# **Exploring the Health Impact on Antimicrobial Contaminants in Leather Industries Effluent in Vellore, India: Distribution and Risk Analysis**

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#### *Abstract*

*This study investigates the presence and risks of common industrial antimicrobial agents used, in the vicinity of Vellore's leather industrial hub, Tamil Nadu, India. Water and sediment samples (n=25 each) were collected from two major tannery clusters, Ranipet and Ambur-Vaniyambadi. Using GC-MS and UPLC-DAD analyses, we attempted to detect six major antimicrobial classes such as chlorinated phenols, 2-(Thiocyanomethylthio) benzothiazole (TCMTB), ortho phenyl phenol (OPP), Para chloro mcresol (PCMC), triclosan (TCS) and n-Octyl-isothiazolinone (n-OIT). Among them, only TCMTB, OPP, and PCMK were recorded in water samples, whereas in sediment samples none of the antimicrobials were found. Detection frequencies in water were 24%, with PCMC levels reaching up to 910 µg/L, TCMTB at 45.3 µg/L, and OPP at 821.3 µg/L. A comparative analysis with global studies showed that contaminant levels in this study are significantly higher than in other country reports. Human health risk assessment, based on USEPA methods, revealed a risk quotient (RQ) of 5.200 for PCMC, indicating a potential health risk, whereas TCMTB and OPP presented lower risks. Environmental risk assessment indicated hazard quotients (HQ) >1 for all detected antimicrobials, suggesting significant ecological risks. The study highlights the need for stringent regulations, improved industrial practices, and better wastewater treatment infrastructure. Continuous monitoring and more comprehensive studies with increased sampling are essential to develop strategies for reducing contaminants from leather industry runoff. These measures are crucial for protecting human health and aquatic ecosystems in the region.*

*Keywords: Antimicrobial Agents, Biocide, Environmental Risk, Leather Industry Human Health Risk, Water Contamination.*

## **Introduction**

Antimicrobial agents, encompassing biocides, preservatives and disinfectants, play a crucial role in various industries to control microbial contamination [1]. They are essential for ensuring product safety and longevity in consumer goods, pharmaceuticals, food processing, and notably, the leather industry. However, their indiscriminate use raises significant environmental and health concerns. Residual antimicrobial compounds from industrial processes often enter water bodies, causing pollution that adversely affects aquatic ecosystems and human health [2]. Broad-range applicability and repeated exposure to biocides confer the creation of tolerant strains of microbes such as antibiotic-resisting MDR (multi-drug resistance strains) [3]. This has dramatically raised the concern of serious lifeimpacting microbial infections in humans and other creatures [4].

Vellore, located in Tamil Nadu, India, is a prominent hub for leather production, particularly known for its high concentration of tanneries [5]. The leather industry in Vellore is a vital contributor to the local economy, offering employment opportunities and contributing significantly to India's leather

exports [6]. This leather hub plays a significant economic role, contributing substantially to the state's GDP and India's overall leather exports [5]. However, this economic benefit comes with environmental costs that must be managed sustainably to ensure long-term industrial viability and ecological balance.

Although numerous chemicals are used in the leather industry and its allied industries at various stages, some of the antimicrobial compounds are contaminants of emerging concern. They include CPs, TCMTB, OPP, PCMC, TCS and n-OIT. Among them, CPs are used as antiseptics but its highly toxic to aquatic life and can cause liver damage and reproductive issues in fish [7]. TCMTB is a fungicide used in leather processing which is extremely toxic to aquatic organisms, leading to high mortality rates among fish and invertebrates [8, 9]. OPP employed as a disinfectant is toxic to fish and disrupts endocrine functions, potentially causing reproductive issues [10, 11]. PCMC known for its biocidal properties is also an endocrine disruptor in aquatic organisms, affecting growth and reproduction [12]. TCS, one of the most commonly used antimicrobial agents is highly toxic to algae and fish, disrupting thyroid hormone metabolism and causing DNA damage [13, 14]. n-OIT, which has antifungal and antibacterial properties can cause severe allergic reactions in aquatic species and contribute to multidrug resistance [15].

