common with all underground mining, but both the high altitude in Peru and the mercury mineral itself created hazardous conditions. During the first 35 years of mine operation in Huancavelica - while the miners were slowly creating underground passageways as they followed the mineral down into an open pit and then out into seams in the ore - the adits in which they worked were not

ventilated (Brown 2017). Mercury vapors are toxic, and by 1600, conditions had grown so hazardous in the Santa Barbara mine that mineral extraction from the underground, unventilated seams was banned by the Viceroy of Spain. Contemporary accounts of conditions in the mine described it as "the most vivid image of hell," writing "we know how one bad night [can] break the strongest and well-nourished man. For these unfortunates [working in the mine], all of the nights are very bad. They rise and descend [the ladders] overloaded with one hundred pounds of weight, through caverns filled with horror and risk, that look like the rooms of devils." [Robins 2011]. By 1604, however, while fully aware of conditions at Santa Barbara, the Crown had replaced the Viceroy and re-opened the mines. In the Crown's calculus of valuing mine output over health or safety, it was not until 1643 that the mine's first ventilation shaft was completed (Brown 2017).

Regarding individual exposures, in both Huancavelica and Potosi the mines were worked principally by indigenous laborers through a system called the *mita*. As detailed by the Crown, the *mita* initially required all men between the ages of 18 and 50 (the *mitayos*) to present themselves for a term of service every seven years, working 2 months in Huancavelica or 12 months in Potosí (Robins and Hagan 2012). In the early years of operation at Huancavelica, the *mita* generated ~ 3,000 laborers annually (Brown 2017). Conditions in the mine proved so dangerous, however, that by the time the ventilation shafts were constructed in the mid-1600s, the *mita* had declined to ~ 600 laborers per year and contemporary critics were reporting that mothers were intentionally maiming their sons to avoid the term of service (Brown 2017). The decrease in *mitayos* over this interval resulted from a combination of factors including disease (mining-related and otherwise), injury (including maiming), migration from the areas to avoid

conscription, and mortality as the result of mine service. It has been estimated that approximately 1/3 of the indigenous miners who were conscripted to work at Huancavelica either perished during their term of service or in the near aftermath (Robins 2011). While free wage laborers also worked the Santa Barbara mine and were not subject to the overt coercion forced upon the *mitayos*, it was recognized even at the time that the indigenous community's needs for income were distorted by practices such as requirements to pay tributes to the Crown for goods, protection and the benefits of Christianization (Brown 2017).

In examining the history of mercury mining in Huancavelia and silver in Potosi (as well as in Mexico), it is important to evaluate both the cultural legacy of mining - in terms of forced labor, physical and psychological injury, out-migration and mortality - with resultant impacts on the household, community, and regional scale – and the environmental legacy of the manner in which these metals were mined and processed. Regarding the spatial scale of the environmental legacy, the form of mercury released into the environment plays a significant role in both the scale of its distribution and its potential hazard for those exposed. In the context of the mining operations considered herein, mercury release into the environment occurred principally in three forms: as particles of cinnabar (HgS) lost during the grinding and processing of mercury ore; as liquid droplets of calomel (Hg₂Cl₂), the form of mercury used in amalgamation and lost during the washing of silver ore; and as Hg⁰, the volatile (vapor) form of elemental mercury that can be lost to the atmosphere when either cinnabar ore or calomel-silver amalgam are heated.

It has been estimated that over the ~ 250 years that mercury was mined and used in the extraction of silver in the New World, $\sim 120,000$ tons of Hg were released to the environment in

a combination of particulate, liquid and gaseous forms (Guerrero 2012; Cooke et al. 2013). Estimates of the relative distribution of releases between these forms of mercury suggest that calomel release into surface water - principally via the re-direction of streams through the silver processing works in Potosi - likely accounted for > 65% of total mercury loss (in Potosi, as well as in Mexico). Loss via erosion of mine tailings (in Huancavelica) and volatilization of Hg⁰ (everywhere it was used in either roasting cinnabar or processing silver ore) accounted for the remaining ~ 35% of mercury release (Guerrero 2012).

