STUDY OF THE CHLORINE AND CHLORINE DIOXIDE ATMOSPHERIC DISPERSION FROM A BLEACHED PULP MILL

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ABSTRACT

The present work intends to synthesize in a methodological way questions related to atmospheric pollution, originated by chlorine dioxide emissions from bleached pulp mill. The objective of this study is to evaluate pollutant concentrations in relation to air quality in regions around the mill. A survey of 19 potential emission sources of these gases was realized. Four sources are placed within the chlorine dioxide production plant, 9 at the bleaching plant, and 6 in the cellulose drying stages. In this study, the mathematical model of pollutant dispersion named ATMOS version 4.1 has been used. This model developed by the Industrial Source Complex – ISC and classified as the best by the Environmental Protection Agency – EPA, uses the Gaussian Method of Plume Dispersion for calculating the pollutant concentration. The methodological framework proposed in the present study has been divided into three steps: Step 1 — Identification of the potentially emission sources of chlorine compounds; Step 2 — Collection and analysis of pollutant concentration data as well as their respective atmospheric emission rates; and Step 3 — Generation of modeled scenarios of air quality. The case study was developed in Celulose Nipo-Brasileira S/A – CENIBRA and the studied area was a 10 km x 10 km square (subdivided into 40,000 square cells with edges of 50 m each) with the industrial unit in its center. The air quality model obtained has been shown as a pollutant concentration net with the maximum concentrations. This were observed as average values during 5 minutes, 1 hour and eight hours, overlapped on a satellite image of the region under study, taking into account the decay effects. These effects were less conservative and the accomplishment of the model without any decay effect was more conservative. Due to the nonexistence of a air quality permit for these pollutants, the results obtained by means of the dispersion mathematical model were compared with the existing permits of occupational health presenting values, lower than the previously established limits. In conclusion, this work proposed a limit to chlorine emission, measured as (Cl) emitted to the atmosphere per ton of cellulose produced in an hour.

Key-Words: model, dispersion, atmosphere, pollution, chlorine, chlorine dioxide, mill, cellulose.

INTRODUCTION

The investigation of the dispersion and transport of pollutants on atmosphere is an fundamental activity on the protection of the air quality. In the last decades, the study of the transport and dispersion of pollutant in the atmosphere became a subject quite studied. The importance of this subject became evident with the increase of the pollutant emissions caused by the urban and industrial areas growth.

Studies have been trying to show, with more and more significant results, morbidity and mortality effects associated to the atmospheric pollutant. However, the difficulty is in the establishment of the cause and effect relation among the exhibition to the several pollutant and the respectives environmental and human health response.

Founded in 1973, CELULOSE NIPO-BRASILEIRA S.A. - CENIBRA, located in the Industrial District of Perpétuo Socorro, municipal district of Belo Oriente - MG, has a built area of 1,207,000 m² and it produces 960,000 tons of elementary chlorine free cellulose annually. In its production process, are used as bleaching agents of the cellulose fiber, oxygen (O_2), hydrogen peroxide (H_2O_2) and chlorine dioxide (ClO_2), being this last one responsible for the chlorine emission on the atmosphere.

Due to the demand of the pulp and paper international market on purchasing cellulose of better quality and free from elementary chlorine, CENIBRA has implemented the eucalyptus cellulose production without using elementary chlorine - Cl_2, substituting it for chlorine dioxide- ClO_2. Because of the large demand of this reagent in the new process, the implantation of a new chemical plant of ClO_2 production was necessary.

The environmental legislation about atmospheric pollution has stablished gradually emissions limits more and more restrictive along the last years. The authorization for installation and operation of an industry demands activities of forecast and control of the pollutants emission and dispersion in atmosphere.

In view of that, the State Environmental Agengy (Fundação Estadual do Meio Ambiente – FEAM) requested, as part of CENIBRA operational license process, the atmospheric emissions evaluation of the new chlorine dioxide production plant and, if necessary, the presentation of adaptation project with a execution schedule for concentrations reduction.

The chemical dissociation of chlorine and their oxides, as of many other atmospehric processes, is controlled by the energy associated to sunlight. According to VOGT et al. (1986) ATSDR (2002), chlorine dioxide gas is unstable and it can be decomposed quickly to chlorine and oxygen in moderate temperatures, even when exposed to sunlight.

Due to the instability of ClO_2 in the atmosphere and the very short dissociation time, the dispersion mathematical modelling was done by using the elementary chlorine - Cl_2 as the only pollutant, including the total concentration of equivalent chlorine - TCC. This model of dispersion is based on the Model ISC (Industrial Source Complex) - classified by EPA as preferential.

