



Expertise on the Environmental Risk Associated with the Co-Incineration of Wastes in the Cement Kiln "Four E" of CBR Usine de Lixhe, Belgium

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1. Introduction

The cement producer CBR Usine de Lixhe has applied for an official permit for the co-incineration of wastes as a secondary fuel during cement production. These wastes which consist of chips of impregnated wood, waste paints and laquers, and waste oils and lubricants are called Résoufuel. Simultaneously, a permit for the use of waste foundry sands as secondary raw material has been applied for.

Apart from Résoufuel, other wastes are utilized by CBR Lixhe. These other wastes include waste tyres (as a secondary fuel) and fly ash (as a secondary raw material).

In the context of the official licensing procedure, an environmental impact assessment (EIA) study has been performed by the Centre Environnement of the Université de Liège:

*Utilisation de combustibles de substitution et valorisation de sables de fonderie
au four à clinker E - Voie sèche de l'usine CBR de Lixhe (March 1996).*

The technical report of this EIA study (Rapport technique) consists of two

volumes with annexes. There exists also a non-technical summary of this technical report (Résumé non technique, 34 pp.).

Okopol has been asked by Stichting Greenpeace Nederland to critically review this EIA study and to elaborate an independent expertise on the question whether any significant environmental risks are associated with the co-incineration of Résosfuel at CBR Lixhe.

2. Critical aspects of cement production in general

2.1 The "inner circuit"

One fundamental feature of cement production in rotary kilns is the counter-current principle: solid material is moving in one direction from the cold side to the hot end of the system while hot gases are moving the opposite way towards the cold end of the system. In a cyclonic pre-heating system as it is installed in Lixhe (like in many other modern cement kilns), the solid material passes through a temperature gradient from less than 300 °C at the entrance of the pre-heating system to more than 800 °C at the point where the material enters the rotary kiln.

Some chemical elements (and also organic substances) will be absorbed to the solid material at 300 °C but will eventually evaporate as the material moves down to hotter zones of the pre-heating system. The evaporated substances are transported back upwards the pre-heater by the hot gases that come from the main burner of the rotary kiln. At lower temperatures, they will condensate again on the surface of new raw material and thus travel down the pre-heating system, until they evaporate again (WINTELER & LOHSE, 1994).


The most prominent element which is kept inside the pre-heating system by this mechanism is thallium (Tl). Continuous addition of traces of thallium which are contained in natural raw materials can thus lead to an enormous enrichment in the "inner circuit" of the pre-heating system. SPRUNG (1987) pointed out that this leads to an increase of thallium emissions over time. Even more volatile than thallium is the element mercury (Hg); depending on operating conditions, Hg will either form its own inner circuit or be almost completely emitted. Other elements like e.g. lead (Pb) may be rather non-volatile in most cases; increased concentrations of chloride, however, will lead to formation of lead chlorides which are significantly more volatile and may thus also form an inner circuit (ibd.).

Depending on their physico-chemical properties, the maximum enrichment levels of different elements will be at different positions along the temperature gradient inside the pre-heating system (JOST, 1996).

In order to interrupt the continuous enrichment of elements in the "inner circuit", SPRUNG (1987) suggested to remove a certain proportion of dust from the pre-heating system and add it directly to the product cement without passing it through the hot rotary kiln. This operational procedure is usually called a "by-pass" and is nowadays regularly installed in many cement kilns in Europe

2.2 Organic emissions

When compared to other large industrial installations, cement kilns emit fairly large amounts of organic substances (Corg.) and carbon monoxide (CO) which are formed from organic matter that has only partially been burnt. This formation of products of incomplete combustion and CO is mainly due to the natural content of organic matter in the raw materials that are fed into the kiln. Their emission is due to the counter-current system in which the hot exhaust gas is moving in a direction contrary to the movement of solid materials.



CO and Corg. emission levels of a cement kiln are between ten and fifty times higher than those of a modern waste incinerator. These organic emissions which are characteristic to the cement process may contain rather toxic constituents such as e.g. benzene and polycyclic aromatic hydrocarbons (PAH).

In specialised plants for waste incineration, the emission levels of organic carbon (Corg.) and CO give relevant information about the destruction efficiency for organic matter. An incinerator for hazardous wastes would immediately be shut down if its Corg. and CO emissions would increase to a level that is typical for cement kilns.

