

IDENTIFICATION OF SOURCES OF CHEMICAL AND BIOAEROSOL EMISSIONS INTO THE WORK ENVIRONMENT DURING SECONDARY TREATMENT OF PULP MILL EFFLUENTS

Nicole Goyer and Jacques Lavoie, Institut de recherche en santé et sécurité du travail du Québec, 505 West De Maisonneuve, Montreal, Quebec, Canada, H3A, 3C2

ABSTRACT

This study identified chemical compounds, namely sulfur compounds and bioaerosols which are present in operations associated with pulp mill effluent secondary treatment systems, and which may be released into workplace atmospheres in concentrations sufficient to warrant worker exposure concern. They are emitted into the air due to their presence in the raw effluent and sludge; therefore, any action that promotes turbulent contact of air with the effluents and sludge will increase release rates of the compounds into the air. As a result, emissions can be minimized by controlling the operations where air-water contact occurs. This information can also be used in designing future worker exposure assessments, and in developing specific control strategies, where warranted.

KEYWORDS: pulp and paper mills, effluent treatment, secondary treatment, biological treatment, emission sources.

APPLICATION STATEMENT

The emission sources of chemical compounds and bioaerosols of potential worker exposure concern have been identified in operations associated with effluent secondary treatment in paper mills. The results were used to identify corrective and preventive measures that could be taken to minimize these emissions. This information may be used to facilitate worker exposure and exposure control assessments.

INTRODUCTION

To comply with new environmental standards relating to the quality of industrial effluents, more than 40 Quebec pulp and paper mills have implemented secondary treatment systems for their effluents. This complex process is based on the action of microorganisms, which convert the organic load of the effluent into more stable products such as water, carbon dioxide, ammoniac, methane and residual sludge, thus allowing water of acceptable quality to be discharged into the receiving stream. However, due to the nature of the compounds present in the effluent, the

variable biodegradability of the chemical structures and the variations in environmental conditions, several chemical compounds are produced. These systems created concerns and complaints from workers and neighboring communities about the possible presence in the air of microorganisms (bioaerosols), chemical substances and disagreeable odors emitted by this biological action. Scientific articles have been published about workers' exposure to chemicals in pulp and paper production plants, and about workers' exposure to bioaerosols at municipal sewage treatment plants, and at facilities recycling or composting solid wastes.⁽¹⁻¹²⁾ But only one publication deals specifically with the emission of chemicals into the environment during the secondary treatment of paper mill effluent.⁽⁹⁾ It discusses a sulfite pulp mill and presents emission rates for two chemical compounds, chloroform and hydrogen sulfide; no measurement were taken in the work environment and no information was provided about the factors that promote their formation or propagation into the air, or about emission control mechanisms.

The objectives of this study were to identify and evaluate process sources of emissions of chemicals and bioaerosols of possible worker exposure concern. The results were used to identify corrective and preventive measures that could be taken to minimize these emissions. This information will also be useful to conduct exposure measurements and control assessment studies at similar facilities.

METHODOLOGY

This field study was carried out in eleven paper mills chosen so as to cover all of the variables related to: 1. the types of manufacturing processes or uses of pulp: kraft pulp, sulfite pulp, thermomechanical pulp and de-inked pulp; 2. the type of treated effluents: process water alone, or process water and sanitary wastewater for the plant or the municipality; and 3. the types of treatment: conventional activated sludge treatment, oxygen-activated sludge treatment, sequential biological reactor, and aerated pond. The overall characteristics of the eleven paper mills are presented in **Table I**.

▪ Description of the measuring stations

At each mill, measuring stations were located as close as possible to all potential sources of emissions associated with the wastewater collection, handling, and treatment systems. **Figure 1** diagrams the steps in the process and indicates the location of the measuring stations. Accessory buildings such as pump rooms and tank rooms, and specific locations such as sumps, sink drains and floor piping, were also evaluated. Simulations for two specific tasks were carried out to verify whether they were contaminant emission sources. This involved the collection of water or sludge samples and the cleaning of orifices in the distribution basin of the cooling towers. For bioaerosols, samples were collected 300 meters upwind in order to measure the background level in the air on the day of the intervention, as well as 100 meters downwind from the secondary treatment plants and land filling sites in order to determine the level of emission into the environment.⁽¹³⁾

