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Field Survey of Reduced-Sulfur Emissions from a Modern Finnish Pulp Mill

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Substantial advances in air emission control technology during the last decade have brought about a decrease in reduced-sulfur emissions from pulp mills. Unfortunately, these reduced-sulfur compounds still cause subjective odor problems at very low concentrations. As a result pulp mills, even with efficient odor abatement systems, may produce a foul odor in the surrounding area. On the other hand, it will not be easy to further reduce the malodorous sulfur emissions at modern pulp mills because all the major emission sources have already been eliminated.

This paper presents the results of a field survey made at one of the major Finnish pulp mills. In the study, all the main emission sources were analyzed, and the ambient air concentrations of reduced-sulfur compounds in the area surrounding the mill were measured. Emission rates from the current sources and measured ambient air concentrations are presented.

The effects of the main continuous emission sources on ground level concentrations are estimated by means of a Gaussian model of emission dispersion. In addition, the emission sources are ranked on the basis of the estimated ground level concentrations and the momentary emission rates. Finally, means of reducing the current continuous and occasional emissions of reduced-sulfur are recommended.

INTRODUCTION

During the past decade revised equipment design changes within the production processes, plus advances in the dilute non-condensable gas (NCG) collection and treatment systems have considerably decreased reduced-sulfur emissions from pulp mills [l]. In Finland, the total reduced-sulfur (TRS) emissions of the pulp industry have decreased by more than 70% during the 90s (Figure I), despite an over 10 % increase in pulp production [2]. As a result, approximately two thirds of the Finnish pulp mills already have emissions below the reasonably low TRS level of 0.5 kilogram of sulfur per ton of pulp. The yearly TRS emissions of the Finnish pulp mills during 1996 and 1997 are shown in Figure 2. In spite of the reduction in TRS emissions, the established regulations for emissions and ambient air concentrations of reduced-sulfur compounds are expected to be tightened in the near future.. Therefore further decreases of reduced-sulfur emission are among the major challenges facing the pulp and paper industry during the next decade [3].

In this paper, a short overview is given describing the formation of reduced-sulfur compounds at pulp mills plus the collection and treatment systems currently in use. The results of an extensive field survey made at one of the major pulp mills in Finland are also presented. The various methods used for data acquisition are first examined, followed by the results of a field survey on current continuous and occasional emissions plus the measured ambient air concentrations. A Gaussian model of emission dispersion used for estimating the ground level concentration of the reduced-sulfur compounds is then discussed, and the results of the modeling are briefly presented. Finally, the emission sources are ranked on the basis of the estimated ground level concentrations and the momentary emission rates, and means of reducing emissions are recommended.

FORMATION OF MALODOROUS REDUCED-SULFUR COMPOUNDS AT PULP MILLS

The organic reduced-sulfur compounds, i.e. methyl mercaptan (CH_3SH), dimethyl sulfide (CH_3SCH_3) and dimethyl disulfide ($CH_3S_2CH_3$), are formed through reactions between the methoxyl groups of dissolved lignin and the hydrogen sulfide ions present in the cooking liquor [4]. Methyl mercaptan (MM) is the main organic reduced-sulfur compound formed during the cooking process. Subsequently, both dimethyl sulfide (DMS) and dimethyl disulfide (DMDS) are formed stepwise MM as the intermediate product. DMS is formed in the reaction between ionized mercaptan, i.e. mercaptide ion, and the methoxyl groups of the dissolved lignin [5, 6]. This partial reaction of the mercaptide results in the formation of either MM or DMS depending on the prevailing process conditions.
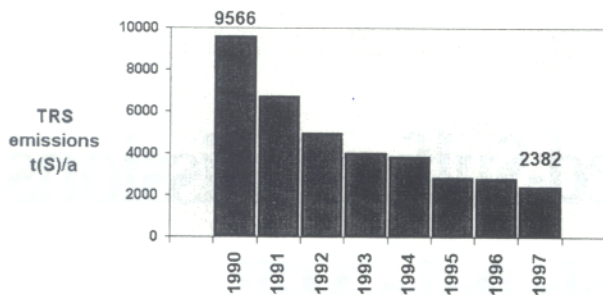
FIGURE 1. TRS emissions of the Finnish pulp industry between 1990 and 1997.

DMDS is not formed directly during the cooking process. However, a significant amount of DMDS may be formed mainly as a result of MM oxidation when the black liquor comes into contact with air in the subsequent processes [7]. Organic reduced-sulfur compounds can also be formed in other parts of the process, e.g. due to the utilization of sulfur-containing foul condensates as makeup and/or spray water. Discharge of the foul condensates or high strength wastewater into the effluent treatment facilities during leaks and/or tank overflows may also cause reduced-sulfur emissions [8].

Inorganic hydrogen sulfide (H_2S) is formed in subsequent processes such as stock washing and black liquor storage when the black liquor pH decreases, although it is not formed during cooking due to the high pH [7]. Consequently, a considerable amount of H_2S is formed during black liquor evaporation when the pH of the black liquor decreases down to 10 or even lower. As the pH decreases the sodium sulfide (Na_2S) present in the black liquor dissociates and is then converted into H_2S according to two-stage hydrolysis reactions.

Hydrogen sulfide is also formed during black liquor pyrolysis in the lower part of the recovery boiler [9]. Normally, almost all of the H_2S formed is oxidized into sulfur dioxide in the upper part of the recovery boiler as a result of the secondary and tertiary air injection. Furthermore, dissolution of the molten smelt, which also contain Na_2S, may produce enormous amounts of H_2S due to reactions with the water vapor from shatter steam jets and the presence of CO_2 [10].

In addition, H_2S is formed in the lime kilns due to the presence of residual Na_2S that has not been sufficiently washed from the lime mud and is therefore fed into the kiln. In the kiln, Na_2S decomposes to form H_2S [11, 12]. In addition H_2S may be formed in the lime kiln if sulfur-containing fuel is used or if NCGs are burned in the kiln.

