

## **NATIONAL INVENTORY OF SOURCES AND EMISSIONS OF FLUORIDE (1972)\***

Pollution Data Analysis Division  
Air Pollution Programs Branch  
Air Pollution Control Directorate

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## ABSTRACT

Atmospheric emissions of fluoride from various Canadian sources have been estimated for the year 1972. The data presented in this report are summarized in Table 1 and Figures 1 and 2. Total emissions of fluoride to the atmosphere are estimated at 15 644 tons. The largest contributor is the primary aluminum industry, which accounts for 57% of the total. Emissions from phosphate fertilizer and elemental phosphorus plants account for a further 17% and the primary iron and steel industry for 16%. On a geographic basis the Province of Quebec accounts for 48% of the Canadian total followed by Ontario with 18%. It is estimated that about 70% of nationwide fluoride emissions, close to 11 000 tons, are in the gaseous state.

Results are first approximations due to paucity of data. The inventory serves to place the various emission sources in perspective and the reader is cautioned not to use the emission estimates out of context of this inventory.

## RÉSUMÉ

Les émissions atmosphériques de fluorure par les différentes sources canadiennes ont été évaluées pour l'année 1972. Le tableau 1 et les figures 1 et 2 présentent un résumé des données du présent rapport. On estime que les émissions atmosphériques totales de fluorure atteignent 15 644 tonnes; 57% de ces émissions sont attribuables à l'industrie de l'aluminium de première fusion, 17% des fabriques d'engrais phosphatés et de phosphore élémentaire et 16%, de l'industrie du fer et de l'acier de première fusion. Du point de vue géographique, la province de Québec fournit 48% du total canadien et l'Ontario, 18%. On estime qu'environ 70% des émissions de fluorure à l'échelle nationale, soit près de 11 000 tonnes, sont à l'état gazeux.

En raison du peu de données disponibles les résultats sont des premières approximations. Le présent rapport est un inventaire permettant de mettre en évidence les différentes sources d'émission, et le lecteur est mis en garde contre l'utilisation hors contexte des données estimatives.

TABLE 1 FLUORIDE EMISSIONS BY PROVINCE AND SECTOR, 1972 (tons)

Province												
Sector	B. C.	Alta.	Sask.	Man.	Ont.	Que.	Nfld.	Maritimes	N. W. T. Yukon	Total Canada	% Total	Gaseous fluoride (% total)
INDUSTRY												
Primary aluminum production	1975	-	-	-	-	6877	-	-	-	8852	56.6	55
Phosphate fertilizer and elemental phosphorus plants	394	912	-	-	628	373	146	215	-	2668	17.1	>96
Primary iron and steel production	13	38	26	53	1253	198	-	837	-	2418	15.5	80-85
Miscellaneous sources	24	77	9	4	295	72	11	42	-	534	3.4	70-75
FUEL COMBUSTION/ STATIONARY SOURCES												
Power generation	-	272	142	27	503	-	-	62	-	1006	6.4	>90
Industrial and commercial	6	17	9	19	66	32	<1	13	<1	162	1.0	>90
SOLID WASTE INCINERATION	-	-	<1	<1	2	1	-	<1	-	4	<0.1	>90
TOTAL	2412	1316	186	103	2747	7553	157	1169	<1	15 644		70
% TOTAL	15.4	8.4	1.2	0.7	17.5	48.3	1.0	7.5	<0.1		100.0	