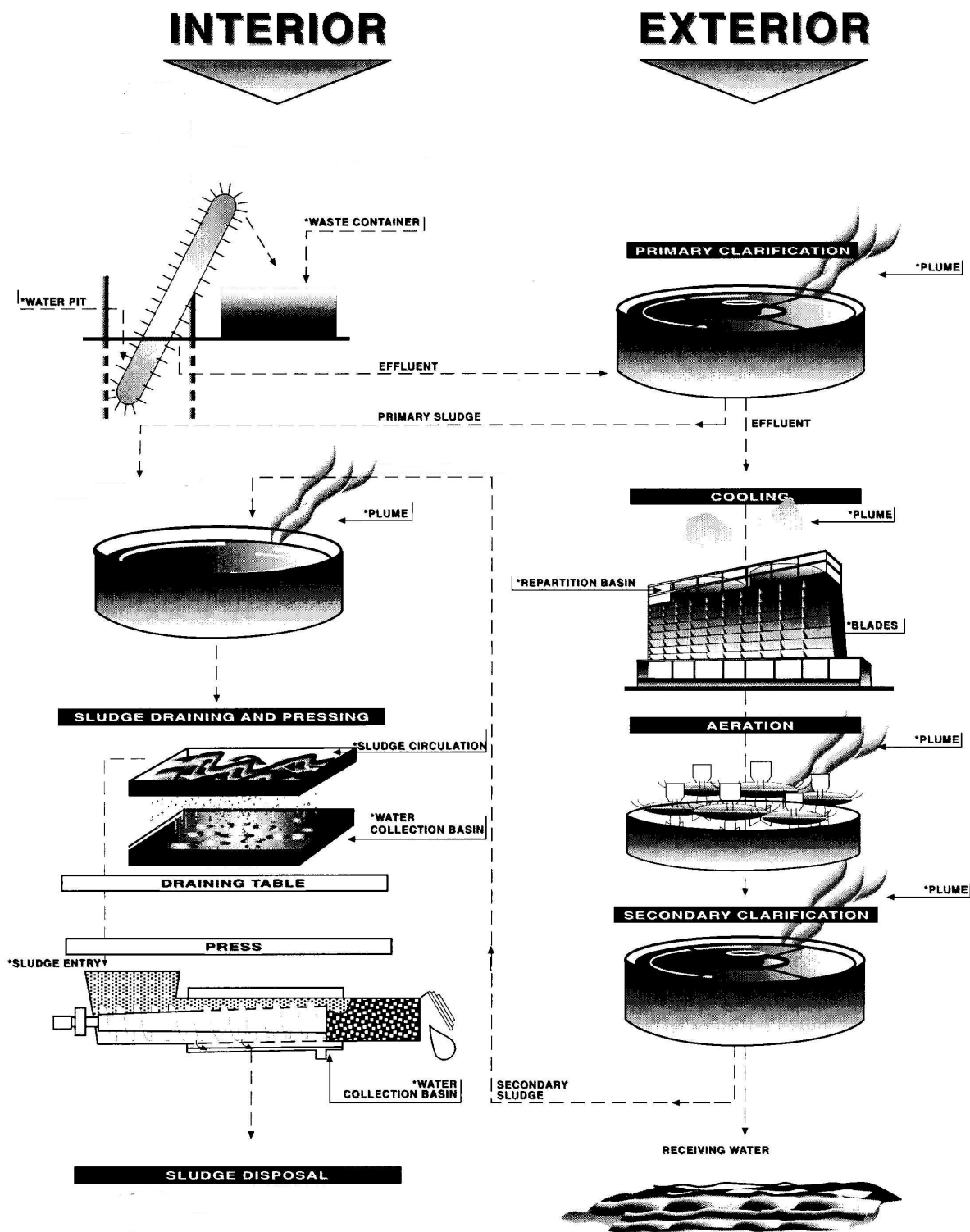
The measurements were taken in the summer during a two-or-three-day period for each mill.