OBJECTIVE

This research aimed to characterize the atmospheric emissions from the chlorine dioxide production plants and from the cellulose bleaching and drying plants, determining their potential impacts in the air quality of the delimited area, by mathematical modelling, in two situations: (1) analyzing the chlorine concentrations considering the effect of chemical removal (decay) and, (2) without the decay effect.

MATERIALS AND METHODS

Stage 1 – Sources Identification

The identification of the potential atmospheric emission sources of chlorinated compounds was done by considering the following stages of the production process: chlorine dioxide production plants and cellulose bleaching and drying processes. Informations about each source such as place, identification number and geographical coordinates were collected.

Stage 2 - Data Collection and Analysis

The methodology used to evaluate the atmospheric emissions of the stationary sources followed the iodometrics methods for the elementary chlorine and for the chlorine dioxide described by FISHER et al. (1987) and the described methods in APHA/AWWA/WPCF (1980) 4500 - Cl and 4500 - ClO2, with the aid of the norms of ABNT.

The sampling system was set up in agreement with the illustration below, in which the vacuum pump was adjusted to operate with a flow of (1.0 + 0.1) L/min during approximately 60 min.

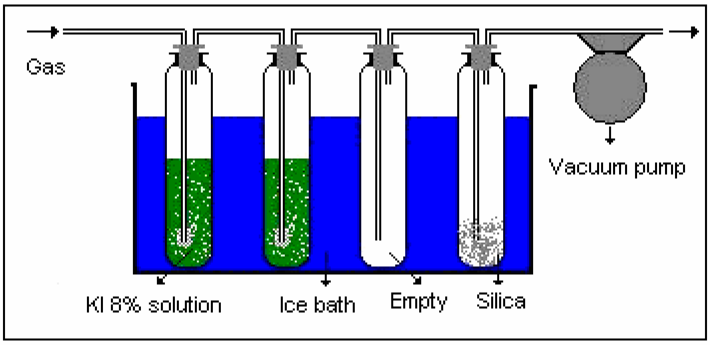

Illustration 1 - Chlorine and chlorine dioxide sampling system in pipes and chimneys

The analysis consisted in reacting slowly 100 ml of sampled solution with sodium tiossulfate 0.01N, using starch as indicator, until the turning point. The wasted volume of sodium tiossulfate 0.01N in the reaction was labeled 'A'. After that, 5 ml of H_2SO_4 20% was added to the erlenmeyer and reacted again with sodium tiossulfate 0.01 N standard, until blue color disappearance. The worn-out final volume was labeled as 'B'.

The mathematical equations used in this study are presented below (the acronyms and symbols list used is in annex).

- **Determination of Chlorine Dioxide Concentration in Normal Conditions (Dry Basis, mg/Nm^3)**

$$C_{ClO_2} = \frac{{}^{5}\!/_{4}\left(B - A\right) \times N_{tio} \times \frac{PM_{ClO_2}}{N^{o}\,Eq} \times V_{total}}{V_{amostra} \times V_{gn}}$$

Eq. 1

- **Determionation of Chlorine Concentration in Normal Conditions (Dry Basis, mg/Nm^3)**

$$C_{Cl_2} = \frac{\frac{\left(5A - B\right)}{4} \times N_{tio} \times \frac{PM_{Cl_2}}{N^{o}\,Eq} \times V_{total}}{V_{amostra} \times V_{gn}}$$

Eq. 2

- **Determination of Total Equivalent Chlorine Concentration in Normal Conditions (Dry Basis, mg/Nm^3)**

$$T_{cc} = \left(C_{Cl2} + C_{ClO2}\right) \times \frac{PM_{Cl2}}{PM_{Cl2} + PM_{ClO2}}$$

Eq. 3

- **Determination of Measured Gas Volume in Normal Conditions (Dry Basis, Nm^3/h)**

$$V_{gn} = \frac{Y \times 0{,}359 \times V_g \times P_g}{T_g + 273}$$

Eq. 4

- **Calculation for Gas Molecular Mass (Wet Basis, g/gmol)**

$$MM_u = \left[MM_s \times \left(1 - P_{wa}\right)\right] + \left(18 \times P_{wa}\right)$$

