GUIDELINES FOR THE MEASUREMENT OF AIR FLOW AND MERCURY IN CELLROOM VENTILATION

Env. Prot. 5

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Euro Chlor

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- improve awareness and understanding of the contribution that chlorine chemistry has made to the thousands of products, which have improved our health, nutrition, standard of living and quality of life;
- maintain open and timely dialogue with regulators, politicians, scientists, the media and other interested stakeholders in the debate on chlorine;
- ensure our industry contributes actively to any public, regulatory or scientific debate and provides balanced and objective science-based information to help answer questions about chlorine and its derivatives;
- promote the best safety, health and environmental practices in the manufacture, handling and use of chlor-alkali products in order to assist our members in achieving continuous improvements (*Responsible Care).*

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This edition of the document has been drawn up by the Environmental Protection Working Group to whom all suggestions concerning possible revision should be addressed through the offices of Euro Chlor.

Summary of the Main Modifications in this version

Section	Nature
Summary	Removed and brought in the introduction
All	Some old stuff was removed
2.2.6 and Annex	Heat balance method reviewed on the base of several sites experience (plus alternative method added)
3.	Reference added to the updated Analytical 6 - Determination of mercury in gasses (and old text removed)

TABLE OF CONTENTS

1. INTRODUCTION

In the frame of the mercury balance that is required to all European producers a least yearly (***Env Prot 12 – Guideline for Making a Mercury Balance in a Chlorine Plant***), this paper presents a summary and evaluation of the present Euro Chlor knowledge on the measurement of mercury emissions in cell room ventilation air.

Both direct and indirect techniques for air flow measurement are available. When practical, direct measurement of air flow using anemometers is preferred.

Techniques for the sampling and analysis of air to evaluate mercury content are also presented (see chapter 3).

Comparisons are made between different types of cell room regarding the number of measurements required and the accuracy that can be expected in the mercury emission results.

The chlorine industry in Europe puts a great deal of effort into minimising the mercury emissions from the amalgam process (***Env Prot 12R – Reduction of Mercury Emissions from West European Chlor Alkali Plants***), and for the last three decades European chlorine producers have been successful in reducing their overall mercury emissions via products, waste water, process exhausts and cell room ventilation.

The mercury losses by cell room ventilation air remain the highest figures in the average European Emission Data of amalgam plants and to be successful, it is necessary to know all sources or means by which mercury can enter the cell room atmosphere. While modelling the cell room ventilation has helped in defining how to measure, continuous monitoring has helped in a quicker detection of leaks and that has led to a reduction of mercury concentration in cell room atmospheres, mainly for closed cell rooms.

However it is more difficult to obtain accurate emission data for cell room ventilation air than for any of the other types of emission.

The basic principle of all techniques is to measure the mercury concentration of the cell room outlet air and simultaneously the air flow rate to obtain the mercury emission rate.

This paper summarises and evaluates the measurement techniques which are known at present.

2. AIR FLOW MEASUREMENT

The volumetric flow rate of air (i.e. m^3/h) leaving the building can only be measured directly for enclosed cell rooms with a limited number of air inlets and outlets.

In open-air cell rooms or those without sidewalls, the air flow pattern is not sufficiently defined to allow accurate measurements. The local atmospheric and climatic conditions can affect both ventilation rates and mercury concentrations.

2.1. Factors Affecting Ventilation Rate

Cell rooms with forced ventilation (by fans) are generally regarded as having a fairly constant air throughput. However the ventilation rate is not totally dependent upon the capacity of the fans, it can be increased dramatically by wind, especially if windows are open. Forced ventilation can also be assisted by natural convection within the building.

Many cell rooms rely totally upon natural convection and some of the important influences on the ventilation rates are:

- geometry of the building in particular the number, type, size and position of air inlet and outlets
- electrical current loading on the cells and resultant heat evolution (effect on internal air temperature)
- general climatic conditions e.g. wind velocity and direction, humidity, outside temperature and atmospheric pressure
- orientation of the building with respect to the prevailing wind and whether the building is free-standing or surrounded by other constructions.

Many of the above variables also apply to cell rooms with forced ventilation.

