The Effect of Particle Composition and Concentration on the Partitioning Coefficient for Mercury in Three Ocean Basins

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The downward flux of sinking particles is a prominent Hg removal and redistribution process in the ocean; however, it is not well-constrained. Using data from three U.S. GEOTRACES cruises including the Pacific, Atlantic, and Arctic Oceans, we examined the mercury partitioning coefficient, $K_d$, in the water column. The data suggest that the $K_d$ varies widely over three ocean basins. We also investigated the effect of particle concentration and composition on $K_d$ by comparing the concentration of small-sized (1–51 µm) suspended particulate mass (SPM) as well as its compositional fractions in six different phases to the partitioning coefficient. We observed an inverse relationship between $K_d$ and suspended particulate mass, as has been observed for other metals and known as the “particle concentration effect,” that explains much of the variation in $K_d$. Particulate organic matter (POM) and calcium carbonate (CaCO₃) dominated the Hg partitioning in all three ocean basins while Fe and Mn could make a difference in some places where their concentrations are elevated, such as in hydrothermal plumes. Finally, our estimated Hg residence time has a strong negative correlation with average log bulk $K_d$, indicating that $K_d$ has significant effect on Hg residence time.

Keywords: partitioning, GEOTRACES, particle concentration, particle composition, marine particles, mercury

INTRODUCTION

Removal of trace elements by particle scavenging has a first-order control on their concentrations in the ocean (Anderson, 2014). This is true for mercury (Hg) as well: particle scavenging represents the ultimate sink for Hg from the ocean over centuries, and it is eventually back to the deep mineral reservoir on the timescales of glacial cycle (e.g., Amos et al., 2013). At present, the global burial flux for Hg in ocean sediments is not very well-constrained but likely lies between the value of 1.2 µg m⁻² y⁻¹ in open ocean conditions and 1,200 µg m⁻² y⁻¹ in hyper-accumulating regions like the Antarctic margin (Soersen et al., 2016; Zaferani et al., 2018). Models have suggested that the global burial flux falls between 1.7 and 7 µg m⁻² y⁻¹ (e.g., Outridge et al., 2018; Tesán Onrubia et al., 2020). Given the current oceanic inventory of Hg, these burial fluxes suggest a residence time of Hg of about 520 years (Outridge et al., 2018).
Mercury likely finds its way from solution onto suspended and sinking particles via processes that John and colleagues have called “regenerative scavenging” (John and Conway, 2014; Lamborg et al., 2016). This mechanism is unlike that exhibited by trace elements such as thorium, for which reversible partitioning appears to occur and results in steady-state particle-/aqueous-phase ratios that are dependent on the composition of the particulate phase (Bacon and Anderson, 1982; Chase et al., 2002; Hayes et al., 2015). In regenerative scavenging, some irreversible process such as biological uptake in surface waters is responsible for the movement of an element from the solution to the particle-phase initially, and the return reaction is only accomplished when the particles themselves are altered or destroyed, forcing the trace elements (and macro-material) back into solution followed by re-association with the altered particles. A combination of non-reversible and reversible processes broadly describes the nature of all “hybrid-type” elements and thus we should expect that their distribution in and removal from the ocean to be controlled by regenerative scavenging. To best understand and describe such cycling, we would benefit from a study of the kinetics of sorption, desorption, uptake, and remineralization as has been done for thorium (e.g., Lerner et al., 2018). However, unlike for thorium, sources of Hg are not well-known making a mass balance inverse approach more difficult, and we are therefore forced to rely on some proxy information to guide our understanding. The empirical distribution coefficient ($K_d$) is one such proxy and has been used extensively in modeling studies for a variety of elements (e.g., Honeyman and Santoshi, 1989). However, in the case of Hg it has been noted that the value of $K_d$ remains poorly constrained (Zhang et al., 2014). We recently examined the magnitude and variability of $K_d$ of Hg in the Atlantic Ocean using data generated from the U.S. GEOTRACES and found that $K_d$ values were substantially higher than had been assumed in the past and that the particle phase with the strongest apparent affinity for Hg was manganese oxide (MnO$_2$), followed by iron oxyhydroxide [Fe(OH)$_3$], particulate organic matter (POM), calcium carbonate (CaCO$_3$), and lithogenic particles in decreasing order (Lamborg et al., 2016). However, as the oxide phases represented only a small fraction of total particulate mass (0.8% for iron and 0.1% for manganese), their apparent contribution to overall Hg sorption (if indeed Hg is to be found in marine particles as a result of sorption) was modest. Instead, POM was the most important phase with its sorption fraction at 36%, CaCO$_3$ next at 30%, and lithogenic phase at 29%. These findings supported the generally held hypothesis that POM is highly influential in determining the phase distribution of Hg in the environment (Fitzgerald and Lamborg, 2014). However, it also suggested that the influence of POM is not exclusive and perhaps not even responsible for the majority of Hg partitioned to the particle phase. In this manuscript, we extended the Lamborg et al. (2016) Atlantic examination to two additional basins including the eastern tropical South Pacific Ocean and the Western Arctic Ocean. We also subject the datasets to additional statistical analyses in search of a predictive model of Hg phase partitioning. The analysis here makes use of total dissolved and particulate Hg but could be performed on monomethylmercury as well. The contributions of monomethylmercury to total dissolved and particulate Hg are relatively small and so were not removed prior to our calculations. Therefore, the $K_d$ values derived using total Hg values are close approximations for the Hg(II) form.

**METHODS**

**Data Used**

All datasets used here are from U.S. GEOTRACES cruises, including the East Pacific zonal transect (nickname EPZT; GEOTRACES transect number GP16, cruise identifier TN303), the North Atlantic zonal transect (NAZT, GA03; KN199-04 and KN204-1), and the Western Arctic (GN01; HLY1502). We used data from 58 stations in total with 22 in the Pacific, 17 in the Atlantic and 19 stations in the Arctic Ocean (Figure 1). All data used were from the BCO-DMO data repository.

The Hg data have all been published previously, with the dissolved and particulate total mercury data from Agather et al. (2019) (GN01) and Bowman et al. (2015, 2016) (GA03 & GP16). The published particle mass and composition data are from Lam et al. (2015, 2018), Xiang and Lam (2020). Due to lack of access to large-size-fraction (>51 μm; LSF) samples for Hg analysis, we only used small-size-fraction (1–51 μm; SSF) particle composition and particulate Hg data in our mercury partition coefficient analysis. Note that the original collected data were linked with “flags” indicating the data quality, in order to have convincing results, we made sure that all parameters we used were identified as either “good” (Quality Flag = 2) or “possibly good” (QF = 3) quality at the same depth.

