

SNAP CODE: **030307**

SOURCE ACTIVITY TITLE: **PROCESSES WITH CONTACT**
Secondary Lead Production

NOSE CODE: **104.12.08**

NFR CODE: **1 A 2 b**

1 ACTIVITIES INCLUDED

This chapter presents information on atmospheric emissions during the production of lead in secondary lead smelters. There are more than 200 secondary lead smelters in the world and a similar number of refineries, producing about 40% of the total lead production (Pacyna, 1989). Various furnaces, including blast, reverberatory and kettle-type ones, are employed in several production processes, such as storage battery production, lead alkyl manufacture, the manufacture of collapsible tubes, ammunition and plumbing equipment, coating of electrical cables and the casting, grinding, and machining of lead alloys, such as brasses and bronzes, in foundries, etc. This chapter describes the methods to estimate emissions of atmospheric pollutants during the secondary lead recovery from products such as battery plates, cable sheathing, type metal, and various slags and drosses.

2 CONTRIBUTIONS TO TOTAL EMISSIONS

The contribution of emissions released from secondary lead production to total emissions in countries of the CORINAIR90 inventory is given as follows:

Table 2-1: Contribution to total emissions of the CORINAIR90 inventory (28 countries)

Source-activity	SNAP-code	Contribution to total emissions [%]							
		SO ₂	NO _x	NMVOC	CH ₄	CO	CO ₂	N ₂ O	NH ₃
Secondary Lead Production	030307	0	-	-	-	-	-	-	-

0 = emissions are reported, but the exact value is below the rounding limit (0.1 per cent)

- = no emissions are reported

There are several trace elements that can be emitted during the secondary lead production. However, these emissions are not very significant on a global scale. Nriagu and Pacyna (1988) concluded that secondary lead production contributes well below 1 % of the total atmospheric emissions of lead, antimony, selenium, and zinc. The estimated contribution of both primary and secondary lead production to European emissions is given in table 2-2.

However, a secondary lead smelter or refinery can be an important emission source on a local scale.

Table 2-2: Contribution to total heavy metal emissions of the OSPARCOM-HELCOM-UN/ECE inventory for 1990 (up to 38 countries)

Source-activity	SNAP-code	Contribution to total emissions [%]							
		As	Cd	Cr	Cu	Hg	Ni	Pb	Zn
Total Lead Production	0403pb	1.0	1.4	-	0.1	0.9	-	1.1	1.1

- = no emissions are reported

Secondary lead production may be a source of polychlorinated dioxins and furans, depending on parameters such as the composition of the raw material (e.g. presence of PVC in battery scrap). European wide emission estimates from this sector are not available.

3 GENERAL

3.1 Description

A secondary lead smelter is defined as any plant or factory in which lead-bearing scrap or lead-bearing materials, other than lead-bearing concentrates (ores) derived from a mining operation, is processed by metallurgical or chemical methods into refined lead, lead alloys or lead oxide. The high proportion of scrap-acid batteries that is re-processed provides feed for the alloy lead market (Barbour et al., 1978).

Secondary lead can be produced using pyrometallurgical or hydrometallurgical processes. Up to now hydrometallurgical processes have only been used at a preliminary stage. The pyrometallurgical processes are subdivided as follows (Rentz et al., 1996):

- battery breaking and processing (scrap preparation),
- smelting of battery scrap materials,
- refining.

In contrast to secondary zinc and copper production, which use a great variety of secondary materials, the recycling of secondary lead materials is concentrated on the processing of scrap batteries, with a world-wide portion of about 80 %. Metal sheets, pipe scraps, sludges, drosses, and dusts play only a minor role as secondary raw materials.

Secondary lead is sometimes combined with primary material for refining. Various pyrometallurgical refining technologies can be applied, depending on the feed material and product specification.

3.2 Definitions

Secondary lead production: - production of lead from materials other than ores.

3.3 Techniques

In general, for the production of secondary lead from battery scrap *two basic process routes* are possible. One route is based on breaking up and dismantling old batteries, and separating the paste, metallics and organics. Melting and reduction is carried out afterwards in different types of furnaces with an additional refining step. The other route is characterised by the direct treatment of complete and undismantled batteries with or without sulphuric acid inside in various smelting furnaces, also with an additional refining step. In detail, in the various stages of pyrometallurgical processing the following technologies are used world-wide (Rentz et al., 1996):

Battery scrap preparation

For battery scrap preparation various processes are possible, which can be differentiated by the degree of separation of single battery components. On an industrial scale, the Penneroya process, the MA process, the Tonolli-CX, and Contibat process are used. Generally heavy metal emissions from battery scrap preparation play a minor role compared to the smelting operation. The Varta and the Bergsoe process are smelting processes carried out without an initial separation, so that the batteries are directly smelted in a furnace.