COLLECTION AND TREATMENT SYSTEMS

At modern pulp mills, virtually all the concentrated and most of the diluted NCGs containing reduced-sulfur compounds are treated by collecting the gases into sealed systems and eliminating odorous compounds by converting them into non-odorous forms [13].

High concentrate and low volume (HCLV) NCGs are formed in the turpentine recovery systems, contin-

uous digester flash steam condensers, foul condensate storage tanks, evaporators and hotwells, as well as in the batch cooking blow heat recovery systems. Typically the HCLV gases contain about 80 to 85% of the total amount of the non-condensible odorous sulfur at the pulp mill, despite the small volume of gas release. Collection systems for the HCLV gases, which may also contain significant amounts of other volatile compounds such as methanol, turpentine and acetone, are typically designed to operate above the upper explosive limit (UEL) by minimizing air entry into the system.

Low concentrate and high volume (LCHV) NCGs are emitted at each stage of the process where the brown stock and the black (or green) liquor come into contact with the air. The main sources of LCHV gases are brown stock washer hoods, seal tanks, knotter hoods, liquor storage tanks, brownstock storage tanks, slakers, lime mud filters, and black liquor oxidizers. The total volume, and especially the concentration of the LCHV gases emitted and which have to be handled varies considerably over time, depending on the production rate, wood species and prevailing process conditions. The collection systems for the LCHV gases are therefore designed to operate well below the lower explosive limit (LEL) by diluting the sources with air.

The main, and most efficient, method for the dilute and, especially for, the concentrated NCGs treatment is thermal oxidation. Traditionally, lime kilns have been the first choice for the thermal oxidation of concentrated NCGs at pulp mills [14]. The main advantage of burning the NCGs in the lime kiln is that most of the SO_2, formed is absorbed on the lime and returned back into the process. On the other hand, burning NCGs in lime kilns is reportedly associated with operational problems [15]. Recovery boilers are the most common site for burning the diluted NCGs. In the recovery boiler the part of the sulfur content is recovered as Na_2S in the smelt and returned back into the process [16]. However, few mills utilize the recovery boiler for concentrated NCGs incineration, due to the potential risk of smelt-water sulfur compounds. A dedicated incinerator eliminates the complications and risks associated with the burning of the NCGs in a process unit such as a lime kiln or a recovery boiler [13].

Moreover, thermal oxidation, absorption with alkali or some other selective absorption media in the specially designed wet scrubber are used for destroying diluted NCGs. Wet scrubbers are also used to reduce sulfur emissions from recovery boilers, lime kilns, tail oil production and dissolving tanks.
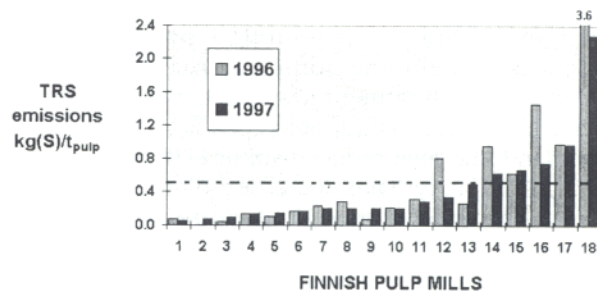

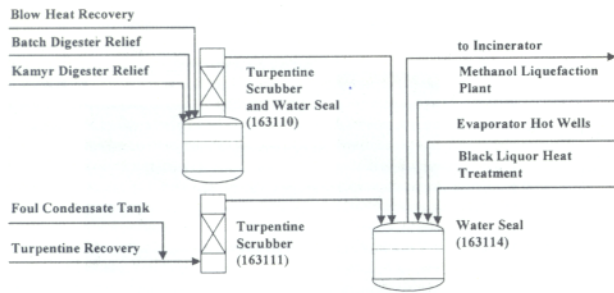**FIGURE 2.** Yearly TRS emissions of the Finnish pulp mills during 1996 and 1997.

FIGURE 3. Collection system for the HCLV NCGs.

THE ODOR ABATEMENT SYSTEMS AT THE WISAFOREST PULP MILL

The Wisaforest pulp mill, which is owned by UPM-Kymmene, is situated in a suburban area near the city of Pietarsaari, in western Finland. The total production capacity of the mill is about 580 000 t/a. Collection and burning of the concentrated NCGs was started at the Wisaforest pulp mill in the mid-70's. Since then, the odor abatement systems have undergone continuous development, intensifying considerably during the 90's.

In 1991, a black liquor heat treatment system was installed in order to increase the dry solids content of the black liquor and to reduce sulfur emissions from the recovery boiler. In the same year, the collection system for the concentrated NCGs was renovated to cover virtually all sources of concentrated NCGs. The new main and reserve incinerator for the concentrated NCGs was installed in 1994. In the incinerator the reduced-sulfur compounds are oxidized to sulfur dioxide, which is washed from the combustion gases by means of a sodium hydroxide scrubber. A methanol column for foul condensates stripper off-gases was installed at the same time. In addition, the systems for collecting the diluted NCGs from cooking and evaporation tank farms were rebuilt. In 1996 the collection system for the diluted NCGs was expanded to cover practically all sources of dilute NCGs. The oxidation capacity was also increased by burning part of the dilute NCGs in the recovery boiler. The collection systems for concentrated and dilute NCGs are illustrated in Figures 3 and 4, respectively.
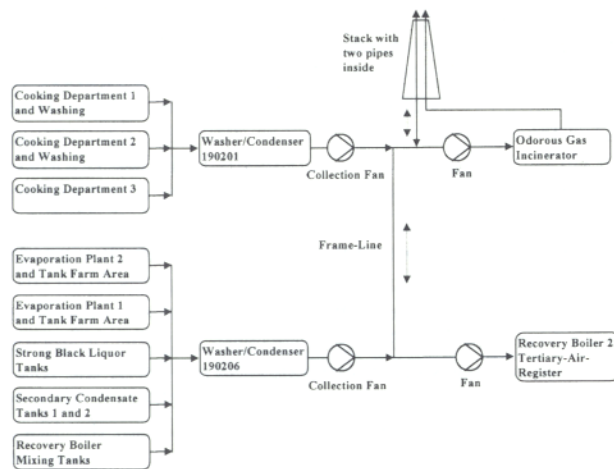

FIGURE 4. Collection System for the LCHV NCGs.

