# CHRONOLOGY OF THE ATMOSPHERIC MERCURY IN LAGOA DA PATA BASIN, UPPER RIO NEGRO REGION OF BRAZILIAN AMAZON

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**ABSTRACT.** We present prehistoric mercury accumulation rates in a dated sediment core from Lagoa da Pata, a remote lake in São Gabriel da Cachoeira, northern Amazon. The sediment samples were subdivided for mercury and radiocarbon analyses. A group of 18 samples have been prepared at ANU for ^{14}C dating by accelerator mass spectrometry (AMS). The dating results show a good correlation with depth in the core, down to 41,500 BP. Three distinct sections are clearly identified in the core. They consist of upper and lower organic-rich layers, separated by an inorganic layer which represents a short period of rapid accumulation around 18 ka BP. The mercury accumulation rate is found to be larger in the upper layer (18 ka to present) than in the lower one (41 ka to 25 ka), by a factor of three. The larger accumulation rate of mercury is probably associated with warmer temperatures and a higher frequency of forest fires during the Holocene.

## INTRODUCTION

This work is part of a project studying paleoclimatic and paleoenvironmental changes in the Brazilian Amazon over the past 40,000 years, using geochemical tracers of natural and anthropogenic processes. Among these, mercury has proved to be a reliable tracer of paleoclimatic conditions, due to its global scale atmospheric transport and long residence time in the atmosphere (Nriagu 1996; Engstrom and Swain 1997). It has been shown that changes in atmospheric mercury deposition reflects changes in the ocean temperature and primary production (Vandal et al. 1993) of average regional temperature (Marínez-Cortizas et al. 1999), and of increases in the frequency of natural fires due to periods of drier climate (Lacerda et al. 1999). In addition, it reflects past mining activities (Lacerda and Salomons 1998), and the evolution of industrialization (Pirrone et al. 1998). Thus, at least in regions where the geological background of mercury is negligible, such as in the Amazon region, the mercury archived in sediment cores of a remote lake may be a powerful tool for the interpretation of paleoclimatology and paleoecology of the region.

Remote Amazon lakes usually have a very low sedimentation rate, and anthropogenic activities are recorded only in the uppermost millimeters of sediment. Anthropogenic sources of mercury in Brazil are mainly gold mining, and industrial activities (Lacerda 1995, 1997; Lacerda and Salomons 1998) during the second half of the 20th century, but this is not the subject of the present work.

The available data for mercury deposition rates in the Amazon region are still too fragmentary to allow any firm conclusions due to a paucity of reliable dating and the large geographical spread of the results. Preliminary studies have suggested that average background mercury deposition rates over the Amazon were fairly constant at 1.7 to 2.6 µg m$^{-2}$ yr$^{-1}$ (Lacerda et al. 1999). During the last glacial maximum (LGM), however, mercury deposition rates more than doubled, reaching about 5.3 µg m$^{-2}$ yr$^{-1}$. Also, during more recent periods of drier climates characterized by a high frequency of forest fires, mercury deposition rates were higher.

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In the present study, results from a detailed core from north Amazonia are presented. The higher frequency of dating (up to 18 layers from a 1.0 m core) has allowed a much more detailed characterization of the sedimentation rates and mercury deposition rates compared to previously published data.

**The Studied Site**

Lake sediment samples were collected from the Lagoa da Pata at the Morro dos Seis Lagos (Six Lakes Hill). Lagoa da Pata is a small (100,000 $m^2$) headwater lake, which occupies a substantial fraction (55%) of its catchment. It is controlled by the flooding dynamics of the great river of the Amazon River, and is therefore expected to be a good atmospheric deposition catcher (Porcella 1996). Lagoa da Pata is important because it is first remote lake studied in Amazon region to span glacial times, which allows the interpretation of Hg deposition rates to pre-historic background. It is located at latitude 0°16′N and longitude 66°41′W, inside the Pico da Neblina National Park, São Gabriel da Cachoeira, northern Amazon State, approximately 600 km northwest from Manaus, not far from the Venezuela border. The lake, which is about 400 m long and 4 m deep, is surrounded by a dense tropical rain forest and is remote from human activities. The climate is warm and humid, with a total annual precipitation of the order of 2900 mm, without a dry season. Other details on lake sediment stratigraphy and palynology have been previously published (Colinvaux et al. 1996a). Lake water is very soft (2 to 5 µS), pH ~5, with a temperature range of 28–30 °C (Gomes et al. 1990; Justo and Souza 1984). The lack of any mercury-bearing geological formations in the lake's basin suggests that atmospheric deposition is the most significant source of mercury to Lagoa da Pata sediments.

