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Research Article

MERCURY CONTAMINATION FROM PULP: COMBINED PAPER MILL UNTREATED EFFLUENT AND SOLID WASTE IN INDIA

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ABSTRACT

Mercury is a major toxic pollutant that is transported over long distances and can bioaccumulate and contaminate. Currently, solid waste of coal fuel and paper mill effluent that emits most to atmospheric Hg and has the high ratio in the industrial outlet. Near the paper mill environment mercury is generally elevated, particularly in effluent, air and water bodies in India. We have selected 20 paper mills and take reference from Google web to finding concentration of mercury. Untreated paper mill effluent and their air spread in local environment and remote areas elevate the concentration of mercury compared than other places of clean environment. Large river estuaries in the world are often heavily affected by upstream industrial effluents. Mercury is also elevated in mud of paper mill effluent, sediments, a direct result of contamination in river systems. Solid waste of paper mill contained high concentration of mercury that can contaminate ground water of local areas of paper mill industry. Studies on mercury in fish showed generally low levels of contamination resulting from low bioaccumulation of Hg in the mostly short food chains. Paper mill worker affected from the air and dust of mercury, and also damage their skin and nails. Due to, mercurous air around the paper mill industry asthma and respiratory diseases observed in the peoples.

INTRODUCTION

The metallic element mercury has been known since ancient times and was certainly mentioned by Aristotle (350 B. C.). Mercury is a global pollutant that can be transported over long distances and can bioaccumulation. Currently, mercury emission from an industrial unit is a global challenging task to the natural environment. Pulp and paper mills generate huge amount of wastewater depending on the type of processes used in the plant whose unsafe disposal can be very hazardous to environment and human health. The production of pulp and paper is increasing worldwide and wastes are therefore being generated in appreciable amounts (Sahinkaya, et al., 2017; Ahirwar, et al., 2015). Due to Hg toxicity, even a minimal content may have a negative impact on biota the levels of mercury emissions from municipal solid waste combustors, emissions of mercury from fossil fuel burning in the United States currently amount to about 300 tonnes of mercury released to the atmosphere. Heavy metal contamination of soil may pose risks and hazards to humans and the ecosystem through, direct ingestion or contact with contaminated soil, the food chain (soil-plant-human or soil-plant-animal human), drinking of contaminated ground water etc. Heavy metals are among the contaminants in the environment. Beside the natural activities, almost all human activities also have potential contribution to produce heavy metals as side effects. Hg in fish and the aquatic environment is also a great problem in the Nordic region of the EU (European Union). Hg in the regulation of waste is regarded as a dangerous substance which, when contained in waste, is one of the properties, leading to a classification of waste as hazardous. Tannery industry is a primary pollutant of the environment and has a strong potential to cause soil and water pollution owing to the discharge of untreated effluent. Generally, Hg is not likely to persist in

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large quantities as dissolved species, since Hg (0) tends to evolve in the gas phase and divalent Hg (II) sorbs strongly to particulate phases including organic carbon and sulfides. However, Hg (II) has the potential to associate with or form colloidal particles that can be mobilized in porous media under high organic carbon conditions. It has been suggested that pulp and paper mill effluent may stimulate bacterial mercury methylation in these sediments leading to contamination of aquatic biota. Metal toxicity has direct effects to flora that forms an integral component of ecosystems. Altered biochemical, physiological, and metabolic processes are found in plants growing in regions of high metal pollution. The data relating to mercury waste, such as inventories, thresholds and mercury concentrations in municipal and hazardous wastes, were limited or did not exist, and the amount of mercury in waste at the global level remains unclear. According to an official European Union directive, mercury is categorized as a threat to aquatic ecosystems, as toxic through inhalation and as hazardous to human's health. Mercury (Hg) emissions from anthropogenic sources pose a global problem. The exact chemical composition of pulp and paper mill effluent is complex and unclear. The chemical compound found in pulp and paper mill effluent are mostly degrading products of lignin, cellulose, hemicellulose and wood extractives. The lignin degradation products found in the pulp and paper mill effluent include a wide variety of compounds such as manomeric phenols, enolethers, mercaptides, stilbene, quinone derivatives, chlorinated phenols, acetic acid, formic acid, acetaldehyde, methanol, furfural and methyl glyoxal. Thus, an overview of Hg pollution in India by paper industry is urgently needed. The present study revealed to contaminated environments and summarizes the environmental status of Hg in India to improve understanding at state and national levels. By Tab. 1. denoted the most paper mill city in India that contaminates soil, river, air and underground water.

