

Mercury Emissions from Steelmaking: A Review

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Keywords:

Mercury, emissions, electric arc furnace, scrap, metal, air pollution

MERCURY EMISSIONS FROM STEELMAKING: A REVIEW

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The emissions of mercury from electric arc furnace steel-plants in the Nordic countries and the corresponding legislation has been reviewed and investigated. The principal analysis methods for continuous and static measurements are discussed as well as other available technologies for mercury measurement, filtering and removal. Analysis of a three-week emissions monitoring study at a single plant showed only weak correlations: raw materials may be critical to emissions, whereas stack temperature showed no correlation with total mercury emissions. In general, mercury emissions were found to be the greatest in steel plants using diverse scrap as compared to primarily automotive scrap.

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1 Introduction

Since the 1980s, emissions of mercury (Hg) from steel plants have been a concern. Although current emission levels are lower than that of earlier measurements, many factors surround the issue today.

Mercury is a toxic element, and the several hundred kilograms of mercury emitted nationwide by industrial sources every year has a tangible effect on the environment and our food quality. It is well known that the bio-accumulative nature of organic forms of mercury affects the fish and arctic animal population [1, 2]. Ingestion of this fish particularly affects pregnant or breastfeeding women, as mercury is known to transcend the blood-brain barrier and adversely affect growth and development of fetuses and young children [3]. As a result, various regulations govern the emissions of mercury in Europe [4]. These regulations will be discussed in the context of the Nordic countries.

Emissions of mercury from steel plants are predominantly elemental mercury [3]. This type of mercury is known to have the lengthiest cycle time in the atmosphere, and it deposits in arctic regions due to air flows [5]. This affects populations of people who are dependent on fish for the primary food source [1]. The positive aspect of mercury emissions to the atmosphere is that although measurements made on arctic ice profiles indicate a very significant increase in mercury deposition over the last 100 years, a decline has occurred within the last ten years [6].

There are three main forms of mercury in flue gases: elemental Hg (Hg^0), particulate Hg (HgP) and reactive (divalent) gaseous mercury (RGM, Hg(II) or $\text{Hg}_{(g)}^{2+}$) such as HgCl_2 [5, 7]. Emissions from steelmaking have historically been believed to be comprised of approximately 80 % Hg^0 , 5 % HgP and 15 % Hg(II) [3]. As compared to other industries, the elemental mercury emissions are an average of 30 % higher and the RGM emissions are 25 % lower. The implication of this is that although point source deposition may be lower for steel plants than other industries, the contributions to mercury depositing in arctic regions is quite high. For Nordic countries, therefore, mercury emissions from steel plants play a significant role in the health of the environment and fish populations.

Mercury emissions are most predominant from the ferroalloy industry and also from Electric Arc Furnace (EAF) steelmaking. In the ferroalloy industry, the source of mercury is from the mined manganese. From EAF steelmaking, the scrap quality is the major factor in mercury emissions. The focus of this work is primarily emissions from EAF steel plants.

Although many attempts have been made to pinpoint the source of mercury emissions in Electric Arc Furnace (EAF) steelmaking, there has not been clear success. Emissions of mercury are shrouded by contributions from various sources. The aim of this paper is to discuss these sources in greater depth. The results of a case study are also presented, which illustrate some of the problems associated with mercury emissions measurements.

The general trend for mercury emissions from EAF steelmaking has shown a decline over the last 12 years. These results are presented and discussed. Also presented are techniques to measure gaseous mercury, both continuously and intermittently. The advantages and disadvantages of each will be briefly reviewed. Current techniques for the treatment of gas phase mercury will also be reviewed.

2 Mercury in EAF steelmaking

The nature of EAF steelmaking involves the recycling of assorted grades of scrap metal in combination with chemical additives, and thus various root sources of emissions are plausible. The first and most obvious source of emissions lies within the types of scrap received. Frequently, old cars, metal from demolished buildings, scrapped submarines and other such sorts of metal are re-melted. Often coming with these types of scrap are small mercury sources, such as switches and thermostats. If a typical device contains 1 gram of mercury, it is clear how emissions of mercury can amount to hundreds of kilograms per year. Providing there is 1 gram of mercury for every 100 tons of scrap melted, a 90 % recycling yield and 450 000 tons of steel produced per year, 5 kg/yr of mercury emissions would be generated. This clearly doesn't account for the mercury emissions seen in steelmaking. However, with 1 gram Hg per ton of scrap, providing for a 90 % yield and the same production rate, 500 kg/yr of mercury would be emitted.

Considering the normalized average yearly emissions reported in Figure 1 and the dramatic drop in emissions after 1990, mercury in scrap has clearly played a significant role. According to the automotive scrap dealers in Sweden, serious attempts at sorting metal scrap to remove mercury sources began in 1992 [8]. Reducing the mercury in scrap from 1 gram per ton to 1 gram per 10 tons reduces emissions due to sources from 500 kg/yr to 50 kg/yr, considering the above calculations.

Despite this dramatic result, it is apparent from Figure 1 that other factors also have a large influence on mercury emissions. This can be seen from Figure 1 by the decrease in emissions after sorting out of mercury began, and then the slightly higher levels observed

between 1994 and 2000. Unfortunately, from the data currently available it is difficult to distinguish which factor is most significant in the emissions levels.

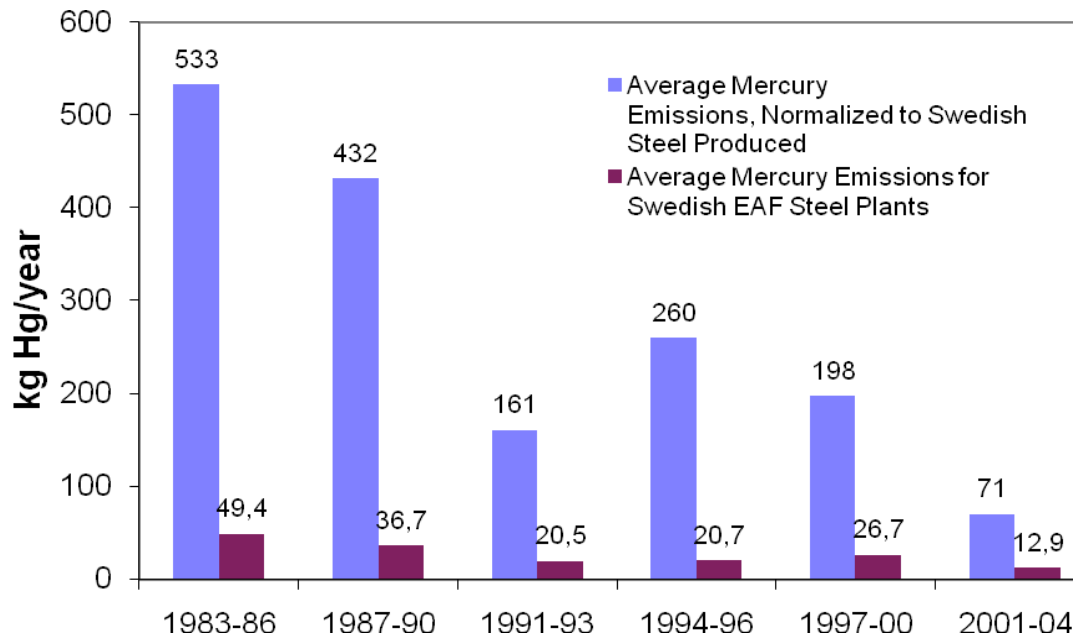


Figure 1. Emissions of mercury from EAF steel plants in Sweden, shown as total average emissions normalized to yearly steel production and by averaged yearly emissions.

Some other factors that may account for the decreased levels are that emissions measurements have been fine tuned. Measurements have been conducted more regularly and the statistics have been improved, i.e. outlying peaks in emissions levels may be filtered out of the data, as peaks can often be due to production stops or instrument calibration issues. Additionally, the testing method has become more uniform throughout Sweden, and the tests are often done for a longer period of time, namely 24 hours.

Although tangible mercury sources such as higher scrap quality and better sorting may be the biggest factor in mercury emissions, there are other sources of mercury that should be considered. Raw materials are known to contain trace amounts of mercury. According to an IVL study [9] in 1988, fluorspar (CaF_2) contains over 1 ppm of mercury, as shown in Figure 2. Other raw materials contain up to 0.2 ppm of mercury, as per a more recent investigation by Mo i Rana in 2004, also shown in Figure 2 [10]. Based on an addition of 150 kg of fluorspar per heat, the concentration of 0.2 ppm of mercury from the raw material becomes 0.3 g of mercury per heat. In stainless steel production, gas and coal are finding increased use. It has been shown that even these raw materials contain significant amounts of mercury [11]. However, the elevated levels of mercury found in residues and

exhaust fumes cannot be accounted for by the trace amounts of mercury that these materials contribute. Previous attempts to correlate mercury emissions with specific raw materials have shown that this is difficult to accomplish [9].

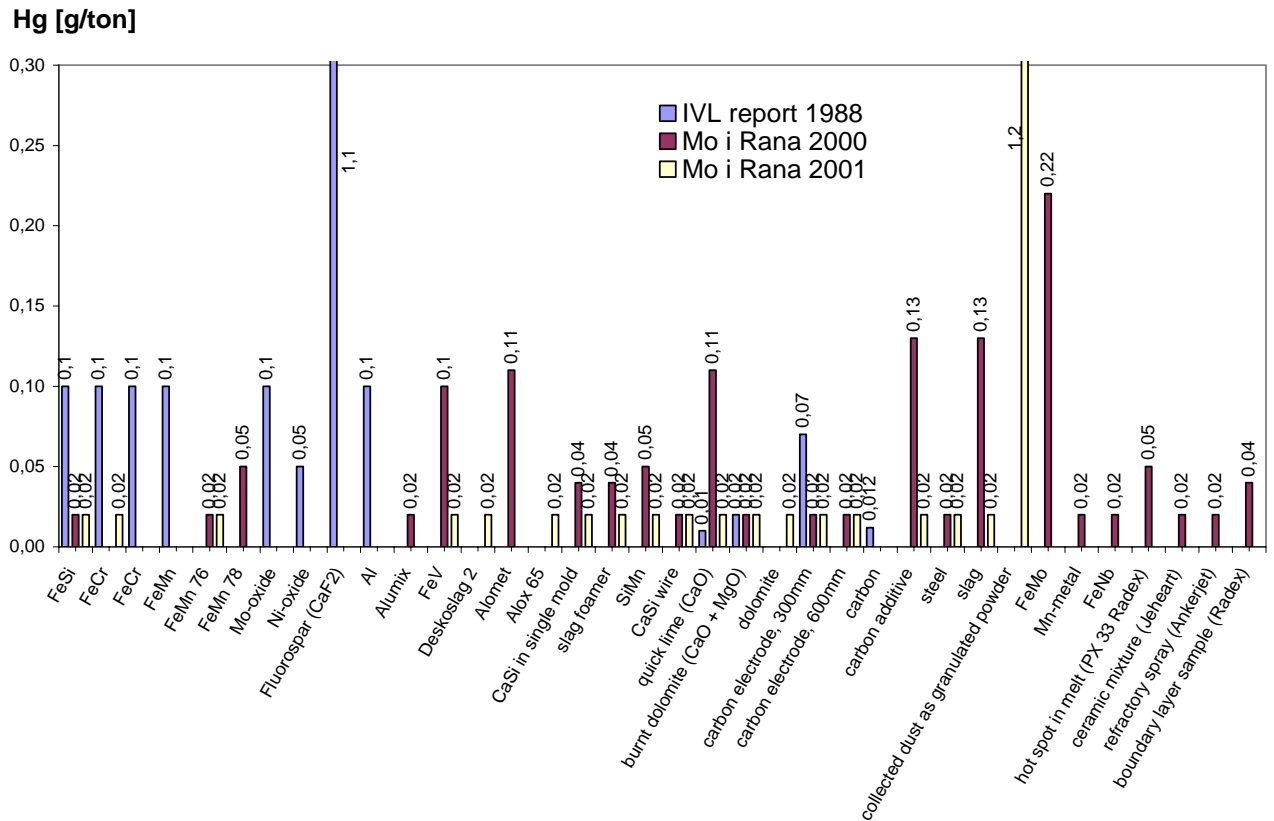


Figure 2. Parts per million of mercury in EAF-related materials. [8, 9].

