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Effects of Mercury Deposition and Coniferous Forests on the Mercury Contamination of Fish in the South Central United States

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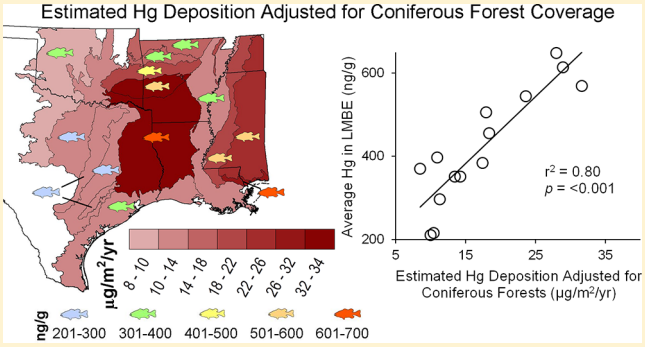
Supporting Information

ABSTRACT: Mercury (Hg) is a toxic metal that is found in aquatic food webs and is hazardous to human and wildlife health. We examined the relationship between Hg deposition, land coverage by coniferous and deciduous forests, and average Hg concentrations in largemouth bass (*Micropterus salmoides*)-equivalent fish (LMBE) in 14 ecoregions located within all or part of six states in the South Central U.S. In 11 ecoregions, the average Hg concentrations in 35.6-cm total length LMBE were above 300 ng/g, the threshold concentration of Hg recommended by the U.S. Environmental Protection Agency for the issuance of fish consumption advisories. Percent land coverage by coniferous forests within ecoregions had a significant linear relationship with average Hg concentrations in LMBE while percent land coverage by deciduous forests did not. Eighty percent of the variance in average Hg concentrations in LMBE between ecoregions could be accounted for by estimated Hg deposition after adjusting for the effects of coniferous forests. Here we show for the first time that fish from ecoregions with high atmospheric Hg pollution and coniferous forest coverage pose a significant hazard to human health. Our study suggests that models that use Hg deposition to predict Hg concentrations in fish could be improved by including the effects of coniferous forests on Hg deposition.



INTRODUCTION

Mercury (Hg) contamination of the environment is hazardous to human and wildlife health.[1,2] Consumption of methyl mercury (MeHg)-contaminated fish is the primary source of MeHg to humans.[1,3] Because fish from most water bodies in the U.S. are not monitored for Hg contamination, it is critical that we develop the ability to predict regions of the environment that may have fish with high concentrations of MeHg.

Most MeHg contamination of fish originates from the deposition of atmospheric inorganic Hg into water bodies and their watersheds.[4] In aquatic ecosystems, bacteria convert inorganic Hg (i.e., Hg^{2+}) to MeHg, which readily bioaccumulates in food chains.[5] Most of the Hg in fish muscle tissue is MeHg,[6] and a positive relationship between Hg wet deposition and the concentration of MeHg in fish has now been established.[7]

Forests have been hypothesized to be an important factor controlling Hg deposition because forests have increased Hg deposition under their canopies compared to open areas.[8,9] Atmospheric Hg adheres to components of the forest canopy (i.e., tree leaves and needles and other material such as bark, branches, fruits, and reproductive structures) and can be incorporated into leaves and needles when Hg enters through the stomata.[9] Mercury is then transported to the ground via throughfall and litterfall.[9] Mercury deposition varies by forest type[9,10] and is higher under coniferous than deciduous tree canopies [9] because of the greater ability of conifers to scavenge Hg from the atmosphere.[9,11] Witt et al. [12] hypothesized that compared to deciduous forests, conifer-dominated systems may be at increased risk for Hg-related water quality issues, but this hypothesis has not been tested.

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Here we examine the relationship between Hg deposition, forest coverage and Hg contamination of fish in 14 ecoregions in the South Central U.S. We found that estimated Hg deposition, adjusted for land coverage by coniferous forests, explained 80% of variance in average Hg concentration of fish between ecoregions. Our study is the first to show that ecoregions with high atmospheric Hg pollution and coniferous forest coverage pose a significant hazard to human health.