**Partition Coefficient ($K_d$) Analysis**

Here, the partition coefficient ($K_d$) is defined as the apparent affinity of mercury for marine particles. This concept was inspired from studies of Bacon et al. (1976) and Baskaran and Santschi (1993). $K_d$ is calculated as:

$$K_d = \frac{C_p}{C_d} \times \frac{1}{SPM}$$

where $C_p$ and $C_d$ represent the concentration of Hg in particulate and dissolved phases, respectively, and SPM is the suspended particulate mass (kg L$^{-1}$). This results in $K_d$ having units of volume per mass, conventionally as L kg$^{-1}$. Here $C_d$ represents the concentration passing a 0.2-μm filter and therefore includes both dissolved and colloidal Hg. Particle sampling depths at each station were not exactly the same as the dissolved phase, thus, to calculate partition coefficients, we estimated concentrations of the dissolved phase by linear interpolation (using the MATLAB routine “interp1”) to the depths of particle phase samples.

In order to better understand how bulk partition coefficients relate to particle composition, a six-end member mixing model was applied, which is similar to the one developed by Hayes et al. (2015) for thorium isotopes. The contribution of each major
FIGURE 1 | (A) Full map of the three GEOTRACES cruises including GP16, GA03, and GN01; (B) Zoomed in map of GN01. Purple dots represent the location of the sampling stations referred to in the text.

Particle phase to the measured bulk $K_d$ is represented by the following formula:

$$K_d = \sum_{i=1}^{6} (K_d)_i \cdot f_i = (K_d)_1 \cdot f_1 + (K_d)_2 \cdot f_2 + (K_d)_3 \cdot f_3 + (K_d)_4 \cdot f_4 + (K_d)_5 \cdot f_5 + (K_d)_6 \cdot f_6$$

where $K_d$ is the observed bulk partition coefficient, $(K_d)_i$ is the apparent partition coefficient for pure phase $i$, $f_i$ is the fraction of certain particle phase $i$ over particulate mass, and $I = 1$ (POM), 2 (CaCO$_3$), 3 (Opal), 4 (lithogenic), 5 (MnO$_2$), 6 (Fe(OH)$_3$). The $(K_d)_8$, which we and Hayes call the intrinsic $K_d$, was calculated using non-negative least-squares regression (“lsqnonneg”) in MATLAB for individual basin datasets, as well as some sub-basin datasets:

$$[f_1^N \ f_2^N \ f_3^N \ f_4^N \ f_5^N \ f_6^N] \times \begin{bmatrix} (K_d)_1 \\ (K_d)_2 \\ (K_d)_3 \\ (K_d)_4 \\ (K_d)_5 \\ (K_d)_6 \end{bmatrix} = \begin{bmatrix} K_d^1 \\ K_d^2 \\ K_d^3 \\ K_d^4 \\ K_d^5 \\ K_d^6 \end{bmatrix}$$

(3)

To estimate the standard error of the derived intrinsic $K_d$ values, we used the jackknife re-sampling technique suggested by Hayes et al. (2015) and Efron and Stein (1981).

Principal Component Analysis of Bulk $K_d$ and Particle Composition

We performed PCA on the partition coefficient and particle composition data to see if a few principal components (PCs), or linear combinations of the original variables, could explain a large proportion of the total variance in dataset being considered. Therefore, PCA was conducted on the following seven variables: bulk log $K_d$, $f_{POM}$, $f_{CaCO_3}$, $f_{opal}$, $f_{lithogenic}$, $f_{MnO_2}$, $f_{Fe(OH)_3}$, in order to analyze the relationship between bulk $K_d$ and particle
composition fraction among three different ocean basins. Since
the units and magnitude of the 7 variables are different, prior
to PCA the sample data were mean-centered and normalized by
standard deviation (“zscore” function in MATLAB). The PCA
itself was performed using the MATLAB function “pca”. In
addition, we constructed biplots of loadings and scores associated
with PC’s 1 and 2 to visualize the results, which interprets
the correlation for each of the original variables as well as the
direction of increasing values.

Principal Component Analysis of Residual
K_{d} and Particle Composition
We did linear regression to model the relationship between
SPM and bulk K_{d}. The residuals K_{d} data were calculated as
the vertical distance in the Y-axis between each data point and
the fitted line and represent the portion of K_{d} not explained
by SPM. Following the same data processing strategy as in
section Principal Component Analysis of bulk K_{d} and particle
composition, we performed PCA on the residuals and particle
composition data to examine whether this would better reveal a
particle composition effect.

RESULTS AND DISCUSSION
In this section, we present our data results and employ a model
to demonstrate the particle concentration and composition effect
on Hg partitioning. In addition to discussion on some special
features of K_{d}, we extended the study of K_{d} on Hg residence time
at the end.

Dissolved and Particulate Hg Profiles
The mean and median concentrations of dissolved and
particulate total mercury for the three ocean basins were
calculated in the surface (above 100 m), as well as every 500 m
depth in the deep water (below 100 m; Figure 2). Overall, the
concentration of dissolved Hg in the Arctic was the highest
(around 1.1 pM) at the surface, as a result of riverine input,
atmospheric deposition and ice melt (e.g., Fitzgerald et al., 2005;
Agather et al., 2019), and decreased to ~0.5 pM at depth. In
contrast, the dissolved Hg in the Pacific and Atlantic Ocean was
distributed in a more nutrient-like way, characterized by a slight
increasing trend with depth. Note that there was a maximum
at 3,500 m in the Atlantic is due to the influence of the TAG
hydrothermal vent where total dissolved Hg increased to 13 pM.