Since the 1980s, the use of galvanized steel products has increased dramatically [12]. As such, higher levels of zinc have been entering the EAF with the steel scrap in non-stainless steel applications. The relationship between the increased use of galvanized scrap metal and emissions of mercury has not been fully defined.

From the calculated stable phases formed at low temperatures between mercury and zinc [13] it has been suggested that background atmospheric mercury may adsorb on the surface of zinc and steel scrap stored at sub-zero temperatures. In this way, mercury may enter the production line with the raw materials [6]. Then, with the high furnace temperatures, mercury is flashed off and passed through the air filtering systems. However, work done on zinc surfaces has shown that this is not likely the case [14]. Similar work done on copper surfaces has shown that mercury easily adsorbs to the

copper surfaces and could therefore enter the EAF with alloying elements or copper sources, which act as a source or a trap for mercury [14].

The same studies have shown that chlorine contamination on zinc and iron surfaces contributes to an increased uptake of mercury [15]. Chlorine contamination on metal may originate in various sources in the environment, but primarily from natural atmospheric deposition. Furthermore, it has been shown that the predominant oxidized form of mercury in flue-gases from coal combustion is HgCl_2 [16, 17, 18].

3 Regulations

In 1999, new and certain existing installations (technical units that have an effect on emissions or pollution) became covered by the IPPC directive [19]. This directive states that, “all the appropriate preventive measures are taken against pollution, in particular through application of the best available techniques,” and such that “no significant pollution is caused.” In Annex 1 of the directive, “Metal ore (including sulphide ore) roasting or sintering installations,” and, “Installations for the production of pig iron or steel (primary or secondary fusion) including continuous casting, with a capacity exceeding 2.5 tons per hour” are listed as activities that should realize “integrated prevention and control of pollution” to prevent or “reduce emissions in the air, water and land.”

The mercury emissions limits currently governing steel plants in Europe are minimal and diverse. However, these are expected to become more restrictive as awareness increases and BATs become available. Recently, a resolution has been passed on the European Mercury Strategy [20]. This states that further legislation is needed to restrict emissions from coal combustion processes. Furthermore, it calls for the proposal of “national emission limits as well as local air quality limits for mercury under relevant existing or separate legislative instruments.”

In Sweden, there are no nationwide regulations citing limits for mercury emissions. Despite this, some companies are still required to monitor and report mercury emissions. There is variability in the frequency of the measurements that must be reported. In most cases, emissions of mercury are measured on an annual basis, and companies measure the emissions in conjunction with other gas and dust emissions. However, the sampling time may vary from 3-6 hours to a 24-hour period, and the sampling time interval may be repeated for accuracy. However, the sampling performed is often limited to a two to three week period once during the year.

In Finland, the requirements for mercury emissions are similar to Sweden, wherein mercury emissions must be measured. In the case of the Ovako Oy AB steel plant in Imatra measurements of mercury must be performed once every two years, with a 24-hour or 15 charge minimum sampling time. Other requirements from Finnish authorities are such that continuous measurements must be employed in reporting emissions levels and attempts must be made to determine and reduce the source of mercury emissions. At Outokumpu Stainless Oy in Torneå, the regulations for their 2005 measurement were that a 20-day minimum continuous mercury measurement must be performed.

The Norwegian pollution control authority, SFT, has imposed several restrictions on Norwegian EAF and ferroalloy plants. Within the ferroalloy industry, the regulatory limits are slightly different between each company but are restrictive enough to require highly efficient gas cleaning systems to remove mercury. Emissions must be kept less than 50 kg/year, and according to the LRTAP Convention protocol, the emissions must be reduced to below 1990 levels with the application of BATs for stationary sources [4, 21]. The emissions limit placed on the single Norwegian scrap-based smelter is currently not strictly enforced, to allow for sufficient time to develop a large-scale or specially tailored gas cleaning system.

4 Environmental impact

Mercury is an element that once mined *viz.* different metals processing, has a long and unstable life, wherein its original stable state is rarely achieved. In steel production, mercury is only a trace metal but it accumulates into hundreds of kilograms of emissions per year. This mercury is eventually absorbed by forest floor beds, sediments in riverbanks, and becomes a part of our food chain. High ingestion levels have been linked to birth defects, learning disabilities and various other disorders. Ingestion of elemental mercury can lead to problems related to the spinal chord and even death. It is therefore important to reduce mercury emissions and capture mercury prior to release whenever possible.

The background level of mercury in the atmosphere is due to both natural and anthropogenic sources [3, 22]. With regards to anthropogenic sources, the deposition of particulate mercury is strongly proportionate to the proximity of the emission source. Recent data has shown that precipitation is an intermediate in mercury deposition [23]. So, although global mercury fluxes accumulate in Arctic regions, its deposition is enhanced in areas with high precipitation, such as southern Sweden.

Elemental mercury is widely transported, up to several tens of thousands of kilometers, and can cycle for up to 2 years before deposition [24]. Inorganic mercuric species are transported up to several hundred kilometers from their source, with a cycle time of a few months. It has also been suggested that there are temporal variations where mercury deposits in higher amounts in winter conditions.

5 Stack gas mercury emissions monitoring techniques

Many techniques are widely available for the measurement of mercury. Most techniques are for the determination of mercury concentration in liquid or solid samples. However, several operable techniques exist for measuring mercury in flue gas streams, both continuously and non-continuously. This discussion presents both types of stack gas measurements, as well as some common techniques for the measurement of mercury in liquid and solid samples.

5.1 Continuous stack gas mercury emissions monitoring techniques

Several automated methods are currently available to monitor mercury emissions continuously from flue gas streams, and these are presented and compared in Table 1. Developed and used commonly in Sweden is the **Semtech analyzer** [25]. This technique uses Cold Vapor Atomic Absorption Spectroscopy (CVAAS) to measure mercury and the instrument employs the Zeeman Effect background correction to reduce interferences from materials such as sulfur dioxide and hydrocarbons. This instrument can be equipped with a reducing solution of tin (II) chloride (SnCl_2) to reduce oxidized forms of mercury. In this way, total mercury can be measured. Without the reducing solution, only elemental mercury is measured. However, this measurement system is only effective with stack gas SO_2 concentrations of less than 50 ppm [26]. At higher SO_2 concentrations, the total mercury is underestimated, likely due to side reactions of mercury with sulfur and SnCl_2 . Therefore only elemental mercury can be accurately measured in this situation.

A second type of instrument is the **Tekran analyzer**, which is made in Canada [27]. This instrument employs Cold Vapor Atomic Fluorescence Spectroscopy (CVAFS), which is more sensitive than CVAAS. The detection limit for AAS is 0.01 – 1 ng/g whereas the detection limit for AFS is 0.001 – 0.01 ng/g of mercury. The Tekran instrument measures particulate, oxidized and elemental forms of mercury by separating the inlet gas streams.

One stream is passed through a thermal filler conversion unit then acid scrubbing while another stream is scrubbed directly to reduce the oxidized mercury.

Another common instrument for continuously measuring mercury from stack gas is the **Sir Galahad analyzer** [28] from PS Analytical in the UK. This instrument also uses CVAFS but instead of a thermal conversion unit, a constant-temperature gold trap is used. Again, the gas stream is split and a conditioning unit is used to separate the particulate mercury whereas the parallel stream is put through an adsorber to reduce the oxidized forms of mercury prior to analysis.

Mercury Instruments (MI) out of Germany and the USA makes an instrument for continuously monitoring mercury emissions, called the **SM-3 Mercury Stack Gas Monitor** [29]. This instrument uses UV-photometry to measure total mercury. The principle is similar that of the Tekran, a sample gas stream is passed through a heating system and then subsequently reduced (thermo-catalytic system). The gas is then dried and measured using CVAAS. Here, the low detection limit is less than 1 % of the measurement range, but as low as $0.45 \mu\text{g}/\text{m}^3$. Elemental, oxidized and particulate forms of mercury are measured.

A fifth instrument, which is produced by Nippon Instruments Corporation, is the **DM-6** [30]. This instrument uses CVAAS to measure mercury in the elemental form and also mercury that has been reduced using a solid reducing agent. QuickSilver Sky Sentinel, made by Genesis Lab Systems in the USA, produces an instrument used for stack gas mercury emissions monitoring. This uses AAS and only measures elemental mercury.

Verewa produces a unit called the **HM-1400 Total Mercury Analyzer** [31] for stack gas mercury monitoring. This instrument measures all forms of gaseous mercury and mercury compounds via fluid-bed charcoal scrubbers followed by wet chemical treatment prior to a secondary reactor tube. Following the secondary reactor is a cooled gas-liquid separation stage. The mercury is then analyzed using UV-absorption spectroscopy.

A final type of continuous stack gas emissions monitor for mercury is the **Mercem instrument** [32] made by SICK in Germany. This instrument measures elemental and oxidized mercury. In a similar fashion as the Semtech, this unit employs a tin (II) chloride reduction unit to determine oxidized mercury contributions. However, an amalgamation unit (gold-trap) is used to pre-concentrate the sample. Post heating, mercury is then measured by AAS.

Table 1. A comparison of available techniques for the continuous monitoring of mercury emissions from stack gases.