METHODS

We focused on Hg contamination of largemouth bass (*Micropterus salmoides*) because it is a widely distributed [13] and economically important species of freshwater game fish [14] that is commonly included in databases of contaminants in fish tissues.[15] Adult largemouth bass are piscivorous top predators, often having high Hg concentrations relative to other fish species.[15,16] We obtained data on largemouth bass from state and federal agencies and Drenner et al.[17] and combined these data with data on largemouth bass and other species from the National Descriptive Model of Mercury in Fish (NDMMF)[18] to produce a total data set of 40,564 fish samples collected over the period 1969−2010 from 893 lentic and lotic sites [Figure S1 of the Supporting Information (SI)] located within 14 U.S. Environmental Protection Agency (USEPA) Level III ecoregions. The ecoregions are located within all or part of six states (Texas, Louisiana, Oklahoma, Arkansas, Mississippi, and Tennessee). Because Hg concentrations in fish vary by species and length, and samples of the same species and length are difficult to obtain from site to site, the NDMMF was utilized to estimate concentration of Hg in 35.6-cm total length (TL) largemouth bass-equivalent fish (LMBE) samples for each site. Adult largemouth bass range in size from 12 to 70 cm TL.[13] For this study, we chose a TL of 35.6 cm for length standardization because (1) creel surveys in Texas from 2005 to 2009 showed this length is a size of largemouth bass commonly caught by anglers (unpublished data, Texas Parks and Wildlife Department, Bobby Farquhar, Personal Communication) and (2) this is a size slightly above 30.5 cm, the smallest minimum length limit commonly used in the U.S. for largemouth bass.[19]

We used ecoregions as the unit of analysis because they are well-suited for spatial studies. Ecoregions denote areas of the environment with similar landscapes,[20] and therefore can serve as a spatial framework for monitoring and management of ecosystems.[21] The 14 USEPA Level III ecoregions [22] examined in this study contained from 10 to 319 fish sampling sites (Figure S1 of the SI). The USEPA ecoregion vector data were converted to a 30 × 30 m raster to overlay and calculate average mercury in fish, Hg deposition, and forest land cover for each ecoregion.

Total Hg wet deposition data were developed from the National Atmospheric Deposition Program's Mercury Deposition Network.[23] The NADP/MDN performs weekly measurements of Hg concentrations and wet deposition in precipitation. All MDN sites are located in open areas away from forest canopies, and represent wet deposition of Hg in precipitation, free from throughfall and litterfall Hg additions. To better relate Hg concentration in multiyear fish tissue data to Hg wet deposition, a long-term average Hg wet deposition was developed. The annualized precipitation-weighted average Hg concentration was calculated over the period 2006−2009 from weekly observations in the NADP/MDN, following the NADP's standard algorithm.[23] Mercury wet deposition was represented as a single average value for each ecoregion. Additional information on calculation of long-term average Hg wet deposition is in the SI.

To determine coverage by deciduous and coniferous forests, we used the National Land Cover Database (NLCD) 2006.[24,25] The NLCD 2006 is a 16-class land cover classification scheme that has been applied consistently across the conterminous U.S. at a spatial resolution of 30 m.[25] Deciduous forests are areas dominated by trees generally greater than 5 m tall, and where deciduous trees account for more than 20% of total vegetation cover. More than 75% of the deciduous tree species shed foliage simultaneously in response to seasonal change. Evergreen areas were used as a proxy for coniferous forest coverage. Evergreen areas are dominated by trees generally greater than 5 m tall, and where evergreen trees account for more than 20% of total vegetation cover. More than 75% of the evergreen tree species maintain their leaves all year and the canopy is never without green foliage.[25]

To further explore the effects of deciduous and coniferous forests on Hg deposition and Hg concentrations in LMBE, we compared the ability of Hg wet deposition collected in open areas to estimated Hg deposition adjusted for coverage by deciduous or coniferous forests to predict average Hg concentrations in LMBE within an ecoregion. Mercury wet deposition in open areas is less than Hg deposition under forest canopies because forest canopies scavenge Hg from the atmosphere and Hg is deposited in throughfall and litterfall.[9] Although only Hg wet deposition in open areas is routinely collected in the United States,[23] there have been several studies in Europe, Canada, Norway, and the United States that have collected Hg deposition under canopies of deciduous and coniferous forests and compared it to Hg deposition collected in open areas (reviewed in ref. 9). We used these studies to compute that the ratio of annual Hg deposition in open areas to Hg deposition under deciduous and coniferous canopies was 1:3.44:5.07 (open:deciduous:coniferous) (Table S1 of the SI). We used this ratio to estimate Hg deposition in ecoregions as a function of deciduous or coniferous forest coverage using the following equations:

