

The choice of technology to reduce emissions of malodorous gases in sulphate pulping production at existing mills

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Pulp and paper mills are characterized by emissions of malodorous gases that commonly create discomfort for people, also in little concentration in the air. This issue is crucial for enterprises that are not actually using the best available technology for reducing non-condensable gases. Thereby, for existing enterprises, it is very important to realize the proper suitable upgrading by choosing the best solution for their specific working process. In this work, a preliminary review of the scientific literature has been done in order to select the best technology for a case study located in Russian Federation. After the technology choice, the pollutant dispersions in the atmospheric air has been calculated by using a specific software ("Prism" NPP "Logus") based on Method OND-86. Obtained outcomes respect the actual Russian regulation.

Keywords: malodorous gases, gases incineration, NCG burner, atmospheric air pollutants, atmospheric dispersion calculation, permissible concentration.

La scelta della tecnologia per ridurre le emissioni di gas maleodoranti nella produzione di cellulosa con tecnologia al solfato in impianti esistenti. Le fabbriche di cellulosa e carta sono caratterizzate da emissioni di gas maleodoranti che, di solito, causano fastidio alle persone anche se presenti nell'aria a concentrazioni molto basse. Questo aspetto è cruciale per le industrie che al momento non utilizzano le migliori tecnologie disponibili per la riduzione dei gas non condensabili. Pertanto, per le industrie già avviate, è molto importante apportare gli aggiornamenti tecnologici scegliendo la migliore soluzione idonea ai propri specifici processi di lavorazione. In questa ricerca, un'analisi preliminare su letteratura scientifica è stata effettuata al fine di scegliere la migliore tecnologia compatibile per un caso studio reale di una industria della Federazione Russa. Dopo la scelta della tecnologia, la dispersione degli inquinanti in atmosfera è stata calcolata mediante un software specifico ("Prism" NPP "Logus") basato sulla procedura Method OND-86. I risultati ottenuti rispettano le norme russe attualmente in vigore.

Parole chiave: gas maleodoranti, incenerimento dei gas, NCG burner, contaminanti dell'aria, calcolo della dispersione atmosferica, concentrazione ammissibile.

1. Best available techniques for reducing emissions of malodorous gases

The kraft (sulphate) pulping process is characterized by emissions of malodorous reduced sulphur compounds, or total reduced sulphur (TRS), namely methyl mercaptan (MM), dimethyl sulphide (DMS), dimethyl disulphide (DMDS) and hydrogen sulphide (H₂S). MM, DMS and DMDS are

formed in the pulping process and they are the main volatile organic sulphur compounds, while hydrogen sulphide is formed in the downstream processes where the pH of the mill streams is reduced to below 10.

The malodorous gases are generally divided into concentrated non-condensable gases (strong, CNCG) and diluted non-condensable gases (weak, DNCG). The weak gases usually have a sulphur concentration below 0.5 g/Nm³, while the more concentrated or

strong gases are normally characterized by a sulphur concentration higher than 5 g/Nm³.

The emissions of the malodorous gases have environmental and public relations issues for the pulp and paper production industry. Consequently, their reduction has great economic and social advantages for the related enterprises. For this reason is very important to find the best economic, technological and effective solution for the existing mills.

In accordance with Suhr *et al.* (2015), 3 techniques are available for existing mills namely collecting both weak and/or strong malodorous gases and subsequently, incinerate them. The incineration phase can be performed in 3 different equipments:

1. recovery boiler;
2. lime kiln;
3. dedicated burner for non-condensable gases (NCG) with scrubbers for SO₂ removal.

By analyzing this reference document, it can be stated that not all the above-mentioned techniques can be successfully used for existing mills. In the following, the incineration approaches are better explained.

