

## Wet Deposition of Mercury and Dissolved Organic Carbon during Pre-Monsoon and Monsoon Periods at Sitapuri Site in Delhi (India)

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### Abstract

This rainwater chemistry study was carried out during monsoon and pre-monsoon seasons in 2018. The rainwater samples were collected in the residential area of Sitapuri which is situated the southwest zone of city Delhi. The rainwater samples were collected with the help of the funnel and bottle assembly on event basis. To refrain any contamination from the ground, the assembly was mounted at 5m above the ground level on a terrace. The assembly was always installed on the onset of rain and retrieved soon after the rain stopped. The collected samples were filtered. The samples were analyzed for Mercury ( $Hg_0$ ), Total Nitrogen (TN) and Dissolved Organic Carbon (DOC). The  $Hg_0$  determination was done using Differential Pulse Anodic Stripping Voltammetry through standard addition methods and DOC species and TN species were determined using DOC/TN analyzer (Shimadzu model LCPH/CPN). The average concentration of  $Hg_0$  was recorded as 54.9  $\mu g/l$ , while that of DOC and TN as 160.2 mg/l and 12.6 mg/l respectively. The study indicated that  $Hg_0$  and DOC were not contributed by common emission sources. Also, the study indicated that pre-monsoon air was more contaminated with  $Hg_0$ , TN and DOC as compared to the monsoon season.



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### Introduction


Atmospheric mercury is mainly found in inorganic forms i.e., a) gaseous oxidised mercury (GOM), (b) gaseous elemental mercury, (c) particulate bound mercury. In addition, its organic form as methyl mercury can be bio-accumulated in aquatic biota.<sup>1</sup> Because of its various states in the atmosphere

and its ability to get adsorbed on the surface of any substance, it becomes difficult to gather comprehensive information about the complete cycle of mercury compounds. As mercury forms the bridge between terrestrial and aquatic ecosystem, its transformation and transport in the troposphere have high importance.

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In rainwater samples, mercury is found in reactive forms such as reactive gaseous mercury (RGM) and dissolved particulate mercury (HgP). Major sources of mercury include waste incineration, ore roasting, coal-fired power plants. It is also emitted by natural sources e.g., geologically bound mercury release by natural biotic/abiotic processes. Hg aerosol dissolution is affected by the droplets pH, aerosol chemical composition, amount of suspended or organic matter which affect Hg oxidation/reduction reactions.<sup>2</sup> Meteorological parameters such as temperature, relative humidity, wind speed, mixing height etc. play very important role in the complex reactions of mercury in the atmosphere. It can be transported to far places. To control Hg emissions from power plants, air toxic standards have been defined by the United States Environmental Protection Agency (USEPA) to limit Hg and other pollutants.<sup>1</sup> Surprisingly, the atmospheric mercury is not extensively studied in India. Only few reports on mercury have been documented. In 2014, Kumari and co-workers have reviewed the south Asian scenario of atmospheric mercury.<sup>3</sup> They have reported particulate mercury in rain samples which was the first report on atmospheric Hg in India.

The process of wet scavenging of pollutants also helps in the removal of carbon species in the form of dissolved organic carbon from the atmosphere through precipitation. According to report, the organic carbon species range from  $10^4$  to  $10^5$  mg l<sup>-1</sup> in rain water.<sup>4</sup> However, the occurrence of total non-methane organic carbon or dissolved organic carbon (DOC) has not been estimated accurately yet. The total reservoir of DOC was initially demonstrated to be about 16 Tg C in the atmosphere, with enormous spatial and temporal inconsistencies.<sup>5</sup> The release of reduced biogenic volatile organic compounds (VOCs) such as isoprene and terpenes, have been estimated around  $1000 \text{ Tg yr}^{-1}$  which makes these emissions as the most predominant terrestrial sources of DOC in the air.<sup>6</sup> Significant anthropogenic contributions of DOC include emissions from biomass burning and hydrocarbons.<sup>7</sup>