The main pigments (chlorophyll a, b, c, carotenoids and phaeopigments) were analyzed, by colorimetry, in the column of water. Concentrations of all these pigments were low: chlorophyll a, 1.56 mg $m^{-3}$; chlorophyll b, 1.09 mg $m^{-3}$; chlorophyll c, 3.01 mg $m^{-3}$; carotenoids, 2.08 mg $m^{-3}$. Phaeopigments (degraded chlorophyll) were also very low. The main chemical and physical-chemical parameters of the Lagoa da Pata water were very similar to those determined in nearby rainwater (Table 1), indicating that there is little addition of nutrients through run off from the catchments. The primary productivity of the lake is probably maintained by the internal cycling of nutrients promoted by the high temperature (28 °C) of the system.

Table 1  Some physical-chemical parameters of the Lagoa da Pata and rain water

| | PH | Conductivity (µs/cm) | $Cl^{-1}$ (µMol/L) | $NO_2$ (µMol/L) | $NO_3$ (µMol/L) | $SO_4$ (µMol/L) |
|---|---|---|---|---|---|---|
| Lagoa da Pata surface | 5.05 | 6.81 | 5.20 | 2.91 | 0.54 | 1.32 |
| Lagoa da Pata 5 m | 4.99 | 6.72 | 2.54 | 3.05 | 0.37 | 0.99 |
| Rainwater 16/05/1997 to 19/05/1997 | 5.23 | 4.78 | 8.80 | 1.91 | 0.59 | 0.90 |
| Rainwater 19/05/1997 to 22/05/1997 | 5.35 | 3.48 | 2.47 | 1.45 | | 1.27 |
| Rainwater 22/05/1997 to 24/05/1997 | 5.57 | 3.49 | 5.34 | 2.02 | 0.38 | 0.97 |

We have assumed that the organic carbon in the sediment was in isotopic equilibrium with the contemporaneous atmosphere at the time of deposition. This assumption is supported by the observation

that the water in Lagoa da Pata has low pH, very low conductivity and therefore low $CaCO_3$, and that the drainage basin has an iron rich crust.

## MATERIAL AND METHODS

Samples also were collected in accordance with accepted protocols (Porcella 1996). A lake sediment core (called LPT IV) approximately 1.0 m in length was collected with a piston corer in a sub-basin of the lake at a water depth of 5 m. The core was sliced in 2-cm layers. Samples were stored in acid-cleaned plastic bags and frozen for transport.

In the laboratory, the sediment samples were dried prior to digestion. Duplicate sub-samples of about 1.0 g were digested with 50% *aqua regia* for one hour at 70 °C in a closed system. Mercury was analyzed through CVAAS (cold vapor atomic absorption spectrometry), in a Bacharach MAS-50D mercury analyzer system. Differences between duplicates were always smaller than 10%. Simultaneous determination of mercury in reference standards (NIST–USA, "Buffalo River sediments"; with 60 ng $g^{-1}$ of mercury) was performed, using the same analytical procedure, and the results were 58 ± 6 ng $g^{-1}$ (n=15). The detection limit of the method, taken to be three times the value of reagent blanks, was 0.4 ng $g^{-1}$.

The bulk density (grams dry sediment/$cm^3$ bulk sediment) of each layer was obtained by taking out 8 $cm^3$ of a wet sediment section and drying it at 60 °C until a constant weight was reached. Total organic carbon (TOC) and nitrogen were measured in a CHN automatic analyzer at Environmental Isotope Laboratory, University of Waterloo, Canada. Mercury fluxes were obtained from the product of the sedimentation rate (cm $yr^{-1}$), bulk density (g $cm^{-3}$), and mercury concentration (ng $g^{-1}$).