MATERIALS AND METHODS

The present study provides an overview of the most recent studies on Hg in the Indian paper mill industries. The literature cited here was extracted from the Google web of Chrome Brower using the following keywords: mercury (Hg) and effluent from Indian paper mill (Fig. 1).

Mercury Outlet

Untreated effluent of paper mill: Discharges of paper mill has huge amount of complex content of mercury which release into environment through pipeline. The toxic mercury concentration was found 0.070 ppm in untreated effluent of Orient paper mill, Amalai, at boiler tunnel. Maximum reduction (66.5%) in Hg content of untreated paper mill effluent was observed using L. minor followed by T. natans (64.8%). Acknowledgements, Concentration of chemical species in sludge waste water of Bhili steel plant and paper mill industry, Hg was 0.12, 0.06, 0.06, 0.09, 0.09, 0.06, 0.06, 0.03 and 0.09 mg/L.

Solid waste and mud of paper mill: Discarded mercury-containing products, if not recycled, ultimately release mercury to air, soil, and groundwater, even after being properly collected and disposed of in MSW management facilities. The paper making has far-reaching environment impacts due to the production of waste in the form of sludge since that is creating major problem to soil, crop and human health due to presences of different toxic chemicals arsenic, cadmium, lead and mercury in India. Semi- solid soil conditions are typically favorable for the formation of inorganic and organic compounds with mercury. The level of mercury in soil is an indicator of its potential to contaminate groundwater and surface runoff. Soil contamination could be caused either by direct dumping or land filling of mercury contaminated wastes. Mercury in water body sediments may indicate the history of contamination.

Fig. 1 Map of various paper mill of India and study sites. **Note:** (----) International boundary; (-----) State boundary; (●) Paper and pulp industry

Tab 1. List of paper mill industry in India.

S.No.	Name of paper mill	State names
1	Yamunanagar	Haryana
2	Saharanpur	Uttar Pradesh
3	Bahadurgarh	Haryana
4	Titagarh	West Bengal
5	Ballarshah	Maharashtra
6	Kaghaznagar	Maharashtra
7	Kamalapuram	Maharashtra
8	Pune	Maharashtra
9	Vikarabad	Andhra Pradesh
10	Sarapaka	Andhra Pradesh
11	Dandel	Karnataka
12	kagithapuram	Tamil Nadu
13	Rajahmundary	Andhra Pradesh
14	Kakinada	Andhra Pradesh
15	Goganpur	Orissa
16	Jaykaypur	Orissa
17	Orient paper mill	Amalai
18	Nepanagar	Amalai
19	Ajanta paper and Board mill	Raipur
20	Jai Mata Di Paper mill	Raipur

The Minamata Bay had to be dredged of toxic mercury contaminated sediments in order to restore the water quality.

Emission of Mercury in Environment

Inorganic mercury: Metallic mercury is an allergen, which may cause contact eczema, and mercury from amalgam fillings may give rise to oral lichen. It has been feared that mercury in amalgam may cause a variety of symptoms. This so-called 'amalgam disease' is, however, controversial, and although some authors claim proof of symptom relief after removal of dental amalgam fillings. Mercury must emit from the paper mill as gases by multiple evaporators, coal burning and boiler heating.

Organic mercury: Methyl mercury poisoning has latency of 1 month or longer after acute exposure, and the main symptoms relate to nervous system damage. The earliest symptoms are parestesias and numbness in the hands and feet. Later, coordination difficulties and concentric constriction of the visual field may develop as well as auditory symptoms. High doses may lead to death, usually 2-4 weeks after onset of symptoms. The Minamata catastrophe in Japan in the 1950s was caused by methyl mercury poisoning from fish contaminated by mercury discharges to the surrounding sea. In the early 1970s, more than 10,000 persons in Iraq were poisoned by eating bread baked from mercury-polluted grain, and several thousand people died as a consequence of the poisoning. Mercury in Aquatic Environment Methyl mercury can be formed in the environment by microbial metabolism. The efficiency of microbial mercury methylation generally depends on factors such as microbial activity and the concentration of bioavailable mercury, which in turn are influenced by temperature, pH, redox potential and the presence of inorganic and organic complexing agents (Amano, et al.,2020; Saadia, et al.,2010; Banana, et al.,2016).

