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Pollution problems in the metallurgical industry: A review

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Abstract

Processing of minerals and production of metals has increased greatly in recent years. As a result, the quantities of waste material and pollutants have also increased. In many cases technology has changed to cope with the problem. Processes have been either modified to decrease emissions, or replaced by others that are less polluting even if at a higher cost. This paper briefly reviews examples in the ferrous and nonferrous industries.

1. Introduction

The mining and metallurgical industries were known to be a source of toxic materials since ancient times. The Romans, for example, used to send convicts to work in mercury mines because it was known that they will die shortly from the air in the mine. Georgius Agricola (1494-1555) the medical doctor in the Renaissance showed in his book De Re Metallica published in 1556 numerous pictures of smelting of ores with extensive fumes being emitted in the work place (Figure 1). Agricola travelled extensively in Saxony and its neighbourhood to visit mines and smelters to examine miners and metal workers.

Nothing was practically done until the first legislation to control obnoxious emissions came into effect in England in 1789 in connection with the chemical industry that became known as Alkali Act. Prior to that time the alkali industry treating salt with H_2SO_4 emitted large amounts of HCl gas in the atmosphere which causes severe destruction to the environment. As a result a method was invented to collect this gas and transform it into a useful product. The process involved the oxidation of HCl by air at 300°C in presence of a catalyst to transform it into chlorine to be used for the bleaching of textiles (Deacon's Process).

At the beginning of this century, the metallurgical industry used to emit waste gases directly at ground level (For example, the roasting of sulfide ores or the making of coke by the beehive method). Later on, stacks were built only high enough to provide adequate draft for furnaces. Operations were usually established in isolated areas. As the scale of operations increased and as lands near industry were inhabited and cultivated by farmers, smoke stacks created problems. Poisonous or irritant gases as well as particles emitted by these stacks, posed serious danger to the vegetation and animal life in their vicinity. Court-ordered shutdowns and compensation to farmers became common.

In the 1950s inhabitants of the industrial town Minamata in Japan suffered many deaths and disease which was attributed to eating fish contaminated with mercury from the nearby chemical factory. This accident alerted public opinion regarding the need to regulate industrial

emissions. In the 1960s the metallurgical industry was severely blamed for its SO_2 emissions. The situation became intolerable and governments were faced with the dilemma when forcing the shutdown of plants. Protect the environment or pay the unemployed as a result of the shutdown. In fact, some companies threatened to shut down if government regulations were so severe. In few cases workers went on strike protesting environmentally unacceptable working conditions in the plant. However, some plants are conscious of environmental problems and do their best to abate pollution even at a high cost. The Scandinavian countries are typical examples.
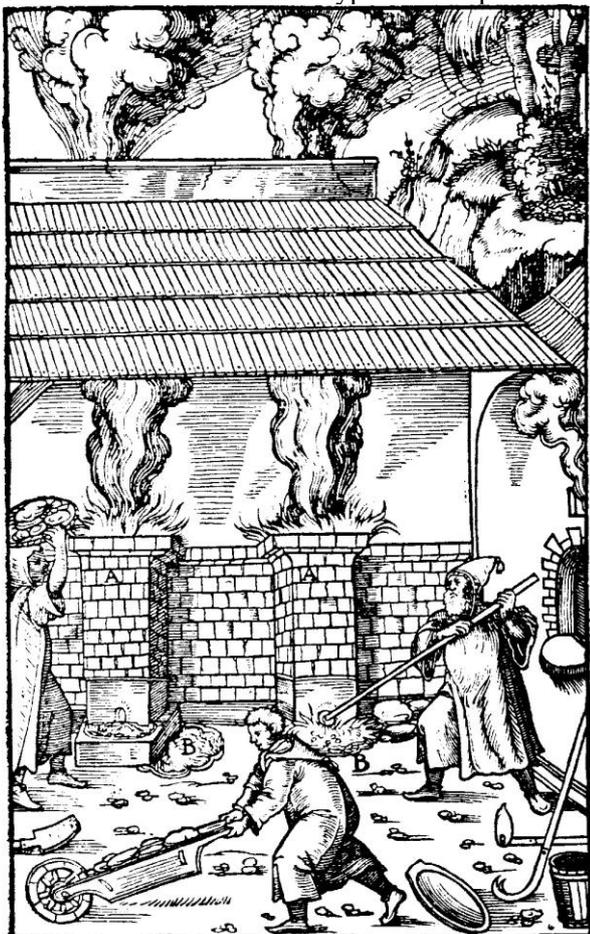