While these contaminants are effective in preventing microbial growth during leather processing, they pose significant environmental risks when discharged into water bodies without adequate treatment. In developing countries such as India, the wastewater treatment facility is inadequate and removal of these contaminants has not been well studied. Their persistence can lead to bioaccumulation in aquatic organisms and potentially enter the human food chain, exacerbating health risks and contributing to antimicrobial resistance [2].

Despite the pivotal role of the leather industry in India, there exists a substantial gap in data regarding the environmental impact of antimicrobial contaminants [6]. Previous studies have predominantly focused on regions outside India, leaving critical gaps in understanding the local impact of these pollutants. Comprehensive data on the concentration levels, distribution, and ecological risks of antimicrobial agents in Indian leather industrial zones are sparse, hindering the development of effective regulatory frameworks and pollution mitigation strategies tailored to the Indian context [1]. Considering the above problems, this study aims to fill these knowledge gaps by investigating the presence and risks of antimicrobial agents in water and sediment samples from the Vellore leather industrial zone. The objectives are (1) to quantify the levels of selected antimicrobial agents (CPs, TCMTB, OPP, PCMC, TCS, and n-OIT) in water and sediment samples, (2) to assess the environmental and human health risks associated with these contaminants, (3) to compare contamination levels with global data to understand their relative impact, and (4) to underscore the need for continuous monitoring and comprehensive studies in the region. By achieving these objectives, the study aims to support the formulation of effective pollution control strategies, thereby safeguarding human health and the environment in Vellore. Moreover, this study emphasizes the critical need to address the environmental impact of antimicrobial agents in the leather industry, particularly in Vellore. By providing robust data on contaminant levels and associated risks, the study aims to support sustainable industrial practices and enhance regulatory frameworks in India.

## **Materials and Methods**

## **Study Area and Sample Collection**

The samples were collected in two major clusters of the industrial area Ranipet and the Ambur and Vaniyambadi of Vellore district in Tamil Nadu, India (Figure 1).



**Figure 1.** Map Showing the Sampling Area and its Industrial Clusters are Located in Vellore District, Tamil Nadu, India

Based on the local source and confluence points, the water and sediment samples were gathered in and around effluent treatment plants of the above two clusters mentioned during the year 2017-19. A total of 25 water and sediment were collected from the sampling points mentioned in Table 1.

The sterile plastic sampling bottles (2L) were rinsed with the sample water thrice and then the sample was collected. In the case of sediment, samples were collected as grab samples, in a sterile and clean polypropylene bag carefully taken to the laboratory and retained at a low temperature (-20℃) until the extraction process. These samples were thoroughly dried subjected to homogenization and extracted as per the standard methods.



**Table 1.** List of Locations from where the Samples were Collected and its Geo-Coordinates



## **Extraction of Antimicrobial Agents from the Surface Water Samples**

For the extraction of five antimicrobial agents such as PCMC, OPP, TCMTB, TCS and n-OIT from water samples, the following method based on EPA 8270 D was employed. Exactly, 100 mL of water sample was adjusted to pH 6.5 by adding formic acid and then transferred into a clean separating funnel. To this, 90 mL of dichloromethane was with intermittent shaking for 10 minutes and this procedure was repeated twice, in which the developed organic layer was gently collected. The collected organic layer was evaporated to dryness by passing nitrogen gas. Then the dried content was reconstituted with 2 mL of acetonitrile and gently vortexed and filtered using a 0.22 µm filter before analysing in the UPLC-DAD system.

For the extraction of chlorinated phenols from the water samples, the method described in ISO 14154-2005 was followed. Briefly, 100 mL was mixed with sodium hydroxide to bring the pH to 9.0. An aliquot (20 mL) of the sample was mixed with 50 µL tetra chloro guaiacol stock (10 mg/mL), 0.5 mL of triethylamine, 0.5 mL of acetic anhydride, and 2 mL of n-hexane (2 mL). Then the entire mixture was gently placed on a mechanical shaker (200 rpm) for an hour and the organic layer was separated, passed through a filter  $(0.45 \mu m)$  and finally analysed in the GC-MS.