Air: Mercury levels in air are in the range 2–10 ng/m3. Study revealed that many types cancer as well as respiratory and cardiovascular diseases to air pollution emitted by pulp and paper mills. The making process of paper sulphur compounds and nitrogen oxides are emitted to the air. In pulp and paper industry air pollution is caused due to odour emitting reduced sulphur compounds such as hydrogen sulphide, methyl mercaptan, dimethyl sulphide and particulate matter, SO_2, NOx present in the gases emitted by different process. The most significant airborne mercury discharge occurs through burning of coal (1-2 lb of Hg/1000 ton of pulp).

Freshwater environments and river: About 75% of marine plants contained the maximum contaminations during the summer season and in fish samples Hg2+ concentrations exceeded the levels provided by international standards. Paper-pulp mills discharge only a very small amount of mercury (about 0.1 lb of Hg/1000 ton of pulp) into the waterways. Actual mercury accumulation in the riverbeds around three mills was found to be around 0.1 ppm. Crop plants collected from the contaminated site showed higher level of residual Hg and Cd and significant depletion in pigment was observed. Plants collected from both the sides of the treated effluent canal showed significant amount residue

mercury and cadmium in the plant leaves near from JK paper mill, Orissa in India. The general population is primarily exposed to mercury via food, fish being a major source of methyl mercury exposure, and dental amalgam. The general population does not face a significant health risk from methyl mercury, although certain groups with high fish consumption may attain blood levels associated with a low risk of neurological damage to adults. Since there is a risk to the fetus in particular, pregnant women should avoid a high intake of certain fish, such as shark, swordfish and tuna; fish (such as pike, walleye and bass) taken from polluted fresh waters should especially be avoided (Gavrilescu, et al., 2012).

Mercury Exposure to Paper Mill Workers

In general, all three forms of mercury (elemental, inorganic and organic mercury) have the potential of causing adverse health effects at sufficiently high doses. According to the quantitative assessment, occupational mercury exposure may cause a great variety of signs and symptoms, in particular in the field of neuro-psychological disorders, such as ataxia, tremor or memory problems. However, many reported symptoms were largely unspecific, such as hair loss or pain. Most of the included studies had a low methodological quality with an overall high risk of bias rating. Workers reported a high exposure to skin irritants, especially when carrying out tasks that caused the hands and feet to become wet from perspiration and having contact with process water. Atopic dermatitis was seen in 3% of the workers. Contact dermatitis was seen in 26% of the workers and 36% were diagnosed with mycosis of the feet. All cases of contact dermatitis and mycosis could be attributed to occupational exposure to skin irritants. One hundred ninety-eight current workers (124 male and 74 female) were included. There were significant associations between both cumulative exposure and years of high exposure to soft paper dust and impaired lung function. Each year of high exposure to soft paper dust was associated with a 0.87% decrease in FEV1 [95% Confidence Interval (CI) -1.39 to -0.35] and decreased FVC (-0.54%, 95% CI -1.00 to 0.08) compared to the lower exposed workers. There was an increased mortality due to obstructive lung disease (asthma and COPD), among high-exposed workers, SMR 1.89, 95% CI 1.20 to 2.83, based on 23 observed cases. High-exposed workers had an increased mortality from asthma, SMR 4.13, 95% CI 1.78 to 8.14, based on eight observed cases. The increased asthma mortality was also observed among high-exposed men, SMR 4.38, 95% CI 1.42 to 10.2, based on five observed cases. Hair loss from hands and fingers without nails (partially and fully) and other dermal problems like rashes and itching on hands were noticed in 9 out of 15 workers at secondary fiber recovery plant of the mill. This study also suggested that paper mill workers are at an increased occupational risk of developing asbestos-related cancers. Because of the long latency period involved with asbestos-related diseases, paper mill workers who worked during the period between the years, 1960 and 1980 may currently receive a diagnosis of serious diseases such as lung cancer, mesothelioma, and asbestosis (Guney, et al., 2020).

RESULTS AND DISCUSSION

We reviewed 20 most paper mill of India in which only 07 paper mills, mercury outlet regarding references are listed in Tab. 2. Saharanpur paper mill located in Uttar-pradesh and their wastewater released various pollutants in environment. A large paper mill, the concentration of mercury in the wastewater could theoretically be as high as 0.015 mg per litre. The sludge showed the abundance of benzene propanoic acid tert- butyldimethylsilyl ester, Octadecanoic acid, TMS, Hexadecanoic acid, TMS, cinnamic acid-α-phenyl-TMS ester, β-sitosterol TMS, 4-mercaptobenzoic acid as residual complex organic compounds along with heavy metal (Hg 0.014 mg/L-1) which were above the prescribed limit of environmental standard. Titagarh paper mill effluent has total mercury level ranged from 0.073 to 0.94 microg/g in both pre and post monsoon season. T. mossambicus in both season and C. mrigela at pre monsoon, cross the Indian recommended maximum limit (0.50 microg/g wet weight) for food consumption and according to World Health Organization guidelines all fish were not. Mercury was in untreated paper mill wastewater a little high at Vikarabad, while at Hyderabad and Valigonda it showed acceptable readings.