In addition to the lately renovated dilute NCGs collection and treatment system, highly diluted relief gases from the fiber lines are collected and treated together with relief gases from the bleach plant by means of a wet scrubber using weak white liquor. Furthermore, the relief gases from tall oil production are washed in a wet scrubber using weak white liquor, and flue gases from the lime kilns and the recovery boilers are treated by electrostatic precipitators and wet scrubbers.

As a result of the environmental investments made during the 90's, the Wisaforest pulp mill complies well with the environmental regulations concerning odor abatement systems and sulfur emissions (Table 1).

DATA ACQUISITION

The data used in the study were gathered in 1996 and 1997. The semiautomatic data acquisition system, also illustrated in Figure 5, was used to collect all the available information related to the sulfur emissions. The methods used for data acquisition, i.e. an on-line TRS analyzer, manual gas samples analyzed in the laboratory, normal process measurements, movable temperature sensors and emission monitoring stations, are described in more detail below.

The TRS concentration in all the main sources of reduced-sulfur emissions were measured with a portable analyzer system. The system, which includes a diluting sampler, a high temperature converter and two calibrated SO_2 analyzers based on UV fluorescence, is illustrated in Figure 6. The diluted sample stream was first split into two equal flows. One flow was directed through the thermal converter prior to the analyzer in order to convert the reduced-sulfur compounds into SO_2 before the actual measurements, and the other

TABLE 1. The environmental regulations and operational results for 1995 and 1997

|	Limit	1995 1997
Total Sulfur Emissions	<3 kg (SO_2)/t_{pulp}	0.5 kg/t_{pulp} 0.4 kg/t_{pulp}
Dilute NCG Disposal	>90% disposal of all dilute NCG relief gases	80.3% 90.0%
Concentration NCG System	In operation >97% of the time	99.9% 99.1%
Recovery Boilers #1	<10 mg H_2S/Nm_3 over 95% of the time on a monthly basis	96.5% 100%
Recovery Boilers #2	<10 mg H_2S/Nm_3 over 95% of the time on a monthly basis	99.1% 100%
Lime kiln #2	<20 mg H_2S/Nm_3 over 90% of the time on a monthly basis	92.4% 97.1%
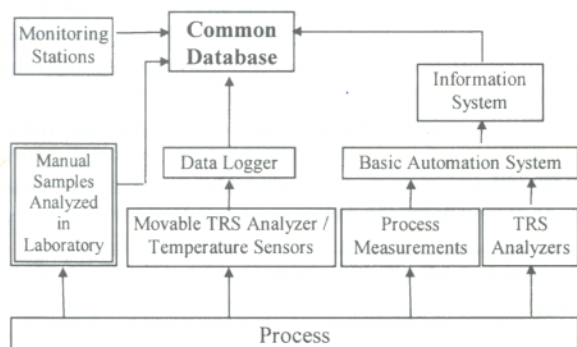
FIGURE 5. Semi-automatic data acquisition system.

FIGURE 7. Reduced-sulfur emissions from the Wisaforest pulp mill during 1996 and 1997.

flow was fed directly into the second analyzer. Both analyzers were connected via a data logger to a PC, which was used to calculate the concentration of reduced-sulfur compounds, based on the difference between the SO_2 concentrations. The result corresponds to the SFS 5727 standard method. The measurements were made on the same wet basis as the initial sample gas thus eliminating the need for additional moisture measurement. It also streamlines the calculation of the mass rate, which was performed on the basis of the measured flow rates. The flow rate measurements were made either by means of a Pitot tube and/or a differential pressure instrument according to SFS 3866. In addition to the on-line measurements made with the portable TRS analyzer system, manual gas samples were taken from a large number of process streams and analyzed in the laboratory. Samples were collected in ethylvinyl acetate-polyamnide bags and then analyzed on-site immediately after collection on a dry basis using a gas chromatography-flame photometric detector (GCFPD). Based on the GC-FPD readings and the corresponding flow rate measurements and also assuming the NTP conditions, the emission rate of each reduced-sulfur component and the total reduced-sulfur emissions were calculated.

Furthermore, normal process measurements such as temperatures, pressures, flow rates and levels related to the emissions or collection and treatment system operation were gathered into the database. Additional temperature sensors (k type wires) were also used to detect short-term emissions. Movable temperature sensors were wired from an 8-channel data logger to sources such as exhaust pipes and pressure / vacuum relief valves from which occasional emissions were expected.
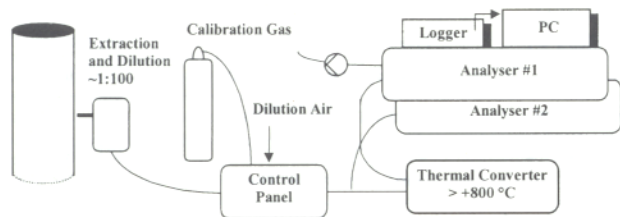

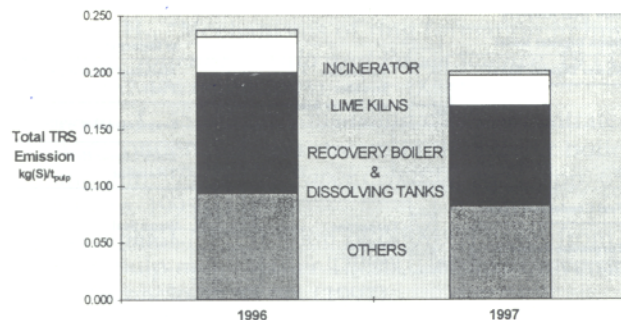**FIGURE 6.** Portable TRS analyzer system.