2.2. Ventilation Rate Measurement Techniques for Enclosed Cell rooms

Direct methods of ventilation rate measurements are available which are based upon the measurement of air velocity. Indirect methods of measurement are mainly based upon the dilution rate of a component blown or injected directly into the cell room atmosphere.

The accuracy of such methods is unlikely to be better than ± 10%.

A more theoretical approach is to calculate the heat balance for the cell room air based on the temperature difference between incoming and exit air.

2.2.1. Hot Wire Anemometer

The measurement principle used in hot wire anemometers is the cooling effect of air flowing past an electrically heated wire. The wire has a known temperature dependent resistance and the resistance change due to cooling corresponds to the flow velocity of air. The wire must be oriented at right angles to the flow direction. The equipment available usually has scales ranging from 0.05 to 15 m/sec.

In order to evaluate the amount of air leaving the ventilation outlets of a cell room building, air flow profiles must be measured, i.e. at each outlet a number of point velocities over the outlet area must be measured. If there are doubts about the direction of air flow, smoke candles could be used. In the case of low flow rates

through large outlet areas, from the top to the bottom of the window or lamellas, a flow reversal can often be observed.

Numerical integration of the measured point velocities gives the average air velocity for the outlet. The average velocity when multiplied by the outlet area gives the volumetric flow rate of air (m^3/h). The total volumetric flow through the building is then the sum of the individual outlet flows.

Hot wire anemometer scales are calibrated in dry air at 20° C. It has been found that air of up to 50 % relative humidity gives a deviation of ± 3 % of the scale range at 20 to 40° C and a flow of 0.1 to 1 m/sec. With higher temperatures and humidity the deviation increases but this seems unimportant for real cell room conditions. Correction charts are supplied with the instruments.

2.2.2. Vane Anemometer

Vane anemometers detect the flow velocity of air directly and usually give a direct reading in m/sec. An advantage of vane anemometers is the fact that the direction of air flow can be seen. Vane anemometers are also subject to the effects of air temperature and humidity.

Again the vanes must be oriented exactly at right angles to the air flow to avoid incorrect measurement.

The measurement procedure and subsequent handling of the data is the same as for hot wire anemometers.

2.2.3. Pitot-tubes, Nozzles, Orifice Plates ...

Pitot-tubes, nozzles or orifice plates are the classical physical equipment which have been used for many decades in the measurement of liquid and gas flow in pipelines. They are all based upon the measurement of pressure drops produced by fluid flow.

At lower velocities these instruments have poorer sensitivity than anemometers and are less practical in their application.

2.2.4. Smoke Method

In cell rooms using natural convection the flow rate of ventilation air can be measured in a more direct way by the smoke method. The technique is based upon the rate of dilution of an artificially produced smoke cloud. Results give the air change rate for the cell room (e.g. number of air changes per hour).

The powder is divided into equal portions and placed on small plates. The plates are distributed evenly through the cell room on the cell covers. Each portion of powder must be ignited at the same instant. A film camera is focused through the smoke onto a black wall and the intensity of the light being dispersed by the smoke cloud is recorded on film.

A calibration test is required using the same film and light. A 20-litre sample of air containing the same concentration of smoke, as used in the cell room test, is diluted by an equivalent volume of clean air. The drop in dispersed light intensity is

photometrically measured on the film. Thus the change in intensity corresponds to one air change.

A comparison is made between the calibration film and the test film. The point on the test film which corresponds to a 50 % drop in dispersed light intensity gives the time in minutes corresponding to one air change in the cell room. It is then a simple calculation to obtain the number of air changes per hour and the total volumetric flow rate is given by the product of the air change rate and the volume of the test area.

Alternative methods of continuous smoke generation now exist based on vegetable oils in aerosol form passed over heater elements. Suitable compact generators are manufactured by Concept Engineering.

2.2.5. Sulphur Hexafluoride Method

Different tracers can be used to determine the air change rate of buildings. Sulphur hexafluoride has been applied in different mercury cell plants to determine the mercury losses.