Plant	Type of Pulp ^A	Effluent Treatment	Quantity of Effluent (m ³ /day)
A	kraft (100%)	conventional activated sludge	municipal: 1,000 plant: 20,000
B	kraft (100%)	aerated pond	70,000
C	kraft (100%)	oxygenated activated sludge	100,000
D	mechanical (70%) / kraft (30%)	sequential batch reactor	60,000
E	mechanical (65%)/ sulfite(28%)/kraft (7%) ^B	sequential batch reactor	35,000
F	mechanical (42%)/ sulfite (42%) /de-inked (15%) ^B /kraft (1%) ^B	conventional activated sludge	40,000
G	mechanical (97%)/ kraft (3%) ^B	conventional activated sludge	70,000
H	mechanical (80%)/ de-inked (20%)	sequential batch reactor	35,000
I	mechanical (48%)/de- inked (48%)/kraft (3%) ^B	oxygenated activated sludge	70,000
J	de-inked (95%) ^B /kraft (5%) ^B	conventional activated sludge	municipal : 2,000 plant : 20,000
K	kraft (70%) ^B /de-inked (30%) ^B	aerated pond	7,000

^A The proportion of the type of pulp is indicated between parentheses

^B This type of pulp is purchased, not produced in the plant

Table I. Description of the Paper Plants.



* Sampling site

Figure 1. Secondary Treatment of Paper Mill Effluents.

▪ Description of the parameters measured

Three sets of parameters were considered, namely chemical compounds, odors and microorganisms.

Chemical compounds

Chemical compounds in dust, gas or vapor form were measured. The techniques were chosen so as to determine known compounds in this industry and identify unknown compounds. They were measured by direct-reading instruments: a portable chromatograph (Voyager, PE Photovac, New York) for sulfur compounds and volatile organic compounds; electrochemical cell dataloggers (Toxilog and Toxi Ultra, Biosystems Inc., Rockfall, CT) for carbon monoxide, nitrogen oxides, sulfur dioxide, chlorine and hydrogen sulfide; and an infrared analyzer (ADC-PM-3, Nortech Control Equipment Inc., Etobicoke, Ontario, Canada) for carbon dioxide. With the electrochemical analyzers, the data were recorded for four to five hours. With the other instruments, three to five instantaneous readings were taken. The instruments were calibrated using IRSST standard methods.⁽¹⁴⁾

For mills A, D, E and G, the chemical compounds were also measured by a mobile laboratory. Detection was done in positive and negative ionization modes using a tandem mass spectrometer (MS/MS) equipped with a source operating at atmospheric pressure (Sciex, TAGA 6000E). The analyses were carried out continuously every 500 milliseconds for a period of approximately 30 minutes. The main contaminants investigated were the following: hydrogen sulfide, mercaptans, sulfides, terpenes, alcohols, ketones, organic acids, ammonia, amines and sulfur dioxide.

Odors

The substances responsible for the odors were identified by comparing the measured concentrations with the odor thresholds reported in the literature.^(4,15-17) The presence of sulfur compounds, terpenes and organic acids was also noted by odor, even when their concentration was below the quantification limit.

Bioaerosols

Sampling was done with impactors (Andersen Instruments Incorporated, Atlanta, USA) on nutrient media for total bacteria, Gram negative bacteria, total molds and the mold *Aspergillus fumigatus*. Two samples were collected simultaneously at each of the measuring sites. The sampling flow was 28 L/min for one minute. After incubation, the colonies formed were counted. The endotoxins produced by the Gram-negative bacteria were sampled on polycarbonate filters for four simultaneous samples. The sampling time was four hours. They were determined by photometry.⁽¹⁴⁾

RESULTS AND DISCUSSION

Parameters measured

Chemical compounds and odors

Table II presents the concentration ranges of the chemical compounds measured at the emission sources in relation to the process steps, specific locations and specific tasks. Only results for plants in which compounds were present in measurable or odor detectable concentrations are presented. Some of these compounds are associated with the production of odors as shown by the odor thresholds also reported in this table. Although the measurements were taken at the emission sources, the ACGIH threshold limit values for an average 8-hour exposure of workers were added as information that could help in evaluating the importance of the emissions.