After getting released into the ambient air, the organic compounds undergo different chemical transformations such as oxidation reactions, fragmentation or oligomerization or dehydration, or hydration etc. along with physical transformations which may include undergoing condensation to

the aerosol phase. Their fate in the atmosphere is determined either through termination by getting converted to  $\text{CO}_2$  and CO, or through dry and wet deposition to the surface in the form of gases or aerosols.<sup>10</sup> Organic compounds can also undergo photolysis with oxidants such as the hydroxyl radical ( $\text{OH}^*$ ), nitrate radical ( $\text{NO}_3$ ), ozone  $\text{O}_3^*$  and halogen radicals (e.g.  $\text{Cl}^*$ ,  $\text{Br}^*$ ) at infinitely altered reaction rates through distinct mechanisms in the atmosphere. These compounds get oxidized in the troposphere, yielding an extensive variability of more oxygenated compounds over intervals of hours to weeks.<sup>8</sup>

Along with other pollutants, N compounds are also scavenged effectively. Major components of total inorganic nitrogen (TIN) in precipitation are ammonium ( $\text{NH}_4^+$ ) and nitrate ions ( $\text{NO}_3^-$ ).<sup>9</sup> In (1995), Galloway has reported that agricultural activities are major contributors of  $\text{NH}_3$  in Asia. Delhi and its neighbouring states such as Punjab, Haryana, and Uttar Pradesh have commonly practiced agrarian occupations which are significant sources of atmospheric  $\text{NH}_3$ . The expeditious urbanization and the transboundary movement of pollutants could be the reason for the increased levels of NOx in Delhi. Singh and Kulshrestha<sup>11</sup> have reported that  $\text{NO}_3^-$  concentrations increased by more than twice in the urban site rain samples as compared to the rural site. The NOx and  $\text{NO}_3^-$  are mostly contributed by automobile emissions.<sup>12-13</sup> In addition,  $\text{NO}_3^-$  concentrations might be higher at the urban as well as the rural sites due to prevalent sources such as emissions from biomass burning and industrial emissions.<sup>11</sup>

Therefore, it is important to measure mercury, carbon and nitrogen in rain water and to identify their possible sources. For the identification of potential sources and the interlinkages between origins of mercury, organic carbon species and TN species, precipitation chemistry is one of the major mediums. This study aims to measure the levels and identify sources of  $\text{Hg}_0$ , DOC and TN in rain water samples at an urban site in Delhi.

## Methodology

### Site Description

The megacity of Delhi is one of the most vulnerable cities of India due to massive population, urban and industrial activities. The sample collection site

Sitapuri is positioned in south-west zone, Delhi. The area can be characterized as residential colony. Geographical coordinates of Sitapuri 28.6126° N,

77.0822° E. The mapping Fig. 1 shows the location of selected site.

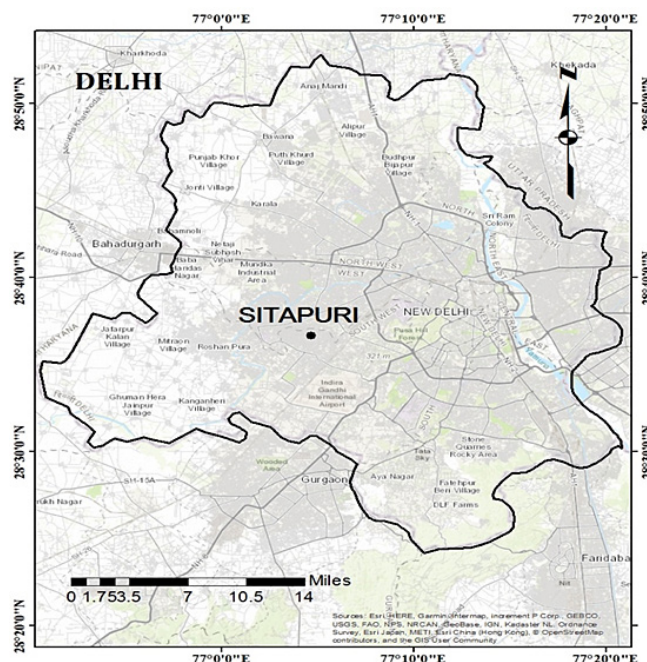


Fig. 1: Location of sampling site

### Sample Collection

The collection of rain samples was done with the help of assembled funnel and bottle. The assembly was mounted on terrace 5m above ground level to refrain any contamination. The collecting assembly was installed just before the rain and was removed as soon as the rain stopped to collect on event basis. The funnel was removed from the bottle to measure the volume of rainwater. The pH and conductivity of the samples were measured as soon as possible using the pH meter and conductivity meter.