Figure 1. A woodcut from Agricola's book De Re Metallica published in 1556 showing intense fumes being emitted at the workplace

Besides government legislation against pollution, residents in many communities now protest against the erection of industrial plants in their regions. Thus, while at one time, smoking chimneys were a welcome sign of prosperity and meant prestige to a country, it is now considered a disaster area by many. Pollution problems may be evident in some cases such as emission of obnoxious smells or dumping of piles of waste. In most cases, however, the problems are hidden and are revealed only to specialists and this renders the problem very serious.

2. Sources of pollution

Processing of minerals and production of metals has increased greatly in recent years. As a result, the quantities of waste material and pollutants have also increased. The general problems in the mineral and metal industries are outlined below. Figure 2 shows the main emissions and sources of pollution in the mineral processing industry. No doubt that in many cases technology has changed to cope with pollution problems. A polluting process has been either modified to decrease emissions, or replaced by another that is less polluting even if at a higher cost; examples are given later.

2.1. Mining

Pollution in mining is mainly due to the waste rock that is brought to the surface from underground and the overburden removed from open pits. As the solids accumulate, a dump covering many acres is formed (Figure 3). Surface drying of the dump and high winds may result in localized dust storms. An effective way of preventing these storms has been to keep the dumps wet at all times. Today, planting and cultivation of shrubs and trees to act as windbreaks are becoming common.

Explosives used in mining produce NO and NO_2 gases usually denoted as NO_x. A commonly used explosive for fracturing purposes is ammonium nitrate which decomposes as follows:

$$NH_4NO_3 \rightarrow N_2O + 2H_2O \qquad (1)$$

Nitrous oxide produced is oxidized by air to NO2 which contributes to the problem of acid rain. Mining radioactive ores is especially hazardous because of the liberation of radioactive gases during shattering the rocks. Excessive ventilation is necessary in underground mining. Mine water contains residual ammonium ion from the explosives used which may be reduced by microorganisms to NH_3. Both species are toxic to fish.

2.2. Mineral beneficiation

Ores supplied by the mines are usually beneficiated to remove as much as possible of the undesired components known as gangue minerals. This operation is essential to decrease the cost of transportation and increase the value of the concentrate obtained. The minerals must first be liberated from the rock by crushing and grinding and then subjected to a separation process that

makes use of differences in a physical or a physico-chemical property. The waste from this operation is known as tailings.
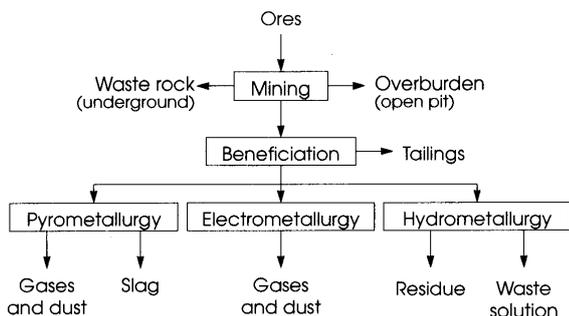

Figure 2. Emissions and waste disposal problems in the mineral industry


Figure 3. Waste rock accumulating from a mining operation

Flotation is a major beneficiation method that makes use of differences in the surface properties of minerals. Organic reagents called collectors are added to the mineral slurry so that they can selectively render certain minerals un-wetted by water and therefore can adhere to air bubbles which float to the surface and are removed as a concentrate in the froth. Collectors are organic compounds containing sulfur, phosphorus, nitrogen, or arsenic. Although their concentration in the slurry is in the parts per million ranges, but because of their toxic nature they must be handled with care.

Tailings from beneficiation processes represent a large disposal problem for the following reasons:

• The presence of pyrite which undergoes aqueous oxidation when exposed to weathering conditions generating sulfuric acid:

$$FeS_2 + H_2O + 7/2O_2 \rightarrow FeSO_4 + H_2SO_4 \quad (2)$$
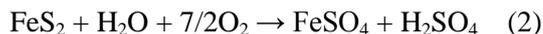
The acid generated will solubilize other minerals thus releasing metal ions in solution.