Step : sampling site ^A	Chemical ^B	Threshold ^C (ppm)	Emission source ^D	
			Plant	Measured concentration (ppm)
Screening: above effluent pit (I)	H ₂ S	OLF: 0.005; TWA: 10	A, C	0.5 – 20.0
	MME	OLF: 0.0005;TWA: 0.5	A, C, D, E	0.03- 4.50
	DMS	OLF: 0.001;TWA: ---	A, C, D	0.05-1.50
	DMDS	OLF: 0.0003;TWA: ---	C, D	1.25 – 2.90
	NH ₃	OLF:5;TWA: 25	A, E	1 - 7
	CO	OLF:100,000;TWA: 25	A, C, H	18 - 44
	NO	OLF: ---;TWA: 25	C, D, E	2 - 11
	SO ₂	OLF:1;TWA: 2	E	2.2
	T	OLF:---;TWA: ---	A, C, D, E, H	P - 0.30
	MOH	OLF:160;TWA: 200	A	7
1 ST Clarification: dispersion plume (E)	H ₂ S	OLF: 0.005; TWA: 10	A, C	0.3 – 0.8
	MME	OLF: 0.0005;TWA: 0.5	A	0.40
	DMDS	OLF: 0.0003;TWA: ---	C	0.08
	NH ₃	OLF:5;TWA: 25	A	7
	Acids	OLF:0.0002;TWA:---	A	0.008
Cooling: dispersion plume (E)	H ₂ S	OLF: 0.005; TWA: 10	A, B, K	0.3 – 2.8
	MME	OLF: 0.0005;TWA: 0.5	A, B, C, D	0.07 – 2.90
	DMS	OLF: 0.001;TWA: ---	A, B, C, D	0.03 – 2.20
	DMDS	OLF: 0.0003;TWA: ---	A, B, C, D	0.06 – 2.40
	NH ₃	OLF:5;TWA: 25	A	6
	Acids	OLF:0.0002;TWA:---	A	0.003

Aeration: dispersion plume (E)	H ₂ S	OLF: 0.005; TWA: 10	A, B	0.5 – 0.6
2 ND Clarification: dispersion plume (E)	CO ₂	OLF:74000;TWA:5000	C, E, I, J	1500- 1800
	NH ₃	OLF:5;TWA: 25	A	15
	Acids	OLF:0.0002;TWA:---	A	0.016
Draining table : above sludge circulation (I)	H ₂ S	OLF: 0.005; TWA: 10	A, B, C, D, E, G, H, I	0.4- 38.0
	MME	OLF: 0.0005;TWA: 0.5	A, C, D, E, G, H	0.05 – 2.7
	DMS	OLF: 0.001;TWA: ---	A, C, D, G	0.06 – 0.41
	DMDS	OLF: 0.0003;TWA: ---	C, D	0.11 – 0.38
	NH ₃	OLF:5;TWA: 25	A	7
	CO	OLF:100,000;TWA: 25	A, E, J	8 - 16
	CO ₂	OLF:74000;TWA:5000	A, D, E	1200 -3770
	SO ₂	OLF:1;TWA: 2	E	1.4
	Acids	OLF:0.0002;TWA:---	A, F	P – 0.007
	T	OLF:---;TWA: ---	C, D, E, H, I	P
Sludge press: sludge entrance (I)	H ₂ S	OLF: 0.005; TWA: 10	A, B, D, E, H, I	0.7 – 12.0
	MME	OLF: 0.0005;TWA: 0.5	A, D, E	0.19 – 0.46
	DMS	OLF: 0.001;TWA: ---	A, D, J	0.07 – 0.35
	NH ₃	OLF:5;TWA: 25	A	29
	CO ₂	OLF:74000;TWA:5000	J	1100
	Acids	OLF:0.0002;TWA:---	A, D, F	P – 0.028
	T	OLF:---;TWA: ---	D, E, H, I	0.60
Sludge loading: above container (E)	MME	OLF: 0.0005;TWA: 0.5	C, F, G	0.07 – 0.18
	DMS	OLF: 0.001;TWA: ---	B, F	0.05 – 0.08
	DMDS	OLF: 0.0003;TWA: ---	B	0.16
	Acids	OLF:0.0002;TWA:---	A, B, C	0.002 – 0.009
Sludge pile or land filling: ambient air (E)	T	OLF:---;TWA: ---	A, C	0.001
	Acids	OLF:0.0002;TWA:---	A, B, C, D, G	0.001 – 0.073
Sludge conveyor: above sludge (I)	H ₂ S	OLF: 0.005; TWA: 10	B, F, H	0.3 – 11.6
	MME	OLF: 0.0005;TWA: 0.5	H	1.14
	DMS	OLF: 0.001;TWA: ---	F	0.08
Sludge tank : inside closed tank (I)	H ₂ S	OLF: 0.005; TWA: 10	B, I	18.0 – 182.0
	MME	OLF: 0.0005;TWA: 0.5	B, D, I	0.20 – 0.75
	DMS	OLF: 0.001;TWA: ---	B, I	0.08 – 0.18
	DMDS	OLF: 0.0003;TWA: ---	B, I	0.10
Sumps/drains/sinks : above	H ₂ S	OLF: 0.005; TWA: 10	C, D	2.0 – 64.0
	MME	OLF: 0.0005;TWA: 0.5	C, D	2.40 – 39.00
	DMS	OLF: 0.001;TWA: ---	C, D	20.80 – 23.90