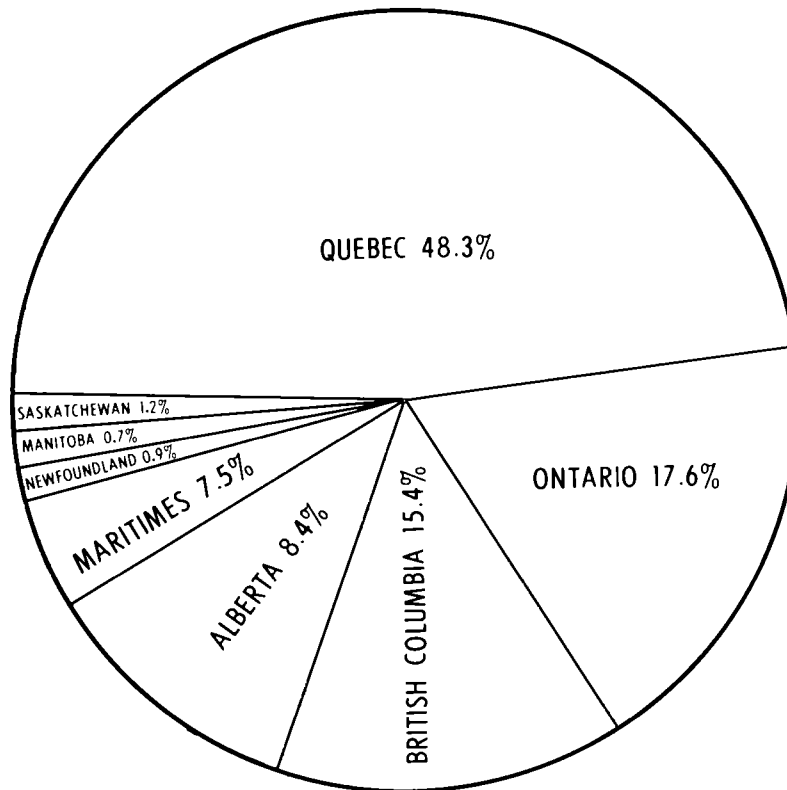


FIGURE 1 PERCENT FLUORIDE EMITTED BY PROVINCE, 1972

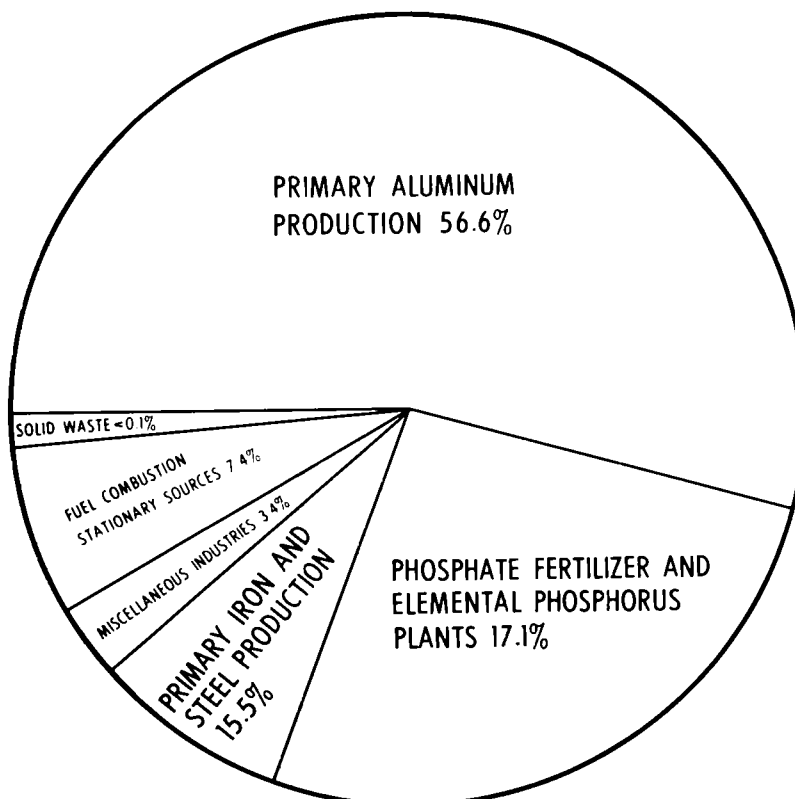


FIGURE 2 PERCENT FLUORIDE EMITTED BY SOURCE, 1972

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## **1 INTRODUCTION**

### **1.1 Scope**

The purpose of this report is to identify and quantify sources of atmospheric emissions of fluoride in Canada for the year 1972. Information contained in this report was obtained from a literature survey, a computer search conducted at the National Science Library, and from previous inventory studies. Information was also gathered from a specifically designed questionnaire which was sent to approximately 1050 plants and received a response of 51%. Details on response rate and quality of returns are given in Appendix . As far as possible emission estimates were based on source testing data applicable to Canadian conditions.

The term fluoride and the symbol F in this report refer to all chemical compounds containing the element fluorine. The term fluorides is occasionally used when referring to several different fluoride salts.

## **2 SOURCES AND USES OF FLUORIDE**

Fluoride is present in trace amounts in soils and rocks, but is most prevalent in active or inactive volcanic regions. It is thirteenth in order of abundance in the earth's crust. The main minerals containing fluoride are fluorspar, cryolite and phosphate rock.

### **2.1 Fluorspar**

All the fluorspar produced in Canada in 1972 was from mines, owned by Alcan, located in the Burin Peninsula in Newfoundland. They produced approximately 167 700 tons in 1972. An additional 71 910 tons of fluorspar was imported (1).

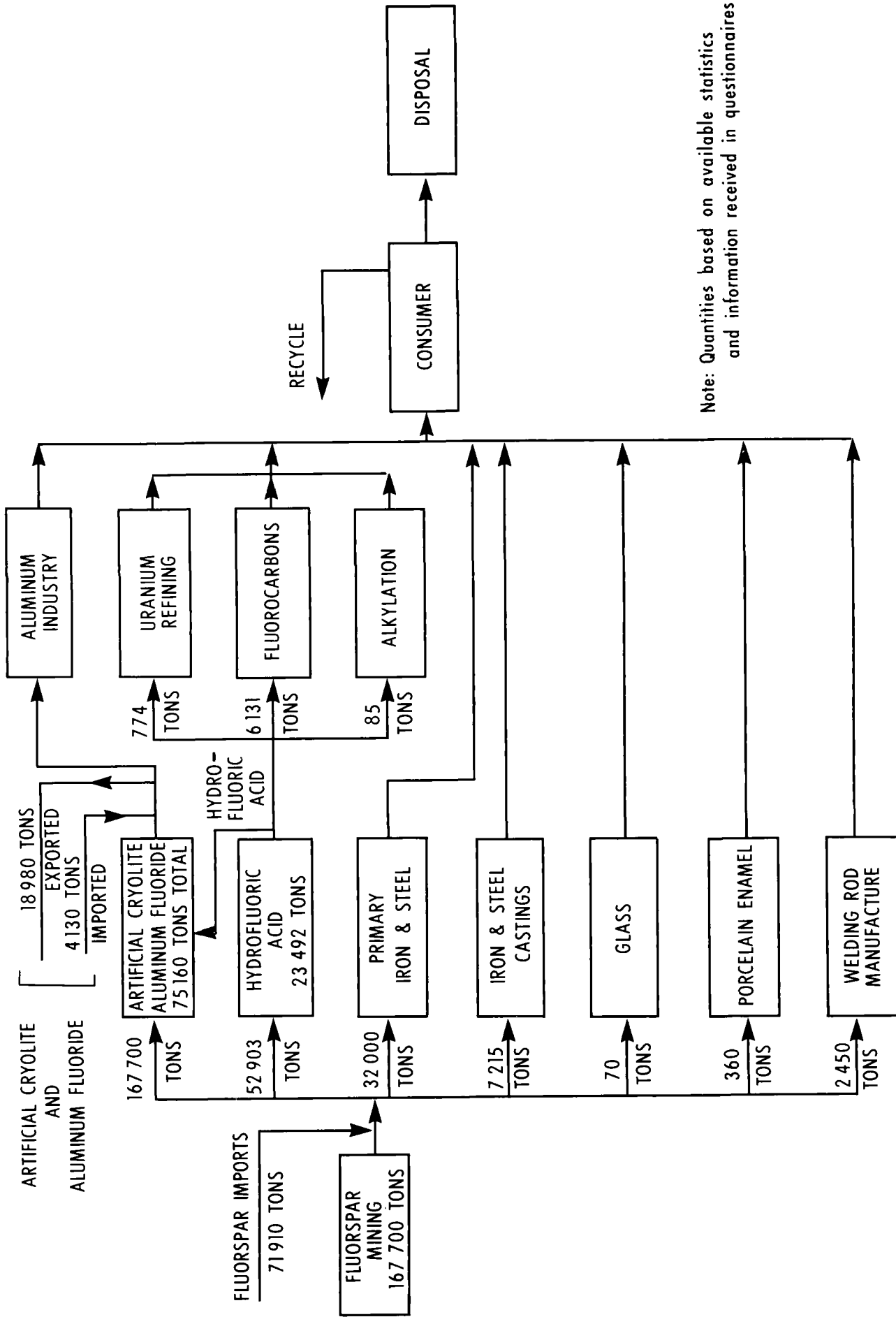
The most important uses of fluorspar are:

- for the manufacture of artificial cryolite and aluminum fluoride, essential ingredients in the electrolytic reduction of alumina to aluminum;
- as a fluxing agent in various metallurgical processes, the most important being steel manufacture;
- for the manufacture of hydrofluoric acid and other fluorine chemicals;
- in the refining of uranium ores;
- and in the glass and ceramics industry.

Figure 3 is a material flow sheet identifying the sources and uses of fluorspar.

### **2.2 Cryolite and Aluminum Fluoride**

The only commercial source of cryolite is in South Greenland. The high price of natural cryolite has led to the production of artificial cryolite which is now generally used in aluminum manufacture. Alcan manufactures all the cryolite and aluminum fluoride in Canada at their plant in



Note: Quantities based on available statistics and information received in questionnaires

FIGURE 3 SOURCES AND USES OF FLUORSPAR, 1972

Arvida, Quebec. The quantities produced in 1972 were 19 160 tons and 56 000 tons respectively. Approximately 4130 tons of cryolite were imported in 1972.