## Radiocarbon Dating

A group of 18 samples was selected for the determination of the chronology of the core. Samples of approximately 1 g of lake sediment were sent to the Australian National University (ANU) to be prepared and dated by AMS. All samples were acid washed with 10% HCl prior to a conventional acid-base-acid (ABA) pretreatment procedure. This was followed by combustion to $CO_2$ at 900 °C in a closed tube containing CuO and silver wire, and conversion to graphite by catalytic reduction of the $CO_2$ on iron powder in the presence of hydrogen (Santos 1999a; Santos et al. 1999b).

The fraction of material remaining after the chemical pretreatment procedure ranged from 20% to 70% of the original amount. Samples at the lower end of this range came from between 20 and 60 cm of the core where the total organic carbon content of the bulk sediments were lowest, as shown in Figure 1. In order to determine the radiocarbon backgrounds for the combustion and graphitization procedures, a sample of Sri Lankan graphite ("infinitely" old) was processed in the same way as the natural samples.

Conventional ^{14}C ages (in BP; Stuiver and Polach 1977) were determined by accelerator mass spectrometry (AMS) using the 14UD accelerator at the ANU. Results are shown in Table 2. The uncertainties shown are the statistical errors only. The reproducibility of the ANU system, as determined from multiple measurements on standards, is approximately 1.5%. Figure 2 shows the ^{14}C chronology of the LPT IV core as a function of cumulative mass g/$cm^2$. Conventional ^{14}C ages rather than calendar years BP have been employed in order to compare our results with the limited data covering the Holocene and prehistoric period (Lacerda et al. 1999; Colinvaux et al. 1996a; Turcq et al. 1998) in the Amazon region, which have been reported as conventional ^{14}C ages. This choice is also more appropriate for ^{14}C ages beyond 20,000 BP, where the calibration in terms of calendar years becomes more uncertain. As ^{14}C ages appear younger than true calendar ages (by about 1000 years

at 10,000 ^{14}C years ago, and by about 4000 years at 20,000 ^{14}C years ago), true calendar ages will be somewhat older than the values reported here, but this does not affect the conclusions of this work.
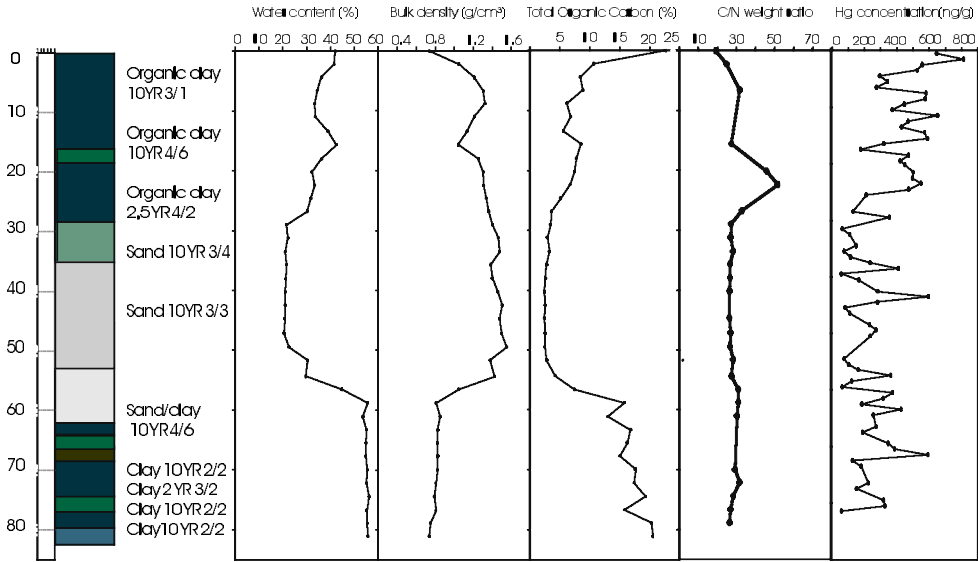