In terms of spatial scale, the release of dust/particles and calomel would have been local and principally controlled by wind-driven erosion (for dust) and stream transport (for calomel and river-borne particles of mineral silt). For Hg^0 , because the residence time of mercury vapor in the atmosphere is on the order of 1 year (Cooke et al. 2009), the spatial scale of volatile Hg^0 distribution - although a small fraction of total mercury release from mine operations - would have been local, regional and global. Overall, during the Colonial period of mercury mining in Huancavelica, it has been estimated that ~ 17,000 tons of mercury vapor (i.e., volatilized mercury) were released into the atmosphere (Robins and Hagan 2012). This mass estimate assumes an emissions rate of ~ 25% of mercury processed, consistent with data from other contemporary locations in which crude-scale cinnabar processing occurs (e.g., Li et al. 2008; Robins and Hagan 2012). Because none of the mercury mined in Huancavelica was intended for export beyond use in Potosi, it is reasonable to assume that the majority of the remaining 75% of mercury processed in Huancavelica - an estimated ~48,000 tons - was ultimately lost to the local environment in Peru as dust/particles and/or lost in Potosi as calomel or vapor emissions.

With respect to the health implications of this mercury release to the environment, along with volatile releases from the ore body itself in Huancavelica, release of mercury vapors during cinnabar processing would have occurred principally during the roasting of HgS ore. During this process, ore is ground and heated in a closed vessel; in a system in which the vessel has a wellsealed lid, the volatile Hg⁰ released during heating would be captured by the lid and condensed to liquid mercury as the vessel cools. In the absence of a well-sealed lid, or if the vessel was opened before it had completely cooled, individuals working in the roasting works would have been exposed to mercury vapors. Because working in the mining district was also often a family occupation as women and children followed their menfolk during their term as *mitayos*, men, women and children all suffered these vapor exposures. Symptoms of mercury vapor intoxication recognized even during that era included uncontrollable tremors, numbness, motor weakness, irritability, emotional lability, difficulty with vision, drooling and anemia (Robins 2011). The contemporary Jesuit priest José de Acosta wrote that "if some smoke or vapor comes to the people who open the pots [early], they get mercury poisoning and die, or remain in a very bad state or lose their teeth." (Robins and Hagan 2012). Additionally, because mercury vapor crosses the placental - as well as the blood/brain – barrier, pregnant women working the roasters were also at increased risk of spontaneous fetal abortion and/or of infant birth with significant mental and physical developmental abnormalities.

What is important to understand with these exposures is that they were both continuous and community – wide. Antonio de Ulloa, the governor of Huancavelica from 1758 - 1764 described the atmospheric thermal inversions that trapped smelter emissions in the valley, writing that "*the sulphourous smoke that they continually breathe, coming from the ovens in*

which they extract the mercury, which are in such abundance, that in summer time with the freezes, form a dense cloud, that covers the area of the town." (Robins and Hagan 2012). Contemporary atmospheric transport modeling has suggested that the near-ground mercury vapor concentration in the air in the Huancavelica valley during the 1600s may have been commonly in the range of 0.01 - 0.1 milligrams per cubic meters (mg/m³), with concentrations in the immediate vicinity of the roasters exceeding 1 mg/m³ (Robins 2011). These modeled concentrations are consistent with concentrations measured in the vicinity of contemporary small-scale gold processing operations,² in which mercury vapor concentrations over an 8-hour period can average 0.2 mg/m³ and reach 6 mg/m³ (Gibb and O'Leary 2014).

As context for evaluating these exposure concentrations, the World Health Organization (WHO) has defined the chronic exposure limit for mercury vapor as 0.001 mg/m³, based on *annual* averaged exposure (WHO 2000); the USEPA has defined a Lowest Observed Adverse Effects Level (LOAEL) of 0.009 mg/m³ associated with the development of hand tremors, memory disturbance and autonomic dysfunction³; and OSHA has defined the acute permissible exposure limit (PEL) for mercury vapor as 0.1 mg/m³, based on an 8-hour work day.⁴ As compared to these threshold exposure values, the *background continuous* near-ground mercury vapor concentration in the Huancavelica valley during the height of mine operations in the 17th

²This mode of mercury use - in what has been termed 'artisanal small-scale gold mining' (ASGM) - continues to expose individuals in Peru, as well as elsewhere, to unsafe levels of mercury. Exposures related to ASGM result principally from vapor released during ore processing (via methods as crude as applying a blowtorch to an open pan of mercury-gold amalgam) and, secondarily, as the result of food chain contamination through conversion of waste mercury to methylmercury in local/downstream aquatic environments (e.g., van Straaten 2000; Maramba et al. 2006).