## 2.3 Phosphate Rock

Phosphate rock is not produced commercially in Canada but large quantities are imported (over 4 million tons in 1972), primarily from the United States, for use in the manufacture of agricultural and industrial phosphate products. The fluoride content of phosphate rock averages about 3% by weight. In order to be suitable for use as a fertilizer or as livestock feed supplement, phosphate rock must be freed from most of its fluorine.

## 2.4 Fluorine Chemicals

In 1972, world consumption of fluorspar reached an estimated 5 million tons. The manufacture of hydrofluoric acid accounted for over 50% of the world's fluorspar requirements. In Canada, hydrofluoric acid production was about 24 000 tons in 1972. One of the important uses of this acid is in the production of artificial cryolite by the aluminum industry (1).

About 30% of the hydrofluoric acid is used to manufacture fluorocarbons. Fluorocarbons are produced by reacting hydrofluoric acid with carbon tetrachloride or chloroform and are used in the manufacture of solvents, resins, plastics, films, refrigerants and aerosol propellants (1).

Another major use of hydrofluoric acid is in uranium refining. Uranium dioxide is reacted with anhydrous hydrofluoric acid to form uranium tetrafluoride ( $UF_4$ ), which is then reacted with elemental fluorine in the form of fluorine gas to form uranium hexafluoride ( $UF_6$ ).

# 3 INDUSTRIAL EMISSIONS

## 3.1 Primary Aluminum Production

Two companies produced approximately 1.0 million tons of aluminum in 1972 at six plant locations. Both companies answered the questionnaire providing accurate emission information for all six plants.

**3.1.1 Process Description.** All of the aluminum in Canada is produced by the Hall-Héroult process. The fundamental principle of the process is the electrolytic dissociation of alumina ( $Al_2O_3$ ) dissolved in a molten bath composed of cryolite ( $Na_3AlF_6$ ) and various salt additives. The electrolysis is performed in a carbon crucible, housed in a steel shell, using the crucible as a cathode, and using carbon anodes.

The electrodes of the electrolysis cell consist of a mixture of petroleum coke and pitch. The prebaked anode cell employs a replaceable, consumable carbon anode formed by baking prior to its use in the cell. Soderberg cells employ continuously formed consumable carbon anodes where the anode paste is baked by the energy of the reduction cell itself. Aluminum metal forms at the cathode and oxygen

gas at the anode where it is released, burning the anode. Molten aluminum is drawn off periodically from the bottom of the cell. The electrolysis cell operates continuously and fresh alumina is added when necessary.

**3.1.2 Emissions.** Fluoride is emitted from the cells due to:

- the high temperatures involved which volatilize some of the fluoride;
- the production of hydrofluoric acid vapours in the cell;
- silicon fluoride and particulate matter containing fluoride entrained in gases produced by the reactions.

Hoods over the electrolysis cells collect the gases which are usually scrubbed before being discharged to the atmosphere; however, some of the gases escape into the cell room and are discharged to the atmosphere through roof ventilators. None of the plants have control equipment on the roof ventilators and these emissions, based on questionnaire replies, account for approximately 66% of the fluoride emissions from the primary aluminum industry.

The uncontrolled emission factor for aluminum plants averages 41 lb fluoride/ton of aluminum. All the plants except one have scrubbers on the cell hood stacks. The control efficiency varies from 40%-85% for solid fluoride emissions and from 75%-99% for gaseous fluoride emissions.

Most of the companies surveyed based their emission estimates on test data and material balance calculations.

Uncontrolled emission factors reported by the United States Environmental Protection Agency are 26.6 lb fluoride/ton of aluminum for gaseous fluoride and 15.6 lb fluoride/ton of aluminum for particulate fluoride (2). The average uncontrolled emission factors for this survey are estimated at 28.1 lb fluoride/ton of aluminum for gaseous fluoride and 13.2 lb fluoride/ton of aluminum for particulate fluoride.

A National Research Council report (3) gives a particulate emission factor for an aluminum plant in Switzerland as 9 lb fluoride/ton of aluminum. The same report gives an emission factor for total fluorides from an aluminum plant in Norway as 40 lb fluoride/ton of aluminum; however, no indication is given of whether the emission factors are controlled or uncontrolled.

Gaseous fluoride emissions were reported as hydrogen fluoride and silicon fluoride. Particulates were reported as aluminum fluoride and sodium fluoride.

Emissions of gaseous fluoride from primary aluminum production in Canada in 1972 were 4867 tons (55% of total) and particulate fluoride emissions were 3985 tons (45% of total), a total fluoride emission of 8852 tons.

**3.1.3 Particle Size Distribution.** No information was received on particle size distribution.

### 3.2 Phosphate Fertilizer and Elemental Phosphorus Plants

In 1972 there were nine companies in Canada producing phosphate fertilizers from a total of 14 plants. Elemental phosphorus was produced at two of these plants owned by one company.

It is estimated that over 3.4 million tons of phosphate rock were used in 1972 for phosphate fertilizer and elemental phosphorus production. Based on questionnaire response and on estimates for plants not reporting, the breakdown of phosphate rock consumption is thought to be:

Phosphate rock used (tons)	Fertilizer produced (tons)	
2 349 090	1 510 917	Ammonium Phosphate
361 510	587 898	Single Superphosphate
177 700	428 147	Triple Superphosphate
545 000	54 500	Elemental Phosphorus
<hr/>		
Total	3 433 300	

At one plant 289 120 tons of phosphate rock were used to produce phosphoric acid prior to manufacturing single and triple superphosphate.

**3.2.1 Process Description.** Phosphate fertilizers are produced by decomposing phosphate rock with a strong mineral acid. In Canada, only sulphuric and phosphoric acids are used commercially, sulphuric being the most common (4).

When phosphate rock is treated with sulphuric acid, either single superphosphate or phosphoric acid (orthophosphoric acid) is produced. For the former, the rock is treated with sufficient acid to convert tricalcium phosphate into water soluble monocalcium phosphate (4). To produce phosphoric acid, larger quantities of sulphuric acid are added to the phosphate rock to maintain a fluid slurry that facilitates removal of calcium sulphate by filtration.

Off-stream phosphoric acid, containing 30%-32%  $P_2O_5$  equivalent, may be used either directly in the manufacture of phosphate fertilizers or concentrated by evaporation to as high as 54%  $P_2O_5$  equivalent (4). Most of the acid is then neutralized with ammonia to form ammonium phosphate fertilizers. At some plants phosphoric acid is used to acidulate phosphate rock, in which case the end product is triple superphosphate.

Elemental phosphorus is produced in Canada by the thermal reduction method which involves the smelting of phosphate rock with carbon (coke) and siliceous flux. Coproducts of the process are ferrophosphorus, carbon monoxide and calcium silicate slag.

**3.2.2 Emissions.** Emissions from the various types of plants are broadly classified as follows:

**Single Superphosphate.** Plants producing this type of fertilizer have different equipment for the acid/phosphate rock reaction. One has a batch operation; the others use a continuous operation.