• The presence of traces of flotation reagents

• Terrains, hundreds of hectares, must be prepared to stock pile this material either dry or under water. Precautions must be taken to avoid breakage of the dams, leaks, seepage to groundwater, etc.

Figure 4 shows a view of a typical tailings pond. Plantation of mining waste and tailings dumps is becoming common to improve the landscape.

2.3. Extractive metallurgy

As shown in Table 1, the major pollution problems in metallurgical plants arise in iron and steel making, in the aluminum industry, in the treatment of nonferrous sulfide ores, in some hydrometallurgical processes, in the treatment of ores containing radioactive elements, in the preparation of certain industrial minerals, and in the electroplating industry. Most of these problems are either solved or can be solved at a high price.

2.3.1. Pyrometallurgy

Pyrometallurgical processing of ores produces dust, slag, and gases. Dust emission may represent a considerable cost to the plant itself because of the loss of particles which are sometimes valuable. The problem of dust has been practically solved by introducing dust-catching equipment such as cyclones, scrubbers, and electrostatic precipitators. Slags are produced in large amounts (Figure 5); only a fraction is used in road construction and in the manufacture of cement.

There is a tendency to install tall stacks to minimize the effect of the irritant or poisonous gases at ground level. Stacks as high as the Empire State Building which is 102 stories high (381 meters) have been constructed (Figure 6); one of these is in a metallurgical plant in Canada to disperse 2500 tonnes of sulfur each day in the form of sulfur dioxide (Figure 7). A stack of this height has a base diameter of about 36 meters; it is made of reinforced concrete 1 meter thick at the base and 26 cm at the top; 1050 tons of steel and 13000 tons of concrete were used in its construction.

A similar but slightly shorter stack (360 meters) was constructed in another metallurgical plant in Utah.

Table 1. Pollution problems in the metallurgical industry

| Industry	Problems
Iron and steelmaking	Gases in coke production, slags, blast furnace, cyanides, electric furnace dust, pickle solution
Ferroalloys production	Arsine and phosphine, silica dust
Aluminum industry	Mercury, red mud, fluorine compounds, toxic organic compounds, cyanides
Sulfide ores: copper, lead, zinc, and nickel	SO_2, mercury, selenium, arsenic
Hydrometallurgical processes: gold, silver, copper, and zinc	Arsine, phosphine, cyanides
Radioactive ores: uranium and thorium	Radon gas (radioactive)
Industrial minerals: coal, phosphate rock, ilmenite, asbestos	Sulfur, ash, trace metals, nitrogen oxides, phosphogypsum, waste acid, toxicity of fibres, tailings
Electroplating industry	Chromium, copper, nickel

Figure 4. A view of a typical tailings pond
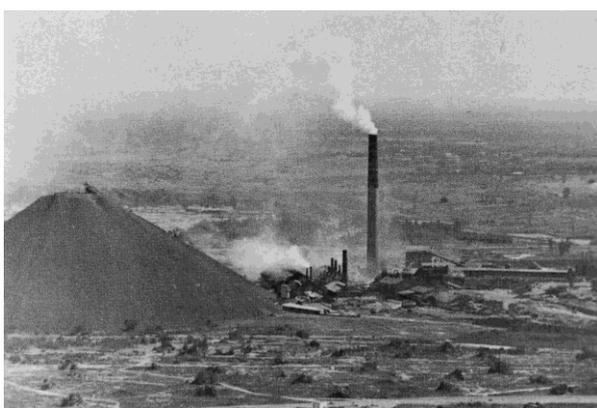
Figure 5. Typical view of a pyrometallurgical plant showing a huge slag pile

Figure 6. INCO chimney is as tall as the Empire State building

Figure 7. INCO'S chimney in Sudbury, Ontario

These stacks are constructed at high costs. They include an inner fibreglass duct, a service elevator, fire-control and water spray systems. Building high stacks to ensure that the poisonous gases are completely dispersed and none can be detected at ground level is not a real solution to the pollution problem because sooner or later these gases will be washed down by rain.

2.3.2. Hydrometallurgy
Treatment of ores by wet methods produces residues and waste solutions. While slags are relatively stable in outside storage, residues filtered off aqueous solutions are not because they usually contain soluble ingredients. Hence, disposal in ponds may be hazardous because of the danger of contaminating surface waters unless they are properly constructed. Also, liquid effluents containing toxic reagents must be treated before being discharged in streams.