The ambient air concentration of the reduced-sulfur compounds in the area surrounding the pulp mill was measured by means of two automatic emission-monitoring stations. One measuring station was moved around the surrounding area and the other one was kept at a fixed location about 2.8 km from the pulp mill, which had been selected according to the most frequently occurring wind direction. The emission monitoring stations were equipped with an automatic SO_2 analyzer based on UV fluorescence and meteorological sensors such as temperature, humidity, wind speed and direction. Both stations were connected to a PC-based monitoring system for automatic data acquisition and reporting.

Supplementary information was also obtained from construction drawings and flow charts, and by interviewing the engineers and operators working at the Wisaforest pulp mill.

FIELD SURVEY RESULTS

Continuous emissions

The main sources of the continuous reduced-sulfur emissions at the Wisaforest pulp mill are the vent gases from the smelt dissolving tanks, the flue gases from the lime kilns and the highly diluted noncondensible relief gases collected from the fiber lines. The relief gases from the fiber lines, i.e. from screening, wash filters and the tanks for wash filtrate, are collected but treated only by means of a wet scrubber, thereby resulting in considerable reduced continuous reduced-sulfur emissions. The other minor sources of continuous reduced-sulfur emissions are the flue gases from the recovery boilers, the vent gases from tall oil production and the off-gases from the odorous gas incinerator. A summary of the reduced-sulfur emissions from the Wisaforest pulp mill during the 1996 and 1997 is presented Figure 7. The flue gases from the recovery boilers and the vent gases from dissolving tanks together account for about 45% the flue gases from the lime kilns about 13% and the off-gases from the odorous gas incinerator about 2% of the total reduced-sulfur emissions. While "others", which includes highly diluted NCGs collected from the fiber lines and the emissions from tall oil production as well as an estimate of the non-collected leaks, accounts for about 40% of the total reduced-sulfur emissions.
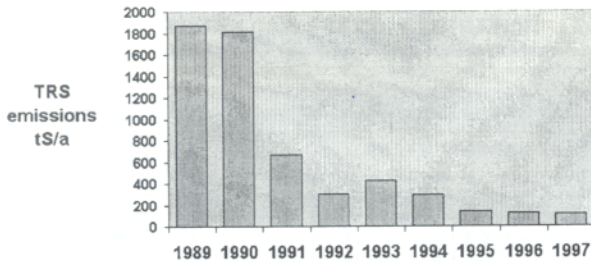
FIGURE 8. Yearly reduced-sulfur emissions of the Wisaforest pulp mill between 1989 and 1997.

The annual reduced-sulfur emissions during 1996 and 1997 were 122 and 112 tonnes of sulfur per year (or 0.24 and 0.20 ton of sulfur per ton of pulp), respectively. Comparison with the statistics from 1990 shows that the reduced-sulfur emissions have decreased during the 90's, (Figure 8), by more than 90%. The major portion of the reduction has been achieved by collecting and incinerating most of the diluted NCGs, which earlier leaked out from different parts of the process. In relation to the other pulp mills in Finland, and also worldwide, the current level of emissions (i.e. about 0.2 $kg(S)/t_{pulp}$) from the mill put it among the mills with low emissions.

Occasional emissions

The operation of the odor-abatement systems covering a large number of the expected occasional emission sources was analyzed on the basis of on-line TRS measurements, manual gas samples analyzed in the laboratory, process history data and temperature readings from the movable sensors. Based on the analysis, the main sources of occasional reduced-sulfur emission at the Wisaforest pulp mill are the chip bin, the turpentine condensers, and the blow heat recovery system in the old batch-cooking department.

The chip bin, depicted in Figure 9, serves as an intermediate storage site before cooking in the digester. At the same time it is also used to remove air from the chips by heating, primarily with flash steam. Occasional reduced-sulfur emissions from the chip bin occurred when the flash steam containing high concentrations of reduced-sulfur compounds is chan-
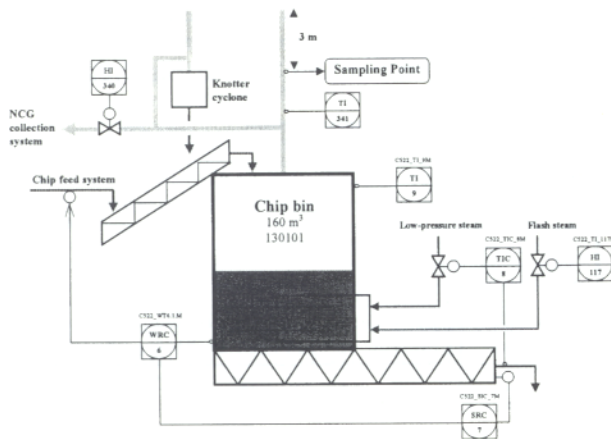

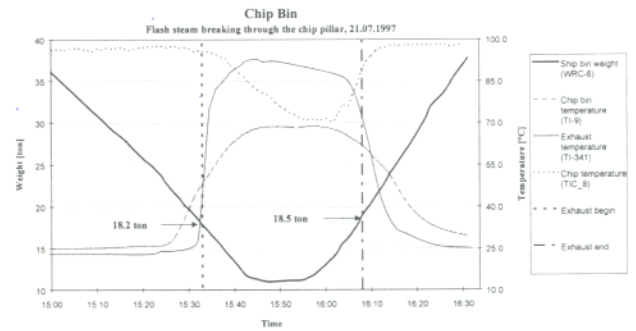**FIGURE 9.** Flow sheet of the chip-streaming process.