Smelting

For the industrial production of secondary lead, various kinds of smelting furnaces are employed. The short rotary furnace is the most extensively used furnace for smelting separated battery scrap materials, while long rotary kilns and reverberatory furnaces are only used in a few applications. In contrast to the short rotary kiln, the long rotary kiln is operated continuously. Reverberatory furnaces may also be used for smelting a lead-rich slag, which has been recovered in a primary furnace. Shaft furnaces are typically used for smelting unprepared battery scrap, together with lead cable scrap, furnace slag and filter dusts.

Refining

The lead bullion from secondary lead production contains various impurities, mainly copper, antimony, and tin, which may require elimination or adjustment by refining. Generally the operations necessary for secondary lead refining are limited compared to those necessary for primary lead refining. Pre-decopperising is not necessary and only final decopperising is carried out. In addition, a removal and adjustment of antimony and the elimination of tin may be necessary.

3.4 Emissions

In the secondary lead production process various direct and fugitive heavy metal emission sources are present (Rentz et al., 1996):

From *battery scrap preparation* only small amounts of particulate heavy metals are emitted as direct emissions if single preparation devices are equipped with a special waste gas cleaning facility.

For the *smelting process*, depending on the type of furnace various kinds of fuels are used. Generally short rotary furnaces and long rotary kilns are equipped with natural gas/air burners or sometimes with oxy-fuel burners, while shaft furnaces use coke as fuel. With the generated

waste gas, irrespective of which kind of furnace is used, considerable amounts of heavy metals contained in the dust, as well as certain amounts of gaseous heavy metals are released, depending on the melting temperature and the vapour pressure.

For *refining and alloying*, several kettles are installed depending on the required lead quality. Because of the ongoing reactions in the waste gas from the refining and alloying kettles various amounts of heavy metals in particulate and gaseous form may be emitted.

Fugitive emissions from secondary lead smelting are released with almost all *stockpiling, transferring, charging, and discharging processes*. The amount and composition greatly depends on the process configuration and operation mode. Values concerning the magnitude of unabated and abated emissions have not been revealed. The smelting furnaces are connected with fugitive emissions during the *charging* of raw materials and the *discharging* of slag and lead bullion. Also the *furnace openings* may be an emission source. Fugitive emissions from refining operations arise mainly during *charging, discharging and metal transfer* operations. Refining vessels not covered with primary hoods, may be a further emission source.

As in many plants, direct emission sources are preferably equipped with emission reduction measures, and the fugitive emissions released into ambient air in secondary lead production are generally much higher compared to direct emissions.

By far the most important SO_2 and NO_x emission source during secondary lead production is the operation of the smelting furnaces. The amount of SO_2 formed is mainly determined by the amount of sulphur contained in the raw materials and in the fuel used. Although a major part of the sulphur remains in the slag formed during the smelting process, a considerable share is also converted to SO_2 .

SO_2 concentrations in the off-gas from reverberatory furnaces and blast furnaces are only available on a volume-percentage basis. During tests carried out at a reverberatory furnace using natural gas as a fuel, the concentration of SO_2 in the off-gas was measured at about 0.1 vol.-%. At a blast furnace using coke as fuel an even smaller off-gas concentration in the range of about 0.03 vol.-% was measured (Rentz et al., 1996b).

The formation of polychlorinated dioxins and furans depends on a number of factors such as scrap composition, process type, temperature, etc.

3.5 Controls

Most of the secondary lead smelters are equipped with dust-removing installations, such as baghouses for the control of direct emissions. The control efficiency of these installations is often very high and can reach 99.9 %. In secondary lead production for most processes it is possible to carry out final dedusting with fabric filters. In this way clean gas dust loads in general below 5 mg/m^3 (STP) are achieved. For covering direct emissions from the refining and alloying kettles, primary suction hoods are arranged above the refining and melting kettles. These hoods are also linked to fabric filters. Waste gases from the furnace and the refining kettles may be dedusted together in one filter. Electrostatic precipitators or wet

scrubbers may be in use for special raw gas conditions. Wet scrubbers are sometimes in place for the control of SO₂. Fugitive particulate emissions can be collected by local systems like hoods and other suction facilities or by partial or complete enclosures (Rentz et al., 1996).