Single superphosphate fertilizer is produced in Canada by either of two processes: the Oberphos process and the Den process. The latter is the common way to manufacture both single and triple superphosphate; here curing dens are used to enable the reaction to continue with the liberation of hydrogen fluoride. If uncontrolled these dens may be a major source of fluoride emissions. The Oberphos process does not make use of curing dens and is used to only a limited extent.

Emissions of fluoride occur mainly from the acid/phosphate rock reaction tank vents and from the curing dens. Small amounts of particulate fluoride emissions also occur from the phosphate rock grinding operation.

In some cases emissions were reported for the rock grinding and reaction vents, but not the curing dens. The curing period is 15-60 days and fluoride emissions occur from such locations as the windows, doors and cracks of the curing den. Emissions from this particular source are, therefore, difficult to evaluate. Because information was lacking, the estimates for this source were based on published data (5).

Emissions from reaction tanks are usually controlled by wet scrubbers with 95% efficiency. Curing dens were estimated to be controlled at about 90% efficiency by venturi scrubbers.

The uncontrolled emission factors estimated for single superphosphate manufacturers using curing dens are given in Table 2. Overall, single superphosphate manufacturers are estimated to account for 255 tons or 9.5% of fluoride emissions from the phosphate fertilizer and elemental phosphorus industry.

*Triple Superphosphate.* Two plants in Canada manufactured triple superphosphate in 1972. Based on information obtained from the survey it was possible to calculate appropriate emission factors for the manufacturing of this type of fertilizer. These factors are shown in Table 2.

Scrubbers with 95% control efficiency are usually installed on the reaction tank vents, conveying, granulating and drying steps. The curing dens are assumed to be controlled by scrubbers at an average rate of 90%.

Triple superphosphate producers are estimated to account for 290 tons or 10.9% of total fluoride emissions from the phosphate fertilizer and elemental phosphorus industry.

*Phosphoric Acid and Ammonium Phosphate.* The first step in the production of ammonium phosphate fertilizers is the production of phosphoric acid. Data reported in the questionnaires enabled an emission factor to be calculated for this process step which involves the phosphate rock/acid reaction tank vents. Emissions also occur from the ammonia/phosphoric acid reaction tank stacks and granulation dryers. Emission factors for both process steps are given in Table 2.

The most important fluoride emission source is the gypsum pond, which is dealt with separately below.

Emissions from phosphoric acid production, including those from gypsum ponds, are estimated to be 1608 tons or 60.3% of the total for phosphate fertilizer and elemental phosphorus plants.

TABLE 2 UNCONTROLLED EMISSION FACTORS FOR PHOSPHATE FERTILIZER AND ELEMENTAL PHOSPHORUS PLANTS

Process	This survey		EPA (5)
	lb F/ton phosphate rock	lb F/ton phosphate in product	lb F/ton phosphate in product
Wet process phosphoric acid			
Grinding	0.71	2.19	
Process	0.90	2.70	0.93 <sup>a</sup>
Gypsum pond	1.05 (5)	3.14 (5)	3.14
Ammonium phosphate			
Process	1.72	5.33	1.30
Gypsum pond	0.003 (5)	0.01 (5)	0.01
Triple superphosphate			
Grinding	0.71	0.64	
Process	6.98	6.28	21.0 <sup>b</sup>
Curing	23.6	21.24	
Gypsum pond	0.22 (5)	0.2 (5)	0.2
Single superphosphate			
Grinding	0.71	2.19	
Process <sup>b</sup>	23.05 (5)	71 (5)	71 <sup>b</sup>
Elemental phosphorus			
Grinding	0.71	2.29*	
Process	3.81	12.31*	30 <sup>a</sup>

\* lb F/ton  $P_2O_5$  in phosphate rock feed

<sup>a</sup> presumed to include grinding

<sup>b</sup> presumed to include grinding and curing



Emissions from ammonium phosphate production (including gypsum ponds) are estimated to account for 235 tons or 8.8% of the same total.

From data reported in the questionnaires, the average degree of control is 95% for phosphoric acid production and 90% for ammonium phosphate manufacturing, excluding the gypsum ponds which are usually uncontrolled. The controls used are wet scrubbers.

*Elemental Phosphorus.* Fluoride emissions occur from the pelletizers, dryers and furnaces usually as gaseous hydrogen fluoride and silicon fluoride. Appropriate emission factors are given in Table 2. Emissions from this source are estimated to account for 280 tons or 10.5% of total emissions from the phosphate fertilizer and elemental phosphorus industry. The degree of control varies from almost nil for one plant to about 95% for the other. When controls are applied, they are usually wet scrubbers.

*Grinding.* Not all fertilizer producing plants grind the phosphate rock prior to processing. From data reported in the questionnaires, an overall uncontrolled emission factor was calculated and is shown in Table 2. It is assumed that companies not reporting have grinding operations controlled by baghouses with 95% efficiency.

*Gypsum Ponds.* The major source of fluoride emissions from phosphate fertilizer manufacturing is the gypsum pond. Gypsum ponds are used for phosphoric acid, ammonium phosphate and triple superphosphate production. Losses from the gypsum pond depend primarily on pH. Although emissions of hydrogen fluoride can be virtually eliminated by the addition of lime to raise the pH, this is a costly practice which does not seem to be used in any plant in Canada. Emissions from gypsum ponds are therefore totally uncontrolled. Questionnaire data are not sufficient to enable calculation of an emission factor, which is consequently based on published data (5). This is given in Table 2.

Emissions from gypsum ponds are estimated to account for 1408 tons or 53% of total fluoride emissions from phosphate fertilizer and elemental phosphorus plants.

Table 3 summarizes emissions for 1972 from the phosphate fertilizer and elemental phosphorus industry, by type of process. It also shows the estimated percentage of emissions in the gaseous state.

Data on particle size were reported by an elemental phosphorus plant, and are shown in Table 4.

### **3.3 Primary Iron and Steel Production**

There were 45 plants in Canada engaged in the production of primary iron and steel in 1972. Primary iron production capacity was about 12 million tons, and steel furnace plant capacity was about 16 million tons (6).

**3.3.1 Process Description.** The manufacture of iron and steel comprises some or all of the following steps:

TABLE 3 SUMMARY OF EMISSIONS FOR PHOSPHATE FERTILIZER AND ELEMENTAL PHOSPHORUS PLANTS, 1972

Process	Total fluoride emissions (tons)	Percent gaseous fluoride (estimated)
Phosphoric Acid (including grinding & gypsum pond)	1608	97.9
Ammonium Phosphate (including gypsum pond)	235	91.5
Triple Superphosphate (including grinding, curing & gypsum pond)	290	99.9
Single Superphosphate (including grinding)	255	95.6
Elemental Phosphorus (including grinding)	280	90.9
<b>TOTAL</b>	<b>2668</b>	<b>96.6</b>

*Iron Production.* This takes place in the blast furnace. In this process, the iron-bearing materials (iron ore, sinter, pellets, mill scale, iron or steel scrap, etc.), fuel (coke), and flux (limestone and/or dolomite) are charged to the top of the furnace.

