



## Wildfires threaten mercury stocks in northern soils

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[1] With climate change rapidly affecting northern forests and wetlands, mercury reserves once protected in cold, wet soils are being exposed to burning, likely triggering large releases of mercury to the atmosphere. We quantify organic soil mercury stocks and burn areas across western, boreal Canada for use in fire emission models that explore controls of burn area, consumption severity, and fuel loading on atmospheric mercury emissions. Though renowned as hotspots for the accumulation of mercury and its transformation to the toxic methylmercury, boreal wetlands might soon transition to hotspots for atmospheric mercury emissions. Estimates of circumboreal mercury emissions from this study are 15-fold greater than estimates that do not account for mercury stored in peat soils. Ongoing and projected increases in boreal wildfire activity due to climate change will increase atmospheric mercury emissions, contributing to the anthropogenic alteration of the global mercury cycle and exacerbating mercury toxicities for northern food chains. **Citation:** Turetsky, M. R., J. W. Harden, H. R. Friedli, M. D. Flannigan, N. Payne, J. Crock, and L. F. Radke (2006), Wildfires threaten mercury stocks in northern soils, *Geophys. Res. Lett.*, *33*, L16403, doi:10.1029/2005GL025595.

### 1. Introduction

[2] Since the Industrial Revolution, coal burning and other industrial activities have been a prominent source of atmospheric mercury (Hg), leading to enhanced Hg deposition and accumulation in northern ecosystems [Fitzgerald *et al.*, 1998; Schuster *et al.*, 2002; Lockhart *et al.*, 1995]. Mercury is a global pollutant that poses public health risks when elevated above natural background levels [Boening, 2000], particularly elevated levels of methyl Hg, a central nervous system toxin that can cause significant damage to reproduction and developing fetuses [Mahaffey, 1999]. Long range atmospheric transport and the concentration of anthropogenic Hg sources in the northern hemisphere have exposed northern ecosystems to large quantities of Hg [Barrie *et al.*, 1992]. Through the accumulation and biomagnification of methyl Hg, Hg introduced to northern ecosystems poses significant health threats to humans, marine mammals, and other animals high on food chains.

Northern ecosystems also are expected to receive the most dramatic changes in climate [Hinzman *et al.*, 2005], though there are great uncertainties about how climate change will influence the emission and deposition of Hg at high latitudes.

[3] The boreal forest biome dominates terrestrial interactions with the earth's climate north of 50° N [Chapin *et al.*, 2000] because of its large extent and the large carbon (C) stocks residing in boreal vegetation and soils. Due to an imbalance between net primary production and C losses (decomposition, dissolved export, fire combustion), boreal soils have served as an important reservoir for terrestrial C since the end of the last ice age (12,000 to 18,000 yr ago) [Harden *et al.*, 1992]. Because reduced sulfur groups in organic matter are effective in binding Hg, boreal ecosystems also can store significant amounts of Hg, particularly in ecosystems with thick organic soil layers such as peatlands [Grigal, 2003]. While boreal peatlands retain the majority of atmospherically deposited C and Hg, catchments with high proportion of wetland cover export more methyl Hg and dissolved organic C to aquatic ecosystems [St. Louis *et al.*, 1996].

[4] Wildfires recently have been recognized as an important transfer of Hg from the terrestrial biosphere to the atmosphere [Friedli *et al.*, 2003; Sigler *et al.*, 2003], and may contribute to the large uncertainties associated with Hg emission estimates from natural sources. Annual burn areas vary globally from 3 to 22 million hectares yr<sup>-1</sup> across the boreal forest region [Conard and Ivanova, 1997; Stocks *et al.*, 2002; Sukhinin *et al.*, 2004], with increasing burn areas over the past several decades in North America [Gillett *et al.*, 2004; Kasischke and Turetsky, 2006]. Drier climatic regimes and more severe fire weather under future climate change are expected to double burn areas and increase fire severity across much of the boreal region [Flannigan *et al.*, 2005]. While industrial activities directly contribute Hg to the atmosphere, here we argue that anthropogenic forcing of our climate system and resulting changes in wildfire cycles also will contribute significantly to Hg emissions from boreal regions.