	DMDS	OLF: 0.0003;TWA: ---	C, D	3.56 – 21.40
Sample collection:	H ₂ S	OLF: 0.005; TWA: 10	A, B, C, E, H	2.2 – 27.0
during sampling	MME	OLF: 0.0005;TWA: 0.5	A, C	1.50 – 4.90
	DMS	OLF: 0.001;TWA: ---	A, C	0.10 – 0.68
	DMDS	OLF: 0.0003;TWA: ---	C	0.50

^A (I) = interior sampling site

(E) = exterior sampling site

^B H₂S = hydrogen sulfide

MME = methyl mercaptan

DMS = dimethyl sulfide

DMDS = dimethyl disulfide

NH₃ = ammonia

CO = carbon monoxide

CO₂ = carbon dioxide

NO = nitric oxide

SO₂ = sulfur dioxide

MOH = methanol

T = terpenes

P = presence (olfactory detection)

Acids = organic acids (acetic, butyric, formic, propionic, valeric)

^C OLF = olfactory⁽¹⁵⁻¹⁷⁾; TWA = 8h-threshold limit value⁽¹⁸⁾

^D Only results for plants where compounds were present in measurable or olfactory detectable concentrations are presented.

Table II. Concentrations of chemical compounds measured at emission sources.

At all the stations where the chemical contaminants measured were significant, one or more sulfur compounds were present, with hydrogen sulfide predominating, followed by methyl mercaptan and dimethyl sulfides. The results show that the type of paper pulp, specifically kraft pulp, was a determining factor for their presence in the atmospheric emissions.

High concentrations of other compounds were occasionally measured. These were: carbon monoxide, measured at three screens and three draining tables; sulfur dioxide, measured at one screen in a sulfite pulp mill; and ammonia, measured at several stations in a kraft pulp mill.

Two classes of substances were detected by odor in several plants, although present in low concentrations. They were terpenes which are components of softwoods, and organic acids, found in sludge environment. These two classes of compounds have very low odor thresholds, and organic acids are known for their very disagreeable odor.

No chlorinated compound was detected and the dust concentrations were all below 1 mg/m³.

The results suggest that the type of secondary treatment, and the combined treatment of sanitary wastewater and mill effluents have no impact on the type of emissions. It should be noted that the proportion of sanitary wastewater was below 10%.