³<u>https://cfpub.epa.gov/ncea/iris/iris_documents/documents/subst/0370_summary.pdf#nameddest=rfc</u> [accessed 11/9/17]

⁴https://www.osha.gov/dts/chemicalsampling/data/CH_250510.html [accessed 11/7/17]

century was likely in the concentration range associated with observable physiological, behavioral and neurological effects and these effects were not limited solely to the miners.

Whereas the mercury was mined in Huancavelica and appears to have resulted in documentable health impacts across the mine area and throughout the local (and regional) indigenous communities, the scale of overall impacts from mercury exposure were likely even greater in Potosi. This increase in impacts across scale in Potosi is due to both the significantly greater population in Potosi during the height of the Colonial era (~160,000 in Potosi in the 1660s versus ~15,000 in Huancavelica) and the specific impacts of the methods used to recover and concentrate the silver ore. Exposure to mercury and release of mercury vapors during silver processing would have occurred both during the amalgamation process - in which elemental mercury was physically mixed with crushed silver ore - and again during the heating (or retorting) of the amalgam to boil off the mercury and recover the refined silver. With these processes - and analogous to conditions in Huancavelica - environmental releases and occupational exposures would have occurred simultaneously. The mixing of elemental mercury and silver ore was typically accomplished by treading the mixture in a trough or *patio*. For those employed in treading the amalgam, exposures resulted principally from direct contact, as work entailed standing barefoot and knee-deep in a mixture of elemental mercury and crushed silver ore for weeks at a time (Robins 2011). A further health risk associated with this labor was frost bite, with the altitude in Potosi and the conditions within the patios rendering this a persistent background concern (Robins 2011).

Retorting the mercury-silver amalgam was accomplished in a manner similar to the roasting of mercury ore and with as little attention paid to the protection of human health. In Potosi, the amalgam – once recovered from the patio – was washed, packed into a cloth bag, squeezed to remove excess water and then placed into a mold and fired. Molds were designed with a conical vented lid that allowed recovery of some, but not all, of the volatilizing mercury vapor. At the height of silver production in Potosi, there were as many as 30 amalgamation processing facilities operating within the city limit and discharging mercury to the environment (Higueras et al. 2012). It has been estimated that \sim 39,000 tons of mercury was released into the environment during the processing of silver ore at Potosi (Robins and Hagan 2012), and while the majority of this release was likely as calomel lost to receiving streams during the washing of the amalgam, some fraction - as in Huancavelica - would have been as mercury vapors lost from the unsealed lids on the retorting molds.

With respect to the question of time, the most significant and acute long term legacies of mercury mining activities in Huancavelica, as well as in Potosi, are local ones. Research examining contemporary mercury vapor concentrations in the air in Potosi has documented average (mean) vapor concentrations in the vicinity of historic retorting facilities that remain elevated relative to regional background concentrations (> 30 nanograms per cubic meter [ng/m³] versus < 4 ng/m³, respectively; Higueras et al. 2012), although the generally consistent and overlapping (and low) range of vapor concentrations between locations adjacent to historic processing facilities and regional background concentrations confirm that mercury is no longer being used in mineral processing in Potosi. In contrast to these low regional contemporary atmospheric vapor phase concentrations of mercury, however, vapor concentration exceeding

 3000 ng/m^3 have been documented following localized disturbance to the soil in a historic processing neighborhood in Potosi (Higueras et al. 2012). For context, and as presented earlier in this paper, this concentration is $3 \times$ the recommended chronic exposure limit for mercury vapor based on an *annual* averaged exposure (WHO 2000).⁵ While this comparison is not to suggest that the mercury vapor concentration measured following soil disturbance necessarily represents a widespread chronic exposure concern, these data do highlight the presence of legacy contamination in the soil of Potosi and suggest that atmospheric volatilization loss and potential resultant exposures may remain as site-specific on-going concerns.