Heated air (blast) and, in some instances, fuel (gas, oil or powdered coal) are blown in at the bottom. The blast air burns part of the fuel to produce heat for the chemical reactions involved and for melting the iron while the balance of the fuel and part of the gas from combustion remove oxygen combined with the metal. A typical blast furnace charge in Canada, per ton of product is:

Ore, Sinter, Pellets	1.6 ton
Coke	0.5 ton
Air	2.0 tons
Limestone/Dolomite	0.1 ton

TABLE 4 PARTICLE SIZE DISTRIBUTION FOR PARTICULATE EMISSIONS FROM AN ELEMENTAL PHOSPHORUS PLANT

Source	Particle size (microns)	Weight of particulates (%)
Silica dryer	<1	1.27
	1-5	84.96
	5-10	12.33
	10-25	1.44
Coke dryer	0.4-1	6
	1-5	86
	5-10	8
Phosphate rock dryer	<1	30
	1-5	44
	5-10	14
	10-50	12

The blast furnace product is known as either hot metal (molten) or pig iron (solid). Pig iron may be remelted with scrap in cupolas to produce cast iron.

*Steel Production.* Blast furnace product and steel scrap are charged to the steel furnace, where further refinement takes place. Careful control of the levels of contaminants such as silica, phosphorus, sulphur and carbon is necessary to impart the desired mechanical and chemical characteristics to the product steel. The chemical and mechanical properties may be changed by alloying with other metals, such as nickel, chromium and manganese.

Fluorspar is used as a flux in the manufacture of steel to remove impurities during melting and to improve separation of metal and slag in the furnace by increasing the fluidity of the slag. Consumption of fluorspar in the steel industry has, in recent years, increased substantially because of increased steel output and changing technology. Steel makers have shifted increasingly from the basic open hearth process to the basic oxygen process. The latter consumes from 10 lb - 15 lb of metallurgical grade fluorspar for each ton of steel compared with 3 lb - 5 lb in the open hearth. The electric furnace process may consume from 8 lb-10 lb of metallurgical grade fluorspar for each ton of steel produced.

The principal steel furnaces used in Canada in 1972 were the basic oxygen, the basic open hearth and the electric arc, listed in decreasing order of overall production capacity. Open hearth furnaces

are becoming obsolete and should eventually be replaced by basic oxygen furnaces. The availability of cheap hydroelectric power in certain areas may sometimes favour the use of electric furnaces.

*Steel Finishing.* This includes a large number of operations, depending on the required final product. The required product shape may be achieved by casting, or in rolling mills, bar mills, railing mills, etc. Other processes such as scarfing, pickling and galvanizing are employed to achieve desired surface properties.

**3.3.2 Emissions.** The main source of fluoride emissions is steel production, which requires fluorspar as a flux. The only source of fluoride in pig iron production is as an impurity in the iron ore. The quantity present in the ore is small and emissions of fluoride are estimated to be 1.7 tons from pig iron production in 1972.

Some emissions probably also occur from certain pretreatment processes but these are employed in specialized cases and emissions are assumed to be negligible. Steel finishing operations are conducted at relatively low temperatures where the steel is heated only to a point where it can be rolled or formed. Fluoride emissions from this source are negligible.

Fluoride emissions in 1972 from steel manufacture were from the furnaces, teeming floor, and slag. Most companies which supplied information only reported losses from steel furnaces. One company reported losses from the teeming floor and none reported losses from slag.

Estimates of fluoride emissions are based on the limited amount of data reported in the questionnaires.

Uncontrolled emission factors estimated for this survey and those reported by the United States Environmental Protection Agency (2) are compared in Table 5.

The total quantity of fluoride emitted to the atmosphere in 1972 from steel furnaces was 1680 tons. The quantity emitted from the slag only was 652 tons and from teeming 84 tons. The total fluoride emissions from primary iron and steel are therefore estimated at 2418 tons in 1972. Some companies reported measured particle size data, summarized in Table 6.

### **3.4 Miscellaneous Industrial Sources**

During the survey many operations were identified which either process fluoride compounds or introduce fluoride into the process as an impurity in the raw materials; however, they were found to only emit small amounts of fluoride. The purpose of this section is to account for these operations.

**3.4.1 Clay and Shale Products.** Products manufactured from clay and shale include refractory bricks, brick and tile, clay sewer pipes, and porcelain and pottery.

No accurate data were found regarding the quantity of clay consumed in Canada during 1972. Statistics Canada reports that approximately 380 000 tons of clays including fireclays and china clays were imported in 1972 (7). Based on information received from the questionnaires and Statistics Canada (8) it is estimated that approximately 3 183 450 tons of Canadian shales and clays were

TABLE 5 COMPARISON OF FLUORIDE EMISSION FACTORS FOR STEELMAKING OPERATIONS

Source	Uncontrolled fluoride emission factor (lb F/ton steel)			
	This survey		E.P.A. (2)*	
	Gaseous	Particulate	Gaseous	Particulate
Basic oxygen furnace	0.0217	1.129	-	0.10
Open hearth furnace	0.172	2.145	0.10	0.014
Electric arc furnace	0.07	2.03	0.012	0.116
Teeming floor	0.012	-	-	-
Slag	0.093	-	-	-

\*The emission factors extracted from reference 2 are based on personal communications and have an average rating, according to EPA.

consumed for the manufacture of clay products in 1972. The average concentration of fluoride in Canadian clays and shales based on data reported in the questionnaires is 0.08%.

Fluoride emissions occur from the kilns of furnaces used in the manufacture of all types of clay products.

An average uncontrolled emission factor from companies reporting emissions is 0.09 lb fluoride/ton of clay products or 10% of the fluoride input. All of the reported emissions are based on actual source testing. The remaining 90% of the fluoride is accounted for in the products. None of the companies report having any control equipment on the kilns.

The United States Environmental Protection Agency reports an uncontrolled emission factor of 1 lb fluoride/ton of bricks which accounts for approximately 90% of the fluoride input (2). Another source reports that between 30%-86% of the fluoride input is driven off during the manufacture of bricks and pottery (9).

It is not possible to rationalize this difference in data. Fluoride emissions are based on reported Canadian data but emissions assuming 30% and 90% loss are given in Table 7 for comparison.

TABLE 6 PARTICLE SIZE DISTRIBUTION FOR PARTICULATE EMISSIONS FROM ELECTRIC ARC STEELMAKING FURNACES

Source	Particle size (microns)	Weight of particulates (%)
Electric arc furnace	<5	20-50
	5-10	30-50
	10-20	10-25
	<20	5-10
Electric arc furnace with oxygen lance	0.2-5	95

TABLE 7 FLUORIDE EMISSIONS FROM CLAY PRODUCTS MANUFACTURE, 1972

Material	Quantity processed (tons)	Quantity of fluoride input (tons)	Fluoride emissions (tons) (Canadian)		
			10% loss	30% loss	90% loss
Canadian clays and shales (0.08% F)	3 183 450	2547	255	764	2292
Imported clays and shales (0.05% F)	380 000	190	19	57	171
Total			274	821	2463

Most of the fluoride is emitted as gaseous hydrogen fluoride and/or silicon fluoride. Particulate fluoride is in the form of calcium fluoride.