COMPANY NAME	EQUIPMENT NAME	TECHNOLOGY	SPECIES MEASURED	DETECTION LIMIT (LOW)	LAG TIME	MEASUREMENT RANGE	OTHER
Genesis Lab Systems	QuickSilver Sky Sentinel	AAS	Hg ⁰ _(g)	0.0001mg/m ³ (0.1 µg / Nm ³)	1 s	---	No false readings
Mercury Instruments	SM-3	Filter and thermo-catalytic reduction, UV-photometry	Hg ⁰ , HgP, Hg ²⁺	< 0.5 µg / Nm ³	<1 min	0-50, 0-75, or 0-500 µg/m ³	No reagents
Nippon Instruments	DM-6 + DM-6B + AM-4 Online Stack Gas System	CVAAS	Hg ⁰ and Hg ²⁺	0.1 µg / m ³		0.1 – 1000 µg/m ³	Solid reducing agent
PS Analytical	10.665, 50.043 (Sir Galahad)	AFS	Hg ⁰ , HgP, Hg ²⁺	0.01 µg / m ³	2.5 mins	0 – 3000 µg/m ³	Gold trap, uses Raoult's law or difference b/w HgP and HgTotal
SEMTECH	Hg 2010	CVAAS	Hg ⁰ , Hg ²⁺		1 min	0-50, 0-150, 0-400 g/Nm ³	Reduces via SnCl ₂
SICK	MERCEM	AAS	Hg ⁰ , Hg ²⁺	< 2% of measurement range	3 min	0-100 µg / m ³	Reduces via SnCl ₂ , gold trap
Tekran	3320 Mercury CEM Sample Conditioner	CVAFS	Hg ⁰ , HgP, Hg ²⁺	< 0.05 ng/m ³	2.5 mins	0.05– 1500 µg/m ³	No reagents
Verewa	HM-1400	Dual-Beam UV-Photometry	Hg ⁰ , HgP, Hg ²⁺	< 3 µg / Nm ³	<1 min	0-100 or 0-500 µg/m ³	Uses reagents to capture ions

5.2 Non-continuous stack gas mercury monitoring

The standard sample collection method for mercury analysis from flue gas streams in Sweden is the European standard method, SS-EN 13211. This method involves the sample collection from the flue gas using a large glass manipulator. The gas is passed through a heated particle filter ($>175^{\circ}\text{C}$), wherein the filter captures particulates and heating reduces any oxidized mercury forms. This is followed by a $\text{H}_2\text{SO}_{4(l)}$ stage to remove moisture. A small iodized activated carbon filter in line after the liquid solutions captures any elemental mercury (Hg^0 will not dissolve in the sulfuric acid). Where more mercury is present than can be dissolved, the excess mercury is not measured. The retained mercury is analyzed using AAS.

The OntarioHydro method [33] is commonly used in North America to efficiently report low concentrations of mercury from $0.5 - 100 \mu\text{g}/\text{Nm}^3$. It was developed for measurement of mercury emissions from flue gas stacks at coal-fired plants, but works effectively for other flue gas stack conditions, provided that the particulate load in the gas stream isn't excessive. A series of impingers are used to reduce and isolate the mercury. Elemental, oxidized, particulate and total mercury can be measured using CVAAS or CVAFS.

Proper gas flow sampling will have an impact on emissions values. The design of the collection tube is significant as it may be prone to clogging or leakage. The ISO method 10780 (similar to withdrawn ISO method 3966-1977) uses a combination of two types of pitot tubes to measure gas volumes and flow rates. However, it applies ideally only for gas streams with constant flow densities, temperature, rate and pressure.

5.3 Wet-lab analytical methods

Several methods exist for the measurement of mercury, as have been previously outlined in the Position Paper on Mercury [34]. These are presented in Table 2.

Table 2. Most frequently used methods for quantification of mercury and their relative detection limits [3, 34].

Method	Reported Detection Limits
Colorimetric methods	0.01 - 0.1 $\mu\text{g}/\text{g}$
AAS graphite furnace (GF AAS)	1 ng/g
cold vapour (CV AAS)	0.01 - 1 ng/g
AFS cold vapour (CV AFS)	0.001 - 0.01 ng/g
NAA instrumental (INAA)	1-10 ng/g
radiochemical (RNAA)	0.01 - 1 ng/g

GC Electron Capture Detector	0.01 - 0.05 ng/g
Atomic Emission Detector	~ 0.05 ng/g
Mass Spectrometer	0.1 ng/g
CV AAS/AFS	0.01- 0.05 ng/g
HPLC UV	1 ng/ml
CV AAS	0.5 ng/ml
CV AFS	0.08 ng/ml
Electrochemical detectors	0.1-1 ng/ml
ICP-MS	0.01 ng/ml
ICP-AES	2 ng/ml
Photo-acoustic spectroscopy	0.05 ng
X ray fluorescence	5 ng/g - 1 µg/g
Electrochemical methods	0.1 - 1 ng/g
Gold-film analyzer	0.05 µg/g

6 Mercury capture and recovery techniques

Common techniques for gas cleaning are bag filter and scrubber systems. According to the global mercury assessment [3], acidic scrubbers or adsorption by active carbon are ideal for the reduction of mercury. In acid scrubbing, HgCl_2 and other forms of oxidized mercury are removed, but elemental mercury is not removed in similarly significant quantities due to its insolubility. Scrubbing may also reduce divalent mercury compounds to Hg^0 [35], which results in a reduction of the overall removal efficiency of mercury in the scrubber. This reduction occurs *viz.* liquid redox reactions. Therefore, the pH and chlorine concentration are important for proper control of mercury removal.

The Odda process is another method for the removal of mercury from flue gases. This process was developed primarily for nitrate removal, but may also be effective for mercury removal [1, 36]. In this process, mercury reacts with chlorine to form mercuric chloride, which is then precipitated out.

At temperatures less than 140°C , oxidized mercury condenses on particles [37]. This particulate mercury can be removed through bag filters, electrostatic precipitation (ESP) or other particulate matter (PM) control devices.

Selective catalytic reduction (SCR) can be used to oxidize elemental mercury prior to scrubbing or PM control, and thus elemental mercury can be removed efficiently. Furthermore, the application of heat exchangers prior to bag filters reduces temperatures which enhance the speciation of elemental mercury and the removal of particulate mercury.

Using a high surface area activated carbon is known to enhance mercury capture. Carbon filters can be used, or powdered activated carbon (PAC) or granulated activated carbon (GAC) can be injected into the gas stream [38 – 41]. This technique can be costly and also creates a large volume of contamination, but the capture efficiency of elemental mercury is high. Sulfur impregnation of the carbon, provided the AC has a large pore diameter, also enhances the uptake of mercury [42]. Where necessary, this technique should be implemented prior to other PM and scrubber equipment.

Some steel plants in Germany currently use carbon-injection methods to meet regulatory limits. Calcium (in the form of dolomite or calcium magnesium acetate (CMA)) is also known to reduce emissions of SO_x and thereby effect the capture of particulate mercury. Sulfate makes the calcium more wettable and similarly sulfur causes a more wettable carbon surface, and thus the surfaces are more available for mercury condensation and/or reaction. The process that uses both calcium and carbon injection, often prior to fabric filters, is called Fisorption [43]. This process is applicable for dry flue gas.

The ferroalloy industry employs several different methods for the reduction of mercury from gas streams. The first method is called the Miltec method [44], wherein the flue gas stream is passed through a scrubber, followed by heating in an oven to remove excess water, then washing with sodium bisulfite. In this stage, the oxidized mercury is removed. After the wash stage, the stream is passed over a limestone bed to particulate elemental mercury, which can then be removed. Another solution is to use an electrostatic filter, followed by heating to remove excess moisture, then passing the stream through an activated carbon adsorption bed. This removes oxidized mercury first, then particulate and elemental mercury.

Another solution used in Sweden for treating mercury emissions from smelter operations includes the use of a multi-stage filtering system. A packed bed reactor captures elemental mercury, followed by a Dowa filter (lead (II) sulfide covered pumice filter) to capture metallic, oxidized and particulate mercury. Another solution used is wet electrostatic precipitation to remove particulate and elemental mercury, followed by low temperature selenium filters to remove additional elemental mercury. This solution is more plant-specific as other chemicals are treated simultaneously. An additional gas cleaning option is to use a filter cake of lime and activated carbon on bag filters [45]. This method is similar to that of activated carbon injection, but occurring at a different stage of the gas cleaning, and employing different conditions of pH and temperature.

The following table taken from the UNEP Global Hg Assessment shows the most commonly available mercury control technologies and their removal efficiencies.

Table 1. Some mercury control technologies [3].

Technology	Mercury control effectiveness	Control of other pollutants	Availability and other notes
Selective Non-Catalytic Reduction	Unknown	30-60% NO _x reduction	Available and used on utility boilers. Minor reduced boiler efficiency.
Selective Catalytic Reduction	SCR + wet scrubber combination may result in substantial mercury reduction (see below)	70->90% NO _x reduction	Available and used on larger power plants. Minor reduced boiler efficiency. SCR catalyst may improve oxidation of elemental mercury to divalent mercury, which can be captured in a wet scrubber used for SO ₂ control.
Low NO _x burners	None	>50% NO _x reduction possible	Available and in use on most coal-fired boilers. SCR and SNCR retrofits provide additional NO _x control beyond low-NO _x burners. It has been postulated that LNBS will improve mercury capture due to the increase in amount of unburned carbon (i.e., carbon loss on ignition [LOI]) in the flue gas stream that may act in a manner similar to activated carbon injection.
Wet Scrubber	Up to 90% removal of oxidized Hg. No removal of elemental Hg.	80->90% SO ₂ removal	Already in use to reduce SO ₂ . Effectiveness for Hg removal highly dependent on mix of chemical species present and on other factors including liquid-to-gas ratio, chlorine content, and coal type.
Combined SCR with Wet Scrubber	>80% removal of overall Hg may be possible for units firing bituminous coals; effectiveness for units firing subbituminous coals is uncertain at this time.	>90% SO ₂ and >90% NO _x removal possible	SCR already in use to reduce NO _x . Helps convert elemental Hg to soluble, oxidized form, thereby allowing for greater removal by downstream wet scrubber. Results are based on limited but encouraging data. The ability of SCR to improve the oxidation of Hg for capture in scrubbers may be highly coal-specific.
Dry scrubber with ESP or FF	6-9% reported removal by NEG/ECP; recent EPA studies reported average removal of approx. 63%)	80-90% SO ₂ removal	In use on only 1% of US boilers (most units apply wet scrubbers). Removal efficiency for Hg depends on speciation, temperature, and chlorine content. Lime scrubbers show better Hg removal in pilot tests.