New scientific evidence has come to light concerning the severe global warming potential of sulphur hexafluoride. Following the December 1997 Kyoto Protocol, Euro Chlor has reviewed the use of this chemical as a tracer gas. The advantages of SF6 are its inertness, harmlessness to health, ease of detection (at ppt levels) and similarity in molecular weight to mercury. The disadvantage of global warming impact is offset by the insignificant quantities used in testing. Overall the conclusion is that, since calibration with SF6 is only required for the initial determination or when a re-evaluation is necessary, the environmental impact is very limited. In any case the method is only recommended in special circumstances (see Section 2.3).

The technique is based upon a physical model of the actual mercury emission sources using the following principles, which can be verified by a preliminary control.

a. When a small known quantity of tracer gas is continuously injected into a cell room atmosphere at or close to a point of known mercury emission the tracer will diffuse and be diluted in a similar manner to the mercury. At steady state conditions (i.e. ventilation rate, mercury concentration and tracer injection rate all constant), the ratio of mercury concentration to tracer concentration in the atmosphere will be the same as the ratio of mercury emission rate to the tracer injection rate.

b. Consider a cell room with a finite number of points from which mercury goes into the atmosphere. Tracer gas is injected continuously into the cell room at each point of known mercury evaporation at constant injection rate. The mercury evaporation rate is regarded to be constant as well. Then at steady state the concentrations of tracer and mercury will be proportional at any point in the cell room.

c. From a) and b) it is possible to calculate the rate of mercury emission to atmosphere from the ratio of tracer concentration to mercury concentration and the rate of tracer injection all being measured:

   The relation can be described by the following formula:

$$\frac{C_{Hg}}{C_{SF_6}} = \frac{M_{Hg}}{M_{SF_6}} \qquad (1)$$

Where C = concentration (g/m^3);
M = mass flow (g/h);
M_{Hg} = mercury emission (g Hg/h);
M_{SF6} = SF_6 injection rate (g SF6/h);

$\frac{C_{Hg}}{C_{SF_6}}$ = the average of all the measured Hg and SF_6 concentration ratios

The mercury emission, given in g Hg/h, is then

$$M_{Hg} = \frac{C_{Hg}}{C_{SF_6}} \times M_{SF_6} \qquad (2)$$

The air flow rate D (m^3/h) of a cell room can also be calculated using M_{SF_6}

$$D = \frac{M_{SF_6}}{C_{SF_6}}$$

where C_{SF6} is the average SF_6 concentration.

d. Sulphur hexafluoride (SF6) was chosen as the tracer gas since it has a molecular weight close to the atomic weight of mercury and also since low concentrations can be easily measured.

Procedure:

Porous lines are installed in the cell room. They consist of tubes with several holes to distribute SF6 near each cell. The tubes are fed with air at 6 m^3/hr at 6 - 8 bar pressure. SF6 is mixed with air by injecting it into an air compressor inlet. The flow of SF_6 from a compressed gas bottle to the compressor is controlled by an electronic mass flow regulator giving a constant accurate flow rate to within ± 1 %.

The flow rate of SF6 is controlled to achieve a concentration of about 1 ppbV (part per billion by volume) near the outlet areas.

In order to obtain a tracer concentration distribution, which is as close as possible to that of mercury, valves, are fitted to the porous lines. The valves are adjusted to obtain the correct SF6 distribution.

SF_6 concentrations in air are measured using portable gas chromatographs fitted with EC-detectors. Mercury concentrations in air are measured using a UV absorption mercury sniffer (real time apparatus).

2.2.6. The Heat Balance Method

There are two heat balance methods which could be used to estimate ventilation air flow rates of a cell room. These are:

a. A full heat balance on the mercury cell process which by difference would give the heat load on the cell room atmosphere.

b. A summation of heat losses to atmosphere in the mercury cell room building and a simpler heat balance on the ventilation air.

A detailed description of the method is given in the Annex.

2.3. Evaluation of Air Flow Measurement Techniques

The deviation of the hot wire anemometer has been found to be ± 10 % overall. This is the most precise among those available.

At lower air velocities the vane anemometer has lower sensitivity than the hot wire instrument and the overall relative error is estimated to be higher.