# **Extraction of Antimicrobial Agents from the Sediment Samples**

The five grams of sediment sample was mixed with 20 mL of acetonitrile and subjected to sonication for an hour at 37 ℃**.** Then the extract was passed through a 0.22 µm filter and condensed to the required volume using nitrogen gas before injecting it into the UPLC-DAD instrument.

The extraction of chlorinated phenol from sediment was performed as per the standard analytical methodology ISO-14154-2005. Briefly, five grams of sediment sample was mixed with potassium hydroxide solution and incubated for 12 hours at 90 ℃. Then, ten millilitres of sample was taken and 50 µL of tetra chloro guaiacol stock (10 mg/mL), acetic acid (0.5 mL) and triethylamine (0.5 mL) were added to it. The entire mixture was placed on a mechanical shaker operated at 200 rpm for an hour and the organic layer gathered was passed via a filter  $(0.45 \text{ µm})$  and analysed through GC-MS.

# **Assessment of Five Antimicrobial Agents in Water and Sediment Samples using UPLC-DAD**

To detect the presence of PCMK, o-PP, TCMTB, TCS and n-OIT, the extracted samples were screened in the Agilent 1260 UPLC-DAD instrument. The instrument was incorporated with a diode-array detector (1290 model),

quaternary pump and an autosampler. The Zodiac column used for the separation of analytes had a dimension of 5 µm x 4.6 µm x 150 mm (length x width x height). Water and acetonitrile were used as mobile phases A and B, respectively, with a flow rate of 1 mL/min. Initially, 50% gradient (acetonitrile) was

retained for 2 minutes and it was further ramped to 80% for 9 minutes, finally a reduction in the gradient was done at 9.01 minutes for 10 minutes. The compounds assessed and the target wavelength applied for the identification and quantification of individual compounds were furnished in Table 2.



26530-20-1 280 279

**Table 2.** Antimicrobials and Biocides (Other than Chlorinated Phenols) were Analysed in Water and Sediment Using UPLC-DAD (Agilent 1260 Infinity) and their Signature Wavelength

During calibration, the linearity range acquired for all the analytes was excellent  $(R^2 >$ 0.992), authenticating the robustness of the method. A good recovery percentage of 89%- 97% was obtained for both the tested water and sediment samples. The limit of detection (LOD) and limit of quantification (LOQ) obtained for the water sample were 0.001 mg/L and 0.01

5. n-Octyl

isothiazolinone

mg.L, respectively. In the case of sediment, LOD and LOQ were 1 mg/kg and 10 mg/kg, respectively. The peaks obtained in the chromatogram were analyzed against the linearity plots to calculate the concentration of analytes. A standard and overlay chromatogram (blank, standard and the screened sample) obtained was shown elsewhere in Figure 2.



**Figure 2.** Standard and Overlay Chromatogram (Blank, Standard and Sample) of Antimicrobial Agents in UPLC

### **Assessment of Chlorinated Phenols in Water And Sediment Samples Using GC-MS**

For analysing chlorinated phenols in water samples, the extracted samples were individually analysed in GC-MS from Agilent Technologies (GC-7890B; MS-5977A) fitted with a capillary column (30 m x  $250 \mu m$  x  $0.25$ ) µm). The injection port temperature was set as 280 °C. A split ratio of 5:1 was used when injecting the sample in split mode. A 70 °C

initial oven temperature was set with a 0.5 minute hold. After that, the temperature was raised to 150°C at a pace of 40°C per minute and then to 200  $\mathrm{^{\circ}C}$  at a rate of 6 $\mathrm{^{\circ}C}$  per minute, with a final ramp up to 280  $\degree$ C at the rate of 50 °C/min with a hold of 1 min. The temperature of the transfer line was retained at 280 ℃ and the 150 ℃ was the temperature of quadrupole. The details of the chlorinated phenols analysed were mentioned in Table 3 along with the target ions which were applied to quantify them.