Electrostatic Precipitator (ESP)	0-82% removal (cold-side ESP) reported by NEG/ECP; EPA found 36% for bituminous and 3% for subbituminous	>99% PM removal	Already in use for particulate removal. Cooler temperature improves ESP performance. US EPA found Hg removal efficiency of 42-83% on oil-fired boilers.
Fabric Filter (baghouse)	0-73% removal reported by NEG/ECP; EPA found removals of 90% for bituminous and 72% for subbituminous	>99% PM removal	Only filters providing particulate collection efficiencies >99% appear to reduce significant amounts of Hg, but data are limited. Again, lower temperatures appear to improve performance. Baghouses are more effective than ESPs in controlling mercury.
Enhanced ESP	0-50% removal at one test unit	>99% PM removal	Enhanced ESPs being developed to capture finer particles may remove more Hg. At one test unit Hg removal improved with lower temperature.
Wet ESP	Around 30% removal in 2 pilot scale studies	56% mean PM removal in pilot studies	Wet ESP being investigated for “polishing” residual emissions from other controls. May improve mercury removal. Lower temperature improves Hg control.
Combined ESP/Baghouse	34-87% removal in 2 pilot facilities	>99% PM removal	Combination technology to achieve very low PM emissions can improve removal of Hg & other toxics when used in conjunction with powdered activated carbon.
Carbon injection	Recent full-scale test results indicate about 80% removal with bituminous coal+ESP+COHPAC and 55-60% removal with subbituminous coal+ESP	Not applicable	Cost and removal effectiveness are directly related to the amount of carbon used. Used carbon may create a hazardous waste disposal issue. Carbon injection on utility boilers is currently under development and demonstration, but is not yet commercially deployed.
Fuel Switching (coal-fired to natural gas)	>99% removal for natural gas	>99% SO ₂ and PM control; 50-75% NO _x reduction	Fuel switching reduces multiple pollutants, incl. NO _x , SO ₂ , particulates and CO ₂ . Accounting for multiple pollutant benefits reduces control costs for mercury alone. Cost affected by several factors, including fuel costs, other pollutant control costs, heat rate, facility age, capacity factor, new plant capital costs and discount rates.

Abbreviations: SNCR - Selective non-catalytic reduction; ESP - Electrostatic precipitator; SCR - Selective catalytic reduction; PM - Particulate matter; LNB - Low-NO_x burner

6.1 Flue gas reactions

The three forms of mercury in flue gases are elemental mercury, Hg^0 , particulate mercury (often elemental mercury bound to particulates in the gas such as soot particles) and oxidized mercury. The most common form of oxidized mercury in flue gases is HgCl_2 [46]. However, the final speciation of mercury will depend on the ratio of Hg(II) to Hg(tot) [47].

Controlling the temperature of the gas stream is critical for the speciation of mercury. At temperatures below 350°C , mercury is not oxidized, and according to Wang *et al.*, this limit is 500°C [26]. However, if mercury has already formed a compound, it will speciate into elemental mercury at temperatures below 140°C [48].

A US EPA study reported that calcium plays a dual role in the oxidation of mercury [49]. Calcium binds mercury (II) but also reacts with flue gas acids that oxidize elemental mercury. This leads to higher values of un-captured elemental mercury in the flue gas. With respect to the Torneå plant and the mercury detected in the AOD, this could mean that the addition of dolomite frees up elemental mercury which is easily read by the Semtech analyzer.

Table 3. Some mercury flue gas combustion reactions [46].

Oxidation Reactions	Oxidation Product
$2\text{Hg}^0 + \text{O}_2$	2HgO
$\text{Hg}^0 + \text{Cl}_2$	HgCl_2
$2\text{Hg}^0 + \text{Cl}_2$	$\text{Hg}_2\text{Cl}_{2(s)}$
$\text{Hg}^0 + 2\text{HCl}$	$\text{HgCl}_2 + \text{H}_2$
$2\text{Hg}^0 + 4\text{HCl} + \text{O}_2$	$2\text{HgCl}_2 + 2\text{H}_2\text{O}$
$4\text{Hg}^0 + 4\text{HCl} + \text{O}_2$	$2\text{Hg}_2\text{Cl}_{2(s)} + 2\text{H}_2\text{O}$
$2\text{Hg}^0 + \text{NO}_2$	$\text{HgO} + \text{NO}$
Reduction Reactions	Reduction Product
$\text{HgO} + \text{SO}_2$	$\text{Hg}^0 + \text{SO}_3$
$3\text{HgCl}_2 + 2\text{Fe}_{(s)}$	$3\text{Hg}^0 + 2\text{FeCl}_{3(s)}$
$\text{HgO}_{(s)} + \text{CO}$	$\text{Hg}^0 + \text{CO}_2$
$\text{HgO} + \text{NO}$	$\text{Hg}^0 + \text{NO}_2$
$\text{HgCl}_2 + \text{SO}_2$	$\text{Hg}^0 + \text{SO}_2\text{Cl}_2$

7 Swedish Hg emissions data

Mercury emissions have been monitored from Nordic steel plants for over thirty years. Various techniques have been used, but in more recent years, two techniques have dominated: continuous Semtech measurements and the non-continuous SS-EN 13211 method. The results from these two techniques are presented and compared. Recent emissions data from Nordic steel producers are also discussed.

7.1 Swedish EAF steel making

The normalized average mercury emissions from Swedish EAF steel plants are shown in Figure 3. These values have been normalized to the yearly steel production and to the averaged yearly emissions of mercury in Sweden. Emissions of mercury from EAF steel making have dropped by over 80 % since the early 1980s, as shown in Figure 3. Sorting of scrap has been the most significant factors influencing this decrease. Other factors include more continuous and representative measurements, the overall decrease in EAF steel plants and efforts to purchase higher quality scrap.

Analysis of the steel plants in operation shows that Domnarvet and Degerfors contributed significantly to the yearly mercury emissions until 1986, and emissions from Hellefors were minor. After 1987, the plants in Domnarvet and Degerfors continued to contribute substantially (~28 %) to the annual mercury emissions, but after 1990, emissions of mercury from Domnarvet were not recorded.

The drop in emissions after 1990 was approximately 60 %, wherein the closed down plants had contributed 55 % of the emissions between 1983 and 1986, and 28% of the emissions between 1987 and 1990. Between 1990 and 2003, Degerfors continued to emit on average, ~24 % of the total emissions to Sweden. In 1996, the high yearly emission from Sandviken, normalized to the total mercury emissions for steel production in Sweden, results in their emissions contributing to 69% of the total mercury emissions in Sweden for that four-year period. Normalizing the data to EAF steel production by individual company only (i.e. Not to all Swedish production), Degerfors, Sandviken and Avesta maintained the highest normalized percentage of contributions to emissions.

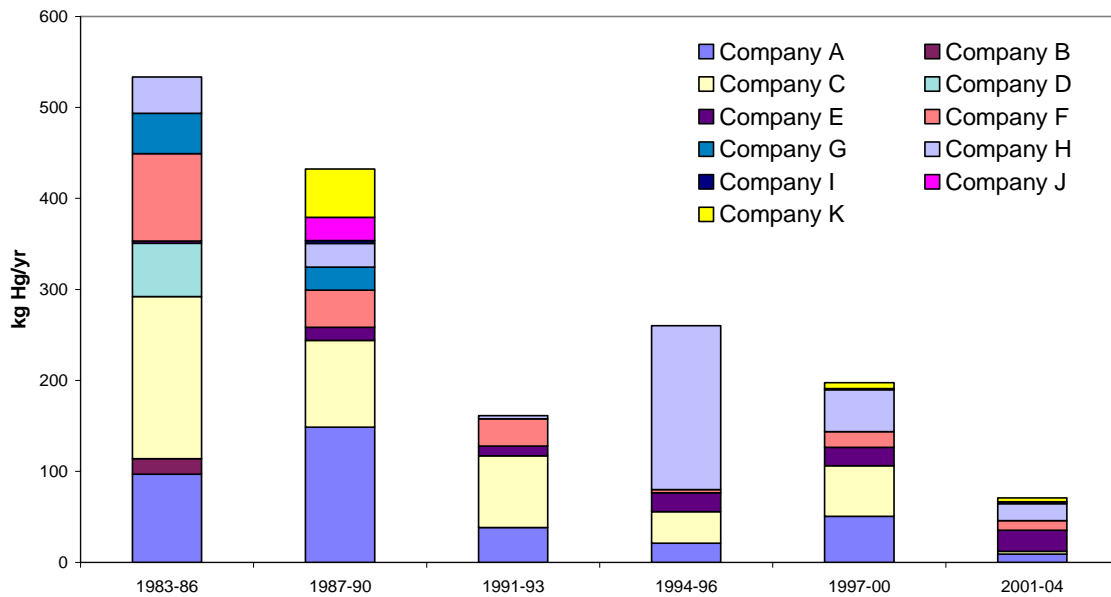


Figure 3. Normalized average emissions of mercury from EAF steel plants in Sweden.

By comparing normalized emissions from individual companies since 2001 as shown in Figure 4, there is a significant variation in the emissions values. Companies A and J are producers of stainless steel. Therefore it cannot be concluded that emissions are lower from this type of steel plant. Furthermore, the scrap metal used by stainless steel facilities does not generally include galvanized scrap. This corresponds well with the results of a study by Sandvik, a stainless steel producer, in which they could not observe a difference in their emissions levels when running clean scrap coming directly from Swedish automobile manufacturers and scrap originating from various other sources that had been stored outside for some time [50].

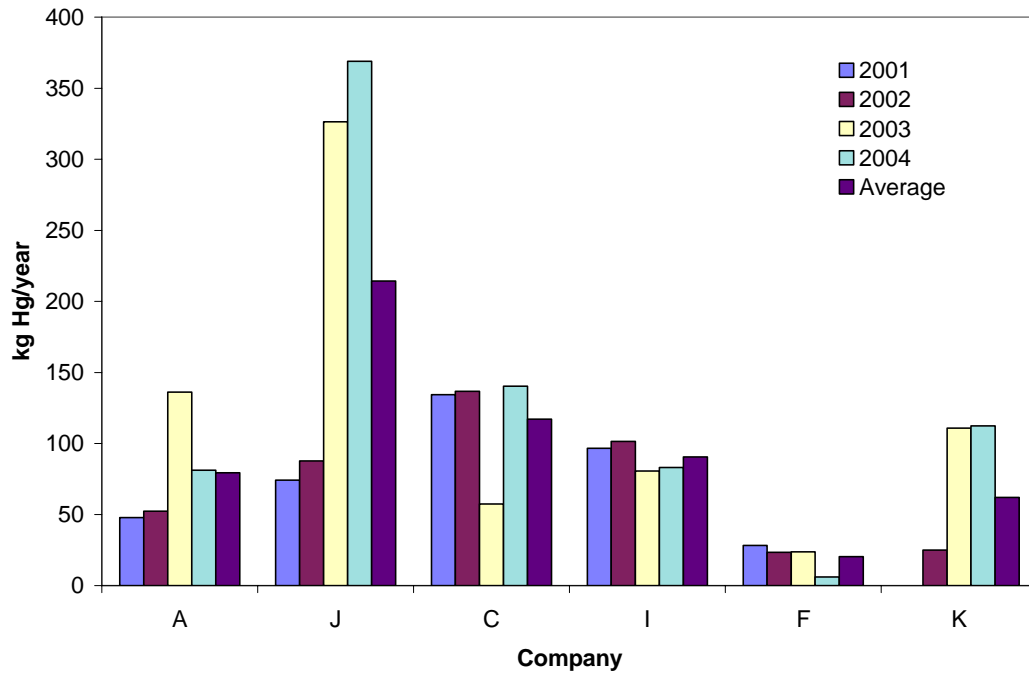


Figure 4. Normalized emissions of mercury from EAF steel plants in Sweden for the years 2001 to 2004. Companies A and J are producers of stainless steel.