The particulate Hg started with the highest concentration at
the surface in all basins, attenuating quickly below the photic
zone and reaching the minimum at about 1,000 m. In the Pacific
Ocean, a strong hydrothermal plume led to abnormally high
concentrations of particulate Hg in deep water (between 2,500
and 3,500 m), accompanied by high variability as well. Similarly,
the SPM concentration in the three ocean basins attenuated
with depth. At the surface, the mean SPM concentration in
the Arctic was around 6 times greater than that in the Pacific
and Atlantic, resulting from the extremely high SPM over the
continental shelf (Xiang and Lam, 2018). The median surface
SPM concentration in the Arctic was the lowest of the three
basins, however, reflecting the very oligotrophic Canadian Basin
in the Western Arctic Ocean (Xiang and Lam, 2020). A maximum
occurred in the deep ocean of the Atlantic due to the influence of
strong bottom nepheloid layers in the Deep Western Boundary
Current (Lam et al., 2015).

Bulk Partition Coefficient (K_{d}) for the Three
Ocean Basins
Bulk partition coefficient values varied widely over the whole
dataset, ranging in order from 10^{4} to 10^{8} L kg^{-1} (Figure 3).
In the Pacific, the bulk partition coefficients were extremely high, with
an average order of magnitude of 10^{7} (range: 5.05 \times 10^{5}-1.63 \times
10^{8} L kg^{-1}; log K_{d} = 5.70-8.21). The bulk K_{d} values from the
Atlantic, in contrast, were lowest and ranged from 2.60 \times 10^{5} to
6.73 \times 10^{7} L kg^{-1} (log K_{d} = 5.41-7.83). Compared to these two
ocean basins, the K_{d} value from the Arctic Ocean were modestly
lower and ranged from 9.70 \times 10^{4} to 6.99 \times 10^{7} L kg^{-1} with an
average on the order of 10^{6}. Summary K_{d} statistics are presented
in Supplementary Table 1. As previously noted by Lamborg
et al., 2016, these K_{d} values are substantially higher than those
previously reported for freshwaters, sediment porewaters over
continental shelves (~10^{2} L kg^{-1}) (e.g., Hammerschmidt and
Fitzgerald, 2004) and coastal waters (10^{5}-10^{6} L kg^{-1}) (e.g.,
Balcom et al., 2008).

Intrinsic K_{d}
End-member coefficients, or intrinsic K_{d}, were calculated for
each of the six particle phases in the SSF in all three cruises,
including POM, CaCO_{3}, opal, lithogenic, Mn oxides, and Fe
oxyhydroxides. The value of intrinsic K_{d} and composition
fractions are summarized in Table 1. Overall, the intrinsic K_{d}
values varied between ocean basins. One commonality, however,
was that the intrinsic K_{d} for opal could not be defined using this
method in any of the three ocean basins. The intrinsic K_{d} for
POM, CaCO_{3}, and lithogenic phases could always be estimated,
but had different values. As for Fe and Mn, the contribution to
sorption of Hg could be evident when their concentrations were
high, typically observed in hydrothermal vents. Otherwise, their
influence on Hg partitioning was negligible.

If there is really is such a thing as an “intrinsic K_{d}” that
expresses the stickiness of Hg to a measured particle phase,
the calculated values should be homogeneous everywhere in the
ocean. However, an examination of our results suggests that this
might not be true. We applied the Z-test based on intrinsic
K_{d} values and their standard deviations (Table 1), setting the
confidence interval to 95%, to test whether the intrinsic K_{d}
values for each particle phase were significantly different between
basins. Generally speaking, intrinsic K_{d} between ocean basins
were similar for some phases but significantly different for others.
For instance, intrinsic K_{d} values of POM in three ocean basins
were significantly different (P < 0.05). In contrast, the values for
CaCO_{3} of three ocean basins were statistically indistinguishable
(P > 0.05). This suggests that the precise composition of POM
with respect to its ability to scavenge Hg varies between ocean
basins but that CaCO_{3} does not (or varies less). Since the
three cruises were sampled in different seasons and different
environments, the dominant plankton were likely not the same
in the three ocean basins, and the components of collected POM
presumably varied with different plankton. The intrinsic $K_d$ of the lithogenic phase in the Arctic Ocean was different from those in the Pacific and Atlantic Oceans. This is perhaps not surprising, since "lithogenic particles" is a category that represents a diverse range of aluminosilicate minerals, which might be expected to have different sorption characteristics for Hg (e.g., Biscaye, 1965; Darby, 1975). Interestingly, intrinsic $K_d$ values of Mn had no significant difference ($P > 0.05$) in the Pacific and Arctic Ocean, while intrinsic $K_d$ values of Fe were significantly different ($P < 0.05$) between the Pacific and the Atlantic Ocean. The Fe intrinsic $K_d$ differences between basins implies a qualitative difference in the Fe itself, but this remains to be explored.

**PCA of Hg Partitioning and Particle Composition**

The PCA results are summarized in Table 2, which includes 7 variables (the bulk $K_d$ value and the fraction of the six particle fractions), loadings of the first three principal components (PC) and their contribution in explaining the total variance. In the East Pacific, the first three principal components explained 77.64% of variation of the seven variables with 35.33% explained by PC1, 22.78% explained by PC2, and 19.53% explained by PC3. The highest loadings for PC1 were the fractions of Fe and Mn, both of which were positively correlated with bulk $K_d$, which could result from the hydrothermal plume (Lam et al., 2018). For PC2, the relative fractions of POM had strong positive loadings, while $f_{CaCO_3}$, $f_{litho}$, and $f_{opal}$ were negatively loaded, which means the organic matter was anticorrelated with minerals. This negative correlation reflects the variation in depth: decreasing POM and increasing lithogenic particles with depth (Lam et al., 2018). The fractions of CaCO$_3$ and opal were strongly anticorrelated in PC3 (Table 2, Figure 4), which might reflect the variation of region: higher opal in the coastal region, abundant CaCO$_3$ in the open ocean (Lam et al., 2018). In the Atlantic, 63.85% of
total variance was explained by the first three components with 30.56% explained by PC1, 17.64% explained by PC2, and 15.65% explained by PC3. Based on our observations, in PC1, bulk $K_d$ and the fractions of POM were anticorrelated with fractions of lithogenic particles and Mn, reflecting the depth variation of POM and lithogenic particles (Lam et al., 2015). For PC2, the fraction of CaCO$_3$ was strongly correlated with bulk $K_d$, and for PC3 the fractions of opal were anticorrelated with Fe and Mn. Additionally, the first three principal components of the Arctic dataset explained 77.92% of the total variance, where PC1 explained 39.15%, PC2 explained 35.61%, and PC3 explained 13.16%. The highest loadings of PC1 were observed from the fractions of lithogenic particles and POM, which could be the consequence of variations in depth and/or region (Xiang and Lam, 2020), whereas PC2’s highest loadings were mainly from bulk $K_d$, and the fractions of Mn, CaCO$_3$ and opal. The $f_{\text{CaCO}_3}$
From the biplots of scores and loadings (Figure 4), the $K_d$ loadings were strongest with PC1 in the Pacific, and PC2 in the Atlantic and Arctic. As already noted above, PC1 in the Pacific was most strongly associated with Fe and Mn oxides, implying a significant influence of those phases on the overall value of $K_d$. This is likely due to the influence of hydrothermal Fe- and Mn-rich particles sorbing Hg from seawater. In the Atlantic and Arctic, PC2 was also strongly loaded with CaCO$_3$, and relatively poorly loaded for POM, implying a major role of CaCO$_3$ in Hg sorption in those basins, but less of a role for POM.