FIGURE 10. Flash steam breaking through the chip pillar.

neled through the chip pillar. Figure 10 shows how the flash steam starts to pass through the chip pillar when the chip weight decreases below a certain limit. In the figure incomplete condensation of the flash steam is shown as an increase in both the chip bin temperature (TI-9) and in exhaust temperature (TI-341). Figure 11 shows how the TRS concentration in the chip bin rises when the chip bin weight (WRC-6) decreases. During the experiment shown in Figure 11 illustrates how the collection valve (HI-340) was intentionally closed.

The purpose of turpentine condensation, depicted in Figure 12, is to recover turpentine from the surplus flash steam from the Kamyr digester. Some of the heat content in the gas is simultaneously utilized for the production of hot and warm water. During normal operations, flash steam coming from the 2nd and the 3rd flashing stages are used in the steaming vessel and to heat the chip bin, respectively. Therefore the amount of surplus flash steam to be condensed is relatively low. During shutdowns and start-ups, however, all the flash steam from the Kamyr digester is directed to the turpentine condensers and therefore the risk for reduced-sulfur emission from the turpentine condensers is increased. A shut down situation is shown as an example in Figure 13. After the screw conveyor had been switched off, all the flash steam was directed to the turpentine condenser (HI-117 = 0%). As the flashing rate decreased to under 30 ls, which is normally over 100 l/s, the temperature controller (TIC- 134) was unable to maintain the set point value (65 °C) and the temperature started to oscillate. Finally, the valve controlling the temperature remained closed and, when the flashing rate was sub-
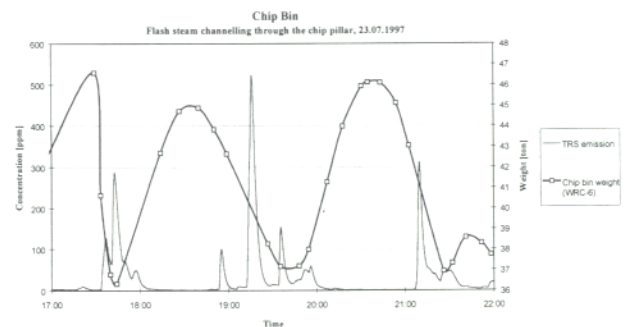

FIGURE 11. Flash steam breaking through the chip pillar.
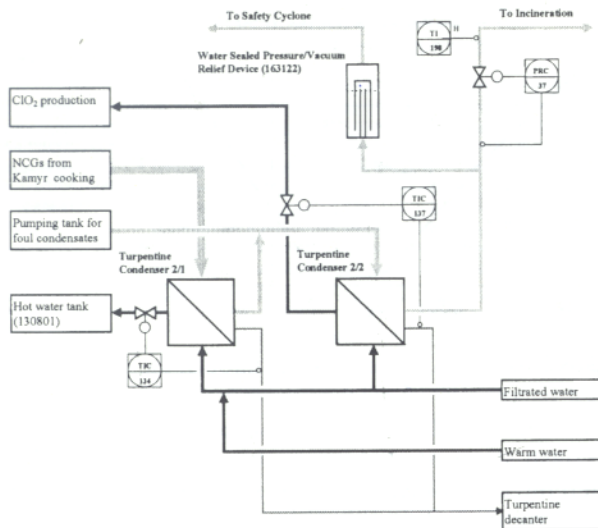
FIGURE 12. Flow diagram of turpentine condensation.

sequently increased to a higher level (around 40 l/s), the valve still remained closed because no turpentine was condensed thus keeping the temperature low as well. As a result, turpentine condenser 2/2 became overloaded, as indicated in the figure by an increase in the temperature (TIC-137). This caused significant reduced-sulfur emissions through the water-sealed pressure/vacuum relief device and the safety cyclone.

Relief gases containing a relative high amount of reduced-sulfur compounds were released from various parts of the blow heat recovery system of the old batch digesters. These short-term emission peaks were mainly a result of the old-fashioned technology used in the system, and the much higher production rate than the blow heat recovery system was originally designed for. Reduced-sulfur emissions from the pressure relief valves (PRV) of the blow tanks were detected during nearly every blow, as shown in Figure 14. The higher temperatures measured at blow-tank 2 than at blow-tank 1 do not indicate higher emission rates, but rather how the sensors were installed relative to the openings of the PRVs. The PRVs of the secondary condenser and the gasholder were also occasionally producing gases containing concentrated reduced-sulfur compounds, as shown in Figure 15.
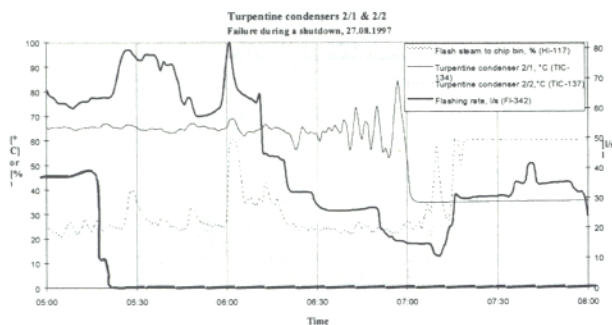
Other minor sources of the occasional reduced-

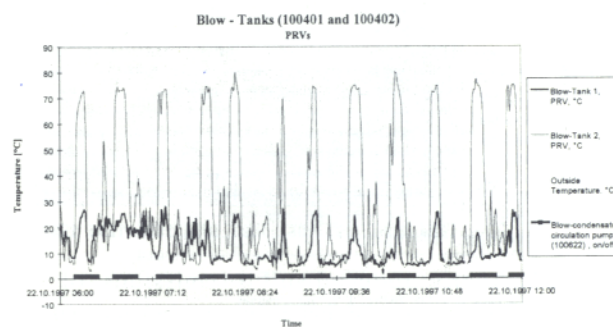**FIGURE 13.** Turpentine condensing failure during a shutdown.