1.1. Incineration in the recovery boiler

The collected malodorous gases are burned in a recovery boiler as secondary air level for strong gases and as secondary or tertiary air for weak gases. This technique is commonly adopted in many mills. In Figures 1 and 2 are shown schematic examples of a treatment system

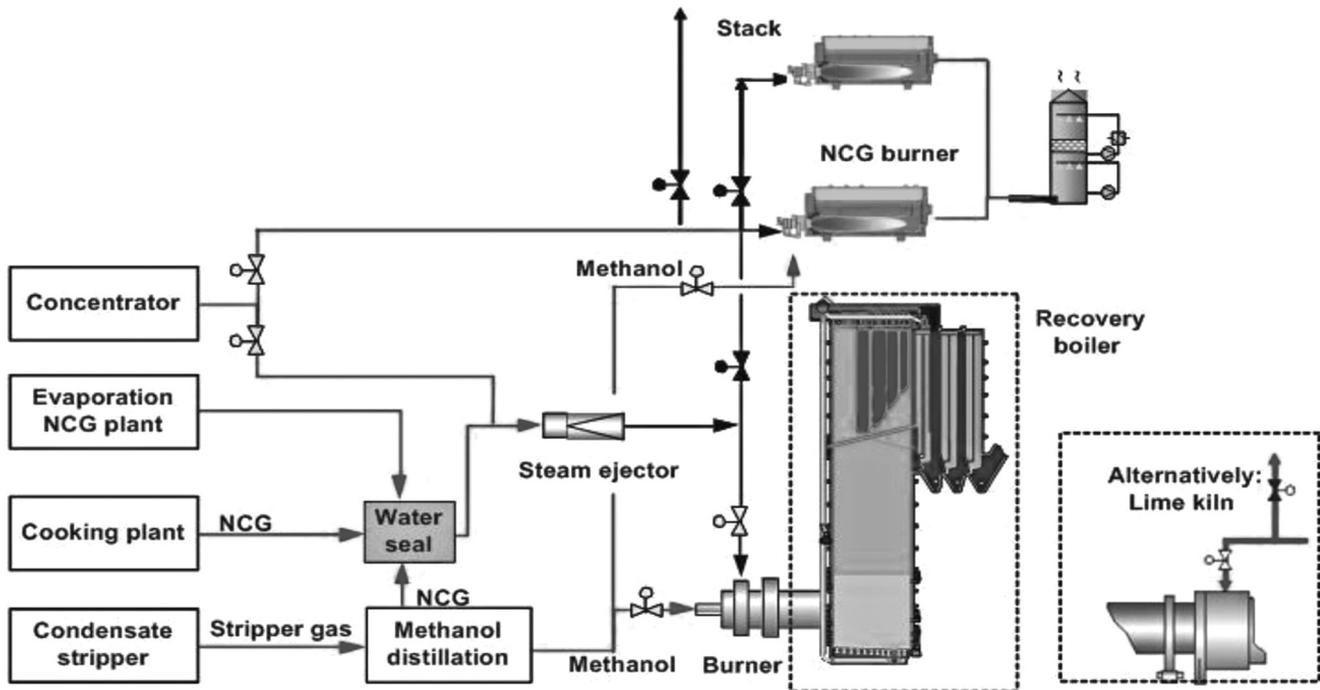


Fig. 1. Example of a CNCG treatment system with the recovery boiler or, alternatively, the lime kiln (Suhr *et al.*, 2015).
 Esempio di un sistema di trattamento CNCG con caldaia di recupero o, in alternativa, con forno a calce (Suhr *et al.*, 2015).

for CNCG and DNCG in the recovery boiler.

The method can be applied in both new and existing mills. However, it is important to highlight the following specific aspects (BAT Bureau, 2015; Gullichsen and Paulapuro, 2008):

1. the gas composition will always change and consequently, the quality and quantitative parameters are not constant, such as volume, concentration, gas pressure;
2. pertaining pulp mills already working to organize the collection and incineration of diluted non-condensable gases sometimes is a hard issue. This is commonly due to the large distances between sources of malodorous gases and the recovery boiler;
3. incineration of the malodorous gases in a recovery boiler is a potential source of additional NO_x emissions, that could be successfully avoided if the malodorous gases are injected in the correct way and in the right location in the furnace of the boiler.