Estimated Hg deposition adjusted for deciduous forest coverage:

$$\text{estimated Hg deposition} = [(1 - \text{proportion}_{\text{deciduous}}) \times \text{deposition}_{\text{Hg}}] + [\text{proportion}_{\text{deciduous}} \times 3.44 \times \text{deposition}_{\text{Hg}}]$$

where $\text{proportion}_{\text{deciduous}}$ is proportion of deciduous forest coverage within an ecoregion, and $\text{deposition}_{\text{Hg}}$ is average Hg wet deposition from the open area in the ecoregion.[23]

Estimated Hg deposition adjusted for coniferous forest coverage:

$$\text{estimated Hg deposition} = [(1 - \text{proportion}_{\text{conifer}}) \times \text{deposition}_{\text{Hg}}] + [\text{proportion}_{\text{conifer}} \times 5.07 \times \text{deposition}_{\text{Hg}}]$$

where $\text{proportion}_{\text{conifer}}$ is proportion of coniferous forest coverage within an ecoregion, and $\text{deposition}_{\text{Hg}}$ is average Hg wet deposition from the open area in the ecoregion.[23]

We tested for ecoregion differences in average Hg concentrations of largemouth bass in the 14 ecoregions using univariate ANOVA (SPSS Ver 20.0.0). We inspected the data