Figure 1 Water content, bulk density, total carbon concentration, carbon/nitrogen ratio and mercury concentration as a function of depth in the core
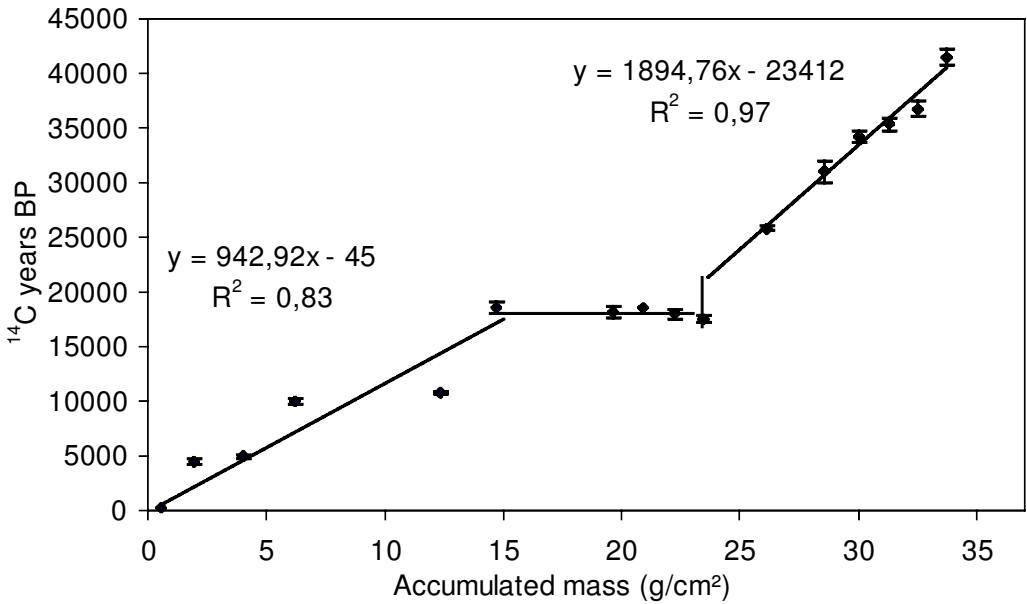


Figure 2 Radiocarbon chronology as a function of the cumulative mass

Table 2  AMS $^{14}$C dating of a lake-sediment core from Lagoa da Pata, São Gabriel da Cachoeira

| Nr | Sample name | Depth (cm) | ANUA[a] number | %M | %M error (1σ) | $^{14}$C age (BP)[b] | Age error (1σ) |
|----|-------------|-----------|----------------|-----|---------------|--------------------|----------------|
| 0 | Sri Lankan graphite | | 13002 | 0.3 | 0.1 | 47,220 | 2300 |
| 1 | LPT4-33 | 4.4–6.6 | 12319 | 57.2 | 1.5 | 4330 | 210 |
| 2 | LPT4-35 | 8.8–11.0 | 12318 | 54.0 | 1.3 | 4800 | 200 |
| 3 | LPT4-37 | 13.4–15.6 | 12317 | 28.6 | 1.0 | 9900 | 270 |
| 4[c] | LPT4-39 | 18.0–20.0 | 12316 | 56.5 | 1.3 | 4430 | 180 |
| 5 | LPT4-43 | 26.8–28.8 | 13426 | 26.1 | 0.6 | 10,650 | 180 |
| 6 | LPT4-45 | 31.2–33.4 | 12315 | 9.9 | 0.6 | 18,420 | 510 |
| 7[c] | LPT4-47 | 35.7–38.1 | 13113 | 6.8 | 0.7 | 21,400 | 810 |
| 8 | LPT4-49 | 40.2–42.4 | 12314 | 10.4 | 0.6 | 18,020 | 485 |
| 9 | LPT4-50 | 42.5–44.7 | 13112 | 9.9 | 0.3 | 18,410 | 275 |
| 10 | LPT4-51 | 44.7–46.9 | 13110 | 10.6 | 0.3 | 17,850 | 210 |
| 11 | LPT4-52 | 47.2–49.4 | 13425 | 11.2 | 0.2 | 17,410 | 155 |
| 12 | LPT4-54 | 51.7–53.9 | 13512 | 4.0 | 0.5 | 25,690 | 950 |
| 13 | LPT4-56 | 56.6–58.8 | 13107 | 2.1 | 0.1 | 30,845 | 510 |
| 14 | LPT4-58 | 61.1–63.3 | 13106 | 1.4 | 0.1 | 34,080 | 640 |
| 15 | LPT4-60 | 65.0–67.7 | 13105 | 1.2 | 0.1 | 35,180 | 680 |
| 16 | LPT4-62 | 70.4–72.2 | 13104 | 1.0 | 0.1 | 36,610 | 780 |
| 17 | LPT4-64 | 74.5–76.7 | 13103 | 0.6 | 0.1 | 41,360 | 1060 |
| 18[c] | LPT4-67 | 81.1–83.3 | 13102 | 1.3 | 0.1 | 34,740 | 690 |

[a]Code number for ANU-AMS measurements

[b]A blank correction of 0.3 ± 0.1 pMC has been subtracted from the measured pMC for each sample. Measurements of δ$^{13}$C for a subset of these samples clustered tightly around –34‰, and this value was assumed for all samples in calculating the $^{14}$C age.