The SF_6 tracer method, although in use in some plants, requires considerable preparation for first implementation but then appears to be reasonably easy to operate and, where comparisons have been done, accurate. Nonetheless, it is not the preferred option when direct air flow measurement is feasible. It can be used when direct flow measurement is impracticable.

Owing to its complexity, the heat balance method is not recommended. The estimation of the radiation and convective heat losses from all the cell room equipment together with measurement of surface temperatures would be extremely time consuming and would likely be subject to large errors.

2.4. Monitoring of Air Flow

Experimental continuous air flow monitors have been installed in several plants. The monitoring systems are preferably based on anemometers (see Sections 2.2.1 and 2.2.2). The output is usually an analogue signal that can be used for monitoring, control or which could be combined with mercury concentration data to show the total emission to atmosphere.

In order to keep the number of measurement points to a reasonable minimum, some investigative work is required to determine the most representative points for measurement. Due to the symmetrical, parabolic airflow pattern across Robertson ridge ventilators only about 10 measurement points are required to give accurate results.

It must be noted that continuous monitoring equipment is used for closed cell rooms to obtain a real time mercury emission rates, giving also a warning in case of abnormal increase of the emission.

2.5. Ventilation Rate Measurement Techniques for Open-air Cell rooms

Because of the influence of other buildings or equipment, the air pattern in an open air cell room may be variable across the room. Accordingly, it is necessary to take several measurements across the area of the cell room and integrate these. The following general procedure is recommended:

a. Each cell room should have a written detailed procedure, tailored to the specific site

b. The cell room should be divided into a regular grid of, say, no more than 10m x 10m. Measurements of upward air velocity and mercury concentration should be taken at the centre point of each grid square at the same height (in Spain, the breathing height of 1,5 m above the plane of the cells has been chosen). A paper plan of the grid should be kept for reference; safe access to each measuring point must be ensured.

c. Sampling for mercury concentration and measuring air velocity should be carried out at the same time.

d. Equipment and analytical methods should be those specified in 2.2, 3.1 and 3.2.

e. The frequency of measurements should be determined by the operation of the cell room. For example, a plant that operates at a steady rate throughout the year may only require a limited number of air flow measurement days. Plants which cycle load between day and night or have a demand-driven load regime will require much greater frequency of measurement. Factors which need to be taken into consideration in determining the frequency include

   - variation of load in the cell room
   - prevailing winds; constancy or variation in intensity and direction
   - seasonal variation
   - rainfall, etc

It may be necessary to establish a typical emission regime for each variable and average these across a representative period; according to experience, a good practice is to realise 2 measurements per month at each load of the cells room; the average emission calculation will integrate the duration of production at each workload.

For example, the weighted mean velocity V_m (m/s) can be calculated as:

$$V_m = \frac{\overline{V}_1 h_1 + \overline{V}_2 h_2 + ... + \overline{V}_n h_n}{h_1 + h_2 + ... + h_n}$$

where $\overline{V}_1$ = air mean velocity at condition 1

h_1 = number of hours worked at condition 1

etc

Similarly, the weighted mean mercury concentration C_m (µg/m³) is given by:

$$C_m = \frac{\overline{C}_1 h_1 + \overline{C}_2 h_2 + ... + \overline{C}_n h_n}{h_1 + h_2 + ... + h_n}$$

where $\overline{C}_1$ = air mean mercury concentration at condition 1, etc

$\overline{V}_1$ and $\overline{C}_1$ are the arithmetic averages of all the velocities and concentrations measured under condition n in the appropriate time period.

If the cell room area is S (m²), the mercury emission E (µg) is given by:

E = V * C * S

3. MEASUREMENT OF THE MERCURY CONCENTRATION IN THE VENTILATION AIR

In order to determine the total mercury emission by cell room ventilation air the airflow measurement and the air sampling for mercury analysis has to be carried out always simultaneously.

3.1. Analytical Methods

Analytical methods to determine the mercury concentration in air are explained in the Euro Chlor recommendation "***Analytical 6 – Determination of Mercury in Gasses***".

3.2. Physical Detection Methods

UV mercury analysers (fixed or portable) have been used in the chlor-alkali industry for many years; they are based on the absorption of a well defined UV wavelength by metallic mercury.