One fundamental problem with co-incineration of wastes in cement kilns is that both Corg. and CO almost completely lose their function as monitoring parameters for the destruction efficiency for toxic organic constituents that may be contained in the waste. It is very difficult to identify any fuel-derived products of incomplete combustion because the level of organic emissions is so high already. In the case of co-incineration of hazardous wastes, organic fragments may leave the stack unnoticed which are far more toxic than the normal organic emissions of the cement kiln (LOHSE, 1996).

3. Re-evaluation of the Environmental Impact Assessment on "four E", CBR Lixhe

The central conclusions of the environmental impact assessment study (EIA) that has been performed by the Centre Environnement of Liège University include the following statements:

For most substances that were detected in the exhaust gas, no variations could be assigned to the use of Résoufuel or foundry sands;

- emissions of heavy metals are very low;
- emissions of toxic products of incomplete combustion (dioxins, furans etc.) are extremely low;
- all emissions are well below any reference limit values which are normally set for this kind of installation;
- the input specifications of the preliminary permit (l'Arrête Ministériel (agrément), 06.12.1994) are very strict and could even be enlarged for certain metals.
- In the following sections, we will re-evaluate the original data from the EIA as a basis for our own interpretation of mass balances and emission levels of the Lixhe kiln E.

3.1 Mass balance data

The central statements of the EIA study of Liège University are mainly based on mass balance investigations that have been performed on the Lixhe cement kiln "four E" between July 29 and August 5, 1993.

In our systematic re-evaluation of these mass balance data, we have made the following observations:

The mass balance investigations of Liège University are full of mathematical and methodological errors. These range from simple calculating errors (confusion of "plus" and "minus") to fundamental balancing errors (element inputs with raw materials into the pre-drying rotary kiln have always been neglected while element outputs of the same kiln are sometimes taken into account in the mass balance and sometimes not). For clarification, the principle of mass-balance calculations and three examples of typical faults shall be explained here:

Principle of mass-balance calculations:

The main material inputs in the mass balance calculations of Annex 2 (Bilans des matières du four) are

- the fuel materials Charbon S, Pneus and Résoufuel
- the raw materials Farine and Tuffeau humide.

The main outputs are Tuffeau sec, exhaust gas (Fumées) and clinker.

The element contents of Tuffeau humide have not been assessed and have therefore always been set to zero. As a compensation for this, the Tuffeau sec element contents which are reported in Annex 2 as outputs have been ignored as outputs in section 4.1.3 Bilans des matières of the EIA main study.

From this procedure (which could be acceptable for a mass balance of the rotary kiln but not for the whole process), the following exceptions have been made that lead to systematical errors:

Example 1: Chromium (in experiment without résoufuel)

In the mass balance of the EIA main study (p. 4.14), outputs with exhaust gas (3,87 g/h) have been subtracted from clinker outputs (8.971,81 g/h) rather than added to give the total outputs of 8.968 grams per hour (as reported on EIA p. 4.14).

Example 2: Thallium (without résoufuel)

In the mass balance of Annex 2, thallium inputs amount to 4.422,47 g/h. Thallium outputs are 24,93 g/h with the exhaust gas and 15,97 g/h with the clinker. The EIA experts decided in this case to add the thallium content of Tuffeau sec (8.962,89 g/h) to the outputs, thus finding a "recovery rate" of 203,5 per cent. However, the thallium content of Tuffeau humide which would have been the counter-balance for Tuffeau sec is still set to zero.

In the mass balance of the EIA main study (p. 4.12), no outputs at all are given, neither the 40,9 g/h (sum of exhaust gas and clinker, equal to a recovery rate of ca. 1%) nor the total output of 9.003,79 g/h (Tuffeau sec included).

Example 3: Nickel (with and without résoufuel)

For the zero measurement, the nickel output with Tuffeau sec (685,94 g/h) is ignored in the mass balance of the EIA main study. The total nickel output in the main study (p. 4.13) was calculated as

2,82 g/h (Fumées) + 4.561,94 g/h (Clinker) = 4.565 g/h (Sorties).

However, for the measurement with résoufuel, the Tuffeau sec outputs (663,37 g/h) are added to the outputs with Fumées (1,36 g/h) and clinker (4.864,7 g/h) in Annex 2 while no nickel outputs at all are reported in the EIA main study.

In summary, equivalent sets of data have illogically been treated in a completely different manner. The corrections for certain elements in certain experiments appear completely arbitrary and have no scientific foundation.