The raw emissions data in Figure 5 shows the emissions of mercury since 1991 to average less than 50 kg of mercury per year from each company, with the exception of Company J, as shown in Figure 5. Companies A, B and J are producers of stainless steel. These emissions values vary with production, but from Figure 4 we saw that no trend could be observed from the differences in emissions between stainless and non-stainless steel producers. However, some of the variability in the yearly emissions may be due to the measurement techniques. If the measurement was a sample taken that occurred during a spike in emissions, this value was weighted to represent the entire years' emissions of mercury. Using the SS-EN 13211 method, a sample is drawn for between three to six and 24 hours and then the result is time-averaged. However, there was variability in that some measurements were only of elemental mercury, while others included oxidized mercury species. In the case of company J in 1996, the measurement was a single measurement period taken to represent the entire year, and thus may have been an anomaly.

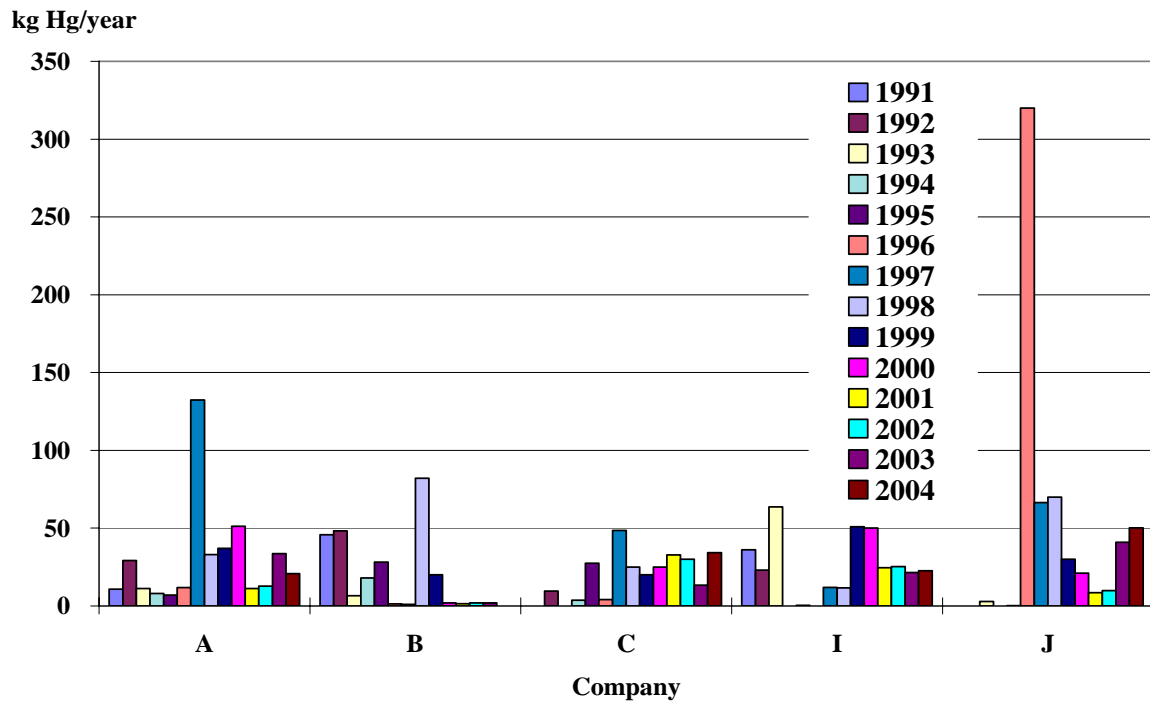


Figure 5. Emissions of mercury from EAF steel plants in Sweden since 1991. Companies A, B and J are producers of stainless steel.

The total emissions from Swedish EAF steel plants are presented in Figure 6. The decline in the mid-nineties was followed by a stable trend. The largest factors in the initial decline is believed to be due to enhanced scrap sorting methods in combination with more specialized and streamlined steel processes. Another factor is the uniformity of the emissions measurement technique. Longer testing intervals have been combined with continuous measurement techniques, thus leading to more accurate and uniform data.

Emission of mercury from Swedish EAF steel plants

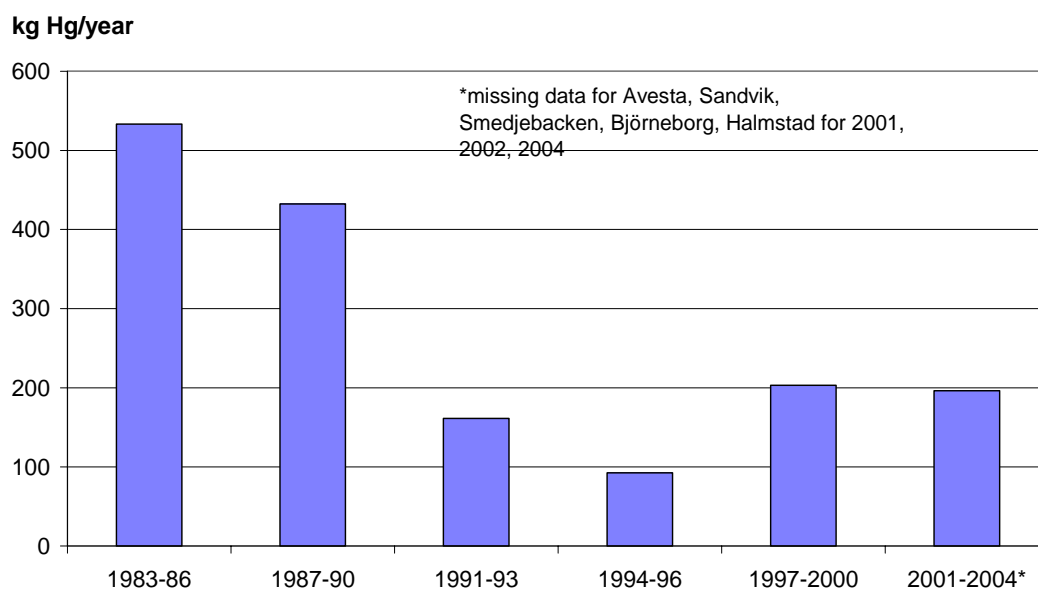


Figure 6. Total emissions of mercury from EAF steel plants in Sweden since 1983.

The variability in the emissions of mercury as grams per ton of produced steel is not as significant as the variability in the yearly emissions values, shown in Figure 7. This may be due to several factors, the most significant being the mechanism for recording emissions.

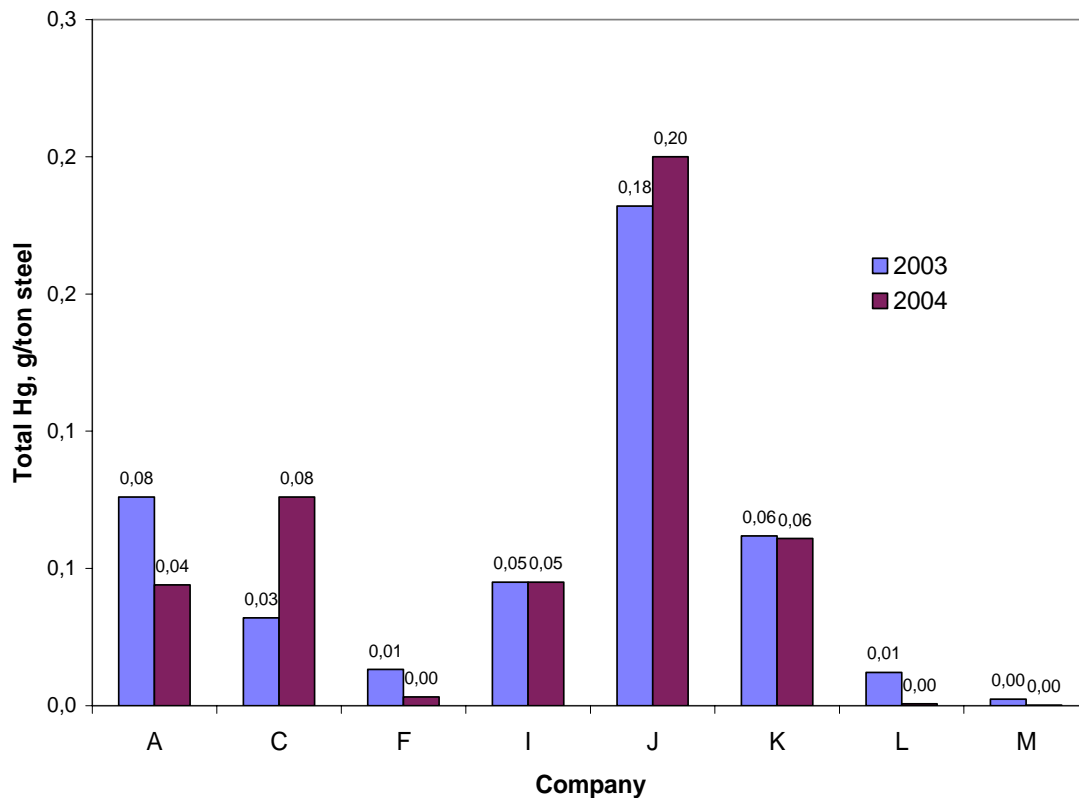


Figure 7. A comparison of mercury emissions in gram of mercury per ton of produced steel from Swedish EAF steel plants in 2003 and 2004.

Separating the mercury emissions into gas phase emissions and emissions from dust as shown in Figure 8, it becomes apparent that the dust contributes significantly to emissions of mercury. The ratio between the dust and the gas emissions varies not only with different companies, but also within each company. Furthermore, the gas to dust ratio has not changed significantly in the decade represented here. It would be expected that temperature control prior to the bag houses would positively impact the removal of mercury, and this would be visible as lower emission rates and higher values for mercury in separated dust.

With respect to the speciation of mercury in the gas phase, many factors are involved. Not only are the bag house temperature and inlet gas temperature factors, the maintenance and operation of the bag houses are significant. Bag houses operating at higher temperatures will naturally detect more oxidized mercury species than particulate species because of the oxidation reactions occurring above 350°C. This means lower capture rates from the bag houses and higher gaseous emission rates.

Furthermore, the presence and quantity of chlorine in the gas stream strongly affect the oxidation and therefore capture of mercury [51]. It has been shown by Li *et al.* [17] that in the presence of chlorine, mercury [I] or mercury [II] will preferentially bind to chlorine as compared to oxygen. By the same reaction kinetics, mercury bonds to available sulfur and thus ease of removal is enhanced.