Eq 5

- **Determination of Atmospheric Emission Average Speed in Conditions of the Chimney**

$$v = K_1 \times C_{pitot} \times \left(\sqrt{\Delta P}\right) \times \sqrt[m]{\frac{T}{P \times MM_u}}$$

Eq. 6

- **Calculation of Gas Volumetric Flow in Normal Conditions (Dry basis, Nm^3/h)**

$$Q_s = \frac{3.600 \times \left(1 - P_{wa}\right) \times v \times A_s \times T_p \times P}{T \times P_p}$$

Eq. 7

Stage 3 - Sceneries Generation

In this study, pollutant emission sources and meteorological informations were introduced in a mathematical model named ATMOS, version 4.1. This model is based in ISC (Industrial Source Complex) and classified by EPA as preferential. Among it main characteristics, the following stages can be outstanding:

- application of the Gauss model in steady state;
- acceptance of all sources types: point, area, line and volume;
- evaluation of short and long period;
- elimination of the pulverization effects ("fumigation");
- use of exponential decay effect for the pollutant that suffer chemical transformations.

The calculation of the pollutant concentration can be obtained by the following equation:

$$C_{(x,y,z)} = \frac{F_c D_{ec}}{2\pi} \times \frac{Q_e}{u_s \sigma_y \sigma_z} \times \exp\left[\frac{-y^2}{2\sigma_y^2}\right] \times \left\{ \exp\left[\frac{-(z-H)^2}{2\sigma_z^2}\right] + \exp\left[\frac{-(z+H)^2}{2\sigma_z^2}\right] \right\} \qquad \text{Eq. 8}$$

The studied area was delimited as being a square of 10 x 10 km, subdivided in square cells of 50 m of side, compounding a total of 40.000 cells, with the industrial unit in the center.

The meteorological informations, used in the present study, were obtained in CENIBRA Meteorological Station, where the following parameters were measured every 5 minutes for the period among 03/10/2001 to 19/11/2003:

- wind direction and speed;
- air temperature;
- precipitation;
- solar radiation;
- standard deviation of wind direction.

RESULTS

Stage 1 – Sources Identification

Nineteen stacionary sources were identified as follow:

- Four sources in the chemical plants of chlorine dioxide production;
- Nine sources in the cellulose bleaching process;
- And six in the cellulose drying process, after pulp washing.

Stage 2 - Data Collection and Analysis

The chemical analyses of the samples of nineteen sources revealed that only three sources are responsible for the release of chlorine and chlorine dioxide for atmosphere: Dioxide Plant I chimney, Dioxide Plant II chimney and chlorate storage tank vents. The concentrations of chlorine and chlorine dioxide obtained for each source are presented in the following Tables.

Table 1 - Dioxide Plant # 1 Concentrations

Parameter	Unit	Sample 1	Sample 2	Sample 3
Y	Not dimensional	1	1	1
V_g	m^3	0.03	0.03	0.03
P_g	mmHg	740	740	740
T_g	^oC	12	12	12
V_{gn}	Nm^3	0.027964	0.027964	0.027964
A	Ml	7.6	7.4	7.1
B	Ml	39.8	39.9	39.9
N_{tio}	Nº Eq/1000ml	0.0089	0.0089	0.0089
PM_{ClO2}	G	67.5	67.5	67.5
PM_{Cl2}	G	35.5	35.5	35.5
V_{total}	Ml	100	100	100
$V_{amostra}$	Ml	100	100	100
C_{ClO2}	mg/Nm^3	172.94	174.55	176.16
C_{Cl2}	mg/Nm^3	–	–	–

Table 2 – Dioxide Plant # 2 Concentrations

Parameter	Unit	Sample 1	Sample 2	Sample 3
Y	adimensional	1	1	1
V_g	m^3	0.03	0.03	0.03
P_g	mmHg	740	740	740
T_g	^oC	12	12	12
V_{gn}	Nm^3	0.027964	0.027964	0.027964
A	ml	8.1	7.9	7.8
B	ml	39.1	39	38.1
N_{tio}	Nº Eq/1000ml	0.0089	0.0089	0.0089
PM_{ClO2}	g	67.5	67.5	67.5
PM_{Cl2}	g	35.5	35.5	35.5
V_{total}	ml	100	100	100
$V_{amostra}$	ml	100	100	100
C_{ClO2}	mg/Nm^3	166.49	167.03	162.73
C_{Cl2}	mg/Nm^3	3.95	1.41	2.54