FIGURE 14. Temperature sensors indicating emissions from blow tanks.

sulfur emissions, such as the pulp tower (A-stage), the PRV of the evaporator's hot wells and the thick black liquor tanks were also detected. A more complete description of the experiments, all detected occasional emission sources, the causes of the emissions and suitable means for early detection of the emissions are presented by Mäenpää [17].

Ambient air concentrations

The second highest and maximum daily average values on a monthly basis measured between Jan. 1996 and Dec. 1997 at the monitoring station kept at a fixed location are shown in Figure 16. The mean value of the second highest daily averages was 1.7 $\mu g(S)/Nm^3$ during the monitored period, and the lowest value and the highest values were 0.2 $\mu g(S)/Nm^3$ and 7.0 $\mu g(S)/Nm^3$, respectively. Comparison of the values with the current Finnish recommendations for ambient air concentration of reduced-sulfur compounds (10 $\mu g(S)/Nm^3$ on the second highest daily average on a monthly basis) shows that all the values were well below the recommended level.

In addition to the daily average values, the measured concentrations over a one-year period were calculated as one-hour average values in order to evaluate the frequency of odor occurrence in the surrounding area. The measured concentrations are the sum of all four reduced-sulfur compounds and, furthermore, the actual proportion of each compound out of the total concentration will vary. Therefore, precise determination of odor occurrence is impracticable on the basis of pure concentrations. However, it can be assumed that concentrations below 2.5
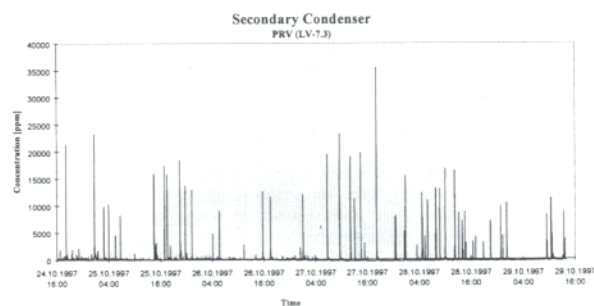

FIGURE 15. PRV of the secondary condenser occasionally releasing odorous gases.

AMBIENT AIR TRS CONCENTRATION
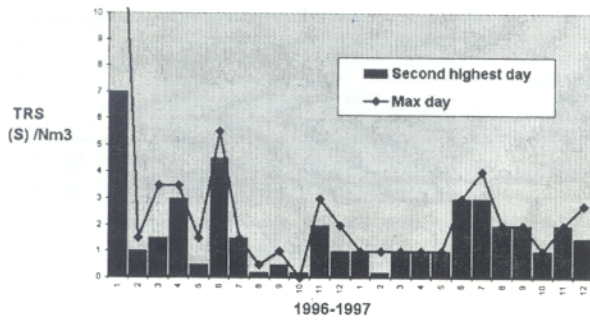MEASURED WITH AUTOMATIC MONITORING STATION

FIGURE 16. The maximum and second highest daily average values of the reduced-sulfur compounds.

$\mu g(S)/Nm^3$ will certainly not cause subjective odor problems. Figure 17 presents the results in a form of a histogram and includes only the data that were measured during the time when the wind was blowing in the direction of the measurement station and the wind speed was over 0.5 m/s. The figure indicates that about 60% of the measured concentrations were below 2.5 $\mu g(S)/Nm^3$ when the raw measurement data were filtered using the wind direction and wind speed.

If all the data, i.e. all the wind directions and wind speeds, are included in the analysis, the proportion of measured concentrations below 2.5 $\mu g(S)/Nm^3$ is much higher, about 95% of the time. The ambient air concentrations in other parts of the surroundings are even lower due to the fact that these values were obtained at the location with the most frequently occurring wind direction.

On the other hand, Figure 17 also indicates that about 5% of the measured concentrations were above 10 $\mu g(S)/Nm^3$ when the wind was blowing in the direction of the measurement station. The values above 10 $\mu g(S)/Nm^3$, which are considerably higher than the normal level and also produce subjective odor problems were most probably caused by the occasional emissions.
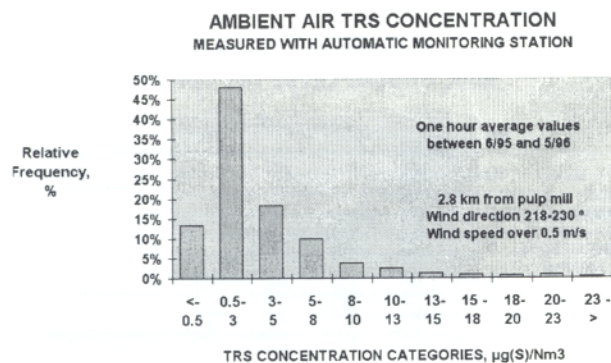
AMBIENT AIR TRS CONCENTRATION
MEASURED WITH AUTOMATIC MONITORING STATION

FIGURE 17. Measured ambient air concentrations, one-hour averages.

EFFECTS OF THE EMISSION SOURCES ON GROUND LEVEL CONCENTRATIONS

In addition to the analysis of the emission sources and the ambient air concentrations, a Gaussian plume model (SCREEN3) was used to study the dispersion of the quantified emissions in the atmosphere [18]. The model is based on the concept of treating each emission source separately as a three-dimensional Gaussian distribution [19,20]. A bell-shaped plume distribution in both the horizontal and vertical directions, uniform emission rates over time, and constant mean wind speed and direction across the area through which the plume is being transported, are assumed in the model. In addition, deposition or chemical reactions are neglected.