Likewise, contemporary analysis of mining residuals in sediment of the Rio Pilcomayo the river receiving stream influx from the watershed in which Potosi is located - has documented elevated concentrations of mercury in river margin (alluvial) deposits over a distance of ~ 50 kilometers (km) downstream from Potosi, with mercury concentrations in these deposits reaching 5 micrograms per gram (or parts per million [ppm]) (Hudson-Edwards 2001). This mercury concentration in river sediments is significantly lower than mercury concentrations measured in adobe bricks in both Huancavelica and Potosi (discussed further below), but is still ~ 2 orders of magnitude higher than background concentrations in sediment from rivers not impacted by mining or industrial discharges (Ames and Prych 1995, Senesi et al 1999; McEwen et al. 2016). Although the highest concentrations of mercury measured in contemporary Rio Pilcomayo sediments are not in the surface layers of the river alluvium, and so are somewhat stably buried by more recent sediment deposition, the potential for erosion of these deposits -

 $^{^{5}3000 \}text{ ng/m}^{3} = 0.003 \text{ mg/m}^{3}$; WHO chronic exposure limit = 0.001 mg/m³ (WHO 2000)

almost 500 years after mining began in Potosi - is an on-going concern for mercury, as well as for other metals mined more recently in this watershed. Moreover, as the biological availability of these sediment deposits is not well constrained, it is not clear to what extent calomel deposits remain as insoluble solids with low biological availability in aquatic environments or whether these deposits exhibit the potential for environmental (microbiological) transformation into more biologically available forms of mercury (such as methylmercury) that could result in food web uptake and trophic transfer to humans. Studies of food web transfer downstream of mercury mining sites in other locations suggest, for example, that when discharged into aquatic environments, cinnabar waste rock can be solubilized and methylated, resulting in elevated fish tissue methylmercury concentrations and fish consumption advisories for downstream watersheds (e.g., Maramba et al. 2006). It should be noted that in countries and locations (such as in rural Peru and Bolivia) in which consumption advisories do not exist, the absence of advisories may speak more to the absence of fish tissue data and associated public health interventions than to any assumptions regarding lack of risks or exposure – related concerns.

Of additional significant concern regarding potential exposures in rural areas of Peru and Bolivia is that communities in both locales still build residences out of unsealed adobe brick. In both Huancavelica and Potosi, studies have documented mercury concentrations in residential mud bricks and dirt floors exceeding ~ 500 ppm (Hagan et al. 2013; McEwen et al. 2016), a concentration that is orders of magnitude higher than 'background' concentrations in the soil of non-mining areas (e.g., Ames and Prych 1995, Senesi et al 1999; McEwen et al. 2016), and consistent with soil concentrations measured in the vicinity of contemporary mercury mine operations (Higueras et al. 2003; Maramba et al. 2006; McEwen et al. 2016). Recent studies in

Huancavelica have documented that the concentration of mercury in samples of human hair, as well as in adobe bricks and surface dust, is significantly higher for residents and households from within historic cinnabar smelting districts (n = 87) than for residents and households from districts in which historic mercury processing did not occur (n = 30) (Hagan et al. 2013; Hagan et al. 2015). Data presented in Hagan et al. (2015) also suggest that mercury concentrations in hair samples are significantly higher (p < 0.001) for men (n = 21) than for women (n = 95). Because the relationships between mercury concentrations in hair samples and in samples of adobe bricks, dirt floors, and surface dust are positively correlated, but not statistically significant (n = 34; p > 0.05), Hagan et al. (2015) suggest that the gender-based relationships observed may be influenced by local and regional occupational choices – such as work in the construction trade – that result in increased potential exposure to contaminated dirt and dust for men.

Although hair is more commonly used as a biomarker for methylmercury exposure (typically via the ingestion of mercury-contaminated fish) rather than as a biomarker for exposure to particulate mercury dust (which is more typically assessed via analysis of blood or urine), and the potential for exposure to mercury as vapor released from adobe brick has not yet been examined, these studies do suggest that, within adobe homes, contact with mercuryenriched dust may create an exposure route analogous to the household exposure route for lead (Pb) from historic use of Pb-based paints. While both mercury and lead are known toxins associated with potential neurological and developmental impacts/delays in children, it is not clear, however, to what extent the similarity between these metals translates into similar biological availability or neurological effects, particularly with hand-to-mouth transfer of dust and/or dirt. Whereas it is likely that the biological availability of mercury associated with mining

dust may be lower than the biological availability of lead associated with house paint and is most certainly lower than the biologically availability of either mercury vapor or methylmercury in fish tissue, the ability to assess the impact of contemporary mercury exposure via contaminated dust is hindered by the current lack of an evaluated reference dose (RfD) for ingestion of particulate mercury.⁶