Table 3 – Chlorate Storage Tank Concentrations

Parameter	Unit	Sample 1	Sample 2	Sample 3
Y	adimensional	1	1	1
V_g	m^3	0,03	0.03	0.03
P_g	mmHg	740	740	740
T_g	^oC	12	12	12
V_{gn}	Nm^3	0.027964	0.027964	0.027964
A	ml	5.7	5.6	5.7
B	ml	0	0	0
N_{tio}	equ/1000ml	0.0089	0.0089	0.0089
PM_{ClO2}	g	67.5	67.5	67.5
PM_{Cl2}	g	35.5	35.5	35.5
V_{total}	ml	100	100	100
$V_{amostra}$	ml	100	100	100
C_{ClO2}	mg/Nm^3	–	–	–
C_{Cl2}	mg/Nm^3	83.33	81.86	83.33

The results of gas molecular mass, in wet basis, of atmospheric emission average speed and of the gas volumetric flow in normal conditions, in dry basis, for the three emission sources, are disposed in Table 4.

Table 4 - Emission sources results

Parameters	Unit	Dioxide Plant I	Dioxide Plant II	Chlorate Storage
P_{atm}	mmHg	740	740	740
P_e	mmHg	0	0	0
P	mmHg	740	740	740
P_p	mmHg	760	760	760
K_1	Not dimensional	34.97	34.97	34.97
C_{pitot}	Not dimensional	0.8555	0.8555	0.8555
T_p	K	273	273	273
MM_U	g/g mol	27.76	27.76	28.3
MM_S	g/g mol	28.84	28.84	28.84
ΔP	mmHg	9.58	6.60	14.74
T	K	285	285	295
P_{WA}	Not dimensional	0.1	0.1	0.05
A_s	m^2	0.049	0.071	0.018
v	m/s	10.9	9.1	13.63
Q_S	Nm^3/h	1,615	1,935	743

The results of chlorine, chlorine dioxide and total equivalent chlorine concentrations obtained for the three emission sources are disposed in the Table 5.

Table 5 - Average Concentrations Results

Local	C_{ClO2} (mg/Nm^3)	C_{Cl2} (mg/Nm^3)	TCC (mg/Nm^3)
Dioxide Plant I Chimney	173	ND	91
Dioxide Plant II Chimney	165	3	90
Chlorate Storage Vents	ND	81	81

Stage 3 - Sceneries Generation

Modeled sceneries of air quality were generated in the form of pollutants concentrations net with the maximum values. This were observed as average values during 5 minutes, 1 hour and eight hours, overlapped on a satellite image of the region under study, taking into account the decay effects. These effects were less conservative and the accomplishment of the model without any decay effect was more conservative.

Due to the inexistence of air quality permits for these pollutants, the results obtained were compared with the effective permits of occupational health listed in Table 6. Table 7 presents the main results of the modeled sceneries.

Table 6 - Chlorine Concentration Limits

Org. (year)	Concentrations [µg/m^3]	Type	Reference Time
ACGIH (1994)	1,500	Occupational health	8 hours (TWA)
	2,900		15 min (STEL)
OSHA (2003)	1,500	Occupational health	8 hours (TWA)
	3,000		15 min (STEL)
NIOSH (1992)	1,500	Occupational health	8 hours (TWA)
	3,000		15 min (STEL)
BRAZIL (1978)	2,300	Occupational health	8 hours (TWA)
Cal/EPA (1999)	210	Toxicological sharp level of exhibition by inhalation – REL	1 hora

Table 7 - Maximum increments in the pollutant concentrations in the atmosphere

Pollutant	Consideration	Concentration [µg/m³]	Reference Time	Date - Hour	Position (relative to the sources)
Cl_2	Without decay	0.145	5 min	03/15/03 09:17	1 km – NE
	With decay	0.068		12/09/02 10:57	1 km – SE
	Without decay	0.067	1 hour	01/19/03 15:37	1 km – SW
	With decay	0.040		01/28/03 15:27	1 km – SW
	Without decay	0.028	8 hour	01/28/03 17:22	1 km – SW
	With decay	0.018		01/28/03 17:22	1 km – SW
TCC	Without decay	0.838	5 min	08/25/02 17:37	1 km – S
	With decay	0.390		11/21/01 12:42	1 km – NW
	Without decay	0.385	1 hour	01/19/03 15:37	1 km – SW
	With decay	0.234		01/28/03 15:27	1 km – SW
	Without decay	0.164	8 hour	01/28/03 17:22	1 km – SW
	With decay	0.103		01/28/03 17:22	1 km – SW

Analysing the modelling results, it was verified that all the values are lower than 1 µg/m³ and the maximum calculated value was 0.838 µg/m³, with the maximum increment of 5 min, staying lower than the concentration international limits, used for occupational health and for toxicological effects by inhalation, as showed on Table 6.