1.2. Incineration in the lime kiln

An alternative equipment for the incineration of malodorous gases is the lime kiln. The collected strong odorous gases are burned in the lime kiln as secondary air level (as in the first case with recovery boiler). Figure 1 shows an example of a system for collecting and treatment CNCG in the lime kiln.

Regarding weak odorous gases, they can be also burned in the lime kiln or, other methods preferably should be used, as a washer or burning in the recovery boiler. Figure 2 shows an example of a system for the collection and treatment of DNCG in the recovery boiler or power boiler (if operated) or in NCG burners.

Nevertheless, numerous mills are nowadays using this technique, particularly in Europe, about 22 enterprises. However, this technique is very disputable and in the modern lime kilns may appear problems in their work due to lack of oxygen (BAT Bureau, 2015; Gullichsen and Paulapuro, 2008).

1.3. Incineration in a dedicated NCG burner equipped with scrubbers for SO_2 removal

In the NCG burner can be utilized both strong and weak gases. After the incineration process, the generated SO_2 can be scrubbed by NaOH absorption (or white liquid) and obtained sodium bisulphite (NaHSO_3) can be used in the bleach plant.

The strong gases are used as fuel to recover heat value for the technological processes. Using this technique helps to reduce over 90% the emissions of malodorous gases. However, NO_x emissions are considerably higher from the NCG burner, if actions for their reduction are not adopted. NCG burner also can be used as a back-up for the incineration of strong gases, an example of this application is shown in Figure 1.

This technique can be approached successfully in new and existing kraft mills (Suhr *et al.*, 2015).