Primary measures for the control of SO₂ aim to reduce the sulphur content in the fuel and in the raw materials used. Accordingly lower SO₂ emissions occur when using natural gas instead of heavy fuel oil for short rotary, long rotary and reverberatory furnace firing. Within blast furnace operation, the use of coke with a low sulphur content reduces emissions.

Oxy-fuel burners have been used in short rotary furnaces resulting in a significant reduction of the fuel input. Accordingly, a smaller pollutant mass flow is observed, although the concentration in the off-gas may be higher than in conventional firing technologies.

Significantly lower emissions occur during secondary lead production if desulphurisation of the lead paste is carried out prior to thermal treating. Within the Engitec-CX process, for example, sulphur is removed from the electrode paste by adding NaOH or Na₂CO₃. According to an operator, a reduction of SO₂ emissions in excess of 90 % can be achieved by this measure (Rentz et al., 1996b).

4 SIMPLER METHODOLOGY

Application of general emission factors with appropriate activity statistics can be regarded as a simpler methodology for estimation of emissions from secondary lead production. However, it should be noted that the chemical composition of input scrap is one of the most important factors affecting the amount of emissions. The chemical composition of input scrap varies considerably from one plant to another and therefore emission factors also vary. Default emission factors in accordance with the simpler methodology are proposed in section 8.

5 DETAILED METHODOLOGY

In this case, different emission factors for various production technologies should be used. An account of the effect of emission controls should be considered. The different emission factors will have to be evaluated through measurements at representative sites.

6 RELEVANT ACTIVITY STATISTICS

Information on the production of lead in secondary smelters is available from the United Nations Industrial commodity statistics yearbooks. This information is satisfactory to estimate emissions with the use of the simpler estimation methodology.

However, in most cases, no information is available from the statistical yearbooks on the quantities of the metal produced by various types of industrial technologies employed in the secondary lead industry. Therefore, the application of detailed estimation methodology may be complicated unless the statistical data are available directly from a given smelter. Some

information in this respect is available from the International Lead and Zinc Study Group (ILZSG) (e.g. ILZSG, 1985).

7 POINT SOURCE CRITERIA

Secondary lead smelters should be regarded as point sources if plant specific data are available.

8 EMISSION FACTORS, QUALITY CODES AND REFERENCES

Information available from the sources cited below does not allow for further differentiation of emission factors with respect to either various industrial processes involved in the secondary lead production or different production technologies used at present. Therefore, the factors in table 8-1 can only be used in a simpler emission estimation methodology applied to the whole sector. As only limited background data are available on the emission factors, such as abatement type etc, a data quality E has been assigned.

The CORINAIR methodology requires the separate reporting of combustion related emissions (SNAP 030307) and process related emissions (SNAP 040300). Table 8-1 also gives emission factors related to the energy input in [g/GJ] based on CORINAIR90 data. In the case of using production statistics the specific energy consumption (e.g. GJ/Mg product) has to be taken into account, which is process and country specific. Within CORINAIR90 values for the specific energy consumption were reported between 38.5 and 100 GJ/Mg product.

9 SPECIES PROFILES

At present no reliable information exists on physical and chemical species of trace elements emitted during the secondary lead production. It can be assumed that the majority of trace elements volatilised from scrap and other lead-containing materials enter the atmosphere on fine particles. Table 9-1 indicates the heavy metal composition of dust from various secondary lead furnaces (Rentz et al., 1996):

Table 9-1: Weight composition of dust from secondary lead furnaces

	Weight composition of dust [wt.-%]	
Short rotary furnace	As 0.002 - 0.4	Ni 0.002 - 0.01
	Cd 0.07 - 0.7	Pb 20 - 54
	Cr 0.01	Sb 0.011 - 1
	Hg 0.01	Zn 0.5
Reverberatory furnace	As 0.1 - 10	Pb 30 - 50
	Cd 0.01 - 0.5	Sb 0.1 - 40
	Cu 0.001 - 0.005	Zn 0.01 - 1
Shaft furnace	As 0.01 - 3	Pb 30 - 55
	Cd 0.5 - 10	Sb 0.1 - 3
	Cu 0.01 - 0.04	Zn 1 - 10