In the EIA, important material and fuel inputs to the Lixhe cement kiln E have not been analysed. This refers mainly to tyres and fly ash. For tyres, literature data only are referred to, while fly ash is neglected altogether.

Literature data on element contents in waste tyres vary widely. E.g. for cadmium in tyres, we found literature values from 1,6 - 8 mg/kg, the EIA

experts chose 8 mg/kg as an estimate. For mercury, literature values that we found are between 0,42 and 1,2 mg/kg, while 0,25 mg/kg was estimated for the EIA mass balance. Lead contents in tyres and / or rubber are reported from 70 - 4.200 mg/kg while the EIA experts estimated 410 mg/kg.

We do not understand why no tyres could be analysed for this EIA, and we do not understand either why no references are given in the EIA as to the literature source where the estimated element concentrations have been published (our references for tyres and rubber: ITU, 1994; FIZ / LAHMEYER, 1993; KIRSCH, 1991).

Heavy metal contents of fly ash can range from almost negligible to very high concentrations. This depends on the industrial process in which the fly ash is generated (e.g. coal fired power plants or municipal waste incineration, MWI), and on the (geogenic) heavy metal concentrations in the input materials to those processes. MWI fly ash is generally carrying more heavy metals than power plant fly ash, but depending on the coal even the latter one can contain high concentrations of e.g. mercury or nickel (FAULSTICH, 1996, TÜV, 1993; MVB, 1990; FIZ/LAHMEYER, 1993).

We do not understand why the fly ash has neither quantitatively nor qualitatively been characterised in the EIA study.

Exhaust air from the clinker cooler was not taken into account in the mass balance although for some elements this emission pathway will be equally important as the oven exhaust gas which was included in the mass balances.

According to p. 4.25 of the EIA study, exhaust air from the clinker cooler is partly used for the pre-drying of Tuffeau humide (100.000 m³/h) and partly emitted (150.000 m³/h) after passage through a gravel filter for dust cleaning. Although no emission concentrations are reported in the EIA, we suspect that this cooling air will contain much higher concentrations of dust than the main source of oven exhaust gas (which is cleaned by electrostatic precipitation). For those elements that will pass the rotary kiln almost quantitatively (e.g. chromium), the emitted air from the clinker cooler will be an equally important pathway as the oven exhaust gas. Consequently, this should have been taken into account in the mass balances for the process.

The EIA experts did not realise that the raw material silos have a storage capacity of approximately one week's raw material input to the kiln. This, in combination with the re-circulation of heavy metal-containing filter dusts, leads to a "memory effect" of the kiln for heavy metals that will presumably last several weeks.

The Lixhe kiln E has a production capacity of 3.500 tonnes per day (EIA, p. 2.3). The raw material is stored and homogenised in two silos with a capacity of 7.000 m³ each. Although we do not know the exact specific weight of the Lixhe raw material, we estimate that this storage capacity will correspond to approximately one week's need for raw material feed to the kiln.

The dusts which are precipitated in the electrofilter are re-circulated into the process at the stage of the pre-dryer, which is before the two storage and homogenisation silos. Due to the inner circuit which is described above (cf. section 2.1), certain elements will accumulate in the storage silos rather than being transported straight along to the output pathways, i.e. exhaust gas (fumées) and clinker.

In spite of this memory effect for toxic metals, the so-called "zero-measurement" without waste incineration was performed only four days after the experiment with extremely polluted Résoufuel waste. Consequently, the relatively high heavy metal outputs with exhaust gas and clinker during the "zero measurement" are a result of the kiln's precedent pollution with Résoufuel, while the full effect of co-incineration of Résoufuel could not be observed during either of the measurements because the kiln was never allowed to attain equilibrium.

The measurement with Résosfuel incineration was performed on the two days of July 29-30, 1993. This two-day measurement is far too short for a mass balance experiment under equilibrium conditions since the internal storage capacity, as pointed out above, is approximately one week.

A Iso, the four day interval between this experiment and the second measurement without résosfuel (performed on August 4-5, 1993) was too short for the Lixhe kiln E to recover from the high dosage of heavy metals that was given in the first experiment.

An accurate "zero measurement" with no wastes, low heavy metal contents and a kiln at equilibrium has never been performed (see Annex 2 for graphic explanation).

Frankly speaking, for the establishment of a mass balance, both experiments are completely meaningless.