### Sample preparation

The rainwater samples were filtered using 0.47 mm nylon filter and were preserved in pre-washed polypropylene bottles with 5% nitric acid. These bottles were soaked in 5% nitric acid overnight before use followed by rinsing with milli-Q water. All glassware and sample containers were washed in the same manner. For quality control (QC), field blanks were collected and analyzed in similar manner as other samples. The mercury analysis was done using 10 ml of

extracted solution. Trace metal determination was done using the remaining solution. Rain samples were preserved at 4°C in the refrigerator and were used for DOC and TN analysis.

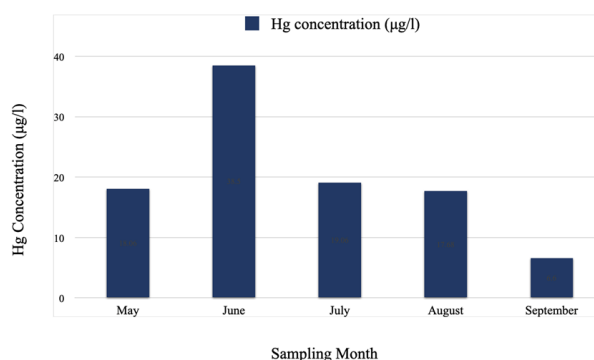
### Determination of Mercury, DOC and TN

Mercury was determined by using 797 VA Computrace (Metrohm, Switzerland) mercury analyser with Differential Pulse Anodic Stripping Voltammetry (DPASV) technique. The voltammetry is widely used technique for mercury determination in environmental samples.<sup>14-18</sup> The instrument consists of working electrode placed in the centre made up using Gold (Au-RDE) of 2 mm, a reference electrode Ag/AgCl which is filled with KCl electrolyte and a glass carbon auxiliary electrode. The gold working electrode was kept in 0.1 M NaOH for at least half an hour for conditioning purpose. In the stripping voltammetry technique, the analyte is determined in two steps- i) in the deposition step, the substance is to be deposited on the working electrode at a constant potential up to a controlled deposition time, ii) stripping step in which the deposited substance

in the first step is dissolved in the solution. During deposition, 370 mV deposition potential was applied for 260 seconds. During stripping step, 0.30 to 0.84 V potential was scanned at a sweep rate of 0.02 V/s. The peak potential was 0.521 V. The Anodic Stripping Voltammetry (ASV) has remarkably low detection limits (0.1  $\mu\text{g/L}$ ) and hence, this technique is proved to be efficient when it comes to analyse sample to find out heavy metal concentration at ppb level.

The stock solution of 1000 ppm  $\text{Hg}^{2+}$  was prepared on weekly basis for which 0.1345 g of  $\text{HgCl}_2$  (Merck) was dissolved in de-ionized water. The working standards of mercury (1 mg/L and 10  $\mu\text{g/L}$ ) were prepared from 1000 ppm stock solution at the time of daily analysis. The electrolyte of voltammetric cell had perchloric acid, ethylenediaminetetraacetic acid (EDTA) and potassium chloride (KCl). In a 10 ml sample, perchloric acid (300  $\mu\text{l}$ ), EDTA solution

(400  $\mu\text{l}$ ) and of KCl solution (100  $\mu\text{l}$ ) were added into the cell followed by nitrogen purging for 300 seconds to remove any dissolved oxygen from the sample. Then as the first standard of addition, 200  $\mu\text{l}$  of 1 mg/L  $\text{Hg}^{2+}$  standard was added to the cell vessel. Further, the replicate voltammograms were taken to ensure the repeatability. Afterwards, second standard of addition (200  $\mu\text{l}$  of 1 mg/L  $\text{Hg}^{2+}$ ) standard was added to the cell vessel, and the corresponding voltammograms were recorded in replicates. Similarly, the water blank was also analysed (Fig. 2).  $\text{Hg}^{2+}$  concentration was calculated using peak height method. The preparation of solutions was done using ultrapure water (Mili-Q). All the chemicals and reagents used in the analysis were purchased from Merck Millipore having EMPARTA grade. Total Nitrogen and dissolved Carbon species were determined by using Total Organic Carbon Analyzer. (Shimadzu model LCPH/CPN) as described elsewhere<sup>19</sup>.