2.3.3. Electrometallurgy
The most important electrometallurgical operation is the aluminum industry which emits gases and dust and is presently undergoing intensive improvement. Copper electrorefining and zinc electrowinning have also pollution problems, but these are manageable.

3. Toxicity Hazard
Too much of anything is a poison. In metallurgical plants, a variety of metals, aqueous solutions, gases, dust, molten slags, etc., are produced. A knowledge of the hazard involved and the threshold limit is essential.

3.1. Ingestion and inhalation
Poisoning can occur by ingestion or inhalation. Stringent sanitary measures are advocated and adopted in certain industrial processes to prevent entrance of industrial poisons by mouth. These measures include change of clothing on entering a shift, careful scrubbing of the hands and face before eating, and a thorough washing and shower at the end of the work period. Inhalation is one of the most dangerous routes of entrance of industrial poisons since many gases are colourless and odourless. Certain gases are dangerous if continuously breathed or if breathed in high concentration. In addition, gases may contain noxious fumes or toxic dusts from certain processes.

3.2. Irritant and asphyxiant gases
Some gases cause irritation, e.g., SO_2 causes excessive flow of saliva as well as eye and respiratory irritation. Others, cause asphyxiation, i.e., when present in high concentrations in air they cause death without other significant physiological effect, for example, acetylene. Thus, the limiting tolerable value is the available oxygen. Carbon monoxide causes a special case of asphyxiation (chemical asphyxiation) because it impairs the vascular oxygen transport. The affinity of CO for hæmoglobin exceeds that of oxygen by about three hundred-fold. Thus 0.1% CO in air is enough to kill a person in few minutes by asphyxiation. A person exposed to an asphyxiant for a short time may be saved by immediately exposing him or her to fresh air.

An asphyxiant may contain a trace impurity of a poisonous gas formed during its manufacture. For example, acetylene, C_2H_2, produced from calcium carbide, CaC_2, is poisonous because it contains traces of the dangerous gas phosphine, PH_3.

The presence of phosphine can be traced to small amounts of phosphorus in the raw materials. Calcium carbide is produced on a large scale by heating lime with coke in an electric furnace. It has two major uses: manufacture of acetylene used in cutting metals and in the carburization of molten iron. When it is decomposed by water to manufacture acetylene, phosphine is formed. Thus technical acetylene contains traces of phosphine

and leaks of this gas can be deadly for that reason. Cases of poisoning have been reported in workshops. Even during storage and handling, the carbide reacts with moisture in the air to form phosphine.

4. Role of microorganisms

Certain microorganisms, marine algæ, and fungus play an important role in transforming a less toxic species into a highly toxic derivative. These organisms produce methyl iodide, CH_3I, as a metabolic product which reacts with metals such as mercury, or metalloids such as arsenic, selenium, and tellurium to formtheir methyl derivatives. The process is known as methylation.

5. Threshold limit

The threshold limit is the maximum amount permitted without causing harm. Table 2 gives the threshold limit for gases that may be present in a metallurgical plant. It can be seen that there are great variation in these values; nickel carbonyl, for example, is extremely toxic while carbon monoxide is comparatively much less toxic.

Nickel carbonyl. This colourless and odourless gas is formed in a nickel refining process whereby impure nickel is reacted at high temperature and pressure with CO. The gas is then decomposed at high temperature and atmospheric pressure to form pure nickel and generate CO for recycle. The process is used principally at Sudbury, Ontario on an industrial scale.

Mercury. The vapour pressure of mercury in air saturated with mercury at 20°C is 1.84 ppm and at 40°C is 8.5 ppm. The degree of atmospheric saturation with mercury depends also on the amount of surface exposed: mercury forms a large number of minute droplets when a drop of mercury falls on the floor. They occupy fine cracks and remain unnoticed. When washed through the drain it may form more toxic compounds by the action of microorganisms. Mercury is used in the electrochemical industry and in gold recovery by the amalgamation process (now abandoned).

Arsine. All arsenic compounds are toxic, arsine is particularly toxic because of its gaseous nature. It has a strong garlic odour. This gas is formed whenever hydrogen is produced in presence of arsenic-bearing solutions. Traces of arsenic in scrap, in ores, and in metallurgical residues find their way as arsine under certain conditions.