Since the EIA experts have failed to realise the time-dependent relationship between inputs and outputs of heavy metals, they were not able to give an adequate interpretation for the apparent "losses" in the mass balances for certain elements. This is most obvious for the toxic element of thallium which was taken into the kiln during both measurements with the raw materials but, as it seems, hardly came out with either the clinker or the exhaust gas. The real explanation for this is that thallium is kept almost completely in an "inner circuit" inside the kilns pre-heating system (cf. section 2.1). Since there is no way out of the system (such as a by-pass), the only mechanism by which such a dramatic enrichment of toxic elements in the pre-heating system will be unloaded is during one of the occasional switch-offs of the electrofilter.

One such filter switch-off has occurred during the experiment without co-incineration of résosfuel (EIA, p. 4.22). As a result, the thallium emissions which had been at 18,8 µg/Nm³ (sec) during the preceding experiment were now found at 56,6 µg/Nm³ (sec). Unfortunately, neither the length of the sampling period, nor the duration of the filter switch-off phase, (nor the overall frequency of such switch-offs) are given with the analytical data. We are therefore not able to estimate the quantities of dust and thallium that were emitted just because of the filter switch-off.

Additionally, the relevant data concerning the thallium inputs to Lixhe kiln E are incomplete: according to Annex 2 of the the EIA, the thallium intake into the Lixhe kiln E with the various fuels is 1,1 - 1,3 g/h (Résosfuel is reported (EIA p. 2.12) to contain between 0,1 and 0,6 mg Tl per kg d.w. although the permit would allow 20 mg/kg). The measured thallium outputs with clinker and exhaust gas are 20 - 40 g/h. Consequently, ca. 95 % of the total thallium intake must have come via the natural raw materials (under the assumption that the kiln E has been close to equilibrium with respect to thallium).

On the basis of the rather poor data presented in the EIA and given the fact that apparently no controlled mechanism exists for the removal of thallium from the inner circuit, it cannot be excluded that several hundred kilograms of thallium (or even more) are emitted each year during the switch-off phases of the electrofilter.

These filter switch-offs can lead to serious environmental and health problems due to thallium poisoning in the surroundings (like it has occurred in the late 1970ies in Lengerich / Germany).

3.2 Absence of a real "zero measurement"

The measurement without Résosfuel ("zero measurement") that should have given a documentation of the kiln's operation without use of wastes as secondary fuel and secondary raw material has been manipulated by feeding

at least two waste materials, fly ash and tyres, into the process during the zero measurement.

Both waste materials have not been analytically characterised in the EIA study. For fly ash, not even the quantity that is brought into the process is mentioned. Depending on its source, fly ash can contain very high contents of heavy metals and also dioxins. Incineration of tyres significantly raises inputs of zinc and lead into a cement kiln (FIZ / LAHMEYER, 1993; ITU, 1994) and also causes between a two-fold and a five-fold increase of dioxin emissions (WINKLER, 1992).

3.3 Emission levels of dust, mercury and dioxins

In the following paragraphs, we will briefly discuss the emission levels of dust, mercury and dioxins of CBR Lixhe kiln E because for these substances we fundamentally disagree with the statements of the EIA experts from Liège University.

Dust

We disagree with the EIA experts in their opinion that the dust emission levels of the Lixhe kiln (around 50 mg/Nm³) are similar to German TA Luft standards.

It is true that the TA Luft emission limit for dust is 50 mg/Nm³. This emission limit, however, must be safely kept during 100 % of all operating days and must not be exceeded in 97,5 % of all half hour average values. In practice this means that the average dust emissions of German cement kilns are somewhere near 15 or 20 mg/Nm³.

On the basis of a proper "mix calculation", German cement kilns that apply for a permit for co-incineration of wastes normally receive a dust emission limit between 35 and 40 mg/Nm³ (dry gas). (Modern cement kilns in Germany are able to emit less than 5 mg/Nm³, that is more a factor of ten below the emissions of CBR Lixhe, kiln E.)

Mercury

In contrast to the opinion of the EIA experts, we find that the mercury emissions that have been observed during co-incineration of Résoufuel are dramatic.

The mercury (Hg) emissions were measured at 36 µg/Nm³ in the experiment with Résoufuel vs. 13,37 µg/Nm³ in the subsequent experiment without Résoufuel (EIA, p. 4.22). However, since the kiln E was not under equilibrium conditions during these experiments, the Hg recovery rate was only 8,2 % during the first experiment but 185 % during the second experiment.