Furthermore, the biplots loadings (Figure 4) for each ocean basins suggested that the PC1 and PC2 could be driven by different variables. The significant strong positive loadings in PC1 for both Mn, Fe and bulk $K_d$ in the Pacific imply that this PC was related to some natural processes that affected bulk $K_d$ in a similar manner with Mn and Fe in the Pacific. Similarly, PC2 has strong positive loadings for Mn and bulk $K_d$ in the Arctic also suggested these two variables behaves similarly and closely correlated with each other, since the high absolute and relative concentrations of manganese oxides were observed in the Arctic halocline and deep water, leading to strong scavenging of various elements (Xiang and Lam, 2020). However, there is no significant correlation between f$_{MnO2}$ and log $K_d$ in the Atlantic, suggesting that f$_{MnO2}$ can’t be the only control on log $K_d$ globally. Meanwhile, f$_{MnO2}$ is very low in the Atlantic, possibly accounting for its little influence in scavenging Hg.

**Particle Concentration and Composition Effects on Partition Coefficient**

**Particle Concentration Effect**

We began our investigation of the potential drivers of $K_d$ by examining the effect of particle concentration on the partitioning of Hg. The linear regression model in Figure 5 shows the relationship between log SPM ($\mu g$ L$^{-1}$) and log bulk $K_d$ (L kg$^{-1}$) for the three ocean basins. A significant inverse correlation was observed in all basins, which is consistent with the findings reported by Moran et al. (1996). Similar inverse relationships were observed between log $K_d$ of $^{210}$Pb and $^{210}$Po and SPM in these transects (Tang et al., 2017; Bam et al., 2020). The inverse relationship between $K_d$ and SPM, also known as "the particle concentration effect” (Honeyman et al., 1988), can be explained by at least three mechanisms. The first one centers

and f$_{Mn}$ were positively correlated with bulk $K_d$ while f$_{opal}$ was negatively correlated with bulk $K_d$. PC3's highest loadings were the fractions of Mn, CaCO$_3$ and opal which were positively correlated with each other.

**TABLE 1** The log intrinsic $K_d$ (L kg$^{-1}$) of Hg for six particle composition in the small size fraction (SSF) in the Pacific, Atlantic, and Arctic ocean, respectively, and their relative standard deviation (r.s.d.) as well as particle composition mass contribution (f, %).

| Variables | Pacific | Atlantic | Arctic |
|-----------|---------|----------|--------|
| POM       | log $K_d$ | r.s.d. (%) | f (%) | log $K_d$ | r.s.d. (%) | f (%) | log $K_d$ | r.s.d. (%) | f (%) |
| Mn        | 7.24    | 0.40     | 48 ± 23 | 6.67    | 0.64     | 42 ± 21 | 6.94    | 1.24     | 45 ± 23 |
| CaCO$_3$  | 7.02    | 0.56     | 33 ± 19 | 6.86    | 1.13     | 28 ± 11 | 8.23    | 15.56    | 2 ± 2  |
| Opal      | n.a.    | n.a.     | 9 ± 8   | n.a.    | n.a.     | 4 ± 3  | n.a.    | n.a.     | 17 ± 13|
| Lithogenic| 7.37    | 1.57     | 6 ± 12  | 6.29    | 5.04     | 25 ± 20 | 6.50    | 0.78     | 34 ± 27|
| Fe        | 7.25    | 27.45    | 6 ± 0.9 | 6.75    | 2.05     | 7 ± 5.0 | 7.90    | 19.46    | 2 ± 2  |
| POM       | 8.20    | 2.08     | 3 ± 10  | 6.75    | 2.05     | 7 ± 5.0 | n.a.    | n.a.     | 1 ± 1  |

n.a., not available yet.

**TABLE 2** The first three principal component loadings, percentage of variance explained by each principal component, and their cumulative percentage of total variance including bulk $K_d$ and six particle compositions in the small size fraction of particles.