### 2. Methods

#### 2.1. Mercury Stocks in Boreal Ecosystems

[5] We quantified organic matter and Hg storage in vegetation and soils in peatland and forested upland ecosystems in western Canada. Our sites varied in canopy type, soil drainage class and stand age (Table S1 included as auxiliary material<sup>1</sup>). At each site, soils were sampled frozen and cut into volumetric samples at depth intervals of 4 to

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**Table 1.** Mercury Concentrations and Stocks Quantified in Vegetation and Soils of Boreal Peatlands and Forested Uplands<sup>a</sup>

	Hg Concentrations, ppm	Hg Stocks, mg/m <sup>2</sup>
<i>Upland Forest Stands</i>		
Canopy overstory <sup>b</sup>	0.08 ± 0.01	0.10 ± 0.00
Canopy understory <sup>c</sup>	0.29 ± 0.06	0.46 ± 0.25
Organic soil to mineral boundary	0.24 ± 0.04	3.41 ± 1.19
<i>Peatlands</i>		
Understory foliage <sup>c</sup>	0.81 ± 0.25	0.63 ± 0.14
Shallow soil <sup>d</sup>	1.18 ± 0.11	11.55 ± 1.10
Shallow + saturated soil <sup>e</sup>	1.02 ± 0.09	41.98 ± 5.48

<sup>a</sup>Sites described in Table S1, which is included as auxiliary material. Data are means±one standard error.

<sup>b</sup>Leaves, bark, bole wood.

<sup>c</sup>Moss and lichen, leaf or needle litter.

<sup>d</sup>Soil in the surface 25 cm.

<sup>e</sup>Includes deeper soil layers to a depth of 70 cm.

20 cm; splits were air dried and analyzed for Hg concentrations using acid digestion and cold-vapor-atomic fluorescence spectroscopy [Environmental Protection Agency, 2001]. Separate oven-dried subsamples were analyzed for carbon concentrations using an elemental analyzer and converted to organic matter concentrations by multiplying by 1.72 [Siltanen *et al.*, 1997]. Mercury and organic matter stocks were calculated by multiplying concentration data by oven-dried bulk densities and cumulative soil thickness to obtain stocks per unit area (g m<sup>-2</sup>). As expected, Hg storage in soils was much greater than Hg stored in vegetation, particularly in peatlands (Table 1 and Figure S1) [Grigal, 2003].

[6] Hg stocks were significantly related to organic matter stocks in both peatland ( $R^2 = 0.857$ ,  $p < 0.0001$ ,  $F = 389.34$ , slope =  $4.8 \times 10^{-7} \pm 2 \times 10^{-8}$ ; intercept =  $0.0028 \pm 0.00147$ ) and upland forest ( $R^2 = 0.884$ ,  $p < 0.0001$ ,  $F = 91.41$ , slope =  $6.06 \times 10^{-7} \pm 6 \times 10^{-8}$ ; intercept = not significantly different from zero) soils. We used these relationships with information from two soil databases to estimate Hg stocks in soils at a regional scale. First, we calculated Hg stocks in upland forest soils using bulk density, thickness, and organic matter concentrations of organic horizons reported in the Soil Profile and Organic C Database for Canadian Forest and Tundra Mineral Soils [Siltanen *et al.*, 1997] ( $n = 390$  sites in the boreal west ecoprovince across AB, MB, SK, and the NWT). Second, we calculated soil Hg stocks in peatlands using bulk density, thickness, and organic matter concentrations of peat layers reported in the Wetland Database for the Western Boreal, Subarctic, and Arctic regions of Canada [Zoltai *et al.*, 2000] ( $n = 336$  sites across AB, MB, SK, and the NWT).

## 2.2. Fire Emission Model

[7] We used emission models to explore the coupling of burn area, fuel availability, and consumption severity as potential controls on Hg emissions from boreal ecosystems. In addition to temporal variability in burn area and landscape variability in upland and peatland burning, our emission models examine controls on ground layer consumption by representing varying levels of combustion efficiency and different water table positions in peatlands that determine fuel loading. These model components are described in more detail below.