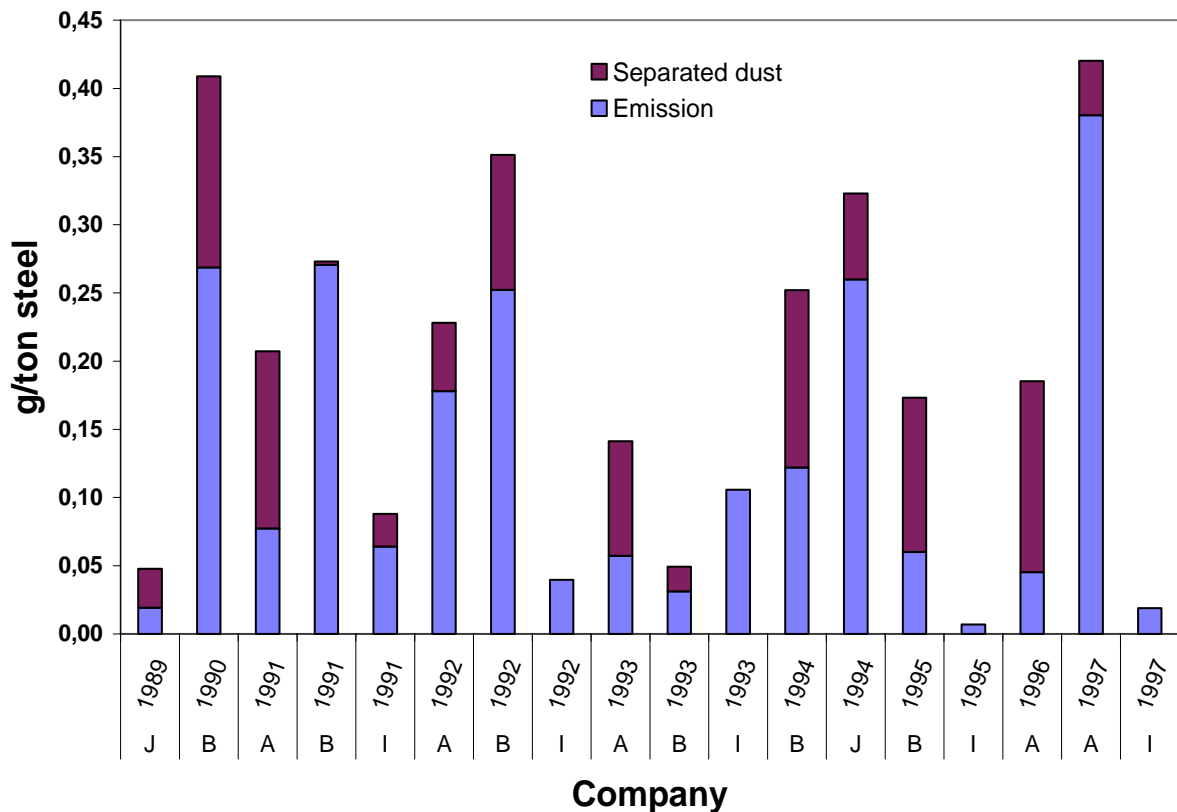


Figure 8. Emissions of mercury from Swedish steel plants from both the gas and the dust.

7.2 Case Study: Outokumpu Stainless AB (Avesta)

An in-depth study has been performed using data collected by IMKAB using a Semtech over a three-week interval. An attempt was made to correlate peaks in emissions data with scrap metal and other raw material inputs as well as production information. The results are presented below.

7.2.1 Experimental details

The data has been captured from the Semtech, where the time lag has been kept constant at five minutes. In other words, after a basket was charged, the emissions data were collected in the stack approximately five minutes after the charging, to allow for the emissions to reach the stack. Data was collected and averaged every sixty seconds. This data was further averaged per heat and per charge. Production and raw materials data was also collected during the same time period.

The data was filtered to sort for peaks in emissions. These peaks were further sorted into steel grade. For each steel grade with more than fourteen peaks, the steel grade was examined against various production and raw material weights using a pivot table.

7.2.2 Results and discussion

An overview of the data is presented in Figure 9. Over the three week period, it is clearly evident that the emissions of mercury vary greatly, from an average of less than 1.7 grams per hour of mercury excluding the peaks (for all values less than 7.2 g/h). For all peaks above 7.2 g/h, the average measured flow rate of mercury was 17.0 g/h.

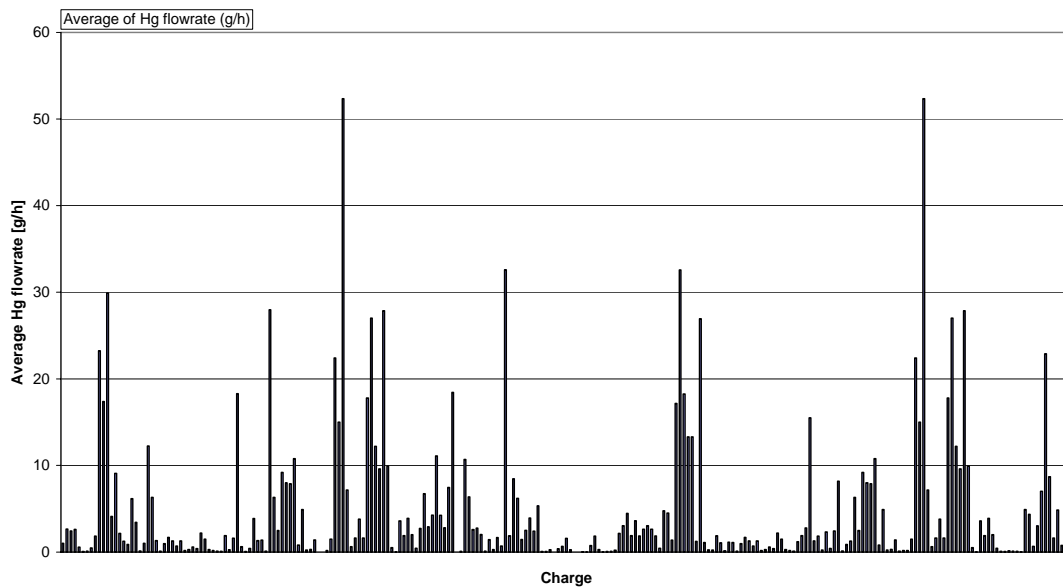


Figure 9. Average mercury flow rate per charge over three week measuring interval.

The data was then plotted against the channel temperature, as presented in Figure 10. The fluctuations in emissions do not correspond with temperature variations in the stack where the emissions were measured.

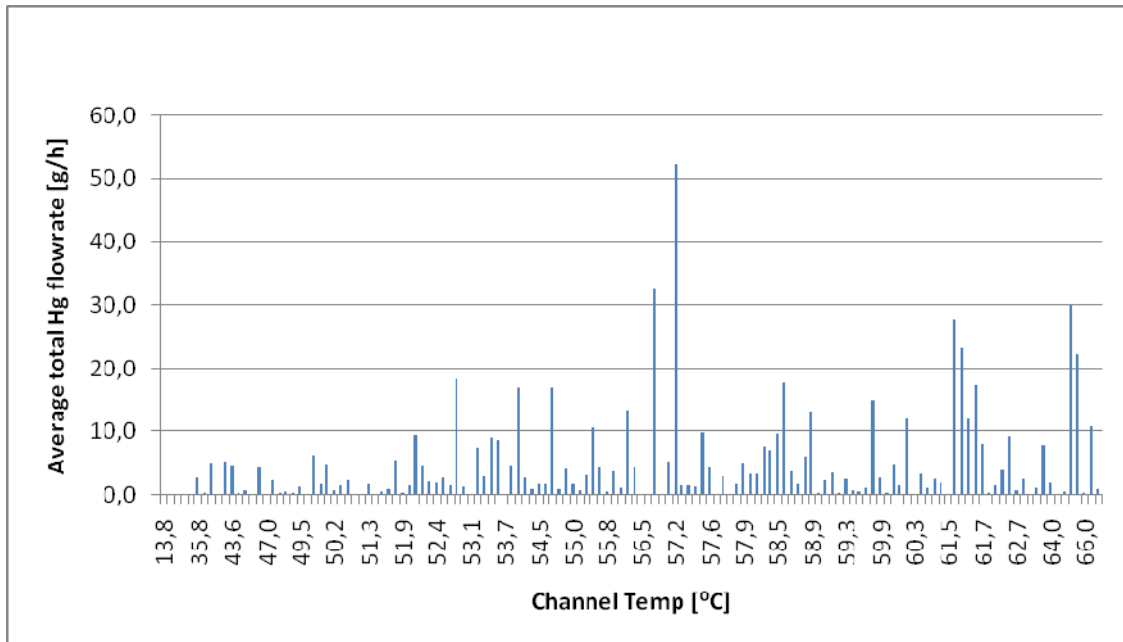


Figure 10. Average mercury flow rate plotted against temperature. Increasing temperature within the temperature range presented has no effect of increased mercury emission.

Pre-drying the heats did not affect the emissions of mercury. From Figure 11, it can be seen that there is no significant difference between the preheated and the non-preheated batch results.

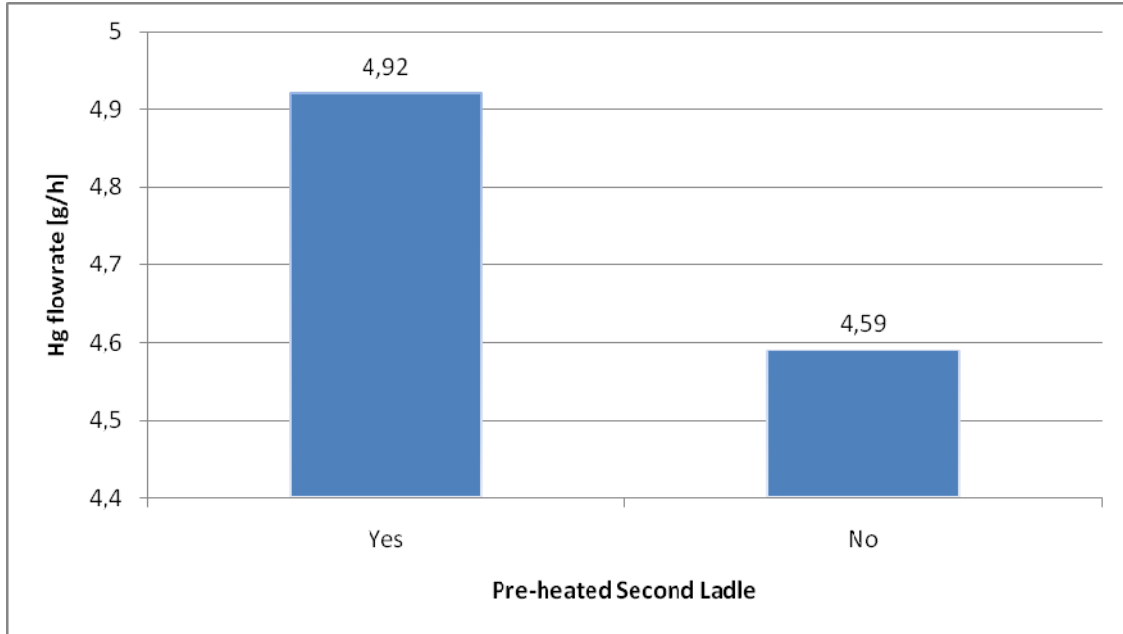


Figure 11. Preheated batch emissions comparison. Within the limits of error, no significant difference is observed between preheating and not preheating.

Upon examination of the average mercury emissions with steel grade, shown in Figure 12, it is evident that the steel grades 3, 4, 5, 8, 10, 13 and 31 have average higher emissions of mercury than other grades, where the grades have been renamed for privacy. These grades all have higher emission values than 8.0 grams per hour.