The emission source related information, such as the heights of the emission sources, exhaust gas velocities and temperatures, and the source emission rates, are presented in Table 2. The local meteorological parameters utilized in the model were taken from the study carried out earlier in the Pietarsaari area [21]. Based on the results of the study, plume dispersion coefficients for neutral atmospheric stability, which is the most frequently occurring condition in the Pietarsaari area, and three different wind speeds, i.e. low (1.5 m/s), moderate (3.5 m/s) and high (6.0 m/s), were chosen for the model.

Figure 18, which shows the influence of the lime kilns emissions on the ground level concentrations, clearly illustrates the combined effect of wind speed and distance on the ground level concentration resulting from an elevated source. An increase in wind speed distributes the emissions over a large area in the downwind direction, thus reducing the estimated ground level concentration. An increase in the distance from the elevated emission source, e.g. the lime kiln, initially increases the estimated ground level concentration until a specific distance is reached, after which the concentration starts to fall as

TABLE 2. The sources of continuous emissions and information related to emission dispersion

SOURCES	Stack height & diameter		Gas temp. & velocity		TRS Emissions
	M	m	C	m/s	g(S)/s
Recov. Boilers	100	5.4	50	5	0.20
Dissolv. Tanks					1.48
Lime Kilns	84	2.5	50	9	0.72
Odorous Gas Incinerator	83	1.2	70	5	0.01
Tall Oil Scrubber	40	0.4	55	9	0.15
Diluted NCGs[1]	56	1.8	55	11	0.77

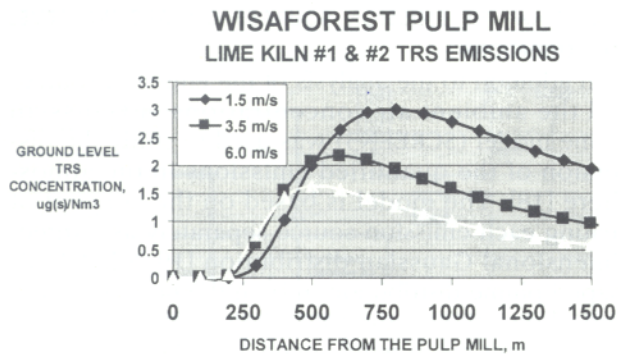[1] *Highly diluted NCG's, which are collected from the fiber lines and treated in the scrubber at the bleaching plant.*

WISAFOREST PULP MILL
LIME KILN #1 & #2 TRS EMISSIONS

FIGURE 18. Effects of the lime kilns on the ground level concentration.

WISAFOREST PULP MILL
GROUND LEVEL TRS CONCENTRATION

FIGURE 19. Concentration profile as a function of the distance from the mill with moderate wind speed.

the distance increases. Figure 18 also shows that the maximum ground level concentration is reached at a distance of about 0.8 km from the lime kilns at low wind speed. An increase in wind speed shifts the maximum ground level impact point closer to the actual emission source, e.g. with a moderate and high wind speed the maxima are reached at a distance of about 0.6 and 0.5 km, respectively.

Figure 19 presents a concentration profile of the ground level concentrations as a function of the distance from the pulp mill with a moderate wind speed (3.5 m/s). The concentration profile, which presents the total ground level concentration and at the same time the proportion of single emission sources out of total emissions at different distances, illustrates the significance of the low level emission sources close to the pulp mill and the increasing importance of the elevated emissions sources with increasing distance from the pulp mill. For instance, the tall oil scrubber (height 40 m) accounts for over 75% of the total ground level concentration, and the lime kilns (height 84 m) have no noticeable effects at the distance of 200 m from the pulp mill. At a distance of 1.5 km the tall oil scrubber accounts for only about 4% and the lime kiln accounts for about 22% of the total ground level concentration.

RANKING OF THE EMISSION SOURCES

In principle, individual emission sources can be ranked on the basis of criteria such as yearly emission rates, and the fulfillment of current or expected environmental regulations. In this case, however, the emission sources were ranked according to the effect of the emission sources on the ground level concentration and the momentary emission rates. These criteria were chosen because the ultimate goal of the study was to find new means of reducing odor occurrence in the surrounding area.

Ranking according to the estimated maximum ground level concentrations of the reduced-sulfur compounds from each individual emission source is shown in Figure 20. As shown in the figure, evaluation of the estimated maximum ground level concentrations emphasizes the importance of lower-level emission sources, such as the vent gases from tall oil production (height 40 m) and highly diluted NCGs (height 56 in), which cause high concentrations near to the actual emission source. How-
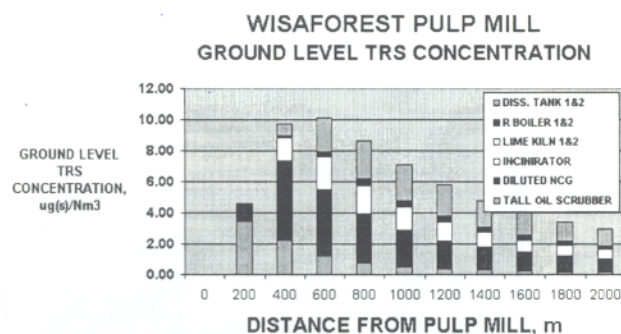
ever, it underrates the importance of elevated-level sources such as the vent gases from the smelt dissolving tanks (height 100 m) and the flue gases from the lime kilns (height 84 in).

Therefore, another way in which the estimated ground level concentrations can be used for ranking the emission sources is presented in Figure 21. The figure shows the share of each emission source out of the total concentration at a distance of 2.5 kilometers from the pulp mill, and moderate wind speed, which is the most frequently occurring wind speed. The emission sources which have a major effect on ground level concentration of the reduced-sulfur compounds in the Pietarsaari city area are highly diluted NCGs, the vent gases from the smelt dissolving tanks and the flue gases from the lime kilns. Together these three sources account for almost 90% of the total concentration.