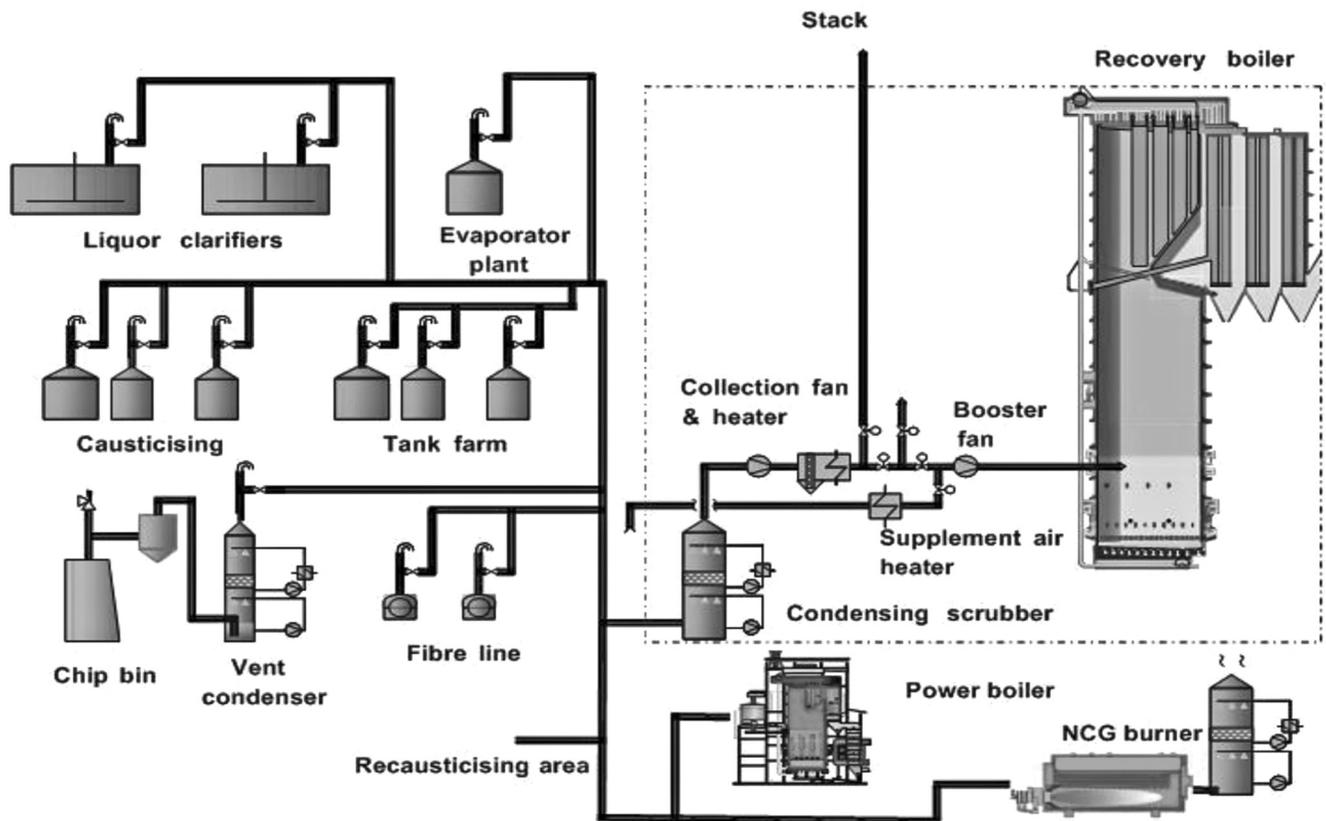


Fig. 2. Example of a DNCG treatment system in the recovery boiler or power boiler (if operated) or in NCG burners (Suhr et al., 2015). Esempio di un sistema di trattamento DNCG nella caldaia di recupero o caldaia semplice (se operativa) o caldaia NCG (Suhr et al., 2015).

2. Dispersion calculation

Emissions from chimney of industrial installations are dispersed in the atmosphere. Nedre *et al.* (2012) highlighted that the dispersion of emission is influenced by:

1. state of the atmosphere;
2. location of enterprise;
3. terrain;
4. physical and chemical properties of emissions;
5. height of the source;
6. diameter of the chimney mouth of the source.

The Government of the Russian Federation has set a list of pollutants that are subject to regulation. For each emission source pollutants and proposed standard emissions must be determined. Proposed standard emissions for the source can be set officially only after performing a dispersion calculation. The dispersion calculation is carried out in order to confirm that the forecast emis-

sions after dispersion in the atmosphere will respect the maximum permissible concentrations on the border of sanitary protection zone (SPZ). The SPZs are recognized in function of the production type. For this research, the localization of the SPZ has been provided from prior studies.

The dispersion calculation has been set in the Methodology for calculating atmospheric concentrations in harmful substances contained in enterprise emissions (Method OND-86). The Method is based on the condition under which the total concentration of each pollutants (C_{Σ} , mg/m^3) shall not exceed the maximum single permissible concentration of this pollutant in atmospheric air:

$$C_{\Sigma} = (C_M + C_B) < C_{MPS}$$

where:

C_M : maximum concentration of pollutant in the surface layer of air created by emission sources, mg/m^3 ;

C_B : background concentration of a pollutant typical for a given area, mg/m^3 ;

C_{MPS} : maximum single permissible concentration, mg/m^3 (GN 2.1.6.3492-17, 2017).

The assessment of impact emissions on the atmospheric air quality of populated areas has been calculated using software package "Prism" (NPP "Logus") based on the Method OND-86.

The program performs a multivariate calculation of concentrations at points on the sanitary protection zone at different wind speeds and directions. Wind speeds are selected automatically according to the program algorithm. The algorithm performs an optimal search of wind speeds (from 0.5 m/s to a dangerous speed) and guarantees the most accurate selection of a dangerous wind speed, taking into account various specific cases. The program automatically determines the maxi-

mum concentration of pollutants and the distance at which they are possible (Nedre *et al.* 2012).