Total fluoride emissions in 1972 from the manufacture of clay and shale products based on Canadian data are estimated at 274 tons.

Particle size from the kiln stack was reported by one company to be:

70% between 1 – 10 microns  
30% between 10 – 100 microns

**3.4.2 Cement and Lime Kilns.** Questionnaire replies from lime users indicate that there is no fluoride in the lime used. A textbook on the chemistry of lime and limestone states that fluorine is only found occasionally in lime and limestone and only in trace quantities (10). Based on this information, fluoride emissions from lime kilns are assumed negligible.

The quantity of clay or shale used in the manufacture of cement is approximately 5% by weight (11). Statistics Canada reports that approximately 10 000 000 tons of cement were produced in 1972 (12). Assuming the same emission factor as that estimated for the manufacture of clay products, (i.e., 10% of the fluoride input) and a control efficiency of 95% the total quantity of fluoride emitted from the manufacture of cement in 1972 is 2 tons.

**3.4.3 Glass Manufacture.** Nearly all glass produced commercially in Canada during 1972 was one of five basic types: soda-lime, lead, fused silica, borosilicate and 96% silica. Of these, modern soda-lime glass accounts for most of the Canadian production because of its suitability for melting and shaping into window glass, plate glass, containers, inexpensive tableware, electric light bulbs, and many other inexpensive, mass-produced articles.

A typical batch charge for making soda-lime flint glass consists of:

	Percentage
Silica sand	54.35
Cullet	15.37
Soda ash	16.30
Limestone	13.48
Niter	0.18
Salt cake	0.29
Decolorizer	0.03

Fluorspar as a flux in glass manufacture had been almost totally phased out by 1972. Only one company reported using fluorspar and this practice was discontinued, at that plant, late in 1972. Only one other plant, which did not reply to the survey, is still known to use fluorspar, but only to a limited extent. Emissions of fluoride were reported to be about 2 tons. An uncontrolled emission factor from data given was calculated to be 2 lb fluoride/ton of glass and the quantity of fluoride emitted uncontrolled was 17% of the original quantity in the fluorspar.

Concentrations of fluoride as an impurity in the raw materials were reported by one company and emissions of fluoride were reported from the glass melting furnace. Based on this information an uncontrolled emission factor was calculated to be 0.004 lb fluoride/ton of glass produced. The quantity

emitted accounts for 3% of the fluoride present in the raw materials and the remainder of the fluoride is in the product glass. The fluoride emitted is estimated at 3 tons assuming no control devices.

The total quantity of fluoride emitted from glass manufacture in 1972 is estimated at 5 tons.

**3.4.4 Hydrofluoric Acid and Anhydrous Hydrofluoric Acid.** It is estimated that 52 903 tons of fluorspar were processed in Canada in 1972 for the manufacture of hydrofluoric acid. The total quantity of hydrofluoric acid produced was 23 492 tons of which 13 070 tons were exported.

The main sources of fluoride emissions from the process are:

- drying and calcining of the fluorspar;
- purification process scrubber;
- aqueous production (not always a process step).

Small amounts are also emitted from fluorspar handling and storage. Another possible source, not mentioned in the questionnaire replies, is possible loss of fluoride from the gypsum residue.

Although some of the data received on emissions are questionable, a controlled fluoride emission factor is estimated to be 2.2 lb fluoride/ton of hydrogen fluoride produced. Total fluoride emissions from hydrogen fluoride production are estimated at 26 tons in 1972.

With the exception of ore handling, ore drying and calcining operations, emissions are in the form of hydrogen fluoride gas. Emissions from the drying and calcining are both hydrogen fluoride gas and calcium fluoride. Ore handling emissions are calcium fluoride only.

**3.4.5 Fluorocarbons.** An estimated 20 900 tons of fluorocarbons were produced in Canada in 1972 by two companies. Fluoride emissions are based on mass balance calculations. The companies have very different processes and emission factors cannot be compared. Total fluoride emissions to the atmosphere in 1972 from fluorocarbon manufacture as reported in the questionnaires are 12 tons. Emissions are all gaseous and were reported as fluorocarbons and hydrogen fluoride.

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**3.4.6 Production of Uranium Hexafluoride.** Hydrofluoric acid is the key chemical in the production of uranium hexafluoride from uranium trioxide. Uranium hexafluoride is used as feed material for the gaseous diffusion enrichment process. Only one company in Canada produced uranium hexafluoride in 1972 with a total output of 1903 tons (expressed as uranium).

Basically the process involves the hydrofluorination of uranium dioxide to produce uranium tetrafluoride followed by fluorination to produce uranium hexafluoride. Fluorine gas required for the process is generated by the decomposition of hydrofluoric acid by electrolysis. The gas produced is used for the reduction of uranium trioxide to uranium dioxide prior to the hydrofluorination stage.



Emissions reported were averages of all source testing done in 1972.

Some fluoride emissions were reported from process stages before any fluorine chemicals were added and were probably from impurities in the original uranium trioxide.

Total emissions of fluoride to the atmosphere in 1972 were reported to be 3.6 tons. The chemical forms of the emissions were not given but all fluoride emitted was gaseous.

**3.4.7 Porcelain Enamel.** Emissions from porcelain enamel were considered under production and use.

*Production of Porcelain Enamel.* There were only two producers of porcelain enamel in Canada in 1972. Fluoride emission information based on source testing was received from one company; however, reported emissions were four times higher than fluoride input. Based on information received and capacity data from Energy, Mines and Resources (1) the total fluorspar consumption in 1972 is estimated at 360 tons with an estimated fluoride content of 122 tons.

Because of the doubtful data received, the uncontrolled fluoride emission was assumed to be the same as that calculated for glass, that is 17% of the fluoride input. This is in agreement with published data (9) which estimate 20% fluoride loss for glass and 10% for enamel frit. No use of emission control devices was reported.

Total fluoride emissions to the atmosphere from the manufacture of porcelain enamel in 1972 are estimated at 21 tons. It is assumed that the emissions are gaseous hydrogen fluoride or fluorine, or particulate sodium fluoride.

*Use of Porcelain Enamel.* No accurate data were reported for losses of fluoride from enamel users. Some losses would be expected from baking operations due to leakage from the furnace and spray application but these should be negligible. Emissions of fluoride from porcelain enamel users in 1972 are assumed negligible.

**3.4.8 Welding Rods.** Emissions from welding rods were considered under both manufacture and use.

*Manufacture.* No information on atmospheric emissions was received in questionnaire responses.