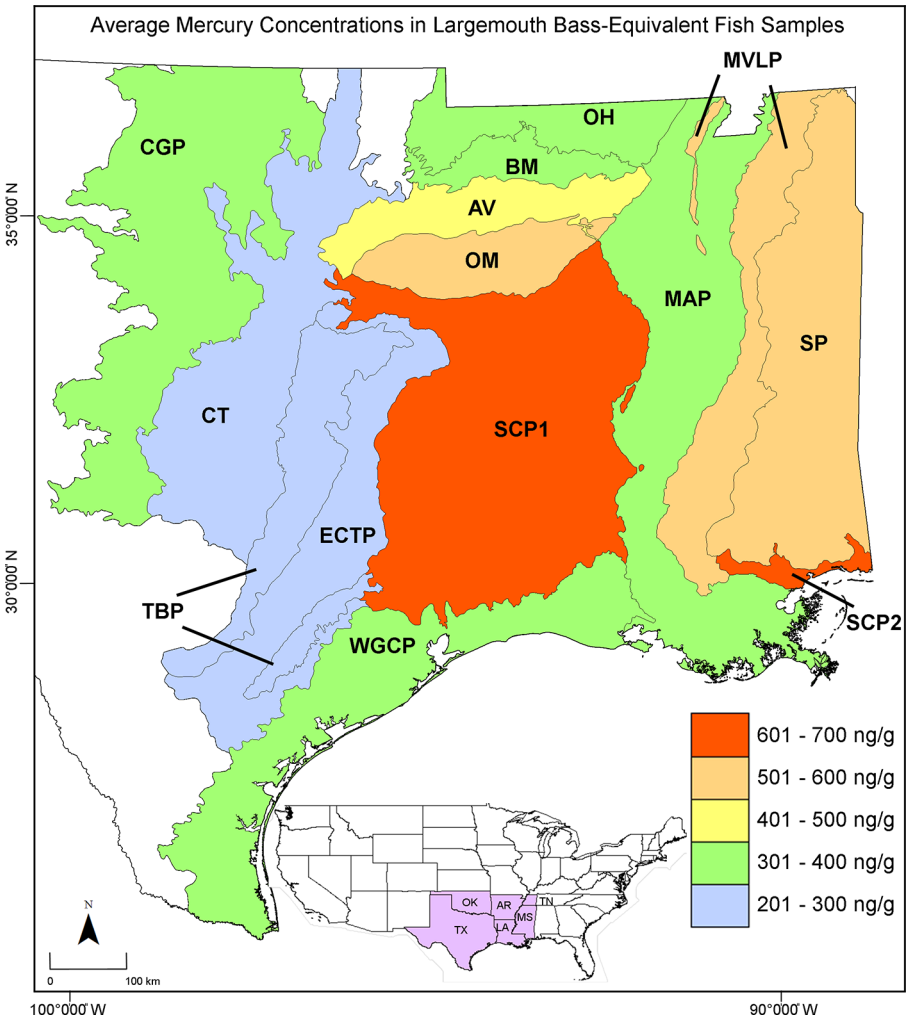

Figure 1. Average Hg concentrations in largemouth bass-equivalent fish from 14 ecoregions in the South Central U.S. Arkansas Valley (AV), Boston Mountains (BM), Central Great Plains (CGP), Cross Timbers (CT), East Central Texas Plains (ECTP), Mississippi Alluvial Plain (MAP), Mississippi Valley Loess Plains (MVLP), Ozark Highlands (OH), Ouachita Mountains (OM), South Central Plains (SCP1), Southeastern Plains (SP), Southern Coastal Plain (SCP2), Texas Blackland Prairies (TBP), and Western Gulf Coastal Plain (WGCP).

set for normality and found that it was positively skewed. We log-transformed the data, which made its distribution more symmetrical and the variance more homogeneous. We detected a significant difference in average Hg concentrations of LMBE between ecoregions using non log-transformed and log-transformed data. Therefore the statistics presented were computed using non log-transformed data. We examined the relationship between percent coverage by deciduous or coniferous forest types and Hg deposition on average Hg concentrations in LMBE using linear regression. Data used in all figures are presented in Table S2 of the SI. Statistical significance was inferred at $p < 0.05$ for all analyses.

RESULTS

We detected statistically significant differences in the average Hg concentrations in LMBE between ecoregions in the South Central U.S. (ANOVA, $p < 0.001$) (Figure 1). Eleven of 14 ecoregions had average Hg concentrations in LMBE above 300 ng/g, the threshold concentration of Hg in fish recommended by the USEPA for the issuance of fish consumption advisories.[26] Five ecoregions in the eastern part of this area (Mississippi Valley Loess Plains [MVLP], Southeastern Plains [SP], Ouachita Mountains [OM], South Central Plains [SCP1] and Southern Coastal Plain [SCP2]) had average Hg concentrations in LMBE above 500 ng/g. Thus the South Central U.S. has levels of Hg contamination in largemouth bass as high as those observed in the Northeastern U.S. [15] and Great Lakes region.[27]

The relationship between forest coverage and average Hg concentration in LMBE differed between forest types. We did not detect a significant linear relationship between Hg concentrations in LMBE and coverage by deciduous forests (Figure 2A). Coverage by coniferous forests had a significant linear relationship with average Hg concentrations in LMBE in the 14 ecoregions (Figure 2B), explaining 73% of the variance.

We compared the ability of Hg wet deposition in open areas to predict Hg concentrations in LMBE to the ability of estimated Hg deposition adjusted for coverage by deciduous or coniferous forests to predict Hg concentrations in LMBE. Mercury wet deposition in open areas had a significant linear relationship with average Hg concentrations in LMBE, explaining 57% of the variance (Figure 3A). We did not detect a significant linear relationship between Hg concentrations in LMBE and estimated Hg deposition adjusted for deciduous forests (Figure 3B). Estimated Hg deposition adjusted for coniferous forest coverage had a significant linear relationship