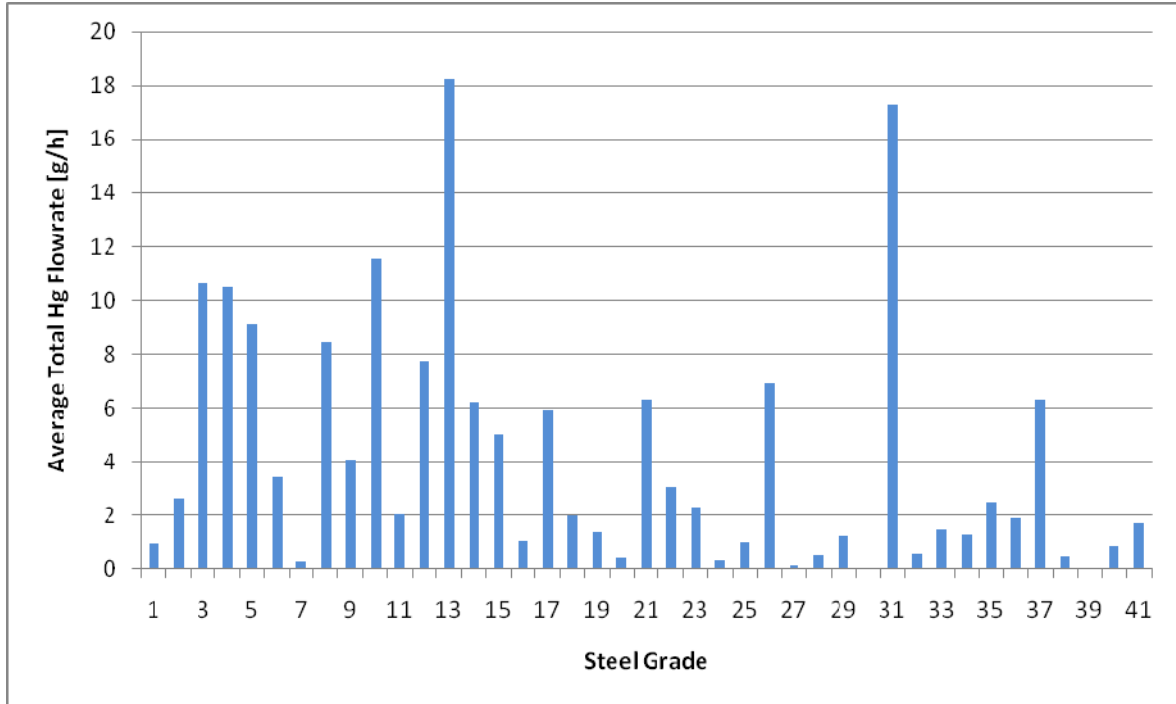


Figure 12. Several steel grades, renamed here in ascending order, have significantly higher average mercury emissions than others. These have been selected out for further evaluation.

This steel grades with higher than 8 grams per hour of mercury emissions were then selected for further analysis. In Figure 13 it is shown that there are some weak correlations with temperature. At temperatures less than 56°C, the emissions of mercury are not as high.

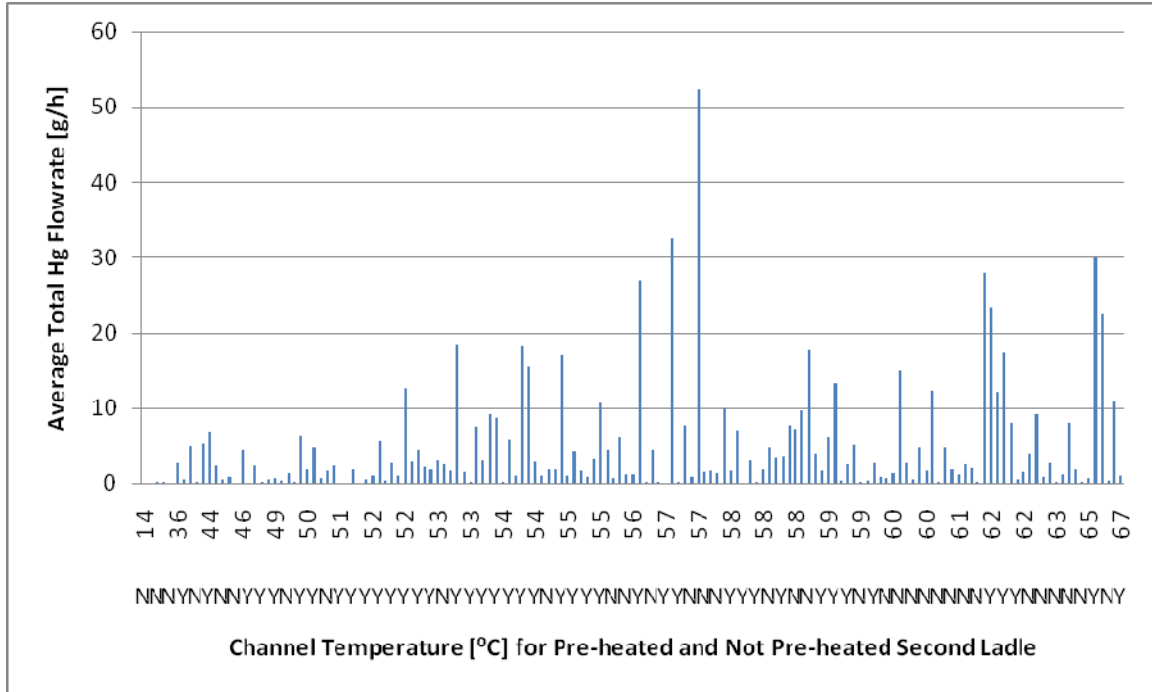


Figure 13. Sorting according to channel temperature and then the preheating of the basket shows that within a certain range of temperatures, 56°C to 62°C, emissions of mercury are highest, whereas preheating has little effect.

Figure 14 shows a selection of the most grades with the highest emissions. These were selected for further analysis.

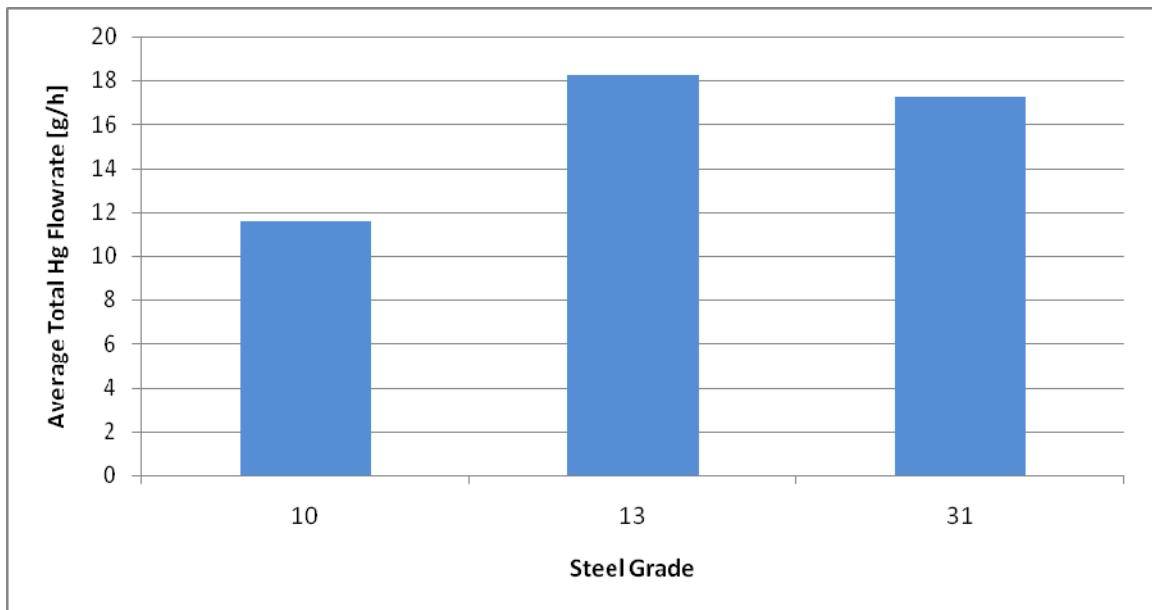


Figure 14. Grades with highest emissions.

A closer examination of the steel grades with the highest emissions was performed. The effect of predrying in Figure 15 is shown. Above 54°C emissions are higher.

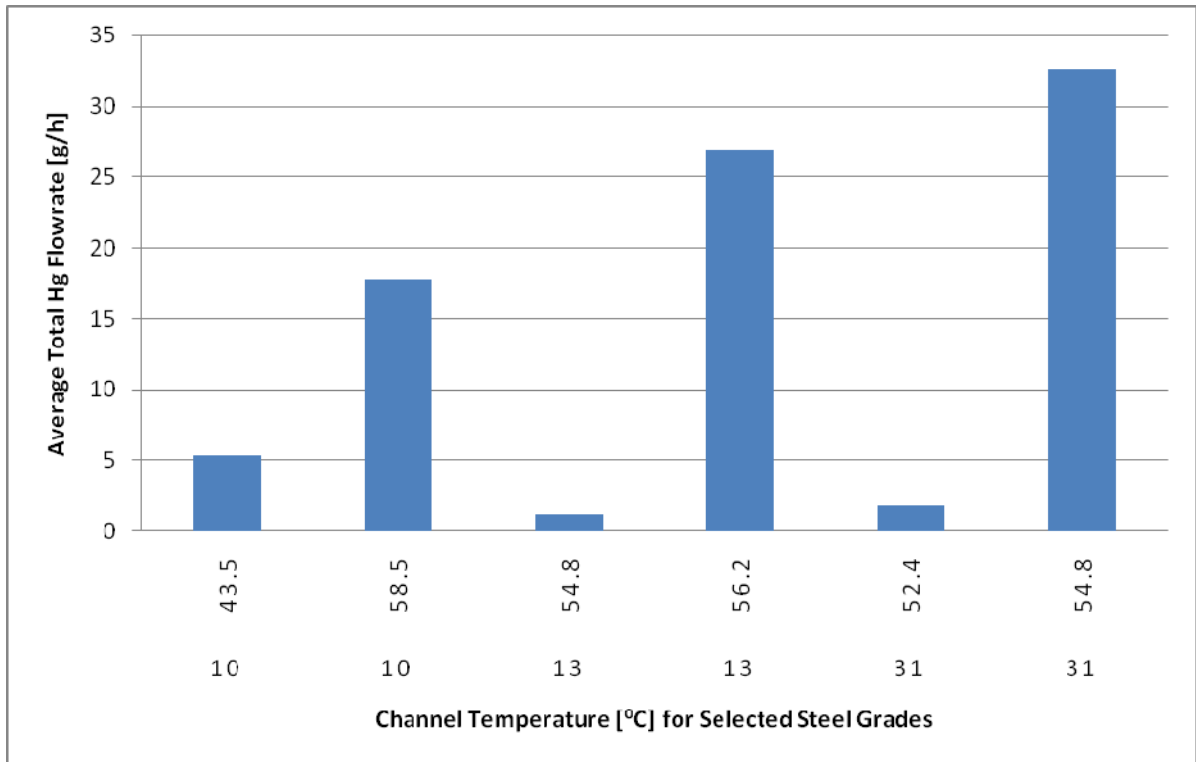


Figure 15. Grades with highest emissions and channel temperature in degrees Celsius are plotted against measured mercury emission flowrate.