Fluorine. A colourless gas with a pronounced odour, heavier than air, produced industrially by the fused electrolysis of HF–KF bath at 100°C, highly corrosive. Used mainly to prepare uranium hexafluoride from uranium tetrafluoride for uranium isotopic enrichment for certain types of nuclear reactors as well as for military purposes.

Phosgene. Also known as carbonyl chloride. It is a colourless volatile liquid at temperatures below its boiling point of 8.2°C. Was used as a toxic gas during World War I. It is formed under certain conditions during the chlorination of metal oxides.

Ozone. A colourless gas of peculiar odour, heavier than air. Sometimes used as a powerful oxidizing agent.

Phosphine. Also known as hydrogen phosphide, a colourless gas with a strong odour resembling decayed fish. It may form during the storage of ferroalloys, handling of scrap iron in acid medium due to the presence of traces of phosphorus in the raw materials.

Arsenic oxide. Arsenic forms two oxides: the trivalent and the pentavalent. Only the trivalent oxide, As_2O_3, is volatile and therefore is dangerous. It is formed during the oxidation of arsenic-containing sulfide minerals under limited supply of air. It is collected as a white dust in the gas filtration system and is usually disposed of by storage in steel barrels. Small amounts are used as insecticide and weed killer. It is soluble in water and therefore its storage should be carefully monitored.

Chlorine. A greenish yellow gas with a pungent, irritating odour, heavier than air. It is used to produce chlorides from ores and concentrates, e.g., production of $TiCl_4$ from rutile, and $ZrCl_4$ from zircon.

Sulfur chlorides. Sulfur monochloride is a yellow liquid that boils at 138 °C while sulfur dichloride, SCl_2, is a red brown liquid that boils at 59°C. Both are formed during the treatment of sulfide concentrates with chlorine. However, no metallurgical processes are operating using this technology.

Hydrogen fluoride. Hydrofluoric acid is a fuming liquid that boils at 19.5°C. It is prepared commercially by the action of concentrated H_2SO_4 on fluorspar, CaF_2. The commercial acid is a solution of hydrogen fluoride in water. The gas is colourless, has a penetrating odour, and is lighter than air. The gas used in the fluorination of oxides, e.g., BeO and UO_2 to prepare the corresponding fluorides.

Table 2. Threshold limit values of toxic gases found in metallurgical plants in decreasing order of toxicity

Gas	Formula	Threshold limit value in air		Remarks
		ppm	mg/m^3	
Nickel carbonyl	$Ni(CO)_4$	0.001	0.007	Toxic
Mercury	Hg	—	0.1	Toxic
Arsine	AsH_3	0.05	0.2	Toxic
Fluorine	F_2	0.1	0.2	Irritant
Phosgene	$COCl_2$	0.1	0.4	Toxic
Ozone	O_3	0.3	0.4	Toxic
Phosphine	PH_3	0.1	0.2	Toxic
Arsenic oxide	As_2O_3	—	0.5	Toxic
Chlorine	Cl_2	1	3	Toxic
Sulfur chloride	S_2Cl_2	1	6	Toxic
Hydrogen fluoride	HF	3	2	Toxic
Hydrogen chloride	HCl	5	7	—
Nitrogen dioxide	NO_2	5	9	—
Sulfur dioxide	SO_2	5	13	Irritant
Hydrogen cyanide	HCN	10	11	Toxic
Hydrogen sulfide	H_2S	10	15	Toxic
Cyanogen	C_2N_2	10	—	Toxic
Carbon disulfide	CS_2	20	60	Toxic
Nitric oxide	NO	25	30	Toxic
Ammonia	NH_3	50	35	Irritant
Carbon monoxide	CO	50	55	Toxic
Carbon dioxide	CO_2	5 000	9 000	—
Acetylene	C_2H_2	—	—	Asphyxiant
Uranium hexafluoride	UF_6	—	—	Toxic

Hydrogen chloride. A colourless gas that forms dense white fumes when exposed to air due to reaction with atmospheric moisture and the formation of hydrochloric acid droplets. Hydrogen chloride is seldom used in metallurgy but forms when gaseous chlorides leak from pipes and equipment and react with moisture in the air, e.g., $TiCl_4$.

Nitrogen dioxide. An orange-red gas at room temperature forms when nitric acid reacts with minerals.

Sulfur dioxide. A colourless gas possessing a characteristic pungent and irritating odour, causing increased generation of saliva. It is generated in metallurgical plants oxidizing or melting sulfide ores and concentrates of copper, lead, zinc, nickel, and mercury or refractory gold ores which are usually pyritic and treated by thermal route.