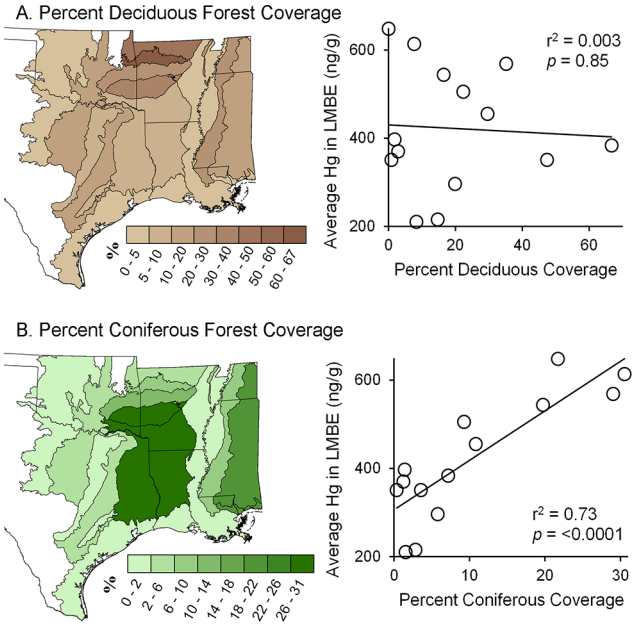

Figure 2. Percent land coverage of deciduous (A) and coniferous (B) forests and their relationships with Hg concentrations in largemouth bass-equivalent fish (LMBE).

with Hg concentrations in LMBE, explaining 80% of the variance (Figure 3C).

DISCUSSION

Atmospheric Hg deposition is the primary source of Hg to most aquatic systems, [4] but the degree of Hg contamination of the food web is dependent on the Hg sensitivity of the landscape.[8,28,29] Hg sensitive landscapes are those in which relatively low levels of atmospheric Hg deposition can cause significant contamination of fish in upper trophic levels.[8,28,29] Hg sensitivity of landscapes is determined in part by land cover type including forests, wetlands, and agriculture.[8,28,29] Other physicochemical factors such as pH, ecosystem productivity, dissolved organic carbon, sulfate, and water-level fluctuations also affect Hg contamination of food chains.[8,28,29]

Here, we examined the effects of deciduous and coniferous forest coverage on Hg concentrations in fish in the South Central U.S. On the basis of studies of Hg deposition under deciduous and coniferous forests,[9] we expected that land coverage by both types of forests would be correlated with average Hg concentration in fish, with coniferous forests having a stronger effect than deciduous forests. We found that coniferous forest coverage had a significant linear relationship with Hg in LMBE but deciduous forest coverage did not have a significant linear relationship with Hg in LMBE.

The spatial patterns of mercury contamination of fish and forest coverage suggest strong effects of conifers on mercury contamination of fish. We found that Hg wet deposition collected in open areas was high (>11 $\mu g/m^2/yr$) and relatively similar in nine ecoregions in the eastern part of our study area (Figure 3A), but Hg concentrations in LMBE varied by almost 2-fold (from 300 to 600 ng/g) across these nine ecoregions (Figure 1). This spatial variation in Hg contamination of LMBE was apparently regulated by coniferous forest coverage. For example, the Mississippi Alluvial Plain (MAP), the ecoregion that contains the Mississippi River, had low (<2%) coniferous forest coverage (Figure 2B) and relatively low (301−400 ng/g)