### 2.2.1. Historical Burn Areas Across Boreal Ecosystems

[8] The Large Fire Database represents a compilation of fire perimeter maps and other fire attributes for all burning events in Canada greater than 200 ha [Stocks *et al.*, 2002]. We overlaid perimeter maps for fire events from 1980–1999 (Figure S2 included as auxiliary material) in the boreal and subarctic ecoregions with distributions of upland ecosystems from SPOT-VGT data of the Canadian Land Cover Classification of 1998 for the Northwest Territories (NWT), Alberta (AB), Saskatchewan (SK), and Manitoba (MB) (study region covers 2.15 million km<sup>2</sup>). We summarized land cover data to the following classes: deciduous upland, evergreen upland, grassland, grassland shrubby upland, mixed upland, recent burns, snow and ice, tundra, water bodies, and wetland/peatlands, and used the deciduous upland, evergreen upland, and mixed upland classes to represent upland forests. Because the Canadian Land Classification Cover Map does not distinguish peatlands from other open ecosystems and appears to underestimate wetland cover, we performed similar overlays with fire perimeter maps and peatland distributions from the Peatlands of Canada database [Tarnocai *et al.*, 2000]. Polygons within the Peatlands of Canada database are not spatially explicit, but provide information on the average percentage of bog, fen, and permafrost peatland across Canada within 1° × 1° polygons. We estimated peatland area by multiplying percent peatland cover by polygon area, and assumed that fire activity within the burned portion of each polygon did not preferentially burn upland systems relative to peatlands [from Turetsky *et al.*, 2002]. Burned area within individual fire events were compiled for each land cover type and then summed within each year to calculate cover specific annual burn areas (Table S2).

### 2.2.2. Surface Fuel Loads

[9] Ground-layer consumption represents a major component of total fuel consumption during boreal fires, and typically leads to C releases ranging from 5 and > 60 t C ha<sup>-1</sup> [Stocks and Kauffman, 1997; Kasischke *et al.*, 2000; Amiro *et al.*, 2001]. Here, we defined potential ground-layer fuels as organic matter situated above the mineral soil boundary in uplands and above the water/permafrost table in lowlands. Our soil core analyses (Table 1) and estimations with regional soil databases (described in section 2.1) showed no difference in Hg stocks

**Table 2.** Organic Matter and Mercury Stocks in Ground Layer Fuels in Boreal Upland Forests and Peatlands Estimated From Organic Matter-Hg Stoichiometry and Regional Soils Databases<sup>a</sup>

	Organic Soil Depth, cm	Organic Matter Stocks, kg/m <sup>2</sup>	Hg Stocks, mg/m <sup>2</sup>
<i>Upland Forest Stands (n = 390 Sites)</i>			
Deciduous <sup>b</sup>	5.8 ± 0.3	4.28 ± 0.24	2.60 ± 0.20
Evergreen <sup>b</sup>	6.2 ± 0.4	4.36 ± 0.26	2.64 ± 0.22
All sites	6.0 ± 0.2	4.32 ± 0.18	2.62 ± 0.15
<i>Peatlands (n = 336 Sites)</i>			
Wet scenario	Surface 15 cm	26.74 ± 0.87	15.73 ± 0.56
Surface organic matter <sup>c</sup>	Surface 25 cm <sup>c</sup>	35.66 ± 1.17	20.04 ± 0.72
Drought scenario	Surface 35 cm	46.21 ± 1.34	25.14 ± 0.81

<sup>a</sup>Data are means ± one standard error; error terms for Hg stocks were compounded using standard deviations of organic matter stocks and slope and intercept terms for organic matter-Hg relationships outlined in section 2.1.

<sup>b</sup>Based on dominant species at each site defined by *Siltanen et al.* [1997].

<sup>c</sup>Based on average water table position from *Zoltai et al.* [2000].

in forest ground-layer fuels dominated by deciduous versus evergreen species (Table 2). Thus, we used an average fuel loading of  $2.62 \pm 0.15$  mg Hg m<sup>-2</sup> for upland forest soils.