These data can only be interpreted as a temporary retention phenomenon in an inner circuit that has been built up during the first experiment and was unloaded in the second experiment. It has to be suspected that in routine operation the mercury emissions will be much higher than 36 µg/Nm³ once that equilibrium conditions between inputs and outputs are established.

Given the facts that

- Résoufuel dosage during the experiment was only 12 % of total energy demand (vs. 20 % in the applied permit) and
- although Lixhe kiln E was far from equilibrium, the Hg emissions with Résoufuel were already measured at 36 µg/Nm³,

we do not see how the Hg limitations of EU directive 94/67 (0,05 or 0,1 mg/Nm³ for new and old plants, resp.) or TA Luft (0,2 mg/Nm³ for the sum of Hg + Cd + Tl) can be safely kept by CBR Lixhe, kiln E during co-incineration of Résofuel.

Organic emissions and dioxins

Stable operating conditions and a high incineration efficiency cannot be guaranteed during co-incineration of waste. Under these circumstances, emissions of toxic organics such as polyhalogenated dioxins and furans, benzene and polycyclic aromatics cannot be effectively controlled. In contrast to what is stated in the EIA, compliance with the EU directive 94/67 on waste incineration is highly questionable.

The dioxin emissions both during zero measurements (0,018 ng Eq/Nm³) and especially during measurement with 12 % waste (0,065 ng Eq/Nm³) are relatively high when compared to emissions of other cement kilns (WINTELER & LOHSE, 1994). A "mix calculation" of maximum allowable dioxin emissions on the basis of EU directive 94/67 would result in a dioxin emission limit somewhere near 0,05 ng Eq /Nm³ (depending on certain assumptions on analytical detection limits). This means that dioxin emissions are far too high during co-incineration, especially given the fact that the dioxin "zero level" has been artificially raised by co-incineration of tyres while waste equivalent to only 12 % of total energy demand was fed in the second experiment.

Like with the element thallium, it has to be suspected that significant amounts of dioxins are retained inside the "inner circuit" of the pre-heating system. This can lead to highly problematic dioxin emissions during filter switch-offs.

4. Summary and Recommendations

In the environmental impact assessment that has been performed by the Centre Environnemental de Liège University, we have identified a number of severe methodological faults and errors. By re-evaluating the original data as they are presented in the EIA and the annexes to it, we have arrived at the following conclusions that differ largely from the conclusions of the EIA experts:

- Methodological faults are the only reason why the EIA experts found no significant variations of emissions that could be assigned to the co-incineration of Résofuel: since the Lixhe kiln E was not allowed to attain equilibrium during the two experiments which were performed shortly after each other, the emission measurements are not representative for the two operational procedures with and without Résofuel.
- In routine co-incineration of Résofuel, it is to be expected that the emissions of heavy metals and potentially dioxins will be much higher than during cement production without co-incineration of Résofuel. Also, the product clinker and cement will carry much higher loads of heavy metals when Résofuel is used as a secondary fuel.
- The emissions of the critical parameters dust, mercury and dioxins are much higher at the CBR Lixhe kiln E than the present state of technology would allow. Compliance with European and German legislation (EU directive 94/67 and TA Luft) that the EIA experts have referred to as a reference is either not given or at least highly questionable.

Based on these conclusions, we recommend the following:

The CBR Lixhe cement kiln four E should be provided with a by-pass to remove toxic heavy metals (and potentially dioxins) from the inner circuit of the pre-heating system.

The maximum allowed contents of mercury and other heavy metals in raw materials and wastes should preferably be further restricted. The Swiss draft (1996) directive on co-incineration of wastes in cement kilns can be taken as a reference here.

Better dust filters should be installed as soon as possible at the Lixhe kiln E.

Good maintenance of operating conditions will help to minimise emissions of organic carbon, dioxins and also NOx. We recommend to investigate the reasons for the rather high level of dioxin emissions, and to make sure that no dioxin enrichment occurs in the inner circuit of the pre-heating system.

Effective measures for minimisation of accidental emissions, filter switch-offs and cyclone cloggings should be taken.

An improved mass balance experiment will respect the following aspects:
A real "zero measurement" will be performed without any inputs of waste to the kiln.