**Fig. 2: Hg concentration ( $\mu\text{g l}^{-1}$ ) from May to September at Sitapuri site**

## Results and Discussion

### Variation of Hg in Rainwater

Table 1 gives the values of mercury concentrations in the samples collected at Sitapuri site. The maximum number of rain samples were collected in the month of July which falls under monsoon season (Fig.2). The Hg concentration was found to be the highest in the month of June followed by May. These are the months of pre-monsoon season in Delhi region when the atmosphere is relatively polluted and dry. This favours an accumulation of particulates leading to the elevated levels of toxins in the atmosphere. The most probable reason for these results can be their emissions from fossil fuel combustion including coal burning in brick kilns, diesel burning in vehicular movement and construction machines etc. Relatively lower values were recorded during

monsoon months i.e., July, August, and September when rain occurrence was frequent which cleaned the atmosphere as a result the atmospheric loadings of Hg were not built up effectively during monsoon period. The concentrations of mercury in precipitation samples are highly affected by the source strength and meteorological factors. Wet deposition is the process helps to eliminate mercury and other contaminants from atmosphere.<sup>20-21</sup>

### Variation of DOC in Rainwater

Table 2 gives the maximum, minimum and average values of DOC and Hg in the month of May. Variation of Hg and DOC is shown in Fig. 3. The figure shows a significant co-variation of Hg and DOC at Sitapuri site of Delhi region. The DOC concentration values were found to be the lowest in monsoon season

and the probable reason is the scavenging effect of rain. When compared with other global studies, organic carbon concentrations have been reported the lowest at Nigata site ( $274 \mu\text{gC l}^{-1}$ ), Tokya  $657 \mu\text{g C l}^{-1}$  and Sado  $273.9 \mu\text{gC l}^{-1}$ . This has been observed that carbonaceous fractions were mostly higher

during non-monsoon season due to higher pollution level and lower mixing height. In the Indian context, crop residue burning in the neighbouring states i.e., Haryana and Punjab makes situation worse during non-monsoon season when organic carbon fractions show high peaks.<sup>22</sup>

**Table 1: Concentration of Hg and DOC in rainwater along with related ratios**

S. No	Date	Hg Concentration ( $\mu\text{g l}^{-1}$ )	DOC mg l <sup>-1</sup>	TN mg l <sup>-1</sup>	DOC/Hg ratio	TN/Hg ratio
1	02/5/18	3.7	10.7	6.2	2.9	1.7
2	03/5/18	0.7	3	2.7	4.2	3.8
3	19/6/18	4.3	6.7	6.2	1.5	1.4
4	27/6/18	2.2	3.3	4.1	1.5	1.9
5	30/6/18	2.6	1.1	2.6	0.4	1.0
6	13/7/18	1.8	1.5	4.2	0.8	2.3
7	22/7/18	1.0	1.4	7.2	1.4	7.2
8	26/7/18	0.4	1.1	8.7	2.7	21.7
9	27/7/18	0.6	1.5	3.0	2.5	5.0
10	28/7/18	0.8	1.4	2.8	1.7	3.5
11	08/7/18	2.6	3.6	5.8	1.4	2.2
12	22/8/18	1.0	2.5	44.4	2.5	44.4
13	28/8/18	0.6	0.7	13.2	1.2	22.0
14	07/9/18	0.7	2.0	20.1	2.8	28.7
15	24/9/18	0.9	2.0	58.8	2.2	65.3
Average		1.6	2.8	12.7	2.0	14.1

**Table 2: Descriptive Statistics of Hg and DOC ( $\text{mg l}^{-1}$ ) for monsoon and non-monsoon samples**

	Hg ( $\mu\text{g l}^{-1}$ )		DOC ( $\text{mg l}^{-1}$ )	
	Monsoon	Pre-monsoon	Monsoon	Pre-monsoon
Average	1.0	2.7	1.8	5.0
Minimum	0.4	0.7	0.7	1.1
Maximum	2.6	4.3	3.6	10.7
<i>n</i>	10	5	10	5