CONCLUSIONS

Based in the accomplished work, it was concluded that:

(i) In the present study, nineteen chlorine emission sources were appraised. Nevertheless, the chemical analyses of the samples from these sources revealed that only three sources are responsible for the release of chlorine and chlorine dioxide for atmosphere: Dioxide Plants I and II chimneys and chlorate storage tank vents.

(ii) Due to the instability of the chlorine dioxide - ClO_2 in the atmosphere, with a very short dissociation time, sceneries of air quality were generated for the real emitted concentrations of elementary chlorine - Cl_2 and for the total chlorine concentration – TCC (by conversion of chlorine dioxide in elementary chlorine and by adding the result to the emitted real concentration).

(iii) The chlorine and chlorine dioxide concentrations analysis methods were considered simple but appropriate.

(iv) It could be concluded that the chlorine concentration in the air, emitted by CENIBRA pulp mill doesn't constitute in a risk to the human health and to the region around the mill, if the current production conditions will be maintained.

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ANNEX - LIST OF ACRONYMS AND SYMBOLS

$\left(\sqrt{\Delta P}\right)m$	average of the square root of ΔP, in mmH_2O;
σ_y	parameter of distribution of Pasquill-Gifford in the lateral direction to the axis of the plume, in
σ_z	parameter of distribution of Pasquill-Gifford in the vertical direction to the axis of the plume, m;
A	volume of sodium thiossulfate wasted in the $1^{st.}$ reaction, in mL;
A_s	area of transversal section of the chimney, in m^2;
B	volume of sodium thiossulfate wasted in the $2^{nd.}$ reaction, in mL;
C_{Cl2}	concentration of chlorine in the Normal Conditions - dry basis, in mg $N^{-1}m^{-3}$;
C_{ClO2}	concentration of Chlorine Dioxide in the Normal Conditions – dry basis, in mg $N^{-1}m^{-3}$;
C_{pitot}	coefficient of the tube Pitot, not dimensional
Dec	term of decay of the pollutant considered, in s^{-1}.
F_c	correction factor for different time of sampling;
H	Effective height of emission, in m;
K_1	Convertion factor = $34{,}97\,\frac{m}{s}\left[\frac{(g/g\ mol)(mmHg)}{(K)\ (mmHg)}\right]$;
MM_s	molecular mass of the gas – dry basis, in g $gmol^{-1}$;
MM_u	molecular mass of the gas - wet basis, in g $gmol^{-1}$.
N_{tio}	normality of Sodium thiossulfate, in Eq L^{-1};
P	absolute pressure of the gas = $P_{atm} + P_e$, in mmHg;
P_{atm}	atmospheric pressure, in mmHg;
P_e	static pressure of the gas in the chimney, in mmHg;
P_g	absolute pressure in the system, in mmHg (the atmospheric pressure is adopted);
PM_{Cl2}	molecular mass of the chlorine, in g;
PM_{ClO2}	molecular mass of the chlorine dioxide, in g;
P_p	standard absolute pressure, in mmHg.
P_{WA}	proportion, in volume of the steam in atmospheric emission, not dimensional;
Q_e	pollutant emission rate, in mg s^{-1};
Q_s	volumetric flow of the gas in the normal conditions - dry basis, in $Nm^3\ h^{-1}$;
T	gas average absolute temperature in the chimney, in K;

TCC	total equivalent chlorine concentration - dry basis, in mg $N^{-1}m^{-3}$;
T_g	average temperature of the gas in the entrance of the system (critical hole), in K.
T_p	standard absolute temperature, in K;
u_s	wind average speed in the emission height, in m s^{-1};
v	average speed of the atmospheric emission in the conditions of the chimney, in m s^{-1};
$V_{amostra}$	bracket volume, in mL;
V_g	volume of the measured gas in the assay conditions, in m^3;
V_{gn}	volume of the measured gas in the normal conditions - dry basis, in Nm^3;
V_{total}	total volume of the solution which a bracket was removed, in mL;
x,y	coordinated cartesian that identify the analyzed point, in m;
Y	factor of correction of the measurer, not dimensional;
Y	orthogonal distance of the analyzed poit to the axis of the plume, in m;
Z	average altitude of the cell where locates the analyzed point, in m.