Most welding rod manufacturers use fluorspar as a flux coating on their electrodes. The approximate quantity, based on one reply, is 50 lb fluorspar / ton of electrodes. One company reported that the temperature involved in the drying of the coatings on the electrodes is too low to volatilize any fluoride. Fluoride emissions from welding rod manufacture are assumed to be negligible.

*Use.* Based on questionnaire replies and the number of people employed by the industry (13) the quantity of fluorspar consumed by welding rod manufacturers in 1972 is estimated at 2450 tons.

Emissions of fluoride from the use of welding rods is primarily an industrial hygiene problem although some fluoride may escape from the welding area and constitute air emissions. No data are available and the fluoride emissions are estimated at 10% of the original quantity of fluoride in the

welding rods. The total quantity of fluoride emitted in 1972 from welding rod use is estimated at 83 tons.

**3.4.9 Primary Nonferrous Metal Production.** Fluoride is introduced to the primary nonferrous processes as an impurity in the ore concentrates. Quantities in the concentrates are reported to range from 0.001% - 0.03%. The following summarizes the information received.

*Zinc.* Fluoride concentrations in zinc ores average 0.005% by weight. The total quantity of fluoride input to primary zinc plants in 1972 is estimated at 52 tons. One company provided information on source testing which indicates that fluoride emitted from the process is approximately 0.3% of the original fluoride input. Total emissions of fluoride from primary zinc in 1972 are estimated at 0.2 ton.

*Lead.* Fluoride concentrations in lead ores average 0.004%. The total fluoride input to primary lead is estimated to be 7 tons. Information from one company indicated a loss of approximately 2.28% of the original quantity in the ore. Emissions of fluoride from primary lead in 1972 are estimated at 0.2 ton.

*Copper and Nickel.* The overall average fluoride concentration for both copper and nickel ores is 0.0135%. Concentrations range from 0.005% - 0.03%. Total fluoride input to primary copper and nickel production in 1972 is estimated at 500 tons. One company provided information which indicates a loss of 3.8% of the original fluoride. Losses of fluoride to the atmosphere in 1972 from primary copper and nickel are estimated at 19 tons. Emissions were reported as particulates in the form of calcium fluoride and/or sodium fluoride.

**3.4.10 Mining Operations.** Only one company mined fluorspar in Canada in 1972 from three mine locations. An estimated 167 700 tons was mined based on reported company data. There was no information on emissions reported for any of the three mines.

Based on information from the United States Environmental Protection Agency (14, 15) an emission factor of 0.2 lb fluoride/ton fluoride in the ore is assumed.

Total emissions of fluoride from fluorspar mining operations in 1972 are estimated at 8 tons.

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**3.4.11 Metallurgical Coke Manufacture.** There were seven coke manufacturing companies in Canada in 1972 with a total coke production of 5.2 million tons (16).

It is assumed that most of the fluoride contained in coal is volatilized during coking and removed in the scrubbing/cooling towers; therefore, fluoride emissions to the atmosphere can only occur during coal handling operations. Since these are low-temperature processes, the content of the particulate emissions would be similar to that of coal.

It is reported (17) that about 1.5 lb of particulates are emitted to the atmosphere per ton of coke produced and the fluoride content of coking coal, reported by one company, is about 50 ppm.

Total fluoride emissions from coke manufacturing in 1972 are estimated at less than 0.5 ton.

**3.4.12 Ferrous Foundries.** Of the companies replying to the questionnaire only three large foundry operations reported using fluorspar. These companies are iron foundries using cupolas. Their raw materials include pig iron, ferromanganese, spiegeleisen and scrap iron. None of these companies provided fluoride emission information.

Iron foundries usually simply melt and cast iron in cupolas with no alloying or purification. This is not the case for steel foundries, however, where alloying is sometimes carried out. Steel foundries in most cases make use of electric arc furnaces.

The three reporting iron foundries had wet scrubbers capturing particulates from their cupolas at an average efficiency of 95%. Steel foundries usually use baghouses to remove particulates from electric arc furnaces, again at an average efficiency of 95% (18).

Total fluorspar consumption by ferrous foundries in Canada in 1972 was 7358 tons, 78.3% of this total was used by four iron foundries in Ontario, 12.8% by three steel foundries in Quebec, 5.7% by one iron foundry in British Columbia and 3.2% by one iron foundry in New Brunswick (18). All except the New Brunswick foundry had control equipment (18).

Emissions from fluorspar-consuming foundries were estimated using an uncontrolled emission factor of 2.1 lb fluoride/ton product (2.03 lb particulate fluoride and 0.07 lb gaseous fluoride), developed for electric furnaces in section 3.3. Lacking better information this factor was used for both cupolas and electric arc furnaces. Foundries not using fluorspar were assumed to have no fluoride emissions. Total emissions of fluoride in 1972 from fluorspar-consuming ferrous foundries are therefore estimated at 78.4 tons.

**3.4.13 Nonferrous Foundries.** None of the nonferrous foundries reported using fluorspar or emitting fluoride from their process. There was no evidence found in the literature of emissions of fluoride from nonferrous foundries.

**3.4.14 Petroleum Industry.** There was no evidence found to indicate the presence of fluoride in crude oils; however, hydrofluoric acid is used as a catalyst for alkylation. Alkylation is a process used for combining an isoparaffin with an olefin, or olefin mixtures, to form a highly branched, high octane paraffinic product. Hydrofluoric acid consumption is between 0.3 lb - 0.8 lb hydrofluoric acid/barrel of alkylate.

No information on hydrofluoric acid consumption or emissions was received.

Losses of hydrofluoric acid from the process may occur at the following points:

**Regenerator Bottoms.** During normal operating conditions and also during upsets, some hydrofluoric acid is carried with the regenerator bottoms. This acid is neutralized with caustic and very little escapes to the atmosphere.

**Hydrofluoric Acid Stripper Bottoms.** As in the regenerator bottoms, some hydrofluoric acid is entrained with the propane. The propane product is neutralized before being sent to storage and very little fluoride emission is expected.

*Defluorinator.* Some hydrofluoric acid and fluoride is absorbed in the defluorinator before entering the debutanizer. No atmospheric emissions are expected from this stage.

*Maintenance Shutdown.* During a scheduled or emergency shutdown some hydrofluoric acid is lost to the atmosphere.

Only trace amounts are carried over with the alkylate and therefore do not present a pollution problem from combustion of these fuels.

Only five companies were using hydrofluoric acid alkylation in 1972 (19). Total consumption of hydrofluoric acid was estimated, from capacity data (19), to be 85 tons. It is estimated that fluoride emissions in 1972 were less than 1 ton.

Fluoride emissions from miscellaneous industrial sources are summarized in Table 8. Also given is the estimated percentage of emissions which are in the gaseous state.