**Table 3.** List of Chlorinated Phenols and their Target Ions (Q1 and Q2) Analysed in Water and Sediment

Samples Using GC-MS	



The linearity achieved for all the screened chlorinated phenols,  $R^2 > 0.99$ , indicates the robustness of the overall procedure. The water sample  $(2 \mu g/L)$  to 50  $\mu g/L$ ) and the collected sediment (10  $\mu$ g/L to 500  $\mu$ g/L) had linearity

plot  $R^2 > 0.99$ . The recovery range for CPs in water and sediment samples was in the range of 86 - 93%. The LOD and LOQ were 2 µg/kg and 20 µg/kg, respectively for the chlorinated phenols. In the GC-MS analysis, the results

were presented as chromatographic signals, which were subsequently analyzed by software. The peaks obtained in the chromatograms were processed along with the standard peak values to find the concentration of CPs. The quantification of the detected samples was performed based on the obtained linearity index. A representative standard and overlay chromatogram (blank, standard, and sample) were given in Figure 3.



**Figure 3.** Standard and Overlay Chromatogram (Blank, Standard and Sample) of Chlorinated Phenols in GC-MS

#### **Result**

The study evaluated the water and sediment samples collected around the leather factories in Vellore were subjected to GC-MS and UPLC-DAD analysis to determine major antimicrobials and biocides such as chlorinated phenols, TCMTP, o-PP, PCMK, triclosan and n-OIT. Based on the levels their environmental and human health risk was also ascertained as per the USEPA regulations.

# **Occurrence and Distribution of Antimicrobials and Biocides**

Among the 25 locations, antimicrobial agents were recorded only in water samples and not in any of the sediment samples, in all three years. The detection frequency of antimicrobial agents in water was 24% which means 6 locations showed at least one contaminant in any of the 3 years. The individual levels of

detected contaminants such as PCMC, TCMTB and OPP were in the range of ND (not detected)  $-820 \mu$ g/L, ND – 224  $\mu$ g/L and ND – 910  $\mu$ g/L, respectively. Three contaminants viz. TCS, n-OIT and CP were not detected in any of the samples. The average concentration of each contaminant recorded in each year was shown in Figure 4 and it indicates that PCMC levels were higher than OPP and TCMTB. During the year 2017, PCMC and OPP were recorded as 821.3 µg/L and 20.1 µg/L, respectively in W22 (Kanigapuram). In the case of 2018, TCMTB, OPP and PCMC were detected in one location each at  $45.3 \mu g/L$  (W18 - Gangapuram), 53  $\mu$ g/L (W19 - Palar riverside -2) and 224  $\mu$ g/L (W20 – Somalapuram), respectively. In 2019, OPP was detected only in the water sample of Kanigapuram (W22;  $35.5 \mu g/L$ ), whereas PCMK was recorded in two locations namely W23 (Pernambet; 910 µg/L) and W24 (150  $\mu$ g/L).



**Figure 4.** Levels of Antimicrobial Agents in Water Samples Collected from Vellore District, Tamilnadu

# **Human Health Risk Assessment of Detected Antimicrobial Agents**

To perform a risk assessment for the detected pollutants, the USEPA risk assessment method for drinking water was followed. It consists of identifying the Reference Dose (RfD), calculating the exposure dose and finally determining the risk. The RfD is an estimate of the daily exposure to the human population (including sensitive subgroups) that is likely to be without an appreciable risk of harmful effects during a lifetime. The exposure dose

(mg/kg/day) is calculated based on the maximum concentration of the contaminant in drinking water, the average daily water intake, and the body weight of the individual based on the formula (1), and the risk quotient (RQ) was determined by the formula (2):

 $Exposure$  do  $=$ [(Concentration in water  $(\mu g/L) \times$ Water intake  $(L/day)$  /  $Body weight (kg) | x 10$  (1)

Risk quotient (RQ) = 
$$
Exposure \frac{dose}{RfD}
$$
 (2)

**Table 4.** Drinking Water Health Risk Assessment of Antimicrobial Compounds Detected in Surface Waters of Vellore District, Tamil Nadu