Table 8-1: Emission Factor Table

Compound	Plant type	Emission factor	Data Quality	Abatement type	Abatement efficiency	Fuel type	Country or region	Ref.
SO ₂	<i>unknown</i>	0.85 - 8 g/GJ	<i>E</i>	<i>n. a.</i>	<i>n. a.</i>	<i>natural gas</i>	<i>Europe</i>	<i>CORINAIR90</i>
SO ₂	<i>reverberatory furnace</i>	40,000 g/Mg charged	<i>E</i>	<i>n. a.</i>	<i>n. a.</i>	<i>n. a.</i>	<i>U.S.A.</i>	<i>EPA 1990</i>
SO ₂	<i>blast furnace (cupola)</i>	26,500 g/Mg charged	<i>E</i>	<i>n. a.</i>	<i>n. a.</i>	<i>n. a.</i>	<i>U.S.A.</i>	<i>EPA 1990</i>
SO ₂	<i>pot furnace heater</i>	17.26 g/l burned	<i>E</i>	<i>n. a.</i>	<i>n. a.</i>	<i>NAPFUE 204</i>	<i>U.S.A.</i>	<i>EPA 1990</i>
SO ₂	<i>pot furnace heater</i>	9,611 g/Mm ³ burned	<i>E</i>	<i>n. a.</i>	<i>n. a.</i>	<i>NAPFUE 301</i>	<i>U.S.A.</i>	<i>EPA 1990</i>
SO ₂	<i>secondary metal production, process heaters</i>	17,209 S g/m ³ fuel	<i>E</i>	<i>n. a.</i>	<i>n. a.</i>	<i>NAPFUE 204</i>	<i>U.S.A.</i>	<i>EPA 1990</i>
SO ₂	<i>secondary metal production, process heaters</i>	19,006 S g/m ³ fuel	<i>E</i>	<i>n. a.</i>	<i>n. a.</i>	<i>NAPFUE 203</i>	<i>U.S.A.</i>	<i>EPA 1990</i>
NO _x	<i>unknown</i>	60 - 110 g/GJ	<i>E</i>	<i>n. a.</i>	<i>n. a.</i>	<i>natural gas</i>	<i>Europe</i>	<i>CORINAIR90</i>
NO _x	<i>reverberatory furnace</i>	150 g/Mg charged	<i>E</i>	<i>n. a.</i>	<i>n. a.</i>	<i>n. a.</i>	<i>U.S.A.</i>	<i>EPA 1990</i>
NO _x	<i>blast furnace (cupola)</i>	50 g/Mg charged	<i>E</i>	<i>n. a.</i>	<i>n. a.</i>	<i>n. a.</i>	<i>U.S.A.</i>	<i>EPA 1990</i>
NO _x	<i>pot furnace heater</i>	2.4 g/l burned	<i>E</i>	<i>n. a.</i>	<i>n. a.</i>	<i>NAPFUE 204</i>	<i>U.S.A.</i>	<i>EPA 1990</i>
NO _x	<i>pot furnace heater</i>	2,242,573 g/Mm ³ burned	<i>E</i>	<i>n. a.</i>	<i>n. a.</i>	<i>NAPFUE 301</i>	<i>U.S.A.</i>	<i>EPA 1990</i>
NO _x	<i>secondary metal production, process heaters</i>	2,396.78 g/m ³ fuel	<i>E</i>	<i>n. a.</i>	<i>n. a.</i>	<i>NAPFUE 204</i>	<i>U.S.A.</i>	<i>EPA 1990</i>
NO _x	<i>secondary metal production, process heaters</i>	6,591.15 g/m ³ fuel	<i>E</i>	<i>n. a.</i>	<i>n. a.</i>	<i>NAPFUE 203</i>	<i>U.S.A.</i>	<i>EPA 1990</i>
VOC	<i>pot furnace heater</i>	0.024 g/l burned	<i>E</i>	<i>n. a.</i>	<i>n. a.</i>	<i>NAPFUE 204</i>	<i>U.S.A.</i>	<i>EPA 1990</i>
VOC	<i>pot furnace heater</i>	44,851 g/Mm ³ burned	<i>E</i>	<i>n. a.</i>	<i>n. a.</i>	<i>NAPFUE 301</i>	<i>U.S.A.</i>	<i>EPA 1990</i>
VOC	<i>secondary metal production, process heaters</i>	23.97 g/m ³ fuel	<i>E</i>	<i>n. a.</i>	<i>n. a.</i>	<i>NAPFUE 204</i>	<i>U.S.A.</i>	<i>EPA 1990</i>