| Variables | PC1     | PC2     | PC3     |
|-----------|---------|---------|---------|
| Pacific   | Bulk $K_d$ | 0.4079  | 0.2223  | 0.119  |
| f$_{POM}$ | -0.4285 | 0.5769  | -0.0387 |
| f$_{CaCO_3}$ | 0.1283  | -0.4739 | 0.6401  |
| f$_{opal}$ | -0.0138 | -0.276  | -0.6346 |
| f$_{lith}$ | 0.1588  | -0.4298 | -0.3948 |
| f$_{Mn}$  | 0.5604  | 0.2188  | -0.0613 |
| f$_{Fe}$  | 0.5432  | 0.2906  | -0.1107 |
| % of variance | 35.33   | 22.78   | 19.53   |
| Cumulative % | 35.33   | 58.11   | 77.64   |
| Atlantic  | Bulk $K_d$ | 0.331   | 0.4279  | 0.2871 |
| f$_{POM}$ | -0.6219 | -0.3042 | -0.1263 |
| f$_{CaCO_3}$ | -0.0499 | 0.8387  | -0.1478 |
| f$_{opal}$ | -0.225  | 0.0063  | -0.4912 |
| f$_{lith}$ | -0.6164 | 0.1421  | 0.1033  |
| f$_{Mn}$  | -0.2649 | -0.0137 | 0.3205  |
| f$_{Fe}$  | 0.0196  | -0.0208 | 0.7247  |
| % of variance | 30.56   | 17.84   | 15.65   |
| Cumulative % | 30.56   | 48.2    | 63.85   |
| Arctic    | Bulk $K_d$ | -0.0604 | 0.6125  | -0.2405 |
| f$_{POM}$ | -0.5367 | 0.09    | -0.2946 |
| f$_{CaCO_3}$ | 0.1441  | 0.4497  | 0.4808  |
| f$_{opal}$ | -0.2857 | -0.4226 | 0.5921  |
| f$_{lith}$ | 0.5816  | 0.0533  | -0.1112 |
| f$_{Mn}$  | -0.167  | 0.4706  | 0.4886  |
| f$_{Fe}$  | 0.4897  | -0.1077 | 0.1501  |
| % of variance | 39.15   | 25.61   | 13.16   |
| Cumulative % | 39.15   | 64.76   | 77.92   |
around the effects of colloidal chemistry on metal distributions within the particulate phase (e.g., Honeyman and Santschi, 1989; Benoit and Rozan, 1999; Bam et al., 2020). Assuming Hg species sorb to colloids and as colloids are too small to be collected by conventional filtration, GEOTRACES sampling methods tend to count colloids as “dissolved” and thereby result in a smaller $K_d$ value for Hg than the ideal circumstance when colloids are properly accounted for in the particle pool. Furthermore, the proportion of colloids over the particulate mass is generally thought to increase with SPM concentrations, leading to a negative $K_d$ vs. SPM slope (Moran et al., 1996). Indeed, based on two observed relationships between colloidal and SPM concentrations, Moran et al. (1996) modeled the percent of colloidal Hg on the particle mass and found that it tends to increase with SPM concentrations if SPM are $<10^4 \mu g L^{-1}$. A second possibility is that particle mass is not a good proxy for available particle surface sorption sites for Hg. The material reported here as SPM at a specific location is made up of many individual particles that vary in size, with the number of small particles generally being much greater than the number of large particles (e.g., Stemmann and Boss, 2012). Particle size distributions vary between different samples, and therefore each SPM sample has varying proportions of large and small constituents. Samples in high productivity regions with high SPM can also be expected to have a relatively high proportion of large particles (e.g., diatom cells), while low SPM samples are more prevalent in oligotrophic regions where cells are mostly small (e.g., picoplankton). Thus, we might expect that the smaller the SPM is, the more Hg scavenged by a given particle mass as a consequence of larger surface area-to-volume ratios for smaller particles. As area-to-volume ratios scale with the inverse of particle size, one might predict that a bulk $K_d$
FIGURE 5 | The linear regression between log bulk Kd (l kg$^{-1}$) and particle concentration (log SPM in µg L$^{-1}$) in the small size fraction for Pacific (blue line), Atlantic (green line), and Arctic (yellow line) oceans. The slopes of linear regression for each ocean basins were $-0.367$, $-0.423$, $-0.629$, with statistics R-squared equals 0.2155, 0.2218, 0.5338, respectively. The slope of the Arctic was significantly different from the slopes of the Pacific and the Atlantic by applying Z-test, with statistics $P < 5\%$.

vs. SPM plot should resemble an inverse curve. However, no discernible trend between the total SPM concentration (Total SPM = SSF SPM + LSF SPM) and fSSF, the mass fraction of small particles over total particles ($f_{SSF} = SSF\ SPM/Total\ SPM$) during the same cruises, was evident (Figure 6). The fSSF is an analog to the slope of the particle mass-size spectrum (Xiang et al., 2020) or the number-size spectrum (e.g., Cael and White, 2020), a higher $f_{SSF}$ implies more abundance of small particles. Hence, it is likely that the particle size did not change with SPM concentrations in particles. Therefore, the “particle-size hypothesis” appears unsupported by these datasets. A third possibility for the apparent particle concentration effect is a kinetic explanation. The ratio of the adsorption to desorption rate constant, normalized by SPM, can be thought of, in some circumstances, as a kinetic representation of $K_d$ (Honeyman et al., 1988). In a kinetic study of the adsorption and desorption of thorium, both the adsorption and desorption rate constants were found to increase with SPM, but at different rates, such that their ratio normalized by SPM also decreased with SPM, much like the classic particle concentration effect (Lerner et al., 2017). Although this kinetic explanation for the apparent particle concentration effect has so far only been observed for thorium, it is a potential alternative to explain the particle concentration effect that does not rely on colloids. This then leaves the influence of colloids, and perhaps the kinetics of particle attachment and detachment, as the possible explanations for the $K_d$ vs. SPM trends observed in our data.

Particle Composition Effect
Additionally, we argue that variations in bulk $K_d$ also partly result from the particle composition effect. The decreasing trend in bulk $K_d$ with SPM (Figure 5) clearly shows the influence of SPM, but the variations around this trend are still worth being investigated. We assumed this variation is from particle composition. Different statistical methods were employed to obtain the information about the impact of particle composition on partition coefficient.

In some specific regions, the value of bulk $K_d$ was clearly driven by the SPM concentration. For example, high concentrations of SPM in the deep water of the western Atlantic (Lam et al., 2015) corresponded to low bulk $K_d$. Similarly, the Arctic continental shelf also had abundant SPM (Xiang and Lam, 2020), coinciding with low bulk $K_d$ values. At the other end of the SPM spectrum, high $K_d$ values were found in Arctic mid-waters where SPM was low. This was not universally true, however, as demonstrated by much higher bulk $K_d$ in relatively low SPM concentrations.
FIGURE 7 | The pie charts (A,C,E,G) described the mean and standard deviation of particle composition fraction of mass in the Pacific (GP16), Atlantic (GA03), Arctic (GN01), and hydrothermal plume in EPZT; chart (B,D,F,H) described the mean proportion of each composition fraction multiplying their intrinsic Kd in four regions above, respectively.
water in the Canada Basin in the Western Arctic Ocean. We take this to be evidence of another factor that influences the bulk $K_d$, namely the particle composition effect. This effect is described by the intrinsic $K_d$ values in each basin and which can be better understood by multiplying the mean compositional fraction for each particle phase by the intrinsic $K_d$ we calculated (Figure 7). The corresponding value, when compared to those of other phases, expresses the relative importance for each phase in sorbing Hg from solution (if that kind of chemistry does in fact occur). The resulting pie charts indicate that for these three oceans, POM and CaCO$_3$ dominate Hg partitioning. This trend holds across all the basins despite there being large variations in the abundance of CaCO$_3$ as a result of compensatory changes in the intrinsic $K_d$ for that phase (Table 1). Moreover, the contributions of lithogenic material to bulk $K_d$ were also quite similar in the three oceans, with percentages from 9 to 11%, while the fraction of it was distinct everywhere.