Plotting channel temperature against preheated batch shows that high levels of mercury were emitted even without preheating.

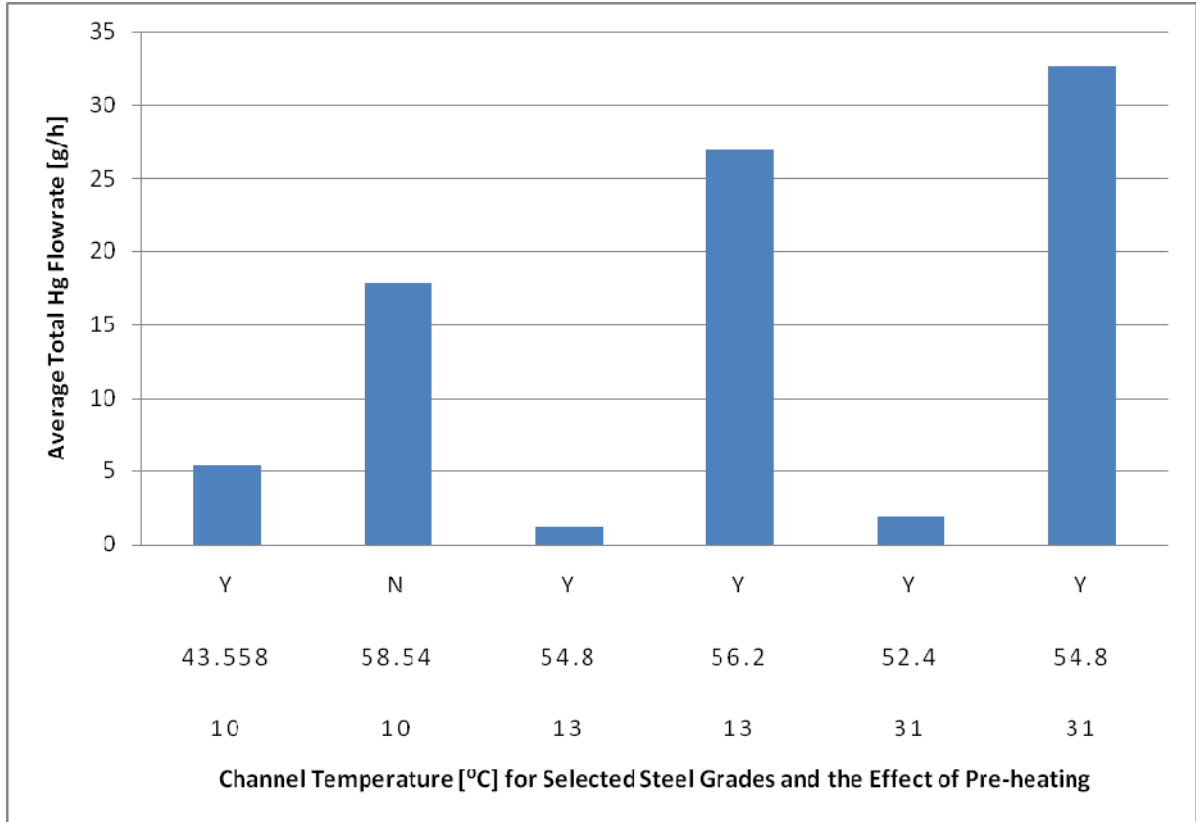


Figure 16. Grades with highest emissions, channel temperature in degrees Celsius and preheating of batch are plotted against measured mercury emission flowrate.

Different additives were compared, but no correlation was found, Figure 17. Here, lime, carbon powder and iron silicate were compared.

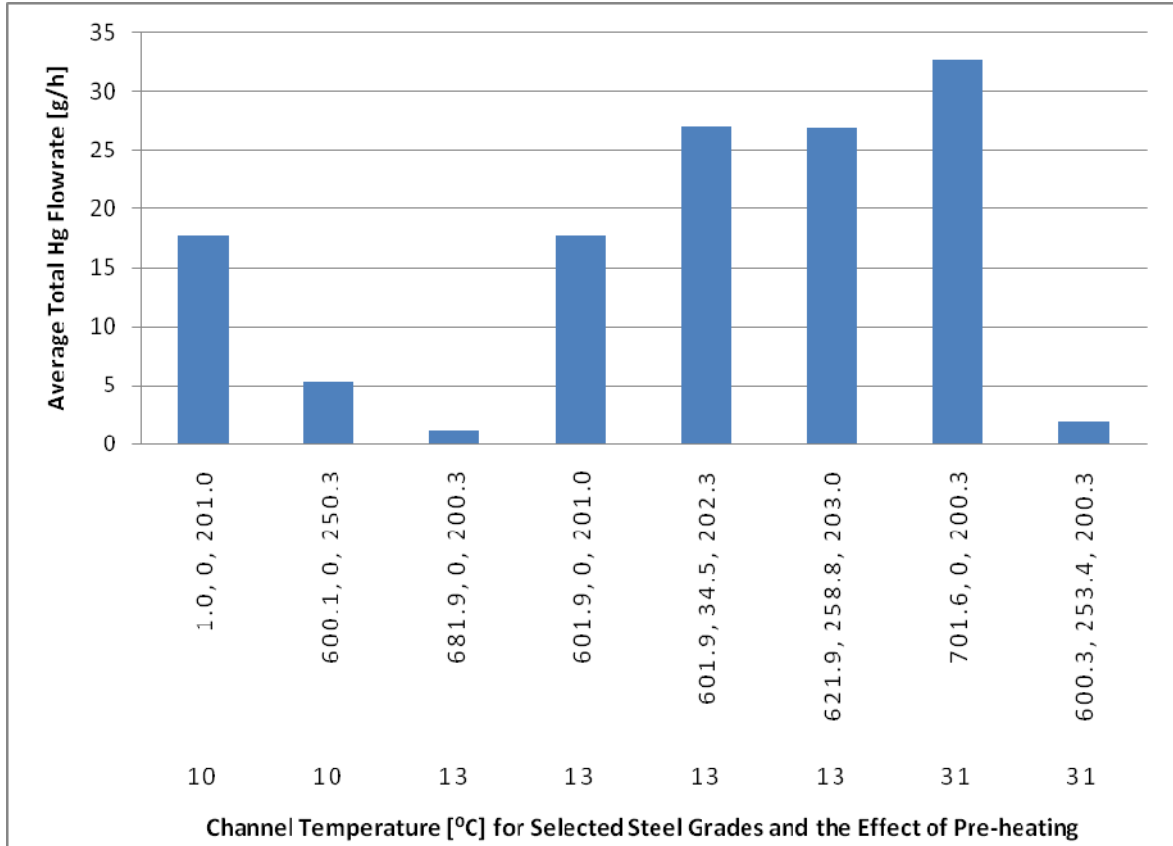


Figure 17. The selected grades with highest emissions were compared with different additive levels (lime, carbon powder and silicate) and plotted against total mercury flowrate.

This process was continued, but unfortunately a correlation was not easily obtainable from the data. An analysis of the mercury emissions closer to the point source would be suggested for the collection of additional data.

7.3 Impact of scrap

An in-depth study of the relationship between scrap metal as a raw material input and mercury emissions from various point sources at the EAF steel plant in Torneå, Finland, has been made without a definitive result [52]. All measurements were conducted with the Semtech Hg 2010 continuous emissions analyzer which measures elemental mercury and mercury (II) converted via a tin chloride solution. It was the hope of the study that peaks in mercury emissions could not only be measured, but also be correlated to specific raw material feeds, and possibly even processes such as heat temperature. In fact, no such relationship was found with scrap metal or specific charges.

A correlation was found with the AOD and the start of the reduction phase for each heat, where material including dolomite is added to the converter. Unfortunately, this finding only adds to the ambiguity of the mercury emission peaks: if they are in fact due to an additive to the process or if they are due to the increased temperature from the heat generated by the exothermic reaction. This correlation was not consistently found in other instances of temperature gradients in the process, where it was examined.

The EAF steel plant in Mo i Rana, Norway has also implemented continuous measurement of mercury emissions in the gas phase. At present, no correlations have been found with specific raw materials.

Despite that both plants have employed continuous mercury emissions monitoring and done extensive plant testing to determine specific contributions, both plants have among the highest overall yearly mercury emissions per ton of finished steel, as compared to other Nordic EAF steel plants. Although it is difficult to find a correlation between emissions of mercury and raw materials, the sorting practices and prohibition of the import and export of mercury have likely contributed to the lower levels of mercury emissions seen in Sweden as compared to other Nordic countries. The non-uniform scrap and scrap sources used in other countries may have a significant impact on the emissions levels.

7 Conclusion

The mercury emissions data from EAF steel plants in the Nordic countries from the last twenty years have been collected where possible and compared. Large variations exist in the data, specifically due to differences in testing and reporting methods. Since the development of online measurement techniques, mercury emissions have been easier to monitor and compare. This effort is very important and should be continued, so that reliable data can continue to be generated and solutions found for emissions of mercury.

The case-study from Outokumpu Stainless AB attempted to correlate specific raw materials to peaks in mercury emissions. However, the variation in the data was too great to be conclusive. Nonetheless, it was seen that the temperature of the gas stream played a small but significant role in the emissions levels that were recorded by the continuous monitor.

The various monitoring methods have been discussed, each with their advantages. A continuous monitor provides more reliable data due to the extended time interval. However, trying to correlate data using a continuous emissions monitor may be deceiving, as the averaged total may be correct, whereas a point method may give more accurate results.

There are many new technologies available for mercury treatment and removal. High chlorine levels in a gas stream can be advantageous for binding elemental mercury at temperatures lower than 440°C. Activated carbon injection is another effective solution for mercury capture. Based on differences in operation techniques and quantities of gas components, specific solutions should be applied.

9 Future work

The measurement of mercury is paramount for controlling the amounts of mercury emitted for the health and safety of communities and the environment. As such, continuous emissions measurements of mercury have been made by several Nordic steel companies, using a Semtech analyzer. The result have clearly demonstrated that mercury is being emitted, and in some cases, in extremely high quantities. However, a lack of correlation is repeatedly found between peaks in mercury emissions and raw material inputs and even process parameters, when measured continuously using a Semtech.

In light of this, it is recommended that other methods for continuous measurements of mercury be examined. A parallel study should be performed using one of more of the techniques discussed previously for measuring mercury continuously. This should be conducted over an extended period of time, such as a week or more. Furthermore, experiments should be conducted using the Semtech Hg 2010 with known amounts of mercury to verify correct quantification of mercury in the gas stream.

It is also recommended that the method of gas capture prior to quantification should be examined. This will clarify whether differences are due to the instrument itself or differences in gas sampling and capture from the gas stream.

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