Hydrogen cyanide. A colourless gas, having an odour resembling that of bitter almonds and is lighter than air. It may form in a hydrometallurgical plant treating gold ores by the cyanidation process as a result of negligence. The gas forms only when the alkalinity of the solutions decreases below pH 10. This may happen due to absorption of CO_2 from the air by the alkaline solution.

Hydrogen sulfide. A colourless gas that has the odour of putrefied eggs. It occurs in natural gas, sometimes in high concentrations, and must be removed before using the gas. It is used in hydrometallurgy to precipitate copper, nickel, and cobalt from solution.

Cyanogen. A colourless gas of distinctive odour that forms when hot air is in contact with carbon under reducing conditions, e.g., in the iron blast furnace. It is soluble in water, therefore, when the

blast furnace gases are scrubbed with water, cyanogen will contaminate the water.

Carbon disulfide. A colourless liquid that boils at 46.2°C has unpleasant odour, used as solvent for elemental sulfur. It forms in small amounts when SO_2 is reduced with carbon at high temperature.

Nitric oxide. A colourless gas that reacts readily with oxygen of the air to form the brown red coloured gas NO_2. It forms in trace amounts whenever a carbonaceous fuel is burnt with air, thus it will be present in all flue gases.

Ammonia. A colourless gas with a pungent odour, highly soluble in water, can form aqueous solutions containing up to 29 % NH_3. Used in hydrometallurgy for leaching sulfides of copper, nickel, and cobalt with which it forms the ammine complexes.

Carbon monoxide. A colourless gas without odour, slightly lighter than air, used as a reducing agent. It is a major component of blast furnace gas and producer gas. The toxic effect of CO is due to decreased oxygen content of the blood because it combines with hæmoglobin to form a stable CO-hæmoglobin complex which prevents oxygen supply to those tissues where oxygen is essential for normal body functions. The oxygen content in blood does not go back to normal as soon as fresh air is inhaled but slowly diminishes as CO is eliminated from blood. Carbon monoxide is not oxidized to CO_2 in the body and is gradually eliminated from the lungs as fresh air is inhaled.

Carbon dioxide. A colourless, odourless gas, with a sharp taste, heavier than air. It is the waste product of combustion of carbonaceous fuels.

Acetylene. A colourless gas, lighter than air, produced from petroleum fractions or from calcium carbide. In the latter case it may be contaminated with arsenic and phosphorus compounds that result in the generation of arsine and phosphine when the calcium carbide is treated with water to form acetylene. The oxyacetylene flame is used for the autogenous welding of metals.

6. Toxicity of metals

The famous alchemist Paracelsus (1493–1541) already recognized that the right dose differentiates a poison and a remedy. Some metals in trace amounts in aqueous solution are essential to the human body, e.g., copper, cobalt, selenium, and manganese. These are generally widespread in foods in trace amounts, and some like cobalt is a component of vitamin B12. Some metals are even used as medicine, e.g., arsenic in Salvarsan and mercury in mercurochrome. However,

amounts in excess to the optimum amount leads to disorder and poisoning. Other metals like the alkali metals, alkaline earth metals, aluminum, and iron are not toxic. Compounds of most other metals are toxic, some of them even in extremely small amounts. Solubility of a compound in water or in body fluids renders it more toxic than insoluble compounds.

Toxicity cannot be evaluated with the ease with which a physical constant such the melting point or the boiling point of a substance may be determined. Solid, massive metals are not toxic, but their vapours are. Vapours may be generated during melting, distillation, and welding. Compounds in the gaseous state are toxic; some are more than others. For example, metallic beryllium pieces are not toxic, but vapours of BeCl2 are highly poisonous as well as vapour of metallic beryllium. Mercury in the metallic state is an especially toxic material. Being liquid at room temperature, it has a high surface tension and when spilled on the ground it forms a large number of extremely small globules, thus high surface area and increased vaporization. A good knowledge of the toxicity of the material being handled is necessary for a metallurgist. For example, handling nickel sulfate or borax does not require any special precaution, but handling nickel carbonyl or boranes (organic boron compounds) requires extensive precautions because of their high toxicity.