Bioaerosols

Table III presents the concentration ranges measured for bioaerosols at emission sources as well as the limits recommended in the literature for a 8-hour exposure. Only results for plants in which bioaerosols were present in significant concentrations (concentrations greater than half of the recommended levels in the case of bacteria and endotoxins, or statistically greater (using the Student “t” test) than the concentrations measured upwind in the case of molds) are presented.⁽¹⁹⁻²²⁾

²²⁾ A comparison of the concentrations at the measuring sites to those upwind identifies whether there is a site of proliferation or an emission source.⁽¹⁹⁾

Step : sampling site ^A	Bioaerosols ^B	Emission source ^C	
		Plant	Measured concentration
Screening: above waste container (I)	Total bacteria (CFU/m ³)	B, H, J	6,980 – 8,300
	Gram negative bacteria (CFU/m ³)	E	680
	Total molds (CFU/m ³)	E, F, J	2,040-6,840
	Endotoxins (EU/m ³)	H	250
1 ST Clarification: dispersion plume (E)	Endotoxins (EU/m ³)	E, G	900 - > 5,000
Cooling: dispersion plume (E)	Total bacteria (CFU/m ³)	B	80,800
	Gram negative bacteria (CFU/m ³)	B, H	750 – 4,670
Aeration: dispersion plume (E)	Total bacteria (CFU/m ³)	A, J	5,640 – 14,980
2 ND Clarification: dispersion plume (E)	Total bacteria (CFU/m ³)	B	6,680
Draining table : above sludge circulation (I)	Total bacteria (CFU/m ³)	B, G, I, J	5,460 – 22,850
	Total molds (CFU/m ³)	J	4,120
	Endotoxins (EU/m ³)	H, J	2,060 - > 5,000
Sludge press: sludge entrance (I)	Total bacteria (CFU/m ³)	B	13,750
	Endotoxins (EU/m ³)	G, J	180 - 200
Sludge loading: above container (E)	Total bacteria (CFU/m ³)	C	6,090
Sludge pile or land filling: ambient air (E)	Total bacteria (CFU/m ³)	A	46,900
Recommended level ⁽¹⁹⁻²²⁾	Total molds (CFU/m ³)	A, G	9,110 – 11,340
	Total bacteria (CFU/m ³)		10,000
	Gram negative bacteria (CFU/m ³)		1,000
	Total molds (CFU/m ³)		Comparison with upwind
	Endotoxins (EU/m ³)		300

^A (I) = interior sampling site (E) = exterior sampling site

^B CFU = colony forming unit ; EU = endotoxin unit

^C Only the results for plants where bioaerosols were present in significant concentrations are presented. Significant concentrations = concentrations greater than half of the recommended levels in the case of bacteria and endotoxins, or statistically greater than the concentrations measured upwind in the case of molds.

Table III. Concentrations of bioaerosols measured at emission sources.

Of 117 measuring stations, 16 (4 of which were located outdoors) had concentrations greater than the recommended values for at least one of the bioaerosols.

▪ **Emission sources**

Each of the steps in the wastewater treatment process was specifically considered in order to determine whether it was a significant emission source and to identify the different development or operational characteristics responsible for propagation into the air, so that corrective and preventive measures could be proposed.

Screening

Two potential emission sources were identified in the screening step. The first was the effluent pit where turbulent screening action results in the release of chemical compounds into the air. The screens operate intermittently according to a predetermined cycle or the water level. Continuous measurement of hydrogen sulfide concentrations above the effluent pits revealed fluctuations that followed the operating cycles of the screens. Their start-up therefore seems to dissipate the contaminants accumulated at the pit into the ambient air. The installation of a removable cover at the top of the pit and a continuous operation minimize the diffusion of these compounds into the air.

The second emission source was the container receiving the solids removed from the wastewater. The organic material accumulated inside these containers is an excellent site for microorganism proliferation. Regular emptying and cleaning of these containers is a good way to limit microorganism proliferation.

Primary clarification, cooling, aeration and secondary clarification

All these steps are carried out outdoors. The fan outlet from the cooling systems is a source of emission of sulfur compounds for mills using kraft pulp. Cooling towers have the potential to be sources of bacterial emission because of considerable aerosolization, particularly when the blades of the towers promote the growth of algae and slime. It is then recommended that exposure to the dispersion plume be avoided and that personal protection (impermeable) be worn by workers working above these basins or in the plume. The other basins give off very low concentrations of chemical compounds and bioaerosols.

Sludge treatment

The purpose of sludge treatment is to dry the sludge before disposal. Two types of equipment are generally used in this step, namely the draining table and the press. For these two steps, the sludge circulation zone is a source of chemical emissions, while the tank for the removed water may release bioaerosols. In addition, the restarting of this equipment following a breakdown or an unexpected shutdown (causing sludge stagnation) can result in the release of significant amounts of substances into the air. Then preventive measures for workers present at start-up must be developed.