[c]Samples 4, 7, and 18 are out of sequence. We suspect that sample 18 was contaminated during the core collection procedure. Further analysis will be require to understand these anomalous results.

## RESULTS AND DISCUSSIONS

The ages obtained show a good correlation with depth in the core with $^{14}$C ages ranging between 4500 and 41,500 BP. Three samples, however, do not follow the general trend. The deepest sample, 18, appears to be younger than sample 17, probably due to contamination from young surface material during the core collection procedure. In addition, samples 4 and 7 are out of sequence. Although there is no evidence in the lithology of this section for a sharp erosion event, which could explain an inversion in the sediment layers, these samples were not used in defining the sedimentation rates (Figure 2). Further analysis will be required to understand these anomalous results.

From this chronology, sedimentation rates could be derived. They were calculated using a linear model correlating ages and depths. Three different sedimentation regimes are evident, which are also reflected in other physical and chemical properties of the deposited material as can be seen in Figure 1. Four time intervals representing different paleoclimatic processes that influenced the characteristics of the deposited material can be identified.

1. From 42 to 26 ka BP, corresponding to depths in the core of 76–52 cm, the sedimentation rate is steady at 0.00165 cm/yr. This facie consists of organic-rich clay with average water content of about 45.6% and an average bulk density of 0.76 g cm$^{-3}$. The average total organic carbon concentration was highest in this section (12.3%) with a C/N weight ratio of about 28.5, suggesting an allochtounous lignocellulosic material as the major carbon source.
2. At about 50 cm in the core, there is a hiatus corresponding to ages between 25,700 and 18,000 BP. The same feature is evident in another core from the same lake (Colinvaux et al.

1996a) as well as in six more sedimentation core records in lowland tropical forests in Brazil (Ledru et al. 1998), but at present, a satisfactory explanation is lacking.

3. At around 18,000 BP, there appears to have been a sudden input of ~20 cm of clastic material. This is represented by a sandy facies, which exhibits lower carbon and water contents (2.7% and 21.2%, respectively) and higher bulk density (1.25 g cm$^{-3}$) than the layers above and below it. It also shows the lowest C/N ratio (26.8). This event may have been due to the occurrence of sudden and torrential rains, typical of dry climate, on a very local scale at about 18 ka BP. The sedimentation rate, therefore, was very rapid over the depth range from 54 to 31 cm and cannot be calculated from the data.

4. During the tardiglacial and the Holocene periods the sedimentation rate has been reasonably constant at 0.00192 cm/yr. This facie of the core is again organic-rich clay with average water content of 37.0% and average bulk density of 1.01 g cm$^{-3}$. Total organic carbon concentrations present the highest value in the top of the core reaching 22.9%. The C/N weight ratio (29.7) in this phase is similar to the precedent phases.