It is worth noting that Fe contributions to bulk $K_d$ were quite variable and were high only in the Pacific, caused by relatively high Fe fraction in particles released in a hydrothermal plume. Similarly, the abundant Mn observed in the Arctic also accounted for a large part of the bulk $K_d$. Therefore, we can see that Mn and Fe had strong adsorption affinities to Hg in some regions despite their relatively low concentration level in particles. The strong sorption affinity of Mn is apparently not only for Hg but also to $^{234}$Th, $^{231}$Pa, $^{210}$Po, and $^{210}$Pb (Hayes et al., 2015; Lerner et al., 2018; Bam et al., 2020). As noted above and discussed below, there was no resolvable intrinsic $K_d$ value for opal and so it does not appear in the pie charts of Figure 7. This implies that opal plays a minor role in the partitioning of Hg into the particle phase, at least in these three basins.

In Figure 5, bulk $K_d$ data from the three oceans were scattered around the regression lines with SPM. We examine the hypothesis that the residuals to the SPM relationship, which are the vertical distance between each data point and the regression lines, might be explained by particle composition. Hence, we applied PCA to see the relationship between residuals of log $K_d$ and particle phases. The PCA results are presented in Supplementary Table 2. In the Pacific, the highest loadings for PC1 were the fractions of Fe and Mn, which were positively associated; for PC2, POM, CaCO$_3$ and the residuals had the highest loadings, where POM correlated positively with residuals whereas CaCO$_3$ correlated negatively with residuals. Information from PC1 suggests that POM was also a crucial factor that led to the variance of $K_d$ in the Atlantic; CaCO$_3$ had a positive association with residuals in the Arctic from PC2.

Besides PCA, we applied the least squares regression of particle composition fraction to the $K_d$ residuals of the SPM regression, which we called the “intrinsic residual $K_d$” ($K_d^*$) (Table 3).

$$\text{residuals} = \sum_{i=1}^{6} (K_d^*)_i \cdot f_i = (K_d^*)_1 \cdot f_1 + (K_d^*)_2 \cdot f_2 + (K_d^*)_3 \cdot f_3 + (K_d^*)_4 \cdot f_4 + (K_d^*)_5 \cdot f_5 + (K_d^*)_6 \cdot f_6$$  (4)

In general, the particle composition explained the 39.72% of variance of residuals in the Pacific Ocean, 6.25% of variance of residuals in the Atlantic Ocean and 12.88% of variance of residuals in the Arctic Ocean. In the Pacific, Mn and Fe had a strong correlation with residuals compared with other particle phases. Mn could result in lower $K_d$ while Fe contributed to a higher $K_d$. CaCO$_3$ and lithogenic particles have little effect in driving bulk $K_d$ to be lower than the regression line (Figure 5), while POM has a positive feedback on bulk $K_d$. In the Atlantic, POM, CaCO$_3$, lithogenic phase and Fe lead to positive residuals that makes bulk $K_d$ higher above the fitting line (Figure 5). Additionally, POM, lithogenic phase have strong positive correlation with residuals in the Arctic, while Mn led to smaller $K_d$ values in the Arctic.

By adding R-squared values that are generated from the linear regression model of SPM and $K_d$ (Figure 5) to R-squared values from least squares regression above, we could have a general view of how much variation of partition coefficient that can be demonstrated by both particle concentration and composition effect. Overall, the SPM combined particle composition explained the 61.27% of variance of $K_d$ in the Pacific, 28.43% of variance of $K_d$ in the Atlantic, and 66.26% of variance of $K_d$ in the Arctic. However, these values were possibly overestimated, since SPM and composition effect are not completely independent in our model. Despite that residuals to the SPM regression remove the effect of particle concentration which represents a pure composition effect on $K_d$, the regression of log $K_d$ against SPM carries some particle composition effect information which we don’t know how to eliminate so far.

### Special Features: Hydrothermal Plume and Coast

Part of the reason for the exceptionally high $K_d$ values observed from the EPZT cruise dataset was the presence of high Fe at stations/depths associated with the continental shelf and East Pacific Rise hydrothermal plume. Thus, we separated the EPZT data into three subsets based on the dissolved Fe concentrations reported by Resing et al. (2015), including a coastal margin (stations 1–5), hydrothermal vents (stations 18, 20, 21, 25, 26, where depth was between 2,200 and 3,000 m) and open ocean (the rest of stations and depths). The bulk $K_d$ values for the

**TABLE 3 |** The coefficients to the least squares regression of the residual $K_d$ (L kg$^{-1}$) of Hg for six particle composition fractions (Equation 4) in the small size fraction (SSF) in the Pacific, Atlantic, and Arctic ocean, respectively.

|        | Pacific   | Atlantic  | Arctic   |
|--------|-----------|-----------|----------|
| POM    | 2.21      | 2.12      | 3.11     |
| CaCO$_3$ | −0.14    | 1.41      | 0.45     |
| Opal   | −0.69     | −3.58     | −1.64    |
| Lithogenic | 2.00     | 0.52      | 0.40     |
| Mn     | −10.97    | 4.39      | −0.93    |
| Fe     | 10.18     | 1.29      | 14.20    |

The statistical R$squared$ for each ocean were 0.3972, 0.0625, 0.1288.
The Lack of a Resolved Intrinsic $K_d$ for Opal

Zaferani et al. have recently documented uncommonly high sediment accumulation fluxes of Hg that are also associated with high opal fluxes along parts of the Antarctic margin (e.g., Zaferani et al., 2018; Zaferani and Biester, 2020). This is seemingly inconsistent with our finding that there is little correlation between the values of bulk $K_d$ and $f_{\text{opal}}$ in overall datasets, resulting in undefined values for the intrinsic $K_d$ for Hg-opal, and implying either that biogenic silica is a poor sorbent material for Hg or that diatoms are not particularly effective at taking up Hg. The first of these two possibilities are more likely as the high Hg fluxes on the Antarctic margin could be due more to high productivity, and therefore high abundance of particulate organic matter, rather than the opal associated with that productivity. Furthermore, diatoms appear to effectively bioaccumulate Hg (Mason et al., 1996; Beldowski et al., 2018). The lack of a resolved $K_d$ contribution from opal in the multivariate linear regression is an expression of an apparent lack of co-variation between $K_d$ and $f_{\text{opal}}$ and is consistent with both the simple correlation matrix view of the data as well as the PCA, both of which show little co-variation of Hg $K_d$ and $f_{\text{opal}}$ in the full datasets. One important caveat to this discussion is that two of the Pacific sub-datasets (open ocean and hydrothermal plume; Table 4), did have resolved intrinsic $K_d$'s for opal, and the values were quite high. This was especially surprising as these were locations in the Pacific with the lowest fraction of particle mass as opal. With our current datasets, this behavior is difficult to explain and is worthy of further study.