7. Dust, fumes, smoke, and aerosols

Dusts are particles or aggregates of particles 1 to 150 microns in diameter, fumes 0.2 to 1, and smokes are less than 0.2 microns. Dust is usually formed as a result of mechanical attrition but fumes and smoke are formed as a result of chemical reaction. Excessive inhalation of mineral dusts causes lung problems. For example, silica causes silicosis, fluorides causes' fluorosis, and others. These cause hardening of parts of the lungs which necessitates surgery. Some dusts, however, like asbestos cause's cancer because the danger is not limited to one region in the lungs — it spreads in other parts of the body. Fumes and smoke are more dangerous because of their small particle size and the ease with which they can enter the respiratory system. Aerosols are produces by condensation of vapours or generated mechanically when a gas escapes from an aqueous solution. The gas bubbles burst on leaving the solutions dispersing fine droplets in the environment. For example, during the electrowinning of zinc from zinc sulfate solution,

oxygen is generated at the anode. During its escape it becomes associated with fine droplets of acid solution that is known as "acid mist". This renders the work place intolerable and causes damage to the equipment. This problem, however, has been totally solved by laying plastic balls or a surface active agent on the top of the electrolyte to permit oxygen escape without acid droplets.

8. Explosion and Fire Hazard

Explosions may take place in a metallurgical plant when handling gases, vapours, and molten materials. Molten material such as molten salts, slags, mattes, and metals are susceptible to explode when improperly handled. Usually contact with water is the source of explosion. While it is safe to pour molten slag or molten metal into water for cooling and granulation purposes, the reverse is not true. Molten mattes are dangerous to contact with water because of the formation of H_2S which is explosive.

Methane and natural gas are often a cause of explosion in underground coal mines. Natural gas may also be present in other mines. Dust particles susceptible to oxidation, e.g., aluminum or iron powders may catch fire or explode due to violent oxidation. Mines where sulfide minerals are being exploited may catch fire when fine sulfides are accumulated in presence of humidity. Certain microorganisms accelerate the oxidation reaction and result in generation of enough heat that the bed may ignite.

9. Recycling and Conservation

Recycling of metal scrap not only conserves the natural resources but also decreases pollution. Table 3 shows the energy savings from the recycling of some metals.

Table 3. Energy savings from recycling metals

Metal	%
Aluminum	95
Copper	85
Steel	74
Lead	65
Zinc	60

10. Conclusions

Industry is now doing its best to improve the working conditions, abate pollution, and safegauarde the environment. The philosophy changed from "making profit" to "be in business". To be in business means that the industry gets involved with the community, increases the level of awareness of her employees, and avoids accidents. Building tall stacks seems to be not the best solution to avoid pollution while adding scrubbers seems to be a better solution. An example of a lead smelter in Mexico shows that emissions decreased after adding scrubbers and this results in decreased lead content in the neighborhood of the smelter, and decreased lead content in the workers' blood (Figures 8-10).

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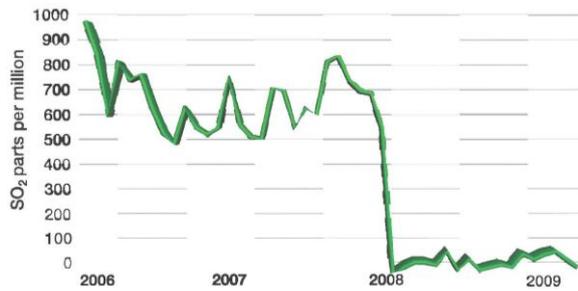
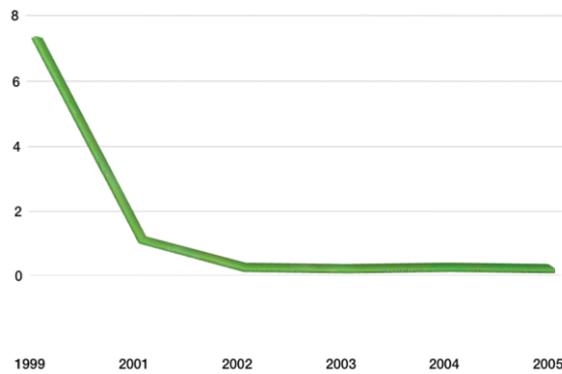**Figure 9. Micrograms of lead / M3 in surrounding air of smelter [1]**

Figure 8. SO2 emission before and after installing scrubbers [1]

Figure 10. Micrograms of lead per deci-Liter of workers' blood [1]

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