Some pollutants have a combined effect with others. In the GN 2.1.6.3492-17 (2017) pollutants with combined effect are established. In our case, this aspect has been taken into account by the software, calculating the concentration for the following pollutants: Sulfur dioxide (SO₂) + Hydrogen sulphide (H₂S) and Nitrogen dioxide (NO₂) + Sulfur dioxide (SO₂).

For the technique NCG burner equipped with scrubbers for SO₂ removal has been calculated the concentration of the pollutant on the border of sanitary protection zone according to the Method (OND-86).

The table 1 shows the pollutants and their concentration from the chimney after the NCG burner with scrubbers.

For the calculation the following parameters of gases emissions were adopted:

- chimney height: 34 m;
- chimney mouth diameter: 0.42 m;
- gas temperature: 28°C;
- flow velocity: 35.73 m/s;
- gas flow rate: 4.95 m³/s.

The calculation as been performed for a pulp and paper mill in Russian Federation adopting the above defined relative gas emissions parameters, sanitary protection zone and recognized control points on it, background

Tab. 1. Emission source parameters equipped NCG burner with scrubbers for SO₂ removal.

Parametri della sorgente di emissione dotata di caldaia NCG con depuratori per la rimozione di SO₂

Pollutant	Concentration (g/m ³)
Hydrogen sulphide (H ₂ S)	0.005
Dimethyl disulphide (DMDS)	0.005
Dimethyl sulphide (DMS)	0.005
Methyl mercaptan (MM)	0.005
NO _x (expressed in NO ₂)	0.400
Nitrogen dioxide (NO ₂)	0.320
Nitric oxide (NO)	0.052
Sulfur dioxide (SO ₂)	0.120
Carbon monoxide (CO)	0.300

concentration of pollutants and climatic parameters typical for this enterprise and geographical zone. The control points were selected in accordance with specific strategic buildings with a higher required protection level. The calculated pollutant concentrations, taking into account the background values, are presented in Table 2 and in Figure 3. Concerning the outcomes presented in Table 2, values are expressed as the ratio between calculated concentration and maximum single permissible concentrations (MPC). Considering that all outcomes are lower than 1, it means that calculated concentrations on the border of SPZ are lower than

permissible concentration, consequently, they respect the Russian regulation.

Dimethyl disulphide, dimethyl sulphide, nitric oxide, sulfur dioxide and carbon monoxide have not been considered for the dispersion calculation, because their maximum concentration in the air after the dispersion is much lower than the permissible concentration prescribed from the regulation (the ratio is lower than 0.1).

After the calculation, the program provides the dispersion maps for each pollutant. An example is shown in Figure 3, corresponding to H₂S. On the map, the point is the emission source (indicate as "1") while the dotted line identifies the sanitary protection zone. Each circle indicates an area characterized by a certain concentration of pollutant.

The following points are indicated on the map:

- point of maximum concentration MAX = 0.783;
- point of maximum concentration on the SPZ = 0.770;
- control points 1 = 0.768 and 2 = 0.769.

3. Conclusions

From the literature analysis, it can be stated that the incineration of gases in the recovery boiler or lime kiln affect the stable

Tab. 2. Calculated pollutant concentrations taking into account the background pollutant values.

Concentrazioni degli inquinanti calcolate tenendo conto dei valori di fondo.