Tab 2. List of paper mill industry in India.

S.No.	Paper mill located in city (India)	Mercury released from paper mill toward various medium			
		Effluent	Air	River	Soil
1.	Yamunanagar	-	-	-	-
2.	Saharanpur	Effluent	-	River	Soil
3.	Bahadurgarh	-	-	-	-
4.	Titagarh	Effluent	Air	River	-
5.	Ballarshah	-	-	-	-
6.	Kaghaznagar	-	-	-	-
7.	Kamalapuram	-	-	-	-

8.	Pune	-	-	-	-
9.	Vikarabad	Effluent	-	-	-
10.	Sarapaka	-	-	-	-
11.	Dandel	-	-	-	-
12.	Kagithapuram	-	-	-	Soil
13.	Rajahmundary	-	-	-	-
14.	Kakinada	-	-	River	-
15.	Goganpur	-	-	-	-
16.	Jaykaypur	Effluent	-	River	Soil
17.	OPM, Amalai	Effluent	-	River	Soil
18.	Nepanagar	-	-	-	-
19.	Ajanta paper and Board mill	-	-	-	-
20.	Jai Mata Di Paper mill	-	-	-	-

The factory had dumped mercury filled glass thermometers in the scrapyard in Kodaikanal and the nearby watershed forest, causing soil and water pollution in the area. Activists alleged that the mercury pollution caused by the now defunct factory unit led to deaths of at least 45 factory workers and 12 children. Organic mercury compounds (especially phenyl mercury acetate) are utilized to prevent microorganisms (bacteria, fungi, algae, etc.) from growing in the pulp, during its digestion and processing. Crop plants collected from the contaminated site showed higher level of residual Hg and Cd and significant depletion in pigment was observed. Plants collected from both the sides of the treated effluent canal showed significant amount residue mercury and cadmium in the plant leaves. The toxic mercury concentration was found 0.070 ppm in sampling station SS-01 in August, 2014 near Orient paper mill, Amalai (Singh, et al.,2019; Singh, et al., 2020; Srivastava, et al., 2017).

Strengths and Limitations

This study provided an overview of the emission and contamination related problems caused by ongoing mercury exposure. In contrast, the major limitation of this study was the failed attempt to carry out a meta-analysis due to the unsuitable data for this procedure. Another restriction was the limited publication period, ending on 1 November 2022. Therefore, articles that were published later and could have also fulfilled the eligibility criteria could not be considered (Jukka, et al., 1994).

CONCLUSION

Least numbers of studies have been carried out to evaluate the Hg pollution from paper effluent mill. These studies generally document well the Hg levels in the nearby environment. demonstrate the mercury pollution by Indian paper mill which are lack of informative data or study not done in mercury outlet from paper mill. Comprehensive monitoring of Hg in the overall environment is lacking. Thus, in the published literature, a bias exists toward studies at contaminated or highly polluted areas. Establishing routine monitoring of Hg in near paper mill industry media is therefore important. Another major uncertainty regarding Hg pollution is the lack of an Hg budget (release-flux) estimate at the national level. Relatively good estimates of atmospheric emissions from major Hg emitting sectors exist, but a comprehensive release inventory is missing. The exact amount of Hg circulating in Indian paper mill and other for intentional use remains unclear. Due to this difficulty is assessing the extent of the Hg pollution problem.

The Hg levels in air and water bodies in large parts of the Indian urban environments are elevated, but their monitoring is done properly. Mercury concentrations in air show seasonal variations resulting maximum from heating with coal in the winter months. Large river estuaries are heavily impacted by upstream industrial sources. Mercury levels in the soil usually depend on local geological conditions. Fish and vegetables, which are considered as the most important source of methyl mercury (R-Hg) exposure from paper mill are generally safe for consumption except for those from a very few mining areas. The available data show that mostly peoples affected various diseases causes' mercury pollution near paper mill industry.

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