The heavy metal contents of all input materials and fuels will be well documented and kept at a reasonably low level during zero measurements.

The time constants of the cement kiln and its surrounding aggregates will be properly assessed. Between the beginning of waste input and the beginning of output measurements in clinker and exhaust gas, the cement kiln will be allowed to attain equilibrium in order to obtain meaningful results.

5. References

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6. Annex 1

Tabelle 1: Gegenüberstellung einiger Alsen-Antragswerte, Forderungen der Biab e.V., Grenzwerte und gemessenen Betriebswerte des Ofen 11

Element	with résosfuel (29/30-7-93)		without résosfuel (4/5-8-93)	
	Input (g/h)	Output (g/h)	Input (g/h)	Output (g/h)
Cadmium	2556	1100	1959	1963
Mercury	238	19,50	12,25	22,7
Thallium	3341	- ¹⁾	4422	- ¹⁾
Beryllium	20,8	12,4	6,82	14,1
Arsenic	1376	1192	1348	1126
Cobalt	3768	2812	2724	2754
Nickel	689,1	- ¹⁾	4032	4565
Selenium	106	94	133,3	212,3
Tellurium	3,16	4,08	4,29	3,84
Antimony	96,3	168,3	84,9	791
Lead	16162	2787	4814	3962
Chromium	9298	8100	7643	8975,7 ²⁾
Copper	8992	10085	4183	5630
Manganese	33456	41717	27654	39095
Vanadium	27795	26282	25653	28140
Zinc	46732	61423	35149	65335
Tin	113	445	- ¹⁾	- ¹⁾

¹⁾ No output given in the EIA

²⁾ corrected by Oekopol on the basis of data given in Annex 2 of the EIA. All figures are from the EIA, section 4.1.3 Bilans de matières de l' unite établi sur les éléments minéraux (except where noted).

7. Annex 2: Graphic explanation of the methodological faults in the EIA on Lixhe kiln E.

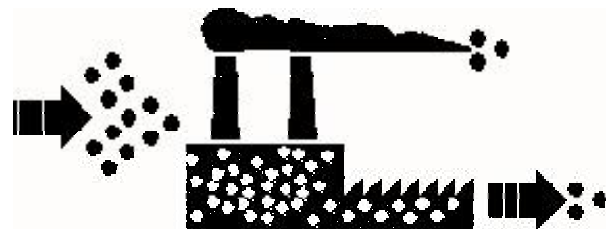


Figure 1: Measurement with Résosfuel (July 29-30, 1993):

A high dosage of heavy metals was fed on the input side of the kiln. Exhaust gas and clinker were sampled before equilibrium conditions were established. A large proportion of heavy metals is therefore stored inside the inner circuit.

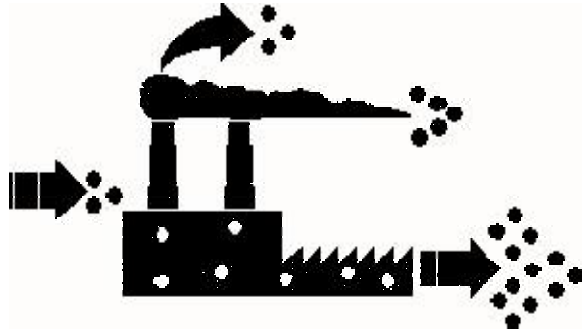


Figure 2: Measurement without Résofuel (August 4-5, 1993):
A lower dosage of heavy metals is fed on the input side. However, elevated heavy metal concentrations that were still stored in the inner circuit are now found in product clinker. Additional emissions occurred due to accidental filter switch-off.

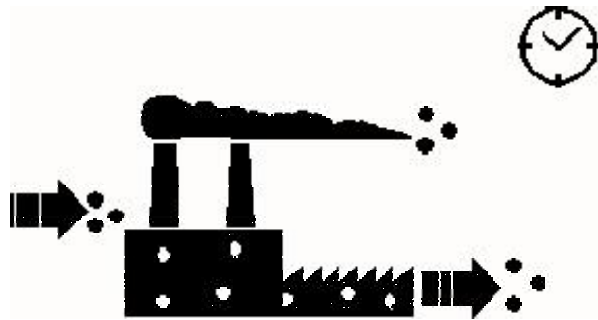


Figure 3: A real "zero measurement" is performed with no waste inputs, low dosage of heavy metals and at equilibrium conditions.

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