[10] To estimate Hg storage in ground layer fuels within peatland environments, we quantified Hg stocks in soils situated above an average water table position of 25 cm (Table 2). This water table scenario is based on mean water table positions reported by *Zoltai et al.* [2000] (AB:  $19 \pm 1$  cm,  $n = 59$  sites; MB:  $24 \pm 1$  cm,  $n = 64$  sites; SK:  $25 \pm 2$  cm,  $n = 74$ ; no measurements for the NWT). However, Hg stored in deeper saturated soil layers may become vulnerable to burning during drought periods. To estimate Hg stocks in peatlands during dry and wet climatic periods, we calculated Hg stocks under scenarios of high (15 cm) and low (35 cm) water tables to represent wet and drought conditions, respectively (Table 2). Given that many peatlands experience much larger water level fluctuations both within and among years [cf. *Branfireun et al.*, 1996; *Warren et al.*, 2001], our fuel load scenarios are conservative for drought conditions. We assumed that Hg stocks in upland fuels do not change during wet or dry climatic periods (but see estimates of varying fuel consumption/fire severity below).

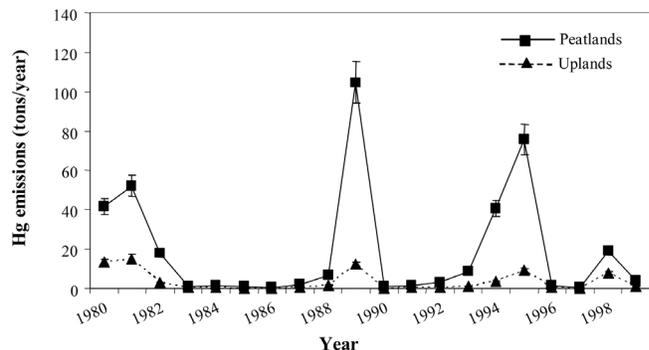
### 2.2.3. Severity of Surface Fuel Consumption

[11] Calculations of fuel consumption rates typically use fractional burning of potential fuels. Here, fractions of fuels consumed during burning are based on combustion rates measured during experimental and wildland boreal fires (*Harden et al.* [2004] and data from J. Harden and M. R. Turetsky [unpublished data, 2005]), which show that approximately 20% of fuels are consumed during average burning conditions across a variety of soil drainage conditions. These empirical results agree well with other conservative estimates of fuel combustion in boreal regions [*French et al.*, 2004]. However, the fraction of fuels consumed varies between wet and dry years as both fuel availability and combustion efficiency increases with declining fuel moisture. Our models use a fractional loss of 30% based on severe combustion estimates established for the boreal region [*French et al.*, 2004] during large fire years and a fractional fuel loss of 10% during small fire years. Estimates of organic matter consumption in our study region averaged 2.9 kg organic matter combusted m<sup>-2</sup> fire<sup>-1</sup> (ranging from 1.9–5.4 and 0.4–2.4 kg organic matter combusted in uplands and peatlands, respectively), which agrees well with published estimates of boreal combustion in uplands

and lowlands (*Harden et al.* [2004], *Benscoter and Wieder* [2003], *Amiro et al.* [2001], and studies reviewed by *Turetsky and Wieder* [2001]).

### 2.2.4. Interactions Among Burn Area, Fuel Loading, and Consumption Severity

[12] While it is difficult to explore variability in fire weather using annual burn areas, components of the Canadian Fire Weather Index System [*Van Wagner*, 1987] were relatively strong predictors of burn area within each province (Table 3). Thus, we followed the approach of *Kasischke and Turetsky* [2006] and classified years into small (less than 1% of land area burned), average (1–2% of land area burned), and large (>2% of land area burned) fire years as the basis for representing interactions among burn area, fuel loading and consumption severity in our model scenarios. From 1980–1999, 3 large fire years occurred in SK (1980,1981,1995), 1 in MB (1989), 1 in the NWT (1995), and 2 in AB (1981,1982). We applied the low water table scenario (35 cm position) and high fractional fuel consumption (30% loss) to large fire years that tend to occur in drought conditions, and the high water table scenario (15 cm position) and low fractional fuel consumption (10% loss) to small fire years (Table 3 and Figure 1). Regional Hg



**Figure 1.** Results of a fire emission scenario coupling annual burn area, fuel loading, and the severity of fuel consumption to explore variability in fire-related Hg emissions from boreal soils. Data are means ± one standard error; errors in regional fluxes were estimated by compounding standard deviation in Hg fuel stocks (Table 2) and a cumulative mapping error (fire perimeter and land cover) of 10%.