Since the early eighties the Jerome Instrument Corp., USA has marketed a portable atmospheric mercury analyser based upon mercury absorption on a gold film - the Jerome 411 analyser. The absorbed mercury changes the electrical resistance of the gold film and in this way enables direct detection of the mercury concentration.

3.3. Monitoring of the Mercury Concentration

Up to now only UV photometers have been used for the monitoring mercury concentrations in cell room ventilation air. Their main advantages are the direct indication of mercury concentration and output of a signal which can be combined with a signal from an air flow measurement device to generate a total mercury emission figure.

The direct UV method has a lower precision than the other analytical methods detailed in 3.1 due to the fact that mercury chloride aerosols are not detected by this method.

Nevertheless this simple method is a useful technique in helping to detect mercury leaks in cell rooms. Each deviation of mercury concentration from normal can be alarmed immediately and the results of housekeeping actions can be monitored.

4. RESULTS OF TRIALS CARRIED OUT IN DIFFERENT CELLROOMS

Investigations in five German cell rooms were done using method 2.2.1 for ventilation rate measurement and 3.1 for mercury analysis. The two monitoring techniques were developed specifically for these trials. Each cell room was monitored for 1 year and measurements were repeated on a monthly basis in order to investigate seasonal influences.

Two of the cell rooms were fitted with Robertson ridge ventilators, two with lantern ventilators and one used fans. The current load was between 80 - 290 kA per cell.

Statistical investigation of the results showed that the deviation in ventilation rate measurement was ± 10 %, that of the mercury analysis was ± 12 % and the resultant deviation in overall mercury emission figures was ± 15 %.

The results also showed that maximum ventilation rates occur in summer, during which period the inlets and outlets are fully open or all fans are operating in order to try to control (reduce) the temperature in the cell room. It was found that the ventilation rate was significantly influenced by:

- heat charge or current load
- inlet air temperature
- difference between cell room temperature and inlet air temperature,
- the degree to which ventilation areas were opened.

The most important finding from these results was the fact that emission results can vary dramatically from one measurement to the next. Thus the precision of annual mercury emission figures is heavily dependent upon the frequency of sampling during the year (therefore it is important to have simple and low cost methods).

Due to the fact that climatic conditions and production demands are frequently changing, the minimum sampling frequency, to obtain realistic emission figures, is once per month.

During the trials the effect of reducing the number of sampling and analyses measurements was assessed, i.e. only selected representative samples were used. It was found that by measurement of flow profiles at representative outlet areas the number of air flow measurements could be reduced by about 20 % without introducing too much error into the overall results. However a corresponding reduction in the number of mercury analyses led to a severe increase in the errors in the final results.

It was found that sample number reduction could only be carried out on cell rooms fitted with Robertson ridge ventilators. On cell rooms with lantern ventilators and with fans the mercury concentrations in the outlet air were non-uniform. Therefore, sample numbers could not be reduced without further investigation into concentration profiles or gradients along the ventilator outlet.

5. ANNEX

The heat balance method

6. REFERENCES

- ***Env Prot 12 – Guideline for Making a Mercury Balance in a Chlorine Plant***
- ***Env Prot 12R – Reduction of Mercury Emissions from West European Chlor Alkali Plants***
- ***Analytical 6 – Determination of Mercury in Gasses***

ANNEX: THE HEAT BALANCE METHOD

1. Derivation

The following simple heat balance can be applied to the cell room atmosphere:

$$Q_{out} - Q_{in} = Q_{losses} \quad (1)$$

where Q_{in} = Heat content of incoming air (J s^{-1})

Q_{losses} = Heat lost to atmosphere from cellroom equipment (J s^{-1})

Q_{out} = Heat content of air leaving cellroom (J s^{-1})

Assuming a constant mass flow rate of air through the cell room (m, kg s^{-1}) and also by defining the enthalpies of incoming air (H_{in}, J kg^{-1}) and exiting air (H_{out}, J kg^{-1}), equation (1) can be rewritten:

$$m H_{out} - m H_{in} = Q_{losses} \quad (2)$$

i.e. $$m (H_{out} - H_{in}) = Q_{losses} \quad (3)$$

Therefore, the mass flow of air into the cell room is given by:

$$m = \frac{Q_{losses}}{(H_{out} - H_{in})} \quad (4)$$

The average volumetric flow rate Vav (m3s-1) of air through the cell room can be calculated as follows:

Let V_E = Volumetric flow of air entering cell room ($m^3 s^{-1}$)

$$V_E = \frac{m}{D_{in}} \quad (5)$$

where D_{in} = Density of air entering cell room (kg m^{-3})

Let V_L = Volumetric flow of air leaving cell room ($m^3 s^{-1}$)

$$V_L = \frac{m}{D_{out}} \quad (6)$$

where D_{out} = Density of air leaving the cell room (kg m^{-3})

The average volumetric flowrate of air, V_{av} , (m^3s^{-1}) through the building is given by:

$$V_{av} = \frac{(V_L + V_E)}{2} \qquad (7)$$

For a cell room building of fixed volume F (m^3), the average number of air changes (N) per hour is:

$$N = \frac{V_{av} \times 3600}{F} \qquad (8)$$

1.1. Estimation of Parameters Used in Derivation

a) H_{in} , H_{out}

The enthalpies of incoming and exiting air can be obtained from psychrometric tables. Determination of the values requires measurement of either:

i) wet bulb and dry bulb temperatures using, for example, a sling psychrometer,

or

ii) the temperature and relative humidity of the air streams entering and leaving the building

Both sets of information can be applied to psychrometric tables to obtain the enthalpy of each stream.

b) D_{in} , D_{out}

The density of air at a given relative humidity and temperature can also be obtained from data tables.

c) Q_{losses}

The key factor in this procedure is the estimation of heat losses to the cell room atmosphere. The term Qlosses in equations (1) to (4) is a summation of these losses. It can be expressed as follows:

$$Q_{losses} = Q_{cells} + Q_{decomposers} + Q_{busbars} + Q_{tanks\ \&\ pipes} + etc \qquad (9)$$

The estimation limited itself to the terms shown. However for a more rigorous estimate, additional terms could be added to the right-hand side of the equation. The methods used to estimate each term are given below.

d) Q_{cells} , $Q_{decomposers}$

Heat losses from the cells and decomposers can be calculated using

$$Q = h_{r+c} A (t_1 - t_0) \qquad (10)$$

where: Q = Heat loss ($J\ s^{-1}$)

H_{r+c} = radiation and convection heat transfer coefficient (J m^{-2} s^{-1} °K^{-1})

A = Surface area (m^2)

t_1 = Mean surface temperature (°K)

t_0 = Mean cellroom temperature (°K)

e) $Q_{busbars}$

Heat losses from busbars can be calculated using measured voltage drops at a given current. This estimate must also include voltage drops in cell top busbars to the anodes.

$$Q_{busbars} = \Sigma(V_{drop} \times I_{busbar}) \qquad (11)$$

f) $Q_{tanks\ \&\ pipes}$

The heat losses to atmosphere from individual tanks and pipework in the cell room can be calculated using equation (10). The total losses are then a summation of the individual results.

1.2. Additional Notes

It is understood that this is not a completely rigorous estimation method. Further refinement in the determination of Q_{losses} in equation (1) would only marginally affect the result whilst greatly complicating the calculation procedure.

1.3. Alternative method

An easier way for calculating the total heat losses in cell room could be based on the following:

$$Q_{losses} = E_{tot} - E_{us} + E_{dec} + \Sigma H_{in} - \Sigma H_{out}$$

Where:

E_{tot} = V_{tot} x I = total supplied electric energy

E_{us} = V_{rev} x I = useful electric energy

E_{dec} = heat balance of decomposer

V_{tot} = total cell room voltage

V_{rev} = 3.15 volts

I = electric load

ΣH_{in} = enthalpy of inlet flows (i.e. inlet brine, demin. water …)

ΣH_{out} = enthalpy of outlet flows (i.e. outlet brine, chlorine, hydrogen …)

Enthalpies of outlet flows should be taken before any possible cooling by external mean (i.e. cooling water), otherwise it have to be considered; enthalpy of water content in chlorine and hydrogen must be taken into account.

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