Ranking based on the momentary, i.e. short-term emission rates (also shown in Figure 22), places emphasis on the occasional emissions. For instance, significant emission peak is generated during failure of the turpentine condenser and almost 9 g(S)/s may be emitted into the air. This alone is more than double the total amount of the continuous emissions. Other noteworthy sources of momentary emissions are the blow heat recovery system of the old batch digesters, the emissions resulting from malfunctioning of the LCHV collection system, and the exhaust gases from the chip bin during flash steam channeling through the chip pillar.
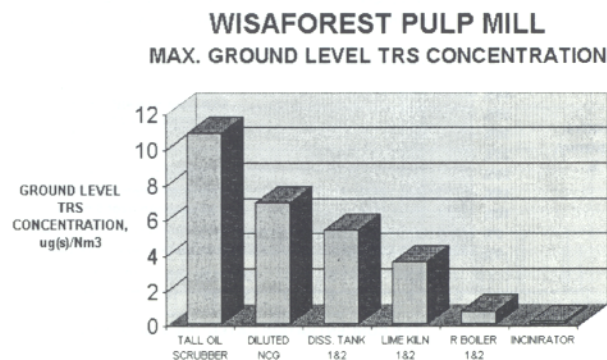
WISAFOREST PULP MILL
MAX. GROUND LEVEL TRS CONCENTRATION

FIGURE 20. Maximum ground level concentration from each emission source.

WISAFOREST PULP MILL
RELATIVE IMPORTANCE OF TRS SOURCES
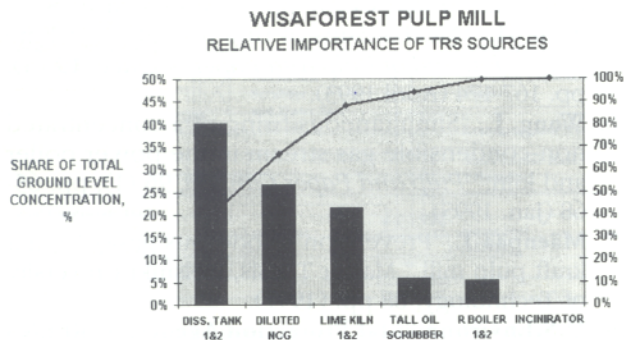
FIGURE 21. Share of each emission source out of the total concentration in the Pietarsaari city area

WISAFOREST PULP MILL
Estimated Momentary TRS Emissions

FIGURE 22. Momentary emissions during periods of higher emission rates.

RECOMMENDATIONS

According to the results of the field survey, the most efficient reduction in the annual reduced-sulfur emissions could be achieved by incinerating the highly diluted NCGs collected from the fiber lines. A marked improvement could also be achieved by connecting the vent gases from tall oil production, which are not as widely dispersed and diluted as the elevated stack emissions, to the dilute NCG collection system. Together these fairly straightforward actions should eliminate over 30% of the total ground level concentration at a distance of 2.5 kin, i.e. the distance to the Pietarsaari city area. Nearer the pulp mill, the effects would be even greater, for instance about 65% reduction at a distance of 0.5 km. To realize the above-mentioned proposals, it will be necessary to construct some new collection lines and provide additional burning capacity. For example, burning in the recovery boiler as secondary air (currently burned as tertiary air) could be one feasible alternative.

The vent gases from the smelt dissolving tanks and the flue gases from the lime kilns together account for about 60% of the ground level concentration of reduced-sulfur compounds in the Pietarsaari city area. The reduced-sulfur emissions from both the lime kilns and the dissolving tanks could be decreased by means of absorption with alkali or some other even more selective absorption media in a wet scrubber. The lime kiln emissions could also be reduced by ensuring both a low residual sulfur content of the lime mud fed into the kiln, and proper combustion conditions in the kiln through improved control of the lime mud filtration and lime kiln process itself.

Even though annual emissions from the occasional sources are low, momentary emissions during process disturbances and different types of failure in the operation of the NCGs collection and treatment system may be very high. Therefore, integration of an enhanced emission monitoring system as part of the process control systems is recommended. Improved monitoring and control using on-line TRS analyzers and other, already available measurements would enhance the performance of the latter systems. There would be a reduction in short-term emissions caused by different types of failure in the collection and treatment system an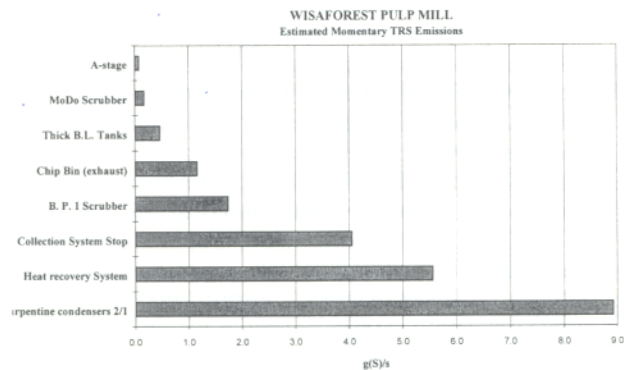d/or process disturbances. Prevention of the short-term emission peaks, released fairly close to the ground level, could have significant influence on the occurrence of odor problems especially in the area near the pulp mill.

CONCLUSIONS

The field survey shows that as a result of the elimination of major emission sources, the other sources such as lime kilns and dissolving tanks that were previously considered to be of minor importance, today account for about half of the overall reduced-sulfur emissions. The field survey also shows that new means, such as prevention of the emissions at their source by means of improved control of the subsequent processes, are required in order to further reduce emissions in the future. The field survey also clearly indicates that occasional emissions deserve special attention, and that improved monitoring and control systems designed to enhance the performance of the installed collection and treatment systems are required in order to eliminate or at least minimize odor occurrence especially in the area near the pulp mill.

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