**Table 3.** Results of Stepwise Multiple Regression Models Exploring Relationships Between Annual Burn Areas and Components of the Canadian Fire Weather Index System Derived as Annual Estimates From Weather Station Data Across Each Province<sup>a</sup>

	Fire Weather Parameter	Model R <sup>2</sup>	F value	p value	Slope
<i>Peatland Annual Burn Area</i>					
Alberta	DMC <sup>b</sup>	0.19	4.18	0.056	0.23 ± 0.12
MB	DMC	0.28	7.09	0.016	0.20 ± 0.07
NWT	SSR <sup>c</sup>	0.52	19.70	0.0003	3.88 ± 1.08
SK	DMC	0.16	3.49	0.078	0.43 ± 0.12
	FWI <sup>d</sup>	0.43	8.01	0.012	0.87 ± 0.31
<i>Upland Forest Annual Burn Area</i>					
AB	DMC	0.19	4.34	0.052	0.20 ± 0.10
MB	DMC	0.29	7.28	0.015	0.19 ± 0.07
NWT	SSR	0.60	27.43	0.0001	2.47 ± 0.47
SK	DMC	0.19	4.28	0.053	0.45 ± 0.15
	FWI	0.39	5.41	0.033	0.85 ± 0.37
<i>Total (Peatland + Forest) Annual Burn Area</i>					
AB	DMC	0.20	4.45	0.049	0.21 ± 0.10
MB	DMC	0.29	7.22	0.012	0.19 ± 0.07
NWT	SSR	0.57	23.66	0.0001	2.44 ± 0.50
SK	DMC	0.18	3.91	0.064	0.44 ± 0.14
	FWI	0.22	6.37	0.022	0.86 ± 0.34

<sup>a</sup>From *Van Wagner* [1987]. Burn areas were log transformed.

<sup>b</sup>The Duff Moisture Code (DMC) is an estimate of fuel moisture in loosely compacted soil organic material.

<sup>c</sup>The seasonal severity rating (SSR) is a seasonal estimate of the severity of the fire season.

<sup>d</sup>The Fire Weather Index (FWI) is an estimate of the potential intensity of a spreading fire.

emissions appear to be most sensitive to variability in burn areas and the severity of fuel consumption (Figure S3).

### 3. Results and Discussion

[13] Soil chronologies collected from diverse wetland and upland ecosystems (Table S1) show that Hg stocks are more than 10-fold greater in boreal peatland soils than in forested soils (Table 1 and Figure S1). Published values for Hg sequestration in peatlands range from 20–450  $\mu\text{g Hg m}^{-2} \text{yr}^{-1}$  [Fitzgerald *et al.*, 1998; Grigal, 2003]. Assuming an average recent sequestration rate of 200  $\mu\text{g Hg m}^{-2} \text{yr}^{-1}$  across the 385 million ha of boreal peatland globally, peatlands may have sequestered up to 770 tons Hg/yr, equivalent to between ~20–40 % of the Hg emitted globally each year from anthropogenic sources (estimates range from ~2000–4150 t/yr [United Nations Environment Programme (UNEP), 2002]). Certainly up to the Industrial Era, northern peatlands have provided an important ecosystem service by binding Hg in accumulating layers of peat. However, it is not clear whether peatlands will continue to sequester Hg as fire behavior is subjected to regional changes in fire weather and drought.

[14] The emissions of Hg during wildfire in northern landscapes have received little attention, though Hg is readily volatilized from consumed organic matter during burning. Fire-emitted Hg is dominated by Hg<sup>0</sup>, although approximately 15% of Hg emitted during fire is in particulate form [Friedli *et al.*, 2003]. Elemental Hg is incorporated into the atmospheric pool with a lifetime of about one year before redeposition, while particulate Hg has a lifetime of days to weeks in the atmosphere prior to local or regional deposition [Schroeder and Munthe, 1998]. Approximately 30% of Hg stored over the past millenia at our peatland sites was stored above the water table (i.e., Figure S1) and therefore is accessible to wildfire and subsequent re-distrib-

ution. Our emission models suggest that Hg emissions to the atmosphere from fires in this region have varied greatly in time and space over the past 20 years (Figure 1). Drought not only exacerbates wildfire severity and the amount of various fuels burned per unit area, it also dramatically increases fire spread and burn area [Kasischke *et al.*, 2000]. Emission scenarios coupling drought, fire severity, and burn area show that boreal wildfire emissions ranged from 0.4 to 116.8 metric tons of Hg between small and large fire years (Figure 1). Over a 20 year period, fire-induced Hg emissions across this region averaged  $22.81 \pm 7.49$  t/yr (median 4.46 t Hg/yr), though emissions during drought years in this region approached industrial emissions of Hg in North America (estimated ~210 t Hg/y [UNEP, 2002]).