Possible Biological Control on $K_d$ in Surface Waters

In surface water, we found higher bulk $K_d$ values in the open ocean compared to the continental margin (water depth < 200 m; Figure 3). This could be the result of the concentration effect as margin water tends to be higher in SPM than open ocean water. It could also be due to a difference in the mechanism by which Hg becomes associated with particles. In deeper waters, this should only be a consequence of sorption, while in surface waters could also be from biological uptake. If this is true, then variations in the value of bulk $K_d$ might be determined by an “uptake efficiency.” In coastal regions, even though the biological activity is high, the uptake efficiency of Hg might be limited by the size of microbes based on Michaelis-Menten kinetics: more effective uptake occurs in the open ocean, where small size microbes with high uptake rate survive. In order to test this hypothesis, we examined the regression of log bulk $K_d$ against log SPM in both shallow water (above 100 m) and deep water (below 100 m), separately, for three ocean basins. Then, we compared regression slopes between shallow and deep water by applying Z-test. Assuming that the particle concentration effect controls the $K_d$ in deep water, if microbes were to passively adsorb Hg (like particle scavenging) in the surface, we would expect to see no difference between slopes in the shallow and deep water. In contrast, if microbes actively taking up Hg in the surface is responsible for the higher open ocean surface $K_d$, we might expect to observe a significantly different slope in shallow water compared to deep water, because $K_d$ was not controlled by particle concentration effect.

The statistical results were summarized in Supplementary Table 3. Through the comparison of slopes of regression in shallow and deep seawater, we noticed that the slopes between shallow and deep water were not significantly different in the Atlantic and Arctic Ocean, suggesting the passive adsorption might take over in the surface seawater, therefore, we can deem that particle scavenging dominate the whole Atlantic and Arctic Ocean. However, the slope analysis failed the

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**TABLE 4** The log intrinsic $K_d$ (l kg$^{-1}$) of Hg for six particle composition in the small size fraction (SSF) in coastal margin, hydrothermal plume, and open ocean of the Pacific, respectively, and their relative standard deviation (r.s.d.) as well as particle composition mass contribution (f; %).

| Coastal margin | Hydrothermal plume | Open ocean |
|----------------|--------------------|-----------|
|                | r.s.d. (%)         | Typical contribution to particle mass (%) | r.s.d. (%) | Typical contribution to particle mass (%) | r.s.d. (%) | Typical contribution to particle mass (%) |
| POM            | n.a.               | 62 ± 19 | n.a. | 26 ± 8 | 7.27 | 0.46 | 50 ± 23 |
| CaCO$_3$       | 7.83               | 9.20 | 14 ± 8 | n.a. | 31 ± 15 | 6.56 | 0.68 | 37 ± 18 |
| Opal           | n.a.               | 22 ± 10 | 7.07 | 7.65 | 11 ± 8 | 7.81 | 5.52 | 6 ± 4 |
| Lithogenic     | 7.62               | 7.17 | 12 ± 17 | n.a. | 3 ± 4 | n.a. | n.a. | 5 ± 10 |
| Mn             | n.a.               | 0.03 ± 0.07 | n.a. | 2 ± 2 | 8.86 | 1.24 | 0.2 ± 0.3 |
| Fe             | n.a.               | 0.6 ± 0.4 | 8.25 | 7.75 | 26 ± 18 | n.a. | n.a. | 1 ± 2 |
Z-test in the Pacific, the result of statistical difference between shallow and deep water is consistent with our hypothesis of active biological uptake control in the surface. Despite this observed difference, more evidence needs to be collected in order to support the phenomenon in the Pacific, we cannot assert the conclusion of surface domination of biological uptake on $K_d$, since the special feature in Hydrothermal Plume could also give rise to this difference between shallow and deep water.

**Model Evaluation: Observed vs. Predicted**

To test the robustness of the six end-member model, we substituted calculated values from Table 1 (intrinsic $K_d$) into Equation 2 to obtain the predicted $K_d$ values for each sample (Tang et al., 2017). We then looked at the correlation between observed and predicted $K_d$ through linear regressions and treating the ocean basins separately. The analysis of the coefficient of determination ($R^2$) revealed that 12% of total variance was explained by the predicted vs. observed $K_d$ regression model in Pacific (without hydrothermal plume data), while 14% of Atlantic $K_d$ and 27% of Arctic $K_d$ were explained by the linear regression of predicted against observed $K_d$ (PO regression), respectively ($P < 0.0001$).

Furthermore, all the slopes were far from 1, the shape of linear regression in Figure 8 suggested that this model overestimates observed $K_d$ at low value while underestimates at high value. The y-intercepts from PO regression were different from 0. Therefore, based on the analysis of $R^2$, slope and y-intercept, this model failed to describe the characteristic of variability of observed $K_d$ well. However, when we re-did the linear regression on extracted data from hydrothermal vents in the Pacific only (Figure 8D), the slope was 0.89 with $R^2$ 0.84. This good agreement occurred because iron oxyhydroxides were a significant fraction of particle mass (26%) and thus dominated the adsorption (97%) in the whole hydrothermal plume area (Figures 7G,H). This evaluation is informative, suggesting we need to look for a new way to express the partition coefficient in the future.