Figure 3 shows mercury deposition rates for the two periods where deposition rates are well defined. Table 3 summarizes, for these two periods, the average values of mercury concentration and deposition rate, as well as the average of the other parameters shown in Figure 1. Although there are significant fluctuations in the mercury flux, it is clear from Figure 3 and Table 3 that the average flux since the LGM was almost a factor of three higher than prior to the LGM. Natural emissions of Hg in Brazil are very small due to a lack of volcanic activity and significant Hg–ore deposits, and arise from re-volatilization of deposited Hg from soils and water, and from long-range atmospheric transport. The observed increase in mercury deposition rate after the LGM, may therefore be linked to drier periods and a higher frequency of forest fires, which release larger amounts of mercury from the biomass and the soils. Evidence for an increased frequency of forest fires comes from charcoal deposits in soils (Pessenda et al. 1998a, 1998b) and sediments (Cordeiro 1995; Cordeiro et al. 1997; Lacerda et al. 1999; Turcq et al. 1998).
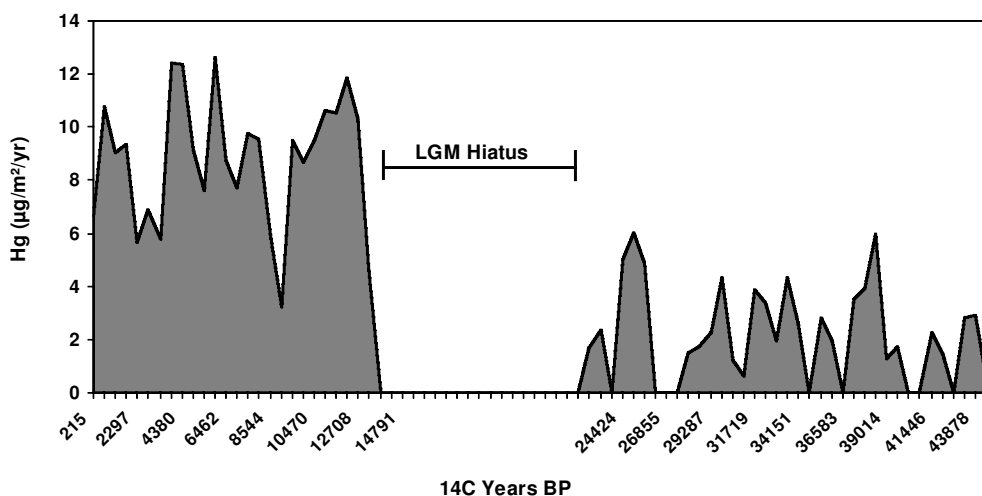


Figure 3  Mercury accumulation rates, in the sediment from Lagoa da Pata at São Gabriel da Cachoeira, as a function of time before and after the Last Glacial Maximum

In the clastic event about 18,000 years ago, mercury concentrations were highly variable, but the average value of 210 ± 160 ng g⁻¹ is similar to the pre-LGM value of 233 ± 130 ng g⁻¹ (Table 2). In contrast, the post-LGM value of 453 ± 155 ng g⁻¹ is considerably higher than either. The pre-LGM mercury concentrations observed here are, however, significantly lower than those observed elsewhere in the world (Lacerda et al. 1999).

Table 3  Average values of water content, bulk density, total organic carbon concentration, carbon/nitrogen ratio, and mercury concentration and flux for three facies corresponding to the periods indicated.

| | %H$_2$O | Bulk density (g cm$^{-3}$) | Total organic carbon (%) | C/N | Hg concentration (ng g$^{-1}$) | Hg flux (μg m$^{-2}$ yr$^{-1}$) |
|---|---|---|---|---|---|---|
| Before LGM 41,500 to ~26,000 | 45.6 | 0.76 | 12.3 | 28,5 | 233 ± 130 | 2.8 ± 1.5 |
| ~18,000 | 21.2 | 1.25 | 2.7 | 26,8 | 210 ± 161 | |
| 18,000 to present | 37.0 | 1.01 | 9.3 | 29,7 | 453 ± 155 | 8.6 ± 2.6 |

## CONCLUSIONS

The distribution of mercury in lake sediment cores can be used as a tracer of paleoclimatic conditions and may be a powerful tool for the interpretation of paleoclimatology and paleoecology of the region. In the present study, mercury accumulation rates were derived from a sediment core from Lagoa da Pata, a remote lake in Northern Amazon. A good chronology for this core, which spanned the age range from 4.3 to 41.4 ka BP was obtained by AMS ¹⁴C dating of the sediments following an ABA pretreatment regime.

The principal finding of this work was that average mercury deposition rates were three times higher after the LGM than before it. This is probably associated with warmer temperatures and a higher frequency of forest fires during the Holocene.

The present results are in broad agreement with the limited data from previous work in the Amazon region (Colinvaux et al. 1996a,b; Ledru et al. 1998; Turcq et al. 1998). The more detailed chronology obtained in the present work, however, allows a more reliable characterization of the sedimentation rates, and hence of mercury deposition rates, than previously published for the Amazon region.

## ACKNOWLEDGMENTS

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