Table 3 gives comparison of mercury concentrations at different cities worldwide. Among all sites, Taichung being an industrial city situated at the western side of Central Taiwan having area  $2,215 \text{ km}^2$  has shown the highest concentration of mercury dispersion.<sup>23</sup> While in case of Kodiak being an Island situated in USA, Alaska state, within the city, The transportation is being done for commercial

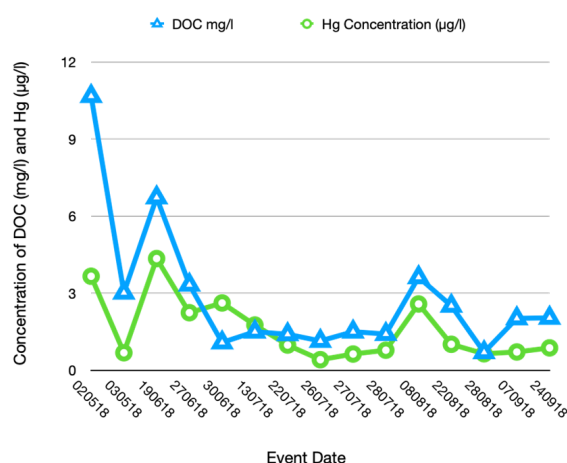
purposes within the Island or outer areas via road, ferryboat, or airline. This site is comparatively less used commercially in terms of industrial activities. The Hg concentration at our sampling site in the present study was found to be low as compared to Taichung city of China but on the other hand it was found to be equal or closely related values such as Tibetan plateau of China, Minamata Bay of Japan,

Mt. Leigong of China, Churchill city of Canada and Southern New Hampshire of USA respectively.

Interestingly, Kodiak Island of USA has recorded the lowest value of Hg concentration.

**Table 3: Comparative data of mercury concentration ( $\mu\text{g l}^{-1}$ ) at different sites worldwide**

Site	country	Site type	Sampling period	Hg ( $\mu\text{g l}^{-1}$ )	Reference
Sitapuri	India	urban	2018	4.35	Present study
Taichung	China	Urban	2017	10,539.1	24
Three Gorges Reservoir	China	Reservoir	2012-2013	18	25
Chongqing	China	Urban	2010-2014	34.25	26
Tibetan Plateau	China	Plateau	2010-2012	4.0	27
Xiamen	China	urban	2012-2013	12.3	28
Lhasa	China	Urban	2010	24.8	29
Tibetan Plateau	China	Plateau	2009-2011	4.8	30
Minamata Bay	Japan	Bay	2009-2010	5.9	31
Mt. Leigong	China	Mountain	2008-2009	4	32
Southern New Hampshire	USA	Urban	2008	12.33	33
Southern New Hampshire	USA	Urban	2007	8.41	34



**Fig. 3: DOC ( $\text{mg l}^{-1}$ ) and Hg ( $\mu\text{g l}^{-1}$ ) concentration at Sitapuri Site**

#### Variation of TN

The maximum and minimum TN was found to be  $58.8\text{mg l}^{-1}$  and  $2.66\text{mg l}^{-1}$  in monsoon and pre-monsoon seasons respectively, at Sitapuri. These values indicated the deposition of total nitrogen in the atmosphere in different forms including  $\text{NH}_3$ ,  $\text{NH}_x$ ,  $\text{NO}_2$  and  $\text{HNO}_3$ .<sup>35</sup>

Among the above-mentioned species, ammonia ( $\text{NH}_3$ ) and ammonium ( $\text{NH}_x$ ) are water soluble species in the atmosphere. The two main reasons because of which atmospheric  $\text{NH}_3$  is hazardous to the environment are; firstly, for the ecological repercussions of its deposition on delicate

ecosystems which causes eutrophication and secondly, it serves as a neutralizing agent in nature. Ammonia reacts rapidly with acidic components such as  $\text{H}_2\text{SO}_4$ ,  $\text{HNO}_3$  in the atmosphere. Neutralization by ammonia results in secondary formation of ammoniumsalts, i.e.  $(\text{NH}_4)_2\text{SO}_4$ ,  $\text{NH}_4\text{HSO}_4$  and  $\text{NH}_4\text{NO}_3$  etc., which play a salient role in radiative forcing. It is to be noted that deposition of  $\text{NH}_x$  leads to acidification of the soil similar to the acidic effect of  $\text{SO}_2$  and  $\text{NO}_x$  due to the ability to form nitrates in the soil as a result of oxidation.<sup>36</sup> Deposition of  $\text{NH}_x$  from the atmosphere leads to excess N input to the ecosystem.<sup>37-38</sup>