Pollutant	Maximum concentration into SPZ, share of MPC	Maximum concentration on the SPZ border, share of MPC	Concentration on control point n. 1 on the SPZ, share of MPC	Concentration on control point n. 2 on the SPZ, share of MPC
Hydrogen sulphide (H ₂ S)	0.783	0.770	0.768	0.769
Methyl mercaptan (MM)	0.079	0.060	0.057	0.059
Nitrogen dioxide (NO ₂)	0.523	0.491	0.484	0.488
Sulfur dioxide (SO ₂) + Hydrogen sulphide (H ₂ S)	0.817	0.800	0.796	0.798
Nitrogen dioxide (NO ₂) + Sulfur dioxide (SO ₂)	0.348	0.326	0.321	0.323

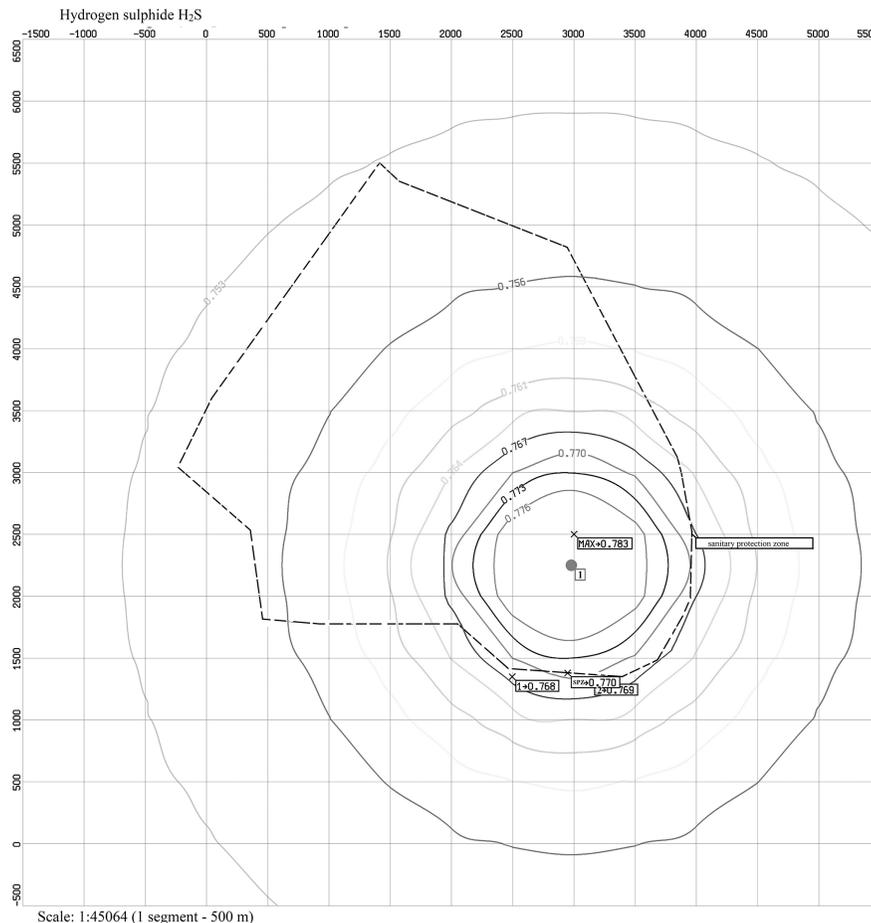


Fig. 3. Example of dispersion map for Hydrogen sulphide (H₂S).
Esempio di mappa di dispersione per acido solfidrico (H₂S).

operating conditions of these equipment.

Having analyzed the techniques proposed in the BAT Reference Document it is possible to conclude that the best solution for existing mills to reduce emissions of malodorous gases is a NCG burner equipped with scrubbers for SO₂. This technique is safe, reliable, available, convenient to use and it ensures an almost complete reduction strong and weak gases.

Considering all the above men-

tioned aspects, the NCG burner equipped with scrubbers for SO₂ has been chosen for the simulation. Thereby, the dispersion was calculated concerning a real case study of a pulp and paper mill.

Outcomes show that concentrations of pollutants from the burner and scrubbers fulfil the limits defined by local regulations, because after the dispersion in the atmospheric air the calculated concentrations on the SPZ are lower than permissible values.

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