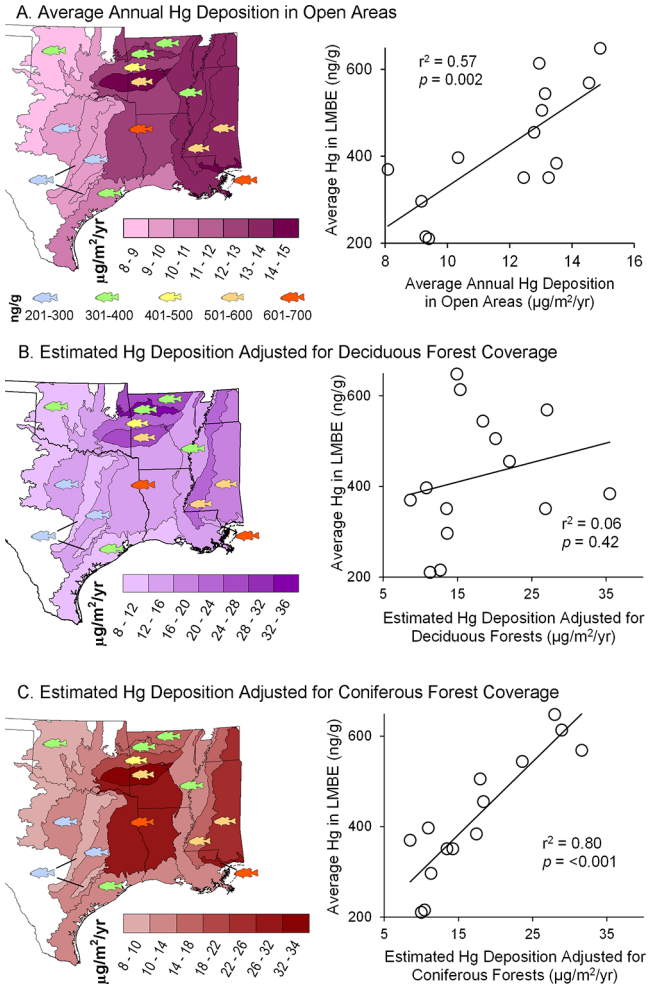

Figure 3. (A) Average annual Hg deposition in open areas and its relationship to average Hg concentrations in largemouth bass-equivalent fish (LMBE). (B) Estimated annual Hg deposition adjusted for deciduous forest coverage and its relationship to average Hg concentrations in LMBE. (C) Estimated annual Hg deposition adjusted for coniferous forest coverage and its relationship to average Hg concentrations in LMBE. Fish symbols represent average Hg concentrations in LMBE from 14 ecoregions in the South Central U.S. and the colors correspond to Hg concentrations in Figure 1.

concentrations of Hg in LMBE (Figure 1). Ecoregions to the west and east of the Mississippi Alluvial Plain (MAP) such as the South Central Plains (SCP1), Southeastern Plains (SP) and Southern Coastal Plain (SCP2) had higher (18−31%) coniferous forest coverage (Figure 2B), relatively high (22−32 $\mu g/m^2/yr$) levels of estimated Hg deposition after adjustment for coniferous forest coverage (Figure 3C), and high (501−700 ng/g) average Hg concentrations in LMBE (Figure 1). Our study is the first to show that LMBE from ecoregions with high atmospheric Hg pollution and coniferous forest coverage pose a significant hazard to human health. Largemouth bass-equivalent fish in these ecoregions had average Hg concentrations two times greater than 300 ng/g, the threshold concentration of Hg in fish recommended by the USEPA for the issuance of fish consumption advisories.[26]

The results of our study are consistent with coniferous forests having elevated Hg deposition rates compared to deciduous forests.[9] Coniferous forests are better scavengers of Hg from the atmosphere than deciduous forests due to higher

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leaf area index (LAI), surface roughness, and density of leaf hairs.[30] Conifers have needles year round, whereas deciduous trees lose their leaves during the cold season. However, there may be other factors associated with coniferous forests that also contribute to elevated Hg in LMBE of the South Central U.S. For example, forested regions have a prevalence of wetlands and unproductive surface waters that promote high concentrations of Hg in freshwater biota.[8] The underlying causal mechanisms of conifer enhancement of Hg concentrations in LMBE need to be explored, perhaps with watershed-level experiments (e.g., ref 31).

The effects of conifers on Hg contamination of fish may extend into other regions of the U.S. The coniferous forest coverage in the ecoregions of the South Central U.S. is dominated by loblolly pine (*Pinus taeda*).[32] Extensive planting and natural regeneration of cutover forest land and abandoned farmland made loblolly pine the leading timber species in the commercial forest land in the Southern U.S., extending from East Texas to the Atlantic Coast.[32] Our study indicates that the enhancement effect of coniferous forests on Hg deposition will be dependent on atmospheric Hg pollution levels, and should be greatest in areas with high Hg wet deposition.

Our study suggests that the amplifying effects of coniferous forest canopies on Hg deposition [9] may result in increased Hg contamination of fish, as hypothesized by Witt et al.[12] Therefore, models that use Hg deposition to predict Hg concentrations in fish [33] could be improved by including the effects of coniferous forest coverage on Hg deposition. The National Atmospheric Deposition Program has initiated a litterfall Hg monitoring program to complement the MDN.[34] This new program operated in cooperation with the U.S. Geological Survey, will provide important information about the role of forests in the Hg cycle and Hg contamination of food webs.

■ ASSOCIATED CONTENT

Supporting Information

Two tables, one figure, and a detailed explanation of the calculation of long-term average Hg wet deposition. This material is available free of charge via the Internet at http://pubs.acs.org.

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Notes

The authors declare no competing financial interest.

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