Table 8-1 (continued)

Compound	Plant type	Emission factor	Data Quality	Abatement type	Abatement efficiency	Fuel type	Country or region	Ref.
VOC	secondary metal production, process heaters	33.55 g/m ³ fuel	E	n. a.	n. a.	NAPFUE 203	U.S.A.	EPA 1990
VOC	secondary metal production, process heaters	44,851 g/m ³ fuel	E	n. a.	n. a.	NAPFUE 301, process gas	U.S.A.	EPA 1990
NMVOG	unknown	10 g/GJ	E	n. a.	n. a.	natural gas	Europe	CORINAIR90
CH ₄	unknown	2 g/GJ	E	n. a.	n. a.	natural gas	Europe	CORINAIR90
CO	unknown	7 - 30 g/GJ	E	n. a.	n. a.	natural gas	Europe	CORINAIR90
CO ₂	unknown	55 kg/GJ	E	n. a.	n. a.	natural gas	Europe	CORINAIR90
N ₂ O	unknown	3 g/GJ	E	n. a.	n. a.	natural gas	Europe	CORINAIR90
As	unknown	8 g/Mg Pb produced	E	n. a.	n. a.	n. a.	Austria	Schneider 1994
Cd	unknown	2.5-3 g/Mg Pb produced	E	ESP	ca. 99 %	n. a.	Europe	Pacyna 1986, Schneider 1994
Cu	unknown	1.0 g/Mg Pb produced	E	ESP	ca. 99 %	n. a.	Canada	Jacques 1987
Pb	unknown	770 g/Mg Pb produced	E	ESP	ca. 99 %	n. a.	Europe	Pacyna 1986
Pb	unknown	100-300 g/Mg Pb produced	E	n. a.	n. a.	n. a.	Europe, Canada	PARCOM 1992, Schneider 1994, Env. Can. 1983
Zn	unknown	150 g/Mg Pb produced	E	n. a.	n. a.	n. a.	Austria	Schneider 1994
Zn	unknown	300 g/Mg Pb produced	E	ESP	ca. 99 %	n. a.	Europe	Pacyna 1986
PCDD/F	unknown	5-35 µg I-TEQ/Mg Pb produced	E	fabric filter/lime injection - limited abatement	range	n. a.	Europe	Bremmer 1995 TNO 1995

n. a.: not available

10 UNCERTAINTY ESTIMATES

It is difficult to assess current uncertainties of emission estimates for pollutants emitted during the secondary lead production. Recently it was concluded that up to 50 % of uncertainties can be assigned for the emission estimates of most of the trace elements emitted from major point sources in Europe (Pacyna, 1994). Higher uncertainty can be assigned for emission estimates of these compounds from the secondary lead production. Information on emission factors and statistics is more limited for the secondary lead smelters than for major point sources, such as primary smelters and power plants.

11 WEAKEST ASPECTS/PRIORITY AREAS FOR IMPROVEMENT IN CURRENT METHODOLOGY

Improvement of emission factors is necessary in order to obtain more accurate emission estimates for the secondary lead production. This improvement should focus on preparing individual emission factors for major industrial technologies currently employed in the lead industry. In this way a detailed approach methodology for emission estimates can be applied. It will be necessary to obtain relevant statistical data on the production of lead in various secondary lead furnaces.

The fuel-specific emission factors provided in table 8-1, are related to point sources and area sources without specification. CORINAIR90 data can only be used in order to give a range of values with respect to point and area sources. Further work should be invested to develop emission factors, which include technical or fuel dependent explanations concerning emission factor ranges.

12 SPATIAL DISAGGREGATION CRITERIA FOR AREA SOURCES

National emission estimates can be disaggregated on the basis of production, population or employment statistics.

13 TEMPORAL DISAGGREGATION CRITERIA

The secondary lead production is a continuous process. No temporal disaggregation is needed.

14 ADDITIONAL COMMENTS

No additional comments.

15 SUPPLEMENTARY DOCUMENTS

UN ECE State-of-the-Art Report on the Heavy Metals Emissions, the UN ECE Task Force on Heavy Metals Emissions, Prague, the Czech Republic, June 1994.

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16 VERIFICATION PROCEDURES

Estimated emission factors could be best verified by measurements at plants using different industrial technologies.

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No additional references.

19 RELEASE VERSION, DATE AND SOURCE

Version : 3.1

Date : March 1999

Updated by: Otto Rentz; Ute Karl
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