### Influence of Meteorological Factors

The direction of wind movement influences the dispersion of mercury. In the atmosphere, the major form of Hg is GEM. Due to its high stability and long residence time (several months to a year), GEM can be effectively carried over long distances.<sup>39</sup> In 2020, Li and co-workers have discussed that mercury is measured as a global pollutant, as it shows transboundary movement in troposphere.<sup>40</sup> The types and forms of Hg metal enter the ambient

air from various sources, and they have the potential to enter the terrestrial and aquatic ecosystems through drainage and rainfall, can be lethal if encountered by humans. So, it becomes essential to monitor mercury concentrations in precipitation. Meteorological data showed that easterly winds had maximum frequency during the month of May and in subsequent months, the frequencies were found to be more from the west direction except in July where it was found to be both easterly as well as westerly.

**Table 4: Hg/DOC ratios for rain samples over Sitapuri site**

S. No	Hg Concentration ( $\mu\text{g l}^{-1}$ )	DOC ( $\text{mg l}^{-1}$ )	Hg/DOC ratio
1	3.7	10.7	0.3
2	0.7	3.0	0.2
3	4.3	6.7	0.6
4	2.2	3.3	0.7
5	2.6	1.1	2.4
6	1.8	1.5	1.2
7	1	1.4	0.7
8	0.4	1.1	0.4
9	0.6	1.5	0.4
10	0.8	1.4	0.6
11	2.6	3.6	0.3
12	1	2.5	0.4
13	0.6	0.7	0.8
14	0.7	2.0	0.3
15	0.9	2.0	0.4

### Hg/DOC ratios

Usually, ratio between two variables ascertains their relationship and helps in identifying the sources by calculating fraction of any given variable. help to identify the sources and to calculate fraction of any variable.<sup>41</sup> Table 4 gives the individual values for mercury concentrations and DOC as well as their corresponding ratios. In case of Hg concentration, the maximum values were found to be  $4.35 \mu\text{g l}^{-1}$  and minimum was  $0.41 \mu\text{g l}^{-1}$  whereas in case of DOC maximum and minimum values were  $10.68$  and  $0.70 \mu\text{g l}^{-1}$  respectively. The ratios of DOC/Hg and TN/Hg indicated different emission sources in Sitapuri. The Hg/DOC ratio had an order of range. It varied from 0.2 to 2.4. In general, the ratio values were less than 1 except two samples. This indicated that the DOC concentrations were much higher than Hg. In addition to the ratios, trend lines in Fig. 3 showed that the possible sources at Sitapuri site contributed

more to DOC concentrations as compared to Hg emissions. This might be due to local source contamination which cannot be explained at present and needs further long-term investigations.

### Conclusions

DOC, TN, and Hg were measured at urban site in New Delhi. The mean values for Hg, DOC and TN were noticed as  $1.6 \mu\text{g l}^{-1}$ ,  $2.8 \text{ mg l}^{-1}$  and  $12.7 \text{ mg l}^{-1}$ , respectively having a range between  $0.4$ - $4.3 \mu\text{g l}^{-1}$ ,  $0.7$ - $10.7 \text{ mg l}^{-1}$  and  $2.7$ - $58.8 \text{ mg l}^{-1}$ , respectively. The values found to be higher during initial precipitation events, especially during pre-monsoon season and dissimilar chemical and physical mechanisms emerge to control the atmospheric concentrations of Hg. DOC/Hg and TN/Hg ratios indicated that the sources present at the site did not contribute uniformly to DOC, TN and Hg levels such that the concentrations of DOC and TN were usually higher

in rainwater. It implies that local influences affect the ambient Hg concentrations to a great degree. The probable sources could be domestic animals, biomass burning, human population; and waste incineration, coal combustion, cement for reactive nitrogen and mercury respectively.

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#### Conflict of Interest

The authors do not have any conflict of interest.

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