#### **4 EMISSIONS FROM FUEL COMBUSTION - STATIONARY SOURCES**

Stationary sources can be divided into the following categories:

- Power generation
- Industrial and commercial
- Domestic

The five principal types of fuels used are gaseous (natural gas, propane, etc.) diesel oils, light oils, heavy oils and coal. Of these only coal contains fluoride (section 3.4.11 and reference 9).

A paper presented in the Journal of the Air Pollution Control Association (9) gives the range of fluoride concentrations in British coals as 0 ppm - 175 ppm. The same paper states that American coals have fluoride concentrations up to 295 ppm. One steel company reported a fluoride concentration in coking coal of 50 ppm. During combustion of the coal most of the fluoride is driven off and a small amount remains in the ash (9). Based on this information the uncontrolled emission factor for coal combustion is estimated to be 0.12 lb fluoride/ton of coal.

##### **4.1 Power Generation**

In 1972, generation of electricity from coal-powered facilities accounted for 62% of total thermal generation; petroleum fuels were responsible for 10%, natural gas for 15.7% and nuclear fuels for 12.3%.

Uncontrolled fluoride emissions from power generation in 1972 are estimated at 1006 tons. Fluoride emissions are assumed to be gaseous and the control devices used are effective only on particulates.

Roughly 60% of the emissions occur during the winter months (20), depending on climate and the extent to which electricity is used for heating purposes.

TABLE 8 FLUORIDE EMISSIONS FROM MISCELLANEOUS INDUSTRIAL SOURCES, 1972

	Total fluoride emissions (tons)	Percent gaseous fluoride (estimated)
Clay and shale products	274	90
Cement and lime kilns	2	unknown
Glass manufacture	5	unknown
Hydrofluoric acid and anhydrous hydrofluoric acid	26	77
Fluorocarbons manufacture	12	100
Uranium hexafluoride production	3.6	100
Porcelain enamel production and use	21	50
Welding rod manufacture and use	83	52
Primary nonferrous metal production	19.4	0
Mining operations	8	0
Metallurgical coke manufacture	<0.5	100
Ferrous foundries	78.4	51
Nonferrous foundries	nil	nil
Petroleum industry	<1	unknown
TOTAL	534	70-75 (approx.)

#### 4.2 Industrial and Commercial

Approximately 2.7 million tons of coal were consumed for industrial and commercial purposes in Canada during 1972. The figure of 2.7 million tons of coal does not include coal used for coke manufacturing (section 3.4.11).

Coal is used throughout the year for process heating. In the winter, however, it is also used for plant heating resulting in a seasonal increase in fluoride emissions.

Fluoride emissions from the combustion of industrial and commercial fuels during 1972 are estimated at 162 tons.

#### **4.3 Domestic**

Very little coal was used for domestic heating in 1972.

### **5 EMISSIONS FROM SOLID WASTE INCINERATION**

In 1972, approximately 1.4 million tons of refuse were disposed of in 17 municipal incinerators (21).

#### **5.1 Composition of Municipal Refuse**

The typical composition of municipal refuse is shown in Table 9 (22).

Of these materials only glass, ceramics, and vegetable food wastes contain appreciable amounts of fluorine. A report by the National Research Council (3) indicates the average fluoride content of vegetable wastes to be 10 ppm. One company reported a concentration of fluorine in glass of 90 ppm.

#### **5.2 Emissions**

Based on the above information the total fluoride input to Canadian incinerators is estimated at 10 tons. Assuming a control level of about 60%, the total fluoride emitted in 1972 in Canada is about 4 tons.

TABLE 9 COMPOSITION OF COMPOSITE MUNICIPAL REFUSE

Material	Percent by weight
Corrugated paper boxes	23.38
Newspaper	9.40
Magazine paper	6.80
Brown paper	5.57
Mail	2.75
Paper food cartons	2.06
Tissue paper	1.98
Plastic-coated paper	0.76
Wax cartons	0.76
Vegetable food wastes	2.29
Citrus rinds and seeds	1.53
Meat scraps, cooked	2.29
Fried fats	2.29
Wood	2.29
Ripe tree leaves	2.29
Flower garden plants	1.53
Lawn grass, green	1.53
Evergreens	1.53
Plastics	0.76
Rags	0.76
Leather goods	0.38
Rubber composition	0.38
Paints and oils	0.76
Vacuum cleaner catch	0.76
Dirt	1.53
Metals	6.85
Glass, ceramics, ash	7.73
Adjusted moisture	9.05
Other	0.01
TOTAL	100.00

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**APPENDIX - SURVEY OF QUESTIONNAIRE RETURNS AND QUALITY INDEX**

Sector	Number of questionnaires		Rate of response (%)	Quality index of questionnaires returned*					Average quality index
	Sent	Returned		1	2	3	4	5	
Primary copper & nickel & assoc.	13	12	92	-	-	-	4	8	4.7
Primary iron & steel	45	28	62	2	2	5	7	12	3.9
Primary aluminum	6	6	100	-	-	-	-	6	5.0
Primary lead & zinc	6	6	100	-	-	-	-	6	5.0
Copper, brass & bronze foundries	84	27	32	2	4	7	8	6	3.4
Die casting	91	51	56	4	18	14	10	5	2.9
Iron & steel foundries	194	88	45	10	16	25	24	13	3.2
Metallurgical coke	7	3	43	-	-	1	-	2	4.7
Phosphate fertilizers & elemental phosphorus	31	28	90	-	1	13	2	12	3.9
Galvanizing	46	26	57	2	3	10	10	1	3.2
Plating	83	25	30	7	9	3	6	-	2.3
Ferroalloys & assoc.	13	4	31	-	-	-	1	3	4.8
Pesticides	58	26	45	9	10	-	4	3	2.3
Ceramics and glass	105	44	42	1	8	6	18	11	3.7
Nonferrous alloys	20	11	55	1	2	-	5	3	3.6
Battery mfg.	5	2	40	-	-	-	1	1	4.5
Rubber mfg.	24	20	83	-	2	5	7	6	3.9
Zinc oxide mfg.	2	1	50	-	-	-	-	1	5.0
Mining	39	21	54	2	2	4	5	8	3.7
Porcelain & enamels	30	17	57	4	4	4	3	2	2.7
Clay products mfg.	57	21	37	7	4	3	4	3	2.6
Petroleum refining	47	35	74	4	5	11	-	15	3.5
Vinyl stabilizers & plasticizers	13	8	62	-	-	2	3	3	4.1
Welding rod mfg.	12	10	83	2	2	-	2	4	3.4
Miscellaneous	16	13	81	-	-	4	3	6	4.2
<b>TOTAL</b>	<b>1047</b>	<b>533</b>	<b>51</b>	<b>57</b>	<b>92</b>	<b>117</b>	<b>127</b>	<b>140</b>	<b>3.4</b>

\* Quality index of questionnaires returned was based on the following rating:

- (1) No emissions reported, no back-up given
- (2) No pertinent information.
- (3) Poor reply, data insufficient
- (4) Medium reply, emissions can be calculated
- (5) Good reply, emissions data given and documented

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