Assessing contemporary health impacts of legacy mining is further complicated by a range of factors including limitations on the public health infrastructure in rural Peru and Bolivia (both in terms of human health evaluation and reporting of observable effects), as well as the necessity of identifying and integrating the range of existing health challenges that may have little to do with mercury exposure, but may either increase susceptibility to mercury-related effects or result in overlapping symptoms that complicate diagnoses. While chronic exposure to inorganic mercury in other forms has been associated with kidney damage and stomach and thyroid tumors in adults, these conditions are not solely correlated with mercury exposures; for children, in potential contrast to adults, it may be prudent to assume that, as with Pb, there may be no safe threshold exposure concentration. Overall, while national and international attention has recently begun to focus on the legacy environmental health concerns resulting from the areas' mining histories,⁷ and actions are being taken to address the potential for chronic mercury exposures that have resulted from this intersection of historical practices and contemporary rural community life, the long term human health and environmental consequences of Colonial-era mercury and silver mining in Latin America remain under-examined.

⁶ US EPA currently defines a maximum daily methylmercury reference dose (RfD) of 0.1μg/kg body weight with the exposure route being dominantly through consumption of fish tissue (USEPA 2001). ⁷e.g., Environmental Health Council (www.ehcouncil.org); Pure Earth (http://www.pureearth.org)

This paper has evaluated the environmental health legacy of Colonial-era mining in Huancavelica, Peru and Potosi, Bolivia via a framework that examined a range of impacts including those on the personal, community, regional and global scales, as well as across time. During the 250-year period that defines the Colonial era in Latin America, ~65,000 tons of mercury was mined in Peru alone, with environmental releases from this industrial activity occurring as dust/mineral particles, as mercury-enriched compounds released into surface water and as mercury vapor released into the atmosphere. On local and regional scales, the impact of Latin American mercury mining was severe, and contributed significantly to mental and physical illness, community decimation via out-migration and mortality from mercury toxicity. Importantly, as a function of how mining and mineral processing operations were conducted in both Huancavelica and Potosi, significant direct occupational exposures extended beyond conscripted mine workers to also include their families and communities. From an environmental social justice perspective, it is interesting to consider that one consequence of community-level exposure to elevated mercury vapor concentrations was that those individuals responsible for mine operations and enforcement of the *mita* – overseers, prelates, and political and economic agents of the Crown - would have lived with similar exposure risks to the *mitayos*. That is, the health impacts of chronic mercury exposure would have transcended class and likely contributed in undefinable ways to the brutalizing conditions that agents of the Crown created and perpetuated and that indigenous communities were forced to endure.

In regard to the temporal scale of impacts, the cultural and environmental legacy of these community-scale exposures continues into the present; ~ 200 years after the Colonial era ended

in Latin America, residents in Huancavelica and Potosi are still affected by legacy mine wastes via residual soil contamination, the potential for fish consumption concerns and use of adobe mud bricks in construction of traditional homes. Moreover, although mercury is no longer used for the commercial extraction of silver, it continues to be used in countries including Peru for the small scale, sometimes illicit, processing of gold. Gibb and O'Leary (2014) report, as example, that for cross-sectional studies assessing health impacts of artisanal small scale gold mining (ASGM) and mercury exposure, the most frequently documented effects are neurological and autoimmune and that, importantly, these effects appear both in mine workers and members of the community.

Addressing these health impacts as well as the overall environmental health consequences of both legacy and contemporary mining and ore processing is a significant global health concern and requires the skills of public health professionals, community health workers, scientists, engineers, non-governmental organizations, international agencies and both local/regional and national governments. Working together, the extent of mercury exposures and resultant toxicological impacts can be evaluated and a framework for mitigating and remediating those impacts can be developed. That during recent decades ~ 50% of the largest single capital investments in mine operations globally have been made in Latin America, with two of these investments in particular occurring in Peru (Bebbington et al. 2008), suggests that the potential for cross-scale impacts– both spatially and temporally – from mine activities remains an on-going global health concern.

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