<b>Pollutant</b>	<b>RfD</b>	<b>Maximum</b>	Average	Average	<b>Exposure</b>	<b>RQ</b>
	(mg/kg/day)	<b>Level in Surface</b>	Water daily	Adult <b>Body</b>	<b>Dose</b>	
		Water $(\mu g/L)$	<b>Intake</b>	Weight (kg)	(mg/kg/day)	
			(L/day)			
TCMTB <sup>a</sup>	0.003	45.3		70	0.00129	0.431
OPP <sub>b</sub>	0.03	52.8		70	0.00151	0.050
<b>PCMC</b> <sup>c</sup>	0.005	910		70	0.02600	5.200

<sup>a</sup>USEPA (1984), <sup>b</sup>USEPA (2006), <sup>c</sup>NTIS (1997)

This RQ ratio indicates the potential health risk, with a value greater than 1 suggesting potential concern. The calculated exposure doses for PCMC (0.02600 mg/kg/day) were the highest (Table 4).

## **Environmental Risk Assessment of Detected Antimicrobial Agents**

The pollutants released into the environment undergo various interactions with the

environmental compartments and affect the habitats of the ecosystem. In this study, the pollutants that are considered to be inadvertently released from industries are reported in the water. So, it's important to understand the risk posed by it to the aquatic organisms as the runoff of the Vellore district often drains into the nearby Palar River. The environmental risk assessment was performed according to the USEPA guidelines, where the

hazard quotient (HQ) is determined by the water quality criterion and the measured environmental concentration as shown in formula (3):

> HQ = Measured Environmental Concentration/PNEC (3)

The maximum concentration reported in this study was taken for the conservative risk assessment and the predicted no-effect concentration (PNEC) was obtained from the USEPA database (USEPA, 2023). The value of  $HO < 1$  indicates that the concentration is below the benchmark and is considered safe for the environment whereas  $\geq 1$  indicates that the concentration exceeds the benchmark, potentially posing a risk to the environment.

In the present study, the HO was  $>1$  for all the three detected antimicrobials and they were in the order of 10s and 100s of magnitude higher than PNEC (Table 5).

**Table 5** Environmental Risk Assessment of Antimicrobial Compounds Detected in Surface Waters of Vellore



District, Tamil Nadu

### **Discussion**

The occurrence and distribution of antimicrobials are minimal and restricted to Ambur and Vaniyambadi Cluster. This cluster has more factories and waste generation than the Ranipet cluster.

The comparison of TCMTB, PCMC, and OPP levels in surface waters and industrial effluents across different regions of other countries reveals significant variations, highlighting the diverse impact of industrial activities on water quality. In India, TCMTB levels ranged from ND to 45.3 µg/L in the surface waters of this study, near leather industries. In Pakistan (Kasur), levels ranged from 5 to 40  $\mu$ g/L [16] and in China (Beijing), from ND to 35  $\mu$ g/L [17], both lower than in India. South Africa (KwaZulu-Natal) reported levels between 8 and 50 µg/L, comparable to India [18]. For PCMC, Indian surface waters near tanneries had levels up to 910 µg/L, significantly higher than in Brazil (5 to 100  $\mu$ g/L) [19], USA (10 to 150  $\mu$ g/L) (Johnson et al., 2022), and Germany  $(15 \text{ to } 110 \text{ µg/L})$ (Schmidt et al., 2021). OPP levels in India ranged from ND to  $821.3 \mu g/L$ , higher than those in Germany (25 to 80  $\mu$ g/L), USA (20 to 70  $\mu$ g/L) and China (10 to 50  $\mu$ g/L) [17].

Comparing the surface water data with effluent studies, the contamination levels are generally higher in effluents. For example, in Spain (Industrial effluents) and South Korea (Tannery effluents), the levels of TCMTB ranged from ND to 50 µg/L and 12 to 48 µg/L, respectively [20, 21]. PCMC levels in these regions were between 6 to 22 µg/L (Spain) and 9 to 28 µg/L (South Korea). OPP levels ranged from 3 to 12  $\mu$ g/L (Spain) and 2 to 11  $\mu$ g/L (South Korea). These concentrations, although significant, are often lower than the maximum levels reported in surface waters near Indian tanneries, especially for PCMC.