**FIGURE 8** | The statistical results of the regression of observed (Obs) vs. predicted (Pre) $K_d$ values in (A) the Pacific Ocean (GP16) without Hydrothermal Plume (HP), (B) the Atlantic Ocean (GA03), (C) the Arctic Ocean (GN01), and (D) Hydrothermal plume in the EPZT. Bule solid line is linear regression fitted line, red dash line is a reference line $y = x$. 
Implication of Hg Residence Time

To some extent, the $K_d$ determines the downward Hg flux from the surface ocean, and further influences the Hg burial flux to marine sediments, which give us insights into the residence time of Hg in the ocean.

Here, we calculated the Hg inventory in each ocean basin by integrating the dissolved Hg with depth (Table 5). Due to a lack of Hg burial flux on a global scale, we used information about the particulate Hg flux into the deep ocean to estimate the burial flux. The particulate Hg flux was derived based on the bulk particle flux from the same cruises (Xiang et al., 2020). To convert to Hg flux, we multiplied the particle flux by the concentration ratios between particulate Hg and SPM for each sample. The underlying assumptions are: (1) particulate Hg follows the particle size distribution of bulk particles; (2) particulate Hg has the same sinking velocity as bulk particles. We then estimated the mean of the logarithmic transformed Hg sinking fluxes below 3,500 m in each ocean basin, and assumed the average sinking flux in the deep ocean equal to burial flux. Overall, our estimates of the Hg burial flux were an order of magnitude higher in the Pacific than the Arctic and Atlantic Ocean (Table 5). In the Arctic Ocean, the Hg burial flux ($20.23, +10.26, −6.81$ pmole m$^{-2}$ d$^{-1}$) was in a similar range compared to Tesán Onrubia et al. (2020) (outer shelf: $51 ± 35$ pmole m$^{-2}$ d$^{-1}$; central Arctic Ocean: $11 ± 8$ pmole m$^{-2}$ d$^{-1}$). Note that the uncertainty of burial flux were not the same because they were transformed back from logarithmic to their original scale. Additionally, the Hg burial fluxes in the Atlantic and Arctic were in line with the globally averaged Hg fluxed from deep ocean to sediments reported by Outridge et al. (2018), which was $0.60$ kt/y.

On the basis of estimated Hg inventory and burial fluxes, Hg residence times in three ocean basins were calculated (Table 5). A strong negative correlation between log bulk $K_d$ and residence time was observed in our analysis, suggesting that $K_d$ could have a significant impact on Hg residence time. Given the higher bulk $K_d$ observed in the Pacific, our estimates of the Hg residence time in the Pacific are the shortest among all oceans, $102$ years, lower than previous studies ($350$ yrs, e.g., Gill and Fitzgerald, 1988). The residence time of Hg is the longest in the Atlantic. It should be noted that, however, our Stokes’ law-based method might overestimate the Hg burial fluxes compared to conventional but limited sediment trap-based observations (e.g., Munson et al., 2015), which could lead to shorter residence time. Future modeling and field observation efforts are needed to better constrain the Hg burial flux and thereby Hg residence time on a global scale.

CONCLUSION

In general, we investigated into the Hg partition coefficient ($K_d$) in the water column across the Pacific, Atlantic and Arctic Ocean, and analyzed the impact of particle concentration and composition on variation of $K_d$. The higher $K_d$ were observed in the Pacific, while modestly lower and lowest $K_d$ were observed in the Arctic and Atlantic Ocean, respectively. The log bulk $K_d$ of Hg was inversely correlated with log SPM concentration, known as “particle concentration effect.” The analysis of intrinsic $K_d$ across three ocean basins indicated that affinity of Hg to particle composition is not constant everywhere, and its variation depends on regions and phase concentration. Fe and Mn oxides have highly strong affinity to Hg in the water column despite their low relative abundance in particles, which could be dominant phases in some special regions like the hydrothermal plume, POM and CaCO$_3$ were important drivers of $K_d$ as well. Further, the end member mixing model has limited ability to describe the observed $K_d$ for the entire ocean possibly as a result of changing water chemistry and mineralogy of particles. Overall, the SPM combined particle composition explained the $28$–$66$% of variance of $K_d$ over three ocean basins. The strong negative correlation between Hg residence time and average log bulk $K_d$ implied that varying $K_d$ might determine the timescale of marine Hg cycling indirectly.

DATA AVAILABILITY STATEMENT

Publicly available datasets were analyzed in this study. This data can be found at: https://www.bco-dmo.org.

AUTHOR CONTRIBUTIONS

XC and CL conceived of the presented idea and took the lead in writing the manuscript. XC performed the computations and analytical calculation. PL and YX collected the SPM datasets. CH and CL collected Hg datasets. All authors provided critical feedback and contributed to the final manuscript.

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### TABLE 5 | Summary of estimated Hg inventory ($10^3$ pmole m$^{-2}$), Hg burial flux (pmole m$^{-2}$ d$^{-1}$) with asymmetric standard deviation, Hg residence time (year), as well as averaged log $K_d$ (log L Kg$^{-1}$) in the Pacific, Atlantic, and Arctic Ocean, respectively.

| Region   | Hg inventory ($10^3$ pmole m$^{-2}$) | log Hg burial flux (log pmole m$^{-2}$ d$^{-1}$) | Hg burial flux (pmole m$^{-2}$ d$^{-1}$) | Residence time (year) | Log $K_d$ (log L Kg$^{-1}$) |
|----------|--------------------------------------|-----------------------------------------------|----------------------------------------|-----------------------|-----------------------------|
| Pacific  | 5.29                                 | $2.15 ± 0.43$                                  | 142.73, $+240.0116, −89.50$            | 102                   | $7.11 ± 0.38$               |
| Atlantic | 5.90                                 | $1.56 ± 0.38$                                  | 36.31, $+14.48, −21.17$                | 445                   | $6.52 ± 0.32$               |
| Arctic   | 2.56                                 | $1.31 ± 0.18$                                  | 20.23, $+10.26, −6.81$                 | 347                   | $6.74 ± 0.53$               |

The correlation between residence time and log $K_d$ was strongly negative correlation coefficient $t = −0.995$, $R^2 = 0.991$. 

The residence time of Hg is the longest in the Arctic Ocean, and further influences the Hg burial flux to marine sediments, which give us insights into the residence time of Hg in the ocean.
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SUPPLEMENTARY MATERIAL

The Supplementary Material for this article can be found online at: https://www.frontiersin.org/articles/10.3389/fenvc.2021.660267/full#supplementary-material
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**Conflict of Interest:** The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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