[15] Our modeling approach presents a simple yet realistic interaction between drought and fire severity that exacerbates Hg release to the atmosphere, particularly in poorly drained ecosystems where climate-driven fluctuations in water tables controls the abundance of easily combustible fuel. Though Hg has been protected in northern wetlands throughout the Holocene period, we suggest that drought conditions will increasingly expose Hg in these ecosystems to burning. Despite larger burn areas in upland stands (Table S2), peatlands dominate Hg emissions to the atmosphere during burning due to their large Hg stocks in surface fuels (Table 2 and Figure S1). Our results demonstrate that drought conditions that lower regional water tables and/or increase fire severity can exacerbate Hg emissions, largely from the burning of peat.

[16] Our estimates of boreal fire emissions are greater than previous estimates of wildfire related Hg emissions that did not consider the large Hg reserves stored in peatlands and permafrost systems [Friedli *et al.*, 2003; Sigler *et al.*, 2003]. Fire-related Hg emissions from spruce and jack pine forests averaged 0.15 mg Hg/m<sup>2</sup> [Sigler *et al.*, 2003], which is lower than our range of upland fire emissions (0.26–

0.86 mg Hg/m<sup>2</sup>). As expected, our estimates of fire emissions from boreal peatlands exceeded upland emissions per unit area, ranging from 1.55–7.01 mg Hg/m<sup>2</sup> owing to their greater stocks of Hg.

[17] While Hg emissions are likely to vary regionally, it is clear that studies that do not account for burning in Hg-rich moderately and poorly drained ecosystems are likely to underestimate the contribution of boreal wildfires to global Hg emissions. For example, Sigler *et al.* [2003] used an average Hg consumption rate of 0.15 mg Hg/m<sup>2</sup> and a global burn area of 15 million ha yr<sup>-1</sup> to estimate that boreal fires world-wide could release an average of 22.5 tons Hg yr<sup>-1</sup>. Estimates using the same circumboreal annual burn area and average Hg emissions from forested uplands across our study region (3.6 ± 1.1 t Hg/yr; 604,000 ha burned/yr) suggests that 53.3 t Hg/yr could be emitted to the atmosphere from boreal wildfires globally. However, average Hg emissions that include the burning of forested upland and peatland soils (22.8 ± 7.5 t Hg/yr; 1.01 million ha burned/yr) suggests that 340.8 t Hg/yr could be emitted across the circumboreal region. While simple extrapolations of fire-related Hg emissions in North America to other boreal regions is problematic due to differences in peatland area, fuel types, and fire weather, the large discrepancies between these global estimates point to the need for additional research on the role of deep organic soils in terrestrial Hg storage and the vulnerability of these Hg stocks to burning. Climate projections predict larger, more frequent and more severe wildfires [Hinzman *et al.*, 2005; Flannigan *et al.*, 2005], which will result in correspondingly larger Hg releases to the atmosphere and redistribution of Hg over high latitudes.

[18] It has long been recognized that human activity has disrupted the global Hg cycle through fossil fuel combustion, but climate change impacts on northern latitudes also may amplify Hg emissions and deposition. Though historically regarded as hotspots for Hg accumulation [cf. Martinez-Cortizas *et al.*, 1999], boreal peatlands may become hotspots for Hg emissions with increased burning. Just as anthropogenic activities redistribute naturally occurring Hg, increasing fire cycles at high latitudes may mobilize relatively harmless Hg stored in saturated soil into potentially more mobile and toxic forms. Climate change and higher water temperatures also are expected to increase methylation rates, thereby increasing methyl Hg concentrations in aquatic systems [Booth and Zeller, 2005]. Post-fire deposition of Hg into aquatic systems combined with warmer surface waters and increased methylation rates [Booth and Zeller, 2005] may exacerbate the already pervasive Hg toxicities found in northern regions [Mahaffey, 1999; Barrie *et al.*, 1992].

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