The installation of collection systems above the draining tables and the confinement of sludge presses are effective measures because in the mills where these controls were implemented, the residual concentrations in the ambient air were very low. However, the

collection systems located solely at sludge entry or discharge only slightly reduce the concentrations in the environment.

The configuration of the collection tank for the removed water has an impact on the phenomenon of aerosolization and therefore on the emission of bioaerosols into the air. Raised edges and deeper tanks avoid splashes.

Sludge accumulated on the ground is also a medium suitable for mold proliferation. Immediate cleanup of any spill is recommended.

Sludge disposal

At the outlet from the presses, the sludge is carried by conveyor or by screw conveyor into containers, trucks, silos or is temporarily piled. The sludge is then buried or incinerated.

Sludge transport and loading generates low concentrations of sulfur compounds. However, piling and land filling are steps in which bioaerosols and chemical compounds are emitted, such as very malodorous organic acids. The residence time of the sludge in piles is a factor that promotes the release of these acids because it allows more complete oxidation and produces an increase in the temperature which facilitates volatilization. The microbial content of these piles is also increased and thus constitutes a major site of proliferation. Minimizing the storing time is recommended and the wearing of respiratory protection with a thin layer of carbon is effective for absorbing odors.

Specific steps or locations

Inside the buildings, chemical contaminants were investigated at certain sumps and floor piping, and sink drains or floor drains. In some cases, although the concentrations at entry to the pipe or above the screen were very high, the compounds were not detected in the surrounding ambient air. In other situations, the compounds (sulfur compounds) were dispersed into the ambient air at low concentrations, but at concentrations clearly greater than the odor limits. These locations must be considered as potential sources and preventive measures must be developed for workers doing the repair or the cleaning. Covering the openings in the floor with a rubber mat may be effective in minimizing the emissions and the odors.

Measurements taken with a telescopic sensor in enclosed or confined spaces like tanks and enclosed conveyors, in some cases revealed very high concentrations, namely of hydrogen sulfide and methyl mercaptan. These locations must be indicated as confined spaces and workers must follow the specific rules applicable to them.

Outdoors, the ventilation vents may sometimes release very high concentrations of chemical contaminants into the environment. These emissions become diluted more or less rapidly depending on the climatic conditions, but contribute to the odor level.

Specific tasks

The cleaning of the orifices of the effluent distribution basins for the cooling systems did not generate any chemical compounds or bioaerosols. Sample collection proved to be a source of

emission of chemical contaminants when the sampling was done in the open air. Depending on the concentrations measured, personal protection may be necessary.

The emission of chemical compounds into the air is due to their presence in the raw effluent and sludge, and therefore any action that promotes turbulent contact of air with the effluents and sludge will increase release rates of the compounds into the air. The same is true for bioaerosols, which propagate by fixation onto water droplets or dust particles. Consequently, emissions can be minimized by controlling the operations where air-water contact occurs.

When direct contact with effluent or sludge is possible (eg., during repair or clean-up) the wearing of personal protective equipment is recommended. With bioaerosols, stringent personal hygiene measures remain the best means of prevention.²¹⁻²²

CONCLUSION

The activities involved in the biological treatment of paper mill effluents result in the emission of chemical compounds, namely sulfur compounds and bioaerosols, into the work environment. For some of these compounds, the concentrations exceed the odor thresholds and are responsible for the odors that are the cause of complaints by the exposed subjects. This information will also be useful to conduct exposure measurements and control assessment studies at similar facilities.

Emission of these substances into the air is promoted by stirring, handling and the projection of water and sludge. The sources can be minimized by controlling the operations where air-water contact occurs.

ACKNOWLEDGEMENTS

The authors sincerely want to thank the paper mill managers and workers, the Quebec public occupational health and safety network hygienists and technicians, as well as their IRSST colleagues for their enthusiasm and professionalism.

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Received: February 8, 2000

Accepted: August 16, 2000

This paper was accepted for abstracting and publication in the February 2001 issue of *TAPPI JOURNAL*.

TAPPI Website: www.tappi.org