Overall, the high levels of TCMTB, PCMC, and OPP in India are likely due to inadequate wastewater treatment and lax regulatory enforcement. The presence of these contaminants in water indicates their continuous usage over time. These findings underscore the need for stringent regulatory measures, better industrial practices, and improved wastewater treatment infrastructure to mitigate contamination and protect water quality.

Also, the PCMC reported was 6.3 times higher than the EC50 value (130  $\mu$ g/L) of fish *Acartia tonsa* (planktonic copepod) [22] and about 60 times higher than PNEC set by the Australian government (15 μg/L) [23].

Since the levels in water are quite significant, the chances of the development of multidrugresistant (MDR) bacteria in the environment are also high. The detected contaminants, including PCMC, OPP, and TCMTB, have been linked to MDR mechanisms in various studies. For instance, prolonged exposure to orthophenylphenol (OPP) can induce genetic changes in bacteria such as *Pseudomonas aeruginosa*, which can upregulate genes associated with drug resistance and virulence [24, 25]. Similarly, fungicide-driven resistance mechanisms involving energy-dependent efflux transporters have been observed in fungi exposed to compounds similar to PCMC and TCMTB, suggesting potential cross-resistance in bacterial populations [26]. This can further pave the way for outbursts of seasonal flu and other contagious diseases.

Although these contaminants are seldom detected in sediment, future monitoring should not be ignored due to their potential factors such as high-water solubility and mobility [27] faster degradation rates in sediment [28] and sediment characteristics that prevent binding [29]. Pollutants from surface water can settle into sediment through adsorption onto particulate matter, potentially accumulating over time if input exceeds degradation. Upwelling and sediment disturbances can also reintroduce contaminants into the water column [30]. Moreover, if the degradability of these pollutants is limited in the sediment, they may transform into by-products, complicating their

identification in sediment. Continuous monitoring in both matrices is therefore crucial to prevent long-term ecological impacts.

Overall risk analysis indicates alarming contaminant levels in water, necessitating immediate action to protect the aquatic ecosystem. A comprehensive study with increased sampling points and frequency is essential. This approach will provide a robust perspective for developing effective monitoring and mitigation strategies to reduce contaminant levels in runoff from leather industries, safeguarding aquatic ecosystems.

### **Conclusion**

In conclusion, this study underscores the critical environmental risks posed by antimicrobial compounds and biocides around Vellore's leather industrial hub. Of the 25 water samples analyzed, antimicrobial agents were detected only in 24% of the samples, with PCMC exhibiting the highest concentration at 910 µg/L, followed by OPP at 821.3 µg/L and TCMTB at 45.3 µg/L. Although these levels are significantly higher than those reported in similar industrial regions globally, the occurrence and distribution of these pollutants are restricted to a few locations. In the case of sediments, none of the targeted pollutants were traced in, indicating other factors involved in the complex sediment matrix such as possible transformation into by-products. Further, the human health risk assessment showed risky levels of PCMC. Environmental risk assessment showed hazard quotients (HQ) well above 1 for all three detected antimicrobial agents, particularly PCMC with HQ > 500. These findings indicate substantial ecological threats and highlight the urgent need for stringent regulatory measures, improved industrial practices, and enhanced wastewater treatment infrastructure. The presence of these biocides as an industrial pollutant raises concerns about multidrug-resistant strains causing serious microbial infections and adverse impacts on aquatic life. Therefore, continuous and comprehensive monitoring, with increased sampling frequency and coverage, is necessary to develop effective strategies for reducing the discharge of these contaminants, thereby protecting both human health and aquatic ecosystems. Additionally, measures must be taken by the government and leather industries to prevent the dumping of biocides into aquatic bodies, without any treatment.

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#### **Conflict of Interests